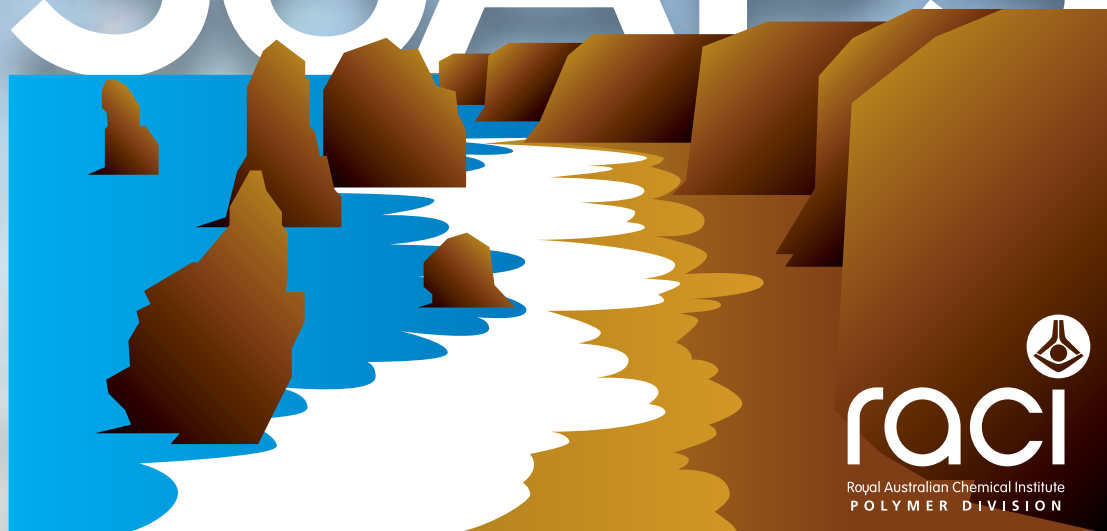


# 36APS



## **36TH AUSTRALASIAN POLYMER SYMPOSIUM MANTRA LORNE, VICTORIA 20-23 NOVEMBER 2016**

[WWW.36APS.ORG.AU](http://WWW.36APS.ORG.AU)

### HANDBOOK

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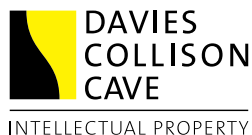
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The American Chemical Society publishes three leading journals in Polymer Science. Macromolecules is the Most Cited and highest impact original research journal in Polymer Science with an Impact Factor of 5.800. Biomacromolecules is the highest impact original research journal interfacing Polymer Science and Biology with an Impact Factor of 5.750. ACS Macro Letters is the highest impact, fastest Letters journal in Polymer Science with an Impact Factor of 5.764.

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MANTRA LORNE, VICTORIA 20-23 NOVEMBER 2016



## WELCOME

On behalf of the Polymer Division of The Royal Australian Chemical Society (RACI), it is my great pleasure to welcome you to the 36th Australasian Polymer Symposium in Lorne.

The themes for 36APS will cover all areas of polymer science, engineering and technologies from the latest innovations and new methods in synthesis, polymeric architectures and nanoparticles, characterization, processing and molecular modelling to the advanced applications of polymers in health, nanomedicine, energy, electronic materials and sustainability as well as polymers in industry and translational research.

The APS has a long established tradition of bringing together the polymer science and engineering communities of Australasia as well as esteemed polymer colleagues from around the world; from the current world-leaders in polymer research to academics, researchers, research students and industrial professionals from universities, research organisations and polymer industry in Australia and around World.

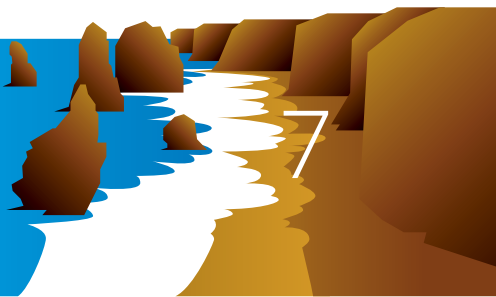
As in past years, the APS will provide an opportunity for national and international networking through an exciting forum of both formal presentations as well as informal idea exchanges in order to contribute towards the development of frontier polymer research. The program has been specifically designed to provide ample opportunities for informal discussions and networking throughout the conference.

Please enjoy the conference and Lorne.

**Greg Qiao**

36APS Convenor

Chair, Polymer Division of the RACI



# Unprofessional Conduct Declaration

The host organization for 36APS is the Polymer Division of the Royal Australian Chemical Institute. Unprofessional conduct breaches the inherent principle of the RACI Code of Ethics (By-Law 26, in particular section 26.2). It has no place at any time, especially at conferences or other meetings of the RACI. Unprofessional conduct includes sexual harassment and discrimination in respect to sex, race, creed and sexual preference or status. The unprofessional conduct may be verbal, written or implied.

The Polymer Division of the RACI expect delegates and members to conduct themselves in a professional manner at all times. The Organising Committee in turn is committed to present and manage 36APS in a professional way.

Substantive complaints against RACI members and other professional organisations will be referred to the RACI Executive Council for action under By-Law 26 or for other recommended action as appropriate.

# RACI Polymer Division Meetings

Website: <http://www.polymer.org.au/>

The Royal Australian Chemical Institute maintains a strong involvement in polymer science and technology through the national Polymer Division, as well as through the RACI State Polymer Groups, and a special liaison with the New Zealand Institute of Chemistry Polymer Group. The Polymer Division concentrates on national and international activities, whilst the Polymer Groups focus on local activities. The Polymer Division is coordinated by a standing committee – it is elected at the General Meeting held at each Australasian Polymer Symposium; the Chairpersons of the State Polymer Groups are ex-officio members of the Polymer Division Committee. The RACI Polymer Division was formed in 1964 with the objective of advancing the theory and practice of polymer science in Australia. Prior to this, the Polymer Group of the Victorian Branch took the initiative, with a National focus, and was responsible for organising the first three Polymer symposia in Australia. As a result, there is some debate as to which was the “first” APS, with legitimate claims by the 1957 Adelaide, 1964 Mildura and the 1966 Canberra conferences.

Since the latter was the first conference following the formation of the Polymer Division, it has been designated 1APS and used to number all subsequent Symposia. More information on the history of the Polymer Division can be found at the Division web site (<http://www.polymer.org.au/index.php/home/history>).

Even though the Polymer Division is part of a Chemical Institute, the Division has always looked beyond the chemistry of polymers and takes a broader view to encompass chemical, physical, engineering and industrial aspects of polymers. A feature of the Polymer Division's activities has been the participation of chemists, physicists, engineers, mathematicians and biologists with an interest in polymers. Polymer Division meetings bring together industrialists and academic and government polymer scientists and technologists from all over Australasia and from overseas.

While the most visible activity of the Polymer Division are the Australasian Polymer Symposia (APS), which are held every 12–18 months, the Division is also the focal point for international interactions and was a foundation member of the Pacific Polymer Federation, which brings together scientists from Japan, Australia, New Zealand, United States of America, South East Asia and many other Pacific nations. The first Pacific Polymer Congress was held in Hawaii in 1989, followed by Japan in 1991, Brisbane, Australia in 1993, Hawaii in 1995, Korea in 1997, China in 1999, Mexico in 2001, Thailand in 2003, Hawaii again in 2005, Kobe in 2007, and Cairns, Australia in 2009 and organized by the Australian RACI Polymer Division. The leadership demonstrated in this area has resulted in the Polymer Division remaining a key entity in world polymer science and has resulted in the cosponsorship of many conferences held around the world and the encouragement of polymer science in developing Pacific Rim countries.

The Division is also a strong supporter of student development, the future of the Division, through the subsidised attendance and participation at national conferences from early stages of degrees. As a means of achieving this objective, the Division provides financial assistance to postgraduate students to allow them to attend Polymer Division symposia and provides the Treloar prizes at each APS for the best paper and poster presented by a polymer scientist under the age of thirty.

Additionally, the Division is a sponsor and supporter of the Polymer Summer School (in conjunction with the CRC for Polymers). It also fosters student development by subsidising travel to related conferences and specialist symposia, usually in collaboration with the State Polymer Groups.

The Division also sponsors a series of forums which are for and organised by Polymer Students. The format includes students giving overviews of their own and of their group's research, along with talks by representatives of industry, academia and government. These forums enable research students to interact scientifically and to start the networking that is an essential part of effective science and technology. The absence of supervisors is a vital component for the success of these meetings!

The Polymer Division provides a range of other services for the Australasian polymer community, including a directory of Polymer Science and Engineering in Australasia, which lists scientists and research establishments, and web pages providing information on polymer science activities in Australia and New Zealand.

The RACI Polymer Division has a number of prestigious awards – see <http://www.polymer.org.au/index.php/home/awards>. The most distinguished is the Australian Polymer Medal for eminence in Australian Polymer Science. In 1992 the Australian Polymer Science & Technology Achievement Award, now called the David Sangster Polymer Science & Technology Achievement Award, was created to recognize the achievement of Australian polymer scientists while their careers are still developing (age limit of 40). Citations are also awarded at Australasian Polymer Symposia for significant service and contributions in any aspect of polymer science, technology or education. There are also awards to encourage young polymer scientists. As already mentioned, Treloar Prizes are awarded for the best presentations at each APS by scientists under the age of 30. The inaugural O'Donnell Young Scientist Award was made at 21APS. This is for a PhD student to undertake a short attachment to another University to perform research towards the PhD, and is given in recognition of the longstanding support and encouragement of young polymer scientists by the late Prof James O'Donnell of the University of Queensland. Prof O'Donnell's vision for polymer scientists in Australia provided many tangible benefits to young Australian polymer scientists, such as reduced conference registration fees, travel subsidies and prizes. This has resulted in increased student participation in the Division at all levels, and consequently in an increased profile in the national and international polymer community.

In 2000 the Division initiated a lecture series aimed at bringing a prominent scientist to the major polymer centres in Australia between APS meetings. It was decided to name the series after David Solomon, recently of the University of Melbourne, in recognition of his long-standing and continuing leadership in the Polymer Division. It was also deemed appropriate that he should also be the inaugural lecturer of the series named after him.

A complete list of the activities and services is included in the Division's webpage ([www.polymer.org.au](http://www.polymer.org.au)). The Polymer Division welcomes suggestions of new initiatives and participation in the organisation of them. Of greatest importance, at all of its meetings the Polymer Division provides an excellent means by which professionals in the field of polymer science can extend their scientific experience and interact with a large group of their professional colleagues.

**RACI Polymer Division**  
Standing Committee



# RACI Polymer Division Standing Committee 2015 - 2016

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# RACI Polymer Division Meetings

## Australasian Polymer Symposia

| APS YEAR | VENUE                 | DIVISION CHAIR                                                                      |
|----------|-----------------------|-------------------------------------------------------------------------------------|
|          | 1957 Adelaide         | <b>D O Jordan</b>                                                                   |
|          | 1963 Mildura          | <b>D H Solomon</b>                                                                  |
|          | 1964 Mildura          | <b>B S Harrup</b>                                                                   |
| 1        | 1966 Canberra         | <b>F W Ayscough</b> (with 3rd Nat. Conv.)                                           |
| 2        | 1968 Canberra         | <b>D H Solomon</b> (with Aust. Biochem. Soc.)                                       |
| 3        | 1969 Sydney           | <b>H J Battaerd</b> (with 22nd IUPAC Conf.)                                         |
| 4        | 1970 Canberra         | <b>D O Jordan</b> (with 4th Nat. Conv.)                                             |
| 5        | 1971 Mildura          | <b>M J Jordan</b>                                                                   |
| 6        | 1973 Broadbeach       | <b>J H O'Donnell</b>                                                                |
| 7        | 1974 Canberra         | <b>J H Bradbury</b> (with 5th Nat. Conv.)                                           |
| 8        | 1975 Terrigal         | <b>I C Watt</b>                                                                     |
| 9        | 1977 Tanunda          | <b>P E M Allen</b>                                                                  |
| 10       | 1978 Surfers Paradise | <b>B A Bolto</b> (with 6th Nat. Conv.)                                              |
| 11       | 1980 Lorne            | <b>C E M Morris</b>                                                                 |
| 12       | 1981 Blackheath       | <b>D F Sangster</b>                                                                 |
| 13       | 1982 Canberra         | <b>D R G Williams</b> (with 7th Nat. Conv.)                                         |
| 14       | 1984 Ballarat         | <b>G B Guise</b>                                                                    |
| 15       | 1986 Leura            | <b>D H Napper</b>                                                                   |
| 16       | 1987 Philip Island    | <b>R A Shanks</b>                                                                   |
| 17       | 1989 Brisbane         | <b>D J T Hill</b>                                                                   |
| 18       | 1990 Bendigo          | <b>E Rizzardo</b>                                                                   |
| 19       | 1992 Perth            | <b>G M Ferguson</b>                                                                 |
| 20       | 1995 Adelaide         | <b>H K Toh</b>                                                                      |
| 21       | 1996 Wollongong       | <b>M Binns</b> (Division Chair); <b>G Spinks</b> (Conf. Chair)                      |
| 22       | 1997 Auckland (NZ)    | <b>R Gilbert</b> (Division Chair); <b>R Cooney</b> (Conf. Chair)                    |
| 23       | 1999 Geelong          | <b>W D Cook</b>                                                                     |
| 24       | 2001 Beechworth       | <b>G Moad</b>                                                                       |
| 25       | 2002 Armidale         | <b>T P Davis</b>                                                                    |
| 26       | 2003 Noosa            | <b>A K Whittaker</b>                                                                |
| 27       | 2004 Adelaide         | <b>D A Lewis</b>                                                                    |
| 28       | 2006 Rotorua (NZ)     | <b>G T Russell</b>                                                                  |
| 29       | 2007 Hobart           | <b>C Barner-Kowollik</b>                                                            |
| 30       | 2008 Melbourne        | <b>G P Simon</b>                                                                    |
| 31       | 2009 Cairns           | (with PPC11) <b>M Stenzel</b> (Division Chair);<br><b>A Whittaker</b> (Conf. Chair) |
| 32       | 2011 Coffs Harbour    | <b>M Stenzel</b>                                                                    |
| 33       | 2012 Hobart           | <b>S Perrier</b>                                                                    |
| 34       | 2013 Darwin           | <b>K Jack</b>                                                                       |
| 35       | 2015 Gold Coast       | <b>A Ellis</b>                                                                      |
| 36       | 2016 Lorne            | <b>G Qiao</b>                                                                       |

# Other Symposia and Courses

| YEAR | VENUE         | TITLE                                                                                                                           |
|------|---------------|---------------------------------------------------------------------------------------------------------------------------------|
| 1968 | Melbourne     | Australian Polymer Workshop                                                                                                     |
| 1969 | Sydney        | Residential Polymer Studies School                                                                                              |
| 1971 | Adelaide      | Tailor Made Molecules                                                                                                           |
| 1973 | Sydney        | Natural and Synthetic Fibres                                                                                                    |
| 1975 | Melbourne     | Polymer Degradation                                                                                                             |
| 1976 | Warburton     | Polymer Degradation                                                                                                             |
| 1977 | Warburton     | Ion Radicals in Growth and Development                                                                                          |
| 1978 | Sydney        | Polymer Identification and Characterisation                                                                                     |
| 1979 | Warburton     | Adhesion and Adhesives                                                                                                          |
|      | Leura         | Polymer Education                                                                                                               |
| 1980 | Melbourne     | Adhesion Science and Technology                                                                                                 |
| 1981 | Melbourne     | Polymer Materials Science                                                                                                       |
| 1983 | Melbourne     | Structure Property Relationships in Synthetic Polymers                                                                          |
|      | Melbourne     | Transitions and Relaxations in Synthetic Polymers                                                                               |
|      | Melbourne     | Poly(Vinyl Chloride)                                                                                                            |
|      | Melbourne     | Patents in Chemical Industry and Research<br>(with Institute of Patent Attorneys)                                               |
|      | Canberra      | Aust. Polymer Discussion Group: Free Radical Polymerization                                                                     |
| 1984 | Sydney        | Polymers as Materials                                                                                                           |
|      | Melbourne     | Polymers as Materials                                                                                                           |
|      | Melbourne     | Deformation, Failure and Strengthening of Polymers (with<br>Materials Engineering, Monash University)                           |
|      | Sydney        | Chemistry and Physics of Elastomers                                                                                             |
|      | Canberra      | Polymerization: Polymer Degradation<br>(Australian Polymer Discussion Group)                                                    |
| 1985 | Melbourne     | POLYMER 85: Characterisation and Analysis of Polymers<br>(with IUPAC, AAA, AATS)                                                |
|      | Melbourne     | Characterisation and Flow of Rheologically Complex Liquids<br>(with Chemical Engineering, Melbourne University)                 |
|      | Sydney        | Electrical, Optical and Acoustic Properties of Polymers<br>(with Physics, Macquarie University)                                 |
|      | Canberra      | Polymerization: Mechanical Properties<br>(Australian Polymer Discussion Group)                                                  |
| 1986 | Sydney        | Emulsion Polymers and Latexes<br>Brisbane Controlled Release Technology<br>(with Australian Pharmaceutical Science Association) |
|      | Canberra      | Polymerization: Kinetics & Mechanism<br>(Australian Polymer Discussion Group)                                                   |
| 1987 | Philip Island | Thermal Characterisation of Polymeric Materials                                                                                 |
|      | Noosa         | Radiation Effects on Polymeric Materials<br>(with ACS Polymer Chemistry Division)                                               |
|      | Sydney        | Copolymerisation (with 8th Nat. Conv.)                                                                                          |
|      | Canberra      | Polymer Spectroscopy (Australian Polymer Discussion Group)                                                                      |
| 1988 | Melbourne     | Controlled Release: Science and Technology                                                                                      |
| 1989 | Brisbane      | Modern Methods for Characterisation of Polymers<br>(with Division of Analytical Chemistry)                                      |
|      | Maui          | Pacific Polymer Congress<br>(organised by the Pacific Polymer Federation)                                                       |

|      |                             |                                                                                                                     |
|------|-----------------------------|---------------------------------------------------------------------------------------------------------------------|
| 1991 | Honolulu<br>Melbourne       | Pacifichem<br>Polymer Materials Preparation, Characterisation, Properties<br>(at POLYMER 91; with IUPAC, AAS, AATS) |
| 1992 | Japan<br>Melbourne<br>Perth | Third Pacific Polymer Congress<br>RACI 9th National Convention<br>Polymers in Medicine                              |
| 1993 | Canberra<br>Brisbane        | Aust. Polymer Discussion Group: Polymer Networks<br>4th Pacific Polymer Congress                                    |
| 1994 | Canberra                    | Aust. Polymer Discussion Group:<br>Adhesion and Advanced Adhesives                                                  |
| 1995 | Adelaide<br>Kauai           | RACI 10th National Convention (Thermal Analysis)<br>5th Pacific Polymer Congress                                    |
| 1996 | Wollongong<br>Canberra      | Polymer Reaction Engineering Workshop<br>Aust. Polymer Discussion Group: Polymer Toughening                         |
| 1997 | Canberra<br>Seoul           | Aust. Polymer Discussion Group: Polymer Dynamics<br>6th Pacific Polymer Congress                                    |
| 1998 | Gold Coast                  | Macro 98 (IUPAC World Polymer Congress)                                                                             |
| 1999 | Coburn                      | Thermal Characterisation of Polymers                                                                                |
| 2001 | Beechworth                  | Biomaterials (with Aust Society of Biomaterials)                                                                    |
| 2002 | Armidale                    | Living Polymer Radical Polymerisation                                                                               |

In addition, the Polymer Division has part sponsored the Radiation Chemistry Conferences organised by the Australian Institute for Nuclear Science and Engineering, held at Lucas Heights every two years from 1972.

## Australasian Polymer Students' Forums

(<http://www.polymer.org.au/index.php/home/activities/student-forum>)

| YEAR | VENUE                    | ORGANISING INSTITUTION        |
|------|--------------------------|-------------------------------|
| 1990 | Mt Tambourine, QLD       | University of Queensland      |
| 1994 | Cunningham's Gap, QLD    | University of Queensland      |
| 1996 | Stradbroke Island, QLD   | University of Queensland      |
| 1997 | Avoca Beach, NSW         | University of New South Wales |
| 1999 | Old Clarendon Winery, SA | University of South Australia |
| 2001 | Lake Hume Resort, NSW    | University of Sydney          |

## Australasian Polymer Summer Schools

(<http://www.polymer.org.au/index.php/home/activities/polymer-summer-school>)

| APS | YEAR | VENUE                  | ORGANISING INSTITUTION                           |
|-----|------|------------------------|--------------------------------------------------|
| 1   | 2000 | Thredbo, NSW           | CRC for Polymers/Key Centre for Polymer Colloids |
| 2   | 2001 | Albury, NSW            | CRC for Polymers/Key Centre for Polymer Colloids |
| 3   | 2002 | Armidale, NSW          | CRC for Polymers/Key Centre for Polymer Colloids |
| 4   | 2003 | Albury, NSW            | CRC for Polymers/Key Centre for Polymer Colloids |
| 5   | 2004 | Wamberal, NSW          | CRC for Polymers/Key Centre for Polymer Colloids |
| 6   | 2004 | Sunset Cove, SA        | CRC for Polymers/Key Centre for Polymer Colloids |
| 7   | 2006 | Ballarat, VIC          | CRC for Polymers/Key Centre for Polymer Colloids |
| 8   | 2007 | Geelong, VIC           | CRC for Polymers/Key Centre for Polymer Colloids |
| 9   | 2008 | Brisbane, QLD          | CRC for Polymers                                 |
| 10  | 2009 | Blue Mountains, NSW    | CRC for Polymers                                 |
| 11  | 2010 | Melbourne, VIC         | CRC for Polymers                                 |
| 12  | 2010 | Wollongong, NSW        | CRC for Polymers                                 |
| 13  | 2011 | Brisbane, QLD          | CRC for Polymers                                 |
| 14  | 2012 | Adelaide               | Flinders                                         |
| 15  | 2013 | Sydney                 | UWS                                              |
| 16  | 2014 | Melbourne              | Monash                                           |
| 17  | 2015 | Monash University, VIC | Monash University                                |

# Sponsored International Scientists

- 1968 **K F O'Driscoll**, State University of New York, Buffalo, USA  
**S Okamura**, University of Kyoto, JAPAN
- 1970 **P Plesch**, University of Keele, UK
- 1971 **P J Flory**, Stanford University, USA
- 1972 **M B Huglin**, University of Salford, UK
- 1973 **H Morawetz**, Polytechnic Institute of New York, USA
- 1974 **M Nagasawa**, Nagoya University, JAPAN  
**H F Mark**, Polytechnic Institute of New York, USA  
**R L Whistler**, Purdue University, USA  
**V T Stannett**, North Carolina State University, USA
- 1975 **F W Billmeyer**, Rensselaer Polytechnic Institute, USA  
**A M North**, Strathclyde University, UK  
**R Takemoto**, Osaka University, JAPAN
- 1976 **J Silverman**, University of Maryland, USA  
**F R Mayo**, Stanford Research Institute, USA
- 1977 **H Benoit**, Centre de Recherches sur la Macromolecules, FRANCE  
**WO Statton**, University of Utah, USA  
**M Swarc**, State University of New York, Syracuse, USA  
**R B Seymour**, University of Southern Mississippi, USA
- 1978 **Y Tabata**, University of Tokyo, JAPAN  
**J P Kennedy**, University of Akron, USA  
**D M Wiles**, Canadian National Research Council, CANADA  
**RA Hochschwender**, American Hoechst Corporation, USA  
**P D Calvert**, University of Sussex, UK
- 1980 **W D Woolley**, Building Research Establishment, Boreham Wood, UK  
**T T Tsuruta**, University of Tokyo, JAPAN  
**G B Butler**, University of Florida, USA  
**A N Gent**, University of Akron, USA  
**W Schnabel**, HahnMeitner Institut fuer Kernforschung, Berlin, GERMANY  
**D H Richards**, Waltham Abbey, UK  
**C E Carraher**, Wright State University, Dayton, USA  
**J E Guillet**, University of Toronto, CANADA  
**C H Bamford**, Liverpool University, UK  
**R D Haward**, Birmingham University, UK  
**D Hull**, Birmingham University, UK
- 1982 **R J Irvin**, Queen's University, Belfast. IRELAND  
**D O Hummell**, University of Koln, GERMANY  
**G Kraus**, Phillips Petroleum, USA  
**D B Wetlaufer**, University of Delaware, USA  
**R Wuthrick**, ETH, Zurich, SWITZERLAND  
**H Ruterjans**, Goethe University, Frankfurt, GERMANY
- 1983 **A M North**, Strathclyde University, UK  
**G Williams**, University College of Wales, UK  
**RA Wetton**, Loughborough University, UK
- 1984 **T Otsu**, Osaka City University, JAPAN  
**G L Wilkes**, Virginia Polytechnic, USA  
**T C Ward**, Virginia Polytechnic, USA  
**J E McGrath**, Virginia Polytechnic, USA  
**C B Bucknall**, Cranfield Institute of Technology, UK  
**A Chapiro**, CNRS, Paris, FRANCE



- 1985 **A Peterlin**, National Bureau of Standards, USA  
**H Brown**, Case Western Reserve University, USA  
**G Williams**, Aberystwyth University College, UK  
**R J Young**, Queen Mary College, UK  
**R A Pethrick**, Strathclyde University, UK  
**J E Mark**, University of Cincinnati, USA  
**C Berry**, Carnegie Mellon University, USA  
**B Billingham**, University of Sussex, UK  
**F Ciardelli**, Università di Pisa, ITALY  
**D T Clark**, ICI Runcorn, UK  
**A E Hamielec**, McMaster University, USA  
**H J Harwood**, University of Akron, USA  
**R W Lenz**, University of Massachusetts, USA  
**J L Koenig**, Case Western Reserve University, USA  
**N Plate**, USSR Academy of Science, USSR  
**J C Randall**, Phillips Petroleum, USA  
**W H Stranes**, Bell Laboratories, USA  
1986 **T Saegusa**, Kyoto University, JAPAN  
**J Economy**, 113M, San Jose, USA  
**R Qian**, Academia Sinica, CHINA  
**D Bassett**, Union Carbide, USA  
**J R Robinson**, University of Wisconsin, USA  
**J Heller**, SRI International, California, USA  
**J Sohma**, Hokkaido University, JAPAN  
1987 **E A Turi**, New York Polytechnic Institute, USA  
**K F O'Driscoll**, University of Waterloo, CANADA  
**V Crescenzi**, University of Rome, ITALY  
**P J Lemstra**, Eindhoven University of Technology, NETHERLANDS  
**W F Maddams**, BP Research and University of Southampton, UK  
**O Vogl**, Polytechnic Institute of New York, USA  
**G N Foster**, Union Carbide, USA  
**J K Gillham**, Princeton University, USA  
**J E McGrath**, Virginia Polytechnic Institute and State University, USA  
**J M C Frechet**, Cornell University, USA  
**H K Hall**, University of Arizona, USA  
**V Percec**, Case Western Reserve University, USA  
**D Y Sogah**, Du Pont, USA  
**R Asami**, Nagoya Institute of Technology, JAPAN  
**G B Butler**, University of Florida, USA  
**R E Cais**, A T & T Bell Laboratories, USA  
**W M Culbertson**, Ashland Chemical Company, USA  
**B Gordon**, Pennsylvania State University, USA  
**H J Harwood**, University of Akron, USA  
**D A Tirrell**, University of Massachusetts, USA  
**J F Rabek**, Royal Institute of Technology, SWEDEN  
1988 **R Duncan**, University of Keele, UK  
**D Williams**, University of Liverpool, UK  
**E Tomlinson**, CIBA Geigy, UK  
**Lord F S Dainton**, Oxford, UK  
**W Schnabel**, HahnMeitner Institute, GERMANY  
**V T Stannett**, North Carolina State University, USA  
1989 **J E Cardotte**, Film Tec Corporation, USA  
**I M Ward**, University of Leeds, UK  
**I Mita**, RCAST, University of Tokyo, JAPAN  
**A F Johnson**, University of Bradford, UK

- D C Sundberg**, University of New Hampshire, UK  
**C Y Kim**, Advanced Institute of Science and Technology, KOREA  
**S A McDonald**, IBM Almaden Research Centre, USA  
**B Tighe**, Aston University, UK
- 1990 **H R Brown**, IBM, USA  
**M Fryd**, Du Pont, USA  
**D O Hummel**, University of Koln, GERMANY  
**M Kamachi**, Osaka University, JAPAN  
**Z Li**, Academia Sinica, CHINA  
**W J MacKnight**, University of Massachusetts, USA  
**W F Maddams**, University of Southampton, UK  
**G Percival**, Rohm and Haas, NZ  
**E Tsuchida**, Waseda University, JAPAN
- 1991 **J M Vergnaud**, University of SaintEtienne, FRANCE  
**A Abe**, Tokyo Institute of Technology, JAPAN  
**J M G Cowie**, HeriotWatt University, UK  
**F Candau**, Institute Charles Sadron, FRANCE  
**A Eisenberg**, McGill University, CANADA  
**W J Feast**, Durham University, UK  
**A D Jenkins**, University of Sussex, UK  
**E T Kana**, National University of Singapore, SINGAPORE  
**H H Kausch**, EPFL, Lausanne, SWITZERLAND  
**C Y Kim**, KIST, KOREA  
**J E McGrath**, Virginia Polytechnic and State University, USA  
**T R Manley**, Newcastleon Tyne, UK  
**E F Oleynik**, USSR Academy of Science, Moscow, USSR  
**E M Pearce**, Polytechnic University of New York, USA  
**R Qian**, Academia Sinica, CHINA  
**R P Quirk**, University of Akron, USA  
**I Soutar**, Lancaster University, UK  
**N Spassky**, University Pierre et Marie Curie, Paris, FRANCE  
**HW Spiess**, Max Planck Institut, Mainz, GERMANY  
**M A Winnik**, University of Toronto, CANADA
- 1992 **D R Williams**, University of Wales, UK  
**K Friedrich**, University of Kaiserslautern, GERMANY  
**H B Feng**, Academia Sinica, CHINA
- 1995 **J Gerlock**, Ford Motor Company, USA  
**J Gillham**, Princeton University, USA  
**C Hoyle**, University of Southern Mississippi, USA  
**T Jacobine**, Loctite Corporation, USA  
**A Ledwith**, Pilkington Technology Centre, UK  
**C C Ho**, University of Malaya Kuala Lumpur, MALAYSIA
- 1996 **M El Aasser**, Lehigh University, USA  
**A German**, Eindhoven University, NETHERLANDS  
**A Holmes**, Cambridge University, UK  
**B O'Shaughnessy**, Columbia University, USA  
**J Scrivens**, ICI, UK  
**A Balazs**, University of Pittsburgh, U.S.A. (with Polymer Blends CRC)  
**M Xanthos**, Stevens Institute of Technology, U.S.A. (with Polymer Blends CRC)
- 1997 **M Sawamoto**, Kyoto University, JAPAN  
**K Friedrich**, Kaiserslautern University, GERMANY  
**D Bassett**, UCAR Emulsion Systems, North Carolina, USA  
**A Halperin**  
**J Vlachopoulos**, McMaster University, Ontario, CANADA  
**J Ebdon**, University of Liverpool, UK

1998

**K Dusek**, Institute of Macromolecular Chemistry, CZECH REPUBLIC  
**W Mormann**, University Siegen, GERMANY  
**T Aida**, The University of Tokyo, JAPAN  
**A C Albertsson**, Royal Institute of Technology, SWEDEN  
**T Asakura**, Tokyo University of Agriculture and Technology, JAPAN  
**D C Bassett**, University of Reading, UK  
**A Bhowmick**, Indian Institute of Technology, INDIA  
**N Billingham**, University of Sussex, UK  
**M Bowden**, R & D Olin Microelectronic Materials, USA  
**C Decker**, Université de Haute Alsace – 3, FRANCE  
**J Economy**, University of Illinois, USA  
**E T Denisov**, Institute of Problems of Physical Chemistry RAS, RUSSIA  
**W J Feast**, University of Durham, UK  
**J M Frechet**, University of California Berkeley, USA  
**T Fukuda**, Kyoto University, JAPAN  
**J L Gardette**, Université Blaise Pascal, FRANCE  
**K Hatada**, Osaka University, JAPAN  
**J N Hay**, University of Birmingham, UK  
**A S Hoffman**, University of Washington, USA  
**K Horie**, University of Tokyo, JAPAN  
**T Kajiyama**, Kyushu University, JAPAN  
**J Karger-Kocsis**, Universität Kaiserslautern, GERMANY  
**A R Khokhlov**, Moscow State University, Moscow, RUSSIA  
**C Y Kim**, Polymer Materials LabKIST, KOREA  
**A J Kinloch**, Imperial College of Science, Technology and Medicine, UK  
**P J Lemstra**, Eindhoven University of Technology, THE NETHERLANDS  
**O Long**, Malaysian Rubber Board, MALAYSIA  
**P A Lovell**, University of Manchester and UMIST, UK  
**A G MacDiarmid**, University of Pennsylvania, USA  
**C Macosko**, University of Minnesota, USA  
**R H Marchessault**, McGill University, CANADA  
**K Matyjaszewski**, Carnegie Mellon University, USA  
**C K Ober**, Cornell University, USA  
**N Ogata**, Sophia University, JAPAN  
**Y Osada**, Hokkaido University, JAPAN  
**J M Piau**, Université de Grenoble, FRANCE  
**M F Refojo**, Harvard Medical School, USA  
**E Rizzardo**, CSIRO Division of Chemicals and Polymers  
**T Russell**, University of Massachusetts, USA  
**S Russo**, Università di Genova, ITALY  
**J Schultz**, University of Mulhouse, FRANCE  
**V P Shibaev**, Moscow State University, RUSSIA  
**H W Siesler**, University of Essen, GERMANY  
**R F T Stepto**, UMIST and University of Manchester, UK  
**S Stupp**, University of Illinois at Urbana-Champaign, USA  
**J Sun**, Changchun Institute of Applied Chemistry, CHINA  
**D N Theodorou**, University of Patras, GREECE  
**Y Tabata**, Japan Atomic Energy Commission, JAPAN  
**S E Webber**, University of Texas, USA  
**R Weiss**, University of Connecticut, USA  
**A Windle**, Cambridge University, UK  
**H Winter**, University of Massachusetts, USA  
**M Wulkow**, Computing in Technology GmbH (CiT), GERMANY  
**W Mormann**, University of Siegen, Siegen, GERMANY  
**G Mitchell**, University of Reading, UK

1999

- D Attwood**, British Aerospace (Operations) Ltd, UK  
**C N Bowman**, University of Colorado, USA  
**P Hodge**, University of Manchester, Manchester, UK  
**F Horii**, Kyoto University, JAPAN  
**M Jianbio**, Nankai University, CHINA  
**D E. Kranbuehl**, University of William & Mary, Virginia, USA  
**M Patel**, IRC in Biomedical Materials, London, UK  
**G Russell**, University of Canterbury, Christchurch, NEW ZEALAND  
**H Sautereau**, Laboratoire des Matériaux Macromoléculaires, Lyon, FRANCE
- 2000 **U Suter**, ETH Zurich, SWITZERLAND  
**D N Theodorou**, University of Patris, GREECE  
**A Windle**, Cambridge University, UK  
**C Holm**, Johannes Gutenberg University, Mainz, GERMANY
- 2001 **A Balazs**, University of Pittsburg, USA  
**J Gerlock**, Ford Motor Company, Dearborn, MI, USA  
**E Malmstrom**, Royal Inzstitute of Technology, SWEDEN  
**R Metheson**, DuPont Performance Coatings, Troy, MI USA  
**A Matsumoto**, Osake City University, JAPAN  
**W van Ooij**, University of Cincinnati, USA  
**H Pasch**, Deutsches Kunststoff-Institut, Darmstadt, GERMANY  
**S Pascual**, Warwick University, UK  
**M Reading**, Loughborough University, UK  
**S Rimmer**, Sheffield University, UK  
**S Sheiko**, University of North Carolina, USA  
**R Ruchards**, Durham University, UK
- 2002 **M Buback**, University of Gottingen, GERMANY  
**S Brocchini**, London University, UK  
**D Haddleton**, Warwick University, UK  
**C McCormick**, University of Southern Mississippi, USA  
**S Holdcroft**, Simon Fraser University, CANADA  
**C Hawker**, IBM, USA  
**M Monteiro**, Eindhoven University of Technology, NETHERLANDS  
**C Bowman**, University of Colorado, USA
- 2003 **K Anseth**, University of Colorado, USA  
**C Bowman**, University of Colorado, USA  
**J M Fréchet**, University of California, USA  
**JF Gerard**, INSA-Lyon, FRANCE  
**C Hawker**, IBM, USA  
**J-I Jin**, Korea University, KOREA  
**J Lacoste**, Universite Blaise Pascal, Aubiere, FRANCE  
**M Mackay**, Michigan State University, USA  
**T McNally**, Queens University, Belfast, UK  
**R Segal**, University of Minnesota, USA  
**U Schelre**, Institute fur Polymforschung, Dresden, GERMANY  
**M Terano**, Japan Advanced Institute for Science and Technology, JAPAN  
**K Wooley**, Washington University, St. Louis, USA
- 2004 **J Hedrick**, IBM, Almaden Research Center, USA  
**A Hoffman**, University of Washington, USA  
**J Mark**, University of Cincinnati, USA  
**V Percec**, University of Pennsylvania, USA  
**L Averous**, University Louis Pascal, Strasburg, FRANCE  
**W Conley**, Sematech, USA  
**D Graiver**, Michigan State University, USA  
**F Groehn**, Max Planck Institute for Polymer Research, GERMANY  
**A Adam Hitchcock**, McMaster University, CANADA

- M Owen**, MMI / Dow, USA  
**V Schadler**, BASF-ISIS Team, Strasbourg, FRANCE  
**M Textor**, ETH Zurich, SWITZERLAND  
**D Tomasko**, Ohio State University, USA  
**Y-Y Yang**, University of Singapore, SINGAPORE  
2006 **P Callaghan**, Victoria University of Wellington, NEW ZEALAND  
**B Charleux**, Université Pierre et Marie Curie, FRANCE  
**D Haddleton**, University of Warwick, UK  
**R Laine**, University of Michigan, USA  
**G Parker**, Wayne State University, USA  
**K Woodhouse**, University of Toronto, CANADA  
**C Bowman**, University of Colorado, USA  
**Prof Mike Coleman**, Pennsylvania State University, USA  
**S Fakirov**, University of Auckland, New Zealand, and University of Sofia, BULGARIA  
**R French**, DuPont Co., USA  
**T Fukuda**, Kyoto University, JAPAN  
**E Harth**, Vanderbilt University, USA  
**K. Anseth**, University of Colorado, USA  
**D. Hutmacher**, National University of Singapore, SINGAPORE  
**I. Lacik**, Slovak Academy of Sciences, SLOVAKIA  
**J van Hest**, University of Nijmegen, NETHERLANDS  
**J Barralet**, McGill University, CANADA  
2007 **AH E Müller**, University of Bayreuth, GERMANY  
**CJ Hawker**, University of California Santa Barbara, USA  
**A Ryan**, University of Sheffield, UK  
**K Matyjaszewski**, Carnegie Mellon University, USA  
**E Malmström**, Royal Technical Institute Stockholm, SWEDEN  
**HD Maynard**, University of California Los Angeles, USA  
**J Torkelson**, Northwestern University, USA  
**P Vana**, University of Göttingen, GERMANY  
**M Buback**, University of Göttingen, GERMANY  
**D Taton**, University of Bordeaux, FRANCE  
**A Buleon**, National Inst. of Agronomic Research, FRANCE  
**J Wendorff**, University of Marburg, GERMANY  
**S Perrier**, University of Leeds, UK  
**G Russell**, University of Canterbury, NEW ZEALAND  
**R Waymouth**, Stanford University, USA  
2008 **V Abetz**, Max-Planck-Strasse, GERMANY  
**A Balazs**, University of Pittsburgh, USA  
**C Beatty**, University of Florida, USA  
**C Bowman**, University of Colorado, USA  
**P Coates**, University of Bradford, UK  
**J Dorgan**, Colorado School of Mines, USA  
**K Haupt**, Compiègne University of Technology, FRANCE  
**W Hayes**, The University of Reading, UK  
**S Howdle**, University of Nottingham, UK  
**M Leeson**, IMEC Center, BELGIUM  
**K Loos**, University of Groningen, NETHERLANDS  
**T McNally**, Queens University, UK  
**T Nagamura**, Kyushu University, JAPAN  
**B Ratner**, University of Washington, USA  
**J Runt**, Penn State University, USA  
**H. W. Spiess**, Max-Planck-Strasse, GERMANY  
**M Stamm**, Leibniz-Institut fuer Polymerforschung Dresden, GERMANY  
2009 **JM Fréchet**, University of California, USA



- T Ikeda**, Tokyo Institute of Technology, JAPAN  
**S Chul Kim**, National Academy of Engineering, KOREA  
**J Ying**, Institute of Bioengineering and Nanotechnology, SINGAPORE  
**M Sawamoto**, Kyoto University, JAPAN  
 and **120 invited keynote speakers**
- 2011 **T Aida**, University of Tokyo, Japan  
**D Castner**, University of Washington, USA  
**R Kaner**, University of California, USA  
**K Landfester**, Max-Planck-Institute Germany  
**C Ober**, Cornell University, USA  
**T Russell**, University of Massachusetts, USA  
 and **18 invited keynote speakers**
- 2012 **H Klok**, Ecole Polytechnique Fédérale de Lausanne (EPFL), Switzerland  
**H Maynard**, University of California, USA  
**G Wallace**, University of Wollongong, Australia  
**D Haddleton**, University of Warwick, England  
**L Leibler**, ESPCI, France  
**X Zhang**, Tsinghua University, China  
**D Solomon**, University of Melbourne, Australia  
**E Rizzardo**, CSIRO, Australia  
 and **20 invited keynote speakers**
- 2013 **C Alexander**, University of Nottingham  
**K Anseth**, University of Colorado  
**A Balazs**, University of Pittsburgh  
**M Forsyth**, Deakin University  
**A Muller**, Universität Bayreuth  
**M Sawamoto**, Kyoto University  
 and **20 invited keynote speakers**
- 2013 **H Klok**, Ecole Polytechnique Fédérale de Lausanne (EPFL), Switzerland  
**H Maynard**, University of California, USA  
**G Wallace**, University of Wollongong, Australia  
**D Haddleton**, University of Warwick, England  
**L Leibler**, ESPCI, France  
**Xi Zhang**, Tsinghua University, China  
**D Solomon**, University of Melbourne  
**E Rizzardo**, CSIRO  
 and **20 invited keynote speakers**
- 2014 **C Alexander**, University of Nottingham  
**K Anseth**, University of Colorado  
**A Balazs**, University of Pittsburgh  
**M Forsyth**, Deakin University  
**A Muller**, Universität Bayreuth  
**M Sawamoto**, Kyoto University  
 and **20 invited keynote speakers**
- 2015 **M Winnick**, University of Toronto  
**Y Yu**, Fudan University  
**B Stokke**, The Norwegian University of Science and Technology  
**B Hsiao**, Stony Brook University  
**W J Blau**, Trinity College Dublin  
**M Adersson**, University of South Australia  
 and **25 keynote speakers**



# RACI Polymer Division Awards

## BATTEARD-JORDAN Australian Polymer Medallists

(<http://www.polymer.org.au/index.php/home/awards/batteard-jordan-medal>)

|      |                                                                                                                          |
|------|--------------------------------------------------------------------------------------------------------------------------|
| 1974 | <b>Prof D O Jordan</b> , University of Adelaide<br><b>Dr J A J Battaerd</b> , ICI Australia, Central Research Laboratory |
| 1978 | <b>Dr D H Solomon</b> , CSIRO, Division of Applied Organic Chemistry                                                     |
| 1982 | <b>Prof J H O'Donnell</b> , University of Queensland                                                                     |
| 1987 | <b>Prof D H Napper</b> , University of Sydney                                                                            |
| 1992 | <b>Dr E Rizzardo</b> , CSIRO, Chemicals and Polymers Division                                                            |
| 1995 | <b>Prof R G Gilbert</b> , University of Sydney                                                                           |
| 2000 | <b>Prof K Ghiggino</b> , University of Melbourne                                                                         |
| 2006 | <b>Prof G George</b> , Technical University of Queensland                                                                |
| 2012 | <b>Dr G Moad</b> , CSIRO                                                                                                 |

## David Sangster Polymer Science and Technology Achievement Award (formerly Australian Polymer Science and Technology Achievement Award)

(<http://www.polymer.org.au/index.php/home/awards/sangster-award>)

|      |                                                            |
|------|------------------------------------------------------------|
| 1992 | <b>Prof G Wallace</b> , University of Wollongong           |
| 1995 | <b>Dr G Meijs</b> , CSIRO Division of Chemicals & Polymers |
| 1997 | <b>Dr G Spinks</b> , University of Wollongong              |
| 2000 | <b>Dr A Whittaker</b> , University of Queensland           |
| 2006 | <b>Prof F Caruso</b> , University of Melbourne             |
| 2008 | <b>A/Prof M Stenzel</b> , University of New South Wales    |
| 2009 | <b>A/Prof S Perrier</b> , University of Sydney             |
| 2011 | <b>Prof M. Coote</b> , Australian National University      |
| 2015 | <b>A/Professor K. Thurecht</b> , University of Queensland  |

## TRELOAR Prize Recipients

(<http://www.polymer.org.au/index.php/home/awards/treloar-prize>)

The Treloar Prize was established in 1980 in recognition of the contributions to polymer research and teaching by the late Dr F E (Ted) Treloar. The Prize is awarded for the best scientific presentation(s) at an Australasian Polymer Symposium by a person less than 30 years of age; usually there are separate awards for best oral presentation and best poster presentation.

|      |            |                                                                                                                                                                                                                              |
|------|------------|------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|
| 1981 | 12 APS     | <b>G Lichti</b> , University of Sydney                                                                                                                                                                                       |
| 1982 | 13 APS     | <b>M Cuthbertson</b> , CSIRO, Div. of Applied Organic Chemistry; jointly<br><b>P O'Sullivan</b> , University of Queensland<br><b>P J Feeney</b> , University of Sydney; jointly<br><b>M R Hatswell</b> , Albright and Wilson |
| 1984 | 14 APS     | <b>A Whittaker</b> , University of Queensland                                                                                                                                                                                |
| 1986 | 15 APS     | <b>M E Adams</b> , University of Sydney                                                                                                                                                                                      |
| 1987 | 16APS      | <b>A P Lang</b> , University of Queensland                                                                                                                                                                                   |
| 1987 | 8NC        | <b>G M Spinks</b> , University of Melbourne                                                                                                                                                                                  |
| 1989 | 17APS      | <b>H A Willis</b> , University of Queensland                                                                                                                                                                                 |
| 1990 | 18APS      | <b>B R Morrison</b> , University of Sydney<br><b>C Hawker</b> , University of Queensland<br><b>P Pascal</b> , University of Sydney                                                                                           |
| 1991 | Polymer 91 | <b>H A Willis</b> , University of Queensland<br><b>M Monteiro</b> , University of Queensland                                                                                                                                 |
| 1992 | 19APS      | <b>B Fox</b> , CSIRO Chemicals and Polymers<br><b>J Forsythe</b> , University of Queensland                                                                                                                                  |
| 1992 | 9 NC       |                                                                                                                                                                                                                              |
| 1993 | PPC 3      |                                                                                                                                                                                                                              |

|      |             |                                                                                                                                                                                  |
|------|-------------|----------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|
| 1995 | 20APS       | <b>R Evans</b> , CSIRO Chemicals and Polymers                                                                                                                                    |
| 1996 | 21APS       | <b>P Dokolas</b> , University of Melbourne<br><b>A Bishop</b> , Monash University<br><b>C Fellows</b> , James Cook University<br><b>M Zammit</b> , University of New South Wales |
| 1997 | 22APS       | <b>E Furness</b> , University of New South Wales<br><b>D Kukulj</b> , University of New South Wales                                                                              |
| 1998 | Macro98     | <b>H Heuts</b> , University of New South Wales<br><b>P Ghi</b> , University of Queensland                                                                                        |
| 1999 | 23APS       | <b>L Yee</b> , University of New South Wales<br><b>N Grigg</b> , Queensland University of Technology                                                                             |
| 2001 | 24APS       | <b>S Harrison</b> , University of New South Wales<br><b>J Hutchinson</b> , University of Melbourne                                                                               |
| 2002 | 25APS       | <b>L Stanton</b> , University of New South Wales<br><b>T Dargaville</b> , University of Queensland                                                                               |
| 2003 | 26APS       | <b>A Ah Toy</b> , University of New South Wales<br><b>B Muir</b> , Monash University<br><b>A Botella</b> , INSA, Lyon                                                            |
| 2004 | 27APS       | <b>J Quinn</b> , University of Melbourne<br><b>G Such</b> , University of New South Wales/C.S.I.R.O.<br><b>R Joso</b> , University of New South Wales                            |
| 2006 | 28APS       | <b>E Prime</b> , University of Melbourne<br><b>G Smith</b> , University of Canterbury<br><b>K Wong</b> , University of New South Wales                                           |
| 2007 | 29APS       | <b>J Wiltshire</b> , University of Melbourne<br><b>L Connal</b> , University of Melbourne                                                                                        |
| 2008 | 30APS       | <b>T. K. Goh</b> , University of Melbourne<br><b>D. Konkolowicz</b> , University of Sydney<br><b>G. Johnston-Hall</b> , University of Queensland                                 |
| 2009 | 31APS/PPC11 | <b>J Kulis</b> , University of Queensland<br><b>R Chapman</b> , University of Sydney                                                                                             |
| 2011 | 32APS       | <b>John Moares</b> , The University of Sydney<br><b>Vien T. Huynh</b> , The University of New South Wales                                                                        |
| 2012 | 33APS       | <b>Zhou Zhang</b> , University of Melbourne<br><b>Adrian Sulistio</b> , University of Melbourne<br><b>Wei Zhao</b> , The University of Sydney                                    |
| 2013 | 34APS       | <b>Shu Jie Lam</b> , University of Melbourne<br><b>Steven Harris-Wibowo</b> , University of Melbourne                                                                            |
| 2015 | 35APS       | <b>Lewis Chambers</b> , The University of Queensland<br><b>Molly Rowe</b> , University of New South Wales                                                                        |

## O'Donnell Young Scientist Award

(<http://www.polymer.org.au/index.php/home/awards/odonnell-prize>)

|      |                       |                                                |
|------|-----------------------|------------------------------------------------|
| 1996 | <b>J Campbell</b>     | Monash University                              |
| 1997 | <b>E Coen</b>         | University of Sydney                           |
| 1998 | <b>K Lenghaus</b>     | University of Melbourne                        |
| 1999 | <b>B Haupt</b>        | Australian National University                 |
| 2000 | <b>S Prescott</b>     | University of Sydney / CSIRO Molecular Science |
| 2004 | <b>L Connal</b>       | University of Melbourne                        |
| 2007 | <b>T Schiller</b>     | Queensland University of Technology            |
| 2008 | <b>J Hodgson</b>      | Australian National University                 |
| 2011 | <b>John Moraes</b>    | University of Sydney                           |
| 2014 | <b>Natasha Ishank</b> | Sydney University                              |

## Polymer Citation Recipients

(<http://www.polymer.org.au/index.php/home/awards/polymer-citations>)

The Polymer Division Citations were awarded for the first time at 16 APS. Up to three Citations may be presented in any year at a national meeting of the Polymer Division. They are awarded for significant contributions and service in any area of polymer science and/or technology.

|      |            |                                                                                                                                                                                                                           |
|------|------------|---------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|
| 1986 | 16 APS     | <b>G B Guise</b> , CSIRO, Division of Textile Industry<br><b>H J Ruddell</b> , Telecom Australia<br><b>D F Sangster</b> , CSIRO, Division of Chemical Physics                                                             |
| 1987 | 8 NC       | <b>D Oldfield</b> , DSTO, Materials Research Laboratory<br><b>I C Watt</b> , CSIRO, Division of Textile Physics                                                                                                           |
| 1989 | 17 APS     | <b>D R G Williams</b> , University of Adelaide<br><b>C E M Morris</b> , DSTO, Materials Research Laboratory                                                                                                               |
| 1990 | 18 APS     | <b>B A Bolto</b> , CSIRO, Division of Chemicals and Polymers<br><b>R A Shanks</b> , RMIT                                                                                                                                  |
| 1991 | Polymer 91 | <b>D J T Hill</b> , University of Queensland<br><b>P E M Allen</b> , University of Adelaide                                                                                                                               |
| 1992 | 19APS      | <b>E Rizzardo</b> , CSIRO, Division of Chemicals and Polymers<br><b>P Pomery</b> , University of Queensland                                                                                                               |
| 1992 | 9NC R      | <b>P Burford</b> , University of New South Wales<br><b>W D Cook</b> , Commonwealth Department of Health                                                                                                                   |
| 1993 | PPC3       | <b>Z H Stachurski</b> , Australian National University<br><b>T Chirila</b> , Lions Eye Institute                                                                                                                          |
| 1995 | 20APS      | <b>K Busfield</b> , Griffith University<br><b>G Moad</b> , CSIRO Chemicals and Polymers<br><b>G M Ferguson</b> , SCM Chemicals                                                                                            |
| 1996 | 21APS      | <b>J G Matisons</b> , University of South Australia                                                                                                                                                                       |
| 1997 | 22APS      | <b>G A George</b> , Queensland University of Technology<br>Special Citation: <b>B Guise</b> , CSIRO                                                                                                                       |
| 1998 | Macro98    | Special Citation: <b>D Hill</b> , University of Queensland<br>Special Citation: <b>P Pomery</b> , University of Queensland<br>Special Citation: <b>D Oldfield</b> , DSTO<br><b>A Whittaker</b> , University of Queensland |
| 1999 | 23APS      | <b>N Edmonds</b> , University of Auckland<br><b>H Toh</b> , Sola Optics                                                                                                                                                   |
| 2001 | 24APS      | <b>G P Simon</b> , Monash University<br><b>I Dagley</b> , CRC Polymers                                                                                                                                                    |
| 2002 | 25APS      | <b>J Hodgkin</b> , CSIRO Molecular Science<br><b>E Senogles</b> , James Cook University                                                                                                                                   |
| 2003 | 26APS      | <b>T Davis</b> , University of New South Wales<br><b>G Russell</b> , University of Canterbury                                                                                                                             |
| 2004 | 27APS      | <b>J P A Heuts</b> , University of New South Wales/University of Murcia<br><b>H Brown</b> , University of Wollongong<br>Special Citation: <b>D F Sangster</b> , University of Sydney                                      |
| 2006 | 28APS      | <b>B Hawkett</b> , University of Sydney<br><b>D Lewis</b> , Carl Zeiss Optical                                                                                                                                            |
| 2007 | 29APS      | <b>C M Fellows</b> , University of New England                                                                                                                                                                            |
| 2008 | 30APS      | <b>C Barner-Kowollik</b> ,<br>University of New South Wales/Karlsruhe Institute of Technology                                                                                                                             |
| 2012 | 33APS      | <b>G. Spinks</b> , University of Wollongong                                                                                                                                                                               |
|      | 33APS      | <b>Prof G Qiao</b> , University of Melbourne                                                                                                                                                                              |
| 2013 | 34APS      | <b>A/Prof M Stenzel</b> , University of New South Wales                                                                                                                                                                   |
|      | 34APS      | <b>Prof S. Perrier</b> , University of Sydney                                                                                                                                                                             |
| 2015 | 35APS      | <b>A/Prof K Jack</b> , The University of Queensland                                                                                                                                                                       |

## CRC for Polymers Prize

(<http://www.polymer.org.au/index.php/home/awards/crc-prize>)

- 1994 **M Singer**, Monash University
- 1995 **A Mayr**, Monash University
- 1996 **M Griffiths**, Sydney University
- 1997 **J McNicol**, Australian National University
- 1998 **B Muir**, Monash University
- 1999 **E Dymke**, University of Wollongong
- 2000 **J Castro**, University of Sydney
- 2001 **S Angus**, University of New South Wales
- 2002 **K George**, Queensland University of Technology
- 2003 **H Chaffey-Miller**, University of New South Wales
- 2004 **K Varcoe**, University of Queensland
- 2006 **J Hodgson**, Australian National University
- 2007 **A J Inglis**, University of New South Wales  
**K Tsang**, Monash University
- 2008 **J van Hensbergen**, University of New South Wales  
**J M Ren**, University of Melbourne
- 2009 **J Tom**, University of Sydney  
**A Widjaya**, University of Melbourne
- 2010 **S Passmore**, University of Queensland 2010  
**R Brooke**, Flinders University
- 2011 **S Harris Wibowo**, University of Melbourne  
**A Pearce**, University of Queensland
- 2012 **S J Lam**, University of Melbourne  
**Y Tang**, Monash University
- 2013 **Adam Sutton**, The University of Western Sydney
- 2014 **Lewis Chambers**
- 2015 **Leo De Waal**, The University of Queensland

## Solomon Lecturers

- 2000 **Prof David Solomon**, University of Melbourne
- 2001 **Prof Julia Higgins**, DBE, Imperial College, UK
- 2005 **Prof Gerhard Wegner**, MPI für Polymerforschung, Germany
- 2006 **Prof David Tirrell**, California Institute of Technology, USA
- 2007 **Prof Mitsuo Sawamoto**, Kyoto University
- 2012 **Prof Krzysztof Matyjaszewski**, Carnegie Mellon

## Bruce Guise Award

- 2011 **H Toh**, Carl Zeiss
- 2013 **Chris Such**, Dulux (Orica)

## The International Award for Promotion of Australasian Polymer Science and Technology

- 2011 **C Bowman**, University of Colorado

# Speakers at 36APS

## PLENARY SPEAKERS AT 36APS

**Professor Takuzo Aida** – The University of Tokyo  
**Professor Frank Caruso** – The University of Melbourne  
**Professor Craig Hawker** – University of California Santa Barbara  
**Professor Heather Maynard** – University of California, Los Angeles  
**Professor Alan Rowan** – Australian Institute of Bioengineering & Nanotechnology  
**Professor Richard Spontak** – North Carolina State University

## KEYNOTE SPEAKERS AT 36APS

**Professor Kristi Anseth** – University of Colorado Boulder  
**Professor Christopher Barner-Kowollik** – Karlsruhe Institute of Technology  
**Dr Remzi Becer** – Queen Mary University of London  
**Prof Christopher N. Bowman** – University of Colorado  
**Dr Ian Dagley** – CRC for Polymers  
**Professor Tom Davis** – Monash University  
**Dr Guillaume Delaittre** – Karlsruhe Institute of Technology  
**Professor Amanda Ellis** – Flinders University  
**Professor Laurent Fontaine** – University of Maine  
**Professor Bronwyn Fox** – Swinburne University of Technology  
**Associate Professor Lisbeth Grondahl** – The University of Queensland  
**Professor Olli Ikkala** – Aalto University  
**Dr Angus Johnston** – Monash Institute of Pharmaceutical Sciences  
**Professor Masami Kamigaito** – Nagoya University  
**Professor Tong Lin** – Deakin University  
**Associate Professor Hideto Minami** – Kobe University  
**Professor Adrian Mouritz** – RMIT University  
**Professor David Officer** – University of Wollongong  
**Professor Costas Patrickios** – University of Cyprus  
**Assistant Professor Emily Pentzer** – Case Western University  
**Professor Sebastian Perrier** – The University of Warwick  
**Dr Julien Poly** – Haute-Alsace University  
**Professor Marie-Françoise Reyniers** – Gent University  
**Professor Thomas Spurling** – Swinburne University of Technology  
**Dr Andreas Walther** – Albert-Ludwigs-University of Freiburg  
**Dr Roger Wepf** – The University of Queensland  
**Professor Andrew Whittaker** – The University of Queensland  
**Dr Wallace Wong** – The University of Melbourne

## ECR SPEAKERS AT 36APS

**Associate Professor Kathryn Fairfull-Smith** – Queensland University of Technology  
**Assistant Professor Brett Fors** – Cornell University  
**Dr Daniel Heath** – The University of Melbourne  
**Dr Kristian Kempe** – Monash University  
**Dr Markus Muellner** – The University of Sydney  
**Dr George Vamvounis** – James Cook University  
**Dr Paul Wilson** – University of Warwick

# General Program Structure

## Sunday 20 November 2016

|             |                                                      |
|-------------|------------------------------------------------------|
| 1400 - 1800 | Registration Open                                    |
| 1600 - 1800 | Conference Opening and Plenary Session               |
| 1800 - 2000 | Welcome Reception at the Lorne Surf Life Saving Club |

## Monday 21 November 2016

|             |                                             |
|-------------|---------------------------------------------|
| 0800 - 1745 | Registration Desk Open                      |
| 0900 - 1000 | Plenary Session                             |
| 1000 - 1030 | Morning Refreshments                        |
| 1030 - 1245 | Concurrent Sessions                         |
| 1245 - 1345 | Lunch                                       |
| 1345 - 1515 | Concurrent Sessions                         |
| 1515 - 1545 | Afternoon Refreshments                      |
| 1545 - 1745 | Concurrent Sessions                         |
| 1800 - 1900 | Symposium Poster Session                    |
| 1930 - 2130 | VIP Cocktail Event at Lorne Beach Pavillion |

## Tuesday 22 November 2016

|             |                                  |
|-------------|----------------------------------|
| 0800 - 1800 | Registration Desk Open           |
| 0900 - 1000 | Plenary Session                  |
| 1000 - 1030 | Morning Refreshments             |
| 1030 - 1300 | Concurrent Sessions              |
| 1300 - 1400 | Lunch                            |
| 1400 - 1530 | Concurrent Sessions              |
| 1530 - 1600 | Afternoon Refreshments           |
| 1600 - 1800 | Concurrent Sessions              |
| 1800 - 1900 | Annual General Meeting           |
| 1930 - 2030 | Student Night at the Lorne Hotel |

## Wednesday 23 November 2016

|             |                                  |
|-------------|----------------------------------|
| 0800 - 1630 | Registration Desk Open           |
| 0900 - 1000 | Plenary Session                  |
| 1000 - 1030 | Morning Refreshments             |
| 1030 - 1300 | Concurrent Sessions              |
| 1300 - 1400 | Lunch                            |
| 1400 - 1530 | Concurrent Sessions              |
| 1530 - 1600 | Afternoon Refreshments           |
| 1600 - 1700 | Plenary Session                  |
| 1900 - 2300 | Symposium Dinner at Mantra Lorne |



# Symposium Information

## Information for Presenters and Session Chairs

All Speakers will be asked to report to the Speakers Preparation Room to load their presentations onto the symposium laptop. This must be done AT LEAST four hours before you are due to present – this may mean the day before your presentation. An audio visual technician will be available throughout the symposium. Please see the staff at the Registration Desk for further assistance or directions.

Speakers are asked to introduce themselves to their session Chair during the break – at least ten minutes before the session, if possible, to familiarise themselves with the room and equipment and check that presentations are working.

## Oral Presentations

### Invited Plenary Presentations

Will be allocated 50 minutes presentation time, which includes 5 minutes for questions and answers.

### Invited Keynote Presentations

Will be allocated 30 minutes presentation time, which includes 5 minutes for questions and answers.

### Submitted Oral Presentations

Will be allocated 15 minutes presentation time which includes 3 minutes for questions and answers to be held at the conclusion of each presentation.

To aid in the smooth running of the sessions, a bell will ring once – indicating the time for questions is approaching. At the second ring you should stop for questions.

## Total Presentation Time      Warning Bell Second Bell – Question Time

|            |            |            |
|------------|------------|------------|
| 15 Minutes | 10 Minutes | 12 Minutes |
| 30 Minutes | 20 Minutes | 25 Minutes |
| 50 Minutes | 40 Minutes | 45 Minutes |

In order that the concurrent sessions remain aligned, all sessions must run according to the program. Speakers who run overtime will be cut off by the session chair. It is the responsibility of the session chairs to ensure that all speakers remain within their allocated presentation time.

## Poster Presentations

Posters will be displayed in the Heritage Ballroom. There will be a poster session on Monday 21 November 2016 from 1800-1900. At least one of the poster authors is expected to present adjacent to their posters to answer questions. Treloar Prize Applicants must be present in person to answer questions during the poster session.



Username: MANTRA LORNE  
Password: 36APS16

## Download the App

To download the mobile app simply follow the URL or scan the QR code

<https://leishman.eventsair.com/attendeeapp/36aps/mobile-app/>



# Social Program

## WELCOME RECEPTION

Date: 20 November 2016  
Time: 1800 - 2000  
Venue: Lorne Surf Life Saving Club

Take a short stroll along the beach or esplanade to the welcome reception at Lorne Surf Life Saving Club. The evening will be a relaxed and enjoyable one, a great opportunity to make new connections and reconnect with old friends.

## POSTER SESSION

Date: 21 November 2016  
Time: 1800 - 1900  
Venue: Heritage Ballroom, Mantra Lorne

Poster authors will be available for questions and answers during this session and beverages will be served.

## VIP COCKTAIL EVENT

Date: 21 November 2016  
Time: 1930 - 2130  
Venue: Lorne Beach Pavilion

This is an invitation only event



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## STUDENT NIGHT

Date: 22 November 2016  
Time: 1930 onwards

All students are welcome to the Lorne Hotel for beverages and a light meal. There will be a bar tab and once that has been exhausted there will be drinks available for purchase at your own cost. Please note students must make their own way to the venue and bring their name badges to receive entry.

## SYMPOSIUM DINNER

Date: 23 November 2016  
Time: 1900 - 2300

Surf's Up! Join your friends and colleagues for the highlight of the APS social program. Pre-dinner refreshments will be held on the lawns of Mantra Lorne and will be followed by a night of food and entertainment.

**HITACHI**  
Inspire the Next



**NewSpec**

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# General Information

## Registration Desk Opening Times:

|                       |             |
|-----------------------|-------------|
| Sunday 20 November    | 1400 - 1800 |
| Monday 21 November    | 0800 - 1745 |
| Tuesday 22 November   | 0800 - 1800 |
| Wednesday 23 November | 0800 - 1630 |

**Accommodation:** If you have any queries relating to your accommodation booking, first please see the staff at your hotel. Your one night deposit has been transferred to the hotel you have selected – please confirm this on check in with your hotel. If you arrive 24 hours later than your indicated arrival day you may find that you have forfeited your deposit.

**Symposium Handbook & Name Badge:** At the symposium you will receive a hard copy pocket program. Each delegate will also receive a name badge upon arrival to the Symposium. The name badge will be your official pass and must be worn to obtain entry to all sessions and to social functions.

**Social Program:** The welcome reception, poster session and symposium dinner are included in the cost of each full symposium registration. Social events ARE NOT included in the cost of day registrations or for accompanying partners. Places for day registrants and additional guests for these events may still be available at an additional cost. Bookings can be made at the symposium registration desk subject to availability.

**Conference Dinner Entry:** All delegates who are registered to attend the symposium dinner will receive a named sticker at registration. You MUST place your sticker on a table located on poster boards at the registration desk.

**Entry to Conference Sessions:** It is suggested that delegates arrive at preferred sessions promptly to ensure a seat. If sessions become full then delegates will not be allowed entry.

**Mobile Phones:** As a courtesy to other delegates, please ensure that all mobile phones and pagers are turned off or in a silent mode during all sessions and social functions.

## Photographs, videos, recording of sessions:

Delegates are not permitted to use any type of camera or recording device at any of the sessions unless written permission has been obtained from the relevant speaker.

**Smoking:** All venues are non smoking - no rooms are designated smoking rooms however guests are allowed to smoke outside the venue.

**Speakers:** Speakers will be asked to bring their presentations with them on a USB, then load their presentations in the speakers preparation room. This must be done AT LEAST four hours before you are due to present – this may mean the day before your presentation. Please see the staff at the registration desk for directions.

**Special Diets:** All catering venues have been advised of any special diet preferences you have indicated on your registration form. Please identify yourself to venue staff as they come to serve you and they will be pleased to provide you with all pre-ordered food. For day catering, there may be a specific area where special food is brought out, please check with catering or Symposium staff.

**Prayer Room:** A prayer room will be available for the duration of the symposium. Please see staff at the registration desk for directions.

**Disclaimer:** The 36APS reserves the right to amend or alter any advertised details relating to dates, program and speakers if necessary, without notice, as a result of circumstances beyond their control. All attempts have been made to keep any changes to an absolute minimum.

# Program – Sunday 20 November 2016

Plenary Session

Chair: Idriss Blakey

Convention 1 & 2

1600 - 1610 **Welcome to the 36th Australasian Polymer Symposium**

*Greg Qiao - The University of Melbourne*

1610 - 1700 **PL1** Accurate control of polymer structure and function

*Craig Hawker - University of California, Santa Barbara*

**Dulux**

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1700 - 1750 **PL2** A new age of functional and responsive thermoplastic elastomers: discovering a buried treasure

*Richard Spontak - North Carolina State University*

1750 - 1755 **IUPAC Update**

*Chris Fellows - The University of New England*

1755 - 1800 **Closing Remarks**

1800 - 2000 **Welcome Reception**

Take a short stroll along the beach or esplanade to the Lorne Surf Life Saving Club

*Lorne Surf Life Saving Club*

# Program - Monday 21 November 2016

## Plenary Session

Chair: Amanda Ellis

Convention 1 & 2

0830 - 1600 Trade Exhibition Open  
Barista coffee served



0900 - 0910 Welcome and Housekeeping

0910 - 1000 **PL3** Biomimetic polymers for protein stabilization: from design to synthesis to applications in food and medicine  
Heather Maynard - University of California, Los Angeles

1000 - 1030 Morning Refreshments & Trade Exhibition Convention Foyer

Visit the exhibitors to have your passport stamped and go into the draw to win some great prizes

Polymers for Electronics, Energy, Sensors and Environmental Applications  
Chair: Simon Moulton  
Convention 1 & 2

Polymeric Nanomedicine, Therapeutics and Imaging Agents  
Chair: Georgina Such  
Convention 3



Polymers in Industry and Translational Research  
Chair: Ramon Tozer  
Heritage Room



Advanced Characterization  
Chair: Hans Griesser  
Heritage 3



1030 - 1045 **M1.1** - Intrinsically self-healing conducting polymers  
Paul Baek

**M2.1** - Combating HIV with RAFT polymers  
Maarten Danial

**M3.1/KN3** - The origin of the polymer banknote  
Tom Spurling

**M4.1** - SAXS studies of self-assembled block copolymer/ionic liquid blends  
Kevin Jack

1045 - 1100 **M1.2** - Advancing the performance of polymer solar cells and photodectors via plasmonic coupling  
Yu Jin Jang

**M2.2** - Synthesis and characterization of an aptamer-targeted hyperbranched polymer for the detection of triple-negative breast cancer  
Nicholas Fletcher

**M4.2** - Monitoring chemical reactions using capillary electrophoresis: some new insights on peptide grafting, anticancer drug binding and starch digestion!  
Patrice Castignolles

1100 - 1115 **M1.3/KN1** - Functionalised polythiophenes for energy, sensing and bio- applications  
David Officer

**M2.3** - Investigations into the role of size for successful cancer nanomedicines  
Anna Gemmell

**M3.2** - Developmental history and future of polymer banknotes in Australia  
Greg Dicinoski

**M4.3/IS1** - Threading or not threading onto polymer chains  
Cecile Dreiss

1115 - 1130

**M2.4** - Protein adsorption on polymeric particles - influence of particle design and biological milieu  
Alessia Clarissa Giuliana Weiss

**M3.3** - Characterisation of industrial adhesives with (solid-state) NMR spectroscopy  
Marion Gaborieau

1130 - 1145 **M1.4** - Surface photovoltage studies of polymer semiconductor films influenced by plasmon-upconversion coupling  
Yu Jin Jang

**M2.5/KN2** - Understanding the secrets of nanoparticle cell interactions  
Angus Johnston

**M3.4** - Five top tips for Australian patent protection  
Grace Chan

**M4.4** - Thin films, deuteration and neutron reflectometry for characterizing cellulose-protein interactions  
Peter Holden

1145 - 1200 **M1.5** - Morphology control in polymer solar cells  
David Lewis

**M3.5** - Functional starch based hydrogels: sustainable material solutions for wastewater and agriculture industries  
Samira Siyamak

**M4.5** - Characterisation of oligoacrylates and their block co-oligomers  
Adam Sutton

1200 - 1215 **M1.6** - Development of organic ionic plastic crystal-polymer nanofibre composites as solid state electrolytes  
Yundong Zhou

**M2.6** - Polyion complex micelles as cancer therapeutic carriers for protein drugs  
Fan Chen

**M3.6** - Polymeric monolithic materials for analytical applications  
Robert Shellie

**M4.6** - Raman Spectroscopy, a powerful tool for polymer analysis  
Taisuke Ota

1215 - 1230 **M1.7** - Designing of electrospun polymer fibers for organic ionic plastic crystal-based solid state electrolytes  
Xiaoen Wang

**M2.7** - Tunable and controlled polymeric drug delivery of NSAIDs  
Felisa Reyes-Ortega

**M3.7** - Sand stabilization along the railway track with polymer as a substitute of crude oil  
Muhammad Umair Shaikat

**M4.7/IS2** - Realtime structure of thin film organic semiconductors during thermally induced diffusion  
Andrew Nelson

1230 - 1245

**M3.8** - Future fibres industry transformation research hub  
Emma Prime

1245 - 1345 Lunch & Trade Exhibition

Convention Foyer



|             |                                                                                                                                           |                                                                                                                                                        |                                                                                                                                                   |                                                                                                                                            |
|-------------|-------------------------------------------------------------------------------------------------------------------------------------------|--------------------------------------------------------------------------------------------------------------------------------------------------------|---------------------------------------------------------------------------------------------------------------------------------------------------|--------------------------------------------------------------------------------------------------------------------------------------------|
| 1345 - 1400 | <b>M1.8</b> - Magnetically-recoverable and Triazole-mPEG Stabilized Transition Metal Nanoparticles for Catalysis<br><i>Pengxiang Zhao</i> | <b>M2.8</b> - Combating multidrug-resistant gram-negative bacteria with structurally nanoengineered star-shaped peptide polymers<br><i>Shu Jie Lam</i> | <b>M3.9/KN6</b> - Translational research in polymer science and the factory of the future<br><i>Bronwyn Fox</i>                                   | <b>M4.8/KN7</b> - Cationic RAFT polymerization: synergetic developments in radical and cationic polymerizations<br><i>Masami Kamigaito</i> |
| 1400 - 1415 | <b>M1.9</b> - Optimising surface functionalisation of carbon fibre to enhance interfacial adhesion<br><i>Kathleen Beggs</i>               | <b>M2.9</b> - Monodispersed polymer encapsulated superparamagnetic iron oxide nanoparticles for cell labeling<br><i>Duc Nguyen</i>                     |                                                                                                                                                   |                                                                                                                                            |
| 1415 - 1430 | <b>M1.10/KN4</b> - Utilizing janus graphene oxide nanosheets for tailored assemblies<br><i>Emily Pentzer</i>                              | <b>M2.10</b> - Understanding the effect of the drug on the performance of drug carriers<br><i>Martina Stenzel</i>                                      | <b>M3.10</b> - Systematic production of high quality and low cost cellulose nanofibres and their potential applications<br><i>Nasim Amiralian</i> | <b>M4.9</b> - Approaches to the RAFT synthesis of multifunctional, multi-armed polymers<br><i>Graeme Moad</i>                              |
| 1430 - 1445 |                                                                                                                                           | <b>M2.11/KN5</b> - Polymer nano- structures for MRI and PET imaging<br><i>Thomas Davis</i>                                                             | <b>M3.11</b> - Spinifex Nanocellulose: the commercialisation story so far<br><i>Phil Nelson</i>                                                   | <b>M4.10</b> - Oxygen tolerant PET-RAFT polymerisation in flow<br><i>Nathaniel Corrigan</i>                                                |
| 1445 - 1500 | <b>M1.11</b> - Specific ion effects on responsive polymer brush coatings<br><i>Erica Wanless</i>                                          |                                                                                                                                                        | <b>M3.12</b> - Patents and your research project: a case study from a polymer-related industry<br><i>Matthew Overett</i>                          | <b>M4.11</b> - Exploring the impact of UV irradiation on traditional RAFT systems<br><i>Oskar Majewski</i>                                 |
| 1500 - 1515 | <b>M1.12</b> - Programmable nucleic acid - polymer conjugates with sensing and signalling capabilities<br><i>Giovanna Sicilia</i>         |                                                                                                                                                        |                                                                                                                                                   | <b>M4.12</b> - Forced gradient co-polymers using continuous flow reactors<br><i>Simon Saubern</i>                                          |
| 1515 - 1545 | Afternoon Refreshments & Trade Exhibition                                                                                                 |                                                                                                                                                        |                                                                                                                                                   | Convention Foyer                                                                                                                           |



Biomolecules, Biofunctional & Responsive Polymers  
Chair: Martina Stenzel  
Convention 1 & 2

Polymeric Nanomedicine, Therapeutics and Imaging Agents  
Chair: Nicholas Fletcher  
Convention 3

Polymers in Industry and Translational Research  
Chair: Bronwyn Fox  
Heritage Room

Plastics and Thermosets  
Chair: Jun Ma  
Heritage 3



|             |                                                                                                                                                                   |                                                                                                                                                                                  |                                                                                                                                                                     |                                                                                                                                                                                               |
|-------------|-------------------------------------------------------------------------------------------------------------------------------------------------------------------|----------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|---------------------------------------------------------------------------------------------------------------------------------------------------------------------|-----------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|
| 1545 - 1600 | <b>M1.13</b> - Substrate independent approach for growing low fouling polymer brushes to inhibit bacterial adhesion<br><i>Yue Shi</i>                             | <b>M2.12</b> - Stable and non-toxic superparamagnetic iron oxide nanoparticles as multimodal (PET/ SPECT-MRI) imaging probes<br><i>Nguyen Pham</i>                               | <b>M3.13/KN9</b> - Industry-driven collaborative polymer research<br><i>Ian Dagley</i>                                                                              | <b>M4.13/KN10</b> - Mechanical energy to electricity conversion of nanofibres and conducting polymers<br><i>Tong Lin</i>                                                                      |
| 1600 - 1615 | <b>M1.14/KN8</b> - Single-Enzyme Nanogels: synthesis and immobilization<br><i>Guillaume Delaittre</i>                                                             | <b>M2.13</b> - Co-delivery system with NO (nitric oxide) and drug (doxorubicin) using nanoparticle to combat doxorubicin-resistant ovarian cancer cells<br><i>Elly Sulhwa Yu</i> |                                                                                                                                                                     |                                                                                                                                                                                               |
| 1615 - 1630 |                                                                                                                                                                   | <b>M2.14</b> - Robust and biodegradable hydrogels for delivery of hydrophobic drugs<br><i>Dunyin Gu</i>                                                                          | <b>M3.14</b> - How to attack a patent/ application<br><i>Richard Grant</i>                                                                                          | <b>M4.14</b> - Structural property and mechanical behavior of Polypropylene –Elemental sulfur (S8) composites: Effect of sulfur loading<br><i>Vijay Kumar</i>                                 |
| 1630 - 1645 | <b>M1.15</b> - 'Click' thiol-yne hydrogels for MSC delivery for the suppression of immunomodulatory responses in osteoarthritic joints<br><i>Richard Fernando</i> | <b>M2.15</b> - Macromolecular donors for controlled and stimulated hydrogen sulfide release<br><i>Francesca Ercole</i>                                                           | <b>M3.15</b> - Why doesn't excellent and well directed research always yield a product in the market place?<br><i>Brian Hawket</i>                                  | <b>M4.15</b> - Graphene/polyacrylic acid composite hydrogels being mechanically robust and pH-sensitive<br><i>Ashrafal Alam</i>                                                               |
| 1645 - 1700 | <b>M1.16</b> - A PEDOT microfibrinous scaffold for neural recordings<br><i>Jason Marroquin Reyes</i>                                                              | <b>M2.16/ECR1</b> - Structured polymer nanoparticles - access to new hybrid materials and nanomedicines<br><i>Markus Muellner</i>                                                |                                                                                                                                                                     | <b>M4.16</b> - A comparison of the material properties of thermosets from itaconic acid based polyesters catalyzed by "classical" catalysis and enzymatic catalysis<br><i>Sara Brannstrom</i> |
| 1700 - 1715 | <b>M1.17</b> - Micropatterned and drug eluting bioelastomers for improved cardiomyocyte beating<br><i>John Forsythe</i>                                           |                                                                                                                                                                                  | <b>M3.16</b> - SAXS study of oxalate salts in the presence of poly (acrylic acid) scale inhibitors<br><i>Christopher Fellows</i>                                    | <b>M4.17/KN11</b> - Multi-scale toughening of polymer composites using carbon nano-particles and carbon macro-pins<br><i>Adrian Mouritz</i>                                                   |
| 1715 - 1730 | <b>M1.18</b> - Kappa-carrageenan-genipin cross-linked chitosan composites for drug release in simulated gastrointestinal fluids<br><i>Kushaal Raj</i>             | <b>M2.17</b> - Enhanced drug delivery system based on Fructose-coated nanoparticles for the treatment of breast cancer<br><i>Mingxia Lu</i>                                      | <b>M3.17</b> - Understanding the fundamentals - investigating the kinetics of a novel class of nN-containing monomers via PLP-SEC<br><i>Katrin Blandine Kockler</i> |                                                                                                                                                                                               |
| 1730 - 1745 | <b>M1.19</b> - Biodegradable antimicrobial polymer containing pendant guanidines<br><i>Jianyong Jin</i>                                                           | <b>M2.18</b> - Magnetic alignment of polymeric nanofibers with embedded sterically stabilized iron oxide nanoparticles for potential cell scaffolds<br><i>Vien Huynh</i>         |                                                                                                                                                                     |                                                                                                                                                                                               |

1800 - 1900 Poster Session

Heritage Ballroom, Mantra Lorne

1930-2130 VIP Cocktail Event  
Lorne Beach Pavilion  
Invitation only event



# Program - Tuesday 22 November 2016

Plenary Session  
Chair: Neil Cameron

Convention 1 & 2

0830 - 1600 Trade Exhibition Open  
Barista coffee served



0900 - 0910 Welcome and Housekeeping

0910 - 1000 **PL4** Building advanced, bioresponsive materials from polymers, macromolecules and metal ions  
*Frank Caruso - The University of Melbourne*

1000 - 1030 Morning Refreshments & Trade Exhibition

Convention Foyer

Visit the exhibitors to have your passport stamped and go into the draw to win some great prizes

Advanced Characterization  
Chair: Marion Gaborieau  
Convention 1 & 2



Polymeric Nanomedicine, Therapeutics and Imaging Agents  
Chair: Markus Muellner  
Convention 3



Polymers for Electronics, Energy, Sensors and Environmental Applications  
Chair: Simon Moulton  
Heritage Room

Biomolecules, Biofunctional & Responsive Polymers  
Chair: John Forsythe & Martina Stenzel  
Heritage 3

1030 - 1045 **T1.1/KN12** - Soft Matter (Cryo) Electron Microscopy and Microanalysis  
*Roger Wepf*

**T2.1** - Intracellular behaviours of theranostic polymers for nanomedicine  
*Joshua Simpson*

**T3.1** - Medium bandgap material for solar cell applications  
*Desta Gedefaw*

**T4.1/KN14** - Photodegradable hydrogels as spatiotemporally controllable cellular niches  
*Kristi Anseth*

1045 - 1100

**T2.2** - Rational design of polymeric nanomaterials for photo-induced molecular imaging and therapeutic function  
*Nicholas Westra Van Holthe*

**T3.2** - Control of active layer architecture in organic photovoltaic devices through block copolymer design  
*Valerie Mitchell*

1100 - 1115 **T1.2** - Universal mass spectrometric analysis of poly(ionic liquid)s  
*Jan Steinkoenig*

**T2.3** - Effect of increased surface hydrophobicity on the clearance of inhaled pegylated polylysine dendrimers  
*Shadabul Haque*

**T3.3/KN13** - Controlled synthesis of semiconducting polymers  
*Wallace Wong*

**T4.2** - Biodegradable and biocompatible glycopolymeric nanoparticles for drug delivery  
*Sylvia Ganda*

1115 - 1130 **T1.3** - Characterization strategies to study the thermal degradation and combustion behaviours of polymeric materials: a critical analysis  
*Paul Joseph*

**T2.4** - Bottle-brush polymer architectures for drug delivery  
*Joaquin Sanchis Martinez*

**T4.3** - Nucleotide functionalised synthetic polymers  
*Michael Wilson*

1130 - 1145 **T1.4** - Chemical characterization of polyelectrolyte complexes: strengths and limitations  
*Lisbeth Grøndahl*

**T2.5** - Oral delivery of folate targeted colorectal cancer therapeutics  
*Dewan Akhter*

**T3.4** - Durable transparent superhydrophobic surfaces via sprayable interpenetrated polymer networks  
*William S. Y. Wong*

**T4.4/KN15** - Static and dynamic bioinspired aqua-materials  
*Andreas Walther*

1145 - 1200 **T1.5** - Database-supported thermal analysis involving automatic evaluation, identification and classification of measurement curves  
*Andrew Gillen*

**T2.6** - Efficient functionalisation of dextran-aldehyde with catechin: potential applications in the treatment of cancer  
*Susan Oliver*

**T3.5** - Stretchable and self-healing conducting polymers  
*Lenny Voorhaar*

1200 - 1215 **T1.6** - Characterisation of shear inhomogeneity of a semicrystalline polymer in couette flow  
*Andrew Phillips*

**T2.7** - Comparison of a divergent and convergent design strategy for non-viral gene delivery  
*Jessica Kretzmann*

**T3.6** - Polymeric interface materials and environmentally friendly processing methods for polymer solar cells  
*Mats Andersson*

**T4.5** - Synthesis of antibacterial peptide analogues using sequence controlled polymerization techniques  
*James Grace*

1215 - 1230 **T1.7** - Morphology of composite nanoparticles assessed by x-ray photoelectron spectroscopy  
*Florent Jasinski*

**T2.8** - Hyperbranched polymer based dual responsive drug delivery system for cancer therapy  
*Yongmei Zhao*

**T3.7** - Nanostructural control via processing kinetics in plastic solar cells  
*Alisyn Nedoma*

**T4.6** - The dynamic and versatile chemistry of amino acid functional polymers  
*Emma Brisson*

1230 - 1300 **T1.8/IS3** - NEXAFS spectroscopy of polymer thin films  
*Chris McNeill*

**T3.8/ECR2** - Synthesis and characterisation of polymer semiconductors: shining new light on plastics  
*George Vamvounis*

**T4.7/ECR3** - Nitroxide-containing materials for biofilm remediation  
*Kathryn Fairfull-Smith*

1300 - 1400 Lunch & Trade Exhibition

Convention Foyer

| Macromolecular Architectures and their Applications<br><i>Chair: Nghia Truong</i><br>Convention 1 & 2 |                                                                                                                                                                                 | Advanced/Smart Polymeric Materials and Nanocomposites<br><i>Chair: Amanda Ellis</i><br>Convention 3                                             |  | Latest Developments and New Methods in Polymer Synthesis<br><i>Chair: Masami Kamigaito</i><br>Heritage Room                             |                  | Polymeric Self-Assembly and Bio-Interfaces<br><i>Chair: Emily Pentzer</i><br>Heritage 3                                                                                                 |  |
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|                                                                                                       |                                                                                                                                                                                 |                                                                |  |                                                                                                                                         |                  |                                                                                                                                                                                         |  |
| 1400 - 1415                                                                                           | <b>T1.9/KN16</b> - Precision glycopolymers and their interactions with lectins<br><i>Remzi Becer</i>                                                                            | <b>T2.9</b> - Functional nanodiamonds by light-triggered reactions<br><i>Anja Goldmann</i>                                                      |  | <b>T3.9/KN18</b> - Recent developments in the field of Light-controlled radical polymerizations<br><i>Julien Poly</i>                   |                  | <b>T4.8</b> - Expanding the scope of visible light mediated polymerization-induced self-assembly<br><i>Jonathan Yeow</i>                                                                |  |
| 1415 - 1430                                                                                           |                                                                                                                                                                                 | <b>T2.10</b> - Polymethacrylate copolymer films containing pendant nitroxides to protect against biofilm formation<br><i>Nathan Boase</i>       |  |                                                                                                                                         |                  | <b>T4.9</b> - Self-assembled sericin/chitosan scaffold: physical and biocompatible properties<br><i>Wirasak Smitthipong</i>                                                             |  |
| 1430 - 1445                                                                                           | <b>T1.10</b> - Triblock copolymers as a new switchable superhydrophobic or superhydrophilic surface coating<br><i>Thorsten Hofe</i>                                             | <b>T2.11/KN17</b> - Preparation of polymeric particles containing reduced graphene oxide by miniemulsion polymerization<br><i>Hideto Minami</i> |  | <b>T3.10</b> - A general strategy to precision oligomers ( $\bar{D} = 1.0$ )<br><i>Jing Ren</i>                                         |                  | <b>T4.10</b> - Non-spherical polymersomes: unusual shapes obtained through fine-tuning of $\pi$ - $\pi$ interactions in perylene-containing diblock terpolymers<br><i>Chin Ken Wong</i> |  |
| 1445 - 1500                                                                                           | <b>T1.11</b> - Novel antimicrobial polymeric nanoparticles<br><i>Edgar Wong</i>                                                                                                 |                                                                                                                                                 |  | <b>T3.11</b> - Order in the chaos of plasmas: retention of complex chemical structures in plasma polymerisation<br><i>Hans Griesser</i> |                  | <b>T4.11</b> - Directed self-assembly of block copolymer nanostructures with tuneable morphologies<br><i>Chao Chen</i>                                                                  |  |
| 1500 - 1515                                                                                           | <b>T1.12</b> - Synthesis of polymeric microcapsules with an aqueous core via membrane emulsification and inverse emulsion periphery RAFT polymerization<br><i>Fumi Ishizuka</i> | <b>T2.12</b> - The improvement of dispersion of surface modified hydroxyapatite in polymer matrix for bone substitute<br><i>Kuanlin Ku</i>      |  | <b>T3.12/ECR4</b> - Synthesis, evaluation and application of arsenical-linked polymer bioconjugates<br><i>Paul Wilson</i>               |                  | <b>T4.12/KN19</b> - Bioinspired materials bridging the length scales<br><i>Olli Ikkala</i>                                                                                              |  |
| 1515 - 1530                                                                                           | <b>T1.13</b> - High glass transition temperature fluoropolymers for hydrophobic surface coatings via raft copolymerization<br><i>Stuart Thickett</i>                            | <b>T2.13</b> - Well-defined colloidal crystal films from the 2D self-assembly of core-shell semi-soft nanoparticles<br><i>Hadi Sabouri</i>      |  |                                                                                                                                         |                  |                                                                                                                                                                                         |  |
| 1530 - 1600                                                                                           | Afternoon Refreshments & Trade Exhibition                                                                                                                                       |                                                                                                                                                 |  |                                                                                                                                         | Convention Foyer |                                                                                                                                                                                         |  |

| Biomolecules, Biofunctional & Responsive Polymers<br><i>Chair: Andreas Walther</i><br>Convention 1 & 2 |                                                                                                                                                             | Advanced/Smart Polymeric Materials and Nanocomposites<br><i>Chair: Per Zetterlund</i><br>Convention 3                                  |  | Reaction Mechanism and Computational Polymer Science<br><i>Chair: Graeme Moad</i><br>Heritage Room                                                                            |  | Polymeric Self-Assembly and Bio-Interfaces<br><i>Chair: Olli Ikkala</i><br>Heritage 3                                                                                              |                  |
|--------------------------------------------------------------------------------------------------------|-------------------------------------------------------------------------------------------------------------------------------------------------------------|----------------------------------------------------------------------------------------------------------------------------------------|--|-------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|--|------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|------------------|
|                                                                                                        |                                                                                                                                                             |                                                       |  |                                                                                                                                                                               |  |                                                                                                                                                                                    |                  |
| 1600 - 1615                                                                                            | <b>T1.14</b> - Anti-fouling polymer films from recombinant biopolymers<br><i>Simon Corrie</i>                                                               | <b>T2.14/KN21</b> - Amphiphilic polymer conetworks: structural evolution during aqueous gelation<br><i>Costas Patrickios</i>           |  | <b>T3.13</b> - Synthesis of ultrahigh molecular weight and sequence-controlled polymers via emulsion polymerization<br><i>Nghia Truong Phuoc</i>                              |  | <b>T4.13/KN24</b> - Recent advances in 19F MRI agents for detection of diseased tissue and cell tracking<br><i>Andrew Whittaker</i>                                                |                  |
| 1615 - 1630                                                                                            | <b>T1.15</b> - Tuning responsiveness of protein polymer via co-assembly approach<br><i>Namita Roy Choudhury</i>                                             |                                                                                                                                        |  | <b>T3.14</b> - Quantitative first principles based kinetic modeling for the synthesis of well-defined macromolecular architectures<br><i>Gilles Desmet</i>                    |  |                                                                                                                                                                                    |                  |
| 1630 - 1645                                                                                            | <b>T1.16</b> - Nitrile oxide-norbornene addition as bio-orthogonal crosslinking chemistry for preparation of 3d cell culture matrices<br><i>Vinh Truong</i> | <b>T2.15</b> - 3D-printing of functional materials<br><i>Milena Nadgorny</i>                                                           |  | <b>T3.15/KN22</b> - Ab initio based microkinetic modelling as a tool to understand and improve controlled radical polymerization processes<br><i>Marie-Françoise Reyniers</i> |  | <b>T4.14</b> - Toroidal nanorattles: self-assembly and on-demand release of free toroids<br><i>Michael Monteiro</i>                                                                |                  |
| 1645 - 1700                                                                                            | <b>T1.17</b> - Designer biomimetic protein polymer: smart biomaterial with unusual responsiveness<br><i>Naba Dutta</i>                                      | <b>T2.16</b> - Nanoporous films from supramolecular block copolymers<br><i>Julien Bernard</i>                                          |  |                                                                                                                                                                               |  | <b>T4.15</b> - Bioinspired polymer catalysts-mimicking the active site of enzymes<br><i>Mitchell Nothling</i>                                                                      |                  |
| 1700 - 1715                                                                                            | <b>T1.18/KN20</b> - Alginate-based materials<br><i>Lisbeth Grøndahl</i>                                                                                     | <b>T2.17</b> - Emulsion-templated porous polymer scaffolds for 3D in-vitro cell culture and tissue engineering<br><i>Ahmed Eissa</i>   |  | <b>T3.16</b> - Structure-property relationships in tactic acrylic ester polymers<br><i>Brad Wells</i>                                                                         |  | <b>T4.16</b> - Temperature-directed self-assembly of multifunctional polymeric tadpoles<br><i>Valentin Bobrin</i>                                                                  |                  |
| 1715 - 1730                                                                                            |                                                                                                                                                             | <b>T2.18</b> - Multi-stimuli responsive polymers based on spiropyran-functionalised poly(N-isopropyl acrylamide)<br><i>Wei Sung Ng</i> |  | <b>T3.17/KN23</b> - Photoinducing dynamic covalent chemistry in covalent adaptable networks<br><i>Christopher Bowman</i>                                                      |  | <b>T4.17</b> - Controlling the self-assembly of polyelectrolyte-cyclic peptides: just add salt<br><i>Tammie Barlow</i>                                                             |                  |
| 1730 - 1745                                                                                            | <b>T1.19</b> - Arylamine-containing block copolymers responsive to nitric oxide<br><i>John Quinn</i>                                                        |                                                                                                                                        |  |                                                                                                                                                                               |  | <b>T4.18</b> - Quaternized P4VP-b-PS honeycomb film with biocidal and antibiofouling Effects<br><i>Ao Chen</i>                                                                     |                  |
| 1745 - 1800                                                                                            |                                                                                                                                                             |                                                                                                                                        |  |                                                                                                                                                                               |  | <b>T4.19</b> - Co-Assembled peptide nanoscaffolds which present multiple bioactive signals demonstrate superior control of cell fate in 3D cell culture<br><i>Richard Williams</i> |                  |
| 1800 - 1900                                                                                            | Annual General Meeting                                                                                                                                      |                                                                                                                                        |  |                                                                                                                                                                               |  |                                                                                                                                                                                    | Convention 1 & 2 |
| 1930 - onwards                                                                                         | Student Night<br>Students must have their lanyard and name badge to receive food and beverages                                                              |                                                                                                                                        |  |                                                                                                                                                                               |  |                                                                                                                                                                                    | Lorne Hotel      |

# Program - Wednesday 23 November 2016

## Plenary Session

Chair: Kris Thurecht

Convention 1 & 2

0830 - 1600 Trade Exhibition Open  
Barista Coffee Served



0900 - 0910 Welcome and Housekeeping

0910 - 1000 **PL5** Biomimetic materials 'controlling the nonlinear mechanics'  
*Alan Rowan - Australian Institute of Bioengineering & Nanotechnology*

1000 - 1030 Morning Refreshments & Trade Exhibition Convention Foyer  
Visit the exhibitors to have your passport stamped and go into the draw to win some great prizes

Biomolecules, Biofunctional & Responsive Polymers  
Chair: Guillaume Delaittre  
Convention 1 & 2

Advanced/Smart Polymeric Materials and Nanocomposites  
Chair: Laurent Fontaine  
Convention 3

Multiple Theme Session  
Chair: Marion Gaborieau  
Heritage Room

Latest Developments and New Methods in Polymer Synthesis  
Chair: Julien Poly  
Heritage 3



1030 - 1045 **W1.1** - Nitroxide containing microspheres as biodegradable drug delivery systems to target bacterial biofilms  
*Christiane Lang*

**W2.1** - Polyethylene as a suitable precursor material for future carbon fibre production  
*Andreas De Palmaenar*

**W3.1/KN25 - Macromolecular Architectures and their Applications**  
Integration of DNA with polymers  
*Amanda Ellis*

**W4.1** - Systematic study of high molecular weight acrylamide polymerisations using aqueous REDOX initiated RAFT  
*Reece Lewis*

1045 - 1100 **W1.2** - Plasma polymer meets surface initiated ARGET-ATRP; A facile way for robust coatings to immobilize antifungals  
*Thomas D. Michl*

**W2.2** - Design and preparation of inverse polymerized high internal phase emulsions (HIPEs) stabilized by amphiphilic diblock copolymers  
*Stuart Thickett*

**W4.2** - Prebiotic chemistry inspired polymers for coatings in material science applications  
*Richard Evans*

1100 - 1115 **W1.3** - Preparation of biodegradable and biocompatible emulsion templated scaffolds for tissue engineering by thiol-acrylate polymerization of polycaprolactone macromers  
*Caitlin Langford*

**W2.3** - Photo-induced self assembly of block copolymers  
*Lewis Chambers*

**W3.2 - Polymeric Self-Assembly and Bio-Interfaces**  
Bioactive nano- and microstructures from self-assembling amphiphilic glycopolymers  
*Neil Cameron*

**W4.3** - Multiblock copolymer synthesis via aqueous room temperature raft polymerisation  
*Liam Martin*

1115 - 1130 **W1.4** - Hydrogels containing hierarchical structures based on poly(2-oxazoline)s and related polymers  
*Tim Dargaville*

**W2.4** - Stimuli-responsive dynamic polymers and their applications  
*Kei Saito*

**W3.3/ECR6 - Latest Developments and New Methods in Polymer Synthesis**  
Shaping the future of polymer molecular weight distributions  
*Brett Fors*

**W4.4/KN26** - Exploiting photochemical selectivity in macromolecular synthesis  
*Christopher Barner-Kowollik*

1130 - 1145 **W1.5** - Light responsive gelatin hydrogel particles for cartilage regeneration  
*Fany Li*

**W2.5** - Stimuli-responsive solid supports for capture and release of analytes  
*Lucy Weaver*

1145 - 1200 **W1.6** - Saving vision with light: Photo-modulated ocular drug delivery  
*Johan Basuki*

**W2.6** - Enhanced carbonisation efficiency of polyacrylonitrile by reinforcement with spinifex nanocellulose  
*Edward Jiang*

**W4.5** - Driving polymer and surface modification using visible light  
*James Blinco*

1200 - 1215 **W1.7** - Hierarchical self-assembly of triblock glycopolymers into multicompartimentalised structures inspired by viruses  
*Alberto Piloni*

**W2.7** - Development of oil palm trunk fiber and polyester fiber composites for sound absorption materials  
*Rungsima Chollakup*

**W4.6** - Photocatalysts; an efficient tool for the control of the polymer architecture  
*Gyrille Boyer*

1215 - 1230 **W1.8** - Addition-fragmentation chain transfer crosslinked hydrogels for rapid photoinduced degradation and stress-relaxation  
*Ian Marozas*

**W2.8** - High internal phase emulsions stabilized by functionalized copolymer: Effect of emulsion formulation and RAFT end-group on the pore structure of polyHIPEs  
*Aminreza Khodabandeh*

**W4.7** - Photopolymerization of RAFT agents under visible light irradiation  
*Tom McKenzie*

1230 - 1300 **W1.9/ECR5** - Biomaterials that promote endothelialization  
*Daniel Heath*

**W4.8/ECR7** - Spontaneous zwitterionic copolymerization of cyclic imino ethers: first steps to biodegradable and biocompatible 2-oxazoline polymers?  
*Kristian Kempe*

1300 - 1400 Lunch & Trade Exhibition

Convention Foyer

| Macromolecular Architectures and their Applications<br><i>Chair: Joaquin Sanshis</i><br>Convention 1 & 2 |                                                                                                                                                                                    | Latest Developments and New Methods in Polymer Synthesis<br><i>Chair: Christopher Barner-Kowollik</i><br>Convention 3                                                                       | Polymeric Self-Assembly and Bio-Interfaces<br><i>Chair: Richard Williams</i><br>Heritage Room                                                                   | Polymeric Nanomedicine, Therapeutics and Imaging Agents<br><i>Chair: Angus Johnston</i><br>Heritage 3                                       |
|----------------------------------------------------------------------------------------------------------|------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|---------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|-----------------------------------------------------------------------------------------------------------------------------------------------------------------|---------------------------------------------------------------------------------------------------------------------------------------------|
| 1400 - 1415                                                                                              | <b>W1.10</b> - Peptide decorated polymeric dendrimers and their biological applications<br><i>Zhongfan Jia</i>                                                                     | <b>W2.9/KN28</b> - Controlled polymer architectures for probing cell uptake mechanism<br><i>Sébastien Perrier</i>                                                                           | <b>W3.4</b> - Protein delivery via self-assembling peptide scaffolds: supporting human embryonic stem cell-derived transplants in stroke<br><i>David Nisbet</i> | <b>W4.9</b> - Engineering of functional nanocapsules by nanoprecipitation<br><i>Julien Bernard</i>                                          |
| 1415 - 1430                                                                                              | <b>W1.11</b> - Synthesis of symmetrical and asymmetrical complex polymer architectures via convergent “graft to” approach<br><i>Faheem Solangi</i>                                 |                                                                                                                                                                                             | <b>W3.5</b> - Polypeptide block co-polymers as versatile platform for drug delivery<br><i>Tushar Borase</i>                                                     | <b>W4.10</b> - Cationic polymers based on fructose and galactose moieties for nucleic acids delivery<br><i>Simon Ting</i>                   |
| 1430 - 1445                                                                                              | <b>W1.12</b> - Engineering shape and core of polymeric nanoparticles via emulsion polymerization and temperature-induced morphological transformation<br><i>Nghia Truong Phuoc</i> | <b>W2.10</b> - Dihydroxyanthraquinone derivatives: do positions of hydroxyl substituents affect their photoinitiating ability of photopolymerization under blue led bulb?<br><i>Pu Xiao</i> | <b>W3.6</b> - The electrokinetic properties of lubricin anti-adhesive protein<br><i>George “Wren” Greene</i>                                                    | <b>W4.11</b> - Using adaptive nanoparticles to target endosomal signalling<br><i>Nicholas Veldhuis</i>                                      |
| 1445 - 1500                                                                                              | <b>W1.13</b> - Hyperbranched polymers by thiol-yne chemistry: Control of molecular weight and branch point distribution<br><i>Alexander Cook</i>                                   | <b>W2.11</b> - Selective photo-activation for precision polymer synthesis<br><i>Jiangtao Xu</i>                                                                                             | <b>W3.7</b> - Controlled attachment of <i>Pseudomonas aeruginosa</i> with colloidal crystal based topographies<br><i>Hitesh Pingle</i>                          | <b>W4.12</b> - Cyclic peptide-polymer conjugates: self-assembling vectors for drug delivery<br><i>Sophie Larnaudie</i>                      |
| 1500 - 1515                                                                                              | <b>W1.14/KN27</b> - Functional nanomaterials via click chemistry: from polymeric nanoparticles to magneto-induced drug-delivery systems<br><i>Laurent Fontaine</i>                 | <b>W2.12</b> - New multifunctional “switchable” RAFT agents for accessing triblock copolymers with less active and more active segments<br><i>Sarah Stace</i>                               | <b>W3.8</b> - Polymeric nano-objects of various morphologies via block copolymer self-assembly using microporous membranes<br><i>Per Zetterlund</i>             | <b>W4.13</b> - 19F magnetic resonance imaging switches: advanced drug delivery and monitoring<br><i>Adrian Fuchs</i>                        |
| 1515 - 1530                                                                                              |                                                                                                                                                                                    | <b>W2.13</b> - Amine retardation of radical mediated thiol-ene<br><i>Dillon Love</i>                                                                                                        | <b>W3.9</b> - Nanoscale patterning of inherently conducting polymers for regenerative therapy<br><i>Iyer Swaminathan</i>                                        | <b>W4.14</b> - Engaging the sharks in a tumour attack - single domain antibodies for specific targeting of nanomedicines<br><i>Pie Huda</i> |
| 1530 - 1600                                                                                              | Afternoon Refreshments & Trade Exhibition                                                                                                                                          |                                                                                                                                                                                             | Convention Foyer                                                                                                                                                |                                                                                                                                             |
| Plenary Session<br><i>Chair: Martina Stenzel</i>                                                         |                                                                                                                                                                                    |                                                                                                                                                                                             |                                                                                                                                                                 |                                                                                                                                             |
| 1600 - 1650                                                                                              | <b>PL6</b> Stimuli-responsive smart soft materials<br><i>Takuzo Aida - The University of Tokyo</i>                                                                                 |                                                                                                                                                                                             |                                                                                                                                                                 |                                                                                                                                             |
| 1650 - 1700                                                                                              | <b>Symposium Conclusion</b>                                                                                                                                                        |                                                                                                                                                                                             |                                                                                                                                                                 |                                                                                                                                             |
| 1900 - 2300                                                                                              | <b>Symposium Dinner</b><br>Mantra Lorne                                                                                                                                            |                                                                                                                                                                                             |                                                                                                                                                                 |                                                                                                                                             |

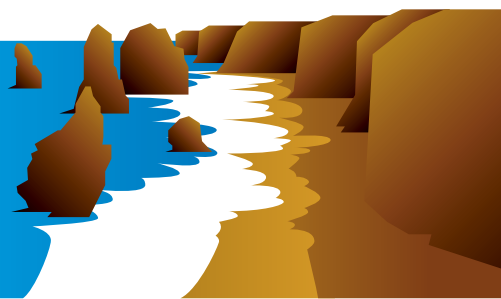
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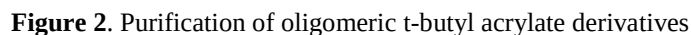


# ORAL ABSTRACTS



Craig J. Hawker

A versatile strategy will be described for the multi-gram synthesis of discrete oligomers from commercially available monomer families, e.g., acrylates, styrenics, siloxanes. Central to this strategy is the identification of reproducible procedures for the separation of oligomer mixtures using automated flash chromatography systems with the effectiveness of this approach demonstrated through the multi-gram preparation of discrete oligomer libraries ( $\text{Đ} = 1.0$ ) (**Fig.1**). Synthetic availability, coupled with accurate structural control, allows these functional building blocks to be harnessed for both fundamental studies as well as targeted technological (**Fig.2**).



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| 1988       | PhD – University of Cambridge                 |
| Since 2004 | UCSB                                          |

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# A New Age of Functional and Responsive Thermoplastic Elastomers: Discovering a Buried Treasure

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With recent advances made in the design and development of multifunctional polymeric materials, elastomers derived from triblock copolymers are in a good position to meet contemporary materials challenges and explore new technological opportunities. In this spirit, we consider the versatility of thermoplastic elastomer (TPE) systems, which provide an attractive alternative to chemically cross-linked materials because of their ability to microphase-separate and form nanostructures connected by a physically cross-linked network that imparts shape memory even after substantial deformation. We first consider the ability of these copolymers to form networks via midblock bridging by examining molecularly asymmetric triblock copolymers grown from a parent diblock copolymer. A combination of experimental methods, theoretical predictions and simulation results reveals the conditions leading to the onset of a load-bearing network. Upon incorporation of a midblock-selective oligomer, the molecular networks formed by triblock copolymers can be swollen to yield highly elastic systems, which exhibit composition-tunable mechanical properties, as well as remarkable electromechanical properties. The morphological features and mechanical properties of swollen TPE gel systems generated from commercial and model styrenic TPEs will be surveyed. These materials, subjected to electrical stimulation as electroelastomers (i.e., dielectric elastomers) between compliant electrodes, are shown to achieve actuation strains greater than 300% and electromechanical coupling efficiencies in excess of 90%. The electrostatic mechanism by which electroactuation proceeds, as well as comparisons with other materials, will be discussed. Unlike conventional dielectric elastomers composed of chemically cross-linked elastomers, these copolymer systems are easily processable and broadly tunable in terms of composition, concentration and molecular weight, and they exhibit little cyclic hysteresis. In the case of acrylic TPEs, electroactuation occurs without the need for mechanical prestrain (often required to thin specimens so that lower voltages can be used to achieve high fields). These same triblock copolymers can be designed in conjunction with liquid metals and phase-change materials to yield ultrastretchable/flexible electronics capable of remaining conductive beyond 1000% strain. Development of stimuli-responsive, hyperelastic and shape-recovering polymeric materials, such as the materials described here, is critical for emerging technologies such as (micro)robotics and microfluidics. As illustrated in Figure 1, shape memory can likewise be achieved through the physical addition of phase-change materials that are midblock-compatible. Incorporation of a charged midblock results in network-forming amphiphilic materials that can be used in ionic polymer-metal composites and photovoltaic devices. Using electron tomography and scattering, we show that the morphological attributes and associated properties of these multiblock ionomers can be solvent-templated and solvent-annealed.

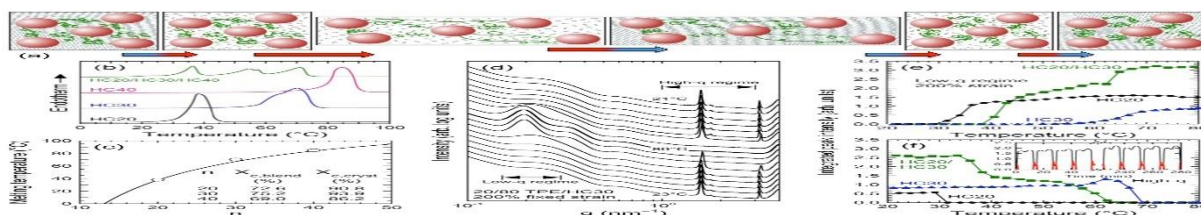


**Figure 1.** Principle of shape-memory polymer blends derived from thermoplastic elastomers.

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Research interests: Polymer nanostructures, nanocomposites and networks with emphases on polymer interfaces and morphology and the use of electron tomographic methods to study them.

# Biomimetic Polymers for Protein Stabilization: From Design to Synthesis to Applications in Food and Medicine

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Proteins and enzymes are important for many applications including in personal care products, food industries and as therapeutics, but their instability can limit application or increase costs.<sup>1</sup> Inspired by the stabilizing capability of the disaccharide trehalose, we synthesized a library of glycopolymers with trehalose side chains (**Fig.1**) and demonstrated that they stabilize various enzymes and proteins to environmental stressors including heat, desiccation, and even electron-beam irradiation better than trehalose itself.<sup>2-5</sup> We have studied the effect of different regioisomers of sugar attachment on the resulting stabilization and investigated the mechanisms of stabilization. Furthermore, we have altered the polymer backbone to render the polymers degradable. Recently, we have extended the application of trehalose polymers by incorporating them into hydrogels and self assembled materials. For example, we prepared hydrogels using a short, cost effective synthesis. We demonstrated that these hydrogels could stabilize enzymes important in animal feed and chemical industry. This talk will discuss this research, as well as other systems that could be employed to stabilize biologics for potential use in the personal care products, medicine and food.

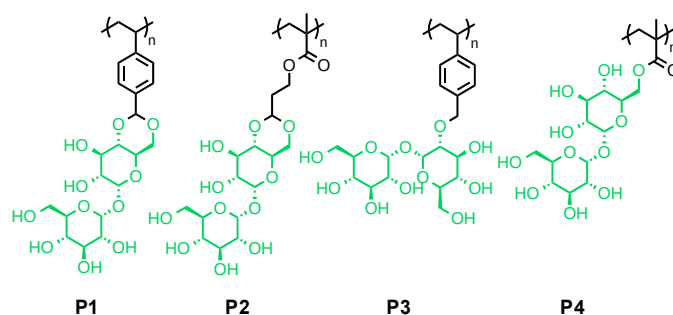


**Figure 1.** Trehalose glycopolymers stabilize proteins and enzymes to extreme conditions.

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<sup>2</sup> R. J. Mancini, J. Lee, H. D. Maynard, *J. Am. Chem. Soc.* **2012**, 134, 8474.

<sup>3</sup> J. Lee, E. W. Lin, U. Y. Lau, J. L. Hedrick, E. Bat, H. D. Maynard, *Biomacromolecules* **2013**, 14, 2561.

<sup>4</sup> E. Bat, J. Lee, U. Y. Lau, H. D. Maynard, *Nature Communications* **2015**, 6, 6654.

<sup>5</sup> J. Lee, J. H. Ko, E. W. Lin, P. Wallace, F. Ruch, H.D. Maynard, *Polymer Chemistry* **2015**, 6, 3443.

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| 2002-present        | Professor, Director of Chemistry Biology Interface Training Program, Associate Director California NanoSystems Institute, University of California, Los Angeles, |
| Research interests: | biomimetic polymer synthesis and application in medicine, hydrogel synthesis, novel polymeric resists, drug delivery                                             |

# Building Advanced, Bioresponsive Materials from Polymers, Macromolecules and Metal Ions

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The design and assembly of responsive materials underpins the development of particle carriers for biomedical applications.<sup>1,2</sup> This presentation will detail various assembly strategies and chemistries to generate multifunctional and responsive particles. Recent studies on the development of nanostructured polymer particles for efficient cargo encapsulation, triggered release and antibody-mediated targeting will be highlighted. It will be shown that engineered particles with well-controlled physicochemical properties, including shape and surface characteristics, can be used to uncover several key principles that govern particle-cell interactions.<sup>3</sup> The assembly of organic ligands with metal ions will also be presented, highlighting metal-ligand complexation as a facile approach to generate particles that can disassemble under cellular conditions.<sup>4,5</sup> It is expected that such engineered particle systems represent a class of novel materials for therapeutic delivery, as well as an interesting paradigm to attain detailed knowledge of complex nano-bio interactions. This knowledge may aid in the rational design of nanoengineered materials (beyond particle systems) for biological applications.

<sup>1</sup> J. Cui, J. J. Richardson, M. Björnalm, M. Faria, F. Caruso, *Acc. Chem. Res.* **2016** DOI: 10.1021/acs.accounts.6b00088.

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<sup>3</sup> G. K. Such, Y. Yan, A. P. R. Johnston, S. T. Gunawan, F. Caruso, *Adv. Mater.* **2015**, 27, 2278.

<sup>4</sup> H. Ejima, J. J. Richardson, K. Liang, J. P. Best, M. P. van Koeverden, G. K. Such, J. Cui, F. Caruso, *Science* **2013**, 341, 154.

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| 2013-present | Bionics Institute, Australia<br><b>Honorary Research Fellow</b>                                                                                                                                                                                           |
| 2003-2010    | Centre for Nanoscience and Nanotechnology, The University of Melbourne<br><b>Founder and Director</b>                                                                                                                                                     |
| 1997-2002    | Max Planck Institute of Colloids and Interface, Germany<br><b>Group Leader &amp; Research Scientist</b> (1999-2002)<br><b>Alexander von Humboldt Research Fellow</b> (1997-1998)                                                                          |
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Research interests: Biomaterials, particle-based delivery systems, protein and polymer assembly

## Biomimetic Materials ‘Controlling the nonlinear mechanics’

Maarten Japers, Zaskia Eksteen Akeroyd, Matthieu Koepf, Vincent A.A. Le Sage, Chris Wilson Eduardo Mendes, Paul H.J. Kouwer, and Alan E. Rowan

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Fibrous networks of biopolymers are found in both the intracellular and extracellular matrix. From the microscopic scale of a single cell to the macroscopic scale of fibrous tissues, biopolymers with different stiffness control cellular processes such as cell differentiation, proliferation, transportation and communication.<sup>1</sup> In recent years, a large number of different hydrogels has been developed, often with the goal to create an artificial extracellular matrix for biomedical applications. However, the mechanical environment inside and outside the cell is not determined by a single component.<sup>1</sup> Multiple biopolymers with different structural and mechanical properties which physically interact with each other, make the mechanical environment of a cell in vivo much more complicated than the environment of a cell in a single-component artificial matrix.

The mechanics of natural biopolymer gels however, are very different from most synthetic hydrogels because they show strain stiffening behaviour.<sup>2,4</sup> Reconstituted networks of cytoskeletal polymers such as actin or intermediate filaments or extracellular biopolymers such as collagen or fibrin show a large increase in stiffness upon an applied stress or deformation.<sup>2,4</sup> The stiffening response prevents these networks from breaking under external stresses and also enables communication between cells growing in these materials. Recently a new biomimetic polymer hydrogel was developed with unique cytomimetic properties, based upon oligo(ethylene glycol) grafted polyisocyanopeptides. These extremely stiff helical polymers form gels upon warming at concentrations as low as 0.005 %wt polymer, with materials properties almost identical to these of intermediate filaments and extracellular matrices.<sup>3,4</sup> The unique ability of these materials and their application in cell growth and drug therapeutics revealed the importance of polymer stiffness and material non-linear mechanics.<sup>5,6</sup>

These nonlinear mechanical properties have been extensively studied in single-component biopolymer networks, but little is known about how the stiffening response is affected by the composite nature of natural biopolymer networks such as the cytoskeleton or the extracellular matrix.<sup>7</sup> Approaches of how to control these nonlinear properties and their importance will be presented.

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# Stimuli-Responsive Smart Soft Materials

Takuzo Aida<sup>1,2</sup>

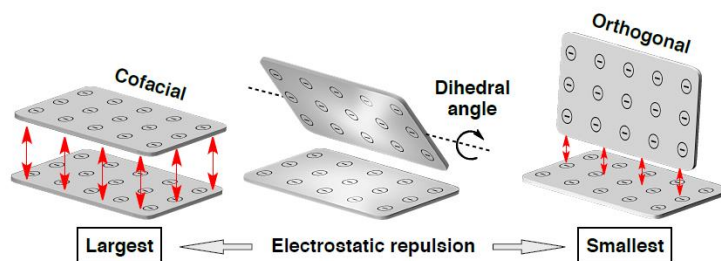
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Machine technology frequently puts magnetic or electrostatic repulsive forces to practical use, as in maglev trains, vehicle suspensions or non-contact bearings. In contrast, materials design overwhelmingly focuses on attractive interactions, such as in the many advanced polymer-based composites, where

inorganic fillers interact with a polymer matrix to improve mechanical properties. However, articular cartilage strikingly illustrates how electrostatic repulsion can be harnessed to achieve unparalleled functional efficiency: it permits virtually frictionless mechanical motion within joints, even under high compression. Here we describe a composite hydrogel with anisotropic mechanical properties dominated by electrostatic repulsion between negatively charged unilamellar titanate nanosheets embedded within it. Crucial to the behaviour of this hydrogel is the serendipitous discovery of cofacial nanosheet alignment in aqueous colloidal dispersions subjected to a strong magnetic field, which maximizes electrostatic repulsion<sup>6</sup> and thereby induces a quasi-crystalline structural ordering over macroscopic length scales and with uniformly large face-to-face nanosheet separation. We fix this transiently induced structural order by transforming the dispersion into a hydrogel using light-triggered *in situ* vinyl polymerization. The resultant hydrogel, containing charged inorganic structures that align cofacially in a magnetic flux, deforms easily under shear forces applied parallel to the embedded nanosheets yet resists compressive forces applied orthogonally. We anticipate that the concept of embedding anisotropic repulsive electrostatics within a composite material, inspired by articular cartilage, will open up new possibilities for developing soft materials with unusual functions.

More recently, we reported a novel carbon nitride polymer, which shows anomalous mechanical responses to minute fluctuations in the ambient humidity.



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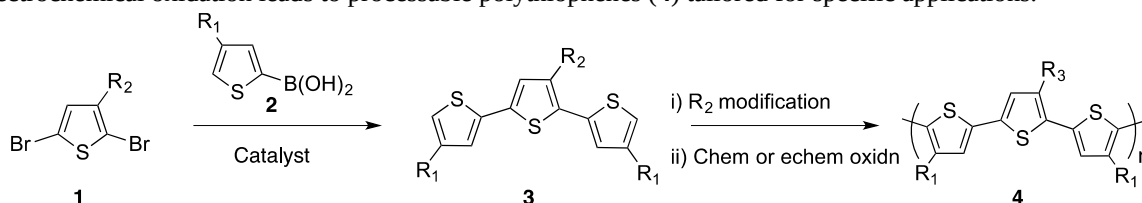
# Functionalised Polythiophenes For Energy, Sensing and Bio- Applications

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Conducting polymers possess properties that make them attractive materials for use in areas ranging from energy storage and conversion,<sup>1</sup> to sensors,<sup>2</sup> drug delivery,<sup>3</sup> and biomedical applications.<sup>4</sup> Predominant amongst this class of polymers are polythiophenes due to the ease of preparation of a wide variety of functionalized polymers. Functionalised thiophene monomers can typically be readily prepared and chemically or electrochemically polymerized. However, while there have been substantial advances in the development of functional, polythiophene-based materials, limitations still exist in the use of these materials, particularly with respect to the production of functionalized polythiophenes with satisfactory solubility and processability. In addition, the introduction of an often-bulky substituent onto each monomer unit of the polythiophene backbone often leads to steric disruption of the backbone that significantly impacts on the polymer properties.

In order to address these issues and develop a general approach to soluble and processable functionalized polythiophenes, we have been investigating the potential of terthiophene as a readily functionalisable precursor to a wide variety of substituted polythiophenes. Solubilising functionality for organic solvents such as alkyl or alkoxy groups ( $R_1$ ) can be introduced onto the periphery of the substituted terthiophene (**3**) by way of thiophene boronic acids (**2**) and reactive functionality ( $R_2$ ) can be introduced using readily prepared functionalized dibromothiophenes (**1**). In a similar fashion, water-solubilising functionality can also be introduced. Modification of the substituted terthiophene (**3**) to give the required substituent followed by chemical or electrochemical oxidation leads to processable polythiophenes (**4**) tailored for specific applications.



This approach not only allows the development of processable polythiophenes but also the introduction of bulky substituents that have a reduced steric impact on the polymer backbone. As a result, we have been able to prepare a number of oligo- or poly-thiophenes with a variety of application specific functionality for sensing,<sup>5</sup> energy applications,<sup>6</sup> and bio-applications.<sup>7</sup> These and other examples will be discussed in this lecture, highlighting the strengths and weaknesses of this approach to functionalised polythiophenes.

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## Understanding the secrets of nanoparticle cell interactions

Angus P.R. Johnston,<sup>1,2</sup> Laura I. Selby,<sup>1</sup> Sarah K. Mann,<sup>1,3</sup> Georgina K. Such<sup>3</sup>

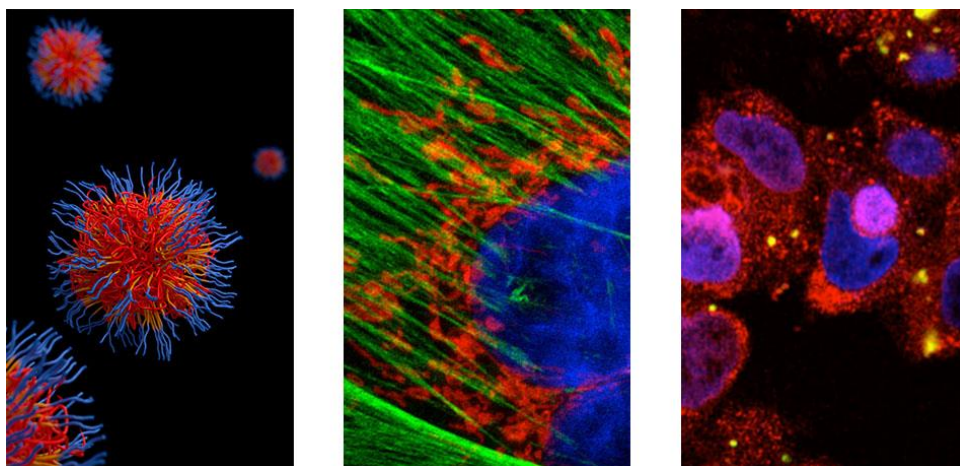
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To engineer 'smart', responsive materials for drug delivery it is essential to understand how nanoparticles interact with cells. Targeted delivery of drugs to specific cells in the body by immobilising therapeutics inside antibody functionalised nanoparticles has the potential to revolutionise the treatment of many diseases. However, our understanding of how these nanoengineered materials interact with cells is limited.

We are developing tools to understanding how these materials interact with cells,<sup>1,2</sup> so we can engineer materials that respond better to the biological conditions they encounter.<sup>3,4</sup> In particular, we are interested in understanding the internalisation, processing and trafficking of nanoparticles in cells. This presentation will focus on understanding the internalisation of polymer nanoparticles into cells, and their subsequent fate once they are inside the cell. It will also outline the progress we are making towards understanding how nanoparticles can induce transport of drugs from the endosomal compartments into the cytoplasm.



<sup>1</sup> Liu, H.; Johnston, A. P. R. A Programmable Sensor to Probe the Internalization of Proteins and Nanoparticles in Live Cells. *Angew Chem Int Edit* **2013**, 52, 5744–5748.

<sup>2</sup> Selby, L. I.; Kongkatigumjorn, N.; Such, G. K.; Johnston, A. P. R. HD Flow Cytometry: an Improved Way to Quantify Cellular Interactions with Nanoparticles. *Advanced Healthcare Materials* **2016**, 5, 2333–2338.

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2012-15 ARC Future Fellow, Melbourne University & Monash University

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Research interests: nanotechnology, cellular interactions, sensors

# **The Origin of the Polymer Banknote**

*T H Spurling<sup>1</sup>*

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This is an account of the Reserve Bank of Australia and CSIRO working together to produce the world's most secure and durable banknotes. It was known within CSIRO as 'the Bank project', and was one of the Organisation's longest and most successful projects.

The project to develop more secure banknotes commenced in 1968 following the 1966 discovery of forgeries of the new Australian decimal currency. Our story ends in 1988 with the release of the \$10 bi-centennial commemorative banknote. Of course the search for more secure banknote technology never ends.

This talk draws upon personal recollections of events and the archived CSIRO records of the project. It is therefore very much the story of the project not only from a CSIRO point of view but from the point of view of people intimately involved in the project.

I hope to make two points. The first is to convey not only the fun and excitement of doing highly applicable science but also the frustration and hard work involved in bringing ideas to the market place.

The second is to show how CSIRO contributes to the nation's economic and social well-being by conducting 'market pull' projects. Such projects were rare at Fishermens Bend in the 1960s and 1970s. Indeed, the Bank project was one of the first genuine 'market pull' projects in CSIRO. In 1968 there were no scientists in CSIRO working on projects to make more secure banknotes with the idea of contacting a banknote producer to see if they were interested in their invention. CSIRO was given the problem by the Bank and had to assemble all the skills and disciplines needed solve the problem. This meant in addition to having to enlist his chemists to work on the project one of the authors had to recruit physicists and engineers to his Division of Applied Organic Chemistry. In those days there was little incentive for other parts of the Organisation to lend their skills to the project. This is not the case in the modern CSIRO. Neither is the flexibility that the 1970s Chief had to muster resources. CSIRO knows that it needs to constantly review and modify its approach to the users of its research.

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1969-2003 CSIRO

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# Utilizing Interfacial Assembly of Graphene Oxide Nanosheets for Tailored Structures

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The Pentzer lab uses fundamental organic chemistry reactions to tune chemical functionality and assembly behavior of organic materials. One multifunctional material studied, graphene oxide (GO), are sheets of  $sp^2$  and  $sp^3$  hybridized carbon atoms decorated with oxygen functionalities throughout.<sup>1</sup> Not only can this material be covalently modified using relatively simple organic chemistry reactions, but the nanosheets also have antimicrobial and gas barrier properties, good mechanical strength, and can serve as precursors to conductive material.<sup>2</sup> We will report our recent advances in using the interfacial assembly of GO to access three higher order structures: 1) Hollow capsules of GO by stitching together of the nanosheets at the oil-water interface with a di-functional small molecule;<sup>3</sup> 2) Polymer particles armored with GO nanosheets that can be covalently functionalized and assembled with inorganic nanoparticles;<sup>4</sup> and 3) Janus GO nanosheets in which each face is functionalized orthogonally with two different polymers or small molecules (Figure 1).<sup>5</sup> First, the Pinner reaction is used to covalently modify the alcohol groups of GO in suspension to give nanosheets with alkene handles on both faces.<sup>6</sup> Then, the alkenylated GO is assembled at the oil-water interface in a Pickering type emulsion, exposing the nanosheet faces to only oil or water. Subsequent thiol-ene click chemistry then covalently modifies each face independently to yield robust structures. A suite of electronic and microscopic techniques are used to characterize the materials.

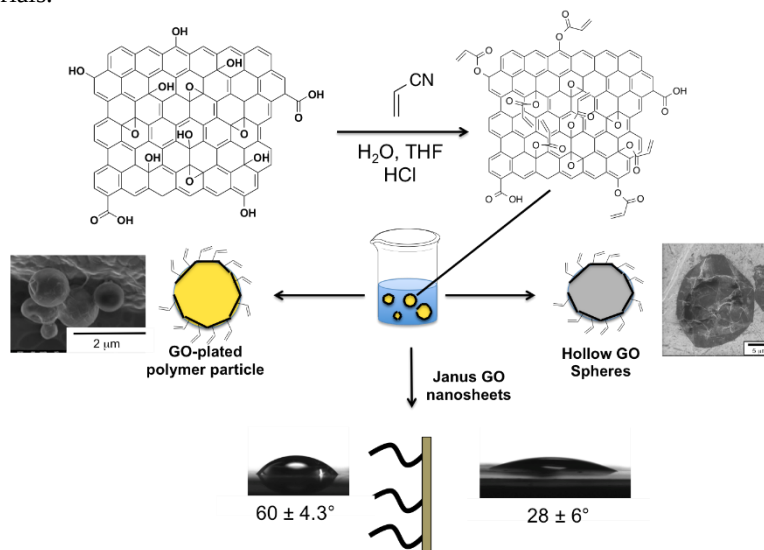


**Figure 1.** Overview of the work to be presented: Assembly of alkenylated graphene oxide (GO) at oil-water interface and modification to give Janus GO nanosheets, hollow GO capsules, and GO-armoured polymer particles.

<sup>2</sup> L. Liao, H. Peng, Z. Liu, *J. Am. Chem. Soc.*, **2014**, 136, 12194-12200.

<sup>3</sup> H. Y. Mao, S. Lauren, W. Chen, O. Akhavan, M. Imani, A. A. Ashkarran, M. Mahmoudi, *Chem. Rev.*, **2013**, 113, 3407-3424.

<sup>4</sup> Q. Luo, P. Wei, E. Pentzer, *Carbon*, **2016**, online.

<sup>5</sup> B. J. Rodier, E. Mosher, S. Burton, R. Matthews, E. Pentzer, *Macromol. Rap. Comm.*, **2016**, online.

<sup>6</sup> B. McGrail, J. Magdalao, B. Rodier, J. Swisher, R. Advincula, E. Pentzer, *Chem Comm.*, **2016**, 52, 288-291.

B. McGrail, B. J. Rodier, E. Pentzer, *Chem. Mater.*, **2014**, 26, 5806-5811.

## Emily Pentzer

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Personal History:

2010-2013 Postdoctoral Scholar, UMass Amherst Polymer Science and Engineering

Since 2013 Assistant Professor, Case Western Reserve University

Research interests: self assembly, Janus particles, interfaces, 2D materials

## Polymer Nano- structures for MRI and PET imaging

*Thomas P. Davis<sup>1</sup>*

<sup>1</sup>ARC Centre of Excellence for Bio-Nano Science

Theranostics, a combination of drug delivery and imaging, is an emerging field, as progress could potentially lead to triggered drug release at precise physical locations (tracking), and provide instantaneous feedback on physical responses to delivery events. Building responses to different delivery vectors using a range of imaging techniques, for example, computer tomography (CT), magnetic resonance imaging (MRI), positron emission tomography (PET), and fluorescence imaging has been described in a wide range of recent publications. MRI responsiveness remains an attractive target for theranostic nanoparticle design, as MRI scanners are readily available in large public hospitals, and MRI scans do not subject patients to ionizing radiation. Two different types of MRI contrast agents (CAs) are commonly employed to build theranostic devices; both CAs have the ability to shorten the surrounding water protons longitudinal (T<sub>1</sub>) and transversal (T<sub>2</sub>) relaxation time to improve MRI contrast. Gadolinium-based CAs yield positive contrast as indicated by an increase in image brightening, while, in contrast, iron oxide nanoparticle-based CAs provide a negative contrast— leading to image darkening. Presently, the most commonly employed CAs in the clinic are based on gadolinium (Gd). As Gd is highly toxic, Gd is chelated with small organic compounds to maintain solution stability while obviating direct interactions of Gd with biological media. Small organic CAs have a number of shortcomings: i) a short biocirculation times, ii) low contrast at high magnetic fields, and iii) systemic delivery (nonspecificity). Recent published work on incorporating Gd into polymeric nanostructures has provided some encouragement that nanoparticle-based CAs can overcome the present application drawbacks. Polymeric nanoparticles can be easily tuned to produce sizes from 10 to 200 nm with bespoke morphologies, potentially to take advantage of the enhanced permeation retention (EPR) effect, for enhanced targeting. In addition, polymeric nanoparticles can be easily decorated with targeting moieties to further improve specific targeting, and they can encapsulate a greater number of Gd per nanoparticles to enhance local contrast. In another significant advantage, encapsulating Gd in nanoparticle structures reduces molecular tumbling, significantly increasing the relaxivity of Gd at high magnetic fields.

In this talk I will discuss the design of novel polymeric agents for imaging applications, and show how potentially combining novel enhanced imaging with VR technology as a data interpretation and manipulation interface can enhance both conventional high street imaging and also help in education and training.

### **Tom Davis**

Professor

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Personal History:

2014 –ARC Laureate Fellow and Director CBNS

2013 - Monash-Warwick Professor

Research interests: Polymer science, NanoBio Science



# Translational Research in Polymer Science and the Factory of the Future

*Bronwyn Fox<sup>1</sup>*

<sup>1</sup>Swinburne University of Technology, Factory of the Future, Faculty of Science, Engineering and Technology  
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The manufacture of polymer products is expected to change radically in the coming years. Swinburne's Factory of the Future is working with multinational companies and Australian SMEs in their supply chains to develop a flexible pilot line to demonstrate state of the art automated production of new polymer matrix composite products. This facility has been intentionally designed to create a platform to demonstrate to Australian manufacturers the principles and benefits of "Industry 4.0" concepts and will exploit Swinburne's existing strengths in design, visualisation, robotics, automation and big data. Industry 4.0 is described as the fourth industrial revolution and utilizes recent advances in communications and digital technologies including the "internet of things" to automate and control production processes in order for them to run seamlessly and smoothly.

In the past two decades, cure cycle times for carbon fibre reinforced polymer matrix composites have been dramatically reduced from hours to minutes (1). At the same time, the global market for carbon fibre composites is confidently expected to grow exponentially to \$36 billion by 2020 (2) (3) with key applications in aerospace, automotive, defence, wind energy and civil engineering. Now that the component cure cycle times are no longer the rate limiting step in the production process, the remaining challenge is to increase production rates by replacement of manually intensive steps with automated production.

An example of the state of the art for automated composite production is the BMW i3 battery electric vehicle manufacturing plant in Leipzig, Germany. The i3's carbon fibre composite "life module" is built using several of the latest technologies for industrial automation. However, it is noted that whilst this process represents a paradigm shift in advanced composite manufacture, there are still significant manufacturing challenges which further innovative research can help overcome. The cure cycle time for the carbon fibre composite components is 8 minutes (automotive targets are under 1 minute to compete with sheet metal forming) and this process requires some manual intervention.

This presentation will describe the current capabilities of Swinburne's Factory of the Future and will highlight the future directions for the development of new capabilities.

## **Professor Bronwyn Fox**

Director Factory of the Future

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Personal History:

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| 2016-     | Swinburne University of Technology, Director Factory of the Future |
| 2001-2015 | Deakin University, Institute for Frontier Materials, Carbon Nexus  |
| 1996-2000 | Australian National University, School of Engineering (PhD)        |
| 1993-1996 | CRC for Polymers (CSIRO)                                           |

Research interests: carbon fibre, polymer composites, structure-property relationships in polymers

# Cationic RAFT Polymerization: Synergetic Developments in Radical and Cationic Polymerizations

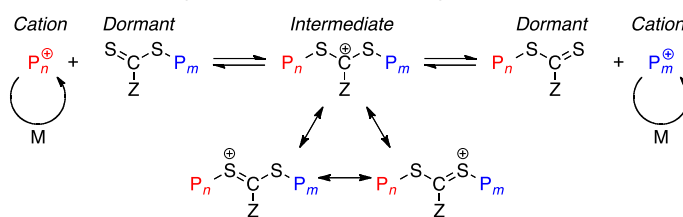
Mineto Uchiyama, Kotaro Satoh, and Masami Kamigaito

Department of Applied Chemistry, Graduate School of Engineering, Nagoya University, Nagoya 464-8603, Japan

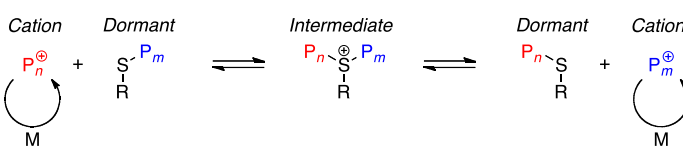
Recent developments in living or controlled polymerization have enabled the synthesis of a wide variety of well-defined polymers with precisely controlled structures. Most of these modern living/controlled polymerizations are based on reversible activation of dormant covalent polymer terminal into the growing species. Therefore, there are many similarities in concepts and strategies for controlling polymerizations irrespective different growing species.

Reversible addition-fragmentation chain transfer (RAFT) polymerization has been originally developed for controlling radical polymerization via reversible chain transfer between propagating radical species and covalent dormant species such as thiocarbonylthio compounds.<sup>1</sup> We have recently found that RAFT mechanism also works for controlling cationic polymerization of various cationically polymerizable monomers such as vinyl ethers and styrene derivatives using similar thiocarbonylthio compounds (**Scheme 1**).<sup>2</sup> The polymerization proceeds via reversible addition of the cationic propagating species to the thiocarbonylthio group to form the cationic intermediate stabilized by the sulfonium ion. Furthermore, thioethers and phosphorous esters are also effective for controlling cationic polymerization via similar reversible degenerative chain transfer (DT) or RAFT mechanisms.<sup>3,4</sup> These novel controlled cationic polymerizations have thus provided new strategies based on RAFT or DT chemistry and will lead to synergetic developments in radical and cationic polymerization for precision polymer synthesis.<sup>5</sup> This lecture will mainly cover the recent developments in cationic RAFT polymerizations.

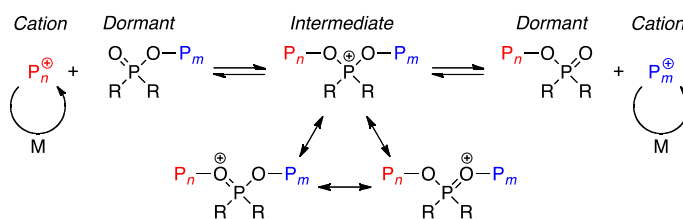
## 1. Cationic RAFT Polymerization with Thiocarbonylthio Compound



## 2. Cationic DT Polymerization with Thioether



## 3. Cationic RAFT Polymerization with Phosphorous Compound



**Scheme 1.** Cationic RAFT and DT polymerizations

<sup>1</sup> G. Moad, E. Rizzardo, S. H. Thang, *Aust. J. Chem.* **2012**, *65*, 985-1076.

<sup>2</sup> M. Uchiyama, K. Satoh, M. Kamigaito, *Angew. Chem. Int. Ed.* **2015**, *54*, 1924-1928.

<sup>3</sup> M. Uchiyama, K. Satoh, M. Kamigaito, *Macromolecules* **2015**, *48*, 5533-5542.

<sup>4</sup> M. Uchiyama, K. Satoh, M. Kamigaito, *Polym. Chem.* **2016**, *7*, 1387-1396.

<sup>5</sup> T. K. McKenzie, Q. Fu, M. Uchiyama, K. Satoh, J. Xu, C. Boyer, M. Kamigaito, G. G. Qiao, *Adv. Sci.* **2016**, *3*, 1500394.

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| 1995-1999  | Assistant Professor, Kyoto University  |
| 1997-1998  | Visiting Scholar, Stanford University  |
| 1999-2003  | Associate Professor, Kyoto University  |
| 2003-2004  | Associate Professor, Nagoya University |
| Since 2004 | Professor, Nagoya University           |

Research interests: Controlled polymerization, Precision polymer synthesis, Renewable vinyl monomers



# Single-Enzyme Nanogels: Synthesis and Immobilization

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Enzymes are not only nature's catalysts but are also essential to many synthetic processes. In the latter, it is often desirable to operate in the presence of organic solvents and at elevated temperatures. However, with respect to activity, enzymes are macromolecules which are sensitive to many environmental parameters such as pH, ionic strength, temperature, and the presence of organic solutes, to name but a few. Immobilization onto solid substrates or within crosslinked enzyme aggregates is typically employed to stabilize enzymes. Yet, common protocols are often lacking precision in terms of enzyme surface chemical modification and may lead to considerable variations in efficiency, depending on the nature of the enzyme.

Single-enzyme encapsulation was shown to stabilize various enzymes at elevated temperatures and in the presence of solvents.<sup>1</sup> It generally consists in carrying out a simple crosslinking polymerization in the presence of an enzyme previously modified in a way that it carries vinyl groups and acts as multifunctional crosslinker (**Fig. 1**). However, precise preparation protocols are scarce and advanced applications have not been pursued. We have recently set out to investigate the synthesis of single-enzyme nanogels (SENs) and particularly the parameters which may affect the formation of well-defined particles with several common proteins.<sup>1</sup>

We found new experimental conditions where pre-functionalization of the enzymes is not necessary, which makes the process simpler and consequently more attractive for applications. In addition, we showed that SENs could be immobilized onto surfaces without specific modification and rapidly by thio-Michael addition (**Fig. 2**).<sup>2</sup> We are currently working on further immobilization protocols, on imparting additional features to SENs, and on applying them in various contexts.

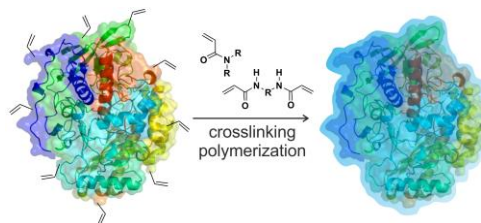


**Figure 1.** Single-molecule encapsulation of an acrylated enzyme by crosslinking polymerization at high dilution.

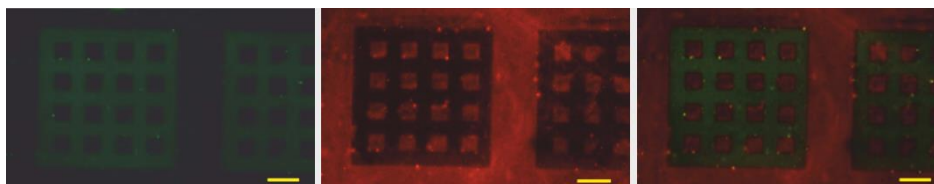


**Figure 2.** Patterns of fluorescent SENs obtained by thio-Michael addition-mediated microcontact printing.

- [1] M. Yan, J. Ge, Z. Liu, P. Ouyang *J. Am. Chem. Soc.* **2006**, 128, 11008; J. Ge, D. Lu, J. Wang, M. Yan, Y. Lu, Z. Liu *J. Phys. Chem. B* **2008**, 112, 14319.
- [2] A. Beloqui, A. Kobitski, J. Bürck, G. U. Nienhaus, G. Delaittre, submitted.
- [3] A. Beloqui, S. Baur, V. Trouillet, A. Welle, J. Madsen, M. Bastmeyer, G. Delaittre, *Small* **2016**, 12, 1716.

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2008 PhD, University Pierre and Marie Curie, Paris, France

2008-2009 Postdoc, Radboud University Nijmegen, The Netherlands

2010-2012 Alexander von Humboldt Postdoctoral Fellow, Karlsruhe Institute of Technology, Germany

Since 2013 Principal Investigator / Group Leader, Karlsruhe Institute of Technology, Germany

Research interests: macromolecular engineering, micro-/nanostructures, biofunctionalization

## Industry-driven collaborative polymer research

Ian J Dagley  
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When it completes its current funding under the Cooperative Research Centres (CRC) Programme in June 2017, the CRC for Polymers (CRCP) will have conducted 25 years of industry-driven collaborative research. The presentation will address: the scale and intensity of the collaboration; the evolution in the expectations of the stakeholders, including the CRC Programme; lessons learnt on how to best manage the process from initial contact with a potential industry partner through to a successful commercial outcome; the legacy of the training in collaborative research provided to its postgraduate students and postdoctoral fellows; and some case studies illustrating both the polymer research carried out and the commercial outcomes.

**Scale and intensity of the collaboration.** The activities of the CRCP have involved more than 40 multidisciplinary projects with a cumulative total of 48 participants comprising 24 businesses, 14 universities, three research organisations (CSIRO, ANSTO and DSTO), and seven end users. Most projects have included an industry partner and at least two universities, and up to six universities have participated in a single project. Over the period 1992-2015, the CRC has attracting \$65 million of CRC Programme funds and conducted over \$250 million of collaborative research activities.

**Evolution of the collaboration.** The evolution of the polymer research conducted by the CRCP has reflected the broader trends across the Australian polymer research community during the past 23 years (**Fig.1**). The CRC commenced as the CRC for Polymer Blends in July 1992 and, during this first term of funding, it broadened the scope of its research to become the CRC for Polymers. In its second funding period (1999-2005), it addressed the broader research needs of the plastics industry, conducting collaborative research on commodity polymers. In its third funding term, it expanded its activities to develop functional polymers to assist businesses in five growth sectors of the economy: manufacturing, biotechnology, mining, agriculture, and energy. The CRC is now in its fourth and final period of funding (2012-2017) which is allowing it to assist Australian manufacturing to develop products to meet emerging global needs in three areas: health therapies and delivery, water and food security, and low-cost solar energy, using enabling advanced polymer technology.

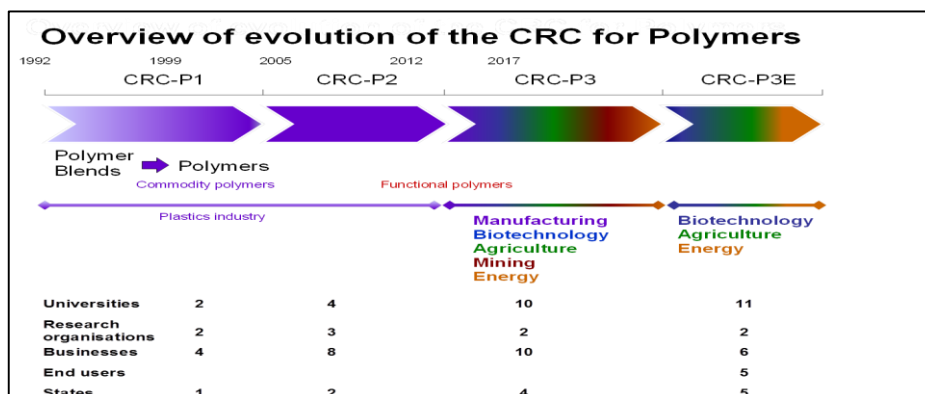


Figure 1: Overview of the evolution of the CRC for Polymers.

**Training researchers to collaborate across sectors.** The CRCP has provided funding support for 74 postgraduate students and ~ 100 postdoctoral researchers who have gained expertise in collaborative research.

**Commercial outputs and publications.** The commercially valuable technologies generated have been protected by more than 45 patent applications, and to date 17 of these technologies have been licensed to CRC participants.

### Ian Dagley

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1979-1981: Postdoctoral Research Fellow, Oxford University

1991-1985: Chief Chemist, Pacific Dunlop subsidiary

1985-1995: Research Scientist at DSTO

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Research interests: Polymer science and engineering

# Mechanical Energy to Electricity Conversion of Nanofibres and Conducting Polymers

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Recently, great efforts have been devoted to finding renewable, green energy sources to mitigate the over reliance on fossil fuels. Mechanical energy such as those from steam flow, wind, body movement or heartbeat exists everywhere. Conversion of those energy into electricity would require special materials and devices. Our recent studies have indicated that two types of materials, electrospun polyvinylidene difluoride (PVDF) nanofibres and conducting polymers, could be used as active layer for making mechanical energy-to-electricity conversion devices<sup>1-5</sup>.

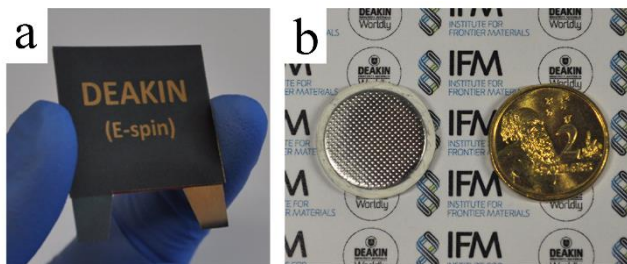


**Figure 1.** Photos of mechanical energy harvesters made of (a) electrospun PVDF nanofibre mat and (b) conducting polymer plate.

High electric field intensity and intensive fibre stretching during electrospinning have contributed to the formation of piezoelectric PVDF nanofibres with high  $\beta$  crystal phase content and oriented electric dipoles. Electrospun PVDF fibre mats with interconnected fibre structures showed not only improved energy conversion efficiency but also increased mechanical strength and structure integrity of during compression/decompression, making them very promising for making mechanical energy harvesters. Needleless electrospinning is efficient to produce PVDF nanofibres on a large scale with enhanced energy conversion behaviours. The nanofibre devices can even convert acoustic waves into electricity.

Most of mechanical energy harvesting technologies produce alternating current (AC) outputs, and an external rectifier has to be used to convert the AC into direct current (DC). The use of rectifier not only reduces the overall energy conversion efficiency but also increases complexity and difficulty in device miniaturization. However, our recent study has shown that self-rectified mechanical-to-electrical energy conversion can be attained when a free-standing conducting polymer plate is combined with two metal electrodes, which form an Ohmic and a Schottky contact separately with the plate. DC voltage with a large output current density can be generated. The electric power generated is sufficient to drive commercial electronic devices.

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1. J Fang, X Wang, T Lin, *Journal of Materials Chemistry*, 2011, 21(30), 11088.
2. J Fang, H Niu, H Wang, X Wang, T Lin, *Energy & Environmental Science*, 2013, 6(7), 2196.
3. H Shao, J Fang, H Wang, C Lang, T Lin, *ACS Applied Materials & Interfaces*, 2015, 7(40), 22551.
4. H Shao, J Fang, H Wang, L Dai, T Lin, *Advanced Materials*, 2016, 28, 1461.
5. C Lang, J Fang, H Shao, X Ding, T Lin, *Nature Communications*, 2016, DOI: 10.1038/ncomms11108.

## Tong Lin

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## Personal History:

- |           |                                                             |
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| 2012-2016 | ARC Future Fellow                                           |
| 2010-2012 | Associate Professor in Fibrous Materials, Deakin University |

Research interests: Nanofibres, Electrospinning, Functional Fabrics, Polymer Materials

# Multi-Scale Toughening of Polymer Composites Using Carbon Nano-Particles and Carbon Macro-Pins

R.B. Ladani<sup>1</sup>, M. Bhasin<sup>1</sup>, A.R. Ravindran<sup>1</sup>, S. Wu<sup>1</sup>, K. Pingkarawat<sup>1</sup>, A.J. Kinloch<sup>2</sup>, C.H. Wang<sup>1</sup>, A.P. Mouritz

<sup>1</sup>Sir Lawrence Wackett Aerospace Research Centre, School of Engineering, RMIT University, Australia

<sup>2</sup>Department of Mechanical Engineering, Imperial College London, England

Improving the delamination resistance of carbon fiber reinforced polymer composites is of significant interest to a broad range of applications, including aerospace, civil infrastructure, marine, automotive and sports goods. Composites with a brittle-like polymer matrix (e.g. non-toughen epoxy) are prone to delamination cracking caused by low interlaminar static or fatigue loads. In the present work, we present a multi-scale toughening approach to increase substantially both the delamination fracture toughness and fatigue strength of brittle polymer composites. The novel approach combines nano- and macro-scale carbon reinforcements which are inspired by natural composite materials such as bone. Carbon reinforcements with two different length scales are used: nano-scale carbon particles (nanofibres or graphene platelets) and macro-scale carbon z-pins to reinforce composites in the through-thickness direction.

The composite, featuring the three-dimensional reinforcement architecture, possesses multiple toughening mechanisms at different scales along delamination cracks thus yielding large improvements to the interlaminar fracture toughness and fatigue resistance. Figure 1 shows the measured improvements to the (a) mode I fracture toughness and (b) mode I fatigue strength of the composite due to nanofibres alone, z-pins alone, and the combination of nanofibres and z-pins. At the nano-scale, the carbon nanofibres promote high delamination resistance by multiple intrinsic and extrinsic toughening processes around the crack tip. The graphene nanoplatelets promote intrinsic toughening at and immediately ahead of the crack tip, although the toughening effect is not as strong as the carbon nanofibres. The macro-size carbon z-pins promote a strong extrinsic toughening mechanism behind the crack tip and over a larger length-scale than the carbon nanoparticles via bridging the crack faces. Under monotonic crack opening or cyclic fatigue loading, the multi-scale composites show a strong improvement in resistance against delamination crack growth.

The manufacture, delamination fracture toughness and fatigue properties, and interlaminar toughening and fatigue strengthening mechanisms of the multi-scale composites are described in the paper.

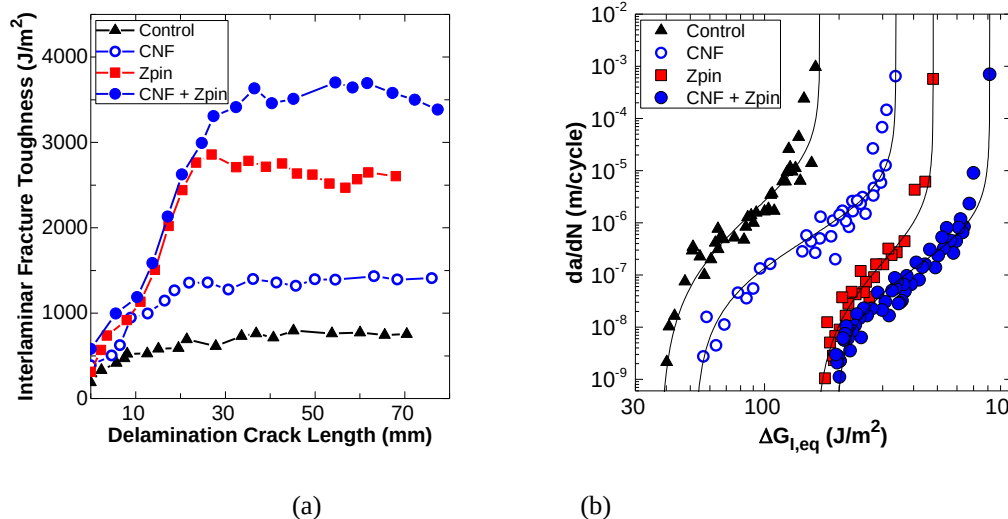


**Figure 1.** (a) Crack growth resistance curves and (b) Paris curves showing respectively the fracture toughening and fatigue strengthening effect of combining carbon nanofibres and carbon macro pins on the mode I delamination properties of carbon-epoxy laminate.

## Adrian Mouritz

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2000- Professor of Aerospace Materials

Research interests: fibre-polymer composites, aerospace materials

## **TitSoft Matter (Cryo) Electron Microscopy and Microanalysis:**

*Roger Wepf<sup>1</sup>, Matthias Floetenmeyer<sup>1</sup>, Kathryn Green<sup>1</sup> and Rick Webb<sup>1</sup>*

<sup>1</sup>CMM; University of Queensland; Brisbane 4072 QLD

Advancement in corrected transmission electron microscopy (cs-S/TEM) and cryo electron microscopy (cryo-TEM, cryo-SEM, cryo-FIB/SEM) have made it recently possible to image objects, such as organic samples and biological samples down to near atomic resolution. These techniques were originally developed for material science or biological science independently but in recent years (cryo)-EM is increasingly used to investigate soft matter of synthetic origin or natural origin.

The key issue for successful structure determination is once more sample preparation of soft matter for these novel electron microscopy opportunities. Again by borrowing methods developed for biological samples - cryo sample preparation - and adapting them to man-made soft matter has lead to an increasing interest in synthetic soft matter EM. In this overview we want to highlight the recent developments in low voltage (cryo) EM and show the potential of (cryo)EM methods on soft matter structure research projects. Finally by combining these novel sample preparation and electron microscopy methods with other analytical techniques we will elaborate on future multimodal sample characterization possibilities appearing on the horizon.

### **Roger Wepf**

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| 2016        | Director CMM – University of Queensland                                                                          |
| 2006 – 2014 | Director EMEZ – ETH Zurich, Electron Microscopy Centre                                                           |
| 1997 – 2006 | Department Head – Beiersdorf AG, First: Analytical Microscopy in R&D/ Structure Research Dept. in Research       |
| 1993 – 1997 | PostDoc Position & Junior Group Leader – European Molecular Biology Laboratory, Cell Biology and Biophysics Unit |
| 1992        | PostDoc Cell Biology ETH Zurich & MIH Biocenter Univ. Basel                                                      |
| 1988 – 1992 | pHD at Cell Biology ETHZ                                                                                         |

Research interests: Rogers major research activities are focused in the field of correlative and multimodal microscopy, automatic sample preparation and imaging techniques for particle imaging and spectroscopy technology. Currently this leads to developing tools for connectivity between imaging and spectroscopy methods and integrative imaging and spectroscopy techniques to explore new frontiers in structure research.



# Controlled Synthesis of Semiconducting Polymers

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Semiconducting polymers is a class of organic material that has been widely employed in organic electronics with applications such as organic light emitting diodes, organic field effect transistors, organic solar cells and sensors. The two key reasons for their popularity are tunable properties through modular structure modifications and solution processability enabling deposition of uniform films. The vast majority of semiconducting polymers are linear rod-like polymers with a conjugated backbone structure. There is a great variety of synthesis methods available for the construction of these conjugated polymers. Most polymerization methods involve step growth reactions that result in poorly defined materials with high polydispersity. To achieve well-defined polymer materials, chain growth polymerization mechanism is required. Currently, there are very few synthetic methods for conjugated polymers that involve chain growth mechanism. The Grignard metathesis polymerization (GRIM) method is commonly used for the controlled synthesis of thiophene homopolymers with the most prominent example being poly(3-hexylthiophene) P3HT. Ring opening metathesis polymerization (ROMP) is a fairly common method for the synthesis of non-conjugated pendant polymers with a couple of conjugated polymer examples that require highly strained cyclic monomer building blocks that are difficult to prepare. Recently, our group prepared a series of well-defined semiconducting bottlebrush polymers using controlled polymerization methods.<sup>1</sup> The electron donor semiconductor, poly(3-hexylthiophene) P3HT, was synthesized using Grignard metathesis polymerization. With appropriate norbornene end caps, the P3HT macromonomers were used to form the bottlebrush material via ring opening metathesis polymerization (**Fig. 1**). The bottlebrush polymers were characterized and compared with linear P3HT materials as well as high molecular weight commercial P3HT. It was found that longer polythiophene chains impart better semiconducting properties resulting in higher solar cell device performance. In this presentation, I will summarise this work and provide an outlook on efforts to improve polymerization control for semiconducting polymers.

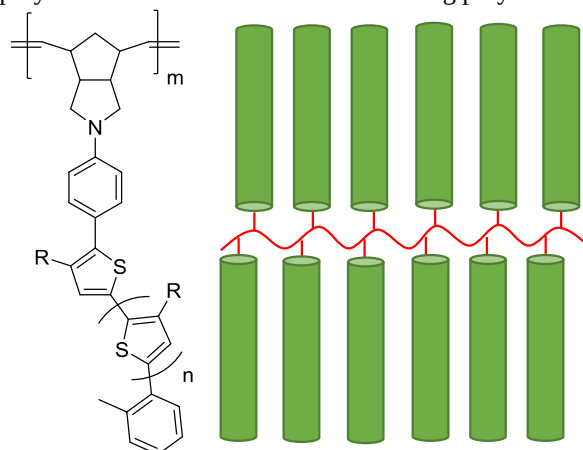


**Figure 1.** Structural illustration of the semiconducting bottlebrush polymer.

<sup>1</sup> van As, D.; Subbiah, J.; Jones, D. J.; Wong, W. W. H., Controlled Synthesis of Well-Defined Semiconducting Brush Polymers. *Macromol. Chem. Phys.* **2016**, 217, 403-413.

## Wallace W. H. Wong

Title: ARC Future Fellow

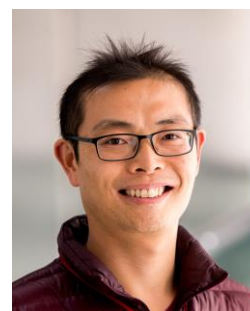
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| 2005       | DPhil (Oxford)                           |
| 2006       | Postdoctoral Research Fellow, ETH Zurich |
| 2007-2011  | Research Fellow, University of Melbourne |
| 2011-2014  | ARENA Research Fellow                    |
| Since 2014 | ARC Future Fellow                        |

Research interests: Organic electronics, organic materials synthesis, flow chemistry



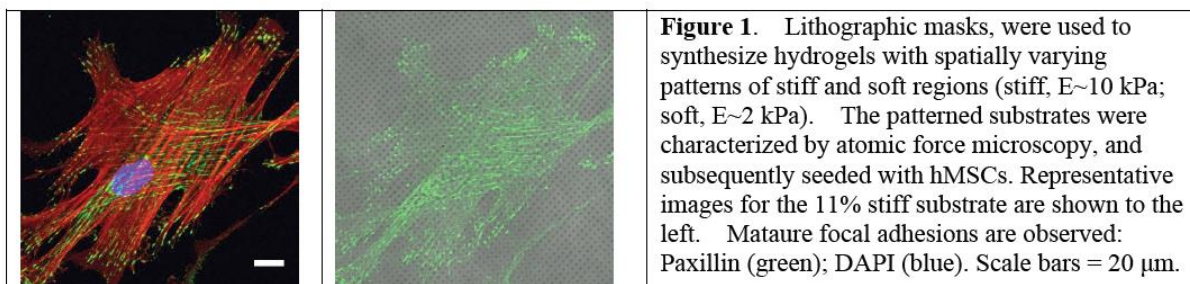
## Photodegradable hydrogels as spatiotemporally controllable cellular niches

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The expansion and differentiation of human mesenchymal stem cells (hMSCs), an important cell type that is frequently used in stem cell based therapies, are regulated by interactions with their local extracellular microenvironment. Matrix stiffness is one of the key regulators, but often varies in space at a range of length scales in the local niche. To better understand and harness cell-matrix interactions, we utilized a photo-tunable hydrogel platform<sup>1,2</sup> to provide compelling evidence that the magnitude and spatial organization of matrix mechanics influence hMSCs through systematic regulation of cytoskeletal tension and transcriptional activation. Specifically, a hydrogel cell culture substrate was fabricated with regions of spatially varied and distinct mechanical properties, which were subsequently mapped and quantified by atomic force microscopy (AFM) (Fig.1)<sup>3</sup>. The variations in the underlying matrix mechanics were found to regulate cellular adhesion and transcriptional events. Highly spread, elongated morphologies and higher Yes-associated protein (YAP) activation were observed in hMSCs seeded on hydrogels with higher concentrations of stiff regions in a dose dependent manner. However, when the spatial organization of the mechanically stiff regions was altered from a regular to randomized pattern, lower levels of YAP activation with smaller and more rounded cell morphologies were induced in hMSCs. We infer from these results that irregular, disorganized variations in matrix mechanics, compared to regular patterns, appear to disrupt actin organization, and lead to different cell fates; this was verified by observations of lower alkaline phosphatase (ALP) activity and higher expression of CD105, a stem cell marker, in hMSCs in random versus regular patterns of mechanical properties<sup>2</sup>. These findings may have important implications in deepening our fundamental understanding of the role of extracellular matrix organization on disease, aging and regenerative processes.



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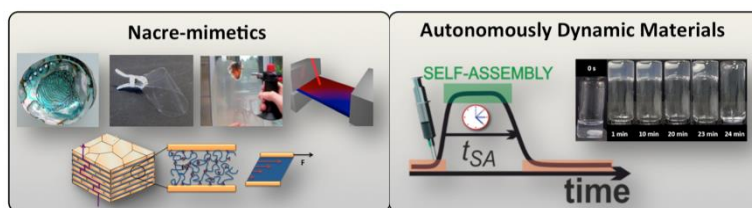
# Static and Dynamic Bioinspired Aqua-Materials

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Biology is a source of inspiration for materials science by demonstrating macroscopic materials with advanced functionalities and excellent mechanical properties. However, although these rather static architectures stimulate large interest, even more thought-provoking are the dynamic, kinetically controlled processes and temporal evolution of structures in complex biological systems. These are orchestrated through feedback loops and require energy input and dissipation to allow non-equilibrium materials and full spatiotemporal control.

In man made self-assemblies we have mastered to a large extent near-equilibrium structure formation in space and have gained an increasing understanding of how to construct very complex, hierarchically structured soft matter by using co-assemblies, competing interactions and hierarchical length scales. This has allowed to create real-life materials with unprecedented functionalities, inaccessible without delicate control over molecular interactions and sophisticated nano- and mesostructuration. The next step is to master temporal control in self-assemblies. This requires kinetic control of opposing reactions (built-up/destruction), internal feedback systems or the use of energy dissipation to sustain structures only as long as a chemical fuel is available. These approaches keep systems forcefully away from equilibrium and potentially allow neat access to temporal control. In this talk I will present concepts for bioinspired materials formed in both static and dynamic conditions. The first part will deal with rather static, nacre-inspired high-performance nanocomposites formed through self-assembly, and in which light-adaptive properties can be encoded through co-assembly and energy transfer approaches. The second part will focus on a platform concept, which allows to program self-assembling systems outside equilibrium with a lifetime by kinetic control of promoter/deactivator pairs and a simple internal feedback system. This will be showcased for different self-assembling systems.



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## Andreas Walther

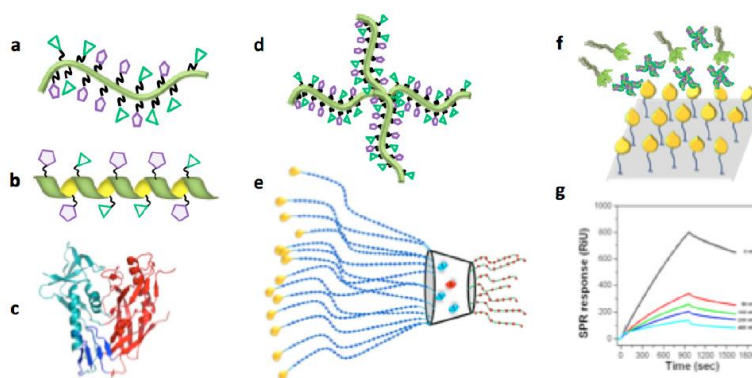
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## Precision glycopolymers and their interactions with lectins

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Sequence controlled polymers have been attracting more and more attention to deliver the desired properties to the advanced materials by the help of their precisely controlled compositions and architectures.<sup>1</sup> Understanding the specific multivalent carbohydrate-protein interactions is crucial to determine the structure-property relationships and to design accordingly the next generation of functional glycomaterials. Therefore, we investigate the structure-property relationships between the mammalian lectins and multivalent carbohydrate polymers, which may have applications for anti-adhesion therapy. Moreover, we have investigated the affinity of poly(mannose-methacrylate), helical glycopolypeptides, gp120, star shaped glycopolymers, and cyclodextrin centered glycopolymers with a selected mannose binding lectin (DC-SIGN) that exists on dendritic cells, using SPR technique.<sup>2-4</sup> Selected members of a glycopolymer library were used to demonstrate the interactions between DC-SIGN and mannose rich polymers. We extend this study to a broader set of polymers to examine the effect of chain length, end group, architecture, thermoresponsive block, and number of arms in the star shaped polymers on the lectin binding.



**Scheme 1.** Schematic representation of (a) linear GP, (b) helical glycopolypeptide, (c) glycoprotein 120 (gp120), (d) star shaped GP, (e) cyclodextrin centred GP, (f) SPR measurement on glycopolymer-DCSIGN competition in solution, (g) SPR response-time for the interaction of DC-SIGN & gp120.

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# Preparation of Polymeric Particles Containing Reduced Graphene Oxide by Miniemulsion Polymerization

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Graphene is an atomically thin two-dimensional form of carbon that has various functionalities (electrical, thermal, and mechanical properties). However, when graphene is used as filler in polymeric materials to enhance their properties, it is difficult to disperse in both a solid matrix or in a solvent due to strong Van der Waals interactions between graphene layers, resulting in aggregation.

Recently, imidazolium-based ionic liquids and poly(ionic liquid)s (PIL) prepared by the polymerization of ionic liquid monomer have received attention as a potential dispersant of graphene due to  $\pi$ - $\pi$  interactions between imidazolium rings and graphene sheets. Graphene dispersions could be obtained in various solvents using PIL as dispersant because of the possibility to conveniently control the solubility of PIL by anion exchange. Additionally, graphene oxide (GO) can be a precursor to composite materials containing reduced graphene oxide (rGO), as the functionality of rGO is similar to that of graphene.<sup>1</sup>

In previous work, we demonstrated the preparation of PIL particles containing rGO by miniemulsion polymerization using an ionic liquid monomer as rGO-dispersant.<sup>2</sup> However, from the point of view of industrial applications, that should be applicable to conventional monomers such as styrenic and methacrylate monomers. In the present work, the preparation of rGO dispersions stabilized by poly(ionic liquid) in a conventional vinyl monomer, and miniemulsion polymerization of the conventional monomer containing rGO was performed to yield composite nanoparticles.<sup>3</sup>

Prior to miniemulsion polymerization, the dispersibility of PIL/rGO in various monomers was investigated, however, PIL/rGO precipitated instantaneously in most common vinyl monomers. To improve the solubility of PIL in monomers, a statistical copolymer of ionic liquid monomer and methyl methacrylate (MMA) was prepared. rGO could be dispersed in ethyl methacrylate (EMA) using the copolymer by ultrasonication, however, polymeric nanoparticles containing rGO were not obtained by miniemulsion polymerization of EMA utilizing statistical copolymer as stabilizer, in which most rGO appeared to have precipitated. This suggested the rGO stability in monomer using this statistical copolymer was insufficient. On the other hand, polymeric nanoparticles containing rGO have been successfully prepared by miniemulsion polymerization of MMA utilizing a block copolymer of the ionic liquid monomer and MMA, which was prepared by RAFT (Fig. 1a). As shown in Figure 1b, the particle size distribution before acetone treatment comprised nanometer-sized particles ( $D_n = 160$  nm), consistent with the TEM imaging. After acetone treatment, significantly smaller nanoparticles ( $D_n = 27$  nm) were detected, consistent with single rGO sheets.

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## Recent Developments in the Field of Light-Controlled Radical Polymerizations

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The introduction of light as an external stimulus in controlled radical polymerizations has been extensively studied in recent years due to the supplementary advantages which can be brought, such as temporal and spatial controls. In this context, two mechanisms recently developed in our group will be presented.

The first one is a photocatalyzed ATRP involving a Cu-based complex with phenanthroline ligands.<sup>8,9,10</sup> The mechanism involves the excited state derived from the activator form of the complex, which is original for a photocatalyzed Cu-based ATRP (Figure 1). This catalytic cycle was completed by an additional regeneration of the activators mechanism, using an amine or a functional comonomer as the reducing agent.

The second one is based on a RAFT mechanism, considering that several thiocarbonylthio compounds were already commonly used as photoinitiators, before being considered as RAFT agents. We report the use of a RAFT agent designed to exploit this dual reactivity, leading to an efficient photoRAFT mechanism based on the reversible photolysis of the terminal functionality. Interestingly, this photodissociation of the chain-ends can be purposely enhanced so as to exploit polymers synthesized by RAFT as efficient and versatile macroinitiators.<sup>11</sup> Narrow polydispersities were reached in both cases either for methacrylates or acrylates using soft LEDs emitting either in the blue or green wavelengths range. Additionally, efficient temporal controls were achieved by simply switching on/off the light source. Light-triggered re-initiation and chain-extension were also successfully applied for the synthesis of block copolymers.

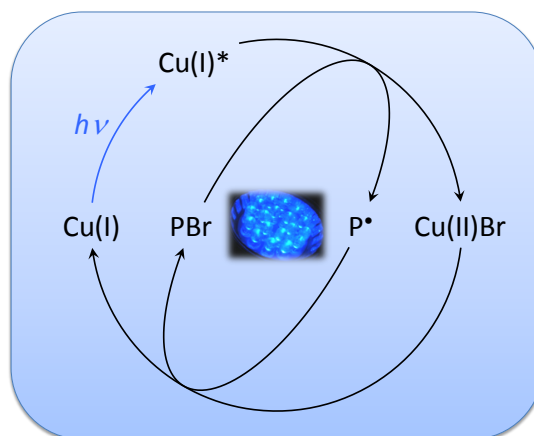


**Figure 1.** Photocatalyzed ATRP based on an oxidative quenching mechanism

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# Bioinspired Materials Bridging the Length Scales

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Self-assemblies bridging the length scales to allow structural hierarchies are amply used in Nature towards achieving materials with functional properties. Here selected examples are given by combining colloidal level structural units and polymeric units. Cellulosic materials have gained revived interest, due to their wide availability as sustainable source for polymers. Therein nanocelluloses as colloidal level fibrillar objects of high mechanical properties hold a particular promise, and they encourage to identify also novel high tech applications beyond those traditionally considered for cellulose. Recently Toivonen et al showed that cellulose nanofibers (CNF) allow transparent aerogel materials with high strength where a method was found which suppressed the need for the classically required supercritical drying or freeze-drying, paving ways towards scalability.<sup>1</sup> The films could be coated by carbon nanotubes to allow conducting transparent aerogel films, which could be a starting point for future devices. A typical problem related to nanocellulose has been moisture sensitivity. Toivonen et al<sup>2</sup> and Mertaniemi et al<sup>3</sup> also prepared nanocellulose films and fibers with increased strength also under wet conditions, which allows new application potential. This opened even possibilities for nanocellulose-based surgery threads with promoted cell adhesion.<sup>3</sup> On the other hand, humidity can also be harnessed to allow actuation, even without incorporation of the common bilayer architecture.<sup>4</sup> Biological inorganic/organic hybrids can allow uncommon combination of high mechanical properties, where a prototypical example is nacre. Previously an evaporation-induced self-assembly method was indentified by Walther et al to prepare nacre-mimetic films with high strength and stiffness.<sup>5</sup> Morits and Verho et al have recently found a self-assembly route towards bulk materials mimicking nacre, showing deflected crack propagation indicating toughness and where the fracture toughness starts to approach to that of nacre.<sup>6</sup> Finally we discuss biomimetic optical properties. Structural colours are inherent in several animals, plants, and organisms, involving chirality of the overall structure. Instead of the macroscopic chiral assembly, we investigate photonic properties of individualized nanoscale objects. Majoinen et al showed that the inherent chiral twisting of individualized cellulose nanocrystals (CNC) in aqueous medium can be harnessed to show chiral plasmonic functions upon binding gold nanoparticles.<sup>7</sup> We expect that the concept is general, suggesting to use different chiral and helical templates for plasmonic materials.

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## Alginate-based materials

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Alginate is an anionic co-polymer composed of (1, 4)-linked  $\beta$ -D-mannuronate (M) and  $\alpha$ -L-guluronate (G) residues and is sourced from nature (eg. seaweed).<sup>1</sup> This polymer has been comprehensively studied for use in various applications in biomaterials and food science because of its reported biocompatibility, non-toxicity and mild gelation property.<sup>1</sup>

The work presented will cover various aspects of developing alginate-based materials including evaluation of the bio-distribution of alginate (Fig. 1A), synthesis of sulfated alginate, encapsulation of bovine lactoferrin in nano-aggregates and micro-gel particles,<sup>2</sup> as well as fabrication and characterisation of alginate hydrogels (Fig. 1B&C)<sup>3</sup> including composite photo-polymerised hydrogels.

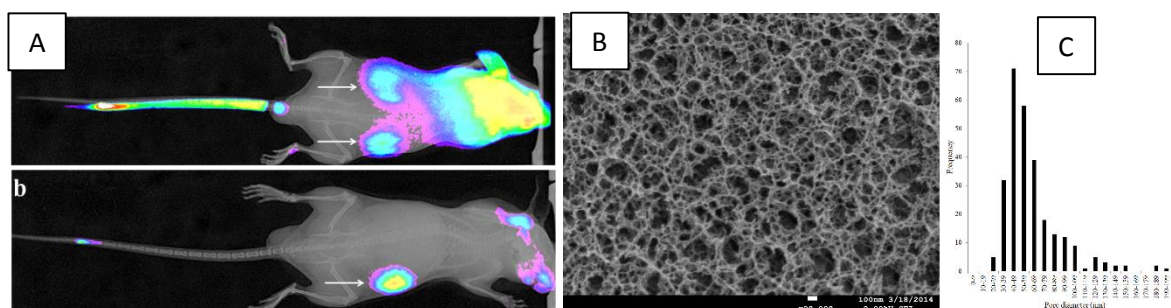


**Figure 1.** A: After 2 days of sample administration, the presence of circulating alginate *in vivo* could still be observed; B: Cryo-SEM image of ionically crosslinked alginate hydrogel; C: Pore size distribution obtained from image in B.

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# Amphiphilic Polymer Conetworks: Structural Evolution During Aqueous Gelation

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Amphiphilic polymer conetworks (APCN) are important materials attracting increasing attention due to their properties and potential applications.<sup>1</sup> These materials comprise hydrophilic and hydrophobic segments, covalently interconnected to a network. When placed in water, the hydrophobic segments rearrange themselves in order to minimize contact with the aqueous environment, leading to phase separation on the nanoscale. This internal organization makes APCNs ideal materials for use in antifouling coatings, modern soft contact lenses, matrices for phase transfer reactions with applications in bio- and organocatalysis, and the fabrication of gas and optical sensors.

Due to the presence of the cross-links, and the inhomogeneity of the polymer chains, the morphologies exhibited by APCNs are not perfect, displaying only short-range order and blurred interfaces. To improve the order in the morphologies of nanophase separated APCNs, we optimized their synthesis by employing block copolymer building blocks and relatively low loadings of cross-linker. However, a non-ideality in this system was the broad distribution in the core functionality. Despite this defect, the system internally self-assembled, resulting in a structure comprising lamellae with relatively long-range order as confirmed via both scattering and microscopy techniques.<sup>2</sup>

This lecture will present a small-angle neutron scattering (SANS) study to kinetically follow the formation of APCNs in deuterium oxide, starting from concentrated solutions of well-defined polymeric building blocks. These building blocks are reactive and lead to APCN formation within 1–2 hours, a time scale that allows monitoring by SANS. Structural transitions during APCN formation were observed and will be discussed in relation to the composition, concentration and architecture of the network building blocks, and the reaction conditions.

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# **Ab initio based microkinetic modelling as a tool to understand and improve controlled radical polymerization processes**

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Precision control of the detailed chemical structure of each individual macromolecule is a key challenge to synthesize next-generation functional polymeric materials that meet predefined end-use properties. Controlled polymerization techniques are ideally suited to meet this challenge.

Full control of the structural characteristics of the individual macromolecules implies that throughout the polymerization the reaction rates are carefully controlled which introduces the need for dynamic synthesis protocols that allow an instantaneous control of reactant concentrations and temperature. This instantaneous rate control pertains not only to initiation, propagation and termination reactions but, more importantly, also includes reactions involving mediating agents and possible side reactions that might lead to structural defects. Hence, a fundamental and quantitative understanding of the reaction mechanism and the availability of accurate values of the relevant rate coefficients are indispensable.

This contribution highlights the application of computational chemistry as a complement to experiment in the development of microkinetic modelling tools that enable to improve control over the polymer microstructure. The potential of this approach to design tailor-made dynamical synthesis protocols is illustrated for reversible addition-fragmentation chain transfer (RAFT) polymerization, nitroxide mediated copolymerization and for the synthesis of well-defined copoly(2-oxazolines) via cationic ring-opening polymerization (CROP).

# Photoinducing Dynamic Covalent Chemistry in Covalent Adaptable Networks

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With a wide array of formulations, chemistries and mechanisms to choose from, the science of making a stimuli responsive material has been studied and documented extensively over the past decade. In particular, the benefits of forming smart polymers that respond to irradiation have been widely studied as the ability to trigger a response based on light exposure provides spatiotemporal control to the process. Herein, we focus on the reaction processes and secondary functionalities that are incorporated into networks to enable tunable dynamic covalent chemistry within these polymer systems. Shape memory polymers (SMPs) with network properties such as high and tunable glass transition temperatures, multiple shape memory transitions, shape memory composites and two-way transition materials are recent examples of new directions that have been undertaken within the broad field of ‘smart’ materials. Among these new directions, the ability to reshape and resize materials repeatedly is particularly appealing. Such shape manipulations are desirable from the standpoint of SMPs applications, especially those requiring the excellent mechanical properties of thermosets. The concept of photoinduced addition-fragmentation chain transfer (AFCT) enables Covalent Adaptable Networks (CANs) in which the presence of photochemically active moieties can be used to induce network rearrangement in a controlled manner. Adopting this concept, it is demonstrated that by incorporating the photochemically active monomers, crosslinked polymer networks can then be induced to repeatedly undergo covalent bond rearrangement, and hence bulk material rearrangement. Further, for other, non-radical mediated catalytic processes that enable dynamic covalent chemistry within CANs, it is also possible to use light as a switch to turn on or off the dynamic covalent chemistry by forming or eliminating the catalyst, respectively. The fundamental mechanisms, benefits and opportunities associated with these photoinducible smart materials will be discussed.

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## Recent advances in $^{19}\text{F}$ MRI agents for detection of diseased tissue and cell tracking

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Recently there has been intense interest in fluorinated molecules to allow tracking of therapeutic particles and cells in vivo. The motivation for this is the very high selectivity of the  $^{19}\text{F}$  imaging experiment, since unlike in  $^1\text{H}$  MRI, the body does not contain a confounding fluorine background signal. In principle therefore, highly-selective  $^{19}\text{F}$  images can be superimposed on high-resolution anatomical  $^1\text{H}$  images, allowing tracking of suitably-labelled cells or biomarker molecules. Here we describe the motivation for the development of new partly-fluorinated copolymers with outstanding potential as  $^{19}\text{F}$  MRI imaging agents, and in particular as agents sensitive to the biological environment.<sup>1-10</sup> A second class of materials for the labelling of potentially-therapeutic stem cells will be described. These are statistical copolymers containing perfluoro-polyether side chains within a PEG-acrylate backbone. Measurement of cell uptake and imaging performance will be described.

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<sup>8</sup> C. Zhang, H. Peng, S. Puttick, J. Reid, S. Bernardi, D. J. Searles, A.K. Whittaker, *Macromolecules*, **2015**, 48, 3310.

<sup>9</sup> K. Wang, H. Peng, K.J. Thurecht, S. Puttick and A. K. Whittaker, *Polym. Chem.*, **2016**, DOI: 10.1039/C5PY01707F.

<sup>10</sup> C. Zhang, S.S. Moonshi H. Peng, S. Puttick, J. Reid, S. Bernardi, D. J. Searles, A.K. Whittaker, *ACS Sensors*, **2016**, 0.1021/acssensors.6b00216.

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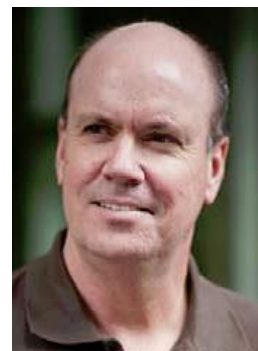
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2006- Group Leader, AIBN, UQ

2016- Deputy Director International, AIBN, UQ

Research interests: polymer physical chemistry, translational polymer science



# Integration of DNA with polymers

Amanda V. Ellis

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The coupling of DNA with polymers offers an entirely new approach to developing unique applications in genotyping, diagnostics, synthetic biomaterials and enhanced polymerase chain reactions. This talk will introduce the fabrication of polymer/nucleotide polymers both with 3' or 5' nucleotides. We show attempts to ligate these nucleotides in order to use the polymer template in DNA amplification.

Thermo-responsive polymers, such as those based on n-isopropyl acrylamide (NIPAAm), have been studied extensively for bioseparation of dilute biomarkers due to their sharp phase transition at the biologically relevant lower critical solution temperature (LCST) of 32 °C. Circulating nucleic acids (cNAs) are emerging as potential biomarkers for early stage cancer diagnosis and treatment of residual disease. However, they are often found in very low levels circulating in a patient's blood and as a result require substantial enrichment to be useful in a diagnostic assay. cNAs are considered to be a dilute biomarker. To successfully detect cNAs tuneable stimuli response (TSR) DNA capture probe (DCP) polymers, using pNIPAAm, are being investigated for their utility in sensitive diagnostic applications. Here, we use RAFT polymerisation to design; tuneable thermo-responsive polymers that have a site specific moiety, a strained alkyne, used for conjugation to model DNA probes. The specificity and simplicity of the strained "click" reaction has the added benefit of eliminating the need for metal catalysts; normally required for traditional cycloaddition chemistry. Tuning of the LCST of pNIPAAm was achieved by incorporation of hydroxyl methyl acrylamide (HMAAm) (0-12.5%) monomer to make a range of pNIPAAm-co-HMAAm copolymers. The polymers displayed a tuneable LCST (**Fig 1(a)**). After functionalisation of the polymer with a strained alkyne, DNA capture probes were attached (**Fig 1(b)**) via a strained alkyne-azide reaction, to enable hybridisation, separation and enrichment of specific cNA transcripts with temperature modulation (**Fig 1(c)**).

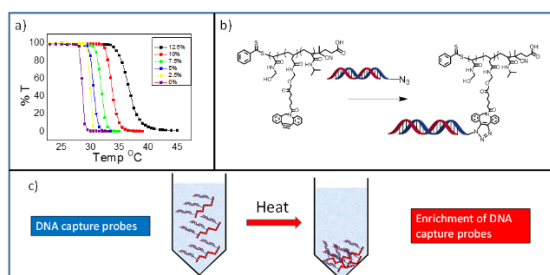


**Figure 1.** (a) LCST measurement of 0-12.5% pNIPAAm-co-HMAAm, (b) Conjugation of strained alkyne functionalised (pNIPAAm-co-HMAAm) with DNA capture probe via alkyne-azide cycloaddition and (c) Schematic diagram of biomarker enrichment process using pNIPAAm-co-HMAAm-co-strained alkyne-DNA capture probe stimuli response polymer.

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# Exploiting Photochemical Selectivity in Macromolecular Synthesis

Christopher Barner-Kowollik<sup>1,2</sup>

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Installing chemical functionality at precise locations within complex macromolecules as well as onto three dimensional direct laser written constructs in the mildest possible fashion (visible light) and without any catalyst becomes possible with powerful light driven, quantitative ligation protocols.<sup>1-6</sup> Advanced light induced processes enable the parallel encoding of multiple molecules onto predefined locations on 3D microstructures in one step<sup>7</sup> as well as the wavelength dependent addressing of specific parts of a macromolecule for its highly orthogonal functionalization (□-orthogonal photochemistry).<sup>8</sup> The lecture will highlight how macromolecular ligations can be directed in their selectivity by the application of photonic fields,<sup>9</sup> how light can be used as an orthogonal trigger in macromolecular chain folding,<sup>10</sup> how the high efficiency of light driven chemistry can be exploited for the synthesis of sequence defined macromolecules<sup>11</sup> or explore – time permitting – how diffraction unlimited laser lithography may become possible based on photo-click chemistry concepts.<sup>12</sup>

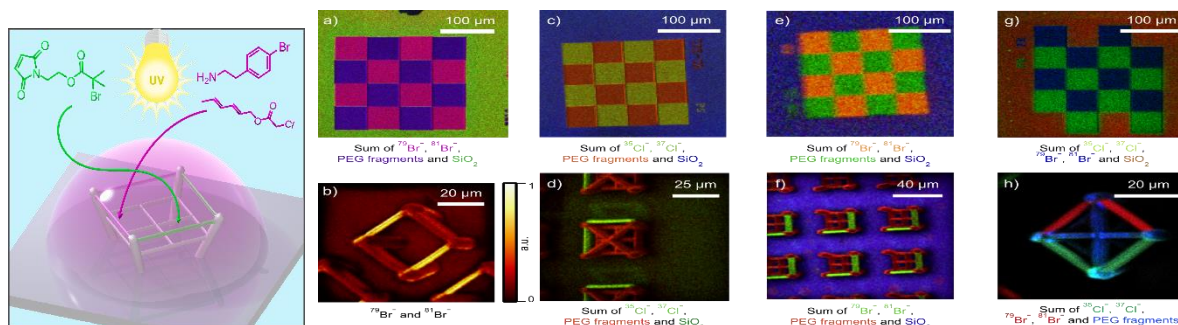


**Figure 1.** Encoding three dimensional direct laser written scaffolds in specific pre-defined positions with chemical functionality via a self-directed light induced molecular sorting process (ToF-SIMS images shown).<sup>7</sup>

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## Christopher Barner-Kowollik

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Since 11/15 Director of the Centre of Excellence (Sonderforschungsbereich) (SFB 1176) ‘Molecular Structure of Soft Matter’ at the Karlsruhe Institute of Technology (KIT)

Since 06/13 Professor (fractional) of **Materials Science**, Queensland University of Technology (QUT)

Since 07/08 Full Professor of Macromolecular Karlsruhe Institute of Technology (KIT)

11/06 – 07/08 Full Professor and ARC Australian Professorial Fellow (2006–2011) at the University of New South Wales, Centre for Advanced Macromolecular Design

Research interests: Light-triggered orthogonal chemistries, novel synthetic approaches to complex macromolecular designs, spatially and temporally controlled surface coding (2D, 3D), polymerization mechanisms, advanced polymer characterization



# Functional Nanomaterials *via* Click Chemistry: From Polymeric Nanoparticles to Magneto-Induced Drug-Delivery Systems

Laurent Fontaine

Institut des Molécules et Matériaux du Mans, UMR 6283 CNRS – Université du Maine, Av. Olivier Messiaen - 72085 LE MANS - FRANCE

This presentation will describe our work to design and to prepare new functional organic (polymer-derived) and hybrid (magnetic iron oxide) nanoparticles by using orthogonal click chemistry reactions with emphasis on our most recent results using azlactone, alkyne, and furan as ‘clickable’ moieties.

The azlactone (oxazolone) ring reacts under mild conditions with primary amines without the formation of a byproduct, a reaction that can be regarded as proceeding with the advantages associated with conventional ‘click’-type reactions.<sup>1</sup> We have investigated azlactone-functionalized polymers as a platform for the design of new reactive thermoresponsive nanoparticles<sup>2</sup> and hybrid functional magnetite nanoparticles.<sup>3</sup>

By using a versatile methodology that employs a combination of orthogonal click reactions, namely copper-catalyzed azide-alkyne cycloaddition (CuAAC) and thermoreversible Diels-Alder reaction, we designed a new class of functional iron oxide magnetic nanoparticles showing unprecedented, active control over drug release using hyperthermia effects. Upon alternating magnetic field exposure, the retro-Diels-Alder reaction enables the release of a conjugated drug without any significant heating of the medium. This method has the potential to improve hyperthermia therapies by expanding the range of polymers and molecules (drugs, dyes, etc) that can be used. The so-obtained functional nanoparticles may become important tools for preparing the next generation of controlled-release devices and nanomedicines for *in vitro* and *in vivo* applications.

*Co-workers and students who have been contributing to this work are gratefully acknowledged.*

<sup>1</sup> H. T. Ho, M. E. Levere, D. Fournier, V. Montebault, S. Pascual, L. Fontaine, *Aust. J. Chem.* **2012**, 65, 970-977.

<sup>2</sup> H. T. Ho, M. E. Levere, S. Pascual, V. Montebault, A. Caruso, N. Casse, L. Fontaine, *Polym. Chem.* **2013**, 4, 675-685.

<sup>3</sup> Y. Prai-In, K. Tankanya, B. Rutnakornpituk, U. Wichai, V. Montebault, S. Pascual, L. Fontaine, M. Rutnakornpituk, *Polymer* **2012**, 53, 113-120.

<sup>4</sup> T. T. T. N’Guyen, H. T. T. Duong, J. Basuki, V. Montebault, S. Pascual, C. Guibert, J. Fresnais, C. Boyer, M. R. Whittaker, T. P. Davis, L. Fontaine, *Angew. Chem. Int. Ed. Engl.* **2013**, 52, 14152-14156.

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| 1988       | Ph.D. Macromolecular Organic Chemistry (Université du Maine)                                                           |
| 1990-2000  | Assistant Professor (Prof. JC. Brosse group)                                                                           |
| Since 2000 | Full Professor (Université du Maine, Le Mans)                                                                          |
| Since 2004 | Deputy-Director of “Institut des Molécules et Matériaux du Mans” (IMMM; UMR 6283 CNRS – Université du Maine, Le Mans). |

Research interests: Macromolecular engineering using controlled / « living » polymerization and application to the synthesis of functional and reactive homopolymers, block and graft copolymers: Ring-Opening Metathesis Polymerization (ROMP), reversible deactivation radical polymerization (ATRP, RAFT); Biomedical applications of polymers; Bioconjugation.

# Controlled polymer architectures for probing cell uptake mechanism

Sébastien Perrier<sup>1,2</sup>

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Chemists are remarkably proficient at directing the synthesis of small molecules, but fine-tuning the structures of large molecules, such as those found in polymers, is far more taxing. Despite many years of research, the field of macromolecular engineering i.e. the preparation of large molecules with strict control over their size and chemical groups has many mountainous challenges yet to overcome. Nature provides endless examples of precisely engineered macromolecules; proteins, for instance, which contain amino-acid side-chains that are accurately positioned, often in a way that determines the proteins' roles. Synthetic chemists have tried to recreate nature's exceptional control over macromolecules, and in so doing they have designed new materials with precisely defined structures, for use in applications ranging from materials to medicine. The lecture will describe our use of complex polymeric architectures designed by controlled polymerisation techniques to probe cellular mechanisms. In particular, we are interested in the design of drug delivery vectors to optimise their uptake by cells.

## Selected Publications

- Gody, G.; Zetterlund, P.B.; Perrier, S.; Harrison, S., *Nat. Commun.* **2016**, 7, 10514, 10.1038/ncomms10514
  - Danial, M.; My-Nhi Tran, C.; Jolliffe, K.A. Perrier, S. *J. Am. Chem. Soc.* **2014**, DOI: 10.1021/ja5024699
  - Danial, M.; My-Nhi Tran, C.; Young, P.G.; Perrier, S.; Jolliffe, K.A. *Nat. Commun.*, **2013**, 4:2780 DOI: 10.1038/ncomms3780
  - Gody, G.; Maschmeyer, T.; Zetterlund, P.B.; Perrier, S. *Nat. Commun.*, **2013**, DOI: 10.1038/ncomms3505
  - Chapman, R.; Warr, G.G.; Jolliffe, K.A.; Perrier, S. *Adv. Mater.*, **2013**, 25, 1170–1172.
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  - Perrier, S.; Takolpuckdee, P.; Mars, C. A., *Macromolecules* **2005**, 38, (38), 2033-2036.
- ## Recent Reviews
- Zetterlund, P.; Thickett, S.; Perrier, S.; Bourgeat-Lami, E.; Lansalot, M. *Chem. Rev.*, **2015**, DOI: cr-2014-00625k.R2
  - Cobo, I.; Li, M.; Sumerlin, B. S.; Perrier, S. *Nature Mat.* 2015, 14 (2), 143-159 Semsarilar, M.; Perrier, S. *Nature Chem*, **2010**, 2, 811-820
  - Boyer, C.; Bulmus, V.; Davis, T.P.; Ladmiral, V.; Liu, J.; Perrier, S. *Chem. Rev.*, **2009**, 109 (11), 5402-5436.
  - Perrier, S.; Takolpuckdee, P., *J. Polym. Sci., Part A: Polym. Chem.* **2005**, 43, (22), 5347-5393.

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Personal History:

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| 2002         | PhD, The University of Warwick                                      |
| 2002         | Postdoctoral Fellow, The University of New South Wales              |
| 2002-2005    | Lecture, The University of Leeds                                    |
| 2005-2007    | Senior-Lecture, The University of Leeds                             |
| 2007-2012    | Associate Professor, The University of Sydney                       |
| 2012         | Professor, The University of Sydney                                 |
| 2012-present | Professor, University of Warwick, UK / Monash University, Australia |
- Research interests: polymer synthesis, nanostructured materials, nanomedicine



# Structured polymer nanoparticles – access to new hybrid materials and nanomedicines

Markus Mueller<sup>1,2,3</sup>

<sup>1</sup> School of Chemistry, The University of Sydney, Australia

<sup>2</sup> Key Centre for Polymers and Colloids, The University of Sydney, Australia

<sup>3</sup> Australian Institute for Nanoscale Science and Technology, Sydney, Australia.

The possibility to precisely engineer at the nanoscale enables the synthesis of materials with tailored chemical composition and physicochemical properties. Within this space, the modular synthesis of cylindrical polymer brushes provides a new and versatile class of materials that are very powerful in molecular templating and sensing,[1,4-7] and which emerge as promising polymer nanomedicines.[2,3,5] Our research focusses on these nanomaterials which we are using to establish new ways of assembling meta and hybrid materials as well as design the next-generation of smart drug delivery devices.

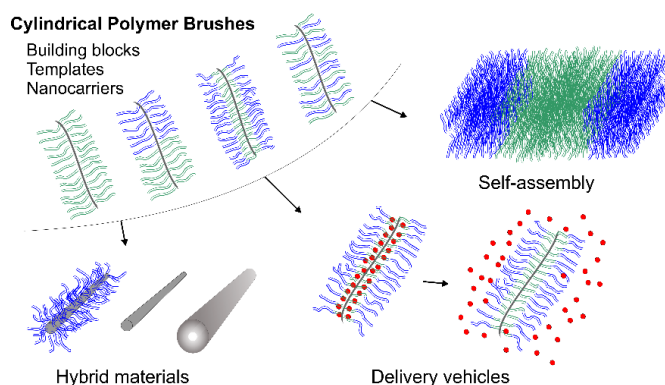


**Figure 1.** Various applications of cylindrical polymer brushes.<sup>1</sup>

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- <sup>1</sup> M. Müllner, A.H.E. Müller; *Polymer* **2016**, DOI: 10.1016/j.polymer.2016.03.076.
- <sup>2</sup> M. Müllner; *Macromol. Chem. Phys.* **2016**, DOI: 10.1002/macp.201600086.
- <sup>3</sup> M. Müllner, D. Mehta, C.J. Nowell, C.J.H. Porter; *Chem. Comm.* **2016**, DOI: 10.1039/C6CC00447D.
- <sup>4</sup> R. Verduzco, X. Li, S.L. Pesek, G.E. Stein; *Chem. Soc. Rev.* **2015**, *44*, 2405.
- <sup>5</sup> M. Müllner, S.J. Dodds, T. Nguyen, D. Senyschyn, C.J.H. Porter, B. Boyd, F. Caruso; *ACS Nano* **2015**, *9*, 1294.
- <sup>6</sup> M. Müllner, T. Lunkenbein, J. Breu, F. Caruso, A.H.E. Müller; *Chem. Mater.* **2012**, *24*, 1802.
- <sup>7</sup> M. Müllner, J. Yuan, S. Weiss, A. Walther, M. Förtsch, M. Drechsler, A.H.E. Müller; *J. Am. Chem. Soc.* **2010**, *132*, 16587.

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Personal History:

- 2012 PhD, University of Bayreuth, Germany.
- 2012-2015 McKenzie Fellow, The University of Melbourne, Australia.
- Since 2015 Lecturer, University of Sydney, Australia, School of Chemistry.
- Since 2016 Foundation Project Leader at the Australian Institute for Nanoscale Science and Technology.

Research interests: polymer architectures, nanomedicine, hybrid materials  
sensors, self-assembly



# Synthesis and Characterization of Polymer Semiconductors: Shining New Light on Plastics

George Vamvounis

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Polymer Electronics has taken science by storm because of their potential to revolutionize today's technologies such as displays,<sup>1</sup> electronics,<sup>2</sup> mechanical actuators,<sup>3</sup> solar cells,<sup>4</sup> and sensory devices.<sup>5-6</sup> In fact, Polymer Electronics are becoming reality through companies such as LG Electronics, Samsung (OLEDs), Heliotech (Solar Cells), Plastic Logic (OFET, RFID), Fido (Explosive Sensors). Understanding the properties of organic semiconductors, as dictated by their chemical structure, is pivotal in producing more efficient devices. For instance, tuning the electron affinity of the macromolecular material can have a dramatic effect on the sensing of various analytes like nitroaromatic compounds. In this seminar, I will discuss the synthesis of new conjugated polymers and how they are applied to the next generation of microelectronic devices like Sensors, LEDs and Transistors (Fig. 1).

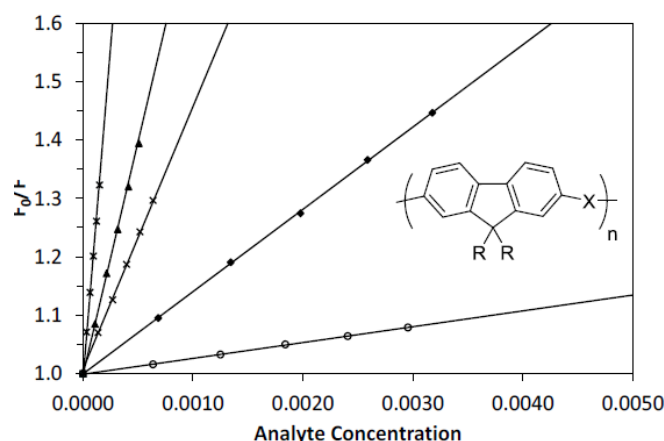


**Figure 1.** Stern-Volmer Quenching using various analytes of fluorene-based polymer semiconductor. These results demonstrate both sensitivity and selectivity.

<sup>1</sup> G. Vamvounis, S. Holdcroft, *Advanced Materials* **2004**, 16, 716-719

<sup>2</sup> G. Vamvounis, Almantas Pivrikas, Paul E. Shaw, Paul L. Burn, *Thin Solid Films* **2013**, 548, 190-194.

<sup>3</sup> D. Gendron, A. Ansaldo, G. Bubak, L. Ceseracciu, G. Vamvounis and D. Ricci, *Composites Science and Technology* **2015**, 117, 364-370.

<sup>4</sup> G. Vamvounis, C. R. Glasson, E. J. Bieske, V. Dryza, *Journal of Materials Chemistry C* **2016**, in press: DOI: 10.1039/C6TC01584K

<sup>5</sup> G. Vamvounis, N. Sandery, *Australian Journal of Chemistry* **2015**, 68, 1723-1726.

<sup>6</sup> G. Vamvounis, M. Fuhrera, A. Koizumia, M. Gao and S. Hu, *submitted for publication*.

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2007-2014 Sr. Research Fellow, Centre for Organic Photonics and Electronics, The University of Queensland

2005-2007 KAMI Post-doctoral Research Fellow, Royal Institute of Technology (KTH)

Research interests: Polymer Chemistry, Photophysics, Photochromism, Solar Cells, Transistors, LEDs

## Nitroxide-containing materials for biofilm remediation

A. D. Verderosa,<sup>1</sup> J. P. Allen,<sup>1</sup> H. Woehlk<sup>1,2</sup> C. de la Fuente-Núñez,<sup>3,4</sup> S. C. Mansour,<sup>3</sup> C. Lang,<sup>1</sup> N. Boase,<sup>1</sup> R. E. W. Hancock,<sup>3</sup> C. Barner-Kowollik<sup>1,2</sup> L. Barner<sup>1,2</sup> and K. E. Fairfull-Smith<sup>1</sup>

<sup>1</sup>Faculty of Science and Engineering, Queensland University of Technology, Brisbane, Australia.

<sup>2</sup>Institut für Technische Chemie und Polymerchemie, Karlsruhe Institute of Technology, Karlsruhe, Germany.

<sup>3</sup>Department of Microbiology and Immunology, University of British Columbia, Vancouver, Canada.

<sup>4</sup>Department of Biological Engineering, Department of Electrical Engineering and Computer Science, Massachusetts Institute of Technology, Massachusetts, United States of America.

The adhesion of bacteria to surfaces and their subsequent ability to aggregate into colonies called biofilms is a significant global problem in numerous applications, particularly as biofilms display an inherent resistance to antimicrobial agents. Low concentrations of the diatomic free radical nitric oxide has been shown to disperse bacterial biofilms to free swimming planktonic bacteria,<sup>1</sup> which has led to the development of coatings that release nitric oxide. However, the non-systemic delivery of gaseous nitric oxide remains challenging as nitric oxide is an extremely reactive molecule with a very short half-life. As an improved strategy, we have recently shown that nitroxides (persistent and stable free radical species that are sterically hindered versions of nitric oxide) can inhibit bacterial biofilm formation and disperse existing biofilms (Figure 1).<sup>2</sup> This presentation will report our current work towards the design and development of novel chemistries to immobilise and/or release nitroxides from surface coatings and microparticles as an innovative strategy for combating biofilm growth in a range of applications.

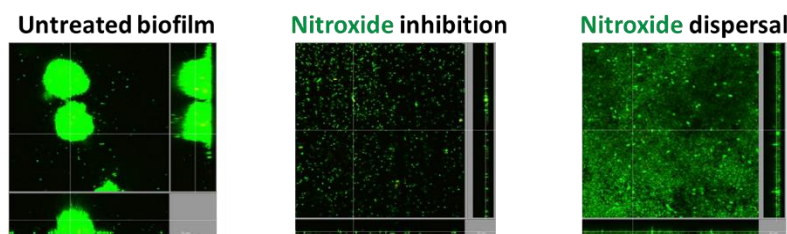


**Figure 1.** Confocal microscopy images showing inhibition and dispersal of *Pseudomonas aeruginosa* biofilms by nitroxides (live cells stained green).

<sup>1</sup> N. Barraud, D. J. Hassett, S. Hwang, S. Kjelleberg and J. S. Webb, *Journal of Bacteriology*, **2006**, 188, 7344-7353.

<sup>2</sup> C. de la Fuente-Núñez, F. Reffuveille, K. E. Fairfull-Smith and R. E. W. Hancock, *Antimicrobial Agents and Chemotherapy*, **2013**, 57, 4877-4881.

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| 2004       | PhD, Griffith University                                               |
| 2004-2006  | Postdoctoral Fellow, University of Sheffield                           |
| 2006-2009  | Postdoctoral Fellow, Queensland University of Technology               |
| 2009-2012  | Vice-Chancellor's Research Fellow, Queensland University of Technology |
| 2012-2014  | Senior Lecturer, Queensland University of Technology                   |
| Since 2015 | ARC Future Fellow and A/Prof, Queensland University of Technology      |

Research interests: Anti-biofilm coatings, fluorescent probes, nanosensors for drug delivery and imaging

## Synthesis, evaluation and application of arsenical-linked polymer bioconjugates

Paul Wilson<sup>1,2</sup>, Kristian Kempe<sup>1,2</sup>, Michael R. Whittaker<sup>1,2</sup>, David M. Haddleton<sup>1,2</sup>, Thomas P. Davis<sup>1,2</sup>

<sup>1</sup>University of Warwick, Department of Chemistry, Coventry, West Midlands, CV4 7AL, UK.

<sup>2</sup>ARC Centre of Excellence in Convergent Bio-Nano Science & Technology, Monash Institute of Pharmaceutical Sciences, Monash University, Parkville, Victoria 3052, Australia

(In)organic arsenic exists as a dichotomous enigma between toxicity and therapy in (bio)chemistry and medicine. Despite being a well-known contaminant of base water supplies, it is an essential dietary element and has historically been employed to treat malignancies, most notably, and recently, in blood born malignancies such as acute promyelocytic leukaemia (APL).<sup>1</sup> Arsenic can exist in two biologically relevant oxidation states, trivalent arsenous acid (As(III)) and pentavalent arsenic acid (As(V)). The toxicity and therapeutic potential associated with arsenic is dominated by trivalent arsenic (As(III)) due to its high affinity for thiols which are present in variable concentrations in the intra- and extracellular milieu. In short peptides, larger functional proteins/enzymes and small molecule cofactors, thiols can exist either as free thiols, or as intra- or intermolecular disulfides. It has been postulated that the affinity of As(III) for thiols is enhanced for chelating vicinal or neighbours-through-space (proximal) dithiols,<sup>2</sup> such as those presented by naturally occurring disulfide bonds.

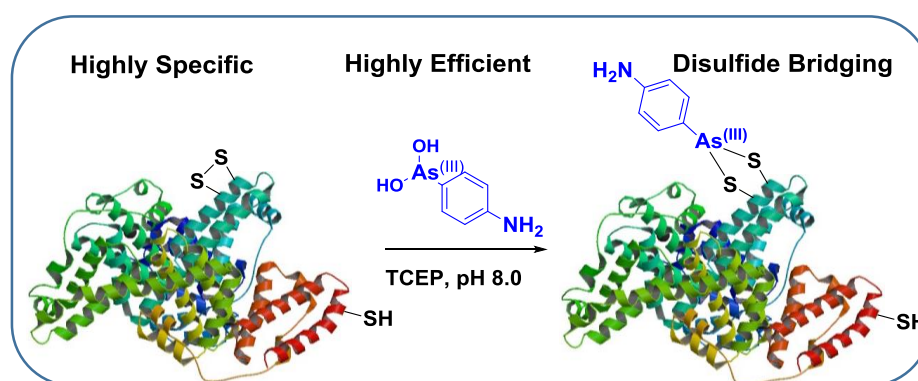


**Figure 1.** Highly specific conjugation of trivalent *p*-aminoarsenous acid to reduced BSA.

Conceptually this has been investigated using trivalent *p*-aminoarsenous acid and reduced BSA as model arsenical and protein respectively (**Fig 1**).<sup>3</sup> The enhanced specificity of As(III) for disulfides was exemplified upon comparison with established thiol-reactive functional groups such as maleimide and dibromomaleimide. Polymeric arsenicals have been synthesised by aqueous single electron transfer living radical polymerisation (AqSET) and reversible addition-fragmentation chain transfer polymerisation (RAFT) using an As(V)-functional initiator and chain-transfer agent respectively. Reduction of the As(V)-functional chain end using glutathione, post polymerisation, furnishes As(III)-functional polymers capable of specifically rebridging disulfide bonds in proteins and peptides. Conjugation to salmon calcitonin (sCT) proceeds rapidly and stoichiometrically to yield stable sCT-polymer conjugates. Upon addition of stronger chelating dithiols (e.g. ethanedithiol, reduced lipoic acid) the sCT is released from the conjugate. This chemistry has been applied to the synthesis of peptide-polymer conjugates that target the somatostatin receptor, a GPCR which is over expressed in neuroendocrine tumours. Conjugates of Somatostatin and analogues Octreotide and Lanreotide have been prepared and the effects of conjugation site and polymer composition on receptor interactions and down-stream cell-signalling events have been investigated.

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### Paul Wilson

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## Biomaterials that promote endothelialization

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Blood coagulates when it contacts foreign surfaces, including biomaterials that are currently used for cardiovascular applications. The development of thrombus (a blood clot) on the material surface can result in failure of devices such as small diameter vascular grafts and artificial heart valves. Additionally, a piece of the clot may detach and cause issues downstream such as stroke. Researchers have attempted to develop blood compatible interfaces for decades, but no surface has achieved great clinical success.<sup>12</sup> Native vasculature is lined with a monolayer of endothelial cells (an endothelium). And this endothelium is what prevents coagulation of blood when traveling through native vasculature. Therefore, creating material systems that foster a confluent and functioning endothelial cell layer is an attractive strategy for generating devices with improved interactions with blood.<sup>2</sup>

Traditionally, mature endothelial cells are used for this purpose. However, a circulating population of endothelial precursor cells has recently been identified. These endothelial progenitor cells can be isolated through peripheral blood, expanded *ex vivo*, and generate large numbers of cells with endothelial phenotype. The outgrowth of the progenitor cells is referred to as endothelial colony forming cells (ECFC).

In this work, we illustrated that substrate topography can improve endothelial cell spreading and proliferation on a biomaterial surface.<sup>3</sup> We then proposed generating a surface that specifically attaches ECFC with the aim of

developing surfaces that can be rapidly endothelialized. In order to achieve this goal, we screened a phage display library to identify polypeptide ligands that bind with high specificity to ECFC but not other commonly occurring blood cell types. These peptides were then covalently immobilized onto a non-fouling biomaterial.<sup>4</sup> The resulting material was able to specifically bind the ECFC but not other cell types including mature endothelial cells (**Fig. 1**).<sup>5,6</sup>

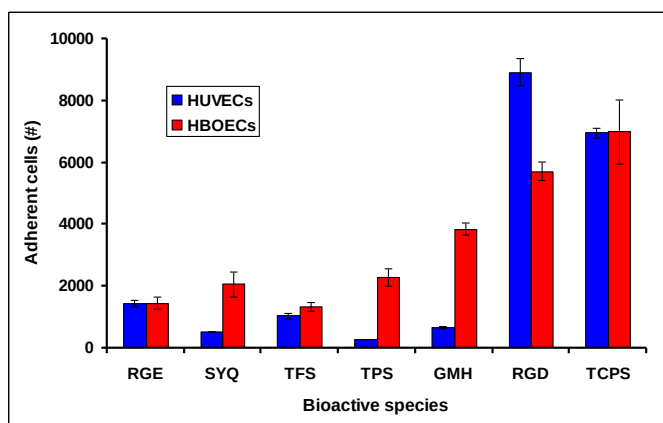


**Figure 1.** ECFC adhesion to non-fouling biomaterials functionalized with novel peptide ligands identified through phage display.

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# Shaping the Future of Polymer Molecular Weight Distributions

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A polymer's molecular weight distribution (MWD) has a profound impact on its properties, from material strength and viscosity to changes in the phase behavior of block copolymers.<sup>1-3</sup> Dispersity is the most common measure of MWD and is described as the ratio of weight-average ( $M_w$ ) to number-average ( $M_n$ ) molecular weights. Importantly, dispersity only provides information on the relative breadth of molecular weights in a sample and is not a comprehensive description of the molar quantities of each chain size.<sup>2</sup> The exact shape of a MWD has been proposed to have a strong influence on polymer properties;<sup>5,6</sup> however, this hypothesis remains relatively unexplored. This presentation will detail the development of a modular synthetic strategy that provides deterministic control over the  $M_n$ , breadth, and composition of polymer MWDs and will examine the influence of MWD shape on polymer properties.<sup>7</sup>

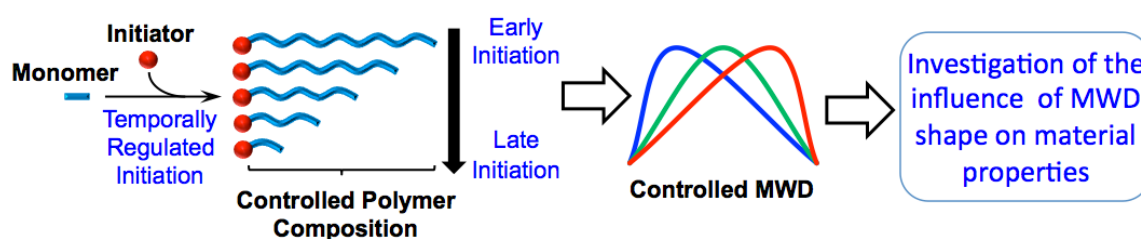


**Figure 1.** Deterministic control of polymer molecular weight distributions.

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# Spontaneous Zwitterionic Copolymerization of Cyclic Imino Ethers: First Steps to Biodegradable and Biocompatible 2-Oxazoline Polymers?

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Five and six membered cyclic imino ethers, also better known as oxazolines and oxazines are highly functional compounds which have been proven to be potent monomers in cationic ring-opening polymerisations (CROP) to yield poly(2-oxazoline)s and poly(2-oxazine)s, respectively.<sup>1</sup> In particular poly(2-methyl-2-oxazoline) (PMeOx) and poly(2-ethyl-2-oxazoline) (PEtOx) have attracted significant attention in the last decades owing to their excellent biocompatibility and antifouling and stealth properties,<sup>2</sup> which has made them one of the top alternatives for the 'gold standard' poly(ethylene glycol) (PEG).

An alternative approach to prepare 2-oxazoline based polymer systems is the spontaneous zwitterionic copolymerisation (SZWIP). This polymerization is believed to occur via a zwitterionic intermediate which is formed upon the reaction between an electrophilic ( $M_E$ ) and nucleophilic monomer ( $M_N$ ) without the addition of an additional catalyst or initiator. Cyclic imino ethers act as  $M_N$  and when combined with a  $M_E$  containing acidic hydrogens they form poly(ester amide)s or more specific N-acylated poly(aminoester)s (NPAE).<sup>3,4</sup>

In this presentation the potential of cyclic imino ethers for the preparation of functional biocompatible and biodegradable materials will be discussed. Particular focus will lie on the synthesis and modularity of functional NPAE based macromonomers and the combination of CROP/SZWIP and reversible-deactivation radical polymerization techniques for the design of responsive comb/brush polymers.

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<sup>3</sup> K. Kempe, P. A. J. M. de Jongh, A. Anastasaki, P. Wilson, D. M. Haddleton, *Chem. Commun.* **2015**, *51*, 16213-16216.

<sup>4</sup> K. Kempe, S. L. Ng, S. T. Gunawan, K. F. Noi, F. Caruso, *Adv. Funct. Mat.* **2014**, *24*, 6187-6194.

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| 2014 – 2016 | Senior Research Fellow (University of Warwick, UK)                                           |
| Since 2016  | NHMRC-ARC Dementia Research Development Fellow (Monash University, Australia)                |

Research interests: stimuli-responsive materials, protein-repellent surfaces, polymer-peptide/protein conjugations, glycopolymers, defined macromolecular architectures: comb/star-shaped polymers, poly(2-oxazoline)s, poly(ester amides), nanoparticles, drug delivery, brain delivery, AD

# Accurate Control of Polymer Structure and Function

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A versatile strategy will be described for the multi-gram synthesis of discrete oligomers from commercially available monomer families, e.g., acrylates, styrenics, siloxanes. Central to this strategy is the identification of reproducible procedures for the separation of oligomer mixtures using automated flash chromatography systems with the effectiveness of this approach demonstrated through the multi-gram preparation of discrete oligomer libraries ( $\bar{D} = 1.0$ ) (**Fig.1**). Synthetic availability, coupled with accurate structural control, allows these functional building blocks to be harnessed for both fundamental studies as well as targeted technological (**Fig.2**).

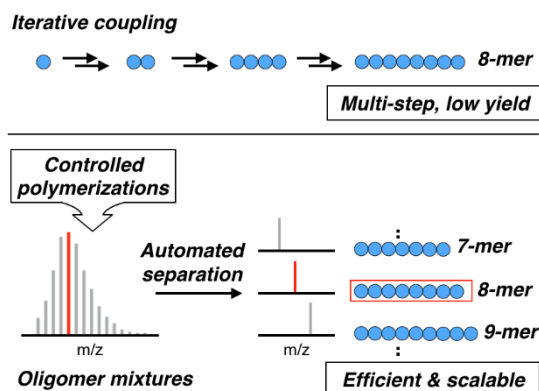


**Figure 1.** Concept for efficient and scalable purification of oligomer mixtures

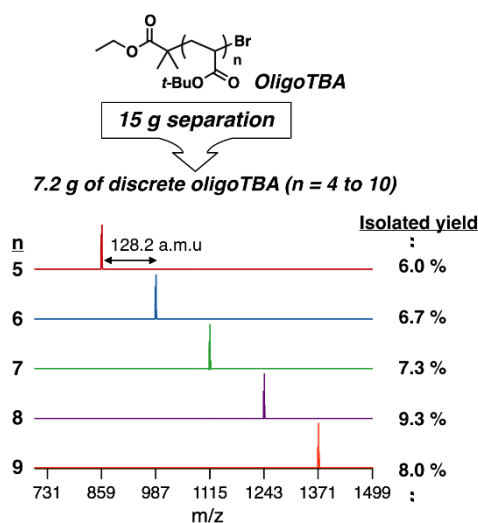


**Figure 2.** Purification of oligomeric t-butyl acrylate derivatives

## Craig J. Hawker

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Research interests: polymer synthesis, controlled polymerization, efficient functionalization reactions

# A New Age of Functional and Responsive Thermoplastic Elastomers: Discovering a Buried Treasure

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With recent advances made in the design and development of multifunctional polymeric materials, elastomers derived from triblock copolymers are in a good position to meet contemporary materials challenges and explore new technological opportunities. In this spirit, we consider the versatility of thermoplastic elastomer (TPE) systems, which provide an attractive alternative to chemically cross-linked materials because of their ability to microphase-separate and form nanostructures connected by a physically cross-linked network that imparts shape memory even after substantial deformation. We first consider the ability of these copolymers to form networks via midblock bridging by examining molecularly asymmetric triblock copolymers grown from a parent diblock copolymer. A combination of experimental methods, theoretical predictions and simulation results reveals the conditions leading to the onset of a load-bearing network. Upon incorporation of a midblock-selective oligomer, the molecular networks formed by triblock copolymers can be swollen to yield highly elastic systems, which exhibit composition-tunable mechanical properties, as well as remarkable electromechanical properties. The morphological features and mechanical properties of swollen TPE gel systems generated from commercial and model styrenic TPEs will be surveyed. These materials, subjected to electrical stimulation as electroelastomers (i.e., dielectric elastomers) between compliant electrodes, are shown to achieve actuation strains greater than 300% and electromechanical coupling efficiencies in excess of 90%. The electrostatic mechanism by which electroactuation proceeds, as well as comparisons with other materials, will be discussed. Unlike conventional dielectric elastomers composed of chemically cross-linked elastomers, these copolymer systems are easily processable and broadly tunable in terms of composition, concentration and molecular weight, and they exhibit little cyclic hysteresis. In the case of acrylic TPEs, electroactuation occurs without the need for mechanical prestrain (often required to thin specimens so that lower voltages can be used to achieve high fields). These same triblock copolymers can be designed in conjunction with liquid metals and phase-change materials to yield ultrastretchable/flexible electronics capable of remaining conductive beyond 1000% strain. Development of stimuli-responsive, hyperelastic and shape-recovering polymeric materials, such as the materials described here, is critical for emerging technologies such as (micro)robotics and microfluidics. As illustrated in Figure 1, shape memory can likewise be achieved through the physical addition of phase-change materials that are midblock-compatible. Incorporation of a charged midblock results in network-forming amphiphilic materials that can be used in ionic polymer-metal composites and photovoltaic devices. Using electron tomography and scattering, we show that the morphological attributes and associated properties of these multiblock ionomers can be solvent-templated and solvent-annealed.

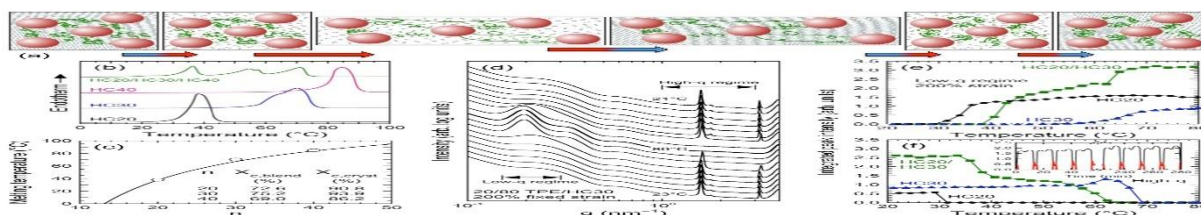


**Figure 1.** Principle of shape-memory polymer blends derived from thermoplastic elastomers.

## Richard J. Spontak

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Research interests: Polymer nanostructures, nanocomposites and networks with emphases on polymer interfaces and morphology and the use of electron tomographic methods to study them.

# Biomimetic Polymers for Protein Stabilization: From Design to Synthesis to Applications in Food and Medicine

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Proteins and enzymes are important for many applications including in personal care products, food industries and as therapeutics, but their instability can limit application or increase costs.<sup>1</sup> Inspired by the stabilizing capability of the disaccharide trehalose, we synthesized a library of glycopolymers with trehalose side chains (**Fig.1**) and demonstrated that they stabilize various enzymes and proteins to environmental stressors including heat, desiccation, and even electron-beam irradiation better than trehalose itself.<sup>2-5</sup> We have studied the effect of different regioisomers of sugar attachment on the resulting stabilization and investigated the mechanisms of stabilization. Furthermore, we have altered the polymer backbone to render the polymers degradable. Recently, we have extended the application of trehalose polymers by incorporating them into hydrogels and self assembled materials. For example, we prepared hydrogels using a short, cost effective synthesis. We demonstrated that these hydrogels could stabilize enzymes important in animal feed and chemical industry. This talk will discuss this research, as well as other systems that could be employed to stabilize biologics for potential use in the personal care products, medicine and food.

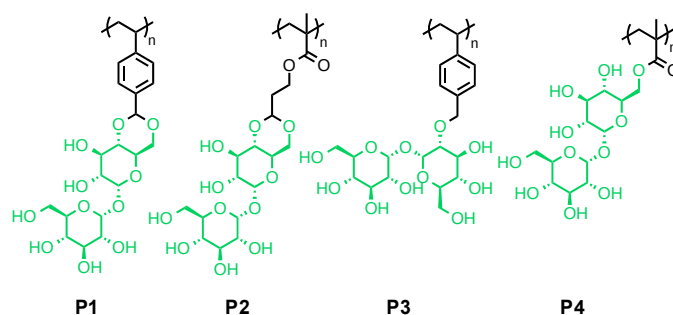


**Figure 1.** Trehalose glycopolymers stabilize proteins and enzymes to extreme conditions.

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<sup>2</sup> R. J. Mancini, J. Lee, H. D. Maynard, *J. Am. Chem. Soc.* **2012**, 134, 8474.

<sup>3</sup> J. Lee, E. W. Lin, U. Y. Lau, J. L. Hedrick, E. Bat, H. D. Maynard, *Biomacromolecules* **2013**, 14, 2561.

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<sup>5</sup> J. Lee, J. H. Ko, E. W. Lin, P. Wallace, F. Ruch, H.D. Maynard, *Polymer Chemistry* **2015**, 6, 3443.

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2000-2002

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Research interests:

biomimetic polymer synthesis and application in medicine, hydrogel synthesis, novel polymeric resists, drug delivery

# Solvent Vapour-Induced Healing of Conducting Polymer Composites

*Paul Baek, and Jadranka Travas-Sejdic<sup>1</sup>*

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Recent advances in organic electronic devices have reached a new milestone in performance and function in various areas. These applications vary from displays to sensory devices. However they still present limitations in mechanical flexibility and electrical durability following the damages caused during their lifetime.<sup>1</sup> Herein, we present a simple route to prepare conducting polymer blends that can address some of these issues through solvent vapour-induced healing of cracks. Conducting polymer blends have been prepared by solution blending of poly(3-hexylthiophene) (P3HT) and poly(dimethylsiloxane) (PDMS)-containing urea segmented copolymer solutions. A freestanding film with 30 wt% of doped P3HT was examined for healing studies of the electrical and mechanical properties. The mechanically bisected film was observed to be readily healed through exposure to solvent vapour, with both electrical and mechanical properties being partially restored. For our blend system, the healing of the blend can be attributed to reversible hydrogen-bonding of PDMS-urea. We propose that exposing the composite to a THF vapour environment allows the PDMS-urea to partially swell, which facilitates the diffusion and rearrangement of PDMS-urea at the cut interface.<sup>2</sup>

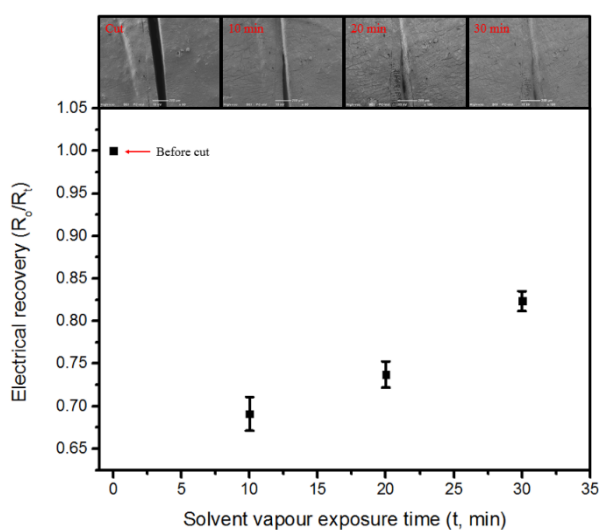


**Figure 1.** Top: SEM images showing healing of free-standing P3HT composite film at the bisected interface as a function of solvent vapour exposure time ( $t$ ). Bottom: Electrical recovery ( $R_t/R_0$ ) of the free-standing composite film as a function solvent vapour exposure time,  $t$ .  $R_0$  and  $R_t$  are the resistance before cutting the composite film and after various solvent vapour exposure time,  $t$ , of the bisected composite film.

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Research interests: conducting polymers, self-healing polymers, polymer synthesis and characterization, neutron scattering techniques, atomic force microscopy.

# Advancing the Performance of Polymer Solar Cells and Photodectors via Plasmonic Coupling

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Polymer solar cells and photodectors constitute representative optoelectronic devices based on polymer semiconductors. Diverse strategies have been demonstrated to advance the performance these functional devices, among which plasmonics have been recognized as a promising platform. Here, we demonstrate a viable and promising optical engineering technique enabling the development of high-performance plasmonic organic photovoltaic devices (OPVs). Laser interference lithography was explored to fabricate metal nanodot (MND) arrays with elaborately controlled dot size as well as periodicity, allowing the spectral overlap between the absorption range of the active layers and the surface plasmon band of MND arrays. MND arrays with ~91 nm dot size and ~202 nm periodicity embedded in poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) hole transport layer remarkably enhanced the average power conversion efficiency (PCE) from 7.52% up to 10.11%, representing one of the highest PCE values and degree of enhancement (~34.4%) among plasmonic OPVs reported to date. The plasmonic enhancement mechanism was investigated both optical and electrical analysis using finite difference time domain simulation and conductive atomic force microscopy studies.

Wavelength-selective photodetection has gained increasing interest in both academic and industrial domains in a ranging number of scientific applications, including imaging, communications, environmental monitoring and chemical/biological sensing. size-controlled silver nanoparticles (AgNPs), gold nanoparticles (AuNPs) and gold nanorods (AuNRs) have been introduced into polymer photodiodes. The evaluated devices exhibit remarkable photocurrent enhancement at corresponding plasmon resonant wavelengths, and a detectivity increase. When compared with a pristine device, AgNPs-, AuNPs- and AuNRs-mediated photodiodes unveil maximum enhancement of detectivity values of 39%, 30% and 54% for blue (450nm), green (525nm) and red (620nm) light detection, respectively. Remarkable selectivity under blue light illumination has particularly highlighted filterless plasmonic AgNPs photodetectors as promising materials in wavelength-selective light detection applications. The mechanism of plasmonics-enhanced photocurrent has been explored combining with the incident photon-to-electron conversion efficiency and steady-state and time-resolved fluorescence analyses. This contribution is believed to showcase the effectiveness and attainable wavelength-selectivity of this strategy by tailoring the size and shape of coupled plasmonic silver, gold or other metal nanostructures.

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| Since 2006 | Ewha Womans University                                          |

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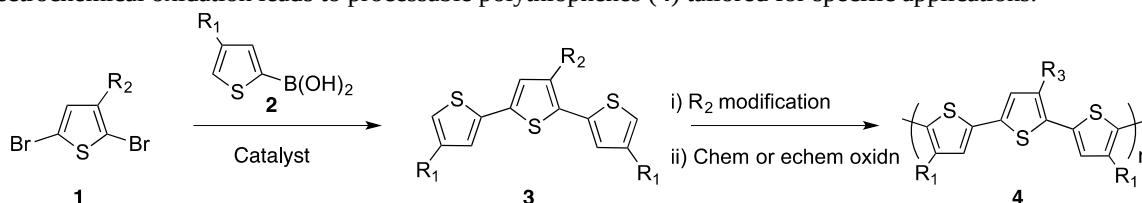
# Functionalised Polythiophenes For Energy, Sensing and Bio- Applications

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Conducting polymers possess properties that make them attractive materials for use in areas ranging from energy storage and conversion,<sup>13</sup> to sensors,<sup>14</sup> drug delivery,<sup>15</sup> and biomedical applications.<sup>16</sup> Predominant amongst this class of polymers are polythiophenes due to the ease of preparation of a wide variety of functionalized polymers. Functionalised thiophene monomers can typically be readily prepared and chemically or electrochemically polymerized. However, while there have been substantial advances in the development of functional, polythiophene-based materials, limitations still exist in the use of these materials, particularly with respect to the production of functionalized polythiophenes with satisfactory solubility and processability. In addition, the introduction of an often-bulky substituent onto each monomer unit of the polythiophene backbone often leads to steric disruption of the backbone that significantly impacts on the polymer properties.

In order to address these issues and develop a general approach to soluble and processable functionalized polythiophenes, we have been investigating the potential of terthiophene as a readily functionalisable precursor to a wide variety of substituted polythiophenes. Solubilising functionality for organic solvents such as alkyl or alkoxy groups ( $R_1$ ) can be introduced onto the periphery of the substituted terthiophene (**3**) by way of thiophene boronic acids (**2**) and reactive functionality ( $R_2$ ) can be introduced using readily prepared functionalized dibromothiophenes (**1**). In a similar fashion, water-solubilising functionality can also be introduced. Modification of the substituted terthiophene (**3**) to give the required substituent followed by chemical or electrochemical oxidation leads to processable polythiophenes (**4**) tailored for specific applications.



This approach not only allows the development of processable polythiophenes but also the introduction of bulky substituents that have a reduced steric impact on the polymer backbone. As a result, we have been able to prepare a number of oligo- or poly-thiophenes with a variety of application specific functionality for sensing,<sup>17</sup> energy applications,<sup>18</sup> and bio-applications.<sup>19</sup> These and other examples will be discussed in this lecture, highlighting the strengths and weaknesses of this approach to functionalised polythiophenes.

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Research interests: conducting polymers, light harvesting materials, solar cells, graphene.

# Surface Photovoltage Studies of Polymer Semiconductor Films Influenced by Plasmon-Upconversion Coupling

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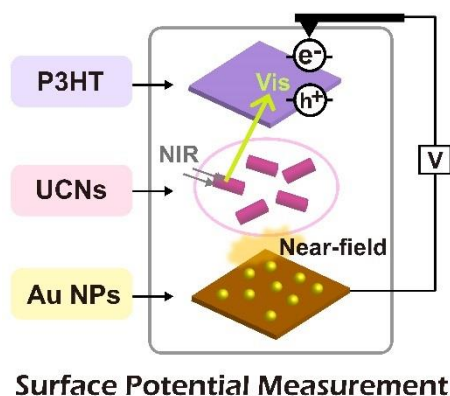
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The integration of upconverters into photovoltaics has attracted much attention owing to an extraordinary optical properties exhibited by a sequential photon pumping.<sup>20-21</sup> Despite of a few studies confirming the expectation, fundamental explanations for a real contribution of upconversion to photovoltaic efficiency are still on demand. In this regard, we suggest a new approach to visualize the upconversion event in terms of surface photovoltage (SPV) by virtue of Kelvin probe force microscopy (KPFM). One of the most conventional polymer semiconductor, poly(3-hexyl thiophene) (P3HT), was employed as a sensitizer to generate photo-induced charge carriers by upconverted light. Along with the upconversion study in P3HT layer, a ternary system which is composed of Au nanoparticles/upconverters/P3HT was also investigated using the same technique (**Scheme 1**). Based on the KPFM results, the upconverted light enabled the formation of near infrared (NIR)-induced charge carriers in P3HT and the integration of Au nanoparticles further promoted the process, doubling the difference in the value of SPV, from 38.5 mV between neat P3HT and upconverters/P3HT films to 76.3 mV between upconverters/P3HT and Au nanoparticles/upconverters/P3HT films. The specific interaction between upconverters and Au nanoparticles also increased a hole collection, which can positively impact on device performance in organic photovoltaics (OPVs).



**Scheme 1.** Ternary system composed of Au nanoparticles, upconverters and P3HT thin layer on conducting substrate for KPFM study.

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Title: Surface Photovoltage Studies of Polymer Semiconductor Films Influenced by Plasmon-Upconversion Coupling

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## Morphology Control in Polymer Solar Cells

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Organic photovoltaics (OPVs) offer a number of advantages, such as fully solid state, compatible with high speed printing and production technologies, but they also a number of challenges related to efficiency and lifetime, many of which can be traced to instability in interfacial layers within the structure.

The ability to create, control and measure nanoscale structures is critical to the development of a range of new technologies. Understanding the properties of a wide range of systems that are generally thought to be “uniform”, “stable” and “consistent” are proving to be critical in the development of a number of options for new, low cost energy solutions which have the potential to change the way in which we harvest and collect energy. All of the approaches rely upon multi-layer structures with layers of the order of 10 - 200nm – a range that materials offer interesting chemical and electronic properties, but are also susceptible to subtle changes in chemistry that can drive significant performance differences.

The structure of the bulk heterojunction – where an exciton is separated into charges – is absolutely critical to the performance of the device. The ideal bulk heterojunction is a co-continuous morphology with a scale length of the order of 20nm. The morphology that is achieved is driven by thermodynamic but limited by kinetics and dependent on the chemical properties and molecular weight of both materials in the heterojunction.

In this presentation, dynamic thermomechanical analysis (DMA) is used in order to follow the development of phases in a bulk heterojunctions fabricated using a range of conducting polymers. As the mechanical properties of these structures is not always optimal, a method using glass fibre reinforcement has been adopted to provide process conditions that are closer to those obtained during conventional OPV fabrication.

A phase separation is clearly observed during annealing processes and information on the composition of the phase has been obtained from the glass transition temperature compared with pure systems.

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# Development of Organic Ionic Plastic Crystal-Polymer Nanofibre Composites as Solid State Electrolytes

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Safer electrolytes for better energy devices such lithium batteries and dye-sensitized solar cells are in urgent need. Organic ionic plastic crystals (OIPCs) are a class of solid-state electrolyte material with good thermal stability, non-flammability, non-volatility and good electrochemical stability. These materials deform easily under stress and can also allow fast transport of ions such as Li<sup>+</sup> through the rotational and translational motions of the matrix ions. Doping with lithium salts can increase the conductivity<sup>1</sup>, and makes them increasingly promising for future energy storage applications<sup>2</sup>.

However, waxy or powdery OIPCs cannot form a flexible membrane. Using electrospun polymer fibres as a matrix is a viable way of developing all-solid-state, free-standing and flexible electrolytes. We have recently reported that the plastic crystal N-ethyl-N-methylpyrrolidinium tetrafluoroborate ([C2mpyr][BF4]), doped with lithium tetrafluoroborate (LiBF4) and combined with electrospun PVdF fibers can successfully support stable lithium cell cycling, with a capacity over 140 mAh g<sup>-1</sup>. The conductivity was also enhanced with incorporation of the PVdF<sup>3</sup>. To further the development of such composite electrolytes, it's important to investigate the influence of polymer nanofibre on the thermal, structural, morphological, and electrochemical properties of OIPCs. Further, understanding the influence of composite composition on electrolyte parameters such as Li transference number, battery cycling performance and stability is key.

In this study, LiFSI doped [C2mpyr][FSI]-PVdF electrospun nanofibers were developed as composite electrolyte membranes (Fig. 1). The effects of PVdF incorporation on the plastic crystal were investigated by DSC, impedance spectroscopy, SEM, synchrotron XRD, and NMR. The optimized ratio was determined to be [C2mpyr][FSI]-10wt%PVdF-10mol%LiFSI, and the transport number in this composite was determined to be 0.11. Use of thicker electrolyte membrane supported more than 500 cycles of a lithium symmetric cell at a current density of 0.13mA/cm<sup>2</sup> at 50°C. Thus, this composite is a promising solid state electrolyte for lithium batteries.

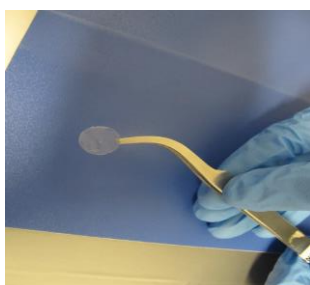


**Figure 1.** The flexible [C<sub>2</sub>mpyr][FSI]-10wt%PVdF-10mol% LiFSI composite electrolyte

<sup>1</sup>M. Forsyth, J. Huang, D. R. MacFarlane, *J. Mater. Chem.* **2000**, *10*, 2259-2265.

<sup>2</sup>L. Jin, P. C. Howlett, J. M. Pringle, J. Janikowshi, M. Armand, D. R. MacFarlane, M. Forsyth, *Energy and Environ. Sci.* **2014**, *7*, 3352-3361.

<sup>3</sup>P. C. Howlett, F. Ponzio, J. Fang, T. Lin, L. Jin, N. Iranipour, J. Efthimiadis, *Phys. Chem. Chem. Phys.* **2013**, *15*, 13784-13789.

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# Designing of electrospun polymer fibers for organic ionic plastic crystal-based solid state electrolytes

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Combining the light-weight of lithium metal and no-leaking, no flammable liquid electrolyte, all solid state lithium metal batteries have been considered as one of most promising power sources for advanced energy technology. Organic ionic plastic crystals (OIPCs), which are homologues of ionic liquids but solid form at room temperature, possess the features of solid, low flammability and negligible volatility, have shown to be one of promising candidates as solid electrolyte materials<sup>1</sup> and have been demonstrated on energy storage devices.<sup>2,3</sup> However, OIPCs normally show relatively slow ionic dynamics especially in low temperature crystal phase, which greatly influences the ionic conductivity and limits their applications in high performance batteries. It has been found that incorporation of nano particles,<sup>4</sup> dendrimers<sup>5</sup> and electrospun polymer fibers<sup>6</sup> can effectively improve the molecular dynamics and ionic conductivity, and this mechanism has been further confirmed in recently developed co-electrospun composite OIPC/polymer fibers.<sup>7</sup> Herein, in order to further improve the ionic conductivity and mechanical properties of OIPC electrolytes towards practical battery applications, different electrospun polymer fibers (as ion conductive matrix) were developed. The effects of interfacial interaction on the ion dynamics, ionic conductivity were investigated by solid state NMR, DSC and impedance spectroscopy techniques, and the lithium batteries performance will be also discussed in this report.

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<sup>3</sup> V. Armel, M. Forsyth, D. R. MacFarlane, J. M. Pringle, *Energy Environ. Sci.* **2011**, 4, 2234-2239.

<sup>4</sup> J. M. Pringle, Y. Shekibi, D. R. MacFarlane and M. Forsyth, *Electrochim. Acta*, **2010**, 55, 8847-8854.

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<sup>6</sup> P. C. Howlett, F. Ponzio, J. Fang, T. Lin, L. Jin, N. Iranipour and J. Efthimiadis, *Phys. Chem. Chem. Phys.*, **2013**, 15, 13784-13789

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# Packing a Punch: Combatting HIV Before and After Host Cell Infection with Polymers

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As of 2015, there are 37 million people infected with HIV<sup>1</sup> and despite the approval of 27 different inhibitors, therapy is often hampered by rapid elimination from the body, high cytotoxicity and poor uptake by infected cells.<sup>2</sup> In addition poor adherence of these inhibitors often lead to resistant HIV strains. Here we present a novel library of polymers that exhibit three independent modes of antiviral activity that potentially could reduce the onset of HIV resistance (Figure 1).<sup>3</sup> The polymers were synthesized via RAFT polymerization and comprise sulfonate side chains as well as pendent self-immolative linkers that permit glutathione-mediated release of lamivudine (3TC), a reverse transcriptase inhibitor. We demonstrate that the polymers can act as (i) an entry inhibitor, (ii) a kinase-independent reverse transcriptase inhibitor mediated by the anionic character of the polymers, as well as (iii) a kinase-dependent reverse transcriptase inhibitor mediated by the release of 3TC. The polymer therapeutics described are envisioned to counter the emergence of resistant HIV strains by providing simultaneous pressures against viral proliferation. The polymers exhibit no cytotoxicity against human T-lymphocytes and epithelial cells and can easily be formulated as a gel and therefore make excellent candidates as virustats that prevent mucosal transmission of HIV.

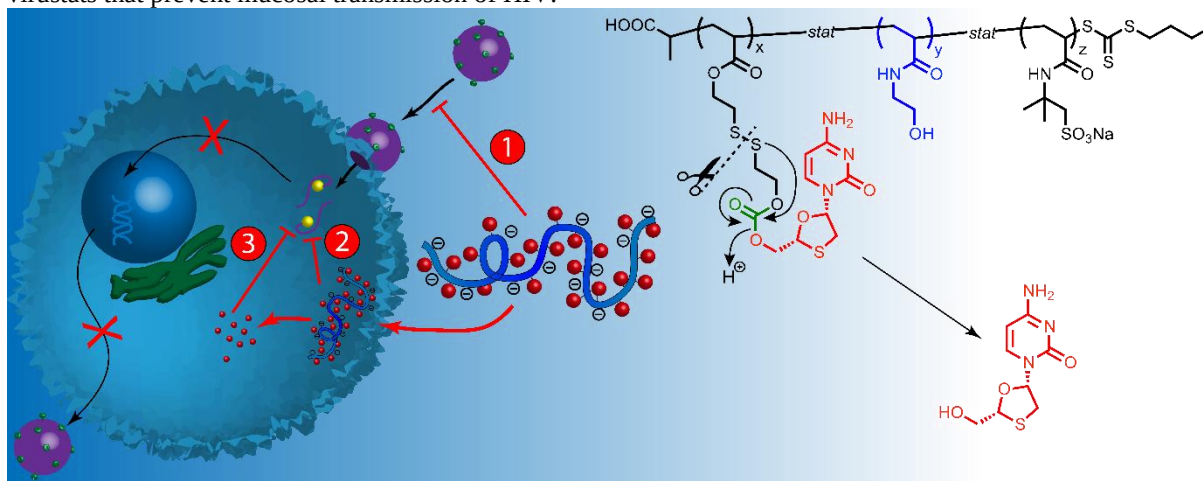


**Figure 1.** Illustration showing the structure of the copolymers and their three independent modes of HIV inhibition, namely (1) entry inhibition, (2) kinase-independent reverse transcriptase inhibition mediated by sulfonated moieties and (3) kinase-dependent reverse transcriptase inhibition mediated by the released 3TC drug.

1. <http://www.who.int/hiv/en/>

2. M. Danial, H.-A. Klok, *Macromol. Biosci.* **2015**, *15*, 9-35.

3. M. Danial, A. H. F. Andersen, K. Zuwala, S. Cosson, C. F. Riber, A. A. A. Smith, M. Tolstrup, G. Moad, A. N. Zelikin, A. Postma, *Mol. Pharmaceutics* **2016**, *13*, 2397-2410.

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# Synthesis and characterization of an aptamer-targeted hyperbranched polymer for the detection of triple-negative breast cancer

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Enhancing clinician's ability to prescribe therapeutic regimes and improve patient outcomes relies on improvements to early detection and diagnosis of cancers. Molecular imaging probes that are targeted to tumour cells are at the forefront of new approaches to image cancers both *in vitro* and *in vivo*. Within our research group we have focused on developing hyperbranched polymers (HBPs) synthesised by RAFT polymerisation which incorporate both targeting and molecular imaging modalities within a single construct.<sup>1, 2, 3</sup> The current work focuses on the production of HBPs targeted to the particularly aggressive triple-negative breast cancer. This was achieved by attachment of an oligonucleotide aptamer which recognises the vascular endothelial growth factors (VEGF) 165 and 121,<sup>3</sup> that are over-expressed in triple-negative breast cancer. Facile polymer synthesis methodologies allowed incorporation of multiple molecular imaging modalities as well as attachment of the aptamer to polymer chain ends utilizing copper-free azide-alkyne 1,3-dipolar cycloaddition. The construct incorporated an optical probe (near-infrared fluorophore), to enable *in vitro* assessment of construct targeting as well as desferoxamine (a <sup>89</sup>Zr chelator) to enable clinically relevant positron emission tomography. The HBP is also composed of a high proportion of PEG-based monomers to enhance biocompatibility and increase circulation time (Fig 1). The targeting efficiency of these aptamer-conjugated HBPs to VEGF expressing human breast cancer cells was assessed both *in vitro* and in xenograft *in vivo* models

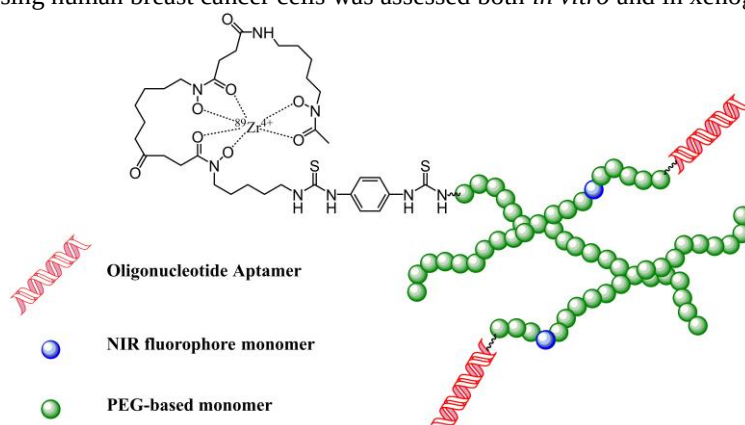


**Figure 1.** Oligonucleotide aptamer-HBP conjugate as a targeted molecular imaging probe

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## Investigations into the role of size for successful cancer nanomedicines

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The development of ‘smart’ nanomedicines capable of visualising stimuli-responsive delivery of therapeutics continues to provide a promising outlook for the future treatment of cancer. Advances in *in vivo* molecular imaging technology, such as MRI, PET and optical imaging, have increased our ability to successfully visualise and monitor the delivery and effect of nanomedicines in the body. However, the effect that specific physicochemical properties, such as nanomedicine size, has on the distribution of nanomaterials across an entire tumour mass *in vivo* remains largely unknown.

Recently, an alternative *in vivo* imaging technique, optoacoustic imaging (OI), has been shown to reveal important pharmacokinetic and pharmacodynamic information about nanomedicines<sup>1</sup>. By combining the high contrast of optical imaging and the high resolution of ultrasound, OI is an emerging technique that surpasses the capabilities of its constituent techniques providing superior deep tissue imaging through sensitive detection of both endogenous and exogenous probes. As has been previously demonstrated<sup>2</sup>, OI is also capable of monitoring molecular interactions through incorporation of an appropriate donor-acceptor FRET pair. Here we report on the development of novel polymeric assemblies through the synthesis of a dual stimuli-responsive amphiphilic crosslinked polymer micelle using RAFT polymerisation. This micelle was subsequently loaded with paclitaxel for cancer therapy and labelled with an optoacoustic FRET pair responsive to cell-death biomarkers, thus enabling visualisation of nanocarrier delivery and uptake as well as real-time monitoring of the cellular response to therapy in tumour tissue. Release of the drug from the micelle is enhanced through cleavage of disulfide crosslinkers, while the subsequent therapeutic effect is monitored through the evolution of an optoacoustic response as a result of FRET that arises following degradation of the nanostructure through a caspase-3-cleavable (DEVD) peptide sequence. By combining this reporter system with polymeric micelles of different size it has been possible to visualise how size can influence diffusion across a heterogeneous tumour mass and how this will ultimately effect therapeutic efficacy of the nanomedicine.

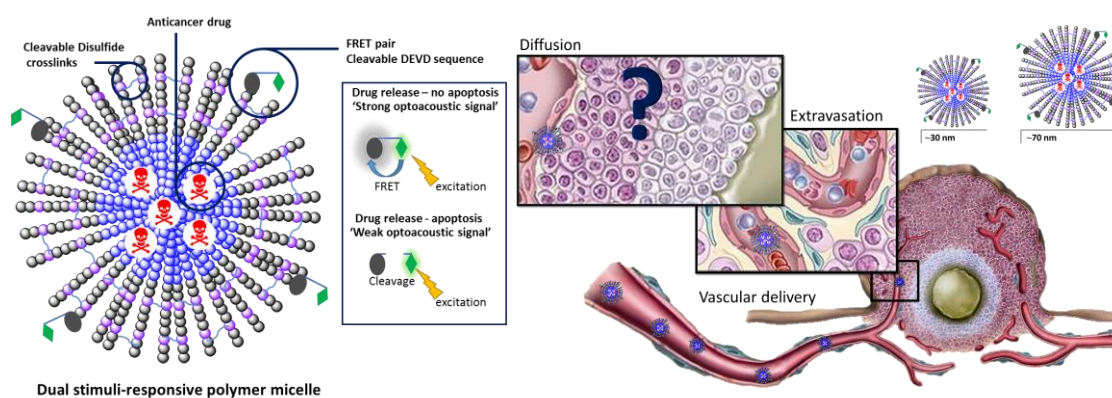


**Figure 1.** Schematic showing dual stimuli-responsive polymer micelle comprising an anticancer drug which releases upon cleavage of disulfide crosslinks and a FRET pair for OI to monitor therapeutic effect through cleavage of a caspase-3 responsive DEVD peptide. Upon intravenous delivery, the effect of micelle size on effective treatment of cells across the tumour mass can be monitored through optoacoustic imaging.

<sup>1</sup> A. Jathoul, J. Laufer, *et al.*, *Nat Photon* **2015**

<sup>2</sup> Y. Wang, J. Xia, L.V. Wang, *J. Biomed. Opt* **2013**, 18 (10), 101316

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## Protein adsorption on polymeric particles –influence of particle design and biological milieu

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There is mounting evidence that the formation of a protein corona around engineered drug delivery vehicles is the primary determinant of their biological behaviour *invitro* and *in vivo*, as this changes the particle's 'synthetic identity' to a 'biological identity'. Therefore, it is crucial to fully understand how particle design parameters such as size, surface material and functionalisation as well as the biological milieu which includes protein identity and concentration, exposure time and adsorption conditions influence this process. This understanding will assist in the design of smarter drug delivery vehicles, which either diminish protein adsorption or take advantage of the adsorbed corona. Furthermore, it will facilitate predicting protein corona formation and composition and therefore bio-nano interactions.

In this study, we investigate the adsorption of proteins from complex media around nano- and micro-sized polymeric particles under varying conditions such as protein concentration and incubation time. Furthermore, we study the impact of flow on the protein adsorption behaviour using defined microfluidic systems. As our main focus lies on protein adsorption around stealth materials, zwitterionic poly(2-methacryloyloxyethyl phosphorylcholine) (PMPC) replica particles are used to examine protein fouling.

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## Understanding the secrets of nanoparticle cell interactions

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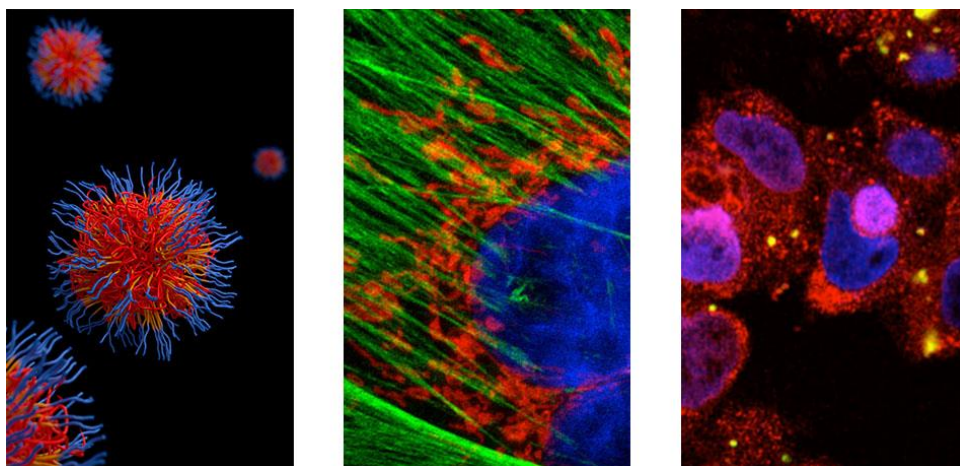
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To engineer 'smart', responsive materials for drug delivery it is essential to understand how nanoparticles interact with cells. Targeted delivery of drugs to specific cells in the body by immobilising therapeutics inside antibody functionalised nanoparticles has the potential to revolutionise the treatment of many diseases. However, our understanding of how these nanoengineered materials interact with cells is limited.

We are developing tools to understanding how these materials interact with cells,<sup>1,2</sup> so we can engineer materials that respond better to the biological conditions they encounter.<sup>3,4</sup> In particular, we are interested in understanding the internalisation, processing and trafficking of nanoparticles in cells. This presentation will focus on understanding the internalisation of polymer nanoparticles into cells, and their subsequent fate once they are inside the cell. It will also outline the progress we are making towards understanding how nanoparticles can induce transport of drugs from the endosomal compartments into the cytoplasm.



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## **Polyion Complex Micelles as Cancer Therapeutic Carriers for Protein Drugs**

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Polyion complex (PIC) micelles of protein model drugs and block copolymers consisting of poly[(ethylene glycol) methyl ether acrylate] (pEGMEA) and either poly(2-carboxyethyl acrylate) (pCEA) or poly(acrylic acid) (pAA) were prepared in aqueous solution as drug delivery carriers. Different particle sizes and different values of zeta potential were obtained from dynamic light scattering (DLS) when varying the molar ratio of the protein and the block copolymer in the system. The binding between the protein and polymer was confirmed by raster imaging correlation spectroscopy (RICS). The structure and activity of incorporated proteins was found to be undamaged. Cellular uptake of the PIC micelles in MCF-7 cancer cells were confirmed by both flow cytometry and laser scanning confocal microscopy (LSCM) after 2-hour incubation. PIC micelles based on different block copolymers were found to have different cellular uptake rates even if the sizes and values of zeta potential of the particles were similar. Lightsheet microscopy was used to obtain the distribution of PIC micelles in 3D MCF-7 cell spheroids with a diameter of 0.5 mm. pEGMEA-*b*-pAA based PIC micelles were found to penetrate deeper in the 3D cell spheroids, whereas drugs were released from pEGMEA-*b*-pCEA based micelles with more ease.

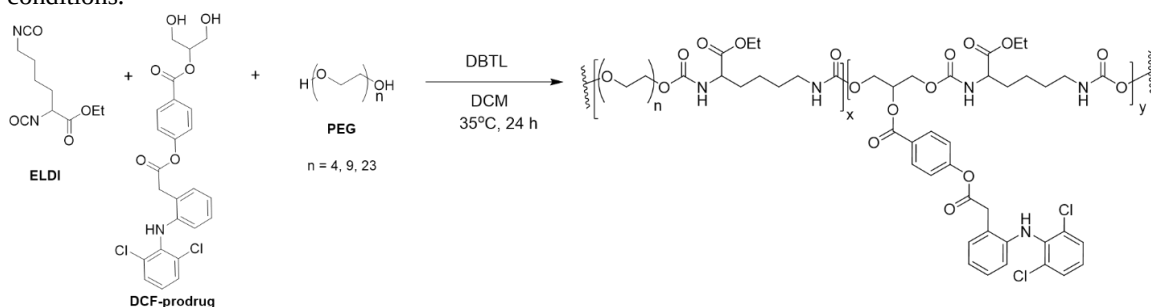
## Tunable and controlled polymeric drug delivery of NSAIDs

*Felisa Reyes-Ortega<sup>1,2</sup>, Adrian Sulistio<sup>1,2</sup>, David Valade<sup>2</sup>, Frank Wang<sup>1,2</sup>, Asha D'Souza<sup>2</sup>, Sarah Ng<sup>2</sup>, Stephen Birket<sup>2</sup>, Andrew C. Donohue<sup>2</sup>, John Lambert<sup>2</sup>, Russell Tait<sup>2</sup>, Michael Whittaker<sup>1</sup>, Thomas Davis<sup>1</sup>*

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To date, polymeric systems used for controlled delivery of non-steroidal anti-inflammatory drugs (NSAIDs) have been based on self-assembled materials such as micelles, vesicles or reversible hydrogels or conventional polymer matrices such as PLGA. These materials rely on physical encapsulation properties to load the NSAIDs and provide a diffusivity barrier to slow the rate of NSAID release. While physical encapsulation can provide a relatively high drug loading, it leads to a sudden burst release of drug which in turn can lead to dose variations and local adverse drug events.<sup>1</sup> Alternatively, NSAIDs can be covalently attached to the polymer via degradable linkers that cleave to release the drug based on the environmental cues of the target area (e.g. hydrolysis). The lability of the degradable linkage provides an alternative kinetic barrier to control the rate of drug release. On example of such a degradable linkers an ester that is subject to hydrolysis to release the drug under physiological conditions.



**Scheme 1.** Synthesis of linear diclofenac (DCF) polymer conjugate *via* step growth approach in a one pot synthesis.

Diclofenac (DCF) is a potent NSAID that is widely used for the treatment of pain and inflammation.<sup>2</sup> We wished to develop a sustained release polymeric delivery system to release therapeutic amounts of DCF over a prolonged period and overcome the burst release associated with encapsulated systems. The diclofenac was covalently modified, by esterification, to form a hydrolysable drug-diol monomer. This monomer was incorporated into a polyurethane system, by step-growth polymerisation with ethyl lysine diisocyanate<sup>3</sup> (Scheme 1). The release of DCF was modulated by varying content of the co-monomer, poly(ethylene glycol) (PEG). By modifying the chain length and content of the PEG comonomer, it is possible to obtain drug delivery systems (DDS) with different drug release rates (Figure 1). These DDS can provide sustained and controlled drug release which can be tailored to suit different treatment regimens.

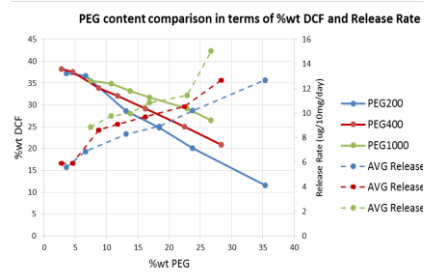


**Figure 1.** Relationship between the PEG content (wt%), DCF loading (wt%) and the average release rate to achieve complete DCF release (ug/10mg/day).

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## **The Origin of the Polymer Banknote**

*T H Spurling<sup>1</sup>*

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This is an account of the Reserve Bank of Australia and CSIRO working together to produce the world's most secure and durable banknotes. It was known within CSIRO as 'the Bank project', and was one of the Organisation's longest and most successful projects.

The project to develop more secure banknotes commenced in 1968 following the 1966 discovery of forgeries of the new Australian decimal currency. Our story ends in 1988 with the release of the \$10 bi-centennial commemorative banknote. Of course the search for more secure banknote technology never ends.

This talk draws upon personal recollections of events and the archived CSIRO records of the project. It is therefore very much the story of the project not only from a CSIRO point of view but from the point of view of people intimately involved in the project.

I hope to make two points. The first is to convey not only the fun and excitement of doing highly applicable science but also the frustration and hard work involved in bringing ideas to the market place.

The second is to show how CSIRO contributes to the nation's economic and social well-being by conducting 'market pull' projects. Such projects were rare at Fishermens Bend in the 1960s and 1970s. Indeed, the Bank project was one of the first genuine 'market pull' projects in CSIRO. In 1968 there were no scientists in CSIRO working on projects to make more secure banknotes with the idea of contacting a banknote producer to see if they were interested in their invention. CSIRO was given the problem by the Bank and had to assemble all the skills and disciplines needed solve the problem. This meant in addition to having to enlist his chemists to work on the project one of the authors had to recruit physicists and engineers to his Division of Applied Organic Chemistry. In those days there was little incentive for other parts of the Organisation to lend their skills to the project. This is not the case in the modern CSIRO. Neither is the flexibility that the 1970s Chief had to muster resources. CSIRO knows that it needs to constantly review and modify its approach to the users of its research.

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1969-2003 CSIRO

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## **Developmental History and Future of Polymer Banknotes in Australia**

*Greg Dicinoski<sup>1</sup>, Phillip Fox<sup>1</sup>*

<sup>1</sup>Reserve Bank of Australia, Note Issue, Craigieburn, Victoria, Australia, 3064

This presentation forms the second part of the series detailing the developmental history of the Australian polymer banknote. The first part, presented by Tom Spurling and David Solomon, will detail the rationale for the change from paper to polymer, the CSIRO development project for the polymer banknote and the transfer of the technology to the Reserve Bank of Australia (RBA). It will conclude with the launch of the world's first circulating plastic banknote; the commemorative Bicentennial \$10 banknote.

In this second part we will focus on the process of translating the technology developed by the CSIRO into a production-ready process for the large-scale manufacture of polymer banknotes, a process we refer to as the “banknotisation” of the technology. In terms of this process the presentation will cover the manufacture of the polymer film along with the printing technology used on banknotes. An important part of this process is the RBA ensuring that the polymer banknotes are fit-for-purpose in circulation, which needs to take into consideration the changing landscape for banknotes. In particular, automated validation, processing and handling of banknotes is far more significant today than when polymer banknotes were first introduced and the RBA needs to ensure that, in addition to providing easily recognised security features for the public, the banknotes also meet the requirements of industry stakeholders.

The polymer material used in Australian banknotes for the past 25-plus years is now not only a security feature in itself, but also a platform technology for the incorporation of sophisticated security features. This presentation will also detail the history and operation of security features used on Australian banknotes dating back to 1988, including the new banknote series launching in 1 September 2016. The discussion around security features will highlight the new technology involved in the application of these to the polymer platform. This presentation will conclude with a discussion around future research directions investigating novel security features for later generations of Australian banknotes. As there are many areas in which research is being undertaken, a number of key areas will be highlighted and these include UV lithography, metamaterials and flexible electronics. Some specific examples will be provided in the discussion to highlight these important areas.

### **Prof Greg Dicinoski**

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## Characterisation of industrial adhesives with (solid-state) NMR spectroscopy

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Adhesives are an important class of industrial polymers with applications ranging from adhesive tape and sticky notes through to resins used for repairing conveyor belts in mines.

The main case study in this presentation will be the development of a new resin to repair conveyor belts in mines. Any downtime in the production process is very costly. A repair tool of choice is in this case a hot melt adhesive: it can be applied quickly, and once applied it does not require a long cure time until it has the desired properties that the process demands. Since there are many possible sources of physical and chemical stress in a mining process, the adhesive needs not only to be strong in its adhesive properties and its flexibility, but also very stable against mechanical stress, heat, UV radiation, acids, bases and other aggressive chemicals, mostly hydrocarbons. The base polyamide resin thus needs to be strengthened by the right combination of additives. ATR FT-IR, solid-state NMR and Raman spectroscopies were assessed for their potential to reveal the composition and homogeneity of the final product. The results were used to evaluate the extrusion process used to form the final product.

NMR spectroscopy also allowed the characterisation on the molecular level of acrylic pressure-sensitive adhesives used in tapes and sticky notes. In particular, <sup>13</sup>C NMR spectroscopy in the solid-, solution- and melt-state were compared in order to obtain with best accuracy and precision the average degree of branching in the samples. A similar methodology is now being applied to poly(acrylic acid) samples with potential applications in scale control and drug delivery.

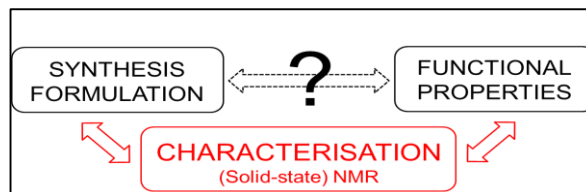


**Figure.** Importance of reliable characterisation methods for the elucidation of the relationship between the synthesis (or formulation) and the functional properties of industrial adhesives.

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## **Five Top Tips for Australian Patent Protection**

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A great idea should be protected. But the patenting process for protecting an idea can be time-consuming and complex.

When pursuing patent protection, some steps taken during the early stages of the patenting process can help to reduce the likelihood of issues arising down the track.

This presentation will provide an overview of some top tips that inventors can use to help improve the prospects of obtaining useful patent rights for an invention.

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# Functional starch based hydrogels: sustainable material solutions for wastewater and agriculture industries

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The continuous application of synthetic fertilizers to crops has become common practice in order to meet the increasing global demand for crop production. This practice has come at the cost of inefficient nutrient delivery and elevated concentrations of nutrients running into surface and ground waters. Oversupply of nutrients entering environmental systems has led to nutrient pollution and environmental degradation<sup>1</sup>. One such example locally of a perturbed environmental system is the enhanced response of algal growth in the GBRMP to nutrient runoff from sugarcane farming. Algal blooms lead to a range of negative impacts on the reef that threaten its long term survival. To protect the reef from this type of environmental stress we must aim to create sustainable food security that mitigates the impacts of intensive farming.

Bioderived materials have recently been of interest to researchers in their effort to develop sustainable solutions for improving farming systems and wastewater treatment technologies<sup>2</sup>. Starch, as a natural biopolymer, has gained attention as an economical feedstock in the development of new technologies for the production of sustainable fertilizers<sup>3</sup> as well as the recovery of nutrients from wastewater<sup>4</sup>. In addition to being chemically modifiable, starch is abundant, renewable and biodegradable. Starch can be processed into three dimensional networks of chemically cross-linked copolymers, known as hydrogel. These hydrogels can be modified to produce functional biopolymers with a unique set of properties such as high water content, flexibility, selectivity, and dynamic responsiveness to external stimuli<sup>5</sup>. For instance, starch based hydrogels have recently been identified as having potential to be successfully employed in a range of unconventional applications such as removal of industrial pollutants from wastewater<sup>6</sup>. To date there have been few advanced nutrient recovery systems and slow release fertilizers based on modified starches<sup>7</sup>. The majority of them however, are not sustainable solutions nor economically viable. In addition, the few existing starch based hydrogels are passive sponges with limited functionality as a sorbent and are only found in the laboratory<sup>8</sup>. Therefore, the development of affordable and smart systems based on starch hydrogels is a promising approach for reducing the environmental impact of industrial pollution. Our vision is to develop a novel processing method based on reactive extrusion for industrial scale production of starch based smart hydrogels, as a full-circle sustainable material solution for wastewater and agriculture industries.

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<sup>6</sup> B. Shahrooie, L. Rajabi, A. A. Derakhshan, M. Keyhani, *J Taiwan Inst Chem Eng.* **2015**, 51, 201-215

<sup>7</sup> H. Peng, S. Zhong, Q. Lin, X. Yao, Z. Liang, M. Yang, G. Yin, Q. Liu, H. He, *Carbohydr Polym.* **2016**, 138, 210-214

<sup>8</sup> C. Xiao, *Starch - Stärke.* **2013**, 65, 82-88

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## **Polymeric Monolithic Materials For Analytical Applications**

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Polymer based monoliths were introduced over 20 years ago. The relatively simple preparation, robustness, high permeability to flow, mass transfer via convection and flexible chemistry has since seen these materials used in a range of applications such as chromatography and as supports for synthesis, catalysis and immobilized enzymes. These same properties make monolithic polymers an excellent choice as materials for sampling and sample preparation, particularly for miniaturized technologies with the potential to produce cleaner extracts and facilitate rapid sample preparation for mass spectrometry (MS).

This presentation will introduce monolithic micro-sampling devices developed for sampling and sample preparation of biological samples, including whole blood. Monolithic micro-sampling devices prepared within disposable pipette tips have been developed for in-tip separation of blood cells from plasma and as immobilized enzyme reactors (IMER) for protein digestion. Subsequent micro-solid phase extraction ( $\mu$ -SPE) was achieved using high surface area polymer monoliths. The  $\mu$ -SPE device was then directly hyphenated with both ESI-MS and nanospray-MS. Microextraction by packed sorbent (MEPS) in which the SPE phase is placed within an exchangeable needle hub integrated into an analytical syringe is also demonstrated for  $\mu$ -SPE. A workflow was developed using the monolith filtration and enzyme reactor technology in combination with an at-line micro SPE-ESI-MS approach enabling both sample preparation and analysis to be completed in < 20 min, facilitating high-throughput sample analysis in a standard bioanalysis workflow. New, high surface area polymeric monolithic sorbents tailored for SPE of small and medium sized molecules will also be introduced and are shown to provide significant advantages over particle-based sorbents, providing greater reproducibility in the sorbent bed. In a further embodiment the selective extraction of small molecules from complex matrices such as plasma will be demonstrated, using new restricted access poly(DVB)-g-PEGMA monoliths.

The majority of this work was undertaken as part of an integrated research program between academic and industry based researchers. This presentation will also describe the research approach taken and demonstrate how using this approach that fundamental research can lead to translation to new commercial products.

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## Sand Stabilization along the Railway Track with Polymer as a substitute of Crude Oil

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The paper describes the applications of Polymer substitute in sand stabilization, As Saudi Railway Organization was using Crude Oil to stabilize the sand along its railway network, but crude oil is not painstaking as an environment friendly solution. Therefore a thorough study was proceeded by **Almobty Company** with the sand stabilization to avoid sand creep towards railway tracks with an environment friendly measure by using polymer as a substitute of crude oil.

For this purpose a study was carried out to check the behaviours of sand movements towards the railway track, and it was suggested to construct new sand berms/dunes as well as the existing sand berms/dunes with Polymer (**Styrene Acrylic Copolymer**) mixing with water with a ratio of 1:9, Polymer sprinkling was carried out in 3 different locations parallel to crude oil spread area to find out the performance of both chemicals in terms of, Penetration depth of each product, Stability of surface and Environmental impact assessment (EIA).

The performance comparison of both chemicals gave the following results;

| Sr.# | Parameter                              | Crude Oil                                                                                       | Polymer                                                                            |
|------|----------------------------------------|-------------------------------------------------------------------------------------------------|------------------------------------------------------------------------------------|
| 1-   | Penetration Depth                      | 2-10 mm with average 5mm, remain close to the surface with shallow penetration ( <b>Fig.1</b> ) | 2-25 mm with average 10mm, uniform penetration ( <b>Fig.2</b> )                    |
| 2-   | Stability of Surface                   | Surface stability lost and easily disturbed with human foot traffic ( <b>Fig.3</b> )            | Stable Surface and capable of handling human foot traffic ( <b>Fig.4</b> )         |
| 3-   | Environmental Impact Assessments (EIA) | No living vegetation is visible ( <b>Fig.5</b> )                                                | No visible embarrassment of plant growth in polymer treated areas ( <b>Fig.6</b> ) |



**Figure 1.** Crude Oil Penetration



**Figure 2.** Polymer Penetration



**Figure 3.** Crude Oil Stability



**Figure 4.** Polymer Stability



**Figure 5.** Crude Oil Impacts



**Figure 6.** Polymer Impacts

Furthermore, Economic analysis with cost benefit ratio (CBR) of three years study was also calculated between crude oil and polymer treatments in order to execute the proposed solution, and it was observed that the cost of sand stabilization by using polymer sprinkling and maintenance was much cost effective as compare to treatment where sand stabilization by using crude oil sprinkling and maintenance were applied.

It has been concluded that sand stabilization by using polymer is an **environmental friendly**, **strong suit**, more stability and ultimate cost effective solution in a long run and future demands.

Based on the study and practical demonstration therefore it has been decided to carry out **172.5 Km** sand stabilization with Polymer as an environmental friendly and substitute of crude oil and later it will be extended by 20 Km, the sprinkled area will be continuously under monitored in future for maximum required out puts.

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## **Future Fibres Industry Transformation Research Hub**

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The Future Fibres Industry Transformation Research Hub is an Australian Research Council (ARC) supported initiative which strives to accelerate the transformation of the Australian manufacturing industry to a vibrant, future fibre-oriented sector. Collaborative projects, which connect leading fibre researchers and highly innovative small and medium enterprises, will tackle industrially relevant research questions. Outcomes will impact on significant global issues, including reducing our environmental footprint and improving public health and wellbeing.

Established in mid-2016 and led by Deakin University, the Hub brings together five industrial partners from different sectors: carbon fibre composites (Carbon Revolution Pty Ltd, Quickstep Automotive Pty Ltd), biomedical (Ear Science Institute Australia Inc), clothing (Draggin Jeans Pty Ltd), and novel materials (HeiQ Australia Pty Ltd). They are joined by experts from Deakin, CSIRO, Swinburne University and several international higher education institutions to conduct research within three streams:

- Nanofibres: Focus on development of the emerging industry to create polymeric nanofibres (with very high surface to volume ratio) including high-throughput production and application in diverse areas such as tissue engineering, clothing, filtration and energy generation.
- Advanced carbon fibre composites: High performance composite materials are light weight and have very high strength and are playing an increasingly important role in various industries including automotive. This stream focuses on addressing some of the industrial challenges associated with carbon fibre composites including cost reduction, performance enhancement and improved tailoring of properties to the intended end-use.
- High-value added applications for fibre technology: Focus on exploring target areas where the innovative use of fibres can add significant value to the market, with particular focus on biomedical uses and protective apparel.

### **Emma Prime**

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# SAXS Studies of Self-Assembled Block Copolymer/Ionic Liquid Blends

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Block copolymers are ubiquitous materials with a vast range of applications, e.g. in drug delivery, photolithography, chemical sensors, surfactants, and as templates for the production of hard materials. The properties of these materials and hence the range of potential applications arise from the ability to combine an array of chemical functionalities and architectures that can be synthesised using contemporary polymerisation techniques<sup>1</sup>. More recently, the addition of low molecular weight diluents has been shown to be a pathway to both modify and allow for the control of the microstructures adopted by these systems.

Recent advancements in characterisation methods have opened up the possibility to interrogate these materials at the molecular- and nano-scale and to, therefore, better understand structure-property-performance relationships within them. This work will focus on recent studies by us carried out in part at the Australian Synchrotron using high-throughput and grazing-incidence SAXS to map the phase diagrams and physical properties of a range of polystyrene-*b*-poly(methyl methacrylate) / ionic liquid (IL) blends.

The addition of ILs, is shown to selectively swell the PMMA blocks, and allows for control of both the accessible block copolymer phases<sup>2</sup> (see for example Figure 1) and the resulting properties of the materials. These studies have provided both insights into physical properties of the materials, e.g. scaling properties of the polymers, and practical methods for fabricating microphases such as the co-continuous phases. Moreover, the effects of the resulting microstructures on mechanical and electrical properties are also discussed.

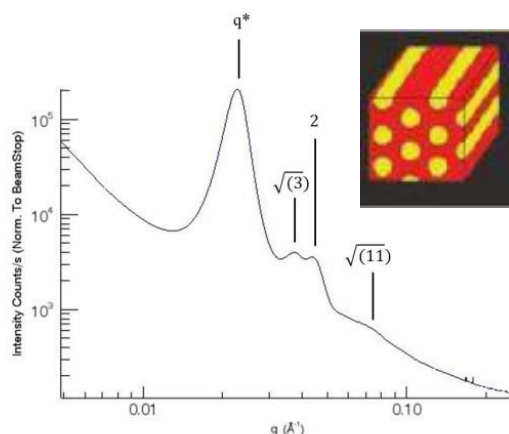


**Figure 1.** SAXS profile a PS-PMMA/EMIM Tf2N blend showing hexagonal packing of cylinders

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1998-2004 Bristol University, UK

1995-1998 Queen's University, Canada

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## Monitoring chemical reactions using capillary electrophoresis: some new insights on peptide grafting, anticancer drug binding and starch digestion!

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Did you ever come across a publication describing an elegant series of chemical reactions, or the manufacturing of an object, only to ask yourself: how do the authors know this is actually happening? How well is the system characterised? How long would it take to accurately characterise it?

The monitoring of chemical reactions involving macromolecules is notoriously difficult, particularly where the characterisation is done in solution. For example, it would be beneficial to perform a reaction directly in a SEC/GPC vial and monitor it. However, this is near-impossible since SEC requires sample preparation by filtration prior to injection. Filtration is not only time consuming, but it can also lead to sample loss. Therefore, we have explored the potential of a novel method that does not require any sample preparation<sup>1,2</sup>: free solution capillary electrophoresis (CE). **The samples do not need to be filtered.** We were able to monitor the **grafting of a peptide on chitosan film**, showing that the peptide forms an oligopeptide while being grafted<sup>3,4</sup> (see figure below). We can also monitor the **binding of an anticancer drug, cisplatin, to poly(acrylic acid)**, despite the incomplete solubility of the former and the variable solubility of the latter. Finally, CE can be used to monitor an even more complex medium: **rice and breakfast cereals while their starch is being digested.**

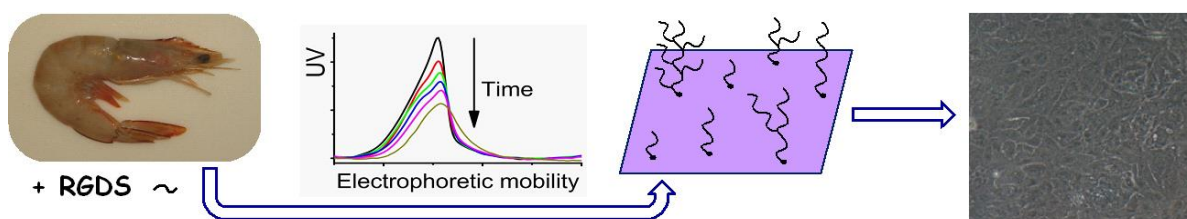


Figure: from prawn shell to chitosan films, to grafting of RGDS peptide monitored by CE to cell culture

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<sup>2</sup> MR Toutounji, MP Van Leeuwen, JD Oliver, AK Shrestha, P Castignolles, M Gaborieau, Quantification of sugars in breakfast cereals using capillary electrophoresis. *Carbohydrate Research* **2015**, 408, 134-141.

<sup>3</sup> DL Taylor, JJ Thevarajah, DK Narayan, P Murphy, MM Mangala, S Lim, R Wuhrer, C Lefay, MD O'Connor, M Gaborieau, P Castignolles, Real-time monitoring of peptide grafting onto chitosan films using capillary electrophoresis. *Analytical and Bioanalytical Chemistry* **2015**, 407, 2543-2555.

<sup>4</sup> JJ Thevarajah, MD O'Connor, P Castignolles, M Gaborieau, Capillary electrophoresis to monitor peptide grafting onto chitosan films in real time *Journal of Visualized Experiments (JoVE)*, **2016**, in press.

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2009: Macromolecular Physical Chemistry Group, University of Mainz, Germany

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## Threading or not threading onto polymer chains

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Cyclodextrins (CDs), cyclic oligosaccharides of glucopyranose units, are widely used in the pharmaceutical industry as drug solubilisers and have also been attracting increasing interest in food products, as flavour carriers, protectants, emulsifiers and dietary fibres. The appeal of cyclodextrins lies in their capacity to form 'host-guest' interactions either with small molecules, or with polymers. They can indeed thread along the chains of macromolecules forming so-called 'pearl-necklaces', a process that is driven by weak interactions and the release of water molecules from inside the CD cavity. These long inclusion complexes – or polyrotaxanes – are the starting point of interesting supramolecular structures that can be used as tools to impart shielding, solubilisation, multi-valency or responsiveness.

In this contribution, I will present some of our work in this area<sup>1-9</sup>, in particular the ability of CDs to modulate the self-assembly of amphiphilic block-copolymers, such as Pluronics<sup>6-8</sup> and their X-shaped counterparts Tetronics.<sup>1,2</sup> We have used small-angle neutron scattering and NMR to resolve the morphology of the complexes on the nanoscale, the kinetics of formation and shed light on the interactions driving these processes, which we then relate to phase behaviour and rheology.

- [1] González-Gaitano *et al.* (2015) *Langmuir*, 31, 4096-4105
- [2] González-Gaitano *et al.* (2015) *Langmuir*, 31, 5645-5655
- [3] Iza *et al.* (2015) *Langmuir*, 31, 2677-2688
- [4] Pérez *et al.* (2014) ***Langmuir***, 30, 11552–11562
- [5] Da Silva *et al.* (2013) *Langmuir*, 29, 7697-7708
- [6] Valero *et al.* (2012) *J. Phys. Chem. B*, 116, 1273-1281
- [7] Castiglione *et al.* (2011) *J. Phys. Chem. B*, 115 9005-9013
- [8] Valero *et al.* (2010) *Langmuir*, 26, **10561-10571**
- [9] Dreiss *et al.* (2009) *Soft Matter*, 5 1888-1896

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## Thin films, Deuteration and Neutron Reflectometry for Characterizing Cellulose-Protein Interactions

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Neutron reflectometry is a powerful tool for probing structure at interfaces including thin films in aqueous environments and offers information on structural changes through depth and molecular disposition. However when components of the system under study have a similar scattering length density then molecular deuteration is required in order to make full use of the technique. Cellulose is an abundant material of great interest not only as a bulk biodegradable structural material but as the basis for nanotechnology applications that take advantage of its regular polysaccharide surface and well-characterised composition. The development of bioactive paper as a platform for low cost diagnostics<sup>1</sup> is a case in point but further advancement requires resolution of interaction between molecules at surfaces and in the aqueous phase. The technology takes advantage of adsorption of protein by cellulose surfaces but characterisation of this phenomenon using neutron reflectometry requires deuterated cellulose as conventional cellulose and protein scatter in a similar fashion.

Bacterial cellulose is readily produced in the laboratory under conditions leading to deuteration of the polymer<sup>2</sup> and we report here for the first time the development of very smooth deuterated cellulose films suitable for neutron reflectometry, and comparison of two film processing methods. Deuterated cellulose was produced by *Gluconacetobacter xylinus* grown on deuterated glycerol in D<sub>2</sub>O and the cellulose was subsequently processed by either acetylation or trimethylsilylation in ionic liquid (1-Butyl-3-methylimidazolium chloride) to produce material soluble in acetone or toluene to facilitate spin coating onto silicon. Acid vapour hydrolysis of material generated using trimethylsilylation in ionic liquid and dissolved in toluene produced thin (10-20 nm) and smooth (RMS= 1 -2 nm) cellulose films which were characterised by ATR-FTIR and AFM. These films were used to investigate adsorption of horseradish peroxidase (HRP), as a model protein, using neutron reflectometry. HRP is an alpha-helical enzyme containing a heme group. It is considered a suitable model system because of its well-known structure, mechanistic understanding, relatively stability and high potential for biotechnological use. Reflectometry data for both unlabelled and deuterated cellulose films and HRP will be presented to show that the latter were thin, smooth films that offer contrast with the HRP enabling quantification of the degree of coverage following adsorption. Analysis and modelling of the reflectometry data showed the thickness of adsorbed HRP layer at the liquid-cellulose interface was  $98 \pm 10$  Å, with roughness of  $17 \pm 5$  Å and with a volume fraction of 20%. This corresponds to partial surface coverage of HRP aggregates. There was insufficient contrast between unlabelled cellulose and HRP to enable useful interpretation.

As the National Deuteration Facility has developed a range of different protocols that produce bacterial cellulose with varying degrees of deuteration<sup>2</sup>, the method described above enables preparation of thin cellulose films with controllable deuteration levels. Control over the level of deuteration can provide a wide range of controllable scattering contrasts that greatly enhances the investigation of the interactions between cellulose and biologically important bio-macromolecules.

**Acknowledgement:** The work conducted at the National Deuteration Facility (<http://www.ansto.gov.au/ndf>) is partially funded by the National Collaborative Research Infrastructure Strategy, an initiative of the Australian Federal Government.

<sup>1</sup>Y.H. Ngo, D. Li, G.P. Simon, G. Garnier, G.. *Adv. Coll. Interface Sci.* **2011**, 163, 23-38.

<sup>2</sup>R.A. Russell, C.J. Garvey, T.A. Darwish, L.J.R. Foster, P.J. Holden. *Meth. Enzymol.* **2015**, 565, 98-121.

### Peter Holden

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Research interests: Structure function analysis using deuteration and neutron scattering; biomolecular labelling.

## Characterisation of Oligoacrylates and their Block Co-Oligomers

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Oligoacrylates and their block co-oligomers synthesised by Reversible Addition-Fragmentation chain Transfer (RAFT) polymerisation are currently used as stabilisers in paints and have applications as surfactants. However, current methods for characterising these oligomers such as Size Exclusion Chromatography (SEC) and Electrospray Ionisation Mass Spectrometry often have limited accuracy. Furthermore these methods can not accurately determine the amount of unreacted RAFT agent, the livingness of the oligoacrylates and whether any branching is present.<sup>1</sup> Free solution Capillary Electrophoresis (CE) has been able to separate and quantify oligoacrylate chains with and without a RAFT end group as well as by chain length and tacticity.<sup>2</sup> In this work free solution CE was used to assess the livingness and block extension of oligoacrylates with styrene, it is then shown that a significant amount of dead oligoacrylate chains were detected in the initial oligoacrylate block but the block co-oligomer as well (Fig. 1) In addition to the livingness, the amount of unreacted RAFT agent was determined for both the initial oligoacrylate block and the corresponding block co-oligomers. The separation of oligoacrylates is shown to be highly reproducible with the separation being conducted over 40 times in the same experimental conditions by more than 10 operators.

Poly(acrylic acid) has been shown to be branched<sup>3</sup> however the chain length required for branching to occur has not been determined leading to assumptions that short poly(acrylic acid) chains are not branched. Using quantitative <sup>13</sup>C Nuclear Magnetic Resonance (NMR) spectroscopy branching has been detected in oligoacrylates with an average chain length of around 10 units (Fig. 2). From these measurements it can be shown that branching occurs in poly(acrylic acid) of even very low molar mass samples. Therefore using a combination of free solution CE and quantitative NMR spectroscopy molar mass, branching and dead chains were determined for oligoacrylates as well as their chain extension to block co-oligomers.

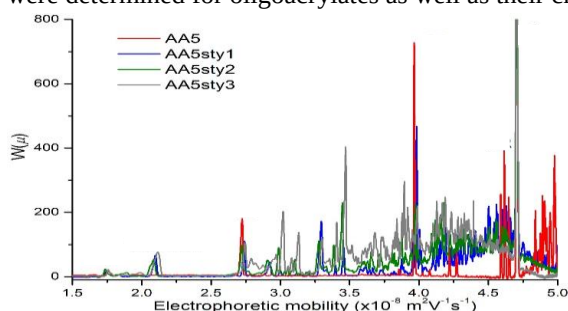


Figure 1. Separation of oligomers by free solution CE

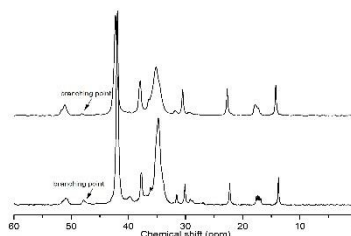


Figure 2. <sup>13</sup>C NMR spectra of oligo(acrylic acid) showing branching point detected

<sup>1</sup> P. Castignolles, M. Gaborieau, E. F. Hilder, E. Sprang, C. J. Ferguson, R. G. Gilbert, *Macromol. Rapid Commun.* **2006**, 27, 42-46.

<sup>2</sup> M. Gaborieau, T. J. Causon, Y. Guillaneuf, E. F. Hilder, P. Castignolles, *Aust. J. Chem.* **2010**, 63, 1219-1226.

<sup>3</sup> A. R. Maniego, D. Ang, Y. Guillaneuf, C. Lefay, D. Gimes, J. R. Aldrich-Wright, M. Gaborieau, P. Castignolles, *Anal. Bioanal. Chem.* **2013**, 405, 9009-9020.

### Adam T. Sutton

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- 2013 BSc Honours, UWS, Australian Centre for Research On Separation Science (ACROSS)
- 2014-2015 MSc, UWS, ACROSS
- 2015-2016 PhD, University of Tasmania, ACROSS
- 2016- PhD, University of South Australia, Future Industries Institute (FII)

Achievements: BSc Honours thesis won the 2013 CRC for Polymers Prize.

Research interests: characterisation of materials and analytical chemistry

## **Raman Spectroscopy, a powerful tool for polymer analysis**

*Taisuke Ota, Kanna Noto, Tomoya Uchiyama, Minoru Kobayashi*

Nanophoton corporation

Raman Spectroscopy is a powerful surface analysis tool for polymer characterisation providing vibrational information about molecular species as well as degree of crystallisation and polymerisation. In conjunction with confocal microscopy, it is capable of providing detailed 3D chemical analysis of complex polymeric structures. The technique has recently been enhanced using line scan technology which increases data acquisition rates and ultimate spatial resolution. We will demonstrate how confocal Raman microscopy can be extended to use in production environments to monitor multi-layer polymer films, crystallisation of PET and other commercial polymer products.

### **Taisuke Ota**

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## Realtime Structure Of Thin Film Organic Semiconductors During Thermally Induced Diffusion

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Stacked multilayered polymer films are an essential structural motif in thin organic semiconductor (OSC) films, such as those found in Organic Light Emitting Diodes (OLEDs) or Organic Photovoltaics (OPVs). For OLEDs the different layers are used for charge injection and transport, as well as light emission. The stability of these layers over time is important for device longevity so the response of such films to thermal stress is an important research topic. In OPVs the structure of the donor/acceptor blends plays a vital role in the performance of the solar cell.

Over the past four years we have pioneered the use of event mode acquisition in Neutron Reflectometry measurements. Neutron Reflectometry is a non-destructive technique for examining thin film structure on the length scale 10-1000 Å. Event mode acquisition has enabled us to make in-situ real time measurements of the multilayer structure in OSC films whilst they are undergoing diffusive processes in response to applied thermal stress. Some of these experiments are only made possible by selective deuteration of individual components, which alters their neutron refractive index.

In this talk I will present some of the highlights of our OSC research examining interlayer diffusivity. These highlights will exhibit the existing and future capabilities of the *PLATYPUS* neutron reflectometer (at ANSTO) for real-time structural characterisation, and underline the necessity of selective deuteration for studying some systems.

The most recent highlight (just accepted in *Advanced Materials Interfaces*) explores how the hole transport material tris(4-carbazol-9-ylphenyl)amine (TCTA) interdiffuses with adjacent charge transport layers that possess different glass transition temperatures (bathocuproine (BCP); 1,3,5-tris(*N*-phenylbenzimidazol-2-yl)benzene (TPBi); *N,N'*-bis(3-methylphenyl)-*N,N'*-diphenylbenzidine (TPD) and *N,N'*-bis(naphthalen-1-yl)-*N,N'*-diphenylbenzidine (NPB)). The overall finding is that the layer with the lowest  $T_g$  acts as a diffusive host for the adjacent layer with higher  $T_g$  and that the high  $T_g$  layer does not necessarily prevent diffusion from taking place. The longevity of OLED devices is increased by choosing materials with high  $T_g$  throughout the device.

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Research interests: Neutron Reflectometry, Real time characterisation of kinetic processes in thin films

# Magnetically-recoverable and Triazole-mPEG Stabilized Transition Metal Nanoparticles for Catalysis

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Nano-gold catalyst is a hot topic in catalysis, thus, understanding the catalytic mechanism and developing efficient and green nano-gold catalysts represent a vibrant field of this research<sup>1-3</sup>. Recently, the triazole-polymer composite stabilized gold nanoparticles (Trz-AuNPs) catalyst has becoming one of the most popular polymer composite based nano-gold catalysts, and exhibits the high catalytic reactivity. However, the easy recovering and recycling the Trz-AuNPs catalyst is still a challenge for scientists<sup>4,5</sup>. In this research, the triazole ligands with carboxylic acid in one termini and poly ethylene oxide (mPEG or PEO) in the other termini are synthesized by “Click” chemistry. Then, the ligands attach on the Fe<sub>3</sub>O<sub>4</sub> nanoparticles *via* the coordination between the carboxylic acid and the Fe<sub>3</sub>O<sub>4</sub> surface. Finally, the AuNPs are stabilized by the triazole ligand and yield the magnetically-recoverable trz-AuNP catalyst<sup>6</sup>.

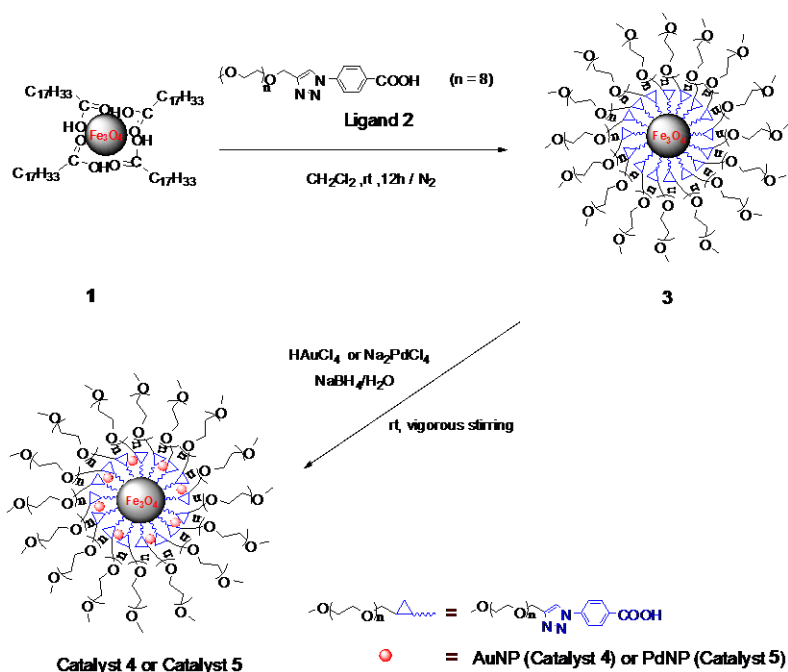


Figure 1. The synthetic route of the polymer composited based catalyst.

<sup>1</sup> M, Brust, M. Walker, D. Bethell, D.J. Schiffrin, J.J. Whyman, *Chem. Commun.* **1994**, 801-802.

<sup>2</sup> D. Astruc, *Nat. Chem.* **2012**, 4, 255.

<sup>3</sup> R.W.J. Scott, O.M. Wilson, S.K. Oh, E.A. Kenik, R.M. Crooks, *J. Am. Chem. Soc.* **2004**, 126, 15583-15591

<sup>4</sup> C. Deraedt, A. Rapakousiou, Y. Wang, L. Salmon, M. Bousquet, D. Astruc, *Angew. Chem. Int. Ed.* **2014**, 52, 8445-8449.

<sup>5</sup> C.J. Serpell, J. Cookson, D. Ozkaya, P.D. Beer, *Nat. Chem.*, **2011**, 3, 478-483.

<sup>6</sup> S. Su, Z. Yue, D. Huang, X. Lai, P. Zhao, *RSC Adv.* **2015**, 5, 44018-44020.

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# Optimising Surface Functionalisation of Carbon Fibre to Enhance Interfacial Adhesion

Kathleen Beggs<sup>1</sup>, Daniel Gunzelmann<sup>1</sup>, Luke O'Dell<sup>1</sup>, Linden Servinis<sup>1</sup>, Thomas Gengenbach<sup>2</sup>, Bronwyn Fox<sup>3</sup>, and Luke Henderson<sup>1</sup>

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Carbon fibres are known for their high strength-to-weight ratio and are used to reinforce a range of polymeric plastics (either thermoset or thermoplastic) to give carbon fibre reinforced plastics (CFRP) commonly referred to as 'composite material'. Carbon fibre reinforced composites are used across a range of industries where there is a requirement for light-weight material such as in the Boeing Dreamliner and BMW i3. However, performance is undermined by failure mechanisms such as fibre-polymer matrix de-bonding. Lack of understanding around the fibre-matrix interface currently limits the development of new composite systems to overcome these problems with interface failure. Building on previous work<sup>1</sup> [ENREF\\_1](#) which shows the introduction of pendent amines onto the fibre surface enhances fibre-to-matrix interaction (**Fig. 1**). This study seeks to optimise existing surface grafting methods and use novel techniques to examine the chemistry at the surface and interface of the modified carbon fibre.

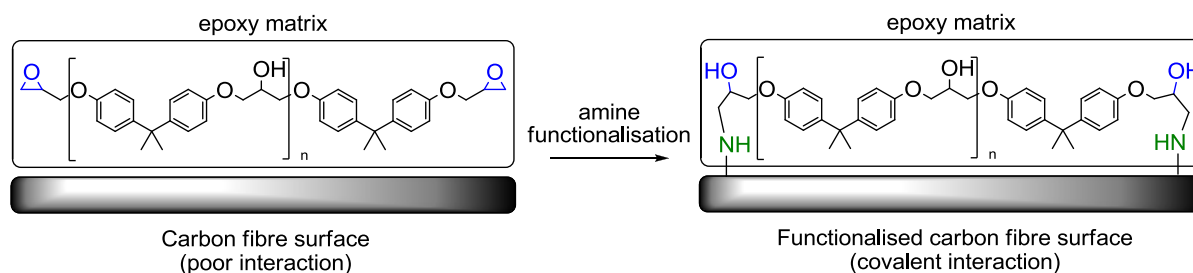


**Figure 1.** General scheme showing improved fibre/matrix interaction following functionalisation

In this study, the surface of the carbon fibre has been modified through *in situ* diazonium formation under both conventional heating and microwave irradiation.<sup>2,3</sup> Electrochemical functionalisation with diazo salts has also been undertaken and offers the fastest reaction rate and superior surface coverage. The modified fibres have been evaluated by XPS and solid state NMR to determine chemical composition. Other key performance parameters (modulus, tenacity, coefficient of friction and interfacial shear strength) have also been determined and compared to control commercial fibres.

<sup>1</sup> Servinis, L.; Henderson, L.; Andrighetto, L.; Huson, M.; Gengenbach, T. R.; Fox, B. L. *Journal of Materials Chemistry A* **2015**, 3, 3360

<sup>2</sup> Beggs, K. M.; Servinis, L.; Gengenbach, T. R.; Huson, M. G.; Fox, B. L.; Henderson, L. C. *Composites Science and Technology* **2015**, 118, 31

<sup>3</sup> Beggs, K. M.; Perus, M. D.; Servinis, L.; O'Dell, L. A.; Fox, B. L.; Gengenbach, T. R.; Henderson, L. C. *RSC Advances* **2016**, 6, 32480

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# Utilizing Interfacial Assembly of Graphene Oxide Nanosheets for Tailored Structures

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The Pentzer lab uses fundamental organic chemistry reactions to tune chemical functionality and assembly behavior of organic materials. One multifunctional material studied, graphene oxide (GO), are sheets of  $sp^2$  and  $sp^3$  hybridized carbon atoms decorated with oxygen functionalities throughout.<sup>1</sup> Not only can this material be covalently modified using relatively simple organic chemistry reactions, but the nanosheets also have antimicrobial and gas barrier properties, good mechanical strength, and can serve as precursors to conductive material.<sup>2</sup> We will report our recent advances in using the interfacial assembly of GO to access three higher order structures: 1) Hollow capsules of GO by stitching together of the nanosheets at the oil-water interface with a di-functional small molecule;<sup>3</sup> 2) Polymer particles armored with GO nanosheets that can be covalently functionalized and assembled with inorganic nanoparticles;<sup>4</sup> and 3) Janus GO nanosheets in which each face is functionalized orthogonally with two different polymers or small molecules (Figure 1).<sup>5</sup> First, the Pinner reaction is used to covalently modify the alcohol groups of GO in suspension to give nanosheets with alkene handles on both faces.<sup>6</sup> Then, the alkenylated GO is assembled at the oil-water interface in a Pickering type emulsion, exposing the nanosheet faces to only oil or water. Subsequent thiol-ene click chemistry then covalently modifies each face independently to yield robust structures. A suite of electronic and microscopic techniques are used to characterize the materials.

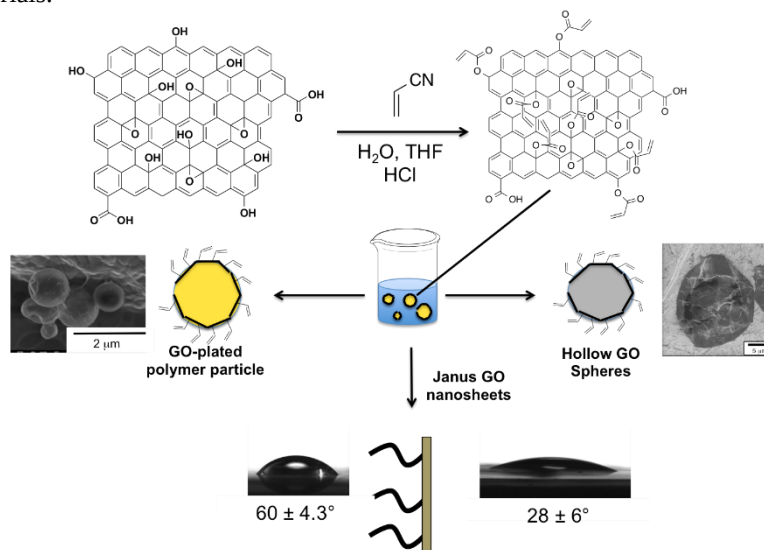


**Figure 1.** Overview of the work to be presented: Assembly of alkenylated graphene oxide (GO) at oil-water interface and modification to give Janus GO nanosheets, hollow GO capsules, and GO-armoured polymer particles.

<sup>7</sup> L. Liao, H. Peng, Z. Liu, *J. Am. Chem. Soc.*, **2014**, 136, 12194-12200.

<sup>8</sup> H. Y. Mao, S. Lauren, W. Chen, O. Akhavan, M. Imani, A. A. Ashkarran, M. Mahmoudi, *Chem. Rev.*, **2013**, 113, 3407-3424.

<sup>9</sup> Q. Luo, P. Wei, E. Pentzer, *Carbon*, **2016**, online.

<sup>10</sup> B. J. Rodier, E. Mosher, S. Burton, R. Matthews, E. Pentzer, *Macromol. Rap. Comm.*, **2016**, online.

<sup>11</sup> B. McGrail, J. Magdalao, B. Rodier, J. Swisher, R. Advincula, E. Pentzer, *Chem Comm.*, **2016**, 52, 288-291.  
B. McGrail, B. J. Rodier, E. Pentzer, *Chem. Mater.*, **2014**, 26, 5806-5811.

## Emily Pentzer

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Personal History:

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Since 2013 Assistant Professor, Case Western Reserve University

Research interests: self assembly, Janus particles, interfaces, 2D materials

# Specific ion effects on responsive polymer brush coatings

Erica Wanless<sup>1</sup>

<sup>1</sup>University of Newcastle

Controlling the interactions between surfaces and their environment is pertinent to the performance of a wide range of formulations and technologies. These interactions can be modified by the presence of a polymer surface coating. Terminally anchored polymer brushes offer an attractive route to regulate surface behaviour through reversible manipulation of colloidal-scale interactions. Through changes in conformation in response to the aqueous phase temperature, pH or electrolyte concentration, a polymer brush coating offers a means, for example, of dictating the wettability, adhesion or friction of a surface.

The response of the polymer brushes to changes in the adjacent aqueous solution have been investigated using in situ ellipsometry, quartz crystal microbalance with dissipation, atomic force microscopy, contact angle and neutron reflectometry measurements. This powerful suite of characterisation methods has enabled us to interpret the coating conformational response in terms of brush thickness, changes in the brush depth profile from neutron reflectometry, and the surface response from contact angle changes.

The focus of our studies has been the family of weakly basic poly(2-dimethylamino)ethyl methacrylate) brushes together with the increasingly hydrophobic poly(2-diethylamino) and poly(2-diisopropylamino) analogues.<sup>1-4</sup> We have characterised the pH-response of this family of polymer brushes through both kinetic and equilibrium measurements. The kinetics of the pH-response was found to be strongly modulated by the monomer hydrophobicity.<sup>2</sup> The delicate balance between the charging behaviour of these brushes and their hydrophobicity led us to investigate the impact of ionic strength on their response.<sup>1</sup> Significantly, the brush behaviour was determined to be not only ionic strength-dependent but is also influenced by the nature of the added salt based on its position in the well-known Hofmeister series.<sup>3-4</sup> Recently we have found similarly strong anion specific effects in the thermoresponse of poly(N-isopropylacrylamide) brushes (**Fig.1**).<sup>5-6</sup> The influence of ion identity on polymer brush response is an area of ongoing investigation and will be the focus of this presentation.

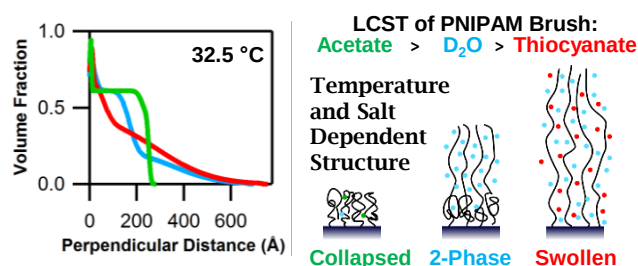


**Figure 1.** Specific anion effects on the internal structure of a poly(N-isopropylacrylamide) brush as determined by neutron reflectometry.<sup>6</sup>

<sup>1</sup> J.D. Willott et al., *Langmuir* **2014**, 30(7), 1827. DOI: 10.1021/la4047275

<sup>2</sup> J.D. Willott et al., *Phys. Chem. Chem. Phys.*, **2015**, 17, 3880. DOI: 10.1039/C4CP05292G

<sup>3</sup> J.D. Willott et al., *Langmuir*, **2015**, 31(12), 3707. DOI: 10.1021/acs.langmuir.5b00116

<sup>4</sup> J.D. Willott et al., *Macromolecules*, **2016**, 49, 2327. DOI: 10.1021/acs.macromol.5b02656

<sup>5</sup> B.A. Humphreys et al., *Phys. Chem. Chem. Phys.* **2016**, 18, 6037. DOI: 10.1039/C5CP07468A

<sup>6</sup> T.J. Murdoch et al., *Macromolecules*, submitted.

## Erica J. Wanless

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Personal History:

- 1990 BSc(Hons) (Australian National University)
- 1995 PhD (Australian National University)
- 1995-1996 Postdoctoral Researcher (University of Otago, New Zealand)
- 1996-2003 Lecturer (University of Newcastle, Australia)
- 2004-2007 Senior Lecturer (University of Newcastle, Australia)
- 2007-2013 Associate Professor (University of Newcastle, Australia)
- Since 2013 Professor (University of Newcastle, Australia)

Research interests: The behaviour of surfactants, polymers, polymer colloids and mineral particles at interfaces.

## Programmable Nucleic Acid – Polymer Conjugates with Sensing and Signalling Capabilities

*Giovanna Sicilia<sup>1</sup>, Adrienne Davis<sup>2</sup>, Sebastian Spain<sup>3</sup>, Kristofer Thurecht<sup>4</sup> and Cameron Alexander<sup>1</sup>*

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The *in vivo* detection of target oligonucleotide sequences is a vital step in diagnosing a wide range of diseases, including bacterial and viral infection, and in numerous cancers. However, the amplification of nucleic acid recognition and binding events to the macroscale using cheap and non-invasive techniques remains an unmet need in the research field of theranostics.

We have developed novel nucleic acid-polymer conjugates capable of sensing DNA binding events through hybridization and de-hybridization processes and amplify these 3-5 nm-scale events into macroscale signals using optical<sup>1,2</sup> and nuclear magnetic resonance techniques<sup>3</sup>. Two classes of hybrid nucleic acid based materials have been screened as potential *in vivo* biosensing platforms of oligonucleotide sequences: hydrogels and multi-nm objects such as polymethacrylamide based polymers functionalised with single stranded DNAs that graft nucleic acid strands bearing MR active nuclei *e.g.* <sup>19</sup>F and Gd(III).

The results from this work demonstrate that fluorine or gadolinium tagged nucleic acid-polymer conjugates can be used to detect very precisely target gene sequences even in extremely low amounts. These systems hold great potential as novel companion diagnostic platforms for the *in vitro* and *in vivo* detection of DNA/RNA-based disease markers. Moreover, the nucleic acid strands that act as reporters of binding events could also carry information which can modify a biological process or act as input signal to trigger release of a drug, or to monitor the progress of a treatment, dependent on what function is needed.

### References:

1. G. Sicilia, C. Grainger-Boulthby, N. Francini, J. P. Magnusson, A. O. Saeed, F. Fernández-Trillo, S. G. Spain and C. Alexander, *Biomaterials Science*, **2014**, 2, 203-211.
2. J. P. Magnusson, F. Fernández-Trillo, G. Sicilia, S. G. Spain and C. Alexander, *Nanoscale*, **2014**, 6, 2368-2374.
3. G. Sicilia, A. L. Davis, S. G. Spain, J. P. Magnusson, N. R. B. Boase, K. J. Thurecht and C. Alexander, *Polymer Chemistry*, **2016**, 7, 2180-2191.

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Personal History: EPSRC Doctoral Prize Research Fellow

Since year 2015

Research interests: theranostic biopolymers, gene delivery, cancer therapeutics, advanced imaging techniques

# Combating Multidrug-Resistant Gram-Negative Bacteria with Structurally Nanoengineered Star-Shaped Peptide Polymers

Shu Jie Lam<sup>1</sup>, Neil M. O'Brien-Simpson<sup>2</sup>, Namfon Pantarat<sup>2</sup>, Adrian Sulistio<sup>1</sup>, Edgar Hoe Hon Wong<sup>1</sup>, Yu-Yen Chen<sup>2</sup>, Jason C. Lenzo<sup>2</sup>, James A. Holden<sup>2</sup>, Anton Blencowe<sup>1</sup>, Eric Reynolds<sup>2</sup>, Greg G. Qiao<sup>1</sup>

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Antibiotic resistance in bacteria has reached alarming levels in many parts of the world, whereby common infections in some settings have become refractory to available treatment options. With the recent emergence of alarming reports on highly resistant Gram-negative 'superbugs', infections caused by multidrug-resistant (MDR) Gram-negative bacteria have been named as one of the most urgent global health threats due to the lack of effective and biocompatible drugs.<sup>1,2</sup>

Herein, a new class of antimicrobial agents, termed 'Structurally Nanoengineered Antimicrobial Peptide Polymers' (SNAPPs), in the form of star-shaped peptide polymer nanoparticles – synthesized *via* ring-opening polymerization of  $\alpha$ -amino acid *N*-carboxyanhydrides – was developed.<sup>3</sup> While most known peptide-based antimicrobials are either selective towards specific bacterial species and/or are toxic towards mammalian cells, these star peptide polymers display sub- $\mu$ M activity against all clinically-relevant Gram-negative bacteria tested with high biocompatibility. SNAPPs were found to be highly effective in combating colistin-multi-drug resistant (CMDR) *A. baumannii* infections *in vivo*, and resistance acquisition by CMDR *A. baumannii* against SNAPPs was not observed. Comprehensive analyses using 3D super-resolution fluorescence imaging (**Fig. 1a**), transmission electron microscopy (**Fig. 1b**), competitive inhibition assays, membrane disruption measurements, and gene expression studies revealed that the antimicrobial activity of SNAPPs proceeded *via* a multi-modal mechanism of cell death by outer membrane destabilization, unregulated ion channel formation in the cytoplasmic membrane and induction of the apoptotic-like death pathway. Overall, SNAPPs show great promise as low-cost and effective antimicrobial agents, and can potentially combat various infectious diseases that are caused by Gram-negative bacteria resistant to conventional antibiotics.

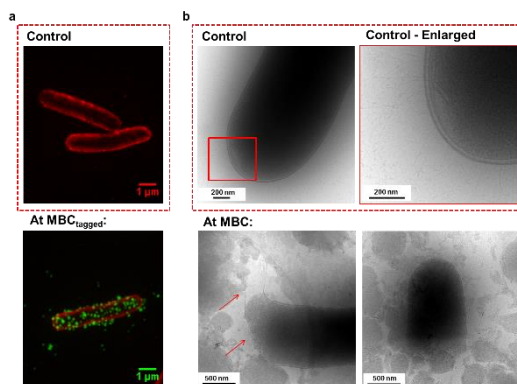


**Figure 1.** (a) 3D-Structured illumination microscopy images of *Escherichia coli* (membrane stained red) before and after treatment with fluorescently-tagged star (green) at the lethal concentration. (b) Cryo-TEM images of *E. coli* before and after treatment with untagged stars at the lethal dose.

<sup>1</sup> G. Taubes, *Science* **2008**, 321, 356-361.

<sup>2</sup> Z. Xu, M. T. Flavin, J. Flavin, *Expert Opin. Invest. Drugs* **2014**, 23, 163-182.

<sup>3</sup> S. J. Lam, N. M. O'Brien-Simpson, N. Pantarat, A. Sulistio, E. H. H. Wong, Y. Chen, J. C. Lenzo, J. A. Holden, A. Blencowe, E. Reynolds, G. G. Qiao, **2016**, *submitted*.

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## Monodispersed Polymer Encapsulated Superparamagnetic Iron Oxide Nanoparticles for Cell Labeling

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Superparamagnetic iron oxide nanoparticles (SPIONs) are the most studied nanoparticles due to their relatively low cost and simple synthesis process.<sup>1</sup> In this work, polymer encapsulation of raw SPIONs ferrofluids was carried out by free radical emulsion polymerization using macro-RAFT copolymers as stabilizers. The iron oxide cores, with positively charged surface at the pH of encapsulation, were dispersed in a solution of negatively charged random macro-RAFT copolymer by ultrasonication. Uniform polymer encapsulated SPIONs were produced by feeding the mixture of methyl methacrylate (MMA) and n-butyl acrylate (BA) monomers into the dispersion containing water soluble initiator 4,4'-azobis(4-cyanopentanoic acid) (V501). Shell thickness could be controlled by adjusting the amount of coating monomer added. Polymer encapsulation of SPIONs aligned in a magnetic field produced polymer encapsulated SPION rods. Monodispersed sterically stabilized crosslinked polymer encapsulated SPIONs were synthesized using diblock and random macro-RAFT copolymers as stabilizers (**Figure 1**). Rhodamine labeled SPIONs were found to be non-cytotoxic and suitable for cell labeling. The coated magnetic particles also show considerable promise for use in cell separation.

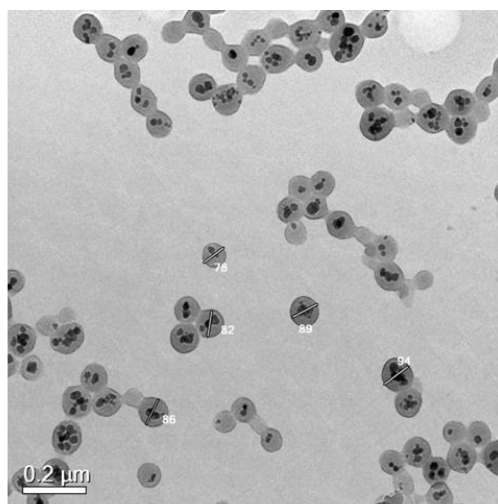
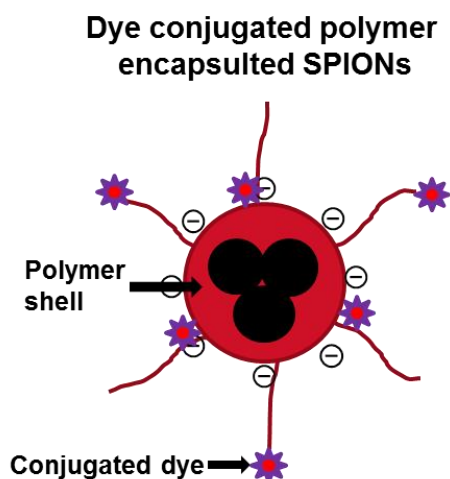


**Figure 1.** Monodispersed sterically stabilized polymer encapsulated iron oxide superparamagnetic particles for cell labelling.

<sup>1</sup> Gupta, A. K.; Gupta, M. *Biomaterials* **2005**, 26, 3995.

### Duc Nguyen

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Personal History:

Since year 2002: KCPC, Sydney University

Research interests: RAFT, nanocomposites, pigment dispersion





# Understanding the effect of the drug on the performance of drug carriers

Manuela Callari<sup>1,2</sup>, Paul L. De Souza<sup>2</sup>, Aditya Rawal<sup>3</sup>, Martina H. Stenzel<sup>1\*</sup>

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Micelles are widely applied for drug delivery purposes. It is commonly assumed that these self-assembled structures take on a core-shell structure with a sharp boundary at the interface. In this paper, we have developed solid state NMR analysis as an analytical tool to understand morphology of these nanoparticles in more detail. As a model system, we employed block copolymers based on a glycopolymers and a block that allows the conjugation of drugs, here platinum drugs. Micelles with high and low platinum loading were prepared and their performance was evaluated *in vitro*. Despite have the same shell, a fructose based polymer that can target GLUT-5 receptors on cell surfaces, the micelles behaved differently with the low Pt content showing better cell uptake and higher toxicity. Solid state NMR revealed the interior structure of the micelle (Figure 1). The high loading micelles showed domain separation which indicates a core-shell type of structure where the platinum complex is localized in the hydrophobic core whereas the sugar forms the outer coating. The low loading micelles showed a gradient type of structure where the hydrophobicity decreases from the centre of the micelles towards the outside where water molecules are still present. We could therefore show that solid state NMR is a valuable tool to understand the morphology of the micelles, which was essential to explain the *in vitro* activity of the two carriers.

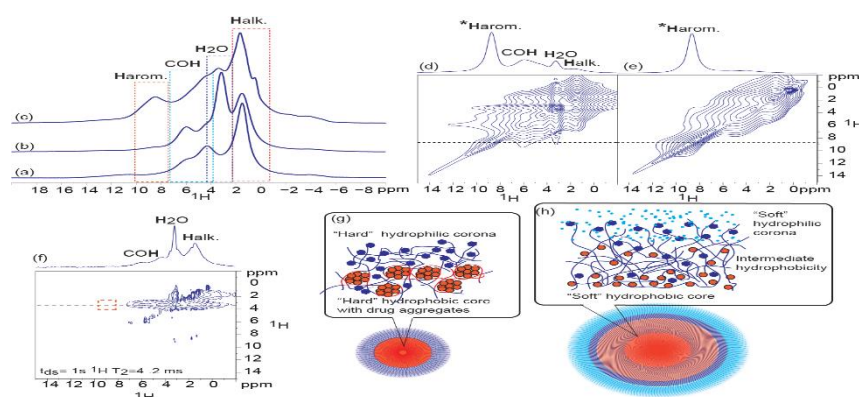


**Figure 1: Phen-Pt dispersion and dynamics in the polymer nanocarriers.** <sup>1</sup>H ultrafast NMR of the neat polymer and platinum-polymer conjugates at 60 kHz MAS. **f**, 2D <sup>1</sup>H-<sup>1</sup>H correlation spectra of the low drug loading with a 4.2 ms <sup>1</sup>H T<sub>2</sub> filter (before the first evolution period) and 1 s of <sup>1</sup>H spin diffusion. **g**, and **h**, schematics of the platinum-polymer assembly in the high and low drug loading systems.

In summary, the structural analysis of the platinum loaded micelles lead to fundamental insights that open up a shift in the current paradigm of maximizing the drug loading of nano-carriers, i.e. in certain cases less can be more. For the present case of platinum-polymer conjugation driven self-assembled micellar systems, higher loading of the platinum complex actually reduces cellular uptake, release and cytotoxicity. The reduced efficacy of the HL micelles can be assigned to reduced potential for interaction between the sugar in the smaller-“hard” micellar structure and the cell wall. At the same time, these micelle show a slower release due to the formation of Phen-Pt aggregates.

## Martina Stenzel

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Personal History

1999 PhD in chemistry at the University of Stuttgart, Germany

2009-2014 ARC Future Fellow

Since 2012 Professor

Research interests: nanoparticles for drug delivery, glycopolymers, macromolecular metal complexes



## Polymer Nano- structures for MRI and PET imaging

Thomas P. Davis<sup>1</sup>

<sup>1</sup>ARC Centre of Excellence for Bio-Nano Science

Theranostics, a combination of drug delivery and imaging, is an emerging field, as progress could potentially lead to triggered drug release at precise physical locations (tracking), and provide instantaneous feedback on physical responses to delivery events. Building responses to different delivery vectors using a range of imaging techniques, for example, computer tomography (CT), magnetic resonance imaging (MRI), positron emission tomography (PET), and fluorescence imaging has been described in a wide range of recent publications. MRI responsiveness remains an attractive target for theranostic nanoparticle design, as MRI scanners are readily available in large public hospitals, and MRI scans do not subject patients to ionizing radiation. Two different types of MRI contrast agents (CAs) are commonly employed to build theranostic devices; both CAs have the ability to shorten the surrounding water protons longitudinal (T<sub>1</sub>) and transversal (T<sub>2</sub>) relaxation time to improve MRI contrast. Gadolinium-based CAs yield positive contrast as indicated by an increase in image brightening, while, in contrast, iron oxide nanoparticle-based CAs provide a negative contrast— leading to image darkening. Presently, the most commonly employed CAs in the clinic are based on gadolinium (Gd). As Gd is highly toxic, Gd is chelated with small organic compounds to maintain solution stability while obviating direct interactions of Gd with biological media. Small organic CAs have a number of shortcomings: i) a short biocirculation times, ii) low contrast at high magnetic fields, and iii) systemic delivery (nonspecificity). Recent published work on incorporating Gd into polymeric nanostructures has provided some encouragement that nanoparticle-based CAs can overcome the present application drawbacks. Polymeric nanoparticles can be easily tuned to produce sizes from 10 to 200 nm with bespoke morphologies, potentially to take advantage of the enhanced permeation retention (EPR) effect, for enhanced targeting. In addition, polymeric nanoparticles can be easily decorated with targeting moieties to further improve specific targeting, and they can encapsulate a greater number of Gd per nanoparticles to enhance local contrast. In another significant advantage, encapsulating Gd in nanoparticle structures reduces molecular tumbling, significantly increasing the relaxivity of Gd at high magnetic fields.

In this talk I will discuss the design of novel polymeric agents for imaging applications, and show how potentially combining novel enhanced imaging with VR technology as a data interpretation and manipulation interface can enhance both conventional high street imaging and also help in education and training.

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### Tom Davis

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Personal History:

2014 –ARC Laureate Fellow and Director CBNS

2013 - Monash-Warwick Professor

Research interests: Polymer science, NanoBio Science

## Translational Research in Polymer Science and the Factory of the Future

*Bronwyn Fox<sup>1</sup>*

<sup>1</sup>Swinburne University of Technology, Factory of the Future, Faculty of Science, Engineering and Technology  
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The manufacture of polymer products is expected to change radically in the coming years. Swinburne's Factory of the Future is working with multinational companies and Australian SMEs in their supply chains to develop a flexible pilot line to demonstrate state of the art automated production of new polymer matrix composite products. This facility has been intentionally designed to create a platform to demonstrate to Australian manufacturers the principles and benefits of "Industry 4.0" concepts and will exploit Swinburne's existing strengths in design, visualisation, robotics, automation and big data. Industry 4.0 is described as the fourth industrial revolution and utilizes recent advances in communications and digital technologies including the "internet of things" to automate and control production processes in order for them to run seamlessly and smoothly.

In the past two decades, cure cycle times for carbon fibre reinforced polymer matrix composites have been dramatically reduced from hours to minutes (1). At the same time, the global market for carbon fibre composites is confidently expected to grow exponentially to \$36 billion by 2020 (2) (3) with key applications in aerospace, automotive, defence, wind energy and civil engineering. Now that the component cure cycle times are no longer the rate limiting step in the production process, the remaining challenge is to increase production rates by replacement of manually intensive steps with automated production.

An example of the state of the art for automated composite production is the BMW i3 battery electric vehicle manufacturing plant in Leipzig, Germany. The i3's carbon fibre composite "life module" is built using several of the latest technologies for industrial automation. However, it is noted that whilst this process represents a paradigm shift in advanced composite manufacture, there are still significant manufacturing challenges which further innovative research can help overcome. The cure cycle time for the carbon fibre composite components is 8 minutes (automotive targets are under 1 minute to compete with sheet metal forming) and this process requires some manual intervention.

This presentation will describe the current capabilities of Swinburne's Factory of the Future and will highlight the future directions for the development of new capabilities.

### **Professor Bronwyn Fox**

Director Factory of the Future

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Personal History:

|           |                                                                    |
|-----------|--------------------------------------------------------------------|
| 2016-     | Swinburne University of Technology, Director Factory of the Future |
| 2001-2015 | Deakin University, Institute for Frontier Materials, Carbon Nexus  |
| 1996-2000 | Australian National University, School of Engineering (PhD)        |
| 1993-1996 | CRC for Polymers (CSIRO)                                           |

Research interests: carbon fibre, polymer composites, structure-property relationships in polymers

## **Systematic production of high quality and low cost cellulose nanofibres and thier potential applications**

Nasim Amiralian, Darren J Martin

*Australian Institute Bioengineering and Nanotechnology (AIBN), Corner College and Cooper Rds (Bldg 75),  
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We have recently discovered<sup>1-3</sup> that a unique, very high grade of cellulose nanofiber can be readily and cost-effectively produced from endemic Australian spinifex grasses from the *Triodia* genus. These fascinating extremophile grasses, with over 30 million years of evolution, have presented us with a most unique source for nanocellulose. Our fibrillation process was performed using a mild pulping procedure followed by a low mechanical energy treatment or a mild sulphuric acid hydrolysis. This novelty means that our method of fibrillation, and more importantly, this source of cellulose (*Triodia* includes 69 species) has the potential to directly address the current technological bottlenecks that have so far limited the widespread translation of nanocellulose technology into more widespread applications. This presentation will give an overview of our group research and technology development activities related to spinifex nanofiber production, with potential applications including filtration membranes, polymer composites and low cost, renewable carbon fibres. Furthermore, the challenges which an early career researcher faces in order to translate the outcome of applied research into the commercial products while pursuing the fundamental science will be discussed.

### **Nasim Amiralian**

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2014 – PhD in Nanomaterials Engineering at the Australian Institute for Bioengineering and Nanotechnology,  
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2014 – 2015 – Postdoctoral research Fellow at the Australian Institute for Bioengineering and Nanotechnology,  
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Since 2016 - Advance Queensland Research Fellow

Research interests: Processing and structure-property performance of novel materials, renewable-based polymers and nanocomposites

## **Spinifex Nanocellulose: the Commercialisation Story So Far**

Phil Nelson<sup>1</sup>

<sup>1</sup>UniQuest Pty. Ltd.

The presentation will focus on the commercialisation aspects of the Spinifex nanocellulose technology developed under the supervision of Professor Darren Martin at the University of Queensland.

Nanocellulose derived from the Australian Spinifex grass plant is a soft, flexible nanomaterial with exceptional toughness versus nanocellulose products derived from other sources. The process for extracting the nanofibres is relatively low cost due to the unique structure of the plant. These technical features played an important role in development of the application and commercial strategies for the material.

Application selection and focus are vital in the development of platform technologies where the list of potential applications is long. Nano-reinforced elastomers were chosen as the “beach head” application due to a number of factors including competitive positioning and the perceived value proposition the technology could offer.

The commercial strategy was built on a solid understanding of the needs of the industry and the presentation will detail how the value proposition of the technology was validated, how to contact potential partners and the how their interest was gained.

### **Phil Nelson**

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Personal History:

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2004-2009 Technical Manager, Nanotecture Ltd. (UK)

2010-2012 Business Development Manager, Hovione Farmaciencia SA (Portugal)

Since 2012 UniQuest Pty Ltd, Director, Commercial Engagement (Materials)

Research interests: N/A

## Patents and your research project: a case study from a polymer-related industry

Matthew J Overett<sup>1</sup>

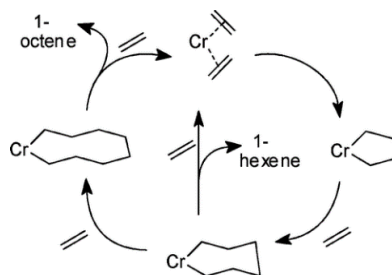
<sup>1</sup>Phillips Ormonde Fitzpatrick

For many researchers, intellectual property considerations only come into play once technical successes have been achieved and attention turns towards commercialisation. Yet a proper appreciation of the intellectual property system, and patents in particular, can be a major source of advantage and a driver of innovation throughout a research project, from early fundamental scientific investigations to commercialisation and beyond.

Some key aspects where patents can inform and influence your research project include:

- Gaining a full understanding of the state-of-the-art in your technology area: in many fields of technology, the most up-to-date and commercially relevant information is not publically available in scientific journals, but only in patent applications.
- Monitoring your competitors: tracking and benchmarking your competitors' activities, as revealed by their patent filings, can point to promising new avenues for exploration and progress.
- Framing your experimental programme: exploring the breadth of your scientific discovery to support a patent filing can lead to unexpected breakthroughs and progress.
- Solving emerging technical challenges: solutions to new problems arising during development and commercialisation may become apparent by considering patents in adjacent technical fields.
- Leveraging your IP to access resources: a patent application can assist you to gain access to the external expertise you need to progress your research project.
- Evaluating your freedom to operate: it is essential to develop an early appreciation of the patent landscape, which may constrain the commercialisation of your research.

The author will discuss these and other lessons acquired during a 12 year R&D career, in which he participated in the development of a new 1-octene production process, from bench scale investigations to commercialisation. 1-Octene, an important co-monomer for commercial grades of linear low density polyethylene and elastomers, is traditionally produced non-selectively via a broad-range linear alpha olefin (LAO) process. In 2003, building on earlier research into ethylene trimerisation catalysts, the first ethylene tetramerisation catalyst (selective 1-octene production) was discovered by researchers at Sasol Technology.<sup>1</sup> The role that patents played in the discovery, early development and ultimate commercialisation of the tetramerisation technology will be used as a case study to highlight the value that the patent system can add throughout the life-cycle of a research project.



(a) US 7,511,183 to Sasol Technology; (b) M.J. Overett et al, *J. Am. Chem. Soc.* **2005**, *127*, 10723; (c) A. Bollman et al *J. Am. Chem. Soc.* **2004**, *126*, 14712

### Matthew Overett

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#### Personal History:

2000-2003     PhD, University of Cape Town  
2003-2015     Sasol Technology R&D (South Africa and USA)  
Since 2015     Phillips Ormonde Fitzpatrick (Melbourne)

Professional interests: working with local inventors in polymer and related fields to protect their IP

# Cationic RAFT Polymerization: Synergetic Developments in Radical and Cationic Polymerizations

Mineto Uchiyama, Kotaro Satoh, and Masami Kamigaito

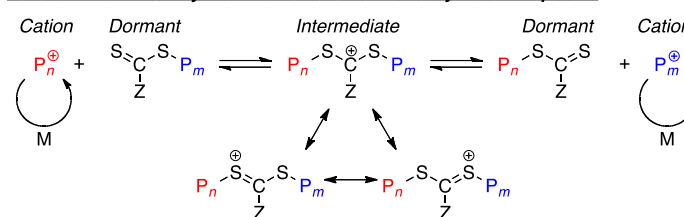
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Recent developments in living or controlled polymerization have enabled the synthesis of a wide variety of well-defined polymers with precisely controlled structures. Most of these modern living/controlled polymerizations are based on reversible activation of dormant covalent polymer terminal into the growing species. Therefore, there are many similarities in concepts and strategies for controlling polymerizations irrespective different growing species.

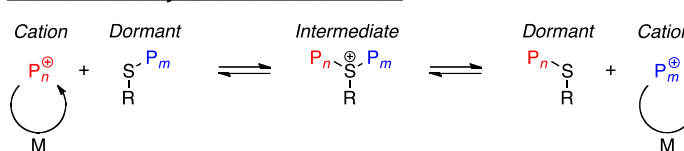
Reversible addition-fragmentation chain transfer (RAFT) polymerization has been originally developed for controlling radical polymerization via reversible chain transfer between propagating radical species and covalent dormant species such as thiocarbonylthio compounds.<sup>1</sup> We have recently found that RAFT mechanism also works for controlling cationic polymerization of various cationically polymerizable monomers such as vinyl ethers and styrene derivatives using similar thiocarbonylthio compounds (**Scheme 1**).<sup>2</sup>

The polymerization proceeds via reversible addition of the cationic propagating species to the thiocarbonylthio group to form the cationic intermediate stabilized by the sulfonium ion. Furthermore, thioethers and phosphorous esters are also effective for controlling cationic polymerization via similar reversible degenerative chain transfer (DT) or RAFT mechanisms.<sup>3,4</sup> These novel controlled cationic polymerizations have thus provided new strategies based on RAFT or DT chemistry and will lead to synergetic developments in radical and cationic polymerization for precision polymer synthesis.<sup>5</sup> This lecture will mainly cover the recent developments in cationic RAFT polymerizations.

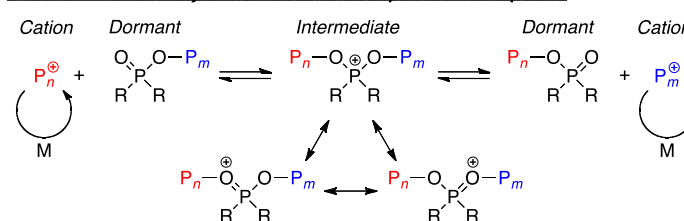
## 1. Cationic RAFT Polymerization with Thiocarbonylthio Compound



## 2. Cationic DT Polymerization with Thioether



## 3. Cationic RAFT Polymerization with Phosphorous Compound



**Scheme 1.** Cationic RAFT and DT polymerizations

<sup>1</sup> G. Moad, E. Rizzardo, S. H. Thang, *Aust. J. Chem.* **2012**, *65*, 985-1076.

<sup>2</sup> M. Uchiyama, K. Satoh, M. Kamigaito, *Angew. Chem. Int. Ed.* **2015**, *54*, 1924-1928.

<sup>3</sup> M. Uchiyama, K. Satoh, M. Kamigaito, *Macromolecules* **2015**, *48*, 5533-5542.

<sup>4</sup> M. Uchiyama, K. Satoh, M. Kamigaito, *Polym. Chem.* **2016**, *7*, 1387-1396.

<sup>5</sup> T. K. McKenzie, Q. Fu, M. Uchiyama, K. Satoh, J. Xu, C. Boyer, M. Kamigaito, G. G. Qiao, *Adv. Sci.* **2016**, *3*, 1500394.

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## Approaches to the RAFT synthesis of multifunctional, multi-armed polymers

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<sup>3</sup>Beijing University of Chemical Technology

The paper will describe approaches to the synthesis of functional star polymers (with potential application as materials, in therapeutic delivery and in other applications) by RAFT polymerization pointing out their scope, advantages and limitations and will be illustrated by examples from recent CSIRO research. These approaches include the arm-first method based on RAFT cross-linking (co)polymerization of a divinyl monomer using macroRAFT agents (1,2), which has recently been applied in the synthesis of Mikto-arm polymers. The other approaches considered are core-first methods (3) where the core may be a precisely defined multi-RAFT agent, a hyperbranched copolymer prepared by RAFT crosslinking (co)polymerization of divinyl monomer using a (small)

RAFT agent or RAFT copolymerization mediated by a so-called “RAFT inimer” (a molecule comprising both RAFT

agent and monomer functionality). Factors which lead to well defined stars vs networks and structures in between will be discussed (4).

1. Wei, X.; Moad, G.; Muir, B. W.; Rizzardo, E.; Rosselgong, J.; Yang, W.; Thang, S. H. *Macromol. Rapid. Commun.* 2014, 35, 840–845.

2. Wei, X.; Gunatillake, P. A.; Moad, G.; Rizzardo, E.; Rosselgong, J.; Yang, W.; Thang, S. H. *Science China Chemistry* 2014, 57, 995-1001.

3. Wilson, J. T.; Postma, A.; Keller, S.; Convertine, A. J.; Moad, G.; Rizzardo, E.; Meagher, L.; Chiefari, J.; Stayton,

P. S. *The AAPS Journal* 2015, 17, 358-369.

4. Moad, G. *Polym. Int.* 2015, 64, 15-24

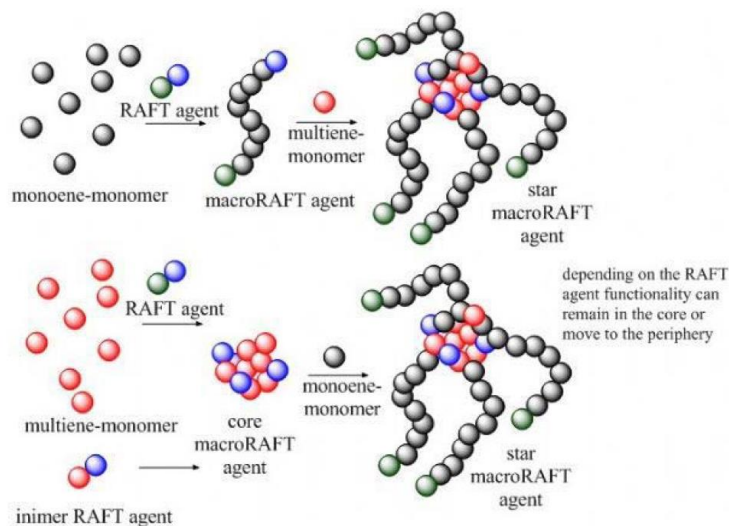


**Figure. Approaches to the RAFT synthesis of multi-armed polymers**

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# Oxygen Tolerant PET-RAFT Polymerisation in Flow

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Conventional and controlled radical polymerisation systems suffer a severe limitation in that polymerisation cannot be effectively performed in the presence of molecular oxygen. Exposure to even small quantities of molecular oxygen leads to inhibition periods, as well as rate retardation and loss of control. Furthermore, applications of polymerization systems in which the reaction media needs to be exposed to the atmosphere is ever increasing. This is especially relevant in photoinduced polymerisation systems, which host a large variety of specialised applications from coatings, printing and medical applications to high-tech applications in stereolithography and nanotechnology, among many others.

Although strategies to reduce oxygen inhibition in photoinduced polymerization systems have been well documented, the development of controlled radical polymerization systems which are inherently tolerant to molecular oxygen has scarcely been demonstrated. Photoinduced electron/energy transfer-reversible addition-fragmentation chain transfer (PET-RAFT) polymerization is one such example of a controlled radical photopolymerization process that exhibits tolerance to molecular oxygen. The development of this technique has led to diverse and efficient systems which use non-toxic and inexpensive photocatalysts in ppm quantities, and which are capable of producing controlled macromolecules under fully open conditions.<sup>1</sup>

In this work we present the production of various macromolecules by the PET-RAFT technique in a flow reactor, using cheap, safe, and commercially available low-energy LED lights as the energy source and without the need for deoxygenation. Without the need for stringent deoxygenation protocols the handling and preparation of reagents becomes effortless, and the production of large quantities of well-defined macromolecules via flow techniques becomes facile. As such, these systems provide an exceptional industrial advantage over previous systems, as an entire process step is eliminated.

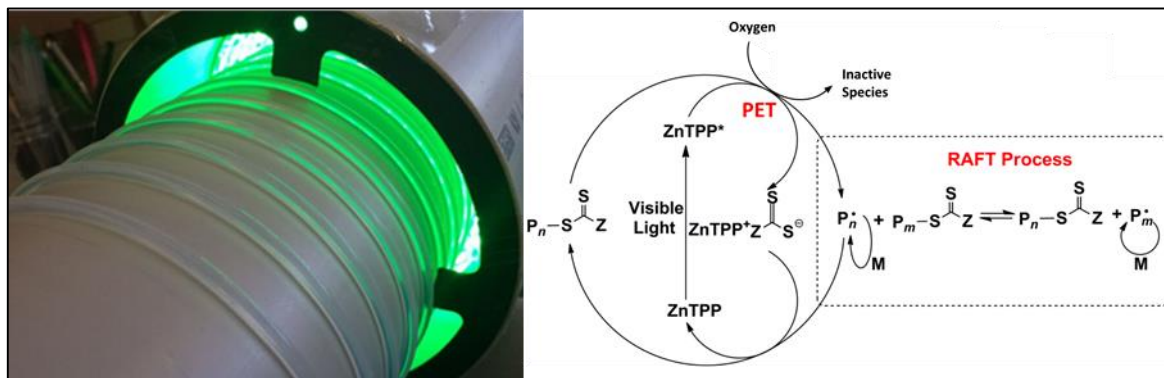


**Figure 1.** Flow reactor with green LED light (left) and PET-RAFT mechanism with ZnTPP as photocatalyst (right)

<sup>1</sup> Shanmugam, S.; Xu, J.; Boyer, C. *Journal of the American Chemical Society* **2015**, *137*, 9174

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## Exploring the impact of UV irradiation on traditional RAFT systems

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<sup>3</sup>Research School of Chemistry, Australian National University

Reversible Fragmentation Chain Transfer (RAFT) polymerization has grown in utility and prominence within the field of polymer science since its discovery in 1998<sup>22</sup>. The ability to employ a wide range of reaction conditions including choice of organic and aqueous polymerization mediums and a tolerance to diverse functionality present on vinyl monomers are the key advantages of RAFT. Traditional RAFT agents have the inherent limitation of usually only being suited to one of two generic classes of vinyl monomer, which have been designated as More Activated Monomers (MAMs) and Less Activated Monomers (LAMs). Advances in this area include the use of acid/base switchable RAFT agents featuring a pyridyl functionality which can successfully polymerize both MAMs and LAMs to yield poly(MAMs-b-LAMs)<sup>23</sup>, and “F-RAFT” agents which theoretically should provide reasonable control over both classes of monomer<sup>24</sup>.

Recently interest has been renewed in other methods of activating RAFT agents, predominantly using light of various wavelengths to achieve direct photolysis and via photo-redox catalysts, along with activation via acids and bases for their application to cationic polymerizations<sup>25</sup>. Employing wavelengths from the ultraviolet to blue allows for direct photolysis of RAFT agents, thus effectively making them function as Iniferter compounds. This means they become capable of Initiating, Transferring and Terminating the polymerization. Moreover in these systems both spatial and temporal control has been demonstrated, with RAFT agent concentration and irradiation power having an influence on polymerization kinetics. Within these systems, irradiation power and wavelength also determine the presence or absence of an inhibition period and the overall control obtained over the polymerization. However, little work has focused on the application of light as an external stimulus on a RAFT polymerization that is run under traditional conditions; i.e. at elevated temperatures with a traditional radical source like AIBN.

Herein we report on our investigations into how UV light affects a typical RAFT polymerization run with an aromatic xanthate as a chain transfer agent. We explore how the application of UV irradiation can be leveraged to accelerate the polymerization of both methyl acrylate and vinyl acetate as models for MAMs and LAMs, respectively. It was found that whilst accelerated rates of reaction are seen for both monomers with the application of UV light, significant deviation from the theoretically predicted molecular weights was not seen, along with only a negligible effect on the polydispersity of polymers formed. The polymerizations showed normal pseudo-first order kinetics, only deviating slightly at very high conversions. The living character of the polymers formed was confirmed via successful chain extension experiments for both PMA and PVAc under both light and dark conditions. We also explore the formation of a range of block copolymers of the type poly(MAM-b-LAM) under traditional conditions and with UV stimulation, yielding interesting insights into the efficiency and limitations of these compounds and approaches.

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## Forced Gradient Co-polymers using Continuous Flow Reactors

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Gradient co-polymers are predicted to have improved physical characteristics, especially those related to surfactants and dispersants, but also for use in more diverse applications such as fiber optics and lenses.<sup>1,4</sup> While the preparation of gradient polymers has received considerable attention using batch reactors,<sup>1–6</sup> the preparation of usable quantities of gradient co-polymers using continuous flow reactors has been hampered by the need to vary the composition of the monomer feedstock continuously during the reaction, and to sample the reaction to determine composition.

Any changes to the composition of the feedstock at the entry of the flow reactor does not effect the composition of available monomer at points further down the reactor tubing.

A reactor that allows for addition of a monomer feedstock continuously at all points along the length of the reactor tubing would allow for the preparation of a subset of forced gradient co-polymers in continuous flow reactors, allowing for the scale-up and bulk preparation of these polymers.

We report here our initial investigation of preparing forced gradient co-polymers using the RAFT methodology in continuous flow reactors.

<sup>1</sup> M. Kryszewski, *Polym. Adv. Technol.*, 1998, **9**, 244–259, doi:10.1002/(SICI)1099-1581(199804)9:4<244::AID-PAT748>3.0.CO;2-J.

<sup>2</sup> K. Matyjaszewski, M. J. Ziegler, S. V. Arehart, D. Greszta, and T. Pakula, *J. Phys. Org. Chem.*, 2000, **13**, 775–786, doi:10.1002/1099-1395(200012)13:12<775::AID-POC314>3.0.CO;2-D.

<sup>3</sup> R. Wang, Y. Luo, B. Li, X. Sun, and S. Zhu, *Macromol. Theory Simul.*, 2006, **15**, 356–368, doi:10.1002/mats.200600007.

<sup>4</sup> U. Beginn, *Colloid Polym. Sci.*, 2008, **286**, 1465–1474, doi:10.1007/s00396-008-1922-y.

<sup>5</sup> A. Feldermann, A. Ah Toy, H. Phan, M. H. Stenzel, T. P. Davis, and C. Barner-Kowollik, *Polymer*, 2004, **45**, 3997–4007, doi:10.1016/j.polymer.2004.04.016.

<sup>6</sup> X. Li, S. Liang, W.-J. Wang, B.-G. Li, Y. Luo, and S. Zhu, *Macromol. React. Eng.*, 2015, **9**, 409–417, doi:10.1002/mren.201500012.

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# Substrate independent approach for growing low fouling polymer brushes to inhibit bacterial adhesion

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Biofilm caused infections of implants has always been a prevalent and persistent problem in the clinic mainly due to the high susceptibility to infection and difficulties in conventional treatment because the biofilm will protect the bacterial from the host immune system and antibiotics<sup>1, 2</sup>. In order to prevent biofilm formation, two main coating methods have been investigated—antibiotic and anti-bioadhesion based coatings<sup>3, 4</sup>. However, due to the antibiotic coatings usually suffer from limited loading, poor bioavailability (if covalently grafted), bacterial resistance and possible adverse effect due to accumulation of dead bacteria become nutrition etc, we chose anti-bioadhesion coatings for this study.

In this work, a substrate independent approach for growing low fouling monomers from the substrate was introduced. Firstly, bromine initiator, allyl 2-bromo-2-methylpropionate, was immobilized on a silicon wafer using plasma polymerization. The halogen species in the monomer retained its reactivity to allow successful brush grafting to be performed later. Then, [2-(methacryloyloxy)ethyl]dimethyl-(3-sulfopropyl)ammonium hydroxide (SBMA), a zwitterionic monomer which was chosen due to its low fouling ability and biocompatibility, was grafted from the surface via atom transfer radical polymerization (ATRP). Successful was confirmed by XPS characterization. The thickness of the grafted polymer could be controlled between 10 nm and 20 nm via different reaction time.

Furthermore, we have shown that the zwitterionic monomer grafted surfaces indeed showed low fouling properties against both protein solution (10% FBS) and the initial adhesion of bacteria (UPEC and PAO1). The surfaces with the thicker brush resulted in improved low fouling properties due to their thicker hydration layer via electrostatic interaction.

1 J.L. Pozo and R. Patel, *Clinical Pharmacology & Therapeutics*, 2007. 82(2): p. 204-209.

2 J.M. Schierholz and J. Beuth, *Journal of Hospital Infection*, 2001. 49(2): p. 8793.

3 P. A. Norowski and J. D. Bumgardner, *Journal of biomedical materials research. Part B, Applied biomaterials*, 2009. 88(2): p. 530-543.

4 K. Bazaka, M.V. Jacob, R. J. Crawford and E. P. Ivanova, *Applied microbiology and biotechnology*, 2012. 95(2): p. 299-311.

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# Single-Enzyme Nanogels: Synthesis and Immobilization

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Enzymes are not only nature's catalysts but are also essential to many synthetic processes. In the latter, it is often desirable to operate in the presence of organic solvents and at elevated temperatures. However, with respect to activity, enzymes are macromolecules which are sensitive to many environmental parameters such as pH, ionic strength, temperature, and the presence of organic solutes, to name but a few. Immobilization onto solid substrates or within crosslinked enzyme aggregates is typically employed to stabilize enzymes. Yet, common protocols are often lacking precision in terms of enzyme surface chemical modification and may lead to considerable variations in efficiency, depending on the nature of the enzyme.

Single-enzyme encapsulation was shown to stabilize various enzymes at elevated temperatures and in the presence of solvents.<sup>1</sup> It generally consists in carrying out a simple crosslinking polymerization in the presence of an enzyme previously modified in a way that it carries vinyl groups and acts as multifunctional crosslinker (**Fig. 1**). However, precise preparation protocols are scarce and advanced applications have not been pursued. We have recently set out to investigate the synthesis of single-enzyme nanogels (SENs) and particularly the parameters which may affect the formation of well-defined particles with several common proteins.<sup>1</sup>

We found new experimental conditions where pre-functionalization of the enzymes is not necessary, which makes the process simpler and consequently more attractive for applications. In addition, we showed that SENs could be immobilized onto surfaces without specific modification and rapidly by thio-Michael addition (**Fig. 2**).<sup>2</sup> We are currently working on further immobilization protocols, on imparting additional features to SENs, and on applying them in various contexts.

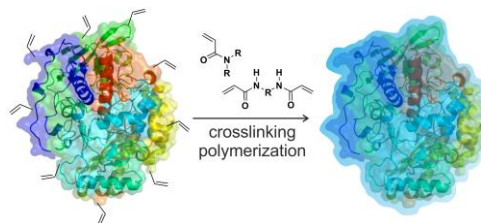


**Figure 1.** Single-molecule encapsulation of an acrylated enzyme by crosslinking polymerization at high dilution.

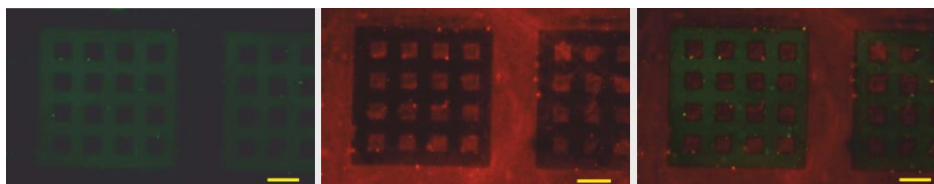


**Figure 2.** Patterns of fluorescent SENs obtained by thio-Michael addition-mediated microcontact printing.

- [1] M. Yan, J. Ge, Z. Liu, P. Ouyang *J. Am. Chem. Soc.* **2006**, 128, 11008; J. Ge, D. Lu, J. Wang, M. Yan, Y. Lu, Z. Liu *J. Phys. Chem. B* **2008**, 112, 14319.
- [2] A. Beloqui, A. Kobitski, J. Bürck, G. U. Nienhaus, G. Delaittre, submitted.
- [3] A. Beloqui, S. Baur, V. Trouillet, A. Welle, J. Madsen, M. Bastmeyer, G. Delaittre, *Small* **2016**, 12, 1716.

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# ‘Click’ thiol-yne hydrogels for MSC delivery for the suppression of immunomodulatory responses in osteoarthritic joints

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Injectable *in-situ* forming PEG-thiol/bispropiolate hydrogels (**Fig. 1**) have great potential for treating osteoarthritis due to its minimally invasive delivery, fast gelation time, convenience in processing, high tenability. The hydrogels also have the ability to release therapeutic molecules produced by the entrapped MSC to suppress immunomodulatory responses of diseased cartilage. In this project, hydrogels were formed from PEG-thiol and PEG-bispropiolate precursors via thiol-yne ‘click’ chemistry. Gelation times of this hydrogel were within minutes at both room and physiological temperatures, which provide a suitable timeframe for injection and localising entrapped molecules/cells. Rheology and unconfined compression tests have shown that this hydrogel possessed storage moduli and compressive moduli of 9.9-14.4 kPa and 7.5-31 kPa respectively at equilibrium swelling conditions (articular native cartilage has a compressive modulus of 500-1,000 kPa). The mechanical properties of this hydrogel are tunable by altering the concentration and molecular weight of the initial reagent to cater tissue regeneration purposes.

Factors secreted by MSC such as TGF- $\beta$  have been previously shown to play an important role in cartilage regeneration and inhibit inflammatory process in osteoarthritic joints<sup>1,2</sup>. Drug release experiments incorporating model drugs that mimic TGF- $\beta$  (FITC-dextran and FITC-albumin) were performed and confirmed that the drug molecules were able to be transported out of the hydrogel by means of diffusion.

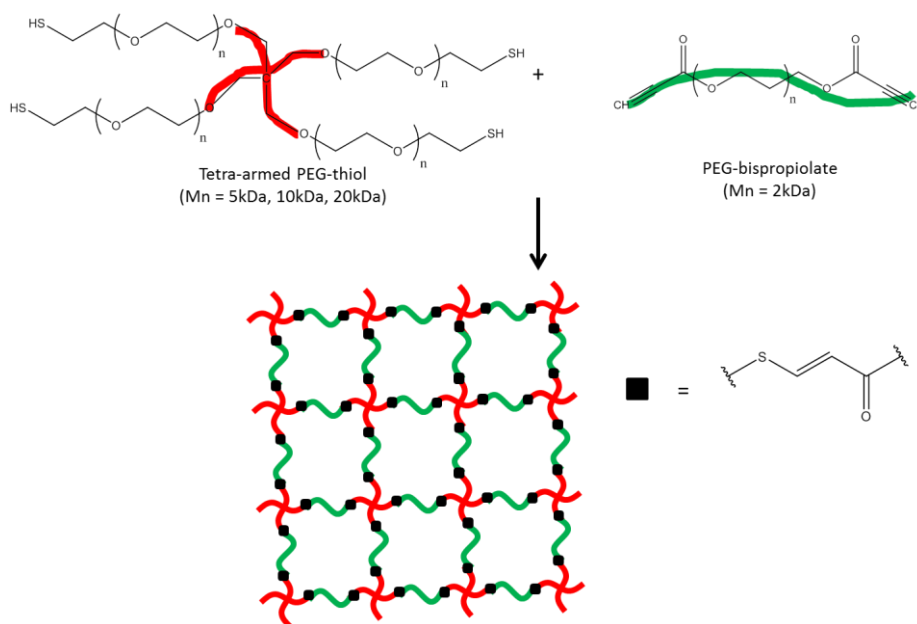


**Figure 1.** Chemical structure of PEG-thiol/bispropiolate hydrogel

<sup>1</sup> E.N.B. Davidson, P.M. van der Kraan and W.B. van der Berg, Osteoarthritis and Cartilage (2007) **15**, 597-604

<sup>2</sup> G.M. van Buul *et al.* Osteoarthritis and Cartilage (2012) **20**, 1186-1196

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## **A PEDOT microfibrinous scaffold for neural recordings**

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Presently there are serious limitations in obtaining stable, consistent and long-term stimulation or recording from current neural interfaces. This is mainly due to an unwanted glial response. The scar increases resistance to current flow as well as the distance between the neurons and the current source<sup>1</sup>. Consequently, higher electrical currents or voltages are required, which result in damage to neural tissue due to Faradaic reactions. Additionally, most neural interfaces have limited proximity to neurons (due to their 2D recording sites), which is greatly compromised once a glial response occurs<sup>2</sup>. In the present study, a microfibrinous 3D neural electrode is under development using an electrospun template. The fibrous template was imbued with electroactive properties by coating with PEDOT, a conductive polymer, via vapour phase polymerization. The electrical properties of the PEDOT-coated substrates were studied in terms of sheet resistance measured by a 4-point probe, and scaffold morphology was investigated by scanning electron microscopy.

Scaffold biocompatibility was assessed through *in vitro* assay of rat primary hippocampal neurons using patch clamp electrophysiology, calcium imaging and immunocytochemistry. Importantly, the *in vitro* assays demonstrated normal spontaneous neural activity, indicating robust neural networking, up to 40 days in culture on PEDOT scaffolds. Additionally, an *in vivo* assay was performed to assess the capabilities of such scaffold as a recording electrode. Accordingly, this neural electrode will provide intimate electrical contact with neurons with reliable long term stimulation and recording.

1. Vadim, S.P., A.T. Patrick, and M.R. William, Response of brain tissue to chronically implanted neural electrodes. *Journal of Neuroscience Methods*, 2005. 148(1): p. 118.
2. Warren, M.G., E.N. Sharon, and V.B. Ravi, Implanted Neural Interfaces: Biochallenges and Engineered Solutions. *Biomedical Engineering*, 2009. 11(1): p. 1-24.



## Micropatterned and drug eluting bioelastomers for improved cardiomyocyte beating

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Heart muscle has limited ability to regenerate following myocardial infarction, which has led to the rapid development of new cell therapies. However a major obstacle is poor survival and engraftment of the transplanted cells. Therefore implantable scaffolds are currently being investigated to assist in cell integration and to ultimately improve tissue function. Bioelastomers based on polyglycerol sebacate (PGS) have mechanical properties that closely match the dynamic properties required for myocardium tissue engineering. In this study, we used lithography to introduce micropatterns of different dimensions on the surface of PGS films. Cardiomyocytes cultured on the micropatterned films exhibited morphologies similar to native cardiac tissue (Fig. 1) resulting in more homogeneous beating characteristics.

To further enhance cardiomyocyte survival, a small molecule tropomyosin-related kinase B (TrkB) agonist was polymerised into the bioelastomer. Release of the mimetic is controlled via hydrolysis of the PGS facilitating long-term and sustained delivery. Controlled release of the mimetic from PGS intensified beating of the cardiomyocytes *in vitro*.

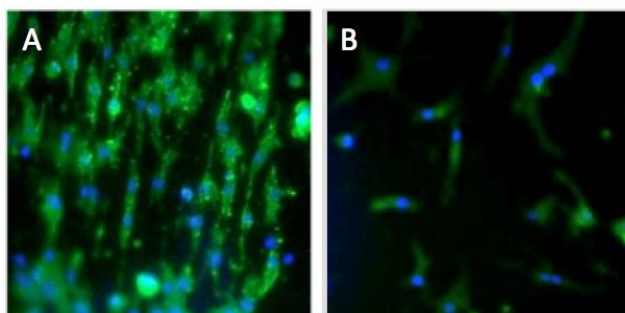


**Figure 1** Fluorescent microscopy images of cardiomyocytes cultures on A) micropatterned and B) flat PGS showing improved alignment on patterned substrates. Cells are stained with troponin I (green) and DAPI (blue).

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# Kappa- carrageenan, genipin and chitosan cross-linked composites for drug release in simulated gastrointestinal fluids.

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$\kappa$ -carrageenan was extracted from the red sea algae, *Kappaphycus alvarezii* using 0.1 and 0.3 M KOH. Blends for drug release were prepared by mixing varying amounts (20, 30 and 40 v/v %) of  $\kappa$ -carrageenan with chitosan and cross-linked with genipin. The optimal genipin concentration for the blend was determined by AFM and swelling studies. FT-IR showed that  $\kappa$ -carrageenan was immiscible with chitosan and hence mostly physical crosslinking had occurred. Swelling and drug release investigated in simulated gastrointestinal fluid (pH of 1.2, 4.5, 6.8) showed similar trends and were responsive to the ionic composition, pH of the media, and the amount of  $\kappa$ -carrageenan in the blend. Blends containing increasing amounts of  $\kappa$ -carrageenan, treated with 0.3 M KOH showed increased and faster drug release. The bio polymeric membranes were responsive to stimuli for the drug release and serves as a possible oral delivery route for Quetiapine fumarate.

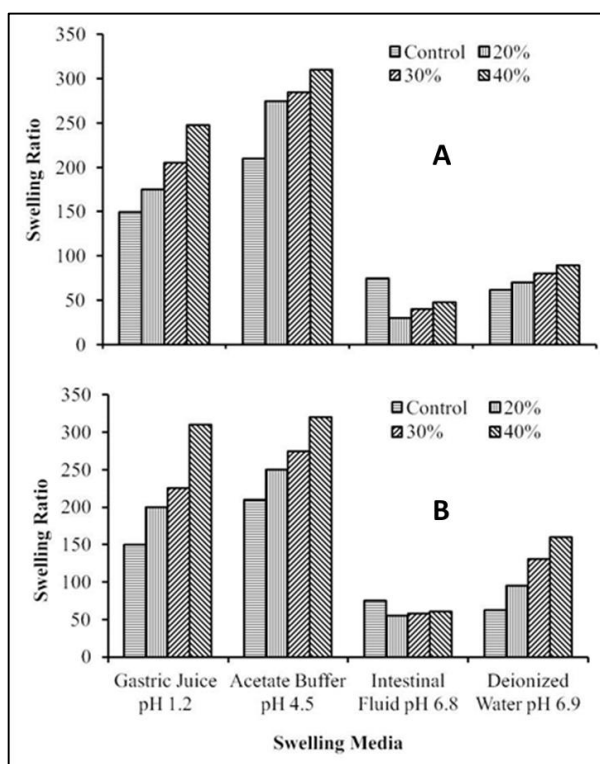


**Figure 1.0: Swelling Profile of the pH responsive composites; 0.1 M treated (A) and 0.3 M Treated (B). Swelling was responsive to changes in the stimuli for the bio polymeric membranes. For the 0.3 M treated KOH, swelling was more rapid than the 0.1 M treated.**

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## Biodegradable Antimicrobial Polymer Containing Pendant Guanidines

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Some microorganisms are serious concern posing great threats to human health which leads to an increasing demand for antimicrobial agents or materials in many application areas.<sup>1</sup> In recent years, polymeric biocides have drawn significant attention as a class of antimicrobial agents with mechanism of action that could potentially overcome bacterial resistance development.<sup>1</sup> Most of these polymers containing cationic functionalities interact with negatively charged microbial cell wall. However, non-biodegradability of the polymer backbone could raise some concerns in future.<sup>2</sup> On the other hand, the guanidine group is common and capable antimicrobial moiety due to its cationic nature. Polyguanidine mimics the arginines found in natural antimicrobial peptides (AMPs) and many researchers have produced antimicrobial polymers through incorporation of this functional group. For instance, Locock at CSIRO reported a series of guanidine based amphiphilic polymethacrylates using RAFT technique and demonstrated its great potency against both *G+* *S. aureus* and fungi *C. albicans*.<sup>3-6</sup>

In this work, we aim to replace polyacrylate with biodegradable/biocompatible aliphatic polycarbonate backbone. The alkyne functionalized six member ring cyclic carbonate monomer was synthesised and used as the building block for further guanidine attachment via CuAAC click reaction. The metal-free, organocatalytic ring-opening polymerization (ROP) was employed to prepare polymers. The antimicrobial activities were investigated on the possible effect of polymers with different molecular weight, guanidine loading density, and hydrophilic/hydrophobic balance etc. The results provided insight to determine optimised composition of polymer for greater antimicrobial efficacy.

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<sup>2</sup> V.P. Shastri, *Current pharmaceutical biotechnology*, 2003, 4(5), 331-337.

<sup>3</sup> K. Locock, et al., *Biomacromolecules*, 2013, 14(11), 4021-4031.

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<sup>5</sup> K. Locock, et al., *Pure and Applied Chemistry*, 2014, 86(8), 1281-1291.

<sup>6</sup> K. Locock, et al., *ACS Macro Letters*, 2014, 3(4), 319-323.

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# Stable and non-toxic superparamagnetic iron oxide nanoparticles as multimodal (PET/SPECT-MRI) imaging probes

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Several approaches to labeling superparamagnetic iron oxide nanoparticles (SPIONs) with radionuclides as imaging agents for PET/MRI or SPECT/MRI have been reported via either chelation or chelate-free techniques.<sup>1,2</sup> However, challenges still remain in the simple and robust synthesis of fully dispersed and stable radiolabeled SPIONs with tightly bound radionuclides.

In our present study, sterically stabilized SPIONs of two different core diameters, 10 and 25 nm, are investigated as potential candidates for multimodal imaging PET/SPECT – MRI techniques. These SPIONs have been shown in our previous study to be excellent negative contrast agents with very-high  $T_2$  relaxivities of 368 and 958 s<sup>-1</sup>.mM<sup>-1</sup>, respectively.<sup>3</sup> In addition, these SPIONs have demonstrated non-cytotoxicity and an excellent bio-distribution profile *in vivo*.<sup>4</sup> The SPIONs were successfully radio-labeled with both <sup>57</sup>Co or <sup>67</sup>Ga in aqueous acetate buffers. The radiolabeling efficiency was strongly dependent on temperature and time for both raw ferrofluid (surface charge stabilized iron oxide cores) and sterically stabilized SPIONs. No detectable loss of radionuclides from the particles to the aqueous phase in various physiological media was observed. The radiolabeled sterically stabilized SPIONs were fully re-dispersed and remained stable in a physiological buffer solution such as saline or PBS (see **Figure 1**), which is a crucial property for an injectable formulation in biomedical applications.

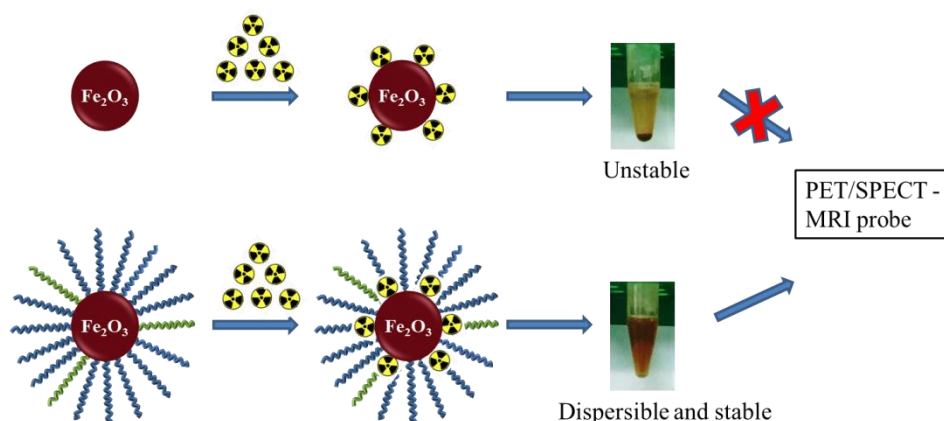


**Figure 1.** Radio-labelling of SPIONs

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2. F. Ai; C.A. Ferreira; F. Chen; W. Cai, *Wiley Interdiscip Rev Nanomed and Nanobiotechnol* **2015**.

3. B.T. Pham; N. Jain; P.W. Kuchel; B.E. Chapman; S.A. Bickley; S.K. Jones; B.S. Hawkett, *Int J Nanomedicine* **2015**, 10, 6645.

4. Pham et al., to be submitted.

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## Co-delivery system with NO (nitric oxide) and Drug (doxorubicin) using nanoparticle to combat Doxorubicin-Resistant Ovarian Cancer Cells.

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Multidrug resistance (MDR) is a phenomenon by which tumour cells that have been exposed to cancer chemotherapy develop cross-resistance to structurally and functionally unrelated compounds.<sup>1</sup> One of the most studied in cancer cell models has been the over expression of energy dependent ATP binding cassette (ABC) transporter proteins, such as P-glycoprotein (P-gp) and MDR-associated proteins (MRP1-6), which causes tumour cells to become resistant to a variety of anticancer drugs.<sup>3</sup> One of the strategies to reverse MDR is the co-administration of an anticancer agent with an inhibitor of these efflux pumps.<sup>2</sup> In human epithelial colon doxorubicin-resistant cells (HT29-dx), a relationship between nitric oxide (NO) and antineoplastic drug resistance has been investigated. Specifically doxorubicin-sensitive and doxorubicin-resistant cells could show a different capacity to produce NO and that feature might contribute to drug resistance.<sup>2,3</sup> HT29-dx overexpress P-gp or MRP3 glycoproteins, which play a significant role in clinical drug resistance.<sup>3</sup> MRP3 contains a higher number of tyrosine residues, which are potentially susceptible to nitration. The greater nitration observed in HT29-dx cells could be explained by the higher expression of MRP3 in this population, induced by exogenously applied NO donors in the MRP3 transporter, consequently leading to doxorubicin (DOX) accumulation and cytotoxic effects in HT29-dx cells.<sup>2,3</sup> This suggests that NO can reduce the number of functionally active MRP3 transporters perhaps by altering the proper conformation of the proteins at a site crucial for drug transport, which affects doxorubicin efflux and consequently increases intracellular DOX accumulation.

However the direct use of NO in combination with chemotherapy agents (DOX) has many limitations. In particular, NO is a gaseous radical species that lacks both stability and specificity. In order to overcome the challenge of controlled NO and DOX delivery/release, NO and DOX could be incorporated into nanoparticles or other polymeric systems.

This project will explore the synthesis of different polymers conjugated to NO donor (S-nitrosothiol) and DOX respectively, which can release NO and DOX into the target cells. Human ovarian carcinoma cell lines (SKOV3) and DOX-resistant (SKOVCR) ovarian carcinoma cell lines were studied in different expression of ABC transporter proteins/MDR-associated proteins, enabling the further investigation of the relationship between NO and DOX accumulation in SKOVCR/SKOV3 cell lines. Polymeric nanoparticle systems for co-delivery NO/DOX will be developed. This nanoparticle system will improve the controlled release/delivery of NO/DOX into target cell lines such as SKOV3/SKOVCR, which leading to further enhancement of cytotoxicity in SKOVCR cell lines.

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### SulHwa YU

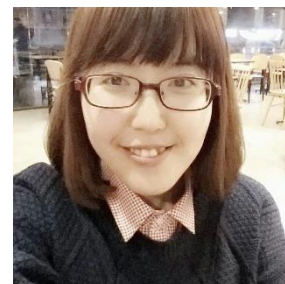
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# Robust and biodegradable hydrogels for delivery of hydrophobic drugs

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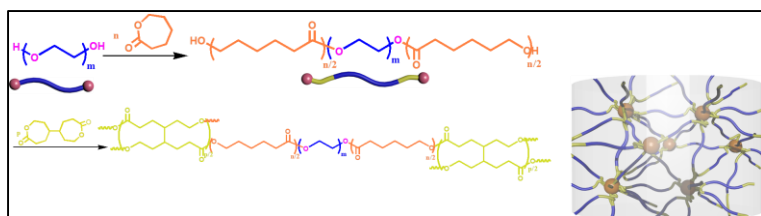
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Hydrogels made of three-dimensional polymeric networks have been widely employed in biomedical applications, such as drug delivery<sup>1</sup>. The applications, however, are significantly hindered by the mechanical weakness and fragility of conventional hydrogels. In terms of drug delivery applications, the hydrogel devices are required to sustain and recover from various deformations in a mechanically dynamic environment<sup>2</sup>, so as to prevent the premature leakage of embedded therapeutic molecules. The other inherent drawback of traditional hydrogels for drug delivery is limited loading capacity for hydrophobic drugs due to their hydrophilic nature. A new hydrogel system with hydrophobic domains has been therefore developed to address these two challenges. The hydrogels presented here were synthesized via one pot ring-opening polymerization (ROP), starting from di-functional poly(ethylene glycol) (PEG) with addition of poly( $\epsilon$ -caprolactone) (PCL), using [4, 4'-bioxepane]-7, 7'-dione (BOD) as a cross-linker (Scheme 1). The uniform distribution of poly(BOD), highly cross-linked PCL hydrophobic domains acting as macro-cross-linkers, can distribute applied stresses and enhance the ultimate strength of the network<sup>3</sup>. Also, they could be used as loading sites for small hydrophobic therapeutics. The linear PCL moieties improve the hydrogel toughness due to the energy dissipation<sup>4</sup> arising from the hydrophobic associations.

Uniaxial compressive testing was performed to reveal increased strength of the hydrogels with increases in the content of hydrophobic domains (Fig. 1A). Upon the addition of linear PCL chains, the hydrogels are able to maintain their integrity up to high compression and over cyclic loading and unloading (Fig. 2B). The excellent anti-fatigue resistance was quantified by the hysteresis recovery rate of the subsequent cycles, with only slight decrease in hysteresis energy as compared to that of the first cycle (Fig. 2C). The drug loading capacity of these hydrogels is heavily influenced by their swelling properties as well as hydrophobicity. The in vitro drug release study illustrated high stability of these drug depots at neutral pH and a fast release profile under simulated in vivo conditions, consistent with the hydrogel degradation profile. Finally cytotoxicity testing of both plain hydrogels and drug loaded hydrogels indicated low toxicity of the hydrogels and effective drug delivery.

In conclusion, a library of PEG-PCL-BOD hydrogels with well-defined structures has been developed, featuring tuneable toughness, hydrophobic drug loading capacities, biodegradability and drug release profile.



**Scheme 1.** Fabrication of PEG-PCL-BOD hydrogels in a two-step ROP process

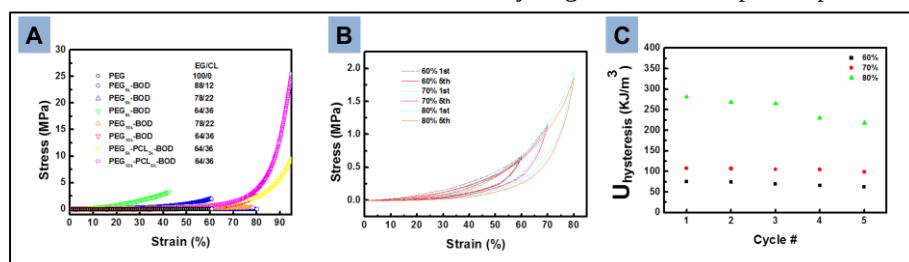


**Figure 1.** (A) Compressive mechanics of PEG-PCL-BOD hydrogels with different compositions; (B) Cyclic loading-unloading curves of PEG-PCL-BOD hydrogel at different strains; (C) Dependence of hysteresis energy on the number of repeated cyclic tests for PEG-PCL-BOD hydrogel

<sup>1</sup> T. R. Hoare and D. S. Kohane, *Polymer*, **2008**, 49, 1993-2007. <sup>2</sup> Y. D. Wang, G. A. Ameer, B. J. Sheppard and R. Langer, *Nat Biotechnol*, **2002**, 20, 602-606. <sup>3</sup> X. Zhao, *Soft Matter*, **2014**, 10, 672-687. <sup>4</sup> Q. Chen, L. Zhu, C. Zhao, Q. M. Wang and J. Zhang, *Adv. Mater.*, **2013**, 25, 4171-4176.

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## Macromolecular Donors for Controlled and Stimulated H<sub>2</sub>S Release

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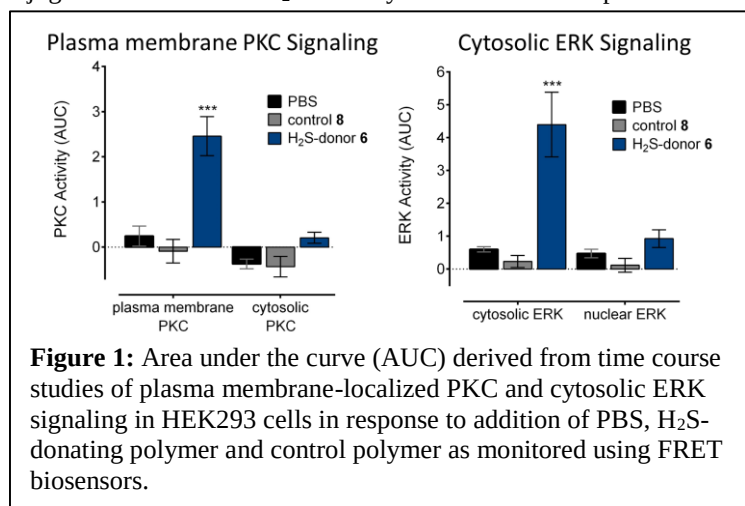
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Hydrogen sulfide (H<sub>2</sub>S) has gained recognition and attention over the last two decades as an important gasotransmitter that is involved in both physiological and pathophysiological conditions. Increasing studies have highlighted its crucial role in the human body, as a mediator and regulator of many physiological processes.<sup>1-4</sup> For example; H<sub>2</sub>S has been shown to have a role in inducing vasodilation, in reducing blood pressure and eliciting an anti-inflammatory responses. Related functions include cardioprotective, neuroprotective and gastroprotective effects. Moreover, a correlation between low levels of H<sub>2</sub>S-generating enzymes with various disease states and pathological changes, such as hypertension, Alzheimer's disease and gastric mucosal injury have been demonstrated. Therefore exogenous delivery of H<sub>2</sub>S may be useful in the management of a large number of diseases.<sup>5</sup>

Reagents that are able to release H<sub>2</sub>S have been applied as useful research tools in studies of biological effects as well as drug development.<sup>6,7</sup> To date inorganic sulfide salts, such as Na<sub>2</sub>S and NaSH (which form H<sub>2</sub>S immediately upon solvation in physiological buffers) have been widely used. However they do not reproduce the effects of continuous and controlled biosynthesis. For therapeutic value, a system that mimics the controlled biosynthetic release of H<sub>2</sub>S with an extended time of delivery and in a physiological concentration range that is of benefit to cells would be ideal. This presentation will report on the development of novel polymeric conjugates for controlled H<sub>2</sub>S delivery which address the problems associated with the therapeutic application of

H<sub>2</sub>S-releasing reagents.

Constructs that release H<sub>2</sub>S slowly via hydrolysis or a thiol- (L-cysteine) mediated mechanism will be presented.<sup>8</sup> We will highlight cellular tests using FRET-based biosensors that demonstrate the ability of donors to trigger spatiotemporally confined cell signaling events (Fig.1). Studies using an H<sub>2</sub>S-selective fluorescent probe in live cells confirmed release of H<sub>2</sub>S from a macromolecular donor over physiologically relevant time scales consistent with signaling observations. Recent constructs that incorporate biodegradable linkers capable of releasing H<sub>2</sub>S in an intracellular



environment will also be highlighted.

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# Structured polymer nanoparticles – access to new hybrid materials and nanomedicines

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The possibility to precisely engineer at the nanoscale enables the synthesis of materials with tailored chemical composition and physicochemical properties. Within this space, the modular synthesis of cylindrical polymer brushes provides a new and versatile class of materials that are very powerful in molecular templating and sensing,[1,4-7] and which emerge as promising polymer nanomedicines.[2,3,5] Our research focusses on these nanomaterials which we are using to establish new ways of assembling meta and hybrid materials as well as design the next-generation of smart drug delivery devices.

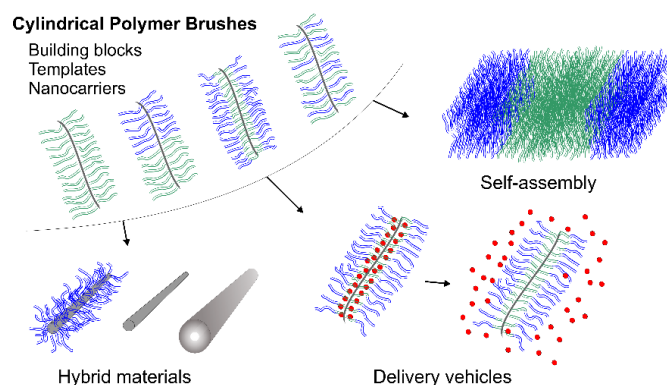


**Figure 1.** Various applications of cylindrical polymer brushes.<sup>1</sup>

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# Enhanced drug delivery system based on Fructose-coated nanoparticles for the treatment of breast cancer

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Breast cancer always seems to spread cancer cells to lungs, bones or brain, therefore, induces extended damages throughout the whole body. Fructose-based glycopolymer has been applied as the hydrophilic moiety to improve the cellular uptake of nanoparticles, due to the nature of triple negative breast cancer cells which overexpress the fructose transporter GLUT5 that can be used as targeting sites.<sup>1</sup> Ruthenium complex—RAPTA-C has been proved to be remarkably effective at suppressing the growth of metastasis.<sup>2</sup> RAPTA-C still faces several problems, such as inadequate bioavailability, low water solubility, and high dosage needed to achieve desired therapeutic outcome with inevitable toxicity to healthy organs, degradation before reaching targeting sites. With the well-known therapeutic benefits of RAPTA-C, identifying an adequate delivery system with high drug loading ratio and high accumulation attraction towards tumour cells would be critical in having a significant contribution towards the chemotherapeutic treatment of metastatic cancer. Polylactic acid (PLA) applied as the hydrophobic block which is able to degrade into smaller polymer chain that will ensure the successful release of macromolecular drug. 2-chloroethyl acrylate (CEA), 1-O-acryloyl-2,3:4,5-di-O-isopropylidene- $\beta$ -D-fructopyranose (1-O-AiPrFru), PLA macro raft agent and azobisisobutyronitrile (AIBN) as initiator were combined in N, N-dimethylacetamide (DMAc) and undergone at 70°C. The obtained glycopolymers were undergone deprotection of hydroxyl groups in trifluoroacetic acid (TFA). The structure and molecular weight and distribution were determined via NMR and GPC. Macromolecular drugs were obtained by attaching free drugs to the polymer chains by three steps procedure. In order to make it easier to attach PTA ligand to the polymer, iodide was substituted with chloride of CEA firstly via Finkelstein reaction. To attach PTA and ruthenium dimer separately can avoid losing arene ligand of the dimer. PTA and polymer were combined in dimethyl sulfoxide-d6 (DMSO-d6) for 7 days. The clear solution was analysed via <sup>31</sup>P NMR until conversion from PTA to PTA-polymer was stable. The ruthenium dimer was subsequently added in DMSO solution. This reaction is quick and the orange solution was analysed via <sup>31</sup>P NMR. Micellisation of block copolymer can form the nanoparticles for delivering drugs. (**Fig.1**) MCF-7, MDA-MB-231, fibroblast and macrophage cell lines were used for Cell work. Cellular uptake of different micelles was determined by inductively coupled plasma mass spectrometry (ICPMS) via calculating the concentration of the ruthenium drug in cells. The efficiency of inhibiting migration of cancer cells was studied by invasion assay.

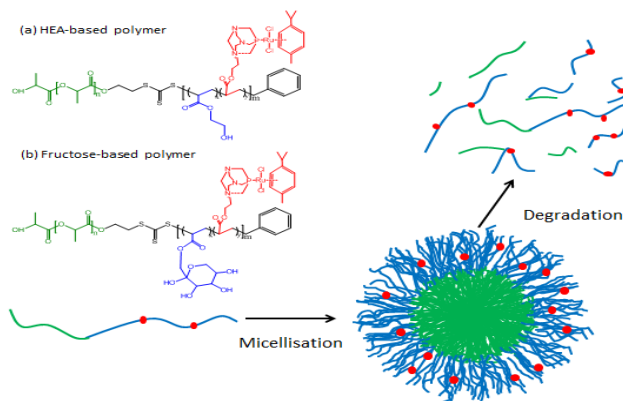


**Figure 1.** RAPTA-C conjugated polymer self-assembled into micelles in aqueous solution and drug release by PLA block degradation.

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<sup>2</sup> B. M. Blunden, M. H. Stenzel, *Biomacromolecules* **2013**, 14 (12), 4177-4188

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Research interests: Polymer materials, Glycopolymer, Nanoparticles for drug delivery, Ruthenium drugs

# Magnetic Alignment of Polymeric Nanofibers with Embedded Sterically Stabilized Iron Oxide Nanoparticles for Potential Cell Scaffolds

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Electrospinning (**Fig. 1-A**) is a versatile technique to fabricate nanofibers from a wide range of synthetic and natural polymers or polymer composites. This has been shown as a potential technique to engineer scaffolds for tissue engineering including bone, cartilage, ligament, skeletal muscle, vascular and neural, and as carriers for the controlled delivery of drugs, proteins, and DNA.<sup>1</sup>

Herein, polyvinyl alcohol (PVA) solution (6.5%, w/w, with average molecular weight of 115,000 g/mol) with or without 0.82% (by weight) sterically stabilized superparamagnetic iron oxide nanoparticles (SPIONs) were electrospayed in a 30 KV electric field. A permanent magnet was placed close to the collector to align the fibres in a desired direction. Characterisations of the nanofibers using optical microscopy, TEM and SEM showed the successful preparation of the PVA nanofibers with relative uniformity. The SPIONs were observed homogenously distributed along the nanofibers and enabled fibre alignment under a magnetic field (**Fig. 1-C**). The ability to align nanofibers in the same direction can yield potential scaffolds for cell adhesion and proliferation.

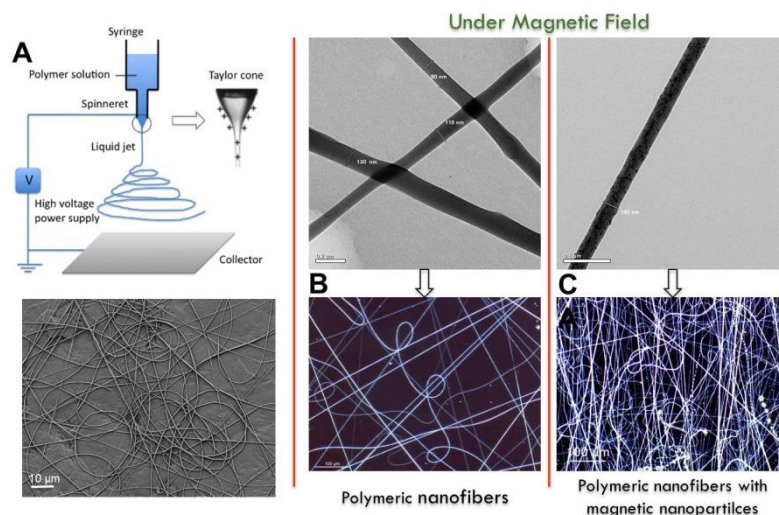


**Figure 1.** (A) Standard set-up of electrospinning<sup>2</sup> and a SEM image of the nanofibers without a magnetic field, (B) PVA nanofibers prepared in a magnetic field, and (C) PVA nanofibers containing the sterically stabilized SPIONs sprayed in a magnetic field.

<sup>1</sup> *Int J Nanomedicine*. **2006**, 1(1): 15–30

<sup>2</sup> *J.Polym.Biopolym. Phys.Chem.*, **2014** 2 (4): 62-66

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## Industry-driven collaborative polymer research

Ian J Dagley  
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When it completes its current funding under the Cooperative Research Centres (CRC) Programme in June 2017, the CRC for Polymers (CRCP) will have conducted 25 years of industry-driven collaborative research. The presentation will address: the scale and intensity of the collaboration; the evolution in the expectations of the stakeholders, including the CRC Programme; lessons learnt on how to best manage the process from initial contact with a potential industry partner through to a successful commercial outcome; the legacy of the training in collaborative research provided to its postgraduate students and postdoctoral fellows; and some case studies illustrating both the polymer research carried out and the commercial outcomes.

**Scale and intensity of the collaboration.** The activities of the CRCP have involved more than 40 multidisciplinary projects with a cumulative total of 48 participants comprising 24 businesses, 14 universities, three research organisations (CSIRO, ANSTO and DSTO), and seven end users. Most projects have included an industry partner and at least two universities, and up to six universities have participated in a single project. Over the period 1992-2015, the CRC has attracting \$65 million of CRC Programme funds and conducted over \$250 million of collaborative research activities.

**Evolution of the collaboration.** The evolution of the polymer research conducted by the CRCP has reflected the broader trends across the Australian polymer research community during the past 23 years (**Fig.1**). The CRC commenced as the CRC for Polymer Blends in July 1992 and, during this first term of funding, it broadened the scope of its research to become the CRC for Polymers. In its second funding period (1999-2005), it addressed the broader research needs of the plastics industry, conducting collaborative research on commodity polymers. In its third funding term, it expanded its activities to develop functional polymers to assist businesses in five growth sectors of the economy: manufacturing, biotechnology, mining, agriculture, and energy. The CRC is now in its fourth and final period of funding (2012-2017) which is allowing it to assist Australian manufacturing to develop products to meet emerging global needs in three areas: health therapies and delivery, water and food security, and low-cost solar energy, using enabling advanced polymer technology.

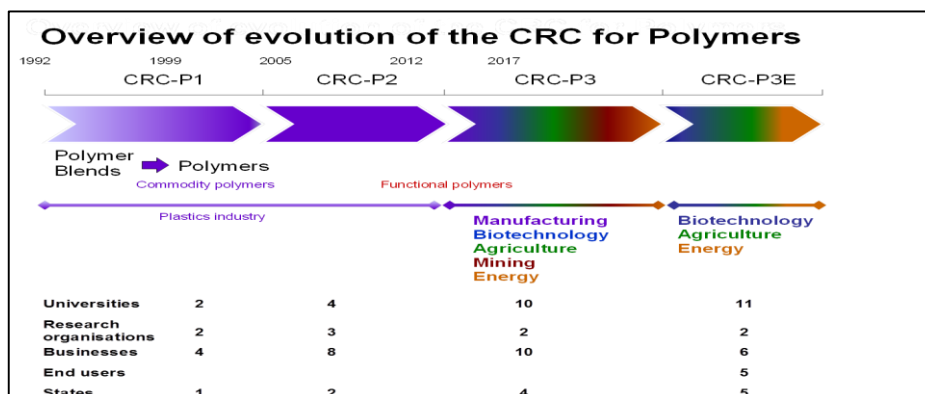


Figure 1: Overview of the evolution of the CRC for Polymers.

**Training researchers to collaborate across sectors.** The CRCP has provided funding support for 74 postgraduate students and ~ 100 postdoctoral researchers who have gained expertise in collaborative research.

**Commercial outputs and publications.** The commercially valuable technologies generated have been protected by more than 45 patent applications, and to date 17 of these technologies have been licensed to CRC participants.

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1991-1985: Chief Chemist, Pacific Dunlop subsidiary

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## How to attack a Patent/Application

*Richard Grant*  
Spruson & Ferguson

There are various options to attack an Australian patent or patent application, including:

- Third party observations: relevant prior art and associated arguments are submitted to the Examiner for consideration before grant of a patent. The submitter takes no further part in the proceedings ;
- Opposition to grant: after acceptance of a patent application, a statement is submitted by the opponent outlining reasons why a patent should not be granted. The applicant and opponent each have the opportunity to submit written evidence and then the matter is argued by both parties. This may be in writing or, more commonly, at a hearing before a Delegate of the Commissioner of Patents, after which a decision is issued by the Delegate;
- Request for reexamination: following grant of a patent, submissions are made outlining why the patent should not have been granted. The Patent Office will reexamine the patent in the light of those submissions. The submitter takes no further part in the proceedings ;
- Revocation proceedings: arguments and evidence are presented by both sides to a judge of the Federal Court regarding the validity or otherwise of a granted patent. Commonly such proceedings are a response to being sued for infringement.

The choice of which of these to choose depends on the stage at which the patent or application is at, the nature of any identified weak points of the application/patent and the budget. The advantages and disadvantages of the different options will be discussed, as will strategies for dealing with problematic third party patents/applications.

## Why Doesn't Excellent and Well Directed Research Always Yield a Product in the Market Place?

Brian Hawkett<sup>1</sup>

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The ultimate success for a researcher working with an industry partner is that that research should result in a product in the market place that is turning a good profit for the industry partner, making life more comfortable and sustainable for the population at large and returning good royalties to the research provider that he or she works for. That is the ultimate success. This talk will examine some of the reasons why good and inventive science does not always reach this ultimate research objective.

The speaker has 25 years of experience working as a contract and collaborative research provider to industry, initially as Technical Manager and then Technical Director of a company providing contract research services to the worldwide agrochemical industry and presently as Director of KCPC. KCPC has a research income of more than \$2M per year, much of which is derived from direct industry funding.

KCPC has been working with RAFT in disperse phase systems in the context of industrial research since about 2000, has worked continuously with 2 of its industry partners for more than 17 years and another for more than 14 years.

This talk will examine some of the reasons why good science generates good outcomes in collaborations with good industry partners but still does not result in products in the market place. Some of the reasons are:

Reactor requirements too sophisticated for available plant, product too complicated to manufacture<sup>1</sup>, outcomes no longer in line with company objectives, changes in company management structure, product unnecessarily complex, improvements in product performance insufficient to warrant new product launch, raw materials required not consistent with long term contracts, etc.

R. Korsmyer, *Regenerative Biomaterials* 2016, 143-147

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## SAXS Study of Oxalate Salts in the Presence of Poly(acrylic acid) Scale Inhibitors

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Scale-forming salts, such as calcium carbonate and calcium oxalate, constrain the operation of many important process streams, including sugar milling and thermal desalination.<sup>26</sup> The use of polyelectrolytes to alter the speciation and morphology of these salts is a common strategy for addressing the problem of scaling on surfaces in industrial systems and we have found that minor changes in the structure of these polyelectrolytes can give dramatic improvements in behaviour. We have previously found via XRD and SEM that poly(acrylic acids) prepared with different end-groups using Reversible Deactivation Radical Polymerisation, as well as altering the kinetics of scale formation, exert dramatic effects on the speciation and morphology of calcium oxalate, calcium carbonate, and calcium sulfate.<sup>2-4</sup>

We are currently using SAXS to follow the nucleation of calcium and barium oxalate under conditions approximating those applicable to sugar mill evaporators, where calcium oxalate scaling is a significant problem. Due to the scale in time and space of these critical nucleation events, SAXS is one of the few methods that can interrogate them meaningfully. Our previous work has been based on analysis of isolated crystals and measuring kinetics through changes in parameters which cannot be easily related to particle size (conductivity, ion-selective electrodes, turbidity, *etc.*<sup>5</sup>), so our understanding of nucleation events in these systems remains speculative, and SAXS experiments are an ideal technique to establish the mechanism of the polyelectrolyte activity on a more solid basis. Our ultimate goal is to follow the heterogeneous nucleation and growth of crystals in these systems *in situ*, but as a preliminary to this work we are examining crystallisation in bulk suspension using SAXS. We are also carrying out powder x-ray diffraction of isolated calcium and barium oxalate to identify how changes in the differential growth of the crystallographic faces may be responsible for the crystallite morphology through a study of size broadening of particular Bragg peaks.

The results of the SAXS study are consistent with our earlier observations of the calcium oxalate system and show that end-groups of moderate hydrophobicity inhibit crystal growth and strongly modify crystal habit, while end-groups of high-hydrophobicity strongly enhance nucleation in the continuous phase. This presentation will show our most recent SAXS and XRD results and their interpretation, with the implications for treatment of scale-forming salts in relevant industries.

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Since 2004            The University of New England

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# Understanding the Fundamentals – Investigating the Kinetics of a Novel Class of N-containing Monomers via PLP-SEC

Katrin B. Kockler<sup>1,2</sup>, Friederike Fleischhaker<sup>3</sup>, Christopher Barner-Kowollik<sup>1,2</sup>

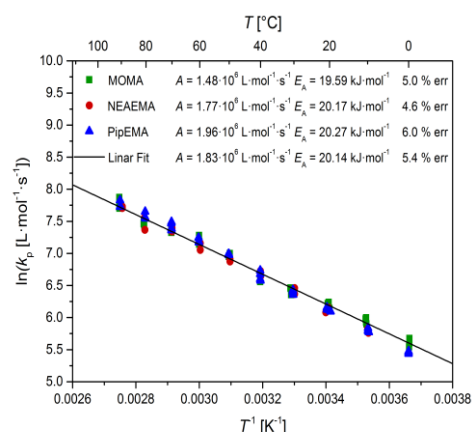
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Functional poly(methacrylates) are important materials for specialty as well as industrial bulk applications. Precision kinetic data are highly desired and the knowledge of temperature-dependant and monomer-specific propagation rate coefficients,  $k_p$ , as well as Arrhenius parameters,  $A$  and  $E_A$ , is mandatory for the synthesis of tailor-made polymers. In the current presentation, the IUPAC recommended pulsed laser polymerization – size exclusion chromatography (PLP-SEC) method<sup>1-2</sup> is used to obtain precision propagation rate coefficients, Arrhenius parameters, and Mark-Houwink-Kuhn-Sakurada (MHKS) parameters for monomers of a to date very rarely investigated group of nitrogen-containing methacrylates, i.e. 2-(N-ethylanilino)ethyl methacrylate, 2-morpholinoethyl methacrylate or 2-(1-piperidyl)ethyl methacrylate. Furthermore, the data are critically compared among each other to analyze for trends or family type behavior between the different monomers (**Fig. 1**).

**Figure 1.** Left: Propagation rate coefficients for 2(N-ethylanilinoethyl methacrylate (NEAEMA), 2(1piperidyl)ethyl methacrylate (PipEMA), and 2morpholinoethyl methacrylate (MOMA) at different temperatures. Right: Joint linear Arrhenius fit for the investigated nitrogen containing methacrylates.<sup>3</sup>



<sup>1</sup> M. Buback, R. G. Gilbert, R. A. Hutchinson, B. Klumperman, F.-D. Kuchta, B. G. Manders, K. F. O'Driscoll, G. T. Russell and J. Schweer, *Macromol. Chem. Phys.*, **1995**, 196, 3267-3280.

<sup>2</sup> C. Barner-Kowollik, S. Beuermann, M. Buback, P. Castignolles, B. Charleux, M. L. Coote, R. A. Hutchinson, T. Junkers, I. Lacik, G. Russell, M. Stach and A. van Herk, *Polym. Chem.*, **2014**, 5, 204-212.

<sup>3</sup> K. B. Kockler, F. Fleischhaker, C. Barner-Kowollik, *Polym. Chem.*, **2016**, in press.

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2008-2014 Studies of chemistry at the KIT Karlsruhe, Degree: Dipl.-Chem.

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Research interests: Free radical Polymerization, PLP, GPC, Kinetics

# Mechanical Energy to Electricity Conversion of Nanofibres and Conducting Polymers

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Recently, great efforts have been devoted to finding renewable, green energy sources to mitigate the over reliance on fossil fuels. Mechanical energy such as those from steam flow, wind, body movement or heartbeat exists everywhere. Conversion of those energy into electricity would require special materials and devices. Our recent studies have indicated that two types of materials, electrospun polyvinylidene difluoride (PVDF) nanofibres and conducting polymers, could be used as active layer for making mechanical energy-to-electricity conversion devices<sup>1-5</sup>.

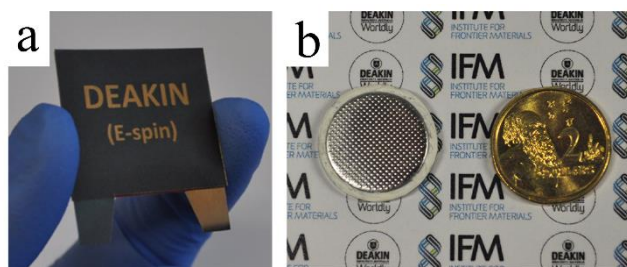


**Figure 1.** Photos of mechanical energy harvesters made of (a) electrospun PVDF nanofibre mat and (b) conducting polymer plate.

High electric field intensity and intensive fibre stretching during electrospinning have contributed to the formation of piezoelectric PVDF nanofibres with high  $\beta$  crystal phase content and oriented electric dipoles. Electrospun PVDF fibre mats with interconnected fibre structures showed not only improved energy conversion efficiency but also increased mechanical strength and structure integrity of during compression/decompression, making them very promising for making mechanical energy harvesters. Needleless electrospinning is efficient to produce PVDF nanofibres on a large scale with enhanced energy conversion behaviours. The nanofibre devices can even convert acoustic waves into electricity.

Most of mechanical energy harvesting technologies produce alternating current (AC) outputs, and an external rectifier has to be used to convert the AC into direct current (DC). The use of rectifier not only reduces the overall energy conversion efficiency but also increases complexity and difficulty in device miniaturization. However, our recent study has shown that self-rectified mechanical-to-electrical energy conversion can be attained when a free-standing conducting polymer plate is combined with two metal electrodes, which form an Ohmic and a Schottky contact separately with the plate. DC voltage with a large output current density can be generated. The electric power generated is sufficient to drive commercial electronic devices.

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10. C Lang, J Fang, H Shao, X Ding, T Lin, *Nature Communications*, 2016, DOI: 10.1038/ncomms11108.

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Research interests: Nanofibres, Electrospinning, Functional Fabrics, Polymer Materials

# Structural property and Mechanical behavior of Polypropylene –Elemental sulfur (S8) composites: Effect of sulfur loading

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Elemental sulfur is currently produced on the level of 70 million tons annually by petroleum refining, majority of which is used in the production of sulfuric acid, fertilizer and other chemicals. Still, over 6 million tons of elemental sulfur is generated in excess, which creates exciting opportunities to develop new chemistry to utilize sulfur as a feedstock for polymers. Development of new polymer composite materials using sulfur is not widely explored and remains an important challenge in the field. Polymer nanocomposites prepared by carbon nanotube, graphene, silica and other nanomaterials were well established. However, utilization of sulfur as filler in the polymer matrix could be an interesting study. This work is to presents the possibility of utilizing elemental sulfur as reinforcing fillers in the polymer matrix. In this study we attempted to prepare polypropylene/sulfur nanocomposite. The physical, mechanical and morphological properties of the newly developed composites were studied according to the sulfur loading. In the sample preparation, four levels of elemental sulfur loading (5, 10, 20 and 30 wt. %) were designed. Composites were prepared by the melt mixing process by using laboratory scale mini twin screw extruder at 180°C for 15 min. The reaction time and temperature were maintained constant for all prepared composites. The structure and crystallization behavior of composites was investigated by Raman, FTIR, XRD and DSC analysis. It was observed that sulfur interfere with the crystalline arrangement of polypropylene and depresses the crystallization, which affects the melting point, mechanical and thermal stability. In the tensile test, one level of test temperature (room temperature) and crosshead speed (10 mm/min) was designed. Tensile strengths and tensile modulus of the composites were slightly decreased with increasing in filler loading, however, percentage of elongation improved by more than 350% compared to neat polypropylene. The effect of sulfur on the morphology of polypropylene was studied with TEM and SEM techniques. Microscope analysis reveals that sulfur is homogeneously dispersed in polymer matrix and behaves as single phase arrangement in the polymer. The maximum elongation for the polypropylene can be achieved by adjusting the sulfur loading in the polymer. This study reviles the possibility of using elemental sulfur as a solid plasticizer in the polypropylene matrix.

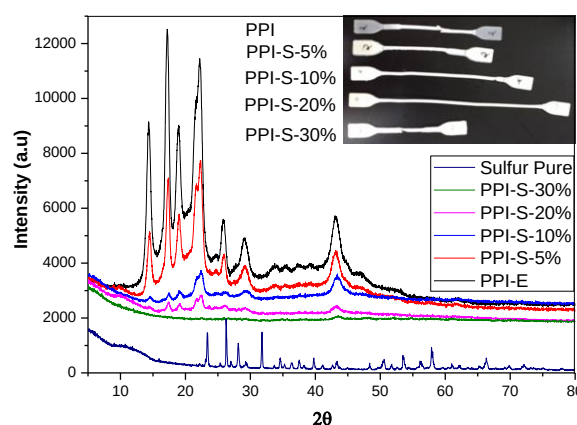


**Figure** XRD results for neat PPI, elemental sulfur and PPI-sulfur composites shows the decrease in crystallinity with the sulfur loading., insight picture shows digital images of the specimens of polymer composites subjected to mechanical test. The sulfur filled materials showed higher elongation with increasing amount of sulfur up to 20 weight percent.

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# Graphene/Polyacrylic acid Composite Hydrogels being Mechanically Robust and pH-sensitive

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Mechanically strong and pH-sensitive hydrogels are highly desired in many applications, and graphene would be the most promising candidate to these hydrogels. While previous research utilized either as-prepared or reduced graphene oxide, we in this study prepared high structural integrity yet solution-processable graphene sheets for the development of graphene/poly(acrylic acid) composite hydrogels. At 1 vol%, Young's modulus and compressive strength were respectively enhanced from 1.64 – 19.03 MPa, 0.37 – 6.9 MPa corresponding to 1060%, 1765% improvements. It is worth noting that our composite hydrogels were found sensitive to pH.

We tailored the oxidation degree of graphene sheets by controlling the sonication time and the acid concentration. It is worth noting that the preparation was made by simultaneously oxidizing and ultrasonically graphite for 60 min. This fabrication method may hold potential for the bulk production of graphene for two reasons: the starting-material graphite is cost effective as no harsh oxidation is involved and the resulting graphene requires no further reduction where dangerous, expensive chemicals are often involved. Raman spectroscopy offers an effective tool to study the disorder of carbon nanomaterials – the structural integrity of graphene. By measuring the ratio (ID/IG) of D band to G band was calculated to be 0.36, far lower than those of graphene oxide. Graphene sheets show a C/O ratio of 3.9.

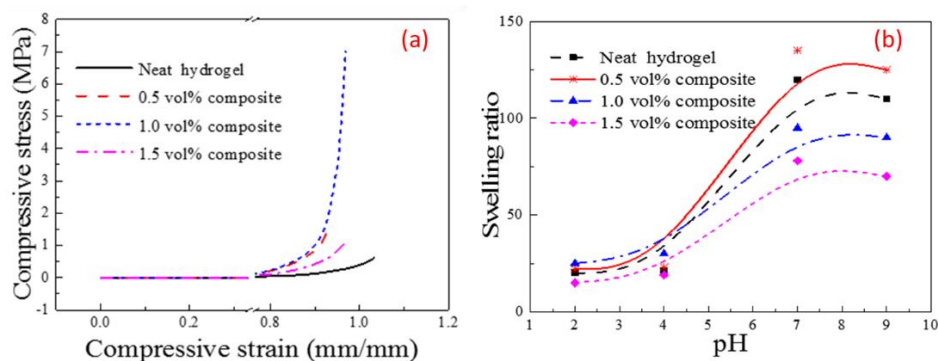


**Fig. 1.** Mechanical and swelling properties of neat and composite hydrogels (a) compressive stress-strain curves, and (b) pH-sensitivity.

**Fig. 1 a-b** contain typical stress-strain graphs and swelling behaviours of hydrogel and its composites with different graphene loadings. The compressive strength and modulus of the graphene/PAA hydrogel composites both increase with graphene fractions (Fig. a). 1.0 vol% graphene improves the gel compressive strength from 0.37 to 6.9 MPa and compressive modulus from 1.64 to 19.03 MPa, corresponding to increments of 1764 % and 1060 %; these may confirm not only good dispersion of graphene but strong interaction between the gel and the surface functional groups of graphene, these may confirm not only good dispersion of graphene but strong interaction between the gel and the surface functional groups of graphene. Fig. 1b depicts the swelling behaviours of hydrogel and its composites with pH values ranging 2–9 at room temperature. At pH > 4.5 the ratio significantly increases because the carboxyl acid groups of both polymer and graphene sheets can ionize at pH over 4.3. The results indicate that our composite hydrogels are pH-sensitive and thus have potential to be used for drug delivery.

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Research interests: Carbon nanomaterials, polymeric hydrogels, nanocomposites

# A Comparison of the Material Properties of Thermosets from Itaconic Acid Based Polyesters Catalyzed by “Classical” Catalysis and Enzymatic Catalysis

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The interest in replacing non-renewable petroleum-based polymeric materials with renewable ones has increased due to environmental awareness. Itaconic acid (IA) is a fully bio-based monomer that is produced industrially through fermentation of carbohydrates using the fungi *Aspergillus terreus*.<sup>1,2</sup> The monomer has multiple functionalities; being a diacid and also having a vinyl group (**Fig. 1**). Previous studies have utilized IA together with different diols to synthesize unsaturated polyesters that have been further cross-linked to form thermoset networks.<sup>1,3</sup> The networks showed great potential in for example coating applications.

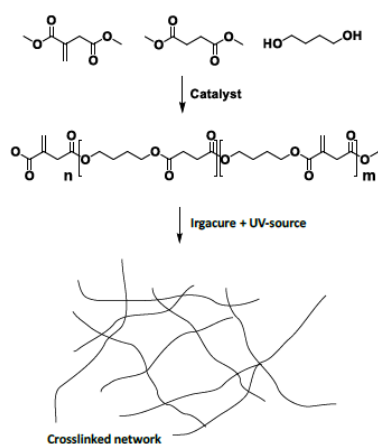


**Figure 1. Unsaturated photo curable polyesters from IA and succinic acid derivatives with 1,4-butanediol crosslinked using UV light and irgacure 651**

The current study aims to provide insight in how the choice of catalyst affects the material properties of networks formed from IA derivative based unsaturated polyesters (**Fig. 1**). Biocatalysis (*Candida antarctica* lipase B), acid catalysis (p-Toluenesulfonic) and an organometallic compound (Titanium (IV) butoxide) were used for the synthesis of unsaturated polyesters. We hypothesized that the choice of catalyst and reaction conditions e.g temperature, affect the selectivity of the reaction and as a consequent a varied distribution of the monomers in the polymeric chains will be obtained. The synthesized unsaturated polyesters were cross-linked into networks by radical polymerization and the material properties of the IA derivative based thermosets were investigated and compared.

1 T. Robert and S. Friebe *Green Chem.* **2016**, 2 S. Choi, C. W. Song, J. H. Shin, and S. Y. Lee *Metab. Eng.*, **2015** vol. 28, pp. 223-239, 3, 3 Dai, Jinyue ; Ma, Songqi ; Liu, Xiaoqing ; Han, Lijing ; Wu, Yonggang ; Dai, Xinyan ; Zhu, Jin *Prog. Org. Coat* **2015** vol. 78, pp. 49-54, 1,

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# Multi-Scale Toughening of Polymer Composites Using Carbon Nano-Particles and Carbon Macro-Pins

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Improving the delamination resistance of carbon fiber reinforced polymer composites is of significant interest to a broad range of applications, including aerospace, civil infrastructure, marine, automotive and sports goods. Composites with a brittle-like polymer matrix (e.g. non-toughen epoxy) are prone to delamination cracking caused by low interlaminar static or fatigue loads. In the present work, we present a multi-scale toughening approach to increase substantially both the delamination fracture toughness and fatigue strength of brittle polymer composites. The novel approach combines nano- and macro-scale carbon reinforcements which are inspired by natural composite materials such as bone. Carbon reinforcements with two different length scales are used: nano-scale carbon particles (nanofibres or graphene platelets) and macro-scale carbon z-pins to reinforce composites in the through-thickness direction.

The composite, featuring the three-dimensional reinforcement architecture, possesses multiple toughening mechanisms at different scales along delamination cracks thus yielding large improvements to the interlaminar fracture toughness and fatigue resistance. Figure 1 shows the measured improvements to the (a) mode I fracture toughness and (b) mode I fatigue strength of the composite due to nanofibres alone, z-pins alone, and the combination of nanofibres and z-pins. At the nano-scale, the carbon nanofibres promote high delamination resistance by multiple intrinsic and extrinsic toughening processes around the crack tip. The graphene nanoplatelets promote intrinsic toughening at and immediately ahead of the crack tip, although the toughening effect is not as strong as the carbon nanofibres. The macro-size carbon z-pins promote a strong extrinsic toughening mechanism behind the crack tip and over a larger length-scale than the carbon nanoparticles via bridging the crack faces. Under monotonic crack opening or cyclic fatigue loading, the multi-scale composites show a strong improvement in resistance against delamination crack growth.

The manufacture, delamination fracture toughness and fatigue properties, and interlaminar toughening and fatigue strengthening mechanisms of the multi-scale composites are described in the paper.

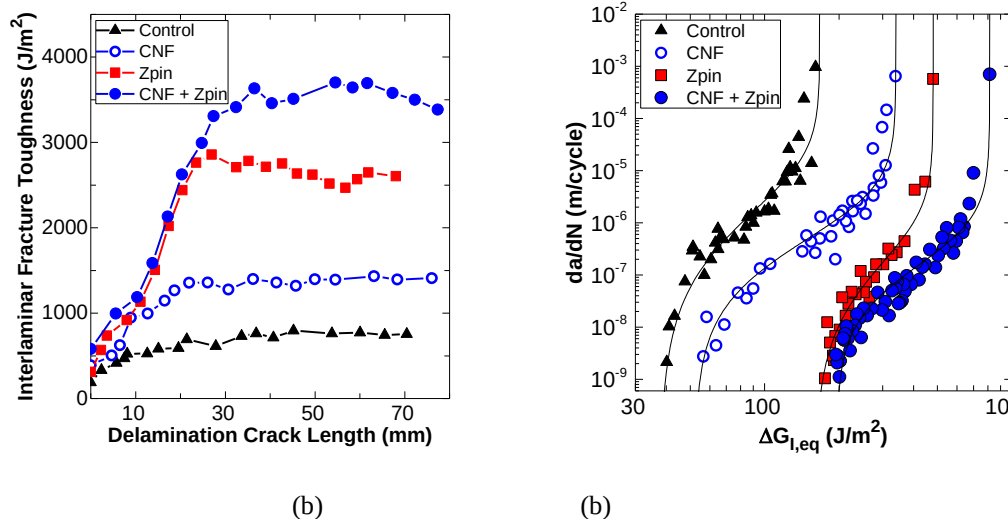


**Figure 1.** (a) Crack growth resistance curves and (b) Paris curves showing respectively the fracture toughening and fatigue strengthening effect of combining carbon nanofibres and carbon macro pins on the mode I delamination properties of carbon-epoxy laminate.

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Research interests: fibre-polymer composites, aerospace materials

## Building Advanced, Bioresponsive Materials from Polymers, Macromolecules and Metal Ions

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The design and assembly of responsive materials underpins the development of particle carriers for biomedical applications.<sup>1,2</sup> This presentation will detail various assembly strategies and chemistries to generate multifunctional and responsive particles. Recent studies on the development of nanostructured polymer particles for efficient cargo encapsulation, triggered release and antibody-mediated targeting will be highlighted. It will be shown that engineered particles with well-controlled physicochemical properties, including shape and surface characteristics, can be used to uncover several key principles that govern particle-cell interactions.<sup>3</sup> The assembly of organic ligands with metal ions will also be presented, highlighting metal-ligand complexation as a facile approach to generate particles that can disassemble under cellular conditions.<sup>4,5</sup> It is expected that such engineered particle systems represent a class of novel materials for therapeutic delivery, as well as an interesting paradigm to attain detailed knowledge of complex nano-bio interactions. This knowledge may aid in the rational design of nanoengineered materials (beyond particle systems) for biological applications.

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<sup>4</sup> H. Ejima, J. J. Richardson, K. Liang, J. P. Best, M. P. van Koeverden, G. K. Such, J. Cui, F. Caruso, *Science* **2013**, 341, 154.

<sup>12</sup> J. Guo, Y. Ping, H. Ejima, K. Alt, M. Meissner, J. J. Richardson, Y. Yan, K. Peter, D. v. Elverfeldt, C. E. Hagemeyer, F. Caruso, *Angew. Chem. Int. Ed.* **2014**, 126, 5546.

### Frank Caruso

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Personal History:

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| 2013-present | Bionics Institute, Australia<br><b>Honorary Research Fellow</b>                                                                                                                                                                                           |
| 2003-2010    | Centre for Nanoscience and Nanotechnology, The University of Melbourne<br><b>Founder and Director</b>                                                                                                                                                     |
| 1997-2002    | Max Planck Institute of Colloids and Interface, Germany<br><b>Group Leader &amp; Research Scientist</b> (1999-2002)<br><b>Alexander von Humboldt Research Fellow</b> (1997-1998)                                                                          |
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## **TitSoft Matter (Cryo) Electron Microscopy and Microanalysis:**

*Roger Wepf<sup>1</sup>, Matthias Floetenmeyer<sup>1</sup>, Kathryn Green<sup>1</sup> and Rick Webb<sup>1</sup>*

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Advancement in corrected transmission electron microscopy (cs-S/TEM) and cryo electron microscopy (cryo-TEM, cryo-SEM, cryo-FIB/SEM) have made it recently possible to image objects, such as organic samples and biological samples down to near atomic resolution. These techniques were originally developed for material science or biological science independently but in recent years (cryo)-EM is increasingly used to investigate soft matter of synthetic origin or natural origin.

The key issue for successful structure determination is once more sample preparation of soft matter for these novel electron microscopy opportunities. Again by borrowing methods developed for biological samples - cryo sample preparation - and adapting them to man-made soft matter has lead to an increasing interest in synthetic soft matter EM. In this overview we want to highlight the recent developments in low voltage (cryo) EM and show the potential of (cryo)EM methods on soft matter structure research projects. Finally by combining these novel sample preparation and electron microscopy methods with other analytical techniques we will elaborate on future multimodal sample characterization possibilities appearing on the horizon.

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| 2006 – 2014 | Director EMEZ – ETH Zurich, Electron Microscopy Centre                                                           |
| 1997 – 2006 | Department Head – Beiersdorf AG, First: Analytical Microscopy in R&D/ Structure Research Dept. in Research       |
| 1993 – 1997 | PostDoc Position & Junior Group Leader – European Molecular Biology Laboratory, Cell Biology and Biophysics Unit |
| 1992        | PostDoc Cell Biology ETH Zurich & MIH Biocenter Univ. Basel                                                      |
| 1988 – 1992 | pHD at Cell Biology ETHZ                                                                                         |

Research interests: Rogers major research activities are focused in the field of correlative and multimodal microscopy, automatic sample preparation and imaging techniques for particle imaging and spectroscopy technology. Currently this leads to developing tools for connectivity between imaging and spectroscopy methods and integrative imaging and spectroscopy techniques to explore new frontiers in structure research.

# Universal Mass Spectrometric Analysis of Poly(Ionic Liquid)s

Jan Steinkoenig,<sup>1</sup>Martina M. Cecchini,<sup>2</sup>Anja S. Goldmann,<sup>1</sup>Francesco De Angelis,<sup>2</sup>Christopher Barner-Kowollik<sup>1</sup>

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High resolution mass spectrometry provides the synthetic polymer chemist with a powerful tool to investigate the molecular structure of macromolecules.<sup>1</sup>In the current presentation, we demonstrate the in-depth mass spectrometric characterization of complex poly(ionic liquid)s (PILs) bearing different core structures, counter ions and end groups (Figure 1).<sup>2,3</sup>The characterization of this highly attractive class of polymers is performed via Orbitrap electrospray ionization mass spectrometry (ESI-MS), using collision-induced dissociation with supercharging technology as well as quadrupole time-of-flight (QToF) MS. To access the PILs, RAFT polymerization was carried out in a highly controlled fashion resulting in polymers with relatively narrow molecular weight distributions ( $2000 \text{ g} \cdot \text{mol}^{-1} \leq M_n \leq 10000 \text{ g} \cdot \text{mol}^{-1}$ ) and dispersities between 1.3 and 1.4. A fast and quantitative salt-exchange to weakly coordinating anions was achieved without further synthetic efforts, employing silver salts for the metathesis and filtration prior to their mass spectrometric characterization was sufficient.<sup>3</sup>In addition, PILs were encoded in a spatially-resolved fashion onto silicon wafers via a photolithographic process followed by the controlled growth of the PILs from the surfaces. The successful surface functionalization was evidenced via X-ray photoelectron spectroscopy (XPS) and time-of-flight secondary ion mass spectrometry (ToF-SIMS).<sup>4</sup>

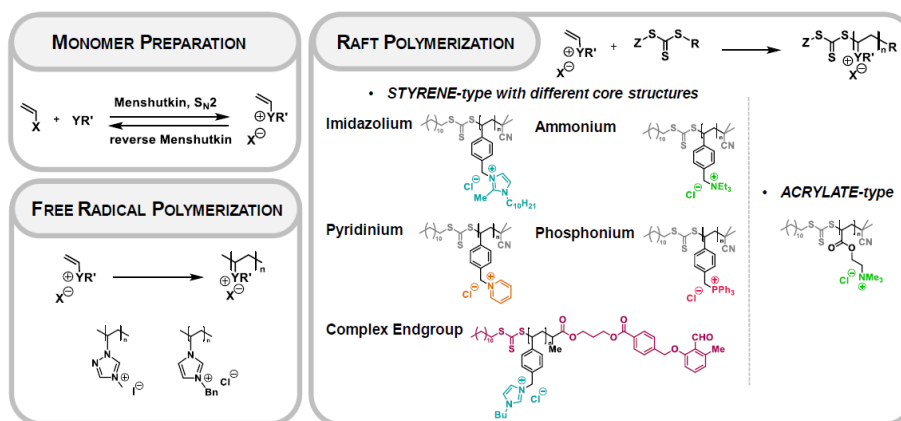


Figure 1 Overview over the IL monomer preparation (X: halide; Y: N or P or S; R': alkyl or aryl) as well as the free radical and RAFT polymerization collating all investigated PILs (Z: stabilizing group, R: leaving group).

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- (2) Steinkoenig, J.; Cecchini, M. M.; Reale, S.; Barner, L.; Yuan, J.; Goldmann, A. S.; De Angelis, F.; Barner-Kowollik, C. *Chem. Sci.* **2016**, DOI 10.1039/C6SC01347C.
- (3) Steinkoenig, J.; Cecchini, M. M.; Goldmann, A. S.; Reale, S.; De Angelis, F.; Barner-Kowollik, C. **2016**, submitted.
- (4) Steinkoenig, J.; Bloesser, F. R.; Huber, B.; Welle, A.; Trouillet, V.; Weidner, S.; Barner, L.; Roesky, P.; Yuan, J.; Goldmann, A. S.; Barner-Kowollik, C. *Polym. Chem.* **2016**, *7*, 451–461.

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# Characterization strategies to study the thermal degradation and combustion behaviours of polymeric materials: a critical analysis

*Paul Joseph<sup>1</sup>, Vasily Novozhilov<sup>1</sup>, Svetlana Tretsiakova-McNally<sup>2</sup> and Rahul Wadhvani<sup>1</sup>*

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Polymers are increasingly being used in domestic, commercial and in public environments as components of fabrics, surface coatings and moulded articles. Most of these polymeric components are synthetic products based on hydrocarbon intermediates and are highly flammable, thus resulting in unwanted fires. Combustion of a polymeric material is a complex phenomenon that involves the solid phase, gaseous phase and interphase. A full characterization of the degradation behaviours and flammability attributes of a polymeric material is not a straightforward task, and therefore several analytical techniques and other prescriptive tests (small-, medium-, and large-scale) were developed over the years<sup>1</sup>. However, the actual behaviour of these materials in a real fire scenario can be quite different from the inferences that we get, especially, through small- and medium-scale tests, and generally there is a lack of correlation among the tests. Furthermore, the various elements of the mechanistic pathways occurring in different phases of a burning solid material remain largely unclear. In this paper, we attempt a critical analysis of, and also seek correlation among, various laboratory-scale test regimes that are used to characterize the degradation and combustion behaviours of polymeric materials<sup>2,3</sup>. Firstly, we describe thermogravimetric analysis (TGA) that is widely used for the study of polymer decomposition processes. However, TGA cannot accurately represent the very complex process of polymer combustion. Furthermore, there is no simple correlation between the flammability of different organic polymers and their thermal stability. Thermal analyses can, however, be used to extract kinetic parameters governing step-wise/multi-step degradation of a polymeric material, either under dynamic, or static heating regimes. The thermodynamic basis and hence the validity, especially, of the former method of measurement needs to be treated with some degree of caution. Secondly, we provide an account of some of the most commonly employed laboratory-scale test methods, such as: pyrolysis combustion flow calorimetry (PCFC), limiting oxygen index (LOI), Underwriters Laboratory test (UL-94) and cone calorimetry. Here, we also present our recent work on PCFC that was aimed at deducing meaningful correlations of the relevant parameters among PCFC and TGA runs.

Ideally, one should be able to predict the behaviours of various solid materials in real scale fire scenarios by looking into the intrinsic thermal properties of the materials and from related empirical parameters without having to resort to actually carrying out expensive, and often time consuming, large-scale tests. Moreover, an accurate knowledge of the composition of volatiles is also necessary to predict the combustion behaviour at large scales. This necessitates a more concerted effort from the scientific community, involved in fire safety research, to try to formulate methodologies that would harmonize the various test regimes, and the ensuing results, performed at different scales. One should be also able to predict realistic behaviours of various materials in real fire scenarios, from the empirical parametric evaluations and associate analytical relations, obtained through scientifically-sound and economically-viable small- to medium-scale laboratory experiments.

<sup>1</sup> Paul Joseph, Svetlana Tretsiakova-McNally and Richao Zhang, *Macromolecular Symposia*. **2016**, 362, 105-118

<sup>2</sup> D. Price, L.K. Cunliffe, K.J. Bullett, G. J. Milnes, J. R. Ebdon, B. J. Hunt and P. Joseph, *Polym. Adv. Technol.* **2008**, 19, 710-723

<sup>3</sup> D. Price, L. K. Cunliffe, K. J. Bullett, T. R. Hull, G. J. Milnes, J. R. Ebdon, B. J. Hunt and P. Joseph. *Polym. Degrad. Stab.* **2007**, 92, 1101-1114

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Research interests: polymer science and combustion chemistry

# Chemical Characterization of Polyelectrolyte Complexes: Strengths and Limitations

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Polyelectrolyte complexes (PECs) form between cationic and anionic polyelectrolytes through attractive electrostatic interactions. The nature of the PEC is affected by many factors including temperature, ionic strength and pH as well as polyelectrolyte type, molecular weight and concentration. By manipulation of these factors it is possible to form either weakly or strongly associated PECs (**Fig.1**) that possess a range of physicochemical properties. Examples of PECs include coacervates and layer-by-layer assemblies. Both have found numerous applications in biomaterials science as, for example, drug delivery systems and coatings with properties tuned through control of the fabrication process.

In the work presented, commonly used chemical characterisation techniques will be critically analysed with reference mainly to PECs formed from polysaccharide-based polyelectrolytes.<sup>1,2</sup> The effect of using a strong (dermatan sulfate<sup>3</sup>) versus a weak (alginate<sup>1</sup>) polyelectrolyte in formation of PECs will be illustrated. In addition, systems in which covalent bonding can form in combination with electrostatic interactions will be described.<sup>2</sup>

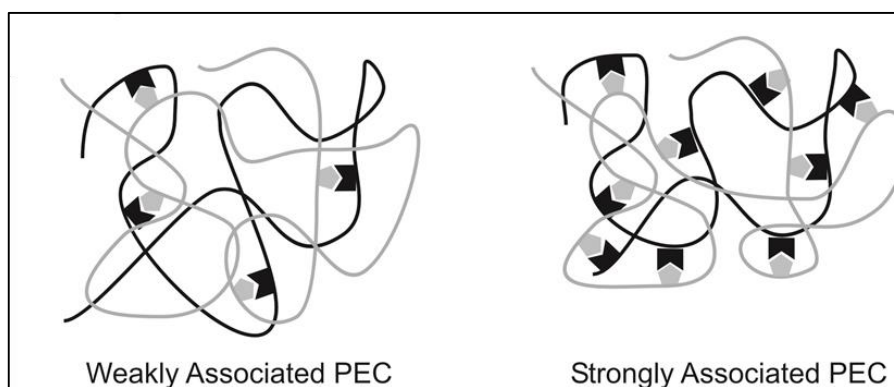


**Figure 1.** Schematic illustration of electrostatic interactions in weakly and strongly associated PECs

4. G Lawrie, I Keen, A Chandler-Temple, B Drew, L Rintoul, P Fredericks, L Grøndahl, *Biomacromolecules* (2007) 8, 2533-2541
5. R Aston, M Wimalaratne, A Brock, G Lawrie, L Grøndahl, *Biomacromolecules* (2015) 16, 1807-1817
6. Y Wen, L Grøndahl, M R Gallego, L Jorgensen, E H Møller, H M Nielsen, *Biomacromolecules* (2012) 13, 905-917

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## Database-supported thermal analysis involving automatic evaluation, identification and classification of measurement curves

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In the history of thermal analysis, the consideration of measurement signals originating, for example, from the methods differential scanning calorimetry (DSC), thermogravimetry (TGA), dilatometry (DIL) or thermomechanical analysis (TMA) required intensive examination by experienced users in most cases<sup>1,2</sup>. Effects such as glass transitions or other caloric effects that may occur in a measurement curve first need to be identified visually before they can be evaluated. Until recently, the possibility of a computer-based identification of a measurement by means of a comparison with a database—as is well known for spectroscopic techniques such as mass spectrometry or Fourier transform infrared spectroscopy (FTIR), for example—did not exist for thermoanalytical curves<sup>3</sup>. In 2014, the first computer-based curve recognition and database system for DSC curves, called *Identify*, was introduced by NETZSCH and was recently introduced for TGA, Cp and DIL/TMA curves. This talk will focus on the philosophy of *Identify*, which also involves a novel, autonomous and automatic evaluation of measurement curves by means of a software function called *AutoEvaluation*. *Identify* and *AutoEvaluation*, which are both part of the NETZSCH *Proteus*® evaluation software, significantly simplify the evaluation and interpretation of thermoanalytical measurement curves.

<sup>1</sup> W.F. Hemminger, H.K. Cammenga, *Methoden der thermischen analyse*. 1989, Heidelberg: Springer.

<sup>2</sup> G.W. Ehrenstein, G. Riedel, P. Trawiel, *Thermal analysis of plastics: theory and practice*. 2004, New York: Hanser Gardner Publications.

<sup>3</sup> J. Fiala, J. Sestak, *J Therm Anal Calorim*. 2000; 60, 1101–10.

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# Characterisation of shear inhomogeneity of a semicrystalline polymer in couette flow

Andrew Phillips<sup>1</sup>,

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Thermoplastic semicrystalline polymers are typically processed into their final form by moulding or extrusion technologies. These processes impart complex flow and temperature histories to the solidifying polymer which in turn can dramatically alter the final physical properties of the component. If the flow intensity is sufficient, a highly anisotropic crystalline architecture, termed 'shish-kebab' can form. A highly oriented thread-like core is considered to form first (shish), which then provides a template for the formation and growth of oriented chain folded crystalline lamellae (kebabs). Understanding the links between processing and morphology will allow for more tightly controlled and optimised properties, as well as reduced processing defects.

Due to its simplicity, one of the most widely used methods to study the effect of flow on semicrystalline polymers is to place molten polymer between two plates and move one plate relative to the other to induce couette (laminar) flow. However, the flow induced from this process may not be uniform through the thickness, potentially impacting the quality of the results obtained from these experiments. For instance wall slip and shear banding phenomenon can occur under certain circumstances which can dramatically change the local flow environment.

This study aimed to shed further light on flow inhomogeneity of molten semicrystalline polymers by using the power of synchrotron radiation to probe in detail the induced morphology gradient in samples subjected to couette flow. Samples 500 micron thick were prepared using a parallel plate rheometer and then examined using micro beam simultaneous wide and small angle x-ray (WAXS/SAXS) scattering as schematically illustrated in Figure 1. The study showed that a skin-core morphology was formed, with the skin representing a considerable (40%) amount of the sample thickness. The skin was dominated with larger and more numerous shish, but the shish had high misalignment when compared to those in the core. These results highlight that surface effects cannot be ignored in couette flow such as that induced by a parallel plate rheometer.

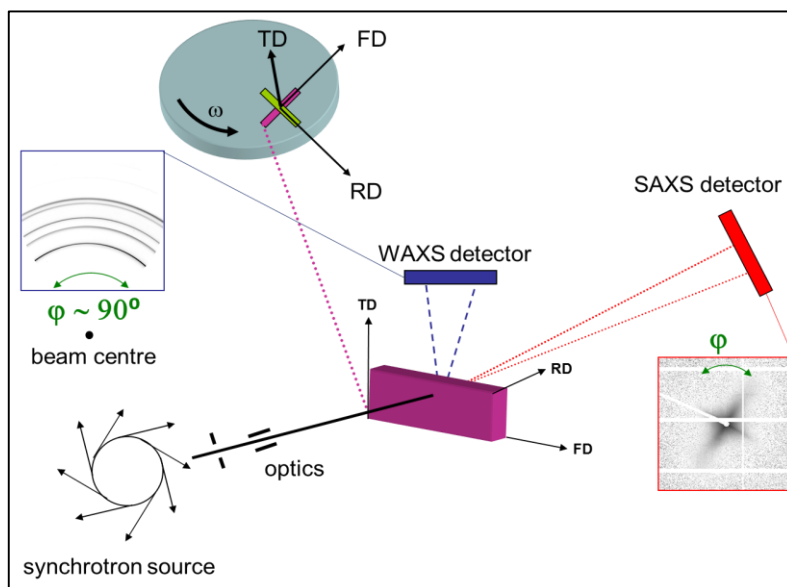


**Figure 1.** Schematic illustration of the simultaneous wide and small angle x-ray scattering setup used. TD,FD and RD correspond to thickness, flow and radial directions.  $\omega$  is angular velocity and  $\phi$  is the azimuthal angle.

## Andrew Phillips

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Personal History:

Since 2012

DST Group

2008-2011

CRC for Polymers, Monash University

2005-2008 & 2011-2012

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Research interests: Polymer composites, processing-structure-property relationships, advanced characterisation

## Morphology of Composite Nanoparticles assessed by X-Ray Photoelectron Spectroscopy

*F. Jasinski,<sup>1</sup> V. Teo,<sup>1</sup> M. Mballa Mballa,<sup>3</sup> S. C. Thickett,<sup>2</sup> R. Brinkhuis,<sup>3</sup> W. Weaver<sup>3</sup> and P. B. Zetterlund<sup>1</sup>*

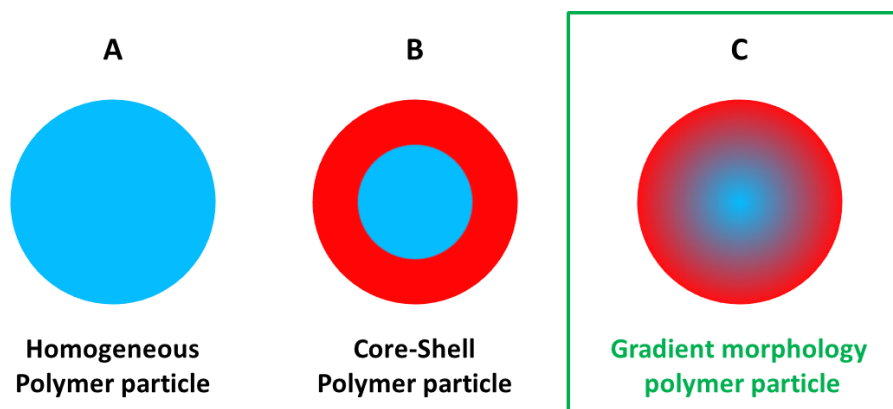
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<sup>3</sup> Nuplex Innovation Centre, Wageningen, Netherlands

Water-borne paints and varnishes made by emulsion polymerization are among the main materials used to protect and decorate surfaces. The microstructural morphology of latex nanoparticles can be advantageously designed in order to (i) remove volatile organic compounds from the formulation and (ii) achieve optimal coating properties such as hardness, flexibility, gloss, water resistance.

We present here an original way to characterize nanoparticle morphology using X-Ray Photoelectron Spectroscopy (XPS) implemented as a surface analysis technique. By comparison with mathematical modelling and control experiments, morphology of different type of nanoparticles were characterized; from the simplest homogeneous morphology to the gradient morphology, through core-shell microstructure (**Scheme 1**). Original feed strategies were developed in order to increase the level of complexity in the microstructure for a high  $T_g$  formulation made from styrene and methyl methacrylate. XPS proved to be a reliable and accurate way to determine the morphology of structured latexes especially gradient morphology latexes.



**Scheme 1.** Different composite morphologies: homogeneous (A), core-shell (B) and gradient morphology (C)

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2012-2014    PhD Candidate in LPIM (UHA, Mulhouse, France)

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Research interests: Photopolymerization, Miniemulsion polymerization, Structured latexes



# NEXAFS Spectroscopy of Polymer Thin Films

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Near-edge X-ray absorption fine-structure (NEXAFS) spectroscopy is a synchrotron-based, X-ray absorption spectroscopy that can provide information about the chemical and structural properties of thin films. For carbon-based polymers, NEXAFS spectroscopy is typically performed about the carbon K-edge where electrons from the 1s state are promoted into antibonding orbitals. The peaks observed in a carbon K-edge NEXAFS spectrum can be assigned to specific bonds, see **Fig. 1**, often with the help of theoretical simulations. For polymer blends, the chemical fingerprinting provided by NEXAFS spectroscopy can be used to measure the chemical composition of blends, or to provide a contrast mechanism for X-ray microscopy. By performing angle-resolved NEXAFS measurements, information about the orientation of molecular orbitals (and hence bonds) can also be obtained. NEXAFS spectroscopy can be performed in a number of detection modes enabling either bulk sensitivity or surface sensitivity. Measuring X-ray absorption via electron yield techniques in particular provides NEXAFS spectroscopy with high surface sensitivity permitting surface analysis.

In this presentation I will highlight some of our recent work employing NEXAFS spectroscopy to understand surface layers in conjugated polymer thin films and their importance for the operation of organic field-effect transistors<sup>27</sup>. In particular, I will demonstrate how NEXAFS spectroscopy can be used to simultaneously measure surface and bulk orientation by combining transmission (bulk sensitive) and electron yield (surface sensitive) measurements. Such an approach has proved the existence of surface layers with a molecular orientation distinct from that in the bulk of the film. I will also discuss the potential for NEXAFS spectroscopy to discern dihedral angles in donor/acceptor co-polymer films, and how NEXAFS spectroscopy can discern surface coverage by solubilising side-chains.

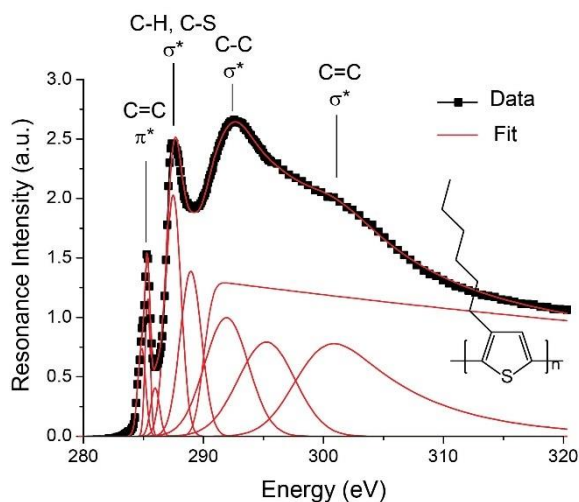


**Figure 1.** Carbon K-edge NEXAFS spectrum of the conjugated polymer poly(3-hexylthiophene) with major peaks identified.

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Research interests: organic electronics, synchrotron characterisation



## Intracellular Behaviours of Theranostic Polymers for Nanomedicine

Joshua D. Simpson<sup>1,2,3</sup>, Nicholas L. Fletcher<sup>1,2,3</sup>, Adrian V. Fuchs<sup>1,2,3</sup>, Kristofer J. Thurecht<sup>1,2,3</sup>

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<sup>2</sup> Centre for Advanced Imaging, University of Queensland, St. Lucia, QLD, 4072

<sup>3</sup> ARC Centre of Excellence in BioNano Convergent Science & Technology

The use of nanomaterials as vectors for improved targeting of chemotherapeutics and nucleic acids to tumour tissue has become a major research focus in oncology.<sup>1</sup> As polymer-based vehicles have begun to translate into the clinical environment, research into more complex structures and mechanisms has led to increasingly elaborate designs; for instance ‘smart’ materials which potentially allow an unparalleled level of control over therapeutic targeting and release.<sup>2</sup> With a growing number of materials being designed to respond to specific intracellular environments or compartments, for example pH-sensitive linkers which cleave in select components of the endolysosomal system, it is becoming increasingly pertinent to understand the phenomena that occur once polymeric materials gain access to the internal cellular environment.<sup>3,4</sup> Due to the requirements of their effective functioning, once these materials have been internalized, the level of control afforded and overall efficacy of the construct become intrinsically dependent upon intracellular fate.<sup>2,4</sup> Moreover, while it is widely acknowledged that size, shape and external charge of materials have the potential to alter ingress, trafficking, sorting and thus intracellular localisation of a construct, in order to better inform the design of smart materials and ensure efficacious therapeutic release,<sup>4,5</sup> we need to better characterise how chemical and physical properties of nanoparticles dictate their post-internalisation behaviours.<sup>5</sup>

This research effort aims to build upon our existing knowledge of how various factors influence the outcome of intracellular sorting events by comparing polymeric nanoparticles that differ by properties such as charge, size or targeting ligand. By examining the behaviours of various materials with respect to labelled cellular features e.g. ribonucleic acids (RNA) and lysosomes, we have begun to collect an array of information that can be linked back to polymer synthesis and design, building upon our understanding of how the components used to fashion nanoparticles may influence their behaviour. Focusing on our hyperbranched polymer systems, we have already identified differences between targeting motifs and cellular response, cell type *versus* the importance of charge and begun to characterise siRNA delivery systems with respect to targeting endogenous RNA circuitry. Utilizing advanced confocal microscopy techniques, we have begun to better understand the behaviours of our polymeric constructs once they have entered the cellular milieu. The results of these studies will be utilised to better inform the design of complex polymeric structures in order to best harness intracellular trafficking pathways and achieve maximum efficacy in terms of therapeutic release.

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# Rational design of polymeric nanomaterials for photo-induced molecular imaging and therapeutic function

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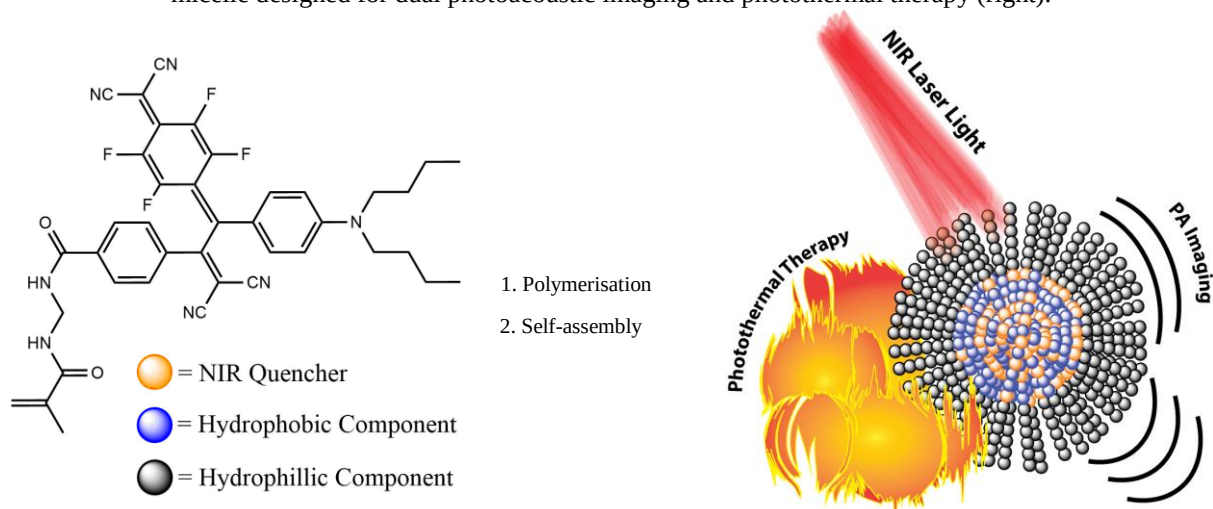
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Modern nanoscience has revolutionised many fields of research, especially in the development of nanomedicines for the advanced treatment of disease. The use of sophisticated chemistry to develop new materials, in particular with specific properties on the nanoscale ( $10^{-9}$ m), offers new prospects for expanding our approach to treating disease. Recent advances in synthetic materials chemistry facilitates specific control over macromolecular structure and offers exquisite control over the manipulation of their specific physicochemical properties. This control provides the foundation for the development of multifunctional macromolecular platforms with well-defined properties on the nanoscale. In the medical context, these multifunctional materials are developed in order to combine a variety of different functions, creating nanomedicines that can achieve a goal more efficiently than many analogous approaches that utilise small molecules. Here we present the synthesis of a custom-designed monomer for the development of polymeric photoacoustic (PA) imaging agents. This includes the design of a near-IR quenching dye with monomer functionality included for RAFT polymerisation. Furthermore, we present the design and synthesis of new polymeric materials that afford photoacoustic contrast in vivo. Through specific irradiation of highly efficient light absorbing species, the proposed nanoparticle also provides the opportunity to act as a photothermal agent for tumour therapy. Thus, new theranostics are proposed that allow simultaneous imaging and therapy utilizing multispectral optoacoustic tomography (MSOT).

**Figure 1.** Target Structure of NIR quenching dye with RAFT polymerisation functionality (left). Polymeric micelle designed for dual photoacoustic imaging and photothermal therapy (right).



S. Huang, P. Kumar Upputuri, H. Liu, M. Pramanik,\* M. Wang\* J. Mater. Chem. B **2016**, 4, 1696-1703

A. Xu, H. Han, J. Li, P. Yang, et. al. Dyes and Pigments **2016**, 125, 392-398

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# Effect of Increased Surface Hydrophobicity on the Clearance of Inhaled PEGylated Polylysine Dendrimers

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## Introduction

Polylysine dendrimers are an attractive inhalable drug delivery platform that have potential to control the release, absorption kinetics and lung retention time of conjugated drugs.<sup>1</sup> Further, the systemic absorption, circulation time, lung retention and enzymatic liability of polylysine dendrimers can be tailored by modulating the molecular weight of the conjugated PEG moiety.<sup>2</sup> Recently, the clearance mechanisms, pharmacokinetic behavior and pulmonary stability of a range of PEGylated polylysine dendrimers (MW 11-78 kDa) was characterised after pulmonary administration in rats.<sup>2</sup> To date however, the impact of substitution of surface PEG groups with a hydrophobic drug on the lung clearance of inhaled polylysine dendrimers has not been evaluated.

## Objective

To examine the impact of increased hydrophobicity, via substitution of 50% surface PEG groups (1100 Da PEG) with alpha-carboxyl OtButylated methotrexate (conjugated via a PVGLIG peptide linker), on the clearance of an inhaled PEGylated polylysine dendrimer (PEG dendrimer) from the lungs of rats and to evaluate the safety of these dendrimers as inhalable drug delivery systems after a single dose.

## Experimental methods

The lung clearance of a <sup>3</sup>H labelled generation 5 fully PEGylated dendrimer (PEG dendrimer, Mw ~70kDa) and the equivalent dendrimer containing 50% surface PEG and 50% modified methotrexate (MTX dendrimer, Mw ~70kDa) were studied over 2 weeks by administering dendrimers (5mg/kg) to the lungs of anaesthetised rats using a Penn-Century microsyringe. Lung retention of the administered dendrimers as well as the levels of inflammatory cytokines and immune cells were evaluated in lung tissue and bronchoalveolar lavage fluid (BALF) 3, 7, and 14 days post dose. Bioavailability of the lung administered dendrimers was evaluated in chronically cannulated rats over 7 days, along with the proportion of the dose excreted via the urine and faeces. Size exclusion chromatography was used to identify <sup>3</sup>H-labelled species in plasma, urine and BALF. The results were compared by one-way ANOVA followed by Tukey's post hoc test for significant differences at p < 0.05.

## Results and discussion

The PEG and MTX dendrimers showed limited systemic absorption (<5%) after pulmonary delivery due to their high molecular weights, but differences in the kinetics of absorption and bioavailability were evident. While higher amounts of inhaled MTX dendrimer were recovered in the urine and feces, greater amount of inhaled PEG dendrimer was retained in the lung tissues 7 day post dose administration, suggesting faster lung clearance for the more hydrophobic MTX dendrimer than the PEG dendrimer. The hydrophobic MTX dendrimer also displayed more efficient lung tissue clearance in comparison to the PEG dendrimer over a period of 14 days. The SEC profiles revealed that both the PEG dendrimer and MTX dendrimer were biodegraded into low molecular weight products containing PEG in the lungs that were subsequently excreted via the urine. The low molecular weight fragments from the MTX dendrimer however, appeared to be absorbed at faster rate than the fully PEGylated fragments. On evaluation of various markers of pulmonary inflammation it was found that the dendrimers were very well tolerated at a single clinically relevant dose (5 mg/kg), as evidenced by the absence of local lung inflammation.

## Conclusion

The substitution of 50% of the surface PEG groups of a fully PEGylated polylysine dendrimer with alpha-carboxyl OtButylated methotrexate accelerated the lung clearance of the dendrimer. Both dendrimers were also well tolerated in the lungs after a single dose despite differences in surface hydrophilicity.

<sup>1</sup> LM. Kaminskas, VM. McLeod, GM. Ryan, BD Kelly, et al. *J. Control Release*. **2014**, 183, 18-26.

<sup>2</sup> GM. Ryan, LM. Kaminskas, BD. Kelly, DJ. Owen, et al. *Mol. Pharm.* **2013**, 10, 2986-95.

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# Bottle-Brush Polymer Architectures for Drug Delivery

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Polymer Therapeutics (PT) is an emerging field within the cutting edge therapies that are being approved in the clinic in the last years<sup>1</sup>. Only in the polymer-drug conjugates family, at least 16 drugs are in clinical trials or the market at the moment. Most of them are based in functionalized linear polymers (HPMA, PEG, PGA) or natural carriers where the control of the architecture of the carrier is very poor (i.e. dextrin).

Therefore, there is a need of obtaining new materials with alternative properties in order to interact with the cell in a different fashion. Novel structures such as dendrimers and star polymers have been incorporated into PT with promising results and have demonstrated the dramatic dependence on the carrier shape in the cell internalization process<sup>2</sup>.

One of the traditional uses of bottle-brush polymers (normally attached to films) has been in water remediation. This complex structure, highly packed due to its polymeric side chains, has demonstrated the ability to interact very effectively with distinct biological membranes of many microorganisms. Therefore, we foretell these molecular cylinders in solution could be perfect candidates for drug delivery applications. However, only few examples of bottle brush polymer-drug conjugates have been reported<sup>3</sup>.

We describe here the implementation of the new Shuttle-CTA RAFT methodology<sup>4</sup> to create two families of molecular brushes with different core properties. While the first family shows a hard and non-degradable styrene core, the second one are built upon acrylate backbones and contains soluble lateral chains (pNAM, pNIPAM and pHEA) that can be pH-dependent hydrolyzed from the backbone (**Fig. 1a**).

In addition to the synthesis, we also report here the toxicity of these materials in human cell lines (HEK and MDA-MB-231). The endosomotropic nature of the cell internalization for these bottle-brushes polymers has also been established and confirm the suitability of these materials for drug delivery applications (**Fig 1b-d**).

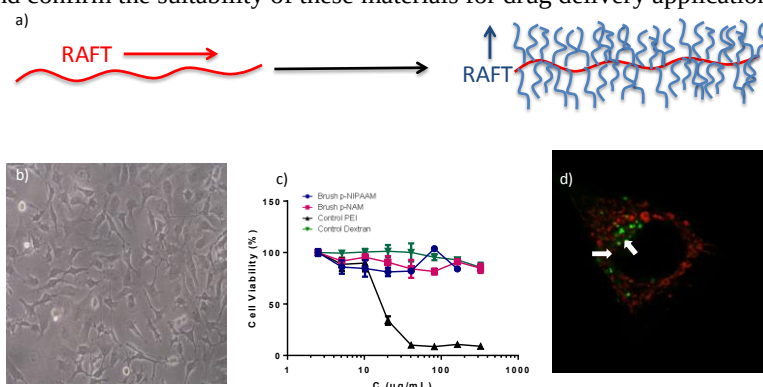


**Figure 1.** A) Brushes are easily synthesized only by RAFT. b) MDA-MB-231 Triple negative breast cancer cell line. c) Toxicity of the brushes in MDA-MB-231 after 72h. d) Intracellular accumulation of the brush in MDA-MB-231

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# Oral delivery of folate targeted colorectal cancer therapeutics

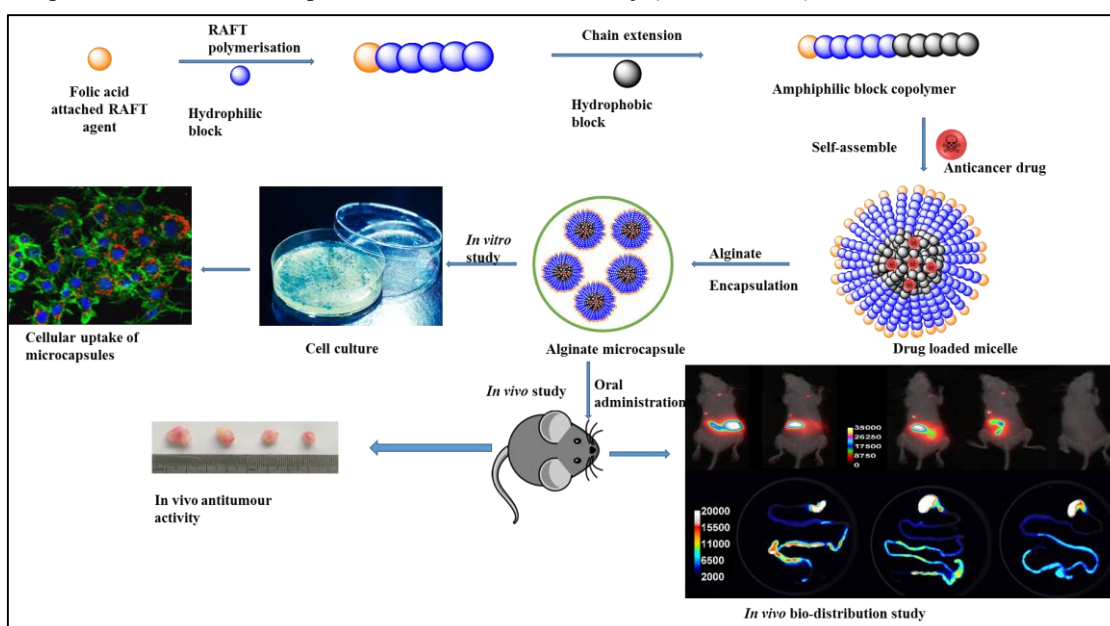
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Colorectal cancer (CRC) is the third leading cause of cancer related mortality after lung and prostate cancer. Every year approximately 700,000 people die of colorectal cancer.<sup>1</sup> Chemotherapy is typically administered by intravenous injection and infusion which often fails to deliver an effective concentration of the drug at the target site, with severe side-effects occurring due to off-target delivery<sup>2</sup>.

Oral chemotherapy has the potential to open a new treatment concept called “chemotherapy at home”<sup>3</sup>. Unfortunately, most anticancer drugs are not suitable for oral delivery due to the myriad of physiological barriers as well as the physicochemical properties of the drugs<sup>4</sup>. The aim of this project is to develop a model carrier for oral delivery of colon targeted anticancer therapeutics that incorporates drug-encapsulated nanoparticles within microcapsules suitable for oral delivery (see Scheme 1).



**Scheme 1:** Experimental design of folate-targeted oral delivery of colorectal cancer therapeutics

An amphiphilic block copolymer of PEGMA-PMMA was synthesised and characterised successfully. Curcumin loaded micelles were formed having sizes of 30 and 70 nm with excellent drug encapsulation efficiency of 85.11% and 87.64%. Further, the micelles were encapsulated into an alginate microcapsule carrier to make a favourable formulation for oral delivery. *In vitro* release studies of the curcumin-loaded micelles showed a sustained diffusional release at pH 5.0 and 7.4.

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## Efficient Functionalisation of Dextran-Aldehyde with Catechin: Potential Applications in the treatment of Cancer

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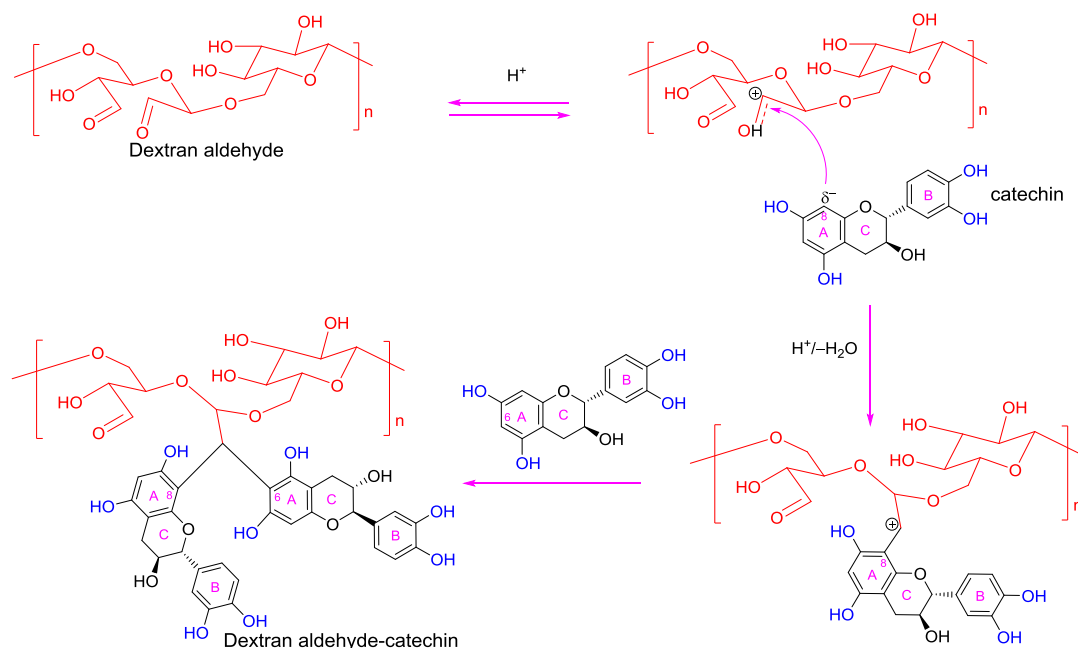
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Here, we report a simple and efficient method to functionalise dextran with catechin - a polyphenol present in red wine, red berries, and green tea. Aldehyde functionalised dextran was reacted with catechin in the presence of acetic acid to yield dextran-aldehyde-catechin conjugates via a condensation reaction. 1D and 2D (DOSY) <sup>1</sup>H NMR and UV-Vis analyses were employed to characterise the polymers. The degree of catechin conjugation was easily manipulated by varying the aldehyde amount in the dextran, the catechin concentration and the reaction temperature and conjugates with up to 38 wt% catechin functionalisation were prepared. In particular, the conjugation reaction was found to be especially sensitive to temperature and this was shown to be partially due to the equilibrium between the aldehyde and the hemiacetal in dextran aldehyde shifting to a greater proportion of aldehyde groups as the temperature increased. The stability of the dextran-aldehyde-catechin conjugates was enhanced in comparison with native catechin in both deionised water and phosphate buffer solution. Finally, the anticancer properties of the dextran-aldehyde-catechin conjugates were investigated *in vitro*, demonstrating cytotoxic efficacy against neuroblastoma cells whilst showing no cytotoxicity towards normal cells.<sup>1</sup>



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# Comparison of a Divergent and Convergent Design Strategy for Non-Viral Gene Delivery

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Non-viral gene delivery systems promise to be the long-term future for delivering gene therapies *in vivo* in a safe and controllable manner. However, non-viral vectors commonly sacrifice gene delivery efficiency in favour of reduced toxicity, struggle to deliver large genetic payloads, and work poorly in particular cell lines.<sup>1</sup> Therefore, clinical implementation of gene therapies is currently limited by the lack of a safe and effective method for delivery of all therapeutic components to the target tissue.<sup>2</sup> Recently, we have developed a non-viral, polymeric system and demonstrated superior *in vitro* transfection performance compared to current gold standard, Lipofectamine® 2000. We have also optimised the polymer for delivery of genome editing tools, namely CRISPR/dCas9 and TALE systems, aimed at upregulating a tumour suppressor gene in an *in vivo* setting, with significant functional outcome.

Here we describe the design and rationale of the novel polymeric system, and compare two different synthesis methods in light of the significant biological outcomes we have demonstrated. First we will discuss the design of the polymeric system (**Fig. 1**), and justify why we chose a dendronized linear polymer architecture instead of investigating a traditional dendrimer architecture. We demonstrate the need for a high degree of structural control in relation to the biological effect we observe with our designs. Since dendronized linear polymers can be synthesized using either divergent or convergent methods of dendron growth, we will discuss the applicability of each method, comparing structural control and purity, and the impact the method of synthesis has on the biological outcome. Specifically, we will compare the stepwise divergent synthesis of poly(amidoamine) (PAMAM) dendrons from the backbone core with copper(I)-catalysed alkyne-azide click reaction of the PAMAM dendrons onto the backbone, and discuss the limitations of each method. Through these experiments we will demonstrate the best synthesis method in terms of structural control and purity, scalability and gene delivery efficiency, to assess how to produce our polymer formulation for large-scale synthesis as we move towards the development of non-viral gene therapeutics.

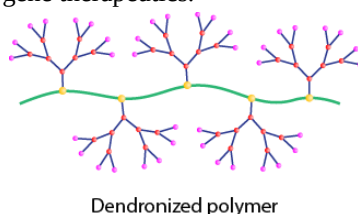


**Figure 1.** Schematic of the dendronized linear polymer system, where control over dendron generation and substitution has a significant biological effect for the delivery of gene therapeutics.

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<sup>2</sup> H. Yin, C.-H. Song, et al., *Nat. Biotechnol.* **2016**, 34, 328-333.

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# Hyperbranched Polymer based dual responsive drug delivery system for cancer therapy

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Currently, nanoparticles have been well developed to deliver drugs to the target sites at accurate timing or specific condition.<sup>1</sup> These systems can trigger drug release by specific stimuli (external or internal) through chemical or physical changes. Based on this concept, we have developed a dual-responsive drug delivery system (DDS) by incorporating pH and redox sensitive units into a hyperbranched polymer to allow the controlled drug release in different chemical environment. As shown in **Fig. 1**, a 13-mer peptide-aptamer that has previously been shown to exhibit high specificity for heat shock protein 70 (HSP70) was conjugated to the polymer.<sup>2</sup> This aptamer is also reported to exhibit a chemosensitizing behaviour against cancer cells. Doxorubicin was also bound to the polymeric carrier utilising a degradable hydrazone bond, enabling pH-controlled release under mildly acidic conditions observed in the intracellular environment of cancer cells. siRNA was also conjugated to the polymer via a reduction-sensitive disulfide bond with the aim of developing a dual gene/chemotherapy delivery system. In addition, Cyanamine-5 (Cy5) was incorporated into the polymer as a diagnostic tag for both *in vitro* and *in vivo* tracking. The DDS shows a significantly enhanced release at low pH and high glutathione concentration values, which exhibits an apparent reduction/pH controlled “on–off” drug release pattern. These results demonstrate that this combination of pH-responsive and redox sensitive functionality, could lead to a real-time tunable drug delivery system with the potential of applying as target release drug nanocarriers for cancer therapy.

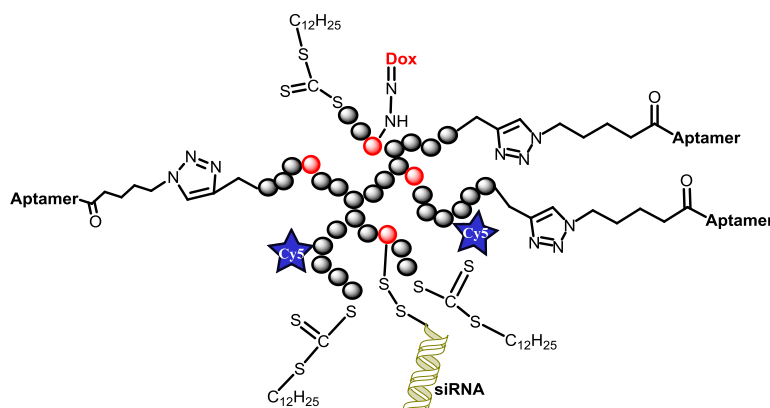


**Figure 1.** Dual gene/chemotherapy delivery system

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<sup>2</sup> A. L. Rerole, J. Gobbo, *Cancer Res.* **2011**, 71 (2), 484-95.

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## Medium bandgap material for solar cell applications

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Polymer solar cells have attracted great attention due to its potential advantages that enables the fabrication of cheap devices on flexible substrates to generate clean and sustainable energy<sup>1</sup>. The synthesis of conjugated polymers that fulfills desired properties is very essential. The donor polymers need to have strong optical absorption with appropriate HOMO and LUMO energy levels and should give a good morphology when blended with electron deficient counterparts (usually C60 derivatives)<sup>2-4</sup>.

Of the different class of conjugated polymers, benzo[1,2-b:4,5-b']dithiophene (BDT) containing polymers have been widely investigated due to its desirable properties such as ease of synthesis, structural rigidity, planarity, extended  $\pi$ -conjugation length and favorable inter-chain  $\pi$ - $\pi$  stacking<sup>5</sup>.

In this work, we synthesized BDT moiety containing medium bandgap polymer and characterized using optical and electrochemical techniques. The polymer was used for the fabrication of organic photovoltaic devices by blending with PCBM[60] and a power conversion efficiency of nearly 5%% was achieved by processing from a non-chlorinated *o*-xylene solvent. The devices were also found to be stable towards ambient conditions which is a one requirement for a good solar cell material.

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# Control of Active Layer Architecture in Organic Photovoltaic Devices Through Block Copolymer Design

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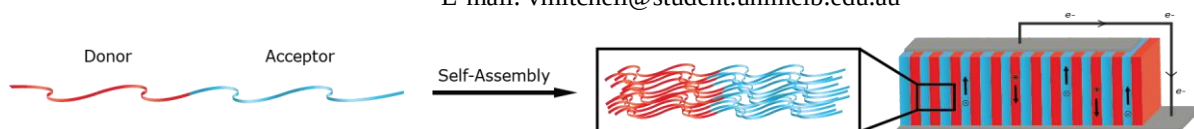


Figure 1. Schematic of block copolymer self-assembly and active layer morphology

Charge generation in bulk-heterojunction organic photovoltaics (BHJ OPVs) depends on the formation of a network of intimately contiguous but distinct domains of a donor and acceptor material. The most common method of obtaining these domains is through blending the donor and acceptor materials and codepositing them onto the chosen electrode. The resultant metastable morphologies are difficult to control and maintain, and small variations in technique and material can lead to suboptimal domain sizes.<sup>1-2</sup>

In this work, an alternative approach is investigated in which the electron donor and acceptor are combined into a single fully-conjugated block copolymer. If properly designed, block copolymers are capable of self-assembly into well-defined domains with size determined by the polymer chain length, a theoretically ideal route to morphological control.<sup>3</sup> However, previous research into block copolymers as active layer materials has shown that achieving complete phase separation into pure donor and acceptor domains can be difficult.<sup>4</sup> Additionally, block copolymer syntheses via polycondensation reactions produce a mixture of donor and acceptor homopolymers as well as di- and tri-block species, a complicated mixture to purify. Thus far analysis of fully-conjugated block copolymers has typically involved polymeric mixtures, with a recent report finding that out of a synthesis of 30 BCPs the percentage of diblock copolymer in the reaction mixture ranged from 33-64%.<sup>5</sup>

Here we report a novel amphiphilic donor-acceptor block copolymer system in which hydrophilic side-chains are incorporated into the acceptor material. This design affords several advantages over previous efforts. The resultant materials showed increased solubility in organic solvents, beneficial for industrial application. The chemical disparity between the donor and acceptor blocks allowed the design of a purification technique able to isolate the block copolymer from the homopolymer contaminants. This is a significant improvement over previously reported systems which employed nonspecific purification techniques incapable of distinguishing between polymer species. Finally, the morphological behavior of these materials was investigated using AFM, SAXS/WAXS, GIWAXS and RSoXS and revealed spontaneous phase separation into lamella-like domains. This indicates that self-assembly in this system is highly favored and thus easily reproduced from film to film. This new amphiphilic strategy is expected to increase the applicability of block copolymers as a viable alternative to traditional blend systems.

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# Controlled Synthesis of Semiconducting Polymers

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Semiconducting polymers is a class of organic material that has been widely employed in organic electronics with applications such as organic light emitting diodes, organic field effect transistors, organic solar cells and sensors. The two key reasons for their popularity are tunable properties through modular structure modifications and solution processability enabling deposition of uniform films. The vast majority of semiconducting polymers are linear rod-like polymers with a conjugated backbone structure. There is a great variety of synthesis methods available for the construction of these conjugated polymers. Most polymerization methods involve step growth reactions that result in poorly defined materials with high polydispersity. To achieve well-defined polymer materials, chain growth polymerization mechanism is required. Currently, there are very few synthetic methods for conjugated polymers that involve chain growth mechanism. The Grignard metathesis polymerization (GRIM) method is commonly used for the controlled synthesis of thiophene homopolymers with the most prominent example being poly(3-hexylthiophene) P3HT. Ring opening metathesis polymerization (ROMP) is a fairly common method for the synthesis of non-conjugated pendant polymers with a couple of conjugated polymer examples that require highly strained cyclic monomer building blocks that are difficult to prepare. Recently, our group prepared a series of well-defined semiconducting bottlebrush polymers using controlled polymerization methods.<sup>1</sup> The electron donor semiconductor, poly(3-hexylthiophene) P3HT, was synthesized using Grignard metathesis polymerization. With appropriate norbornene end caps, the P3HT macromonomers were used to form the bottlebrush material via ring opening metathesis polymerization (**Fig. 1**). The bottlebrush polymers were characterized and compared with linear P3HT materials as well as high molecular weight commercial P3HT. It was found that longer polythiophene chains impart better semiconducting properties resulting in higher solar cell device performance. In this presentation, I will summarise this work and provide an outlook on efforts to improve polymerization control for semiconducting polymers.

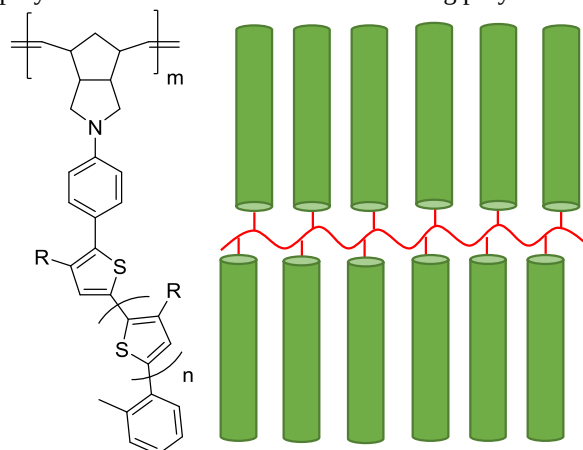


**Figure 1.** Structural illustration of the semiconducting bottlebrush polymer.

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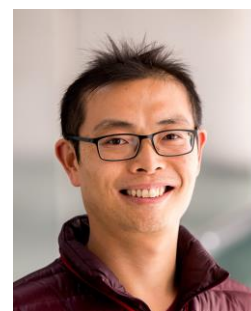
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# Durable Transparent Superhydrophobic Surfaces via Sprayable Interpenetrated Polymer Networks

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Since the advent of biomimetics, natural superhydrophobic plants<sup>1,2</sup> such as the native Australian mottlecane (*Eucalyptus macrocapra*) have received much industrial and research interest.<sup>3</sup> Their durability stems from the strongly cohesive properties of multi-scale organic tissue. Today, although a myriad of synthetic replicas has been achieved in laboratories, severe practical drawbacks exist.<sup>3-5</sup> [ENREF 2](#) Primarily, real-world impact of such materials continues to be diminished by the poor chemical-structural stability of nano-microstructures required for retaining highly dewetting superhydrophobic states.<sup>6</sup> Here, we present a unique bi-layer composite design that biomimics the soft but tough nature of organic tissues in plants, enabling enhanced abrasion resistance. This was achieved through the synthesis of a reactant-product stabilized polyurethane-acrylic colloid (Figure 1).<sup>7</sup> The colloid possesses excellent shelf-life stability, retaining its meta-stable state for extended periods (tested up to 6 months). Upon spray-forming, the colloidal solution self-assembles into a micro-structure roughened surface that is reminiscent of the epidermal layers of superhydrophobic plants. More importantly, the self-assembling polymer realizes complete interpenetration, forming stable; tough and elastic interpenetrated polymer networks (IPNs) during controlled solvent-degassing. Notably, these IPN films can be developed on practically limitless material-types and -geometries.<sup>7</sup> Sequential deposition and integration of an inherently superhydrophobic nanomaterial, such as fluorinated-silica then imparts superhydrophobic functionalities, while preserving micro-roughness. To our knowledge, this technique represents the first exemplification of rapidly developed template-free micro-nano textured IPNs for bioinspired durable superhydrophobicity. These hierarchically textured IPN films possess superior thermal-mechanical properties, with an elongation at break of up to 210 % (3 times its length!). IPN-assisted superhydrophobicity also demonstrated outstanding anti-abrasion properties when analysed with the ASTM D4060 rotary abraser. The pristine Cassie-Baxter<sup>8</sup> effect was well-preserved, with sliding angles of below 10° for up to 120 continuous abrasion cycles (Figure 2). Coatings also demonstrated superior chemical- and photo-stability, preserving initial performance upon more than 50 h exposure to intense UVC light (254 nm, 3.3 mW cm<sup>-2</sup>), 24 h of oil contamination, and highly acidic conditions (1 M HCl).<sup>7</sup>

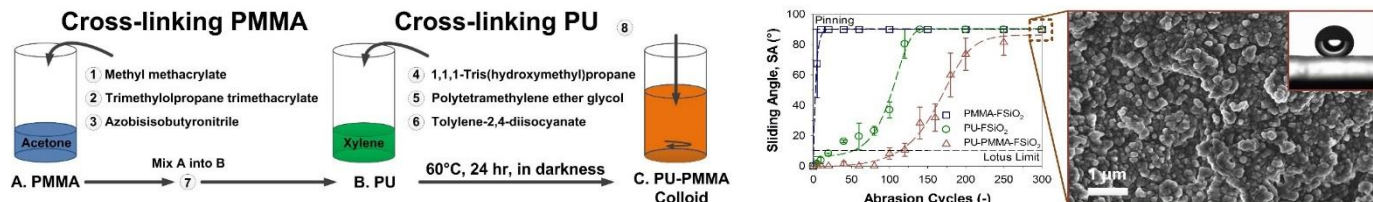


**Figure 1. Synthesis of a sprayable stable PU-PMMA colloidal pre-IPN.**

**Figure 2. Abrasion Resistant Superhydrophobic PU-PMMA-based interface vs. Controls**

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<sup>5</sup> Lu, Y.; Sathasivam, S.; Song, J.; Crick, C. R.; Carmalt, C. J.; Parkin, I. P. *Science* **2015**, *347*, 1132-1135.

<sup>6</sup> Liu, G.; Wong, W. S. Y.; Nasiri, N.; Tricoli, A. *Nanoscale* **2016**, *8*, 6085-6093.

<sup>7</sup> Wong, W. S. Y.; Stachurski, Z. H.; Nisbet, D. R.; Tricoli, A. *ACS Appl. Mater. Interfaces* **2016**, *10*, 1021/acsami.6b03414.

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## Stretchable and self-healing conducting polymers

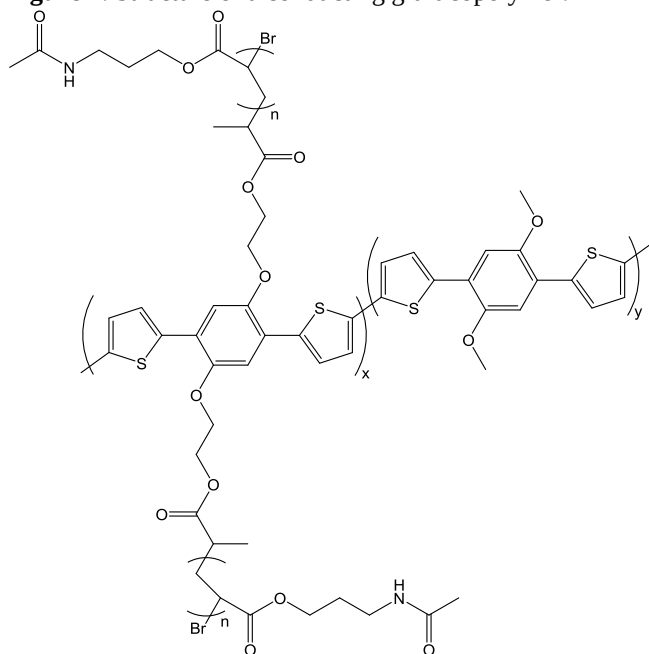
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Stretchable electronics can find many applications in fields of medical devices, implanted sensors and soft robotics. Combining stretchable electronics with self-healing properties would increase the lifetime and usefulness of these devices. While some examples of stretchable and self-healing electronics have been reported, these are always formed by composites or layers of different materials, with one material providing conductivity and another providing self-healing properties. Here we aim to create a single material that has intrinsic conducting properties, as well as being stretchable, self-healing and adhesive.

Building on a recently developed platform of functionalizable polythiophene phenylenes,<sup>28</sup> we can combine a rigid conducting polymer chain with flexible grafted side chains (**Fig.1**), which are polymerized via controlled radical polymerization from initiating groups on the polythiophene phenylene backbone. By using acrylates with hydrogen-bonding functionality, such as amides, self-healing properties can be implemented into the material.<sup>29</sup> Additional properties, such as adhesiveness to surfaces and thermal properties, are also studied.

**Figure 1.** Structure of a conducting graft copolymer.



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# Polymeric Interface Materials and Environmentally Friendly Processing Methods for Polymer Solar Cells

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Polymer solar cells have gained considerable interest during the last decades. The most promising active layer material comprises so-called bulk-heterojunction blends of a donor polymer that is finely mixed with a fullerene acceptor. The photovoltaic performance has increased rapidly over the last years with high power conversion for lab-scale single-junction devices. One advantage of polymer solar cells is the possibility to use roll-to-roll coating techniques for large-scale device production. However, several issues must be resolved before this can be realised commercially. Two critical aspects concern (1) the inverted geometry of large-area devices that typically require stable modification of the bottom electrode with interlayers, and (2) environmentally friendly deposition of the active material.

So far a number of different types of materials have been used as interface materials for work function modifications. We have synthesised and evaluated the stability of a number of such materials having different functional groups. The materials include modified fullerenes, polyfluorenes and the use of ethoxylated polyethylenimine (PEIE) and poly(4-vinyl pyridine) (P4VP). To evaluate the stability of the work function modification of ITO, UPS measurements have been performed on samples exposed to ambient conditions for up to four weeks. For most of the materials the work function was found to stabilise after two weeks and the largest changes were found for the materials with conjugated backbones. In devices we have seen a clear effect on the open circuit voltage and in some cases an improved thermal stability of the morphology as well. When using P4VP as the interphase layer on top of ZnO we have seen an increase in the power conversion efficiency from 5.1% to 6.5% for TQ1 devices.

In our search for environmentally friendly deposition methods we have concentrated our efforts on the preparation of water dispersion of nanoparticles. The most widely used preparation method involves the use of charged surfactants, for example sodium dodecyl sulfate (SDS). The potential shortcoming of this approach is that the surfactants will remain fugitive and trapped within the active material, decreasing the stability/lifetime of the final device. We have focused our efforts on making materials that can be used to prepare nanoparticles with and without the addition of surfactants. One of the methods we have used is to incorporate tertiary amine side groups on the polymer backbone. After protonation of the amino groups with acid, stable aqueous miniemulsions and nanoparticles could be prepared. The morphology of the active layers is studied using different techniques including dynamic mechanical analysis (DMA) to determine a suitable annealing temperature for the coalescence of the nanoparticles. These methods present a promising way to deposit the active material in the solar cells from aqueous dispersions.

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## Nanostructural control via processing kinetics in plastic solar cells

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Plastic solar cells hold the promise of inexpensive, flexible energy generation; however, pioneering attempts to scale-up production have generated solar cells with power conversion efficiencies that decay rapidly over the short device lifetimes. The ideal plastic solar cell is a nanostructured, two-phase mixture comprising, i.e. a polymeric electron donor and a fullerene-based electron acceptor (**Fig.1a**). The growth of micron-sized fullerene crystals is largely responsible for the loss of performance during the operation of plastic solar cells (**Figs.1b and c**). The glass transition temperature of the polymer component, commonly poly-3-hexylthiophene, is  $\sim 40^\circ\text{C}$ , well below the expected operation temperature of solar cells,  $80^\circ\text{C}$ . During operation, kinetically trapped fullerene crystals continue to grow within the rubbery polymer matrix, reducing the amount of interfacial area available for charge separation and decreasing the cell efficiency. Two general strategies are proposed for inhibiting fullerene crystal growth: 1) nucleating a large population of nanoscale fullerene crystals, which prevents any individual crystal from growing to the micron scale and 2) inducing a higher degree of crystallinity in the polymer matrix, which mechanically prevents the growth of fullerene crystals. Both these strategies depend on the processing conditions of the polymer/fullerene composite.

The two most common processing techniques, spincoating and printing, were translated from thin films to bulk samples to minimize the effects of surface forces; rapid quenching into a non-solvent and drop-casting, respectively, were chosen as the analogous bulk techniques. Small-angle neutron scattering was used to examine the structures that resulted in polymer/fullerene blends (**Fig.1d**). Slowly dried samples exhibited a small population of polymer crystals and a homogeneously mixed polymer/fullerene phase. Upon annealing, these samples evolved micron-sized fullerene crystals. Rapidly quenched samples exhibited a large population of both polymer and fullerene crystals; upon annealing, no structural changes were observed. These results suggest that rapid phase transitions can be harnessed to build a population of fullerene nuclei that controls further growth.<sup>5</sup> Current work seeks to translate these fast-quenches into a scalable deposition technique for thin films.

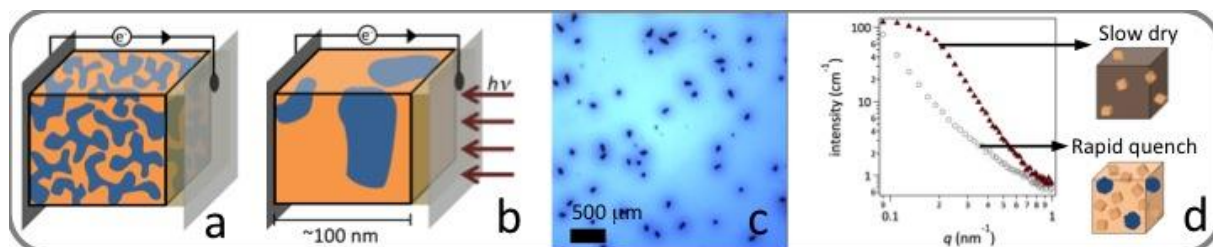


**Figure 1.** a) and b) are schematics of an idealized solar and one in which the fullerene crystals have aggregated, respectively. c) Optical micrograph of fullerene crystals. d) Small-angle neutron scattering spectra of polymer/fullerene composites showing schematics of the measured structures.

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## Synthesis and Characterization of Polymer Semiconductors: Shining New Light on Plastics

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Polymer Electronics has taken science by storm because of their potential to revolutionize today's technologies such as displays,<sup>1</sup> electronics,<sup>2</sup> mechanical actuators,<sup>3</sup> solar cells,<sup>4</sup> and sensory devices.<sup>5-6</sup> In fact, Polymer Electronics are becoming reality through companies such as LG Electronics, Samsung (OLEDs), Heliotech (Solar Cells), Plastic Logic (OFET, RFID), Fido (Explosive Sensors). Understanding the properties of organic semiconductors, as dictated by their chemical structure, is pivotal in producing more efficient devices. For instance, tuning the electron affinity of the macromolecular material can have a dramatic effect on the sensing of various analytes like nitroaromatic compounds. In this seminar, I will discuss the synthesis of new conjugated polymers and how they are applied to the next generation of microelectronic devices like Sensors, LEDs and Transistors (Fig. 1).

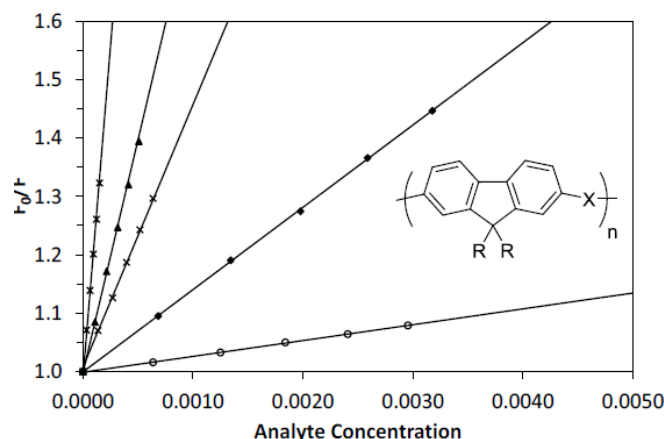


**Figure 1.** Stern-Volmer Quenching using various analytes of fluorene-based polymer semiconductor. These results demonstrate both sensitivity and selectivity.

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<sup>3</sup> D. Gendron, A. Ansaldi, G. Bubak, L. Ceseracciu, G. Vamvounis and D. Ricci, *Composites Science and Technology* **2015**, 117, 364-370.

<sup>4</sup> G. Vamvounis, C. R. Glasson, E. J. Bieske, V. Dryza, *Journal of Materials Chemistry C* **2016**, in press: DOI: 10.1039/C6TC01584K

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<sup>6</sup> G. Vamvounis, M. Fuhrera, A. Koizumia, M. Gao and S. Hu, *submitted for publication*.

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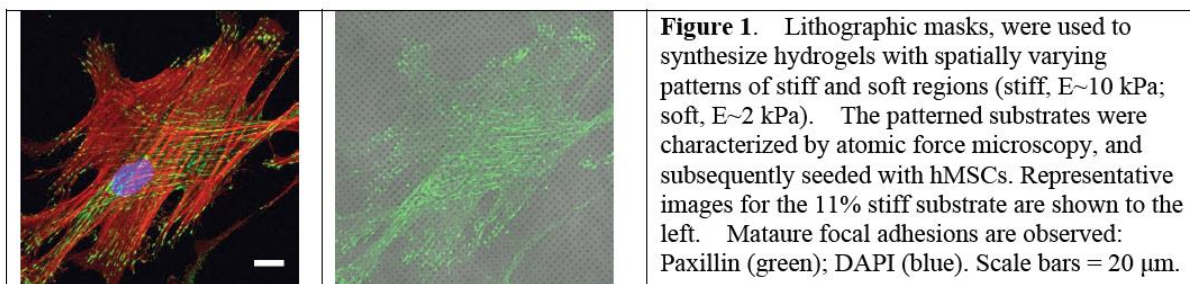
## Photodegradable hydrogels as spatiotemporally controllable cellular niches

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The expansion and differentiation of human mesenchymal stem cells (hMSCs), an important cell type that is frequently used in stem cell based therapies, are regulated by interactions with their local extracellular microenvironment. Matrix stiffness is one of the key regulators, but often varies in space at a range of length scales in the local niche. To better understand and harness cell-matrix interactions, we utilized a photo-tunable hydrogel platform<sup>1,2</sup> to provide compelling evidence that the magnitude and spatial organization of matrix mechanics influence hMSCs through systematic regulation of cytoskeletal tension and transcriptional activation. Specifically, a hydrogel cell culture substrate was fabricated with regions of spatially varied and distinct mechanical properties, which were subsequently mapped and quantified by atomic force microscopy (AFM) (Fig.1)<sup>3</sup>. The variations in the underlying matrix mechanics were found to regulate cellular adhesion and transcriptional events. Highly spread, elongated morphologies and higher Yes-associated protein (YAP) activation were observed in hMSCs seeded on hydrogels with higher concentrations of stiff regions in a dose dependent manner. However, when the spatial organization of the mechanically stiff regions was altered from a regular to randomized pattern, lower levels of YAP activation with smaller and more rounded cell morphologies were induced in hMSCs. We infer from these results that irregular, disorganized variations in matrix mechanics, compared to regular patterns, appear to disrupt actin organization, and lead to different cell fates; this was verified by observations of lower alkaline phosphatase (ALP) activity and higher expression of CD105, a stem cell marker, in hMSCs in random versus regular patterns of mechanical properties<sup>2</sup>. These findings may have important implications in deepening our fundamental understanding of the role of extracellular matrix organization on disease, aging and regenerative processes.



<sup>1</sup> A.M. Kloxin, A.M. Kasko, C.N. Salinas, K.S. Anseth, *Science*. **2009**, 324, 59-63.

<sup>2</sup> C. Yang, M.W. Tibbitt, L. Basta, K.S. Anseth, *Nature Materials*. **2014**, 13, 645-652.

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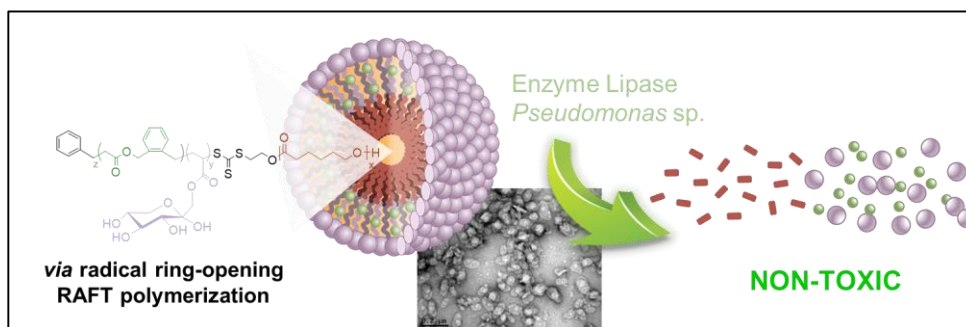


# Biodegradable and Biocompatible GlycopolymERIC Nanoparticles for Drug Delivery

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The design and synthesis of a type of entirely biodegradable carbohydrate-coated nanoparticles will be presented. This design relies on the utilization of RAFT-controlled radical ring-opening polymerization (rROP) technique to afford multiple insertions of cleavable ester linkages onto the backbone of the glycopolymer as the corona-forming block of the nanoparticles.<sup>1</sup> This approach enables the generation of biologically reducible nanoparticles *via* enzymatic attack, yielding non-toxic degradation products as investigated by using *in vitro* cell proliferation assay, thus opening opportunities to the design of new bioactive nanoparticles that are biocompatible and biodegradable for drug delivery purposes. The building blocks employed in the design of the nanoparticles include poly( $\epsilon$ -caprolactone) (PCL) that forms the hydrophobic core of the nanoparticles, which was used as the macroRAFT agent to control the polymerization of well-defined statistical glycopolymers of 1-*O*-acryloyl-2,3:4,5-di-*O*-isopropylidene- $\beta$ -D-fructopyranose (1-*O*-AiPrFru) and 5,6-benzo-2-methylene-1,3-dioxepane (BMDO) monomer. Self-assembly of the deprotected block copolymers generated non-spherical elliptical shaped micelles. The biodegradation of these micelles were investigated under physiological condition *via* incubation with enzyme Lipase *Pseudomonas* sp., indicating entirely degradable architectures which are no longer visible *via* dynamic light scattering (DLS) and further confirmed by SEC analysis. *In vitro* cell proliferation assay of the micelles and their degradation products revealed no toxicity against healthy human fibroblast HS27 and breast cancer MDA-MB-231 cell lines. The polymer concentration range tested was up to 0.20 mg·mL<sup>-1</sup> with the cell viabilities of  $\geq 95\%$ . Further analyses and physical characterizations were conducted to reveal the morphology of the nanoparticles to be flat lamellae disks resembling nanoplatelets.



S. Ganda, Y. Jiang, D. S. Thomas, J. Eliezar and M. H. Stenzel, *Macromolecules*. **2016**, *final corrections*.

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| 2011-2015  | B.Eng (Chemical Engineering), Honours, UNSW Australia                                        |

Research interests: the synthesis and development of carbohydrate-based polymers and nanoparticles to be used as drug delivery vehicles and studying the morphological transition behaviour and physical properties of crystalline-based polymeric nanoparticles to find use in biomedical applications.

## Nucleotide Functionalised Synthetic Polymers

*Michael Wilson<sup>1</sup>, Elizabeth G. Williams<sup>2</sup>, Greg Simpson<sup>2</sup>, Amanda V. Ellis<sup>1</sup>*

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This paper outlines the synthetic methods for producing nucleotide functionalised synthetic polymers using reversible addition-fragmentation chain-transfer (RAFT) polymerisation. First a modified phosphoramidite approach is used to produce a monomer for the RAFT polymerisation. Four different monomers were prepared using protected nucleotides of adenine, thymine, guanine and cytosine. This was achieved by coupling 2-hydroxypropyl methacrylate (HPMA) to a 3'-deoxynucleotide (**Fig. 1a**), 5'-dimethoxytrityl-2'-deoxy nucleobase, 3'-[(2-cyanoethyl)-(N,N-diisopropyl)]-phosphoramidite (DCP), resulting in a HPMA-3'-deoxynucleotide monophosphate bioconjugate (**Fig. 1b**). Coupling of the HPMA to the DCPs was determined using <sup>1</sup>H nuclear magnetic resonance (NMR) spectroscopy and electrospray ionisation-mass spectrometry (ESI-MS) to confirm the substitution of the diisopropylamine with the HPMA. The coupling yield was determined to be >80% for each of the 4 nucleotides.

Each individual HPMA-3'-deoxynucleotide monophosphate bioconjugate was then polymerised using an azobisisobutyronitrile initiator and 4-cyano-4-(phenylcarbonothioylthio) pentanoic acid *N*-succinimidyl ester as a RAFT chain transfer agent (CTA) at 70 °C. The kinetics of the polymerisation were measured with <sup>1</sup>H NMR spectroscopy using the change in peak area ratio of the vinyl peak at 5.4 ppm relative to the initiator and RAFT CTA agent peaks at 1.4-1.5 ppm indicating a 55% conversion in 5 hours. Following this time size exclusion chromatography was used to determine the polydispersity index of 1.3 and Mw of 13.5 kDa.

Deprotection of the bioconjugate was conducted with 2 successive workups. Firstly, heating with excess ammonia at 50 °C for 48 hours for deprotection of the nucleobase and phosphate. This was followed by isolation and treatment with concentrated acetic acid to remove the 5' hydroxyl protecting group. <sup>31</sup>P NMR spectroscopy was used following workup to confirm oxidation of the phosphorous from the P3 to P5 state. Current work is focussed on the conversion of the bioconjugate from a monophosphate to a triphosphate for use in enzymatic systems, specifically the polymerase chain reaction, for template polymer synthesis.

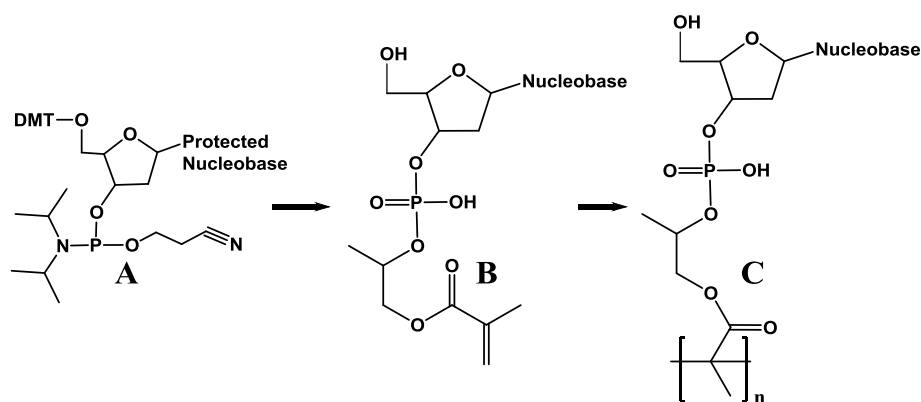


**Figure 1.** Major steps in the synthesis of a nucleotide functionalised synthetic polymer showing A) 5' phosphoramidite, B) bioconjugate monomer, and C) bioconjugate polymer

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2007-2011 Bachelor of Science- Flinders University

2013-2014 Honours in Nanotechnology- Flinders University

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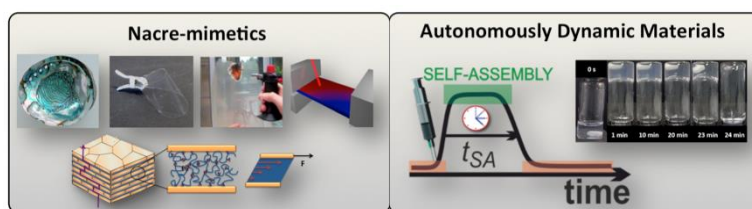
# Static and Dynamic Bioinspired Aqua-Materials

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Biology is a source of inspiration for materials science by demonstrating macroscopic materials with advanced functionalities and excellent mechanical properties. However, although these rather static architectures stimulate large interest, even more thought-provoking are the dynamic, kinetically controlled processes and temporal evolution of structures in complex biological systems. These are orchestrated through feedback loops and require energy input and dissipation to allow non-equilibrium materials and full spatiotemporal control.

In man made self-assemblies we have mastered to a large extent near-equilibrium structure formation in space and have gained an increasing understanding of how to construct very complex, hierarchically structured soft matter by using co-assemblies, competing interactions and hierarchical length scales. This has allowed to create real-life materials with unprecedented functionalities, inaccessible without delicate control over molecular interactions and sophisticated nano- and mesostructuration. The next step is to master temporal control in self-assemblies. This requires kinetic control of opposing reactions (built-up/destruction), internal feedback systems or the use of energy dissipation to sustain structures only as long as a chemical fuel is available. These approaches keep systems forcefully away from equilibrium and potentially allow neat access to temporal control. In this talk I will present concepts for bioinspired materials formed in both static and dynamic conditions. The first part will deal with rather static, nacre-inspired high-performance nanocomposites formed through self-assembly, and in which light-adaptive properties can be encoded through co-assembly and energy transfer approaches. The second part will focus on a platform concept, which allows to program self-assembling systems outside equilibrium with a lifetime by kinetic control of promoter/deactivator pairs and a simple internal feedback system. This will be showcased for different self-assembling systems.



## Recent references:

**Self-Assembled Artificial Nacre:** *Angew. Chem. Int. Ed.* **2015**, 54, 8653; *Nat. Commun.* **2015**, 6, 5967; *ACS Appl. Mater. Interfaces* **2013**, 5, 3738; *Adv. Mater.* **2013**, 25, 5055;

**Time Programmed Dynamic Materials:** *Nano Lett.*, 2015, 15, 2213, *Angew. Chem.* **2015**, 54, 13258 ,

**Review:** *Soft Matter* **2015**, 11, 7857.

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# Synthesis of Antibacterial Peptide Analogues using Sequence Controlled Polymerization Techniques

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The evolution of antibiotic resistant bacteria and a lack of new antibiotics continues to cause well-publicised concern world-wide.<sup>1</sup> In the quest for new antibiotic candidates, researchers have refocused on naturally occurring antibacterial lipopeptides. These low molecular weight peptides have a broad spectrum of activity and low toxicity to mammalian cells. However, they are costly to produce in large amounts and have poor pharmacokinetic properties: they exhibit low half-lives in vivo due to their degradation by proteases. As such, the development of synthetic analogues to antimicrobial peptides is of considerable interest. Compared to antimicrobial peptides, antimicrobial polymers with a balance of cationic and hydrophobic moieties (essential for their activity) can be produced cost-effectively in large quantities, are more adaptable to drug-delivery methods, and provide a flexible framework for systematic chemical pharmacophore modification and adaptation. Importantly, recent progress in the sequence control of synthetic polymers has opened the opportunity for full translation of the critical structural features of antibacterial peptides that afford effective bacterial killing to a wholly synthetic polymer system.<sup>2</sup>

Following from our initial work<sup>3</sup>, we report the synthesis of a library of cationic polymers with uniquely low degrees of polymerization and tuneable monomer sequence, via Cu(0)-mediated polymerization. We employed two biologically relevant cationic monomers, and two polyether comonomers. The polymers possessed molecular architecture-dependent antibacterial activity when tested against *Staphylococcus aureus* ATCC 29213, *Klebsiella pneumoniae* ATCC 13883, *Acinetobacter baumannii* ATCC 19606, and *Pseudomonas aeruginosa* ATCC 27853 as a representative palette of Gram-positive and Gram-negative ESKAPE pathogens. A superior compound was identified, and its mechanism of action investigated via membrane permeability testing against *Escherichia coli*. Through the control of monomer sequence, mammalian cell toxicity could also be manipulated.

<sup>1</sup>CDC-Get Smart: Know When Antibiotics Work 2013 [17 April 2014]. Available from: <http://www.cdc.gov/getsmart/antibiotic-use/antibiotic-resistance-faqs.html#how-bacteria-resist>.

<sup>2</sup> A. Anastasaki; V. Nikolaou; G. Nurumbetov; P. Wilson; K. Kempe; J. F. Quinn; T. P. Davis; M. R. Whittaker; D. M. Haddleton, *Chemical Reviews* **2016**, 116, 835–877

<sup>3</sup> J.L. Grace; J.X. Huang; S.E. Cheah, N.P. Truong; M.A Cooper, J. Li; T.P. Davis; J.F. Quinn; T. Velkov; M. R. Whittaker, *RSC Advances*, **2016**, 6, 15469-15477

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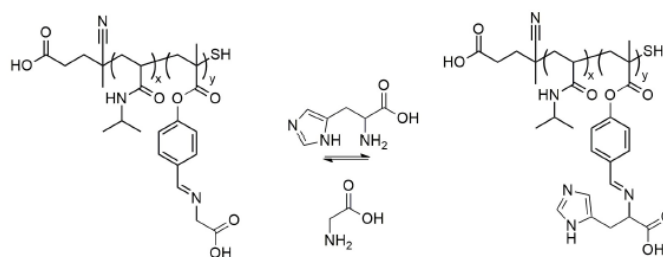
Research interests: Polymer chemistry

## The Dynamic and Versatile Chemistry of Amino Acid Functional Polymers

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The copolymerization of N-isopropylacrylamide with aldehyde functional monomers facilitates post polymerisation functionalisation with amino acids via reductive amination.<sup>[1]</sup> Reductive amination is the reduction of an imine formed from the condensation reaction between an amine and an aldehyde. In this work, we characterize amino acids into four groups based on the reactivity of their side chains (neutral, polar neutral, acidic and basic) and react their amine groups in condensation reactions with aldehyde functional polymers. The dynamic nature of the imine as well as the versatility of reductive amination to functionalise a polymer with a range of amino acids is highlighted. In this manner, amino acid functional polymers are synthesized without the use of protecting groups with high yields, demonstrating the high functional group tolerance of carbonyl condensation chemistry and the subsequent reductive of the imine. The resulting temperature responsive, amino acid functional polymers are characterised and their temperature response is explored. Prior to reduction of the imine bond, Scheme 1 shows the transimination reactions used to shuffle between a glycine functional polymer and a histidine functional polymer.



**Scheme 1.** Transimination shuffling reactions between glycine and histidine functional polymers.

<sup>1</sup> E. R. L. Brisson, Z. Xiao, L. Levin, G. V Franks, L.A. Connal, Polym Chem 2016, 7, 1945-1952.

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Research interests: Biomimicry, Polymer Science, Polymer Self-Assembly

## Nitroxide-containing materials for biofilm remediation

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The adhesion of bacteria to surfaces and their subsequent ability to aggregate into colonies called biofilms is a significant global problem in numerous applications, particularly as biofilms display an inherent resistance to antimicrobial agents. Low concentrations of the diatomic free radical nitric oxide has been shown to disperse bacterial biofilms to free swimming planktonic bacteria,<sup>1</sup> which has led to the development of coatings that release nitric oxide. However, the non-systemic delivery of gaseous nitric oxide remains challenging as nitric oxide is an extremely reactive molecule with a very short half-life. As an improved strategy, we have recently shown that nitroxides (persistent and stable free radical species that are sterically hindered versions of nitric oxide) can inhibit bacterial biofilm formation and disperse existing biofilms (Figure 1).<sup>2</sup> This presentation will report our current work towards the design and development of novel chemistries to immobilise and/or release nitroxides from surface coatings and microparticles as an innovative strategy for combating biofilm growth in a range of applications.

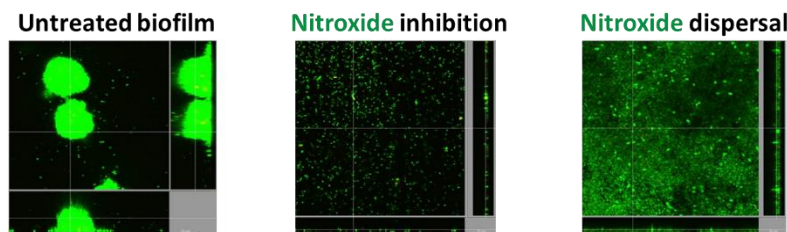


**Figure 1.** Confocal microscopy images showing inhibition and dispersal of *Pseudomonas aeruginosa* biofilms by nitroxides (live cells stained green).

<sup>1</sup> N. Barraud, D. J. Hassett, S. Hwang, S. Kjelleberg and J. S. Webb, *Journal of Bacteriology*, **2006**, *188*, 7344-7353.

<sup>2</sup> C. de la Fuente-Núñez, F. Reffuveille, K. E. Fairfull-Smith and R. E. W. Hancock, *Antimicrobial Agents and Chemotherapy*, **2013**, *57*, 4877-4881.

### Kathryn E. Fairfull-Smith

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| 2009-2012  | Vice-Chancellor's Research Fellow, Queensland University of Technology |
| 2012-2014  | Senior Lecturer, Queensland University of Technology                   |
| Since 2015 | ARC Future Fellow and A/Prof, Queensland University of Technology      |

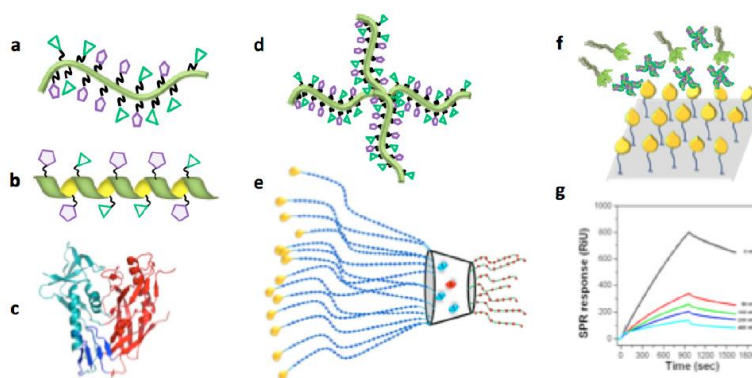
Research interests: Anti-biofilm coatings, fluorescent probes, nanosensors for drug delivery and imaging

## Precision glycopolymers and their interactions with lectins

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Sequence controlled polymers have been attracting more and more attention to deliver the desired properties to the advanced materials by the help of their precisely controlled compositions and architectures.<sup>1</sup> Understanding the specific multivalent carbohydrate-protein interactions is crucial to determine the structure-property relationships and to design accordingly the next generation of functional glycomaterials. Therefore, we investigate the structure-property relationships between the mammalian lectins and multivalent carbohydrate polymers, which may have applications for anti-adhesion therapy. Moreover, we have investigated the affinity of poly(mannose-methacrylate), helical glycopolypeptides, gp120, star shaped glycopolymers, and cyclodextrin centered glycopolymers with a selected mannose binding lectin (DC-SIGN) that exists on dendritic cells, using SPR technique.<sup>2-4</sup> Selected members of a glycopolymer library were used to demonstrate the interactions between DC-SIGN and mannose rich polymers. We extend this study to a broader set of polymers to examine the effect of chain length, end group, architecture, thermoresponsive block, and number of arms in the star shaped polymers on the lectin binding.



**Scheme 1.** Schematic representation of (a) linear GP, (b) helical glycopolypeptide, (c) glycoprotein 120 (gp120), (d) star shaped GP, (e) cyclodextrin centred GP, (f) SPR measurement on glycopolymer-DCSIGN competition in solution, (g) SPR response-time for the interaction of DC-SIGN & gp120.

1. C. R. Becer, R. Hoogenboom and U. S. Schubert, *Angew. Chem. Int. Ed.*, 2009, **48**, 4900-4908.
2. C. R. Becer, *Macromol. Rapid Commun.* 2012, **33**, 742-752.
3. C. R. Becer, M. I. Gibson, J. Geng, R. Ilyas, R. Wallis, D. A. Mitchell and D. M. Haddleton, *J. Am. Chem. Soc.*, 2010, **132**, 15130-15132.
4. Q. Zhang, J. Collins, A. Anastasaki, R. Wallis, D. Mitchell, C. R. Becer, and D. M. Haddleton, *Angew. Chem. Int. Ed.* 2013, **52**, 4435-4439.

### Dr. Remzi Becer

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Research interests: polymer chemistry, glycopolymers, biomaterials

# Triblock copolymers as a new switchable superhydrophobic or superhydrophilic surface coating

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Multifunctional polymer surfaces based on the hierarchal surface structure, show Ultrahydrophobic and Superhydrophilic behavior. To achieve this multifunctionality it is necessary, to combine different functionalities in one material or in this case in one polymer molecule. This implies the production of core-shell-nanoparticle layers on a variety of substrates as polymers and metals with easy-to-clean (anti-soil and anti stain behavior) and self-cleaning properties.

The Ultrahydrophobic properties of a lotus leaf are based on a hierarchal surface structure in the  $\mu\text{m}$  and nm range. The lotus leaf surface can be copied by the use of a polymer particle in the  $\mu\text{m}$  range and a polymer coating in the nm range.

The synthesis of tri block copolymers using controlled living polymerization techniques and the preparation of the core-shell-nanoparticles will be presented <sup>1,2</sup>.

The superior advantages and possibilities of 2 - dimensional separation techniques including FT-IR identification will be discussed to determine the molar mass and the composition of the polymers

The discussed polymers consist of a block responsible for the anchoring to the substrate, a “hydrophilic” and a “hydrophobic” block. This offers the possibility to combine responsive switching of the wettability behaviour and the anchoring to a substrate in one molecule. The wetting behaviour of the prepared polymer brushes can be switched by external stimuli: temperature, pH-value, solvent atmosphere, salt concentration etc. (figure 1).

Superhydrophilic and Ultrahydrophobic surface behavior was achieved using  $\mu\text{m}$  based hierarchical silica particles and nm scaled polymer surface structure. Applications for the possible industrial use of these new surface properties will be shown. .

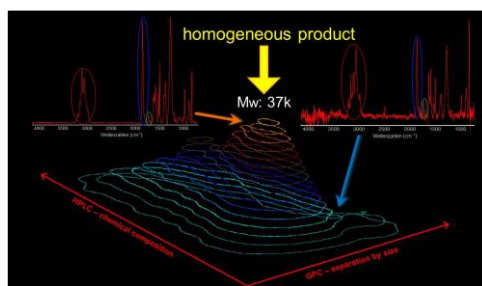
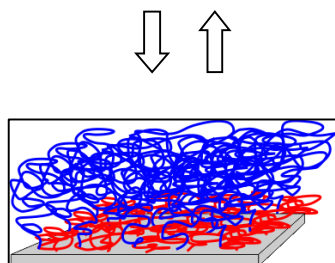


**Figure 1: switching principle of a binary (mixed) polymer brush** **Figure 2: 2D analysis of a triblockcopolymer**

The references should be cited in the text as superscripted number<sup>30</sup> and the reference should be added after the text using the format below

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Research interests: controlled polymerization, polymer characterization

## Novel Antimicrobial Polymeric Nanoparticles

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The development of novel reagents and antibiotics for combating multi-drug resistance bacteria has been receiving significant attention in recent years. The use of synthetic biomacromolecules/nanoparticles prepared via controlled polymerization techniques is regarded as one of the most promising approaches to overcome such challenge because of the ability to precisely engineer the chemical functionality, and hence the biological properties, of the nanomaterial. Several research groups have reported on the synthesis of antimicrobial polymers to combat the rise of antibiotic-resistant pathogens.<sup>1,2,3</sup> However, some materials are not compatible with mammalian cells which therefore hinder their potential for clinical applications. Nonetheless, these early developments provide the inspiration for future technologies. Herein, a new class of antimicrobial polymeric nanoparticle that exhibit selective killing of bacteria over mammalian cells is developed using controlled polymerization methodologies. The technology described in here represents an innovative approach in fighting deadly infectious diseases.

<sup>1</sup> D. J. Coady, Z. Y. Ong, P. S. Lee, S. Venkataraman, W. Chin, A. C. Engler, Y. Y. Yang, J. L. Hedrick, *Adv. Healthcare Mater.* **2014**, 3, 882-889.

<sup>2</sup> J. L. Grace, J. X. Huang, S. -E. Cheah, N. P. Truong, M. A. Cooper, J. Li, T. P. Davis, J. F. Quinn, T. Velkov, M. R. Whittaker, *RSC Adv.* **2016**, 6, 15469-15477.

<sup>3</sup> E. H. H. Wong, M. M. Khin, V. Ravikumar, Z. Si, S. A. Rice, M. B. Chan-Park, *Biomacromolecules* **2016**, 17, 1170-1178.

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Research interests: biopolymers, antimicrobial, anticancer, complex macromolecular architectures, controlled polymerization

# Synthesis of polymeric microcapsules with an aqueous core via membrane emulsification and inverse emulsion periphery RAFT polymerization

*Fumi Ishizuka<sup>1</sup>, Rhiannon Kuchel<sup>2</sup>, Hongxu Lu<sup>3</sup>, Martina Stenzel<sup>3</sup> and Per B. Zetterlund<sup>1</sup>*

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Hollow polymeric particles have a wide range of application in various areas such as the biomedical field, cosmetics, food and water treatment. A number of encapsulation techniques utilizing hollow particles, liposome and emulsion droplets have been developed for the encapsulation of various materials such as anticancer drugs, enzymes, nucleotides, fragrances and catalysts. Hollow polymeric particles have been extensively studied for biomedical applications due to their high capacity for encapsulation of therapeutic agents. A number of approaches to synthesize hollow particles have been reported, however it is still challenging to control the size, shell thickness and surface functionality. Previously, we developed a facile synthesis method for nanocapsules via inverse miniemulsion periphery RAFT polymerization (IMEPP).<sup>31</sup> This approach provides a reaction-free core environment for encapsulation of fragile molecules such as proteins since the polymerization occurs on the periphery of the droplets as opposed to within the droplets. However, the size range of polymeric nanocapsules has been limited to approximately 50-500 nm because a conventional emulsification method was used to create the initial inverse miniemulsions.

In this study, we have explored the use of “Shirasu Porous Glass (SPG) membrane emulsification” to create a wide size range of water droplets stabilized with amphiphilic macroRAFT species comprising poly(di(ethylene glycol) methyl ether methacrylate) and poly(methyl methacrylate). SPG membrane emulsification is a relatively novel technique which can produce monodisperse droplets with lower energy compared to conventional techniques such as ultrasonication. This technique was applied for the preparation of inverse emulsions. By changing the pore size of the SPG membrane (0.2-3  $\mu\text{m}$ ), we have succeeded in controlling the droplet size from submicron to tens of microns (**Fig. 1**). The IMEPP technique was applied to create polymeric shells around water droplets, leading to hollow microparticles with similar size to the initial droplets and relatively narrow size distribution. Water-soluble guest molecules such as a fluorescent dye and a protein were successfully encapsulated within these microcapsules.

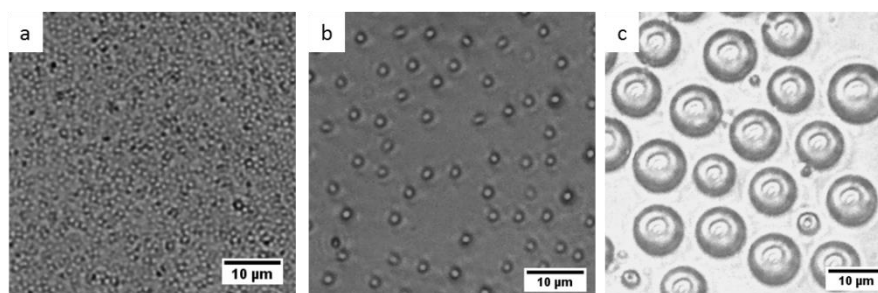


**Fig. 1** Optical micrographs of microcapsules prepared by SPG membrane with pore size of 0.2 (a), 0.8 (b) and 3.0  $\mu\text{m}$  (c).

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Since 2014 Ph.D. in Chemical Engineering, University of New South Wales, Australia

Research interests: nano/microcapsules, drug delivery, membrane emulsification



# High Glass Transition Temperature Fluoropolymers for Hydrophobic Surface Coatings via RAFT Copolymerization

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The preparation of polymer thin films or surface coatings that display a static water contact angle  $> 95^\circ$  often requires hierarchical roughness features or surface functionalization steps. In addition, inherently hydrophobic polymers such as fluoropolymers often possess low glass transition temperatures, reducing their application where thermal stability is required.

In this work, we report the first synthesis of 2,3,4,5,6-pentafluorostyrene (PFS) and *N*-phenylmaleimide (NMI) via reversible addition–fragmentation chain-transfer (RAFT)-mediated free radical polymerization.<sup>1</sup> The copolymerization of these monomers was specifically chosen with a view towards the preparation of inherently hydrophobic polymers with a high glass transition temperature. A suite of copolymers were prepared and characterized, and owing to the inherent rigidity of the maleimide group in the polymer backbone and  $\pi$ – $\pi$  interactions between adjacent PFS and NMI groups, very high glass transition temperatures were achieved (up to  $180^\circ\text{C}$  at a mole fraction of NMI = 0.37). The polymers also exhibited relatively low dispersities ( $D < 1.26$  in all cases). Side-chain interactions within the PFS-NMI copolymers were confirmed by FTIR and 2D NMR spectroscopy. The copolymerization of *N*-pentafluorophenylmaleimide was also performed, also resulting in extremely high glass transition temperature copolymers; however, these polymers did not exhibit characteristics of being under RAFT control. Thin films of PFS-NMI copolymers exhibited a static contact angle  $\sim 100^\circ$  upon a simple thermal annealing step, essentially independent of the amount of NMI incorporated into the polymer.

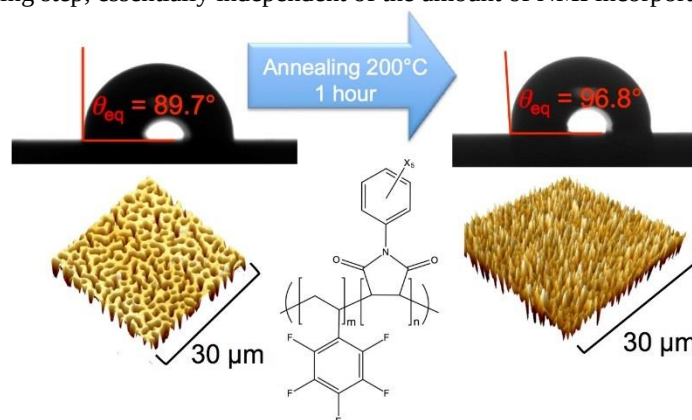


**Figure 1.** PFS-NMI copolymers as high glass transition polymers for hydrophobic surface coatings. Thermal annealing reduces the surface roughness and increases the static water contact angle.

1. M. Rowe, G. H. Teo, J. Horne, O. Al-Khayat, C. Neto and S. C. Thickett, *Aust. J. Chem.*, 2016, DOI: <http://dx.doi.org/10.1071/CH15787>, -.

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## Functional Nanodiamonds by Light-Triggered Reactions

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Nanodiamonds (NDs) are accessible in large quantities and with low cost from the detonation of carbon containing explosives. To offer novel hybrid nanomaterials for e.g. biological applications, the nanodiamond surface needs to be selectively functionalized. Employing light to modify the nanodiamonds' surfaces has the advantage of accessing high spatial resolution, which is of potential interest for the development of ND patterned surfaces or ND based 3D structures. To introduce targeting functions and enhance the colloidal stability, the light-triggered modification in the absence of any catalyst and at ambient temperature is reported. For instance, *o*-methyl benzaldehyde groups – forming o-quinodimethanes (photoenols) upon UV irradiation – were attached to the surface of the NDs. The photoenol groups undergo Diels-Alder reactions with electron deficient double bonds, such as maleimides. For this purpose, various maleimide end group functional polymers (poly(styrene), poly(*N*-isopropylacrylamide) and poly(2-(2',3',4',6'-tetra-*O*-acetyl- $\beta$ -D-mannosyloxy)ethyl methacrylate)) were linked covalently to the NDs showing the modularity of this approach. The resulting polymer coated NDs were characterized by FT-IR, X-ray photoelectron spectroscopy, thermogravimetric analysis and dynamic light scattering. Additionally, the effect of polymer chain length on grafting efficiency as well as the lectin binding behavior of the glycopolymer functional NDs were investigated.

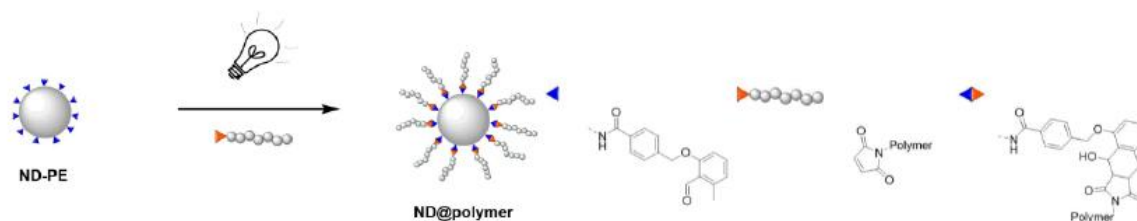


**Figure 1.** Example for the light-induced preparation of polymer coated NDs (ND@polymer), i.e. with poly(styrene), poly(*N*-isopropylacrylamide) or poly(2-(2',3',4',6'-tetra-*O*-acetyl- $\beta$ -D-mannosyloxy)ethyl methacrylate).

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Research interests: modular (light-induced) ligation chemistries, modern controlled/living radical polymerization techniques, surface modification protocols, interface design, advanced characterization methods

# Polymethacrylate copolymer films containing pendant nitroxides to protect against biofilm formation

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Biofilms are highly organised colonies of bacteria and other microorganisms on a surface. These colonies show a high level of structural complexity, which helps to protect the microorganisms from removal from the surface by both physical and chemical processes. Biofilms can cause major issues for industry, for example through the fouling of ship hulls or food processing plants. They are also implicated in the majority of nosocomial infections where they can show increased resistance against conventional antibiotic therapy.<sup>1,2</sup>

Our research group has shown that small molecule nitroxides (persistent and stable free radical species) are able to inhibit the formation of biofilms on a surface, as well as being able to disperse existing biofilms.<sup>3</sup> In this work we look to extend these molecules further by designing polymeric materials bearing pendant nitroxide groups, to allow the coating of a range of different materials with an antifouling polymer film.

This presentation will detail our efforts to synthesise and characterise films generated from polymethyl methacrylate (PMMA) copolymers containing pendant nitroxide units. These copolymers were synthesised using RAFT polymerisation, in order to control the concentration of nitroxide molecules in the polymers and ultimately the films. The physical characterisation and biological evaluation of the prepared films as anti-biofilm coatings will be presented and compared to the small molecule systems. Implications for the structure-function relationship of these nitroxides to inhibit biofilm formation will be discussed, and how this information is influencing design of the next generation of both small molecule drugs and polymeric materials.

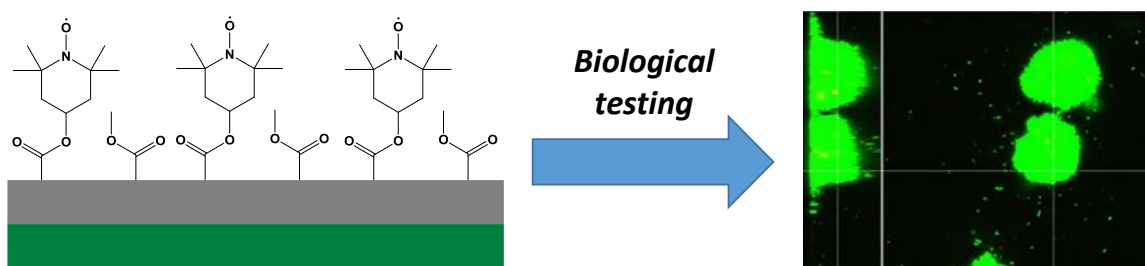


**Figure 1.** RAFT synthesised copolymers of MMA and a monomer nitroxide were spin coated onto silicon wafers to test their ability to inhibit biofilm formation.

(1) Davies, D. *Nat Rev Drug Discov* **2003**, 2, 114.

(2) O'Toole, G., *et al. Annual review of microbiology* **2000**, 54, 49.

(3) de la Fuente-Núñez, C., *et al. Antimicrobial Agents and Chemotherapy* **2013**, 57, 4877.

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# Preparation of Polymeric Particles Containing Reduced Graphene Oxide by Miniemulsion Polymerization

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Graphene is an atomically thin two-dimensional form of carbon that has various functionalities (electrical, thermal, and mechanical properties). However, when graphene is used as filler in polymeric materials to enhance their properties, it is difficult to disperse in both a solid matrix or in a solvent due to strong Van der Waals interactions between graphene layers, resulting in aggregation.

Recently, imidazolium-based ionic liquids and poly(ionic liquid)s (PIL) prepared by the polymerization of ionic liquid monomer have received attention as a potential dispersant of graphene due to  $\pi$ - $\pi$  interactions between imidazolium rings and graphene sheets. Graphene dispersions could be obtained in various solvents using PIL as dispersant because of the possibility to conveniently control the solubility of PIL by anion exchange.

Additionally, graphene oxide (GO) can be a precursor to composite materials containing reduced graphene oxide (rGO), as the functionality of rGO is similar to that of graphene.<sup>1</sup>

In previous work, we demonstrated the preparation of PIL particles containing rGO by miniemulsion polymerization using an ionic liquid monomer as rGO-dispersant.<sup>2</sup> However, from the point of view of industrial applications, that should be applicable to conventional monomers such as styrenic and methacrylate monomers. In the present work, the preparation of rGO dispersions stabilized by poly(ionic liquid) in a conventional vinyl monomer, and miniemulsion polymerization of the conventional monomer containing rGO was performed to yield composite nanoparticles.<sup>3</sup>

Prior to miniemulsion polymerization, the dispersibility of PIL/rGO in various monomers was investigated, however, PIL/rGO precipitated instantaneously in most common vinyl monomers. To improve the solubility of PIL in monomers, a statistical copolymer of ionic liquid monomer and methyl methacrylate (MMA) was prepared. rGO could be dispersed in ethyl methacrylate (EMA) using the copolymer by ultrasonication, however, polymeric nanoparticles containing rGO were not obtained by miniemulsion polymerization of EMA utilizing statistical copolymer as stabilizer, in which most rGO appeared to have precipitated. This suggested the rGO stability in monomer using this statistical copolymer was insufficient. On the other hand, polymeric nanoparticles containing rGO have been successfully prepared by miniemulsion polymerization of MMA utilizing a block copolymer of the ionic liquid monomer and MMA, which was prepared by RAFT (Fig. 1a). As shown in Figure 1b, the particle size distribution before acetone treatment comprised nanometer-sized particles ( $D_n = 160$  nm), consistent with the TEM imaging. After acetone treatment, significantly smaller nanoparticles ( $D_n = 27$  nm) were detected, consistent with single rGO sheets.

<sup>1</sup> T. Kim, H. Lee, J. Kim and K. S. Suh, *ACS Nano*, **2010**, 4, 1612-1618

<sup>2</sup> M. Tokuda, S. Thickett, H. Minami, P. B. Zetterlund, *Macromolecules*, **2016**, 49, 1222-1228

<sup>3</sup> M. Tokuda, M. Yamane, S. Thickett, H. Minami, P. B. Zetterlund, *Soft Matter*, **2016**, 12, 3955-3962

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| 2002       | Doctor's degree of engineering from Kobe University |
| 1996-2004  | Research Associate at Kobe University               |
| Since 2004 | Associate Professor at Kobe University              |

Research interests: Polymer colloids, Ionic liquid, Colloidal structure



# The Improvement of Dispersion of Surface Modified Hydroxyapatite in Polymer Matrix for Bone Substitute

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Poly(1,6-bis-(p-carboxyphenoxy hexane)-co-(sebacic anhydride)) (PANH), is a polyanhydride copolymer which has good biocompatibility, and degrades to non-toxic products with a predictable rate of degradation. Nano-sized hydroxyapatite (HAP) is a well-known biomaterial which has been applied in bone regeneration due to its osteoconductivity. However, nano-sized HAP has poor colloidal stability which leads to agglomeration when incorporated into polymeric composites. In this work we describe the surface grafting of poly( $\epsilon$ -caprolactone) to HAP (PCL-gHAP) to improve the interfacial adhesion and dispersion of HAP particles in a PANH matrix to form a composite material. The use of scanning electron microscopy-backscattered electron (SEM-BSE) detector and the combination of focused ion beam (FIB)/ transmission electron microscopy (TEM) provided a powerful approach for observing the dispersion of HAP particles in the polymer matrix. From TEM results (in Fig. 1) show that HAP agglomerate and PCL-gHAP disperse homogeneously in PANH matrix. This study shows that surface modification of HAP with PCL improved the homogeneity of the dispersion of HAP particles in the composites and affected the composite morphology during hydrolytic degradation. Composites with high HAP content displayed high compressive strength and a fast rate of degradation. The PCL-gHAP/PANH composites showed superior maintenance of mechanical properties compared with HAP/PANH composites during degradation. These results suggest that the newly developed PCL-gHAP/PANH composite materials may be a potential substrate for bone tissue engineering.

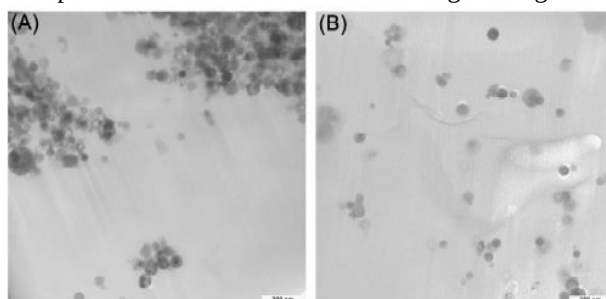


**Fig. 1** TEM results of particles in PANH matrix. (a) HAP, (b) PCL-g HAP

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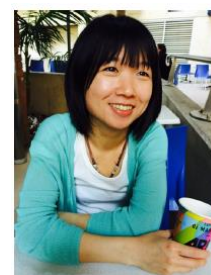
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2012 Master of Engineering, Chemical Engineering, National Tsing Hua University, Taiwan

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# Well-defined colloidal crystal films from the 2D self-assembly of core-shell semi-soft nanoparticles

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Core-shell hybrid nanoparticles are promising candidates for a wide range of applications such as waveguides, chemical and biological sensors and photonic crystals.<sup>1-4</sup> Here we report a facile and straightforward approach to fabricate 2D arrays of colloidal crystal film. A combination of an inorganic core comprising silica nanoparticles (SiNP) with a poly(methyl methacrylate) (PMMA) shell, obtained by reversible addition fragmentation chain transfer (RAFT) polymerization, was used to fabricate core shell nanoparticles. The surface-modified SiNP were used to mediate RAFT polymerization of methyl methacrylate (MMA) by the ‘grafting from’ approach to form core-shell hybrid particles with high polymer grafting density. The use of RAFT allowed the design of two families of particles of different grafting densities, in order to establish the effect of grafting density on the particle self-organisation. We find that high grafting density enables these hybrid particles to form two-dimensional (2D) arrays in a colloidal crystal film on a liquid substrate as evidenced by TEM, SEM, EDX, and AFM. Figure 1 shows a three-dimensional photograph of SiNP-PMMA monolayer film on a silicon wafer acquired by an Olympus laser microscope. In Figure 2 the Bragg’s reflection of a colloidal crystal film on a silicon wafer is captured by a digital camera.

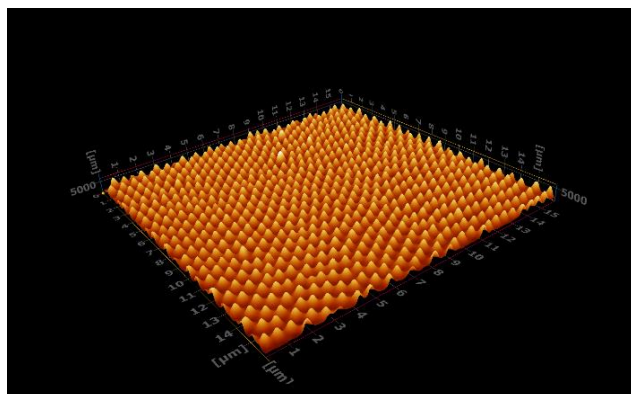


**Figure 1.** A 3D image of SiNP-PMMA monolayer film on silicon wafer acquired by an Olympus laser microscope.

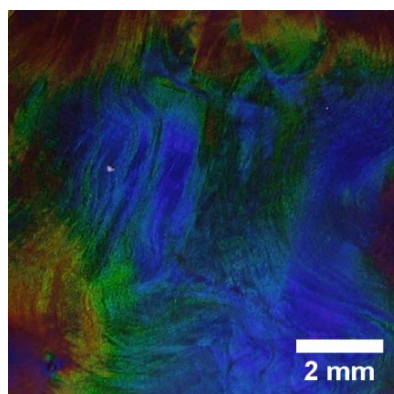


**Figure 2.** A digital photograph of colloidal crystal film of SiNP-PMMA1 with reflected pattern

## References:

- <sup>1</sup> Alam, M.M., J.-Y. Kim, and W.-G. Jung, *J. Colloid and Interface Sci*, 2011. 363(2): p. 446-449.
- <sup>2</sup> Nicolaou, C., et al., *Optics Express*, 2013. 21(25): p. 31698-31712.
- <sup>3</sup> Ye, X.Z. and L.M. Qi, *Science China-Chemistry*, 2014. 57(1): p. 58-69.
- <sup>4</sup> Ye, X. and L. Qi, *Nano Today*, 2011. 6(6): p. 608-631.

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2009-2012 R&D Chemist, SGE Analytical Science, VIC, Australia

Research Interests: RAFT polymerization, Magnetic nanoparticles and Ferrofluids  
Self-assembly, Colloidal Crystals, Core-Shell hybrid particles



## Recent Developments in the Field of Light-Controlled Radical Polymerizations

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The introduction of light as an external stimulus in controlled radical polymerizations has been extensively studied in recent years due to the supplementary advantages which can be brought, such as temporal and spatial controls. In this context, two mechanisms recently developed in our group will be presented.

The first one is a photocatalyzed ATRP involving a Cu-based complex with phenanthroline ligands.<sup>32,33,34</sup> The mechanism involves the excited state derived from the activator form of the complex, which is original for a photocatalyzed Cu-based ATRP (Figure 1). This catalytic cycle was completed by an additional regeneration of the activators mechanism, using an amine or a functional comonomer as the reducing agent.

The second one is based on a RAFT mechanism, considering that several thiocarbonylthio compounds were already commonly used as photoinitiators, before being considered as RAFT agents. We report the use of a RAFT agent designed to exploit this dual reactivity, leading to an efficient photoRAFT mechanism based on the reversible photolysis of the terminal functionality. Interestingly, this photodissociation of the chain-ends can be purposely enhanced so as to exploit polymers synthesized by RAFT as efficient and versatile macroinitiators.<sup>35</sup> Narrow polydispersities were reached in both cases either for methacrylates or acrylates using soft LEDs emitting either in the blue or green wavelengths range. Additionally, efficient temporal controls were achieved by simply switching on/off the light source. Light-triggered re-initiation and chain-extension were also successfully applied for the synthesis of block copolymers.

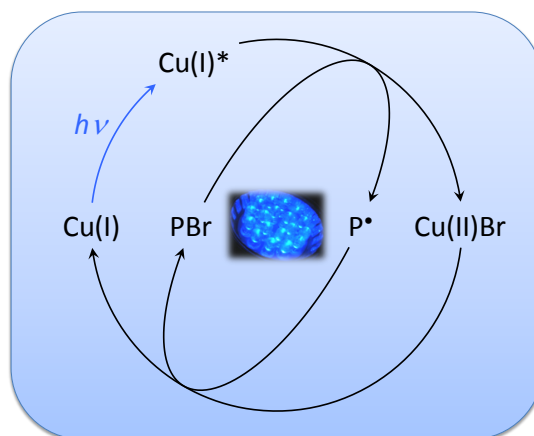


**Figure 1.** Photocatalyzed ATRP based on an oxidative quenching mechanism

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Research interests: photocontrolled radical polymerizations, macrophotoinitiators, self-healing polymers...



## A General Strategy to Precision Oligomers ( $\mathcal{D} = 1.0$ )

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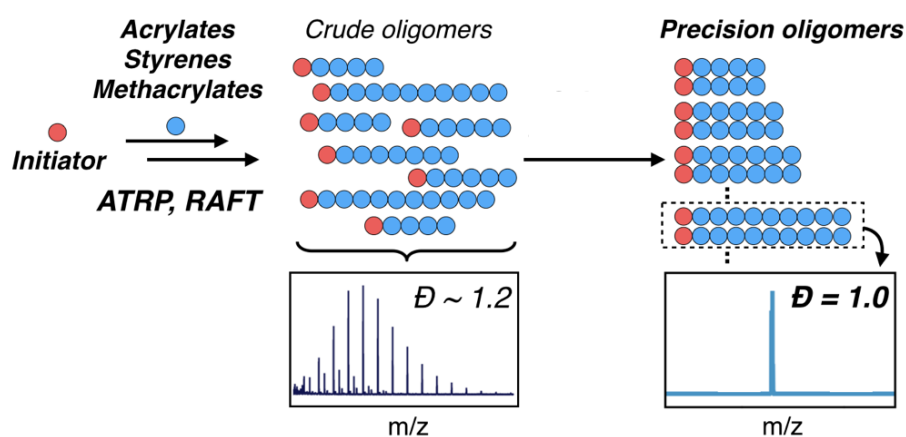
<sup>1</sup>Materials Research Laboratory and Departments of Materials, Chemistry and Biochemistry, University of California, Santa Barbara, California 93106, United States,

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Due to their statistical chain growth, all synthetic polymers are polydisperse materials containing a distribution of polymer chains of unequal length. While many strategies have been developed to prepare low dispersity polymers with diverse functionalities, they remain statistical mixtures of varying molecular weights. This presentation discloses a general strategy to obtain a library of near to truly monodisperse oligomers.

**Figure 1.** Diagram illustrating the separation of precise oligomers.



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Research interests: stereospecific polymerization, polymer architecture, and functional polymers for therapeutics and energy storage.

# Order in the Chaos of Plasmas: Retention of Complex Chemical Structures in Plasma Polymerisation

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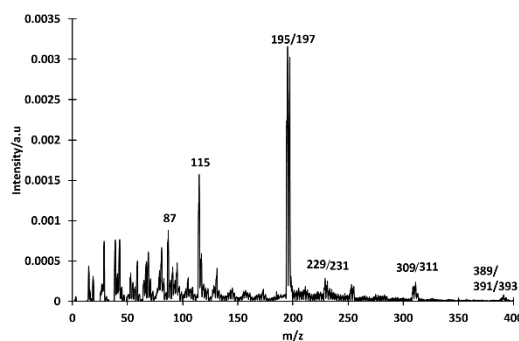
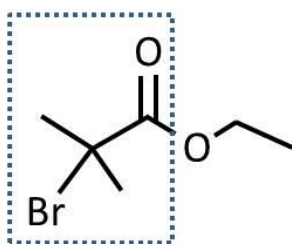
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Among polymerisation methods, plasma polymerisation has occupied a strange and unique position. On one hand, this method for producing thin polymeric coatings has found several large-scale industrial applications on the basis of advantages such as excellent uniformity and adhesion of coatings combined with ease of process control and reproducibility. On the other hand, plasma polymerisation has long had a black art connotation due to the fact that the chemical composition of plasma polymer films was not predictable and there were no scientifically rigorous ways of tailoring coating chemistries. The volatile organic precursor molecules used for plasma polymerisation are thought to undergo in the plasma gas phase a wide range of plasma-activated (i.e., non-thermodynamic) reactions, often apparently starting with homolytic scission of C-H bonds but other bonds can also be scissioned. The resultant fragment radicals, and in some cases also ions, then recombine in various ways to produce a coating, which typically displays several chemical structures not found in the “monomer”. This “scrambling” of chemical structures and the crosslinked nature of plasma polymers sets them apart from polymers produced by conventional methods.

Attempts to gain more specificity in plasma polymerisation have focused on empirical screening of plasma parameters such as input power combined with characterisation of deposited films mainly by XPS and IR. Clearly, for rational optimisation and to achieve predictive capability, one needs to analyse the deposition process itself. We have done this with ethyl- $\alpha$ -bromoisobutyrate (EBIB) as the process vapour, so as to deposit coatings bearing  $\alpha$ -bromoisobutyryl surface groups, capable of serving as ATRP initiation surfaces. Such motifs usually fragment easily in plasmas, but we have found that efficient retention is possible when plasma conditions are steered towards maximising the generation of the protonated molecular ion in the plasma, combined with reducing fragmentation and “soft landing” of hyperthermal ions. Using mass spectrometry of the plasma phase, ion flux, distribution of ion energies, and deposition rate measurements, we have been able to steer the plasma polymerisation of EBIB towards a dominant role of protonated molecular ions and hence high incorporation of  $\alpha$ -bromoisobutyryl groups. Analogous investigations with other volatile ester compounds have shown that they too are capable of being directed towards a dominant role of protonated molecular ions in the plasma gas phase. The resultant coatings are much less scrambled chemically than comparable plasma polymers, thus possessing a degree of chemical specificity previously unattainable in plasma polymerisation (though not matching conventional polymers).



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## Synthesis, evaluation and application of arsenical-linked polymer bioconjugates

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(In)organic arsenic exists as a dichotomous enigma between toxicity and therapy in (bio)chemistry and medicine. Despite being a well-known contaminant of base water supplies, it is an essential dietary element and has historically been employed to treat malignancies, most notably, and recently, in blood born malignancies such as acute promyelocytic leukaemia (APL).<sup>1</sup> Arsenic can exist in two biologically relevant oxidation states, trivalent arsenous acid (As(III)) and pentavalent arsenic acid (As(V)). The toxicity and therapeutic potential associated with arsenic is dominated by trivalent arsenic (As(III)) due to its high affinity for thiols which are present in variable concentrations in the intra- and extracellular milieu. In short peptides, larger functional proteins/enzymes and small molecule cofactors, thiols can exist either as free thiols, or as intra- or intermolecular disulfides. It has been postulated that the affinity of As(III) for thiols is enhanced for chelating vicinal or neighbours-through-space (proximal) dithiols,<sup>2</sup> such as those presented by naturally occurring disulfide bonds.

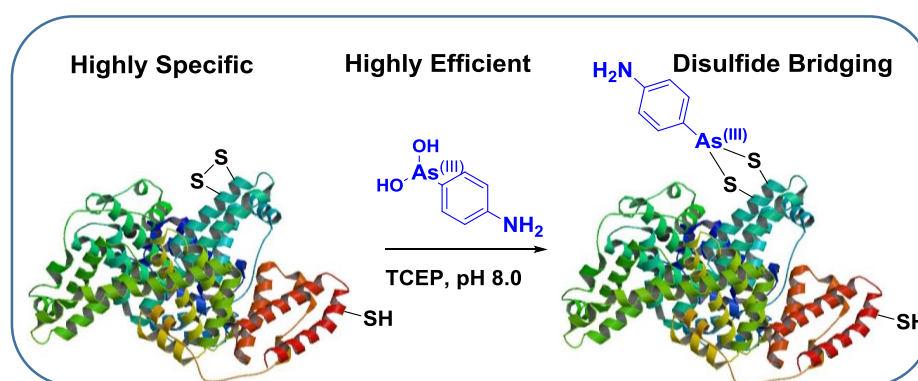


**Figure 1.** Highly specific conjugation of trivalent *p*-aminoarsenous acid to reduced BSA.

Conceptually this has been investigated using trivalent *p*-aminoarsenous acid and reduced BSA as model arsenical and protein respectively (**Fig 1**).<sup>3</sup> The enhanced specificity of As(III) for disulfides was exemplified upon comparison with established thiol-reactive functional groups such as maleimide and dibromomaleimide. Polymeric arsenicals have been synthesised by aqueous single electron transfer living radical polymerisation (AqSET) and reversible addition-fragmentation chain transfer polymerisation (RAFT) using an As(V)-functional initiator and chain-transfer agent respectively. Reduction of the As(V)-functional chain end using glutathione, post polymerisation, furnishes As(III)-functional polymers capable of specifically rebridging disulfide bonds in proteins and peptides. Conjugation to salmon calcitonin (sCT) proceeds rapidly and stoichiometrically to yield stable sCT-polymer conjugates. Upon addition of stronger chelating dithiols (e.g. ethanedithiol, reduced lipoic acid) the sCT is released from the conjugate. This chemistry has been applied to the synthesis of peptide-polymer conjugates that target the somatostatin receptor, a GPCR which is over expressed in neuroendocrine tumours. Conjugates of Somatostatin and analogues Octreotide and Lanreotide have been prepared and the effects of conjugation site and polymer composition on receptor interactions and down-stream cell-signalling events have been investigated.

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# Expanding The Scope of Visible Light Mediated Polymerization-Induced Self-Assembly

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Traditionally, the self-assembly of polymers into polymeric nanoparticles has only been accomplished in dilute solution making large-scale production relatively inefficient. Progress in the field has led to research into processes capable of generating materials at high concentrations and with relatively good control over the morphology. Polymerization-Induced Self-Assembly (PISA), a process whereby a living polymerization technique is used to induce self-assembly in situ allows for the production of nanomaterials at relatively high solids content. Judicious selection of PISA parameters can also lead to the formation of more complex micelle morphologies, such as cylindrical micelles and vesicles, which may be desirable for a number of applications. For example, several recent publications have demonstrated a strong dependence of nanoparticle morphology on cell uptake and cytotoxicity.<sup>1-3</sup> However, significant challenges still exist in synthesising high purity polymeric nanoparticles with well-defined morphologies particularly intermediate morphologies such as worm-like micelles.

Recently, we reported for the first time the use of a visible light-mediated polymerization technique known as Photoinduced Electron/Energy Transfer RAFT (PET-RAFT) to drive the PISA process.<sup>4</sup> In this system, activation of a PET-RAFT process by the ruthenium-based photocatalyst, Ru(bpy)<sub>3</sub>Cl<sub>2</sub> could be used to form different nanoparticle morphologies under blue light and at room temperature. Interestingly, the formation of worm-like micelles could be monitored in situ by observing the increase in reaction viscosity which is attributed to the presence of multiple inter-worm contacts. Furthermore, we recently extended upon this work by conducting a PISA process mediated by blue or green light in the absence of external catalysts or initiators.<sup>5</sup> In this process, a poly(oligo(ethylene glycol) methyl ether methacrylate) (POEGMA) macromolecular RAFT (macroRAFT) agent acts simultaneously as a radical photoinitiator, chain transfer agent and particle stabilizer yielding nanoparticles of different morphologies. In particular, the formation of worm-like micelles was readily observable even when using POEGMA macroCTA's of different lengths due to gelation of the polymerization mixture in situ. Such a result appears to be a simple method by which to more reproducibly isolate the worm-like morphology which should facilitate further studies particularly in the biological arena. Research is currently underway to further expand the scope of "Photo PISA" by conducting such polymerizations under a range of wavelengths for a variety of applications.

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## Self-assembled sericin/chitosan scaffold: physical and biocompatible properties

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This research aims to utilize sericin, which is the waste from boiling silk cocoon, for mixing with chitosan in order to produce the self-assembled scaffold. The optimum conditions on morphological and physical properties of scaffold are studied. The suitable method for forming the scaffold between sericin and chitosan at 1:1 stoichiometry is presented. Briefly, the self-assembled sericin/chitosan scaffold is prepared in a cylindrical glass mold and then treated in a 1:1 (v/v) methanol / 1N NaOH solution to change the secondary structure of protein sericin and also to adjust pH to neutral. Finally, the scaffold is incubated in phosphate buffer solution (PBS). The production parameters of this scaffold are the stoichiometry between sericin and chitosan, and the molecular weight of chitosan. The results show that sericin increases the porosity of scaffold while chitosan increases the rigidity. The self-assembled scaffold of sericin and chitosan is non-toxic to the skin cell and the cells can adhere and spread very well on it. For the effect of molecular weight of chitosan (MW15000 and MW100000 g/mole), the scaffold made from lower MW chitosan provides less viscosity, which shows better homogeneous distribution with sericin. Based on this research, sericin, which is a silk waste from textile industry, can be utilized to produce the self-assembled scaffold by mixing with chitosan in order to increase the porosity of scaffold. This type of scaffold is favourable to the adhesion of fibroblast cell and non-toxicity (Figure 1).

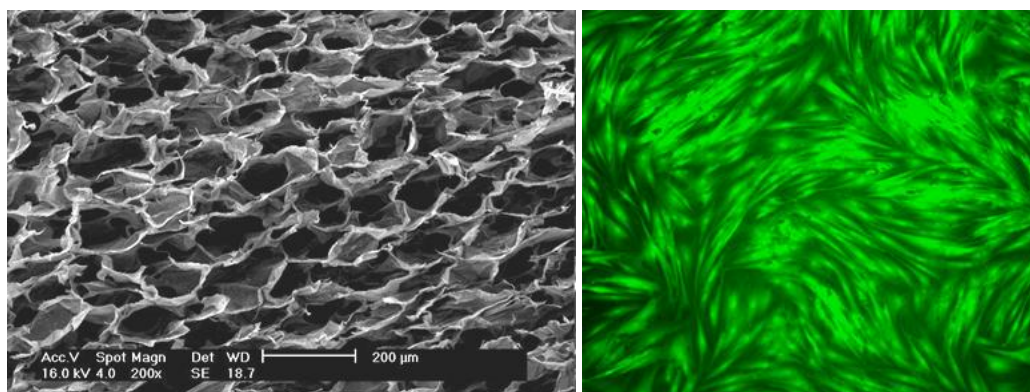


**Figure 1.** Self-assembled sericin/chitosan scaffold at 1:1 stoichiometry: (left) SEM picture at X200 and (right) positive result of cytotoxicity test with fibroblast cell in the media after 8 days.

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# Non-Spherical Polymersomes: Unusual Shapes Obtained Through Fine-Tuning of $\pi$ - $\pi$ Interactions in Perylene-Containing Diblock Terpolymers

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The complex biological processes that we see all around us in Nature are, by and large, performed in compartmentalised spaces, i.e. within membrane-bound organelles, endosomes and the outer cellular boundary, the cell membrane which is formed by small amphiphilic molecules such as phospholipids. Amphiphilic block copolymers represent an interesting class of building blocks that can self-assemble to form polymersomes which are cell-resembling structures that consist of an aqueous interior/core surrounded by a hydrophobic bilayer membrane.<sup>1</sup> Favoured for their structural analogy to cell membranes and their superior stability/versatility over liposomes (their lipid counterparts), polymersomes have been widely explored in the field of biomimicry.<sup>2,3</sup> Although commonly used as a basis for preparing synthetic cells, conventional polymersomes suffer from a lack of shape control and are usually found to be spherical in nature. As a result, such polymersome-based synthetic cells fail to mimic (and exemplify the structural importance of) the non-spherical morphology of most existing cells and organelles.<sup>4,5</sup>

Recently, we have managed to fabricate polymersomes with uncommon shapes, using polymers that have perylenes (a class of polycyclic aromatic hydrocarbons) attached via the side chains. By changing the solvent quality used in the fabrication/self-assembly process (**Fig 1A**), we could obtain polymersomes that were either ellipsoidal- (with different sizes and aspect ratios) or tubular-shaped (**Fig 1B-F** for TEM images). Interestingly, spectroscopic studies revealed that these differently shaped polymersomes are stabilised by different degrees of  $\pi$ - $\pi$  interactions between perylene moieties within the polymer. Different membrane thicknesses revealed by cryo-TEM led us to hypothesise that the complex shapes of these polymersomes arise from the different supramolecular arrangement of perylene moieties within the bilayer membranes. The self-assembly/formation pathways of these polymersomes will be discussed in detail.

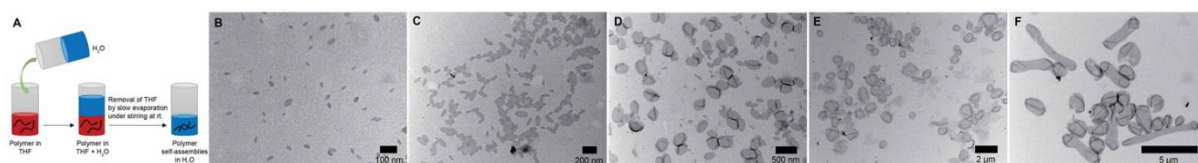


**Figure 1.** Cartoon representation of the polymersome fabrication process and TEM images of (B) small ellipsoidal micelles, (C) medium ellipsoidal polymersomes, (D) large ellipsoidal polymersomes, (E) giant ellipsoidal polymersomes and (F) tubular polymersomes.

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# Directed Self-assembly of Block Copolymer Nanostructures with Tuneable Morphologies

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Manipulation of block copolymer (BCP) nanostructures holds great academic interest as the functionality and performance of BCP assemblies crucially depend on their overall shape and morphology<sup>1</sup>. Therefore, facile and versatile strategies to access novel structures of BCPs are desired. In aqueous solution self-assembly, the control of the interface between BCP colloidal and the surrounding medium has been proved to be a powerful tool to access BCP nanostructures with tuneable features.<sup>2</sup> We developed a facile strategy to achieve morphological evolution of BCP nanoparticles using a mixture of surfactants to tune the interface. The influence of block weights was also investigated and a non-conventional structure of a prolate spheroid nanoparticle with a unique phase separated interior morphology can be synthesised.<sup>3</sup> The morphological evolution and their corresponding conditions are summarised in **Figure 1**.

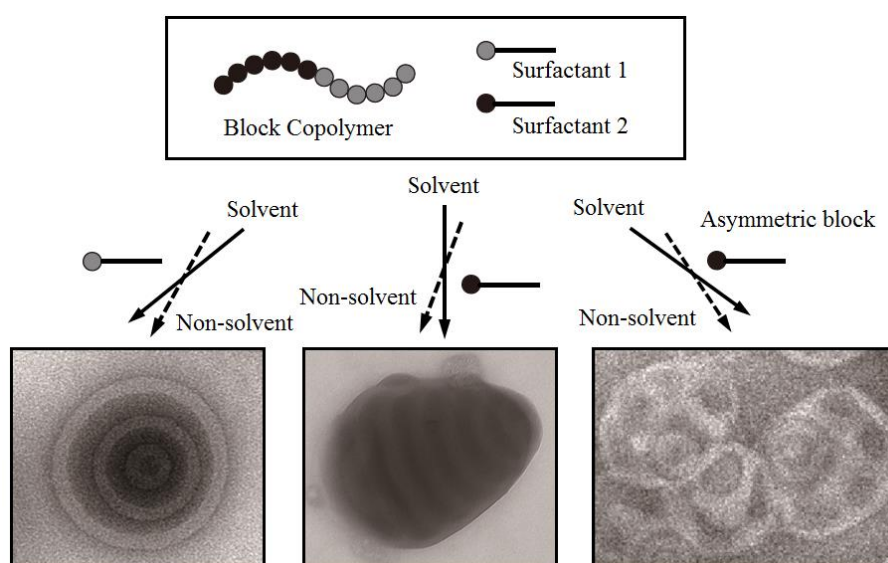


**Figure 1.** Morphologies of diblock copolymers can be tuned using functional surfactants which have preferential wetting to the respective block (indicated by the colour of surfactants and blocks). For symmetric block copolymers, it forms either onion-like or axially stacked particles depending on the composition of surfactant mixtures. As for asymmetric copolymers, it forms a nonconventional patchy particle when one of the surfactant is used.

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# Bioinspired Materials Bridging the Length Scales

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Self-assemblies bridging the length scales to allow structural hierarchies are amply used in Nature towards achieving materials with functional properties. Here selected examples are given by combining colloidal level structural units and polymeric units. Cellulosic materials have gained revived interest, due to their wide availability as sustainable source for polymers. Therein nanocelluloses as colloidal level fibrillar objects of high mechanical properties hold a particular promise, and they encourage to identify also novel high tech applications beyond those traditionally considered for cellulose. Recently Toivonen et al showed that cellulose nanofibers (CNF) allow transparent aerogel materials with high strength where a method was found which suppressed the need for the classically required supercritical drying or freeze-drying, paving ways towards scalability.<sup>1</sup> The films could be coated by carbon nanotubes to allow conducting transparent aerogel films, which could be a starting point for future devices. A typical problem related to nanocellulose has been moisture sensitivity. Toivonen et al<sup>2</sup> and Mertaniemi et al<sup>3</sup> also prepared nanocellulose films and fibers with increased strength also under wet conditions, which allows new application potential. This opened even possibilities for nanocellulose-based surgery threads with promoted cell adhesion.<sup>3</sup> On the other hand, humidity can also be harnessed to allow actuation, even without incorporation of the common bilayer architecture.<sup>4</sup> Biological inorganic/organic hybrids can allow uncommon combination of high mechanical properties, where a prototypical example is nacre. Previously an evaporation-induced self-assembly method was indentified by Walther et al to prepare nacre-mimetic films with high strength and stiffness.<sup>5</sup> Morits and Verho et al have recently found a self-assembly route towards bulk materials mimicking nacre, showing deflected crack propagation indicating toughness and where the fracture toughness starts to approach to that of nacre.<sup>6</sup> Finally we discuss biomimetic optical properties. Structural colours are inherent in several animals, plants, and organisms, involving chirality of the overall structure. Instead of the macroscopic chiral assembly, we investigate photonic properties of individualized nanoscale objects. Majoinen et al showed that the inherent chiral twisting of individualized cellulose nanocrystals (CNC) in aqueous medium can be harnessed to show chiral plasmonic functions upon binding gold nanoparticles.<sup>7</sup> We expect that the concept is general, suggesting to use different chiral and helical templates for plasmonic materials.

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## Anti-fouling polymer films from recombinant biopolymers

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Anti-fouling polymer films are used to protect surfaces from protein adsorption in the context of medical devices, biosensors, nanoparticles, and other materials.<sup>1</sup> The most commonly employed polymer to date is poly(ethylene glycol).<sup>2</sup> More recently, zwitterionic polymers (e.g. those based on carboxybetaine, sulfobetaine) have emerged, and we previously showed that they possess a similar anti-fouling effect as PEG, with the added advantage of stabilising immobilised antibodies for high efficiency antigen capture.<sup>3</sup> Immobilised biomolecules often include proteins, peptides, or antibodies, all of which require a stable conformation to maintain their biological activity. Thin films that comprise “motifs” that promote surface interaction, anti-fouling, and bioconjugation, in a sequence-controlled manner, are therefore highly desirable.<sup>4</sup>

Protein engineering through recombinant approaches allows the synthesis of a DNA, protein or peptide sequence with monomer-level precision. We have recently designed several simple peptides that contain surface-reactive, anti-fouling, and bioconjugation motifs in the one sequence. Herein we will describe the results of a screening process to express and purify peptides in high yield, along with results on surface attachment, anti-fouling, antibody immobilization, and specific antigen binding and detection, using previously developed polymer-coated surfaces (PEG, sulfobetaine-methacrylate)<sup>3</sup> as controls.

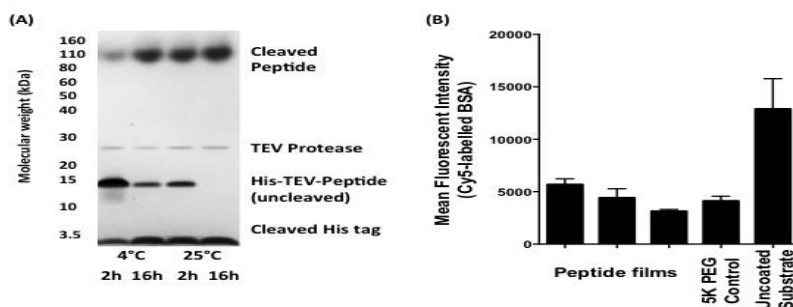


**Figure 1.** Expression and anti-fouling behaviour of recombinant peptides. (A) Coomassie-stained protein gel showing expression of peptide and cleavage from column using TEV protease tag under different conditions. (B) Fluorescence scan of surfaces coated with different polymer or peptide films after incubation in Cy5-labelled BSA solution in PBS (1 $\mu$ M).

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# Tuning Responsiveness of Protein Polymer via Co-assembly Approach

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The emergence of genetic engineering of synthetic polypeptides has recently enabled to synthesize protein-based polymers artificially. Rec1-resilin is a recombinant form of resilin that is derived from the first exon of the *Drosophila melanogaster* CG15920 gene. It exhibits responsiveness to multiple stimuli, including temperature, pH, and light, which allows for precise control of the self-assembly behaviour.<sup>1-7</sup> Rec1-resilin exhibits dual phase transition behaviour (DPB), with both a lower critical solution temperature (LCST) and an upper critical solution temperature (UCST). Below the UCST (~6 °C) and above the LCST (~70 °C), the protein undergoes phase separation in solution, whereas in between these temperatures the protein is soluble with a hydrodynamic diameter of approximately 11 nm<sup>2,3</sup>. However, both the UCST and LCST are outside the physiological temperature range, which limits the use of this unique behaviour for biomedical applications. The need to lower the LCST of Rec1-resilin has stimulated our interest into investigating its co-assembly with other rigid materials. The ability to tune the thermo-responsiveness of recombinant resilin protein, Rec1-resilin through a facile co-assembly system was investigated in this study. The effects of change in conformation and morphology with time and the responsive behaviour of Rec1-resilin in solution were studied in response to addition of a rigid model polypeptide, or other biopolymer using scattering, microscopy and circular dichroism. The results from this study indicate that the phase transition behaviour of Rec1-resilin can be tuned to more physiologically relevant temperatures (Fig 1) through co-assembly with rigid biomolecules. Time and temperature were found to be critical parameters in controlling the co-assembly behaviour of Rec1-resilin in both the model polypeptide and more complex polymer systems. Such unique properties are useful for a wide range of applications, including drug delivery and soft tissue engineering applications.

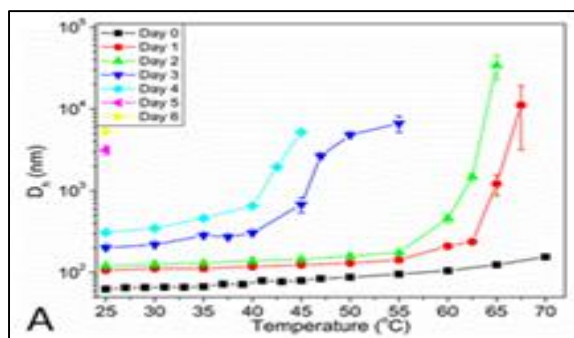


**Figure 1.** DLS data showing the shift in LCST of the Rec1-resilin coassembled system with time (0 to 6 days)<sup>7</sup>

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## Nitrile oxide-Norbornene Addition as Bio-orthogonal Crosslinking Chemistry for Preparation of 3D Cell Culture Matrices

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Hydrogels prepared from naturally derived gelatin can provide a suitable environment for cell attachment and growth, making them favourable materials in tissue engineering. However, physically crosslinked gelatin hydrogels are not stable under physiological conditions while chemical crosslinking of gelatin by radical polymerization may be harmful to cells. This presentation outlines the strategy for facile preparation of injectable gelatin-based hydrogel which can encapsulate living cells in 3D. We utilized the click cycloaddition between a norbornene and nitrile oxide for crosslinking of the polymer precursors under physiological conditions.<sup>1</sup> The hydrogels properties including mechanical strength, swelling and degradation, can be tuned by varying either the ratio of the reacting groups or the total concentration of the polymer precursors. Murine embryonic fibroblastic cells cultured in soft gels with the elastic modulus of approximately 150 Pa demonstrated high cell viability as well as similar phenotypic profiles (PDGFR $\alpha$  and MTS15) to cells culture in commercial Matrigel, which is typically used as 3D cell culture in biology lab, over 5 days.<sup>2</sup>

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<sup>1</sup>Truong, V. X.; Zhao, K.; Simon G. P.; Forsythe, J. S., *Macro. Rapid Comm.*, 2015, DOI: 10.1002/marc.201500314.

<sup>2</sup>Truong, V. X.; Hun M. L.; Li, F; Chidgey, A. P.; Forsythe, J. S., *Biomater. Sci.*, 2016, DOI: 10.1039/C6BM00254D

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**Consensus** GGRFSDSYGAPGGGN  
MHHHHHHFEP EVNISTLP

FSDSYGAPQSGP  
GGRFSDSYGAPGGGN  
GGRFSDSYGAPQGGQGGQGGGVA  
FSDSYGAPGGNGN  
GGRFSSYGAPGGGN  
GGRFSDTYGAPGGN  
GGRFSDTYGAPGGGNGN  
GGRFSSSYGAPQGGNGN  
GGRFSSSYGAPGGN  
GGRFSDTYGAPGGN  
GGRFSSSYGAPGGN  
GGRFSDTYGAPGGNGNS  
GGRFSSSYGAPQGGG  
GGRFSDSYGAPQGNQ  
FSDSYGAPSGNGN  
GGRFSSSYGAPSGP  
GGRFSDSYGSPASG

Scheme-1: Structural consensus and alignment of amino acid repeat sequence in rec1-resilin. Single letter code for amino acid residues has been used.

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## Alginate-based materials

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Alginate is an anionic co-polymer composed of (1, 4)-linked  $\beta$ -D-mannuronate (M) and  $\alpha$ -L-guluronate (G) residues and is sourced from nature (eg. seaweed).<sup>1</sup> This polymer has been comprehensively studied for use in various applications in biomaterials and food science because of its reported biocompatibility, non-toxicity and mild gelation property.<sup>1</sup>

The work presented will cover various aspects of developing alginate-based materials including evaluation of the bio-distribution of alginate (Fig. 1A), synthesis of sulfated alginate, encapsulation of bovine lactoferrin in nano-aggregates and micro-gel particles,<sup>2</sup> as well as fabrication and characterisation of alginate hydrogels (Fig. 1B&C)<sup>3</sup> including composite photo-polymerised hydrogels.

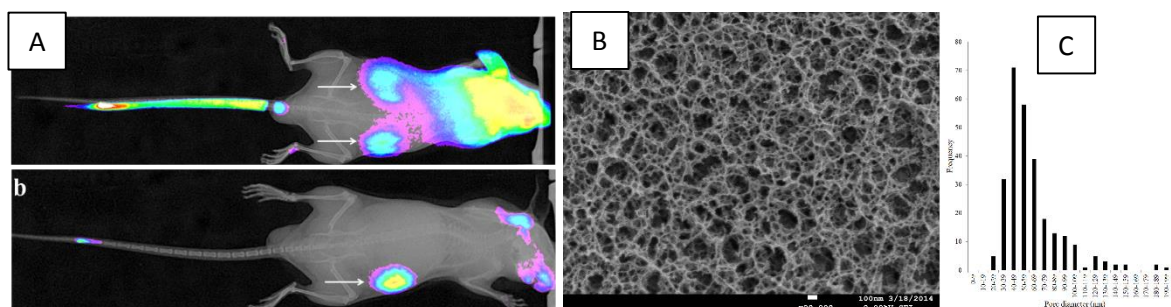


**Figure 1.** A: After 2 days of sample administration, the presence of circulating alginate *in vivo* could still be observed; B: Cryo-SEM image of ionically crosslinked alginate hydrogel; C: Pore size distribution obtained from image in B.

1. KY Lee, DJ Mooney (2012) *Progress in Polymer Science* 37, 106
2. H Bokkhim, N Bansal, L Grøndahl, B Bhandari (2016) *Food Hydrocolloids* 52, 231
3. R Aston, K Sewell, T Klein, G Lawrie, L Grøndahl (2016) *European Polymer Journal* 82, 1

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# Arylamine-Containing Block Copolymers Responsive to Nitric Oxide

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Nitric oxide (NO) is a lipophilic, diatomic species with multiple signalling functions in human physiology.<sup>36</sup> For instance, nitric oxide is involved in pain perception, sleep control and regulation, vasodilation, and mucus production. Additionally, misregulation of NO is associated with numerous pathologies, such as preeclampsia-related hypertension and Prinzmetal's angina. Elevated production of NO is also related to processes in transplant rejection and septic shock. As a result, materials capable of modulating the physiological levels of NO may be of considerable benefit.<sup>2</sup>

Inspired by the chemistry employed in fluorogenic probes for nitric oxide sensing,<sup>3</sup> we have recently been exploring polymers which include an *o*-phenylenediamine motif as NO-responsive materials.<sup>4-5</sup> In view of the mechanism through which *o*-phenylenediamine moieties react in the presence of NO,<sup>6</sup> we hypothesized that materials with a *p*-phenylenediamine functionality may also be suitable candidates for application as NO responsive materials. This presentation will describe our recent results demonstrating that *p*-phenylenediamine functional monomers (and polymers synthesized therefrom) react in the presence of NO to yield materials with a phenyldiazohydroxide moiety (see Figure 1). Moreover, we will also demonstrate that the product formed has a unique propensity to decompose under UV irradiation to yield phenol substituted polymers, and discuss the use of these materials in triggering changes to the LCST of a thermoresponsive diblock copolymer.

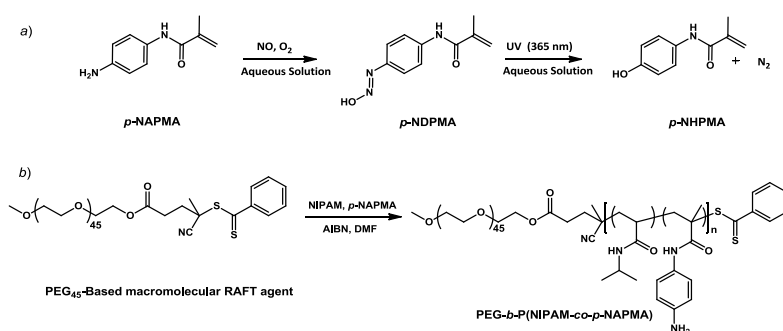


**Figure 1.** (a) Reaction of *p*-NAPMA monomer in the presence of nitric oxide, and subsequent loss of the diazohydroxide moiety under UV irradiation. (b) Synthesis of arylamine-containing block copolymers using RAFT polymerization.

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## Amphiphilic Polymer Conetworks: Structural Evolution During Aqueous Gelation

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Amphiphilic polymer conetworks (APCN) are important materials attracting increasing attention due to their properties and potential applications.<sup>1</sup> These materials comprise hydrophilic and hydrophobic segments, covalently interconnected to a network. When placed in water, the hydrophobic segments rearrange themselves in order to minimize contact with the aqueous environment, leading to phase separation on the nanoscale. This internal organization makes APCNs ideal materials for use in antifouling coatings, modern soft contact lenses, matrices for phase transfer reactions with applications in bio- and organocatalysis, and the fabrication of gas and optical sensors.

Due to the presence of the cross-links, and the inhomogeneity of the polymer chains, the morphologies exhibited by APCNs are not perfect, displaying only short-range order and blurred interfaces. To improve the order in the morphologies of nanophase separated APCNs, we optimized their synthesis by employing block copolymer building blocks and relatively low loadings of cross-linker. However, a non-ideality in this system was the broad distribution in the core functionality. Despite this defect, the system internally self-assembled, resulting in a structure comprising lamellae with relatively long-range order as confirmed via both scattering and microscopy techniques.<sup>2</sup>

This lecture will present a small-angle neutron scattering (SANS) study to kinetically follow the formation of APCNs in deuterium oxide, starting from concentrated solutions of well-defined polymeric building blocks. These building blocks are reactive and lead to APCN formation within 1–2 hours, a time scale that allows monitoring by SANS. Structural transitions during APCN formation were observed and will be discussed in relation to the composition, concentration and architecture of the network building blocks, and the reaction conditions.

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<sup>2</sup> E. J. Kepola, E. Loizou, C. S. Patrickios, E. Leontidis, C. Voutouri, T. Stylianopoulos, R. Schweins, M. Gradzielski, C. Krumm, J. C. Tiller, M. Kushnir, C. Wesdemiotis, *ACS Macro Lett.* **2015**, 4, 1163-1168.

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## 3D-Printing of Functional Materials

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Three-dimensional printing (3DP) is a world leading additive manufacturing technology, which attracts a lot of attention in the fields of tissue engineering<sup>1</sup>, pharmaceuticals<sup>2</sup> and electronics<sup>3</sup>. Various 3DP technologies are available, among which fused deposition modeling (FDM) is the most popular one due to its affordable cost and simple operation.<sup>2,4</sup> In FDM, a polymeric filament is introduced to a heated nozzle and melts to allow a layer-by-layer fabrication of a 3D object<sup>5</sup>. Traditionally, polymers applied in FDM are thermoplastics, such as acrylonitrile-butadiene-styrene (ABS) and polylactide (PLA),<sup>4</sup> while there is still a limited number of “smart” materials with versatile functionalities<sup>6</sup>. Recently, stimuli-responsive materials gain momentum as prospective candidates for 3DP.<sup>7</sup> Their responsiveness to external stimuli, such as light or temperature, is associated with dynamic molecular rearrangements, which make these polymers applicable for fabrication of smart devices with advanced architectures.<sup>7,8</sup> Despite the operational simplicity of FDM, production of new, 3D-printable smart materials remains a great challenge. To comply with FDM, materials are required to meet strict processing requirements, such as thermal stability, well-defined flow properties, strength, structural integrity and low surface roughness.<sup>9</sup>

In this work, we are aiming to address these challenges and to develop novel, 3D-printable materials with advanced chemical functionalities for applications in FDM. We synthesize and characterize functional polymers, apply rheological tools to examine their dynamic flow properties. We characterize the mechanical properties and the surface morphology of extruded filaments and aim to determine a set of optimal conditions for their extrusion and 3DP. We demonstrate the applicability and benefits of these 3D-printable materials and show their wide range of applications.

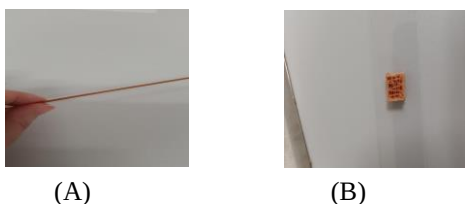


**Figure 1.** 3D-printable, functional filament and object: (A) A 3D-printable filament made of a functional polymer and produced by hot-melt extrusion. (B) A 3D-printed object: a macroporous, functional membrane.

<sup>1</sup> I. Zein, D.W. Hutmacher, K. C. Tan, S.H. Teoh, *Biomaterials* **2002**, 23,1169-1185

<sup>2</sup> A. Goyanes, J. Wang, A. Buanz, R. Martínez-Pacheco, R. Telford, S. Gaisford, A. W. Basit, *Mol. Pharma.* **2015**, 12, 4077-4084

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<sup>6</sup> W. Oropallo, L. A. Piegl, *Eng. Comput.* **2016**, 32, 135-148

<sup>7</sup> Z. X. Khoo, J. E. M. Teoh, Y. Liu, C. K. Chua, S. Yang, J. An, K. F. Leong, W. Y. Yeong, *Virtual Phys. Prototyp.* **2015**, 10, 103-122

<sup>8</sup> G. I. Peterson, M. B. Larsen, M. A. Ganter, D. W. Storti, A. J. Boydston, *ACS Appl. Mater. Interfaces* **2015**, 7, 577-583

<sup>9</sup> Y. Yang, Y. Chen, Y. Wei, Y. Li, *Int. J. Adv. Manuf. Technol.* **2015**, 1-7.

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# Highly Ordered Nanoporous Films from Supramolecular Diblock Copolymers with H-Bonding Junctions

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Herein, we report on the design of H-bonding precursors that combine complementary associative groups with exceptional binding affinities and thiocarbonylthio moieties enabling RAFT polymerization. Well defined PS and PMMA supramolecular polymers with molecular weights up to 30 kg.mol<sup>-1</sup> are synthesized and shown to form highly stable supramolecular diblock copolymers (BCPs) when mixed, in non-polar solvents or in the bulk. Hierarchical self-assembly of such supramolecular BCPs by thermal annealing affords morphologies with excellent lateral order, comparable to features expected from covalent BCP analogues. Washing of the resulting materials with protic solvents affords nanoporous materials without resorting to crosslinking or invasive chemical degradations (Figure 1).<sup>1</sup>

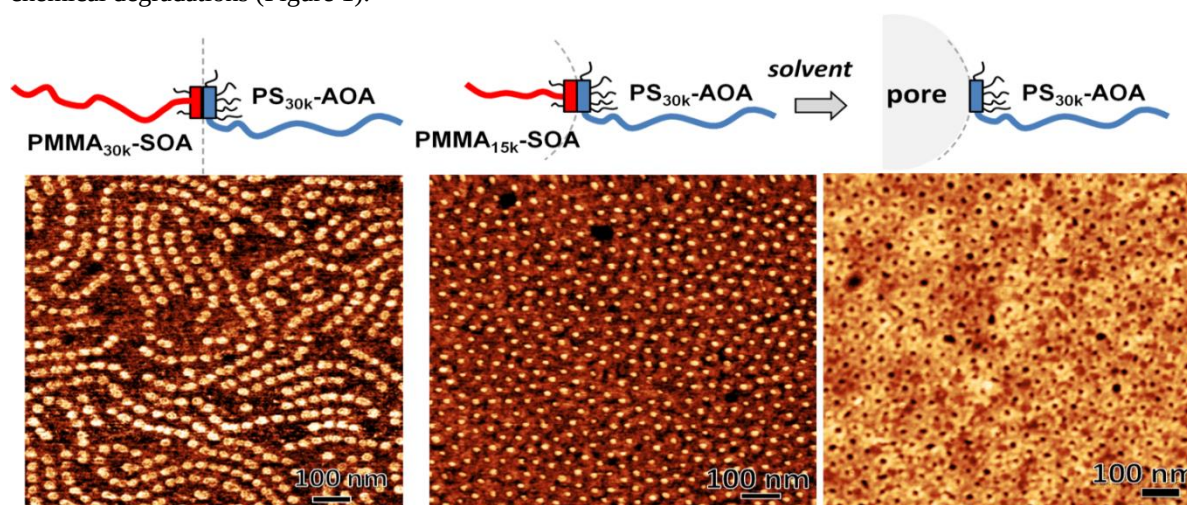


Figure 1. Left and middle: AFM phase images of **PS<sub>30k</sub>-supra-PMMA<sub>30k</sub>** and **PS<sub>30k</sub>-supra-PMMA<sub>15k</sub>** thin films, Right: AFM height image of **PS<sub>30k</sub>-supra-PMMA<sub>15k</sub>** after washing. Top: General sketch of the packing at the interface.

1. Montarnal, D.; Delbosc, N. Chamignon, C.; Virolleaud, M-A.; Luo, Y.; Hawker, C. J.; Drockenmuller, E.; Bernard J. *Angew. Chem. Int. Ed.* **54**, 11117-11121, 2015.

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# Emulsion-Templated Porous Polymer Scaffolds for 3D *In-Vitro* Cell Culture and Tissue Engineering

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Highly porous polymeric materials can be created by the emulsion templating technique, whereby a high internal phase emulsion (HIPE) is created in which the continuous (non-droplet) phase contains polymerisable monomer(s). Polymerisation then results in a well-defined macroporous polymer (foam), known as a polyHIPE. The nature of the formation of polyHIPEs creates a highly porous, interconnected monolith structure, the architecture of which can be tightly controlled. These polyHIPE materials can serve as scaffolds for the culture of cells and, ultimately, tissues in 3D, allowing cells to mimic their native structure and function *in-vitro*. Applications of these polymeric scaffolds are foreseen in 3D cell culture, tissue engineering and regenerative medicine. However, one potential limitation of these polyHIPE materials as 3D cell scaffolds is surface chemistry. Cells *in-vivo* are typically surrounded by a complex extracellular matrix that contributes to cell anchorage and function. For example, it is well known that hepatocytes (liver cells) possess a galactose-binding cell receptor (asialoglycoprotein). The presence of galactose residues on the surface of polyHIPE scaffolds could improve hepatocyte adhesion and function while growing on the scaffold, leading to enhanced performance in liver tissue engineering applications.

This presentation will describe the surface functionalisation of polyHIPE materials by incorporating pentafluorophenyl acrylate (PFPA) into the initial styrene emulsion. Galactose-functionalised scaffolds are formed via displacement of the labile PFPA group by post-polymerisation treatment with 2'-aminoethyl- $\beta$ -D-galactopyranoside (**Fig. 1**). Experiments with primary rat hepatocytes show that the galactose-functionalised scaffolds offer enhanced hepatocyte adhesion and functionality compared to the unfunctionalised/glucose-functionalised scaffolds.<sup>1</sup>

The surface functionalisation of the recently developed biodegradable polyHIPE scaffolds which are made by photopolymerisation of HIPEs containing multifunctional thiols and acrylates will also be presented. The resulting thiol-ene polyHIPE materials have been found to contain residual thiols which are reactive towards a range of (meth)acrylate, maleimide and allyl functionalities providing a facile route to functionalisation with biomolecules.<sup>2</sup> Initial cell culture experiments with human endometrial stem cells will furthermore be shown. Lithographic 3D printing approaches to create scaffolds with complex architectures and features are currently underway.

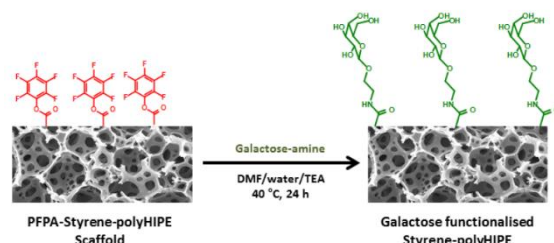


**Figure 1.** Schematic showing the surface functionalisation of a polyHIPE material.

<sup>1</sup>A. S. Hayward, A. M. Eissa, D. J. Maltman, N. Sano, S. A. Przyborski and N. R. Cameron, *iomacromolecules* **2013**, 14, 4271.

<sup>2</sup>C. R. Langford, D. W. Johnson and N. R. Cameron, *Polymer Chemistry* **2014**, 5, 6200.

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# Multi-stimuli responsive polymers based on spiropyran-functionalised poly(N-isopropyl acrylamide)

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Over the past few decades, significant progress has been made on the application of temperature-, pH-, and ionic-responsive polymers as capsules, gels, and brushes, typically in the biomedical field as a vehicle for targeted drug delivery systems. This has been spurred by recent advancements in the synthesis of multi-stimuli responsive polymers, which offer several distinct advantages over more traditional materials responsive only to a single stimulus. With these novel materials, a phase transition can be tuned to occur only when a combination of two or more environmental conditions are met, affording a higher degree of control. Alternatively, these polymers may be tailored such that a change in any one of its stimuli groups beyond a given threshold will trigger a conformational change in the polymer, providing increased flexibility. One such material of interest is poly(N-isopropyl acrylamide) functionalised with spiropyran groups, which is known to exhibit temperature-, light-, and pH-responsiveness<sup>1</sup> (**Fig 1**). The aim of this work is to examine in more detail the multi-stimuli responsive behaviour of these materials, and how its properties can be tuned to serve in a wide range of practical applications.

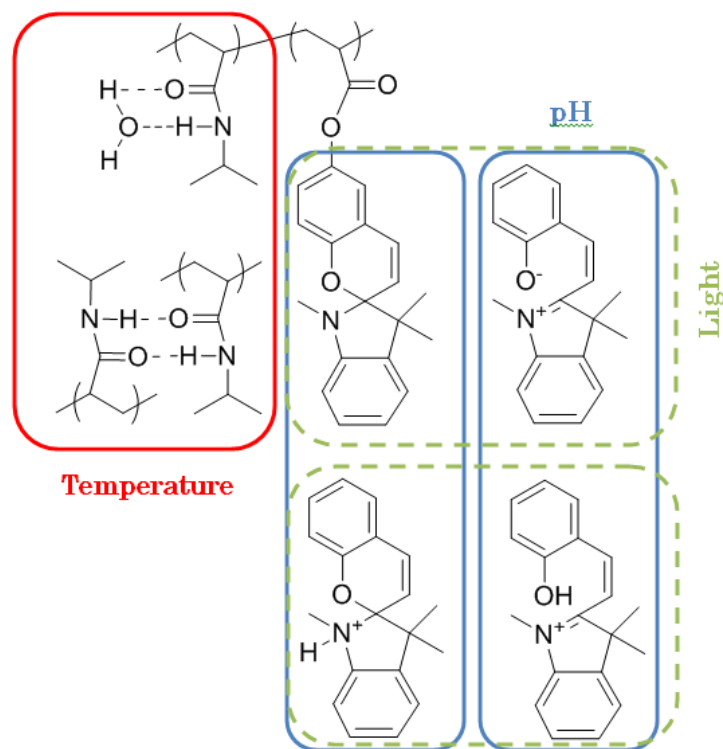


**Figure 1.** Multi-stimuli responsive nature of spiropyran-functionalised poly(N-isopropyl acrylamide)

<sup>1</sup> K. Sumaru, M. Kameda, T. Kanamori and T. Shinbo, *Macromolecules*, **2004**, 37, 4949-4955.

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# Synthesis of ultrahigh molecular weight and sequence-controlled polymers via emulsion polymerization

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Reversible-deactivation radical polymerization (RDRP) have provided powerful methods for preparing well-defined macromolecules with predetermined molecular weight, low polydispersity, and precisely defined end groups.<sup>1</sup> However, RDRP techniques still fall well short of producing ultrahigh molecular weight (UHMW) and sequence-controlled polymers from monomers that have low propagation rate coefficients (such as styrene and methacrylates). The key challenge in the synthesis of these polymers stems from the interplay between polymerization rate and side reactions such as bimolecular termination and chain transfer to the monomer. To produce UHMW or sequence-controlled polymers, a high polymerization rate for the polymer chains is needed which often correlates with an increase in terminated chains by side reactions. In this contribution to APS36, we present our recent works on using emulsion polymerization, an environmentally friendly technique, to both increase the polymerization rates and reduce side reactions. In particular, we have applied the emulsion polymerization technique, for the first time, to synthesize well-defined polystyrene diblock copolymer with a molecular weight up to a million g mol<sup>-1</sup> (Fig. 1A), and well-defined, sequence-controlled polymethacrylates with quantitative conversions attained throughout all the iterative monomer additions (Fig. 1B).<sup>2,3</sup> By understanding the RDRP mechanisms and overcoming their key challenge, our works represent a step forward in the synthesis of well-defined, large, and sequence-controlled macromolecules with the same size and degree of precision as nature.

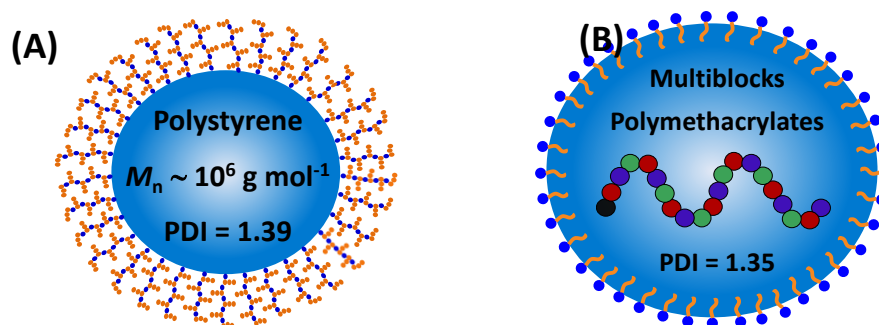


**Figure 1.** (A) Ultrahigh molecular weight polystyrene diblock copolymers and (B) sequence-controlled multiblock polymethacrylates prepared via emulsion polymerization.<sup>2,3</sup>

<sup>1</sup> K. Matyjaszewski and N. V. Tsarevsky, *Nat. Chem.*, **2009**, 1, 276.

<sup>2</sup> N. P. Truong, M. V. Dussert, M. R. Whittaker, J. F. Quinn and T. P. Davis, *Polym Chem-Uk*, **2015**, 6, 3865.

<sup>3</sup> Manuscript is currently under revision.

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# Quantitative first principles based kinetic modeling for the synthesis of well-defined macromolecular architectures

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Accurate multi-scale modeling is essential for the further development of advanced and well-defined macromolecular architectures.<sup>1</sup> From this perspective, a fundamental and complete understanding of the reaction mechanism and accurate values of the relevant parameters are indispensable. The exponential increase of computational resources and the continuous development of advanced quantum chemical procedures brought us to the verge of an era in which *ab initio* calculated rate coefficients can be used in kinetic models to predict quantitatively and *a priori* the responses of chemical reactions with fairly large reactants, even in the condensed phase.<sup>2</sup> Using carefully selected model systems, this allows the reliable description of macromolecular reactions, enabling further process optimization and control as well as providing a deeper insight in the reaction mechanism.<sup>3,4</sup>

In this work, this is illustrated for the case of reversible addition - fragmentation chain transfer (RAFT) polymerization (Fig. 1).

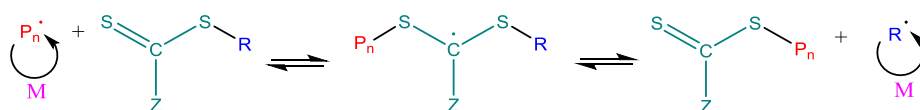


**Figure 2. Principle of RAFT polymerization.**

A first case is presented in which the parameters of the addition - fragmentation reactions pertaining to the RAFT process are solely obtained using *ab initio* methods. Subsequently these are readily used in a kinetic model to describe conversion, degree of polymerization and polydispersity in an excellent agreement with obtained experimental information.

A second case shows how *ab initio* calculations are applied to obtain a detailed reaction mechanism for the aminolysis of RAFT-polymers (Fig. 2), an often used technique to modify the RAFT-end group allowing further functionalization towards the synthesis of advanced polymeric structures.<sup>5</sup>

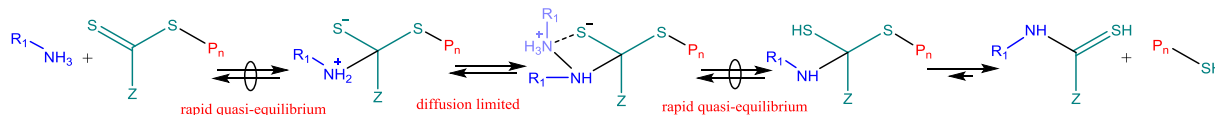


**Figure 3. Reaction mechanism for the aminolysis of a RAFT-macromolecule.**

<sup>1</sup> D. R. D'hooge, P. H.M. Van Steenberge, M. F. Reyniers, G. B. Marin, *Prog. Polym. Sci.* **2016**, dx.doi.org/10.1016/j.progpolymsci.2016.04.002

<sup>2</sup> G. B. Desmet, D. R. D'hooge, M. K. Sabbe, G. B. Marin, F. E. Du Prez, P. Espeel, M. F. Reyniers, *J. Org. Chem.* **2015**, *80*, 8520-8529

<sup>3</sup> B. B. Noble, M. L. Coote, *Int. Rev. Phys. Chem.* **2013**, *32*, 467-513

<sup>4</sup> M. L. Coote, E. H. Krenske, E.I. Izgorodina, *Macromol. Rapid. Commun.* **2006**, *27*, 473-497

<sup>5</sup> C. Boyer, A. Granville, T. P. Davis, V. Bulmus, *J. Polym. Sci., Part A: Polym. Chem.* **2009**, *47*, 3773-3794

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## **Ab initio based microkinetic modelling as a tool to understand and improve controlled radical polymerization processes**

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Precision control of the detailed chemical structure of each individual macromolecule is a key challenge to synthesize next-generation functional polymeric materials that meet predefined end-use properties. Controlled polymerization techniques are ideally suited to meet this challenge.

Full control of the structural characteristics of the individual macromolecules implies that throughout the polymerization the reaction rates are carefully controlled which introduces the need for dynamic synthesis protocols that allow an instantaneous control of reactant concentrations and temperature. This instantaneous rate control pertains not only to initiation, propagation and termination reactions but, more importantly, also includes reactions involving mediating agents and possible side reactions that might lead to structural defects. Hence, a fundamental and quantitative understanding of the reaction mechanism and the availability of accurate values of the relevant rate coefficients are indispensable.

This contribution highlights the application of computational chemistry as a complement to experiment in the development of microkinetic modelling tools that enable to improve control over the polymer microstructure. The potential of this approach to design tailor-made dynamical synthesis protocols is illustrated for reversible addition-fragmentation chain transfer (RAFT) polymerization, nitroxide mediated copolymerization and for the synthesis of well-defined copoly(2-oxazolines) via cationic ring-opening polymerization (CROP).

# Structure-Property Relationships in Tactic Acrylate Ester Polymers

*Brad A. Wells<sup>1</sup>, Baichuan Sun<sup>1</sup>, Amanda Barnard<sup>1</sup>*

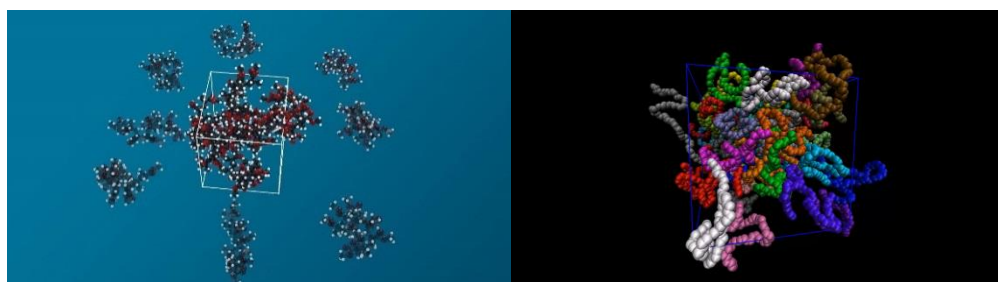
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Directly calculating the bulk properties of polymers presents a significant computational challenge. Polymers are generally amorphous, and thus able to adopt a wide range of configurations. Despite this complexity however, the fundamental chemical information contained within a polymer is generally quite small, as the polymer is made up of a small number of repeating units. The bulk properties of polymers can therefore be seen as being effectively encoded within a relatively small number of parameters. These may include structural features of the polymer chains, internal chain flexibility and non-bonding interactions between chain segments. Understanding how each of these factors influences the bulk properties of the polymer may lead to the ability to create polymers tailored to specific applications.

In this presentation structure-property relationships for a range of acrylate ester polymers is discussed. A particular focus of this work is in determining how the tacticity of the polymer chains can be included in the descriptive model. In order to elucidate the correlation of the structural features of the polymer models with their corresponding bulk properties, datasets of 100 randomly generated structures were created for each polymer and tacticity. Each structure contained 8 oligomers, with each oligomer made up of 8 monomers. An example structure for poly(methyl methacrylate) is shown in (Fig.1). These structures were optimised using tight-binding electronic density functional theory.<sup>1</sup> Molecular dynamics simulations using empirical potentials were also employed for each polymer to estimate the internal dynamics of the polymer chain motions.

Using these datasets a range of descriptors were then calculated to characterise each polymer and tacticity combination. Focus was given to descriptors that are able to distinguish between both the chemical composition and stereochemistry of the polymer chains. To determine the relevance of these descriptors, a correlation model predicting the glass transition temperature was created based on available experimental data. This glass transition temperature has been shown to vary substantially based on the tacticity of the polymer chain.<sup>2</sup> The validity of the model was then tested based on comparisons to the results of large-scale molecular dynamics simulations on acrylate ester polymers. These simulations involve models in excess of 20,000 atoms, an example of which is also shown in (Fig.1).

**Figure 1.** Example molecular models for poly(methyl methacrylate) for both structure-property datasets (left) and molecular dynamics simulations showing only backbone atoms (right).



M. Gaus, Q. Cui, M. Elstnew, *J. Chem. Theory Comput.* **2011**, 7, 931-948

<sup>2</sup> C. Duce, A. Micheli, A. Starita, M. Tiné, R. Solaro, *Macromol. Rapid Commun.* **2006**, 27, 711-715

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# Photoinducing Dynamic Covalent Chemistry in Covalent Adaptable Networks

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Main text: Times New Roman, 10pt, single spaced, intention left 0.5 cm (first line only), margins top, left, right, bottom all 2.5 cm With a wide array of formulations, chemistries and mechanisms to choose from, the science of making a stimuli responsive material has been studied and documented extensively over the past decade. In particular, the benefits of forming smart polymers that respond to irradiation have been widely studied as the ability to trigger a response based on light exposure provides spatiotemporal control to the process. Herein, we focus on the reaction processes and secondary functionalities that are incorporated into networks to enable tunable dynamic covalent chemistry within these polymer systems. Shape memory polymers (SMPs) with network properties such as high and tunable glass transition temperatures, multiple shape memory transitions, shape memory composites and two-way transition materials are recent examples of new directions that have been undertaken within the broad field of ‘smart’ materials. Among these new directions, the ability to reshape and resize materials repeatedly is particularly appealing. Such shape manipulations are desirable from the standpoint of SMPs applications, especially those requiring the excellent mechanical properties of thermosets. The concept of photoinduced addition-fragmentation chain transfer (AFCT) enables Covalent Adaptable Networks (CANs) in which the presence of photochemically active moieties can be used to induce network rearrangement in a controlled manner. Adopting this concept, it is demonstrated that by incorporating the photochemically active monomers, crosslinked polymer networks can then be induced to repeatedly undergo covalent bond rearrangement, and hence bulk material rearrangement. Further, for other, non-radical mediated catalytic processes that enable dynamic covalent chemistry within CANs, it is also possible to use light as a switch to turn on or off the dynamic covalent chemistry by forming or eliminating the catalyst, respectively. The fundamental mechanisms, benefits and opportunities associated with these photoinducible smart materials will be discussed.

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## Recent advances in $^{19}\text{F}$ MRI agents for detection of diseased tissue and cell tracking

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Recently there has been intense interest in fluorinated molecules to allow tracking of therapeutic particles and cells in vivo. The motivation for this is the very high selectivity of the  $^{19}\text{F}$  imaging experiment, since unlike in  $^1\text{H}$  MRI, the body does not contain a confounding fluorine background signal. In principle therefore, highly-selective  $^{19}\text{F}$  images can be superimposed on high-resolution anatomical  $^1\text{H}$  images, allowing tracking of suitably-labelled cells or biomarker molecules. Here we describe the motivation for the development of new partly-fluorinated copolymers with outstanding potential as  $^{19}\text{F}$  MRI imaging agents, and in particular as agents sensitive to the biological environment.<sup>1-10</sup> A second class of materials for the labelling of potentially-therapeutic stem cells will be described. These are statistical copolymers containing perfluoro-polyether side chains within a PEG-acrylate backbone. Measurement of cell uptake and imaging performance will be described.

<sup>1</sup> H. Peng, I. Blakey, B. Dargaville, F. Rasoul, S. Rose, A.K. Whittaker, *Biomacromolecules*, **2009**, 10 374.

<sup>2</sup> K.J. Thurecht, I. Blakey, H. Peng, O. Squires, S. Hsu, C. Alexander, A.K. Whittaker, *J. Amer. Chem. Soc.*, **2010**, 132, 5336.

<sup>3</sup> H. Peng, K.J. Thurecht, I. Blakey, E. Taran and A.K. Whittaker, *Macromolecules*, **2012**, 45, 8681.

<sup>4</sup> K. Wang, H. Peng, K.J. Thurecht, S. Puttick and A. K. Whittaker, *Polym. Chem.*, **2013**, 4, 4480.

<sup>5</sup> K. Wang, H. Peng, K.J. Thurecht, S. Puttick and A. K. Whittaker, *Polym. Chem.*, **2014**, 5, 1760.

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<sup>7</sup> C. Zhang, H. Peng, A.K. Whittaker, *J. Polym. Sci. A: Polym. Chem.*, **2014**, 52, 2375.

<sup>8</sup> C. Zhang, H. Peng, S. Puttick, J. Reid, S. Bernardi, D. J. Searles, A.K. Whittaker, *Macromolecules*, **2015**, 48, 3310.

<sup>9</sup> K. Wang, H. Peng, K.J. Thurecht, S. Puttick and A. K. Whittaker, *Polym. Chem.*, **2016**, DOI: 10.1039/C5PY01707F.

<sup>10</sup> C. Zhang, S.S. Moonshi H. Peng, S. Puttick, J. Reid, S. Bernardi, D. J. Searles, A.K. Whittaker, *ACS Sensors*, **2016**, 0.1021/acssensors.6b00216.

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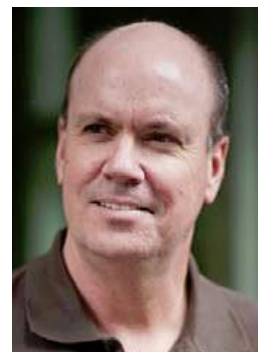
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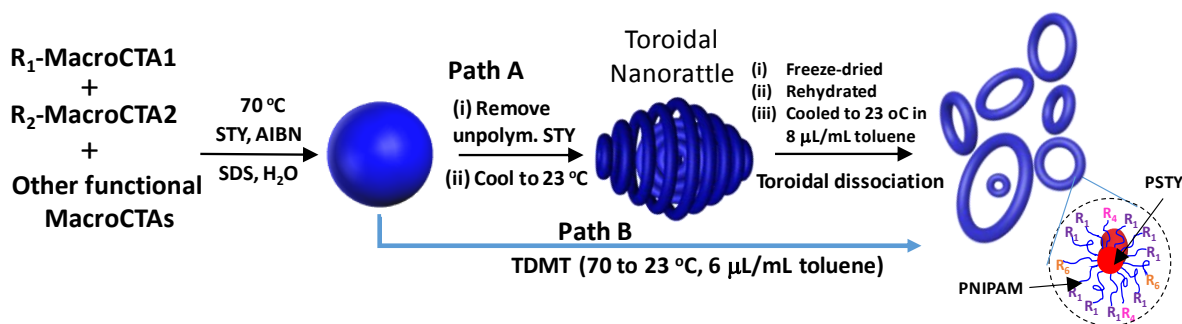


# Toroidal Nanorattles: Self-assembly and On-demand Release of Free Toroids.

Zhongfan Jia,<sup>1</sup> Valentin A. Bobrin<sup>1</sup> and Michael J. Monteiro<sup>1</sup>

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Nanorattles are complex structures consisting of an outer shell and a detached inner core. Examples include the self-assembly of surfactants, amphiphilic dendrimers or block copolymers into onion-like nanorattles. Our group developed a method which utilizes the reversible addition-fragmentation chain transfer (RAFT) emulsion polymerization of styrene with a thermoresponsive (poly(N-isopropylacrylamide), PNIPAM) MacroCTA in water to produce spherical particles at a temperature well above the lower critical solution temperature (LCST) of PNIPAM. Upon cooling, a temperature directed morphology transformation (TDMT) results in the formation of a wide range of nanostructures, ranging from spheres, worms, rods, vesicles, donuts, lamellae, and recently tadpoles; with the additional feature of controlled surface functionality on the particles from the R-group on the MacroCTA.1-5 All these structures could be freeze-dried and rehydrated in water with maintenance of their original nanostructure. In this work, we report a method to produce chemically multifunctional toroidal nanorattles directly in water at a high weight fraction of polymer (~10 wt%) using our method, which can be then triggered with the addition of a plasticizer for polystyrene (PSTY) to produce individual toroidal micelles (Scheme 1).



**Scheme 1.** Synthesis of toroidal nanorattles.

<sup>1</sup> Kessel, S.; Urbani, C. N.; Monteiro, M. J. *Angew. Chemie. -Int. Ed.* **2011**, *50*, 8082-8085.

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<sup>5</sup> V.A. Bobrin, M.J. Monteiro. *J. Am. Chem. Soc.*, **2015**, *137* (50), 15652–15655.

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## Bioinspired polymer catalysts- mimicking the active site of enzymes

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Enzymes as nature's catalysts exhibit remarkable control over biological reactions and their use in industrial applications is widespread<sup>1,2</sup>. The utility of enzymes as catalysts however, is reduced by their low stability, limited operating range and high cost. Past studies, including pioneering work by Breslow et al., have aimed to mimic the complex chemistry of enzymes while reducing their limitations as industrial catalysts, with limited success<sup>3,4</sup>. Despite the complex structure of enzymes, they employ a relatively small functional area- the active site- to mediate their important reactions (**Figure 1**)<sup>5</sup>. In this presentation I will outline our efforts towards mimicking the structure and function of a proteolytic enzyme active site. Our artificial active site (AAS) incorporates the key functional groups present in the enzyme's functional core, and exhibits a significant hydrolytic effect in our selected catalysis assays. Furthermore, the AAS moiety has been used to functionalise a hydrophobic polymer backbone to deliver greater control over the AAS secondary structure and self-assembly in solution. These materials exhibit a wider operating range and increased stability over the natural enzyme, and have application in detergents, antibacterial coatings and biodiesel production.

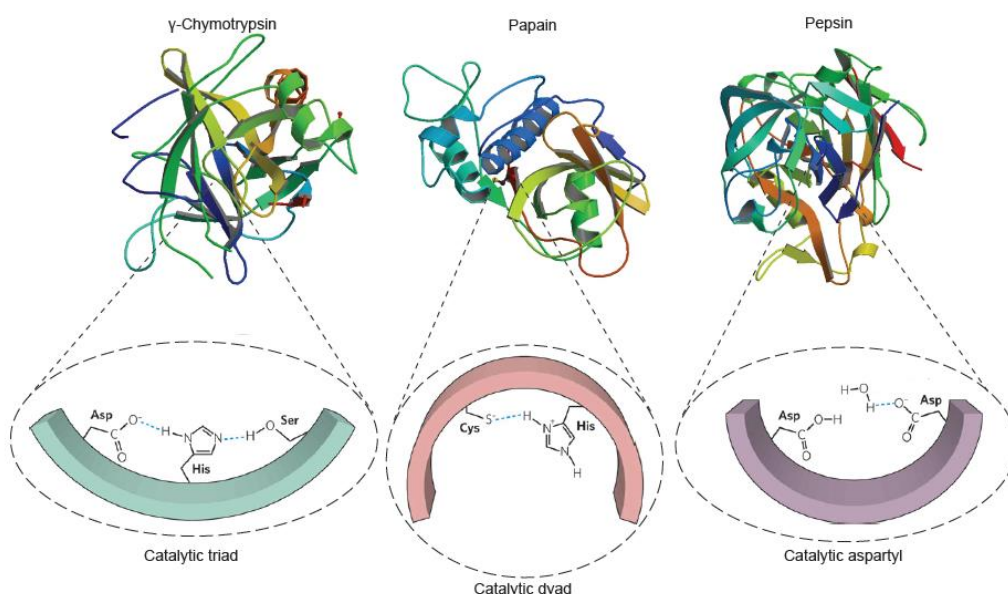


**Figure 1:** The amino acid residues present in the active site of some proteolytic enzymes. The interplay of these functional groups is chiefly responsible for the catalysis observed by the parent enzyme<sup>6</sup>.

1. K. E. Jaeger, T. Eggert *Current Opinion in Biotechnology*, **2002**, 4, 390-397.
- 1 H.P. Meyer, E. Eichhorn, D. Hanlon, S. Luetz, M. Schuermann, R. Wohlgemuth, R. Coppolecchia, *Cat. Sci. and Tech.* **2013**, 3, 29.
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- 5 Elinor Erez, E., Fass, D. & Bibi, E. *Nature* 2009, 459, 371-378

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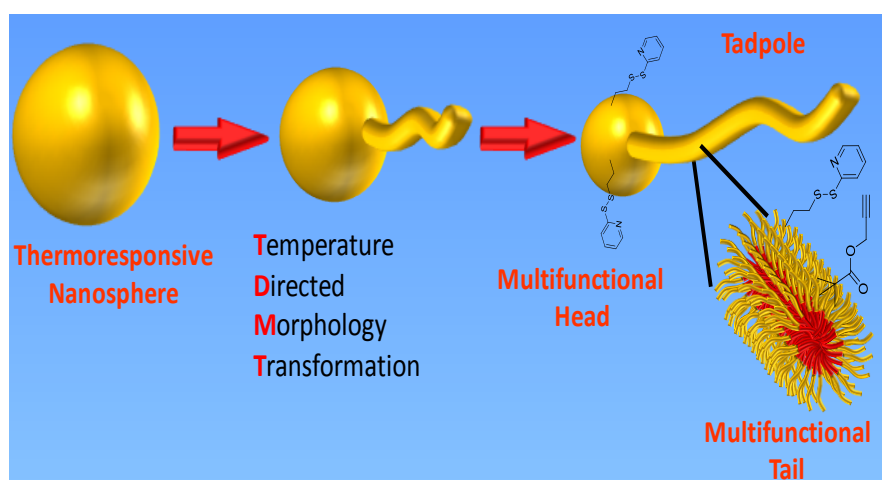
Biomimetics, catalysis, green chemistry

# Temperature-Directed Self-Assembly of Multifunctional Polymeric Tadpoles

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Building nanostructures with controlled geometry and multiple compartments with a variety of chemical functionalities represent an important development in the design of nano-objects<sup>1</sup>. These types of new structures will further extend the use of nano-objects in biological<sup>2</sup>, photonics<sup>3</sup> and chemical<sup>4</sup> applications. Here, we demonstrate the synthesis of chemically multifunctional tadpoles made at high weight fraction of polymer (> 10 wt. %). The tadpoles are synthesized using two different thermoresponsive MacroCTAs with either alkyne or pyridyldisulfide end-groups, allowing chemical functionality in the head, tail, or both. Water-soluble molecules or polymers were coupled to the head, tail, or both without a change in tadpole configuration. In addition, the tadpoles can be dried, rehydrated, and stored in water for 5 months without a change in shape. This method represents a new and an important synthetic development in the design of nano-objects.<sup>5</sup>



**Scheme 1.** Synthesis of Multifunctional Nano-Tadpoles

<sup>1</sup> A.H. Groschel, A.H.E. Muller. *Nanoscale*, 2015, 7, 11841–11876.

<sup>2</sup> S. Mitragotri, J. Lahann. *Nat. Mater.*, 2009, 8, 15–23.

<sup>3</sup> W.U. Huynh, J.J. Dittmer, A.P. Alivisatos. *Science*, 2002, 295, 2425–2427.

<sup>4</sup> Y. Nishihata, J. Mizuki, T. Akao, H. Tanaka, M. Uenishi, M. Kimura, T. Okamoto, N. Hamada. *Nature*, 2002, 418, 164–167.

<sup>5</sup> V.A. Bobrin, M.J. Monteiro. *J. Am. Chem. Soc.*, **2015**, 137 (50), 15652–15655.

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# Controlling the Self-Assembly of Polyelectrolyte-Cyclic Peptides: Just Add Salt

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Self-assembling peptide nanotubes formed by the stacking of cyclic peptides (CPs) offer a versatile scaffold for formation of nanotubular structures (**Fig. 1**)<sup>1</sup>.

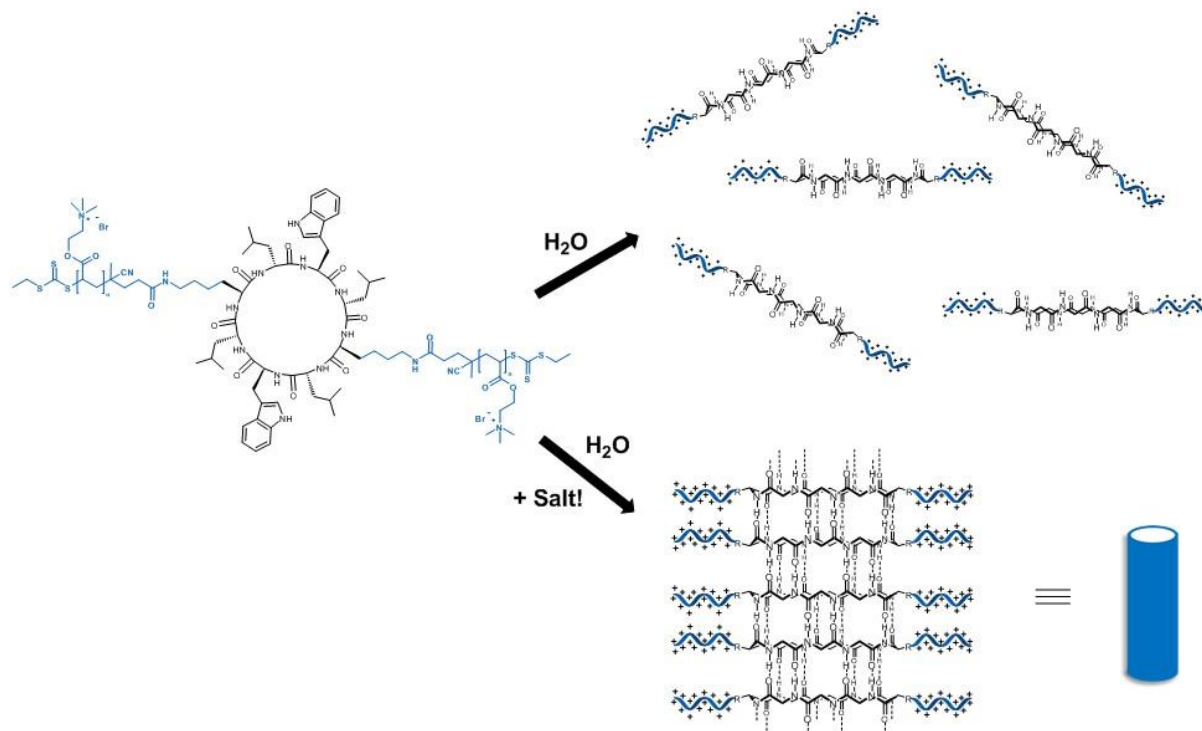


**Figure 1:** Polyelectrolyte-peptide conjugates form  $\beta$ -sheet hydrogen bonded assemblies in the presence of salt.

Conjugating polyelectrolytes to the CP building block will result in nanotubes with an external surface charge. The high charge densities of these polymers cause inter-chain repulsion, which competes with CP H-bond assembly and prevents self-assembly in water (**Fig. 1**). By adding salt to the solution of polyelectrolyte conjugates, the charge on the polymers are effectively shielded, diminishing inter-chain repulsion and allowing assembly (**Fig. 1**).

Using RAFT, amide coupling and post-polymerisation substitution, we have developed novel polyelectrolyte materials with remarkable self-assembly properties that we have analysed by small angle neutron scattering.

R. Chapman, M. Danial, M. L. Koh, K. A. Jolliffe, S. Perrier, *Chem. Soc. Rev.* **2012**, *41*, 6023-6041

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# Quaternized P4VP-*b*-PS Honeycomb Film with Biocidal and Antibiofouling Effects

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Biofilm contamination has been one of the most serious nosocomial situations. Antimicrobial polymer films have been widely used to reduce bacterial surface contaminations. However, most antimicrobial polymers exert either biocidal or antibiofouling effects. We here present an amphiphilic polymer quaternized poly(4-vinylpyridine)-*block*-polystyrene (quaternized P4VP-*b*-PS) honeycomb film which combines both biocidal and antibiofouling functions.

The polymer P4VP-*b*-PS was synthesized through RAFT polymerizations (**Fig. 1**). Then the polymers are cast through breath figures method to form honeycomb pattern (**Fig. 2**).<sup>1</sup> Finally the P4VP blocks are quaternized to bear biocidal ability. The influences of different film casting factors were further investigated on surface morphology and antimicrobial performances against common nosocomial bacteria. Both biocidal (through quaternized P4VP) and antibiofouling (through honeycomb pattern) effects of the film should be expected.

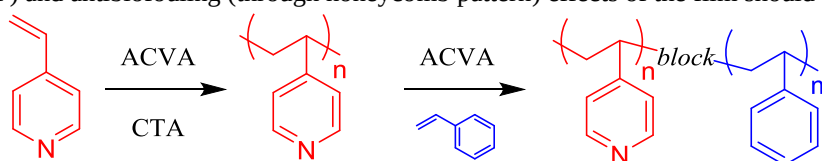


**Figure 1:** P4VP-*b*-PS Synthesis Route

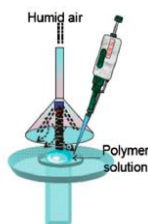


**Figure 2:** Breath Figures Casting Method of P4VP-*b*-PS Honeycomb Film (the figure to the left reprinted with permission from Reference 1)

<sup>1</sup> Maribel Hernandez-Guerrero, Martina H. Stenzel, *Polym. Chem.* **2012**, 3, 563

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# Co-Assembled Peptide Nanoscaffolds which Present Multiple Bioactive Signals Demonstrate Superior Control of Cell Fate in 3D Cell Culture

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The nanofibrillar structures that underpin self-assembling peptide (SAP) hydrogels offer great potential for the development of finely tuned cellular microenvironments suitable for tissue engineering.<sup>1</sup> However, biofunctionalisation without disruption of the assembly remains a key issue. SAPs present the peptide sequence within their structure, and studies to date have typically focused on including a single biological motif, resulting in chemically and biologically homogenous scaffolds. This limits the utility of these systems, as they cannot effectively mimic the complexity of the multicomponent extracellular matrix (ECM).

In this work, we show for the first time the successful co-assembly of multiple bioactive SAP sequences into a single nanofibre scaffold and demonstrate that this approach is more bioactive than either of the individual systems alone.

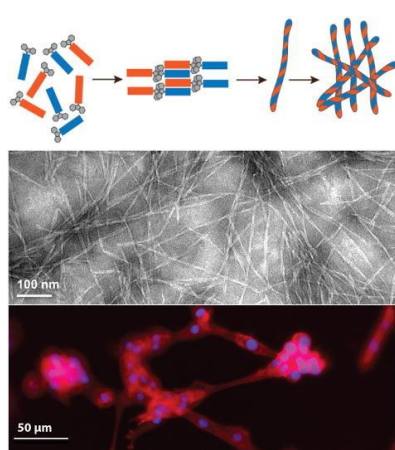


Fig 1: top: cartoon of assembly, middle: nanofibrils formed via coassembly: bottom multinucleate C2C12 cells in culture

We designed two SAPs that Assembled to clear hydrogels Under physiological conditions: Fmoc-FRGDF (from fibronectin) and Fmoc-DIKVAV (IKVAV from laminin). We used a range of mechanical and spectroscopic techniques to characterise the system (AFM, TEM, FTIR, CD, and SAXS). Our results demonstrate that when mixed prior to assembly, these SAPs are able to co-assemble to form stable hybrid nanofibres containing dual epitopes. Comparison of the co-assembled SAP system to the individual SAP hydrogels and to a mixed hydrogel system demonstrated superior stable, transparent, shear-thinning hydrogels at biological pH, ideal characteristics for tissue engineering applications.

Importantly, *in vitro* 3D cell culture shows that only the co-assembled hydrogel was able to induce multinucleate myotube formation with muscle precursor cells (C2C12 cells). This study similarly demonstrates the distinct biochemical properties of a co-assembled system for the concomitant presentation of unique signals to individual C2C12 myoblasts stimulating distinct cellular behaviours with regards to cell spreading and the formation of elongated, multinucleated myotube-like structures.<sup>2</sup>

In conclusion, this co-assembled system will provide a platform for a tuneable biomimetic material for future *in vitro* and *in vivo* tissue engineering applications.

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## Biomimetic Materials ‘Controlling the nonlinear mechanics’

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Fibrous networks of biopolymers are found in both the intracellular and extracellular matrix. From the microscopic scale of a single cell to the macroscopic scale of fibrous tissues, biopolymers with different stiffness control cellular processes such as cell differentiation, proliferation, transportation and communication.<sup>1</sup> In recent years, a large number of different hydrogels has been developed, often with the goal to create an artificial extracellular matrix for biomedical applications. However, the mechanical environment inside and outside the cell is not determined by a single component.<sup>1</sup> Multiple biopolymers with different structural and mechanical properties which physically interact with each other, make the mechanical environment of a cell in vivo much more complicated than the environment of a cell in a single-component artificial matrix.

The mechanics of natural biopolymer gels however, are very different from most synthetic hydrogels because they show strain stiffening behaviour.<sup>2,4</sup> Reconstituted networks of cytoskeletal polymers such as actin or intermediate filaments or extracellular biopolymers such as collagen or fibrin show a large increase in stiffness upon an applied stress or deformation.<sup>2,4</sup> The stiffening response prevents these networks from breaking under external stresses and also enables communication between cells growing in these materials. Recently a new biomimetic polymer hydrogel was developed with unique cytomimetic properties, based upon oligo(ethylene glycol) grafted polyisocyanopeptides. These extremely stiff helical polymers form gels upon warming at concentrations as low as 0.005 %wt polymer, with materials properties almost identical to these of intermediate filaments and extracellular matrices.<sup>3,4</sup> The unique ability of these materials and their application in cell growth and drug therapeutics revealed the importance of polymer stiffness and material non-linear mechanics.<sup>5,6</sup>

These nonlinear mechanical properties have been extensively studied in single-component biopolymer networks, but little is known about how the stiffening response is affected by the composite nature of natural biopolymer networks such as the cytoskeleton or the extracellular matrix.<sup>7</sup> Approaches of how to control these nonlinear properties and their importance will be presented.

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# Nitroxide Containing Microspheres as Biodegradable Drug Delivery Systems to Target Bacterial Biofilms

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The ability of bacteria to form protective colonies known as biofilms can be observed in all parts of life from large ships and pipes to food-packaging or medical implants. Besides antibiotic resistance, the formation of biofilms is a major hindrance to effectively target diseases of bacterial origin or bacterial infections. The extracellular matrix of a biofilm protects the inhabiting bacteria from the penetration of immune cells and the traditional antibiotics therapy. Most often bacterial infections are introduced by medical implants which need to be replaced when inflamed followed by a harsh antibiotic treatment. Genetic disorders such as cystic fibrosis, however, can cause bacterial infections within the patient's body. Cystic fibrosis mainly effects the lungs where the biofilm (mucus) additionally hinders the ability to breathe.

The dispersal of biofilms is a vital part of the bacterial biofilm life cycle where mature biofilms disperse into planktonic bacteria in order to spread and proliferate. Nitric oxide has been shown to effectively induce biofilm dispersal in sub-toxic concentrations.<sup>1</sup> In combination with an antibiotic treatment, the biofilm can be eradicated completely.<sup>2</sup> The targeted delivery of a gaseous compound, however, remains challenging. Our research group has investigated the biofilm dispersing effects of nitroxides as solid and stable nitric oxide mimics.<sup>3</sup>

As a next step, we aim to design drug delivery systems enabling the transport of the active substances into the lungs of the patients by their incorporation into biodegradable polymeric microspheres. Various synthetic strategies and nitroxide compounds have been tested to produce biodegradable microspheres. In this way, we are able to identify and directly compare the limitations and potential of each technique. Tunable release profiles and nitroxide contents enable the design of an effective and highly flexible pulmonary drug delivery platform.

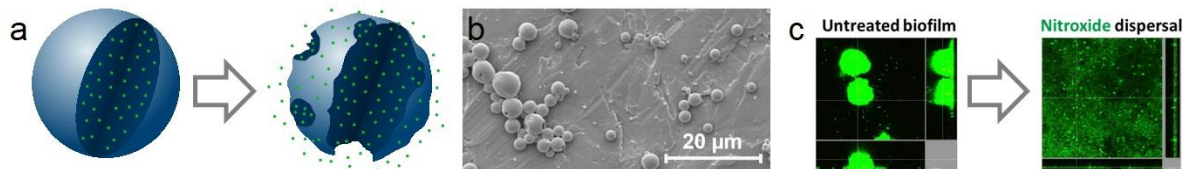


**Figure 1.** Schematic display of a drug delivery system composed of biodegradable microspheres (a), SEM image of nitroxide containing microspheres produced using oil-in-water emulsion technique (b), and biofilm dispersal effect of free nitroxide molecules in preliminary tests (c).<sup>3</sup>

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<sup>3</sup> C. de la Fuente-Núñez, F. Reffuveille, K. E. Fairfull-Smith and R. E. W. Hancock, *Antimicrobial Agents and Chemotherapy*, **2013**, 57, 4877-4881.

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# Plasma Polymer meets surface initiated ARGET-ATRP; a facile way for robust coatings to immobilize antifungals

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Fungal infections are an underestimated killer, being the fourth leading cause of hospital-acquired bloodstream infections<sup>1</sup>; even worse, systemic infections, candidiasis, have mortality rates of up to 40%.<sup>2</sup> Fungi prove to be formidable foes, partly due to being eukaryotic like human cells and partly due to their polymorphic adaptability, arguably making them harder to fight than bacteria. This struggle is further exacerbated due to the small pool of antifungal drugs of which many can't be administered systematically due to their toxicity to the host.<sup>3</sup> All these aspects taken together call for a shift in paradigm where these cytotoxic drugs are covalently tethered onto surfaces to prevent fungal colonization without the drug leaching into the host. This tethering, however, requires to be adjustable in length and chemical nature to reach important cell wall and membrane targets protected by a very thick cell envelope. Surface initiated ARGET-ATRP is a well-controlled polymerization process which has the flexibility and precision to control the tether but requires the use of laborious chemistry to install the ATRP initiator.<sup>4</sup> In contrast, we hereby report the use of an industrial grade process, plasma polymerization, to generate a thin and well adherent covalent coating which then is further used to immobilize an ATRP initiator. This coating is then used to grow poly-(Hydroxyethyl)methacrylate (pHEMA) in air via surface initiated ARGET-ATRP up to thicknesses of 100nm. Despite the non-fouling properties of pHEMA, pathogenic *C. albicans* can

colonize these surfaces within 24h which is why, for proof of concept, they were further modified to attach the antifungal drug caspofungin. In contrast, caspofungin modified surfaces reliably resist fungal attachment and colonization. This novel combination of non-classical plasma polymerization and ARGET-ATRP holds the promise to provide a platform for novel antifungals or "rediscovering" off-patent or toxic antifungal drugs as surface active agents to prevent implant associated

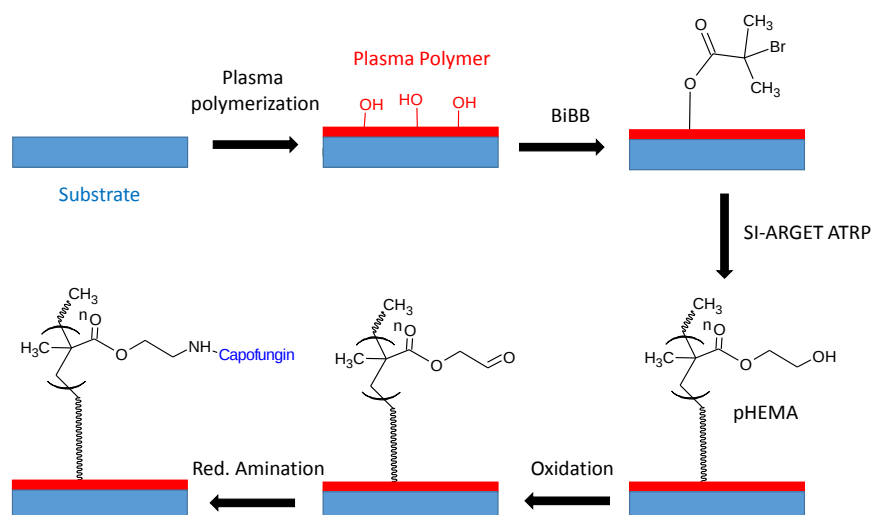


Fig. 1: pHEMA brushes were grown from an initiator-modified plasma polymer coating and post-functionalized with an antifungal drug (caspofungin)

fungal infections.

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## **Preparation of biodegradable and biocompatible emulsion templated scaffolds for tissue engineering by thiol-acrylate polymerization of polycaprolactone macromers**

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The emulsion templating process offers a route to highly porous polymers with well-defined morphologies<sup>1</sup>. The process, whereby the continuous phase of a high internal phase emulsion (HIPE) is polymerized, results in materials referred to as polyHIPEs. These porous materials have found application in a wide range of areas including catalysis<sup>2</sup>, organic synthesis<sup>3</sup>, and in biotechnology<sup>4, 5</sup>.

Recent developments in the photo-initiated polymerization of HIPEs have allowed for the preparation of polyHIPE polymers from monomers which form highly unstable emulsions, such as multifunctional thiol and alkene (typically acrylate) monomers<sup>6</sup>, including polycaprolactone triacrylate (PCL-TA)<sup>7</sup>. The resulting polyHIPE materials have well-defined and interconnected porosities, allowing for cell penetration into the polymer.

The preparation of biodegradable thiol-PCL-TA polyHIPEs will be described along with initial biocompatibility results. Fibroblast cell lines have been successfully cultured on the thiol-PCL-TA polyHIPEs for periods of up to 14 days, indicating they are biocompatible and the degradation products of these materials have been shown to be non-toxic to cells.

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# Hydrogels Containing Hierarchical Structures Based on Poly(2-oxazoline)s and Related Polymers

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The rise in popularity of Tissue Engineering and Biofabrication as disciplines over the last decade has driven demand for highly sophisticated materials to mimic human tissue. In particular, hydrogels have contributed significantly to these fields thanks to their high water content and other physical properties similar to native tissue. Despite significant efforts, methods for making hydrogels containing hierarchical features (nano- to macro-) found in native tissues have not become wide-spread. This presentation will focus on our recent attempts to use a 3-dimensional contact printing method to produce biocompatible hydrogels with controlled porosity and spatially controlled patterning of chemical signals.

The polymer chosen for this study is from the family of poly(2-oxazoline)s, a class of polymers with potentially superior protein resistance to the gold standard, polyethylene glycol. Poly(2-oxazoline)s are also attractive to polymer scientists as their synthesis is relatively straightforward and can include a range of possible monomers to generate various architects from one-pot reactions.<sup>1</sup>

In this work porosity has introduced into hydrogels based on poly(2-oxazoline)s using sacrificial moulding methods based on melt electrospun written and fused deposition modelling templates. This approach (**Fig 1**) allows for independent printing of the porogen, encapsulation of the template and subsequent dissolution of the template to leave an exact negative replica of the porogen with fidelity at the sub-micron level. The presentation will conclude with progress on incorporation of small molecules into the template for transfer to the hydrogels in a 3D micro-contact printing process.

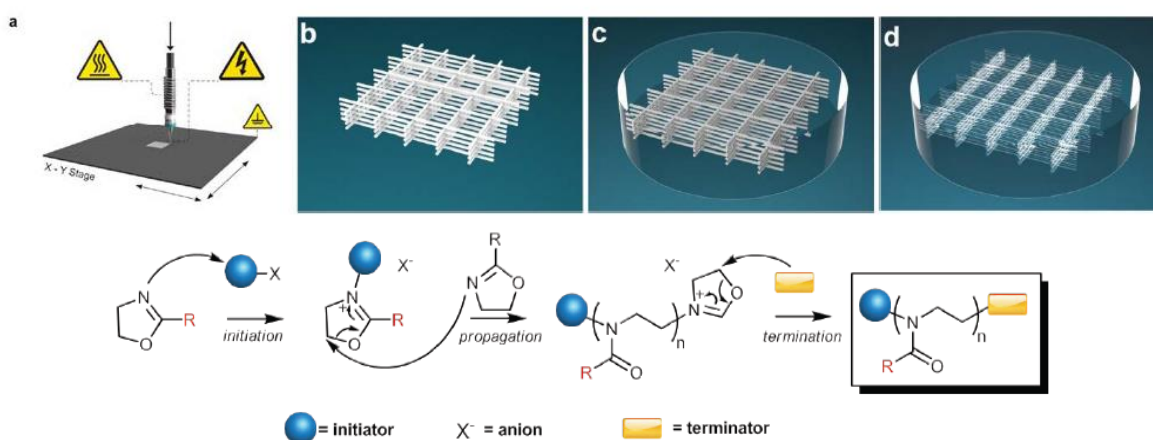


**Figure 1.** Schematic of 3D sacrificial micro-moulding. A) The 3D micro-fibre porogen is printed; B) the porogen template is C) encapsulated in a hydrogel; D) then sacrificed by dissolution to reveal the microchannel architecture. Below is the general CROP mechanism of poly(2-oxazoline)s.

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## Light responsive gelatin hydrogel particles for cartilage regeneration

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Human mesenchymal stem cells (hMSCs) are regarded as a promising candidate in cell therapies for cartilage regeneration, however limitations still exist for effective tissue regeneration – these include (but are not limited to) the requirement for large cell numbers, inability to efficiently control MSCs differentiation, low retention in the tissue post implantation and difficulties in evaluation or monitoring of regenerated tissues <sup>1</sup>.

In order to address these problems, we have employed gelatin, a collagen-derived polydisperse biomaterial, to construct a bulk hydrogel via rapid and highly biocompatible thiol-ene photoclick chemistry. hMSCs were encapsulated, *in situ*, in a gelatin hydrogel and cultured in the presence of chondro-inductive supplements (**Fig.1.a**). Live/dead staining confirmed that MSCs remained viable after initial encapsulation and longer term cultures. Further to this, when cultured in the presence of chondrogenic supplements, cells could efficiently differentiate into the chondrogenic lineage, as determined by immunostaining for type -I and -II collagens and qPCR. A droplet microfluidic device was used to fabricate non-immunogenic gelatin hydrogel particles for *in situ* hMSCs encapsulation and chondrogenesis culture (**Fig.1.b**) <sup>2</sup>. The hydrogel particles not only present a high surface area to volume ratio to acquire higher mass transfer rate, but also act as a micro 3D culture environment to protect cells from the immune system and assist high cell retention in the targeted tissue area. Furthermore, incorporation of the photolabile *o*-nitrobenzyl ester moiety within the network structure allows low UV doses for rapid solubilisation of the gel facilitating the collection of cells without sacrificing the extracellular matrix or other biomolecules by enzymatic digestion.

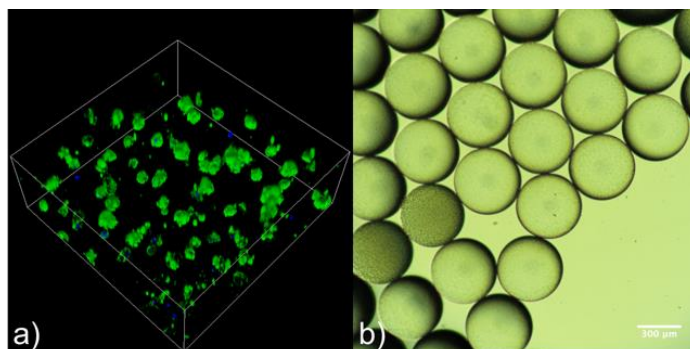


**Figure 1. a)** 3D Laser confocal fluorescent imaging of hMSCs culture in a gelatin based hydrogel (stained with phalloidin/actin and hoechst 33342/nuclei); **b)** Droplet microfluidic device generated gelatin hydrogel particles.

<sup>1</sup> P. A. Parmar, L. W. Chow, J.-P. St-Pierre, C.-M. Horejs, Y. Y. Peng, J. A. Werkmeister, J. A. Ramshaw and M. M. Stevens, *Biomaterials* **2015**, 54, 213-225

<sup>2</sup> F. Lapierre, N. R. Cameron and Y. Zhu, *Journal of Micromechanics and Microengineering* **2015**, 25, 035011.

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# Saving Vision with Light: Photo-modulated Ocular Drug Delivery

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Age-related macular degeneration (AMD) is the leading cause of blindness in Australia today.<sup>1</sup> The only way to save vision requires monthly injection to the back of the eye with biomacromolecular anti-angiogenics, such as bevacizumab (Avastin®) and ranibizumab (Lucentis®).<sup>2</sup> This invasive procedure, with the potential for sight threatening complications, has obvious negative impacts on patient's quality of life as well as posing a major burden on the health care system. A drug delivery system that serves as a reservoir to enable sustained release or 'on-demand' release of therapeutic protein/antibody would be beneficial, since it will significantly reduce the number of injections into the back of the eye. The release of therapeutic payloads from the drug reservoir can be modulated on demand by light, particularly due to its external spatiotemporal control.<sup>3</sup> In order to fabricate a photo-responsive drug delivery system a thermo-responsive polymer was formulated into a hydrogel containing light sensitive nanoparticles and therapeutic payloads. The release of different payloads, ranging from small molecules (doxorubicin, trimacrinolone acetonide) to biomacromolecules (lysozyme, bovine serum albumin, and IgG/antibody), can be switched on and off upon exposure to light (**Fig. 1**).

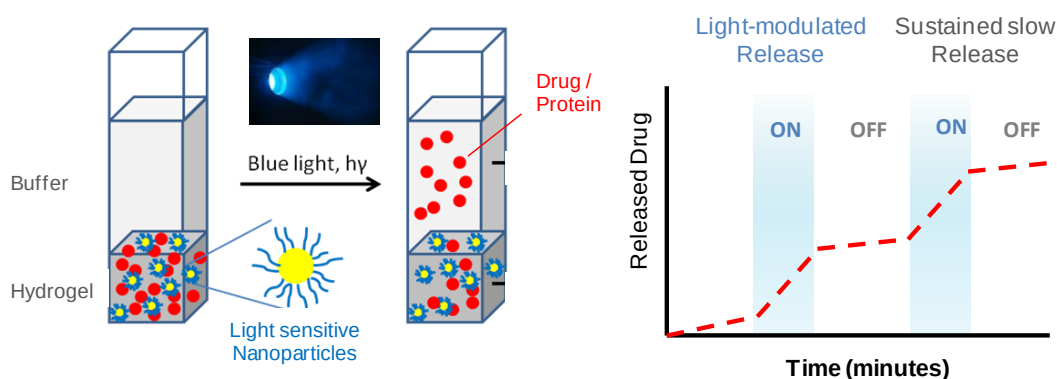


**Figure 1.** Schematic representation of photo-modulated drug/protein release from the hydrogen, and the release profile of the drug into the buffer in the absence (OFF) and presence of light (ON).

Importantly, the released lysozyme and bevacizumab retained more than 85% of their original biological activity as measured by lysis and ELISA assays, respectively. In addition, the formulation did not show *in vitro* toxicity to ocular cells, and can be implanted via minimally invasive subconjunctival injection through a 30-gauge needle. Safety assessments against the ocular injection and visible light exposure have been carried out as well. Due to its minimum toxicity, tuneable drug release rates and high versatility, this photo-modulated drug delivery system has a potential to improve the treatment of chronic ocular diseases.

<sup>1</sup> H. R Taylor, J. E Keefe, H. T. V Vu, J. J. Wang, E. Rohtchina, M. L. Pezzullo, P. Mitchell, *Med. J. Aus.* **2005**, *182*, 565-568.

<sup>2</sup> O. A. Anderson, J. W. B. Bainbridge, D. T. Shima, *Drug Discov. Today* **2010**, *15*, 272-282.

<sup>3</sup> S. Sortino, *J. Mater. Chem.* **2012**, *22*, 301-318.

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# Hierarchical self-assembly of triblock glycopolymers into multicompartimentalised structures inspired by viruses

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Spherical particles are the most investigated morphology as polymeric drug delivery vessels due to their versatility and relative ease of preparation. On the other hand viruses often have elongated worm-like morphologies with carbohydrate derivatives decorating the outer layer and demonstrate a high efficiency in penetrating the cellular membrane.

We propose the use of linear triblock terpolymers to imitate these worm-like and multicompartimentalised structures through a hierarchical self-assembly process described in the literature<sup>1</sup>. Well defined terpolymers with sugar functionalities on the side chain are synthesised using RAFT polymerisation. The self-assembly process outlined in **Fig.1** initially produces spherical micelles which are subsequently used as ‘building blocks’ for more sophisticated aggregates upon solvent exchange to aqueous solution. Cellular uptake of particles with different morphologies is tested and compared with traditional core-shell spherical particles and targeting ability of sugar moieties that cover the outer surface is evaluated on various cell lines.

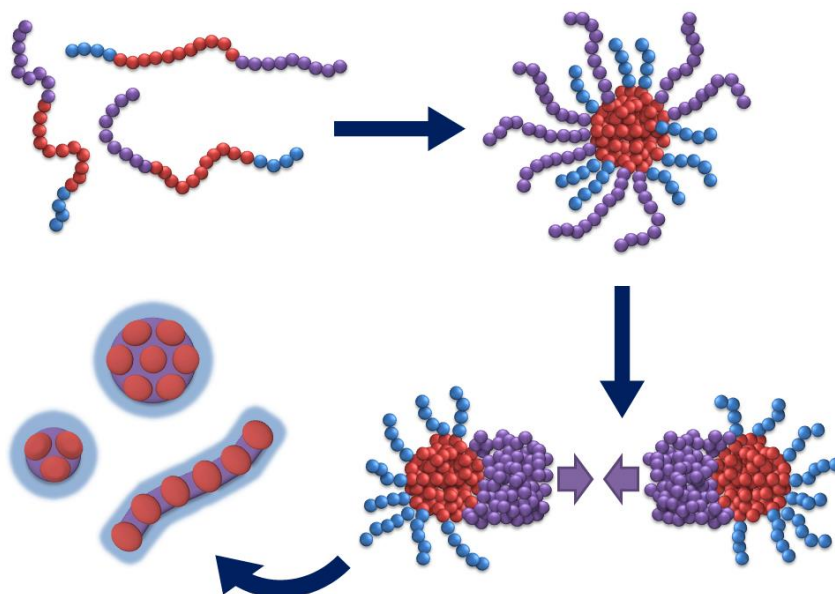


**Figure 1.** Hierarchical self-assembly process. Spherical micelles are formed first with a solvophobic B block core (red). Upon solvent exchange the responsive C block (purple) collapses triggering aggregation into more complex structures while the hydrophilic carbohydrate-based block (blue) forms the shell stabilising the nanoparticles.

A. H. Groschel, F. H. Schacher, H. Schmalz, O. V. Borisov, E. B. Zhulina, A. Walther and A. H. Muller, *Nat. Commun.*, **2012**, 3, 710

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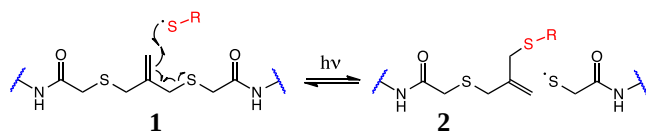
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Research interests: Glycopolymers, RAFT polymerisation, self-assembly

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In addition to rapid photodegradation properties, the AFCT crosslinker also allows for photoinduced viscoelastic material properties<sup>2</sup>. With a growing appreciation of the influence that viscoelastic materials have on cellular differentiation, it is important to have a tool to probe the spatial and temporal effects of viscoelasticity on cellular behavior. By swelling in a photoinitiator with a stoichiometric amount of a thiol, the network transitions from purely elastic to viscoelastic upon exposure to light and recovers elasticity upon removal of the light.



<sup>2</sup> H. Park, C. Kloxin, T. Scott, C. Bowman, *Macromolecules*. **2010**, 43(24), 10188-10190

**Research interests:** My research interests lie in utilizing material chemistries to create hydrogel scaffolds that mimic the biochemical and physical properties of the extracellular matrix.

## Biomaterials that promote endothelialization

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Blood coagulates when it contacts foreign surfaces, including biomaterials that are currently used for cardiovascular applications. The development of thrombus (a blood clot) on the material surface can result in failure of devices such as small diameter vascular grafts and artificial heart valves. Additionally, a piece of the clot may detach and cause issues downstream such as stroke. Researchers have attempted to develop blood compatible interfaces for decades, but no surface has achieved great clinical success.<sup>38</sup> Native vasculature is lined with a monolayer of endothelial cells (an endothelium). And this endothelium is what prevents coagulation of blood when traveling through native vasculature. Therefore, creating material systems that foster a confluent and functioning endothelial cell layer is an attractive strategy for generating devices with improved interactions with blood.<sup>2</sup>

Traditionally, mature endothelial cells are used for this purpose. However, a circulating population of endothelial precursor cells has recently been identified. These endothelial progenitor cells can be isolated through peripheral blood, expanded *ex vivo*, and generate large numbers of cells with endothelial phenotype. The outgrowth of the progenitor cells is referred to as endothelial colony forming cells (ECFC).

In this work, we illustrated that substrate topography can improve endothelial cell spreading and proliferation on a biomaterial surface.<sup>3</sup> We then proposed generating a surface that specifically attaches ECFC with the aim of

developing surfaces that can be rapidly endothelialized. In order to achieve this goal, we screened a phage display library to identify polypeptide ligands that bind with high specificity to ECFC but not other commonly occurring blood cell types. These peptides were then covalently immobilized onto a non-fouling biomaterial.<sup>4</sup> The resulting material was able to specifically bind the ECFC but not other cell types including mature endothelial cells (**Fig. 1**).<sup>5,6</sup>

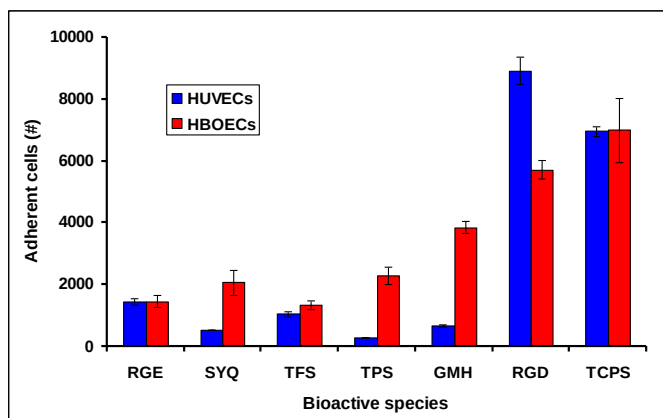


**Figure 1.** ECFC adhesion to non-fouling biomaterials functionalized with novel peptide ligands identified through phage display.

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## Polyethylene as a suitable precursor material for future carbon fibre production

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Carbon fibre impresses with its excellent mechanical properties and low density. Due to the complex production process, production costs (15 – 25 €/kg) are high and carbon fibre is not appropriate for mass usage in the automotive or energy sector (renewable energy) so far <sup>1</sup>. The main cost factor (50 % to 65 % of the carbon fibre production costs) is the production of the polyacrylonitrile (PAN) precursor. These high costs mainly arise because of the complex solvent spinning procedure, the solvent's preparation and PAN-Copolymer's high commodity price (2 – 5 €/kg) <sup>2</sup>. Alternative polyolefin based precursors are a possibility to reduce costs for the carbon fibre production and increase the value added of polyolefins. Especially polyethylene (PE) is, due to its high availability, the low material price (1 – 1.5 €/kg) and the good suitability as precursor, a promising material <sup>3,4,5</sup>. Furthermore, the production costs could be reduced as a melt spinning process is used to produce the PE-precursor instead of a solvent spinning process to produce the PAN-precursor. By the use of a polyethylene based precursor more than 30 % of the carbon fibre production cost can be economized, compared to the costs of a PAN based carbon fibre <sup>6</sup>. Since the complex and cost-effective solvent's preparation does not apply, melt spinning is faster (up to 6,000 m/min), more energy efficient and more environmentally friendly. First investigations on polyolefin based carbon fibres took place in the early 1970's <sup>7</sup>. The mainly resulting conclusions in this research show that the process of sulphonation might be to cost intensive due to high investment costs, handling of high amounts of sulphuric acid and related exhaust gas treatment <sup>8</sup>.

This presentation shows the manufacturing of PE based carbon fibres by sulphonation process and the investigation of the mechanical properties of the obtained sulphonated and carbonized fibres. Furthermore a possible process setup including a sulphuric acid recovery and a production and investment cost calculation for an industrial scale process is presented. As result PE based carbon fibres presents a possible low cost alternative to commercial PAN based carbon fibres.

<sup>1</sup> M. Warnecke, A. De Palmenaer, D. Veit, G. Seide, T. Gries, *Faserstofftabelle Carbonfasern*. **2013** Aachen, Shaker Verlag

<sup>2</sup> C. Wilms, *Kosteneffizientes Produktionsverfahren zur Herstellung von Carbonfaser-Precursoren*. **2014** Aachen, Shaker Verlag

<sup>3</sup> Platts McGraw Hill Financial, *Polymerscan*. **2013**, Vol. 36, Issue 25, pp. 1-32

<sup>4</sup> J. Morales, *Polyethylene Global Overview SPI Flexible Film & Bag*. **2013**, Nashville: IHS: SPI Flexible Film and Bag Conference

<sup>5</sup> E. Sagel, *Polyethylene Global Overview*. **2012**, Mexico city: IHS: Expo Foro Pemex

<sup>6</sup> F. Drissen, A. De Palmenaer, *Marktübersicht und Erstellung eines Kostenmodells für die Polyolefin-basierte Precursorherstellung*, **2014**, Aachen: Eigenverlag

<sup>7</sup> S. Horikiri, J. Iseki and M. Minobe, *Process for Production of Carbon Fibre*. US Patent 4070446. **1978**. Available online: <http://www.google.com/patents/US4070446> (accessed on 24 February 2015)

<sup>8</sup> B. E. Barton, J. T. Patton, E. J. Hukkanen, M. T. Bernius, *Two-step sulphonation process for the conversion of polymer fibers to carbon fibers*. Dow Global Technologies LLC, WIPO/PCT, WO002014011460A1, **2014**

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# Design and Preparation of Inverse Polymerized High Internal Phase Emulsions (HIPEs) Stabilized by Amphiphilic Diblock Copolymers

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The design of macroporous polymer monoliths that possess a highly interconnected structure represent a useful class of materials for potential use in numerous fields, such as separation science, tissue engineering, and catalytic supports, amongst others. One of the most common methods towards producing such structures is through the polymerization of a high internal phase emulsion (HIPE), usually termed a polyHIPE. HIPEs are commonly stabilized via commercial non-ionic surfactants, and are typically used to produce water-in-oil (w/o) HIPEs to afford the polymerization of hydrophobic monomers.

In this work, we report the preparation of hydrophilic, ‘inverse’ (i.e. oil-in-water, o/w) HIPEs stabilized by amphiphilic diblock copolymers prepared by reversible addition fragmentation chain transfer (RAFT) polymerization for the first time.<sup>1</sup> The resulting monoliths were characterized by EDX-SEM, Raman and FTIR spectroscopies to demonstrate full coverage of the walls of the material with RAFT moieties. This approach has two main benefits; rational design of the stabilizer affords polyHIPEs with different porous structures, and retention of the trithiocarbonate end-group allows for further surface polymerization and modification. When prepared *in situ* within a narrow internal diameter capillary, these materials demonstrate applicability as a stationary phase in high pressure liquid chromatography, demonstrating promising performance for small molecule separation.

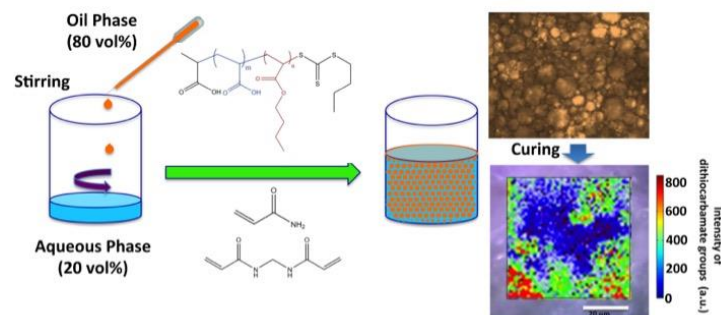


**Figure 1.** Schematic of inverse (oil-in-water) polyHIPE production stabilized by an amphiphilic block copolymer.

- (1) Khodabandeh, A.; Dario Arrua, R.; Desire, C. T.; Rodemann, T.; Bon, S. A. F.; Thickett, S. C.; Hilder, E. F. *Polymer Chemistry* **2016**, *7*, 1803.

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## Photo-Induced Self Assembly of Block Copolymers

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Block copolymers can self-assemble into well-defined, nanoscale morphologies, which are typically isotropic and lack long range order. Chemical or physical templates generated through optical lithography can direct self-assembly to create morphologies with enhanced long range order and fashion them into hierarchical patterns. This ability allows the patterns formed to be optimized to be more suitable for a desired application. Alternatively, the incorporation of stimuli-responsiveness into these materials has the potential to exert similar control over long range order and pattern complexity, while also creating dynamic nanostructures with expanded functionality. Here, photo-responsive block copolymers are synthesized and their ability to undergo a change in morphology triggered by light exposure is investigated. This will allow direct spatial control to be exerted over self-assembled nanostructures produced in thin block copolymer films.

Photo-responsive block copolymers were synthesized through successive RAFT polymerisations and post-polymerisation modification reactions to yield poly(benzyl methacrylate)-*b*-poly(*o*-nitrobenzyl methacrylate). When the *o*-nitrobenzyl group is exposed to UV light, it degrades to reveal a carboxylic acid functional group, allowing the polymer to be switched from a structure with two aromatic blocks and low segregation strength to an aromatic and polar acid block with a large segregation strength and induce self-assembly as shown in **Figure 1**.

Samples of this material were exposed to UV radiation for varying amounts of time to achieve different degrees of *o*-nitrobenzyl group deprotection and different levels of segregation strength as a result. Small Angle X-ray Scattering was used to characterize the evolution of morphology during this process, taking advantage of the high sample throughput achievable through the use of a synchrotron X-ray source to characterize a large number of samples with fine resolution between their levels of UV exposure. By mapping the transition in such fine resolution and for a range of different molecular weights and block ratios targeting different morphologies, we have been able to construct a phase diagram describing the system's transition from a disordered to well-ordered morphology with increasing UV exposure.

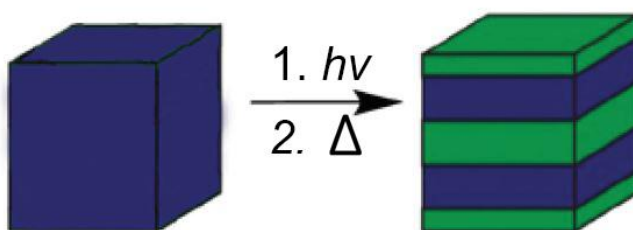


**Figure 1.** Exposure to UV radiation induces an increase in the  $\chi$  parameter of the initially disordered block copolymer, allowing it to self-assemble following annealing.

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## Stimuli-responsive dynamic polymers and their applications

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Dynamic bonds are defined as any type of bond that can be formed and broken selectively under equilibrium conditions. The incorporation of dynamic bonds into polymeric materials gives the polymer the dynamic properties. The term dynamic bonds include the two categories supramolecular interactions and dynamic covalent bonds. Dynamic covalent bonds are favored in the design of reversible polymers as they can be triggered only by a specific stimulus. There are different dynamic covalent bonds that can be triggered by heat, chemicals, and light. Hence, the type of stimulus can be controlled when designing the dynamic polymer. Thymine, one of the nucleic bases in DNA, exhibits the ability to form dynamic covalent bond. The photo-dimerisation of thymine, a UV-induced  $2\pi + 2\pi$  photo-cyclisation, results in the covalent dimerisation of adjacent thymine and disrupts the helical structure of DNA. This process has been linked with the development of certain forms of skin cancer. Photo-crosslinking of thymine occurs with irradiation at wavelengths above 270 nm. Crosslinking is reversed either by irradiation below 249 nm or enzymatically. Our research exploits this dynamic covalent reaction, reversible dimerization of thymine, to develop a novel, photo-responsive reversible polymer system.

We have used the reversible  $[2\pi + 2\pi]$ -cycloaddition of a bioinspired bis-thymine monomer to topochemically synthesize a polymer. The polymer can be fully photo-depolymerized to the monomer, and then reversibly and repeatedly photo-polymerized and photo-depolymerized. This is the first demonstration of complete photo-depolymerization and subsequent monomer recycling using this mechanism. The design and synthesis of various di-thymine monomers, determination of monomer crystal structures, and characterization of the photoproducts using Nuclear Magnetic Resonance, UV-Visible Spectroscopy, Gel-Permeation Chromatography, and other polymer characterization techniques will be discussed in the presentation.

We also have used above photo-reversible reaction to create self-healing polymers. The self-healing property of the new polymers using crosslinkers with photo-reversible unit was shown by complete elimination of cracks after the process of photo-reversible reactions. The appropriate conditions required to heal surface scratches of a micrometer size-scale were also studied.

<sup>1</sup> P. Johnston, C. Braybrook, K. Saito, *Chemical Science*, **2012**, 3, 2301-2306.

<sup>2</sup> P. Johnston, D. Wheldale, C. Braybrook, K. Saito, *Polymer Chemistry*, **2014**, 5, 4375-4384.

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# Enhanced Carbonisation Efficiency of Polyacrylonitrile by Reinforcement with Spinifex Nanocellulose

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Ultra-lightweight and high strength carbon fiber composites have recently found considerable use in the automotive and aerospace sectors. However due to the high cost of the polyacrylonitrile (PAN) precursor, as well as the energy input required for the carbonisation process, uptake of carbon fibers in other areas have been severely limited. The use of nanoparticles and nanofibers to reinforce the polyacrylonitrile precursor offers a new avenue for improving the efficiency of the carbonisation process to reduce costs. This work involves investigating new high-performance carbon fibers prepared from PAN composite precursors reinforced with high-aspect ratio nanocellulose fibers derived from Australian spinifex grass<sup>1</sup>. To this end, the effect of a spinifex nanocellulose reinforcing agent on PAN-based carbon nanofibers was explored by electrospinning composite dispersions of PAN and nanocellulose, followed by carbonisation of the nanofibers at 1200°C. Raman spectroscopy and X-ray diffraction characterisation of the carbon nanofibers showed that the nanocellulose additive incorporated into the precursor at specific concentrations led to an improvement in the crystallinity and graphitic carbon structure (Fig 1) – properties which typically increase with higher carbonisation temperature. This result successfully demonstrates the improved carbonisation efficiency of PAN without the need to increase heat input. A potential mechanism for this accelerated carbonisation is thought to be due to a molecular alignment effect on the bulk PAN matrix, induced by the high-aspect ratio nanocellulose fibers. This work will provide the basis for future thermomechanical studies on larger-scale wet spun nanocellulose-enhanced composite carbon fiber precursors.

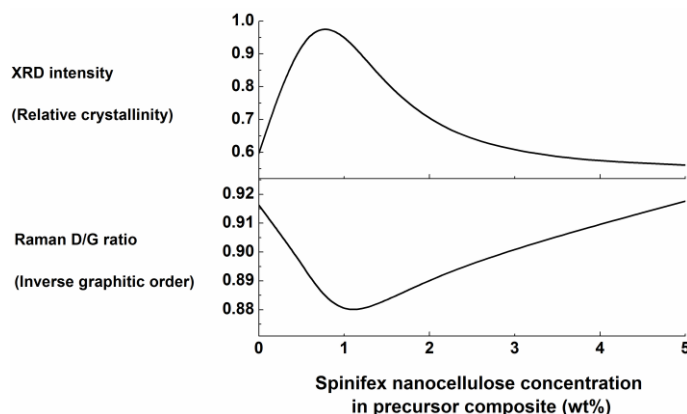


**Figure 1.** The effect of spinifex nanocellulose additives on the graphitisation efficiency of electrospun PAN nanofiber precursors.

<sup>1</sup> Amiralian, N.; Annamalai, P.K.; Memmott, P.; Taran, E.; Schmidt, S.; Martin, D.J. Easily deconstructed, high aspect ratio cellulose nanofibres from *tridodia pungens*; an abundant grass of australia's arid zone. *RSC Advances*, 2015, 5, 32124-32132.

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## Development of oil palm trunk fiber and polyester fiber composites for sound absorption materials

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In this research, oil palm trunks of 25 year-old trees which cannot be extracted oil from fruit bunch were utilized as fibers to reinforce composite for sound absorption materials. Oil palm trunk fibers were extracted by chemical treatment using 25 %w/w NaOH and then bleached by hydrogen peroxide. Sound absorption materials were formed as non-woven fabric by thermal bonding method using 2 formulas. First sound absorber composed of 60% of oil palm trunk fibers and 40% of low melt polyethylene terephthalate (LPET) fibers with composite thickness of 30 mm and weight per area of 2000 g/m<sup>2</sup> (P1). Second one composed of 50% of oil palm trunk fibers, 25% of polyester fibers type polyethylene terephthalate (PET) and 25% of LPET with composite thickness of 15 mm and weight per area of 1000 g/m<sup>2</sup> (P2). Two times increasing of thickness and weight per area of the composite led to two times increasing of noise reduction coefficient (NRC) and thermal resistance. In addition, tensile strength, air permeability, and sound absorption coefficient for all frequency also increased. In order to study effect of composite thickness and weight per area, oil palm trunk fiber composites of P2 were formed with thermal bonding method to composites with two times of thickness and of weight per area. Both composites provided slight increasing of sound absorption. Increasing of composite thickness yielded to increase thermal resistance. Meanwhile, increasing of weight per area affected on decreasing of thermal resistance and air permeability. In order to improve sound absorption efficiency, bilayer technique of PET needle punching nonwoven fabric (2 mm of thickness) and the oil palm trunk fiber composites were formed by heat press for two composites (LP1 and LP2). It is found that bilayer technique of PET nonwoven fabric on oil palm trunk fiber composites improved composite strength and sound absorption efficiency compared to two formulas of the oil palm trunk fiber composites.

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# High internal phase emulsions stabilized by functionalized copolymer: Effect of emulsion formulation and RAFT end-group on the pore structure of polyHIPEs

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High internal phase emulsions (HIPEs) are concentrated mixtures of liquid droplets dispersed in another liquid, defined by a minimum droplet volume fraction of 74%. These HIPEs are commonly stabilized by commercially available non-ionic surfactants, where these amphiphilic species decrease the interfacial tension between the oil and the aqueous phases, allowing emulsification. However stabilization of a new system often requires a different choice of surfactant.

Novel amphiphilic polymeric emulsifiers with versatile synthesis and ease of functionalization are highly desired, as this may allow the stabilization of multiple systems by simply altering the ratio of hydrophilic to hydrophobic portions of the molecule. The reversible addition-fragmentation chain transfer (RAFT) process allows the synthesis of such amphiphilic polymers, with the most common method through sequential polymerization of two monomers.

In this study, we illustrate the potential of RAFT-derived diblock copolymers as emulsifiers for water-in-oil HIPEs, in which the oil phase contained styrene (Sty) and divinylbenzene (DVB). Poly(styrene)-b -poly(acrylic acid) copolymers of varying block ratios were synthesised with the use of the chain transfer agent 2-[[[(butylsulfanyl)-carbonothioyl]sulfanyl]propanoic acid (PABTC). These were then evaluated as the sole emulsifier for a water-in-monomers HIPE. The presence of these species allowed the successful preparation of a polyHIPE upon polymerization. The pore structure of polyHIPEs was closed or non-droplet shaped, depending on the amount of the cross-linker monomer used in the oil phase.

The FTIR mapping technique confirmed the presence of the C=O groups in the same physical location as the walls of the polyHIPE voids, which are due to the carboxylic group of the acrylic acid segment (**Fig.1**). Further study shows that by removing the RAFT-end group of the amphiphilic macro-RAFT agent, the obtained polyHIPE possess an open structure within voids. Novel technology was developed to allow straightforward functionalization of the obtained polyHIPE via surfactant-assisted functionalization strategy as a new functionalized porous media.

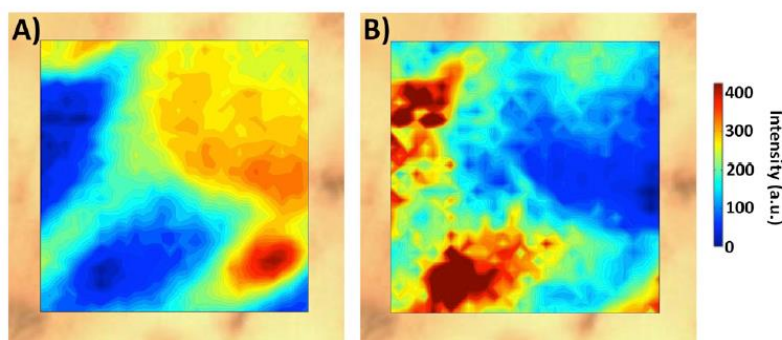


**Figure 1.** A) FTIR mapping based on signal to baseline from 2800 to 3000  $\text{cm}^{-1}$  (dark blue regions are void locations within the polyHIPE). B) FTIR mapping is based on the carboxylic peak at 1650  $\text{cm}^{-1}$  of macro-RAFT agent on the surface of polyHIPE at the same area.

# Integration of DNA with polymers

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The coupling of DNA with polymers offers an entirely new approach to developing unique applications in genotyping, diagnostics, synthetic biomaterials and enhanced polymerase chain reactions. This talk will introduce the fabrication of polymer/nucleotide polymers both with 3' or 5' nucleotides. We show attempts to ligate these nucleotides in order to use the polymer template in DNA amplification.

Thermo-responsive polymers, such as those based on *n*-isopropyl acrylamide (NIPAAm), have been studied extensively for bioseparation of dilute biomarkers due to their sharp phase transition at the biologically relevant lower critical solution temperature (LCST) of 32 °C. Circulating nucleic acids (cNAs) are emerging as potential biomarkers for early stage cancer diagnosis and treatment of residual disease. However, they are often found in very low levels circulating in a patient's blood and as a result require substantial enrichment to be useful in a diagnostic assay. cNAs are considered to be a dilute biomarker. To successfully detect cNAs tuneable stimuli response (TSR) DNA capture probe (DCP) polymers, using pNIPAAm, are being investigated for their utility in sensitive diagnostic applications. Here, we use RAFT polymerisation to design; tuneable thermo-responsive polymers that have a site specific moiety, a strained alkyne, used for conjugation to model DNA probes. The specificity and simplicity of the strained "click" reaction has the added benefit of eliminating the need for metal catalysts; normally required for traditional cycloaddition chemistry. Tuning of the LCST of pNIPAAm was achieved by incorporation of hydroxyl methyl acrylamide (HMAAm) (0-12.5%) monomer to make a range of pNIPAAm-co-HMAAm copolymers. The polymers displayed a tuneable LCST (**Fig 1(a)**). After functionalisation of the polymer with a strained alkyne, DNA capture probes were attached (**Fig 1(b)**) via a strained alkyne-azide reaction, to enable hybridisation, separation and enrichment of specific cNA transcripts with temperature modulation (**Fig 1(c)**).

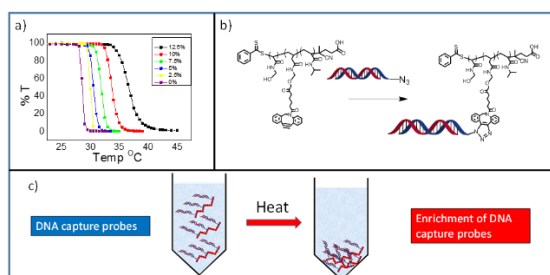


**Figure 1.** (a) LCST measurement of 0-12.5% pNIPAAm-co-HMAAm, (b) Conjugation of strained alkyne functionalised (pNIPAAm-co-HMAAm) with DNA capture probe via alkyne-azide cycloaddition and (c) Schematic diagram of biomarker enrichment process using pNIPAAm-co-HMAAm-co-strained alkyne-DNA capture probe stimuli response polymer.

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## Bioactive Nano- and Microstructures from Self-assembling Amphiphilic Glycopolymers

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Carbohydrates mediate many processes in biology, particularly those that involve contacts between surfaces (for example cell-cell and cell-virus particle contacts). Nature overcomes the intrinsic weakness of individual carbohydrate-protein (lectin) interactions by means of multivalency, of both the carbohydrate ligand and the lectin. Thus the preparation of multivalent, high-affinity carbohydrate-bearing macromolecules, including glycopolymers, is an attractive strategy for investigating and inhibiting such interactions, leading to applications in therapeutics, diagnostics, protein purification and other areas of biotechnology. We present our work on the development of biologically active, amphiphilic glycopolymers from simple monosaccharide-containing monomers using RAFT polymerization. Depending on the exact molecular characteristics of the polymers and the self-assembly process employed, the polymers can be assembled into a variety of nano- and microstructures, including micelles, nanoparticles and polymersomes. The presentation will discuss two recent projects in our lab:

1. The production of self-assembled glyconanoparticles for the delivery of antibiotics ('glyconanobiotics'); and
2. The development of glycosylated giant polymersomes as simple minimal cell mimics.

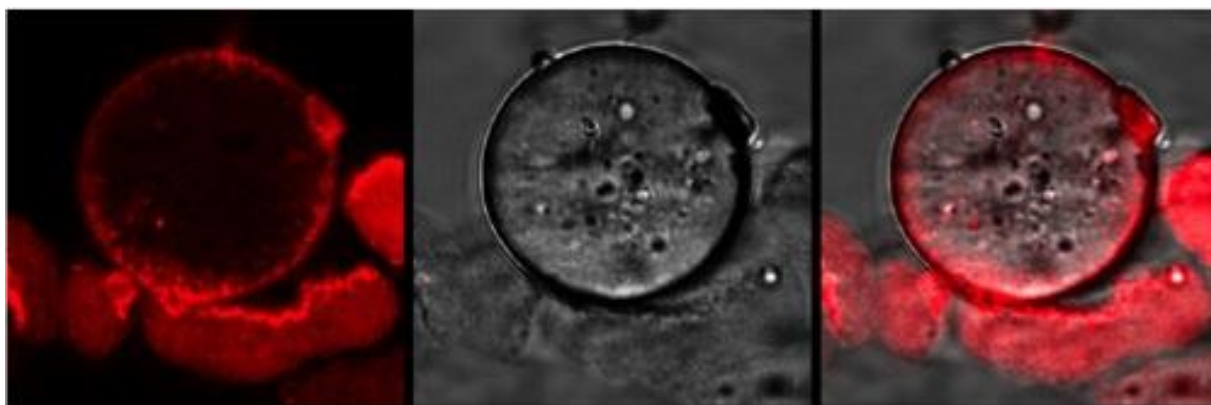


**Figure 1.** Light microscopy images of a glycosylated giant polymersome. Left to right; red fluorescence channel (rhodamine B octadecyl ester perchlorate dye); middle: optical channel; right: overlay.

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# Shaping the Future of Polymer Molecular Weight Distributions

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A polymer's molecular weight distribution (MWD) has a profound impact on its properties, from material strength and viscosity to changes in the phase behavior of block copolymers.<sup>1-3</sup> Dispersity is the most common measure of MWD and is described as the ratio of weight-average ( $M_w$ ) to number-average ( $M_n$ ) molecular weights. Importantly, dispersity only provides information on the relative breadth of molecular weights in a sample and is not a comprehensive description of the molar quantities of each chain size.<sup>2</sup> The exact shape of a MWD has been proposed to have a strong influence on polymer properties;<sup>5,6</sup> however, this hypothesis remains relatively unexplored. This presentation will detail the development of a modular synthetic strategy that provides deterministic control over the  $M_n$ , breadth, and composition of polymer MWDs and will examine the influence of MWD shape on polymer properties.<sup>7</sup>

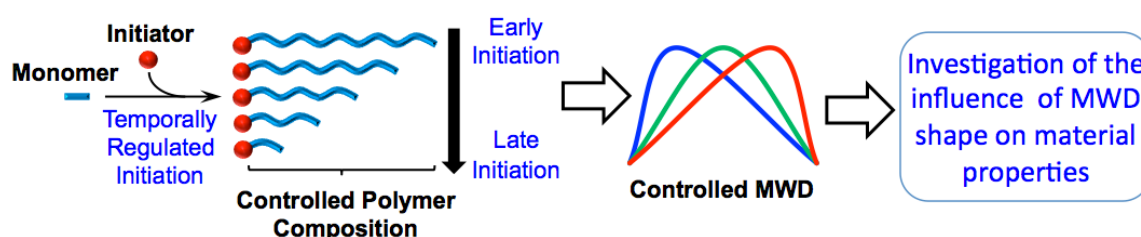


**Figure 1.** Deterministic control of polymer molecular weight distributions.

1. Nichetti, D.; Manas-Zloczower, I. *Polym. Eng. Sci.* **1999**, 39, 887–895.
2. Collis, M. W.; Mackley, M. R. *J. Non-Newtonian Fluid Mech.* **2005**, 128, 29–41.
3. Lynd, N. A.; Meuler, A. J.; Hillmyer, M. A. *Prog. Polym. Sci.* **2008**, 33, 875–893.
4. S. S. Rane and P. Choi, *Chem. Mater.*, 2005, **17**, 926–926.
5. Lynd, N. A.; Hillmyer, M. A.; Matsen, M. W. *Macromolecules* **2008**, 41, 4531–4533.
6. Cooke, D. M.; Shi, A.-C. *Macromolecules* **2006**, 39, 6661–6671.
7. Gentekos, D. T.; Dupuis, L. N.; Fors, B. P. *J. Am. Chem. Soc.* **2016**, 138, 1848–1851.

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# Systematic study of high molecular weight acrylamide polymerisations using aqueous REDOX initiated RAFT

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Due to the industrially relevant properties of high molecular weight ( $M_w$ ) polyacrylamides, developing controlled polymerisation strategies which deliver significant livingness to high  $M_w$  (>500 kDa) has received increasing interest. In 2014 Destarac et al.<sup>1</sup> synthesised polyacrylamide ( $M_w > 1,000$  kDa,  $D_M < 1.2$ ) using low temperature RAFT/MADIX gel polymerisation.

In this work, we used a systematic approach to optimise conditions for the polymerisation of acrylamide to a target  $M_w = 500$  kDa. We employed an automated parallel synthesiser (Chemspeed SWING XL) to quickly scan a range of temperatures, trithiocarbonate RAFT agents, redox initiator systems, CTA to initiator ratios and monomer concentrations.

Preliminary results indicate CTA3 (**Fig.1**) allows for excellent conversion ( $\geq 90\%$ ) and controlled polymerisation ( $D_M = 1.22 - 1.44$ ) across a range of polymerisation conditions over 24h. Other RAFT agents trialled resulted in less controlled polymerisations ( $D_M = 1.32 - 1.62$ ) or no polymerisation across the conditions studied. The APS/NaFS (Ammonium persulfate/ Sodium formaldehyde sulfoxylate) redox couple gave the most consistent and controlled polymerisation when using the Chemspeed synthesiser. Utilising a low oxidant concentration relative to CTA was shown to be the most significant variable for improving control of polymerisation (**Fig.1**). These results allow for the identification of ideal RAFT polymerisation conditions for controlled, high molecular weight polyacrylamides. These conditions can then be applied in the synthesis of advanced polymeric materials.

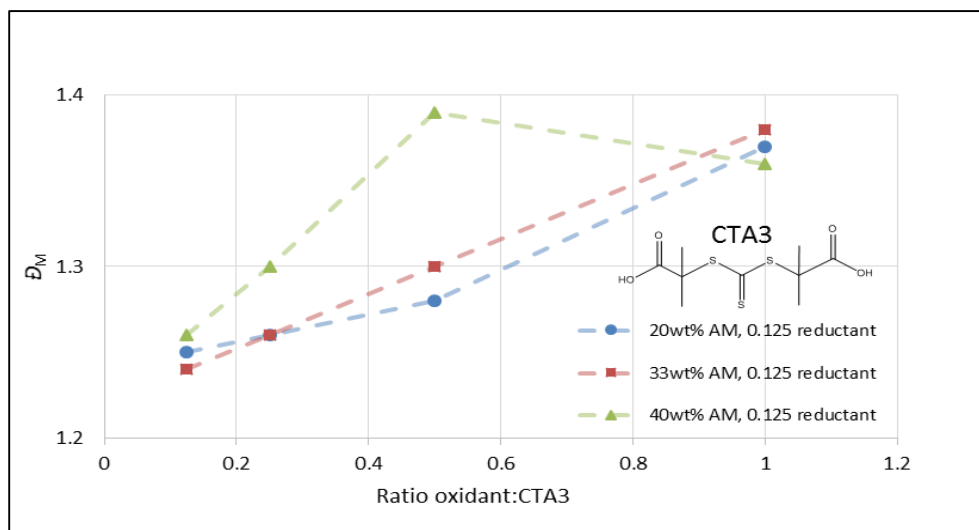


**Figure 1.** Effect of both oxidant and monomer concentration on dispersity for polyacrylamide with 500 kDa target molecular weight at 20°C (all conversions  $\geq 90\%$ ).

<sup>1</sup> E. Read, A. Guinaudeau, D. J. Wilson, A. Cadix, F. Violleau and M. Destarac, *Polym Chem-Uk*, **2014**, 5, 2202-2207

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# Prebiotic Chemistry Inspired Polymers for Coatings in Material Science Applications

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Christopher D. Easton, Aylin Koegler, Richard A. Evans.

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Prebiotic chemistry is the study of the chemistry required for the origin of life to have arisen.<sup>1</sup> Various polymers have been studied for many years as possible sources for the precursors of proteins and nucleic acids and so have been proposed potentially relevant to the origin of life. However, the application of prebiotic chemistry and specifically prebiotic polymers to materials science has not been reported.

Polymers of HCN have been proposed as one class of polymers of possible prebiotic relevance.<sup>2</sup> We have drawn upon the work investigating HCN polymers in the prebiotic field and applied it as a solution to material science challenges. Here we have used aminomalononitrile (**1**),<sup>3</sup> as a convenient precursor to HCN polymers, which spontaneously polymerizes to give a brown complex nitrogenous polymer (**Fig. 1**). We found that the polymerization, carried out in simple aqueous buffered solutions, can be used to coat a wide range of organic and inorganic substrates.<sup>4,5</sup> The robust, non-cytotoxic coatings also provide outstanding cell attachment, thereby providing access to a range of biomedical applications. This discovery opens the door to the re-examination of 60 years of experimental prebiotic chemical research from the point of view of its application to polymer, material and life sciences. Recent work on control of morphology and interaction with biology will be presented.

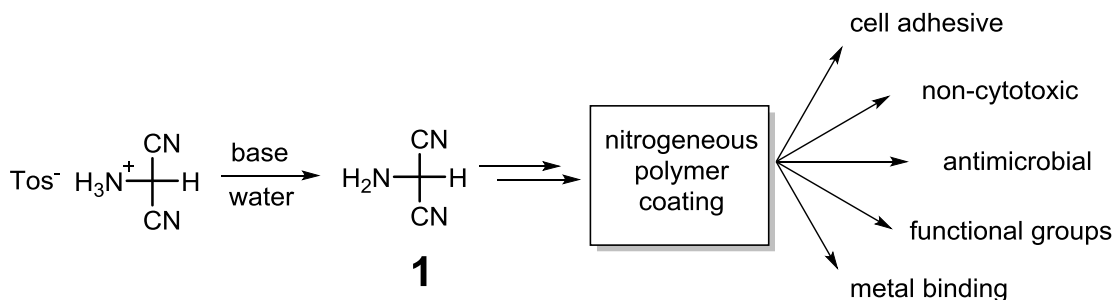


**Figure 1.** Polymerization of aminomalononitrile **1** to produce a highly nitrogenous polymer coating with a variety of applications.

<sup>1</sup> S. L. Miller, *Science* **1953**, *117*, 528; R. M. Lemmon, *Chem. Rev.* **1970**, *70*, 95; J.-F. Lambert, M. Sodupe, P. Ugliengo, *Chem. Soc. Rev.* **2012**, *41*, 5373; J. L. Bada, *Chem. Soc. Rev.* **2013**, *42*, 2186

<sup>2</sup> C. N. Matthews, R. E. Moser, *Nature* **1967**, *215*, 1230; C. N. Matthews, R. D. Minard, *Faraday Discuss.* **2006**, *133*, 393; J. P. Ferris, W. J. Hagan, *Tetrahedron* **1984**, *40*, 1093

<sup>3</sup> R. E., Moser, A. R. Claggett, C. N. Matthews, *Tetrahedron Lett.* **1968**, *9*, 1605

<sup>4</sup> Thissen H., Evans R.A., Koegler A. (2013) WO 2013/170308 A

<sup>5</sup> Thissen H., Koegler, A., Salwiczek, M., Easton, C.D., Qu, Y., Lithgow, T., Evans, R.A. *NPG Asia Materials* **2015**, *7*, e255; doi:10.1038/am.2015.122

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Personal History:

1992-present. CSIRO. Synthetic polymer chemist.

Adjunct Professor Swinburne University of Technology.

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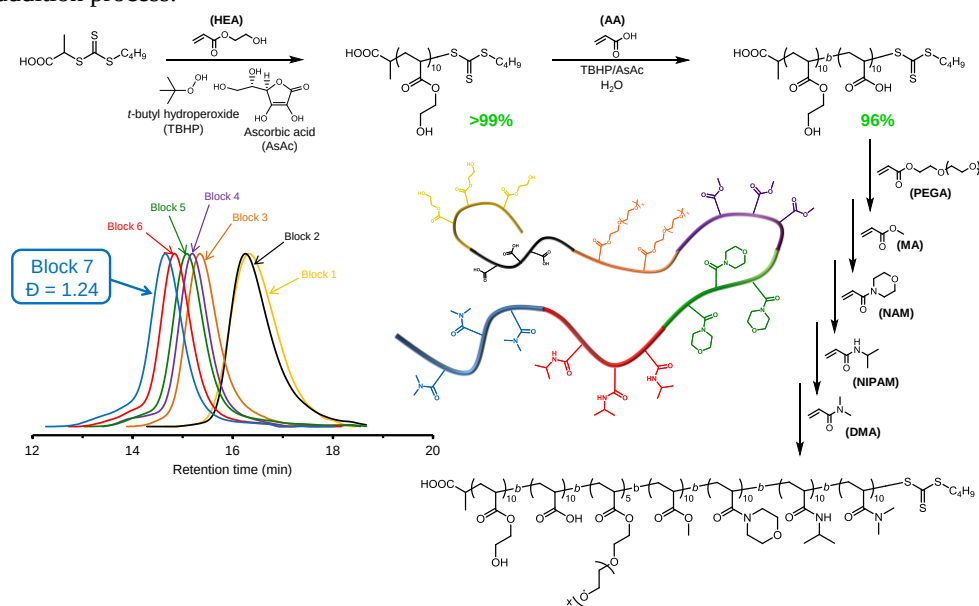
# Multiblock Copolymer Synthesis via Aqueous Room Temperature RAFT Polymerisation

Liam Martin<sup>1</sup>, Sébastien Perrier<sup>1, 2</sup>

<sup>1</sup>University of Warwick, CV47AL, United Kingdom

<sup>2</sup>Faculty of Pharmacy and Pharmaceutical Sciences, Monash University, VIC3050, Australia

Progress in the understanding of RAFT has granted the ability to push polymerisation of certain monomers to near-quantitative conversion (>99 %) whilst retaining an extremely high fraction of the  $\omega$ -chain end.<sup>1-3</sup> This has been exploited for the preparation of well-defined multiblock copolymers using a simple, high-yielding one-pot sequential-addition process.



**Scheme 1 – Demonstrating the one-pot synthesis of an acrylate/acrylamido multiblock copolymer via room temperature aqueous RAFT polymerisation.**

We have investigated the use of redox initiation as an approach for aqueous RAFT polymerisation at ambient temperatures in order to incorporate certain challenging monomer classes such as acrylates (side reactions at elevated temperatures) and polymers exhibiting low LCST behavior in water (e.g. NIPAAm, DEA) into multiblock copolymer “sequences”.<sup>4</sup> We demonstrated aqueous multiblock copolymer synthesis at room temperature by preparing a range of acrylamido, acrylate and acrylate/acrylamido multiblock copolymers with relatively narrow molar mass distributions ( $\bar{D} \leq 1.3$ ) using the *t*-butyl hydroperoxide/ascorbic acid redox couple. We are also investigating the use of different redox couples and the oxidizing agent/reducing agent stoichiometries to control the polymerisation rates, allowing full monomer conversion to be achieved with lower initiator concentrations (increased “livingness”) whilst reducing polymerisation times.

1. G. Gody, T. Maschmeyer, P. B. Zetterlund and S. Perrier, *Nat Commun*, 2013, **4**.
2. G. Gody, T. Maschmeyer, P. B. Zetterlund and S. Perrier, *Macromolecules*, 2014, **47**, 639.
3. G. Gody, T. Maschmeyer, P. B. Zetterlund and S. Perrier, *Macromolecules*, 2014, **47**, 3451.
4. L. Martin, G. Gody and S. Perrier, *Polym. Chem.*, 2015, **6**, 4875.

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# Exploiting Photochemical Selectivity in Macromolecular Synthesis

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Installing chemical functionality at precise locations within complex macromolecules as well as onto three dimensional direct laser written constructs in the mildest possible fashion (visible light) and without any catalyst becomes possible with powerful light driven, quantitative ligation protocols.<sup>1-6</sup> Advanced light induced processes enable the parallel encoding of multiple molecules onto predefined locations on 3D microstructures in one step<sup>7</sup> as well as the wavelength dependent addressing of specific parts of a macromolecule for its highly orthogonal functionalization ( $\square$ -orthogonal photochemistry).<sup>8</sup> The lecture will highlight how macromolecular ligations can be directed in their selectivity by the application of photonic fields,<sup>9</sup> how light can be used as an orthogonal trigger in macromolecular chain folding,<sup>10</sup> how the high efficiency of light driven chemistry can be exploited for the synthesis of sequence defined macromolecules<sup>11</sup> or explore – time permitting – how diffraction unlimited laser lithography may become possible based on photo-click chemistry concepts.<sup>12</sup>

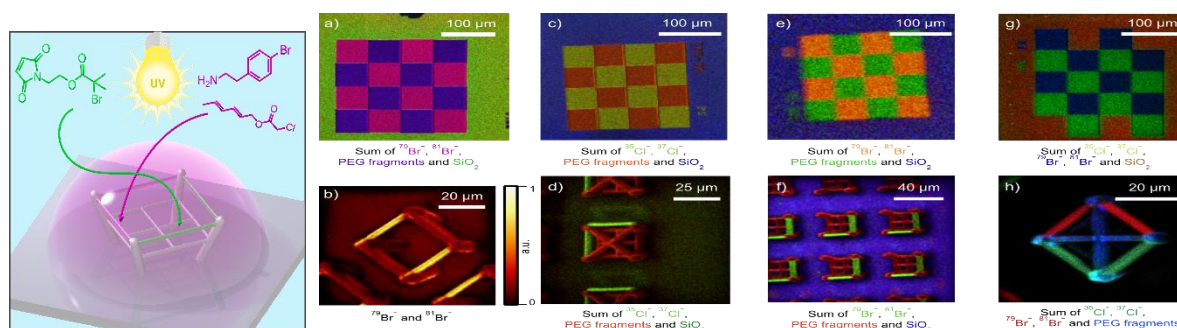


**Figure 1.** Encoding three dimensional direct laser written scaffolds in specific pre-defined positions with chemical functionality via a self-directed light induced molecular sorting process (ToF-SIMS images shown).<sup>7</sup>

[1] Delaittre, G.; Goldmann, A. S.; Mueller, J. O.; Barner-Kowollik, C. *Angew. Chem. Int. Ed.* **2015**, 54, 11388–11403. [2] (a) Mueller, J. O.; Schmidt, F. G.; Blinco, J. P.; Barner-Kowollik, C. *Angew. Chem. Int. Ed.*, **2015**, 54, 10284–10288; (b) Lederhose, P.; Wuest, K. N. R.; Barner-Kowollik, C.; Blinco, J. P. *Chem. Commun.* **2016**, 52, 5928–5931. [3] Altintas, O.; Glassner, M.; Rodriguez-Emmenegger, C.; Welle, A.; Trouillet, V.; Barner-Kowollik, C. *Angew. Chem. Int. Ed.* **2015**, 54, 5777–5783. [4] Pauloeuhl, T.; Welle, A.; Bruns, M.; Linkert, K.; Börner, H. G.; Bastmeyer, M.; Delaittre, G.; Barner-Kowollik, C. *Angew. Chem. Int. Ed.* **2013**, 52, 9714–9718. [5] Frick, E.; Anastasaki, A.; Haddleton, D. M.; Barner-Kowollik, C. *J. Am. Chem. Soc.* **2015**, 137, 6889–6896. [6] Pauloeuhl, T.; Delaittre, G.; Winkler V.; Welle, A.; Bruns, M.; Börner, H. G.; Greiner, A. M.; Bastmeyer, M.; Barner-Kowollik, C. *Angew. Chem. Int. Ed.* **2012**, 51, 1071–1074. [7] Claus, T. K.; Richter, B.; Hahn, V.; Welle, A.; Kayser, S.; Wegener, M.; Bastmeyer, M.; Delaittre, G.; Barner-Kowollik, C. *Angew. Chem. Int. Ed.* **2016**, 55, 3817–3822. [8] Hiltebrandt, K.; Pauloeuhl, T.; Blinco, J. P.; Linkert, K.; Börner, H. G.; Barner-Kowollik, C. *Angew. Chem. Int. Ed.* **2015**, 54, 2838–2843. [9] Hiltebrandt, K.; Elies, K.; d’Hooge, D.; Blinco, J.; Barner-Kowollik, C. *J. Am. Chem. Soc.* **2016**, in press. [10] Greb, L.; Mutlu, H.; Barner-Kowollik, C.; Lehn, J. M. *J. Am. Chem. Soc.* **2016**, 138, 1142–1145. [11] (a) Zydziak, N.; Feist, F.; Huber, B.; Mueller, J. O.; Barner Kowollik, C. *Chem. Comm.* **2015**, 51, 1799–1802; (b) Zydziak, N.; Konrad, W.; Feist, F.; Barner-Kowollik, C. **2016**, submitted. [12] Müller, P.; Barner-Kowollik, C.; Wegener, M. **2016**, submitted.

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Since 07/08 Full Professor of Macromolecular Karlsruhe Institute of Technology (KIT)

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Research interests: Light-triggered orthogonal chemistries, novel synthetic approaches to complex macromolecular designs, spatially and temporally controlled surface coding (2D, 3D), polymerization mechanisms, advanced polymer characterization

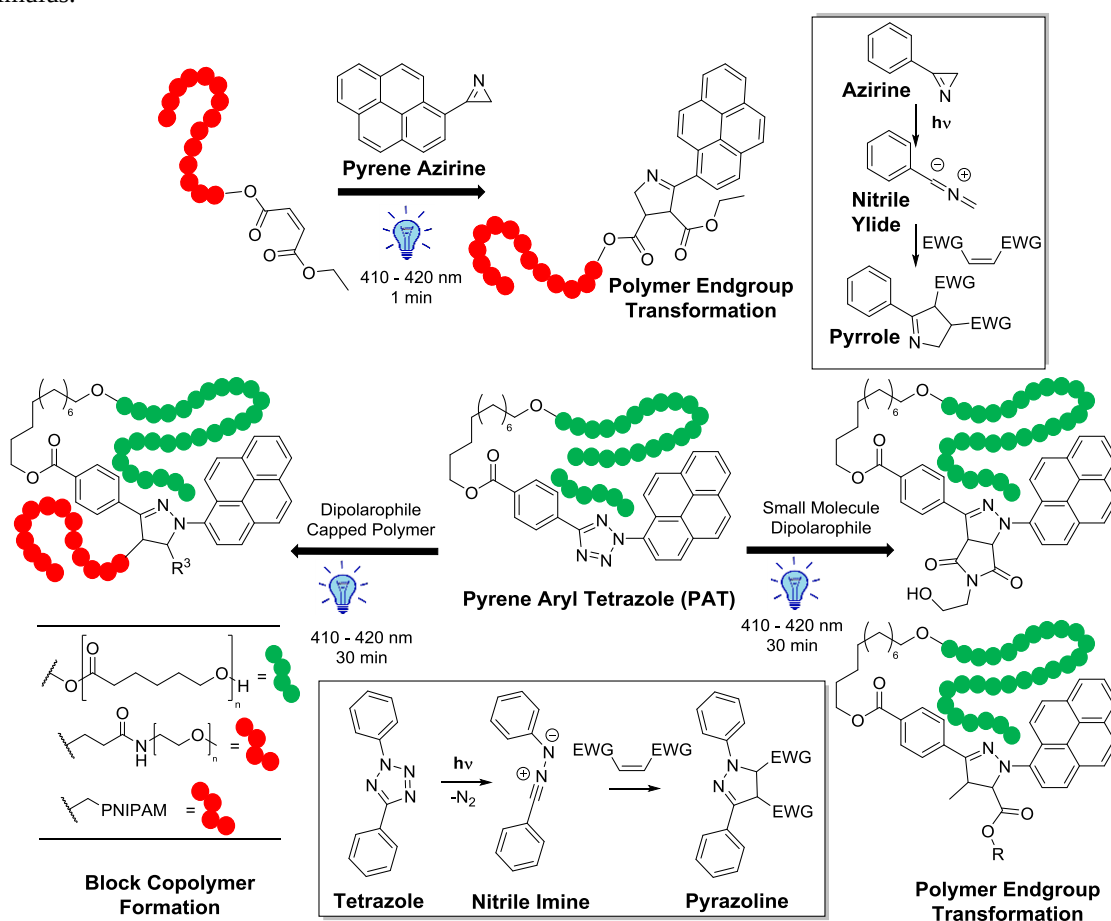
# Driving Polymer and Surface Modification Using Visible Light

Paul Lederhose,<sup>1,2</sup> Jan Mueller,<sup>2</sup> Christopher Barner-Kowollik<sup>1,2</sup> and James Blinco<sup>1</sup>

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Rapid and catalyst-free cycloaddition systems for visible-light-induced modification of polymers and surfaces are reported. Photo-reactive azirine<sup>39</sup> and tetrazole<sup>40</sup> moieties were designed to absorb light at wavelengths longer than 400 nm through incorporation of polyaromatic chromophores. Irradiation with low-energy light sources enabled quantitative ligation of the photoactivatable unit with functional polymeric substrates achieving full conversion with irradiation times of only 1 min at ambient conditions. Utilising hydroxyl functional derivatives it was also possible to attach the photoactivatable unit directly to polymer and surface substrates for the synthesis of AB block copolymers and patterned polymer grafted surfaces using visible light as the only stimulus.



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## Photocatalysts; An efficient Tool for the Control of the Polymer Architecture

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The emergence of efficient photocatalysts using transition metals, such as iridium and ruthenium, has been recently applied to perform organic reactions under visible light.<sup>1</sup> Such catalysts have been utilized to perform complex organic reaction. In this presentation, we present our recent results showing that these catalysts can be employed in polymer synthesis for the preparation of complex macromolecules and the post-modification of macromolecules under visible light. According to the nature of photocatalysts, different polymerizations can be achieved using different wavelengths. We demonstrated that a large range of photocatalysts can activate a photoinduced electron/energy transfer (PET) process, which initiates a reversible addition fragmentation chain transfer polymerization (RAFT). For instance, by selecting the photocatalysts, we were able to control the tacticity of the polymer and control the polymer architectures.<sup>2</sup>

1. Corrigan, N.; Shanmugam, S.; Xu, J.; Boyer, C. *Chemical Society Reviews* **2016**, DOI: 10.1039/C6CS00185H.
2. Xu, J.; Shanmugam, S.; Fu, C.; Aguey-Zinsou, K.F.; Boyer, C. *Journal of the American Chemical Society*, **2016**, 138 (9), 3094-3106

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## Photopolymerization of RAFT agents under visible light irradiation

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Reversible addition-fragmentation chain transfer (RAFT) radical polymerization has become one of the most prominent methods for synthesizing polymers of controlled molecular weight, low dispersity, defined chemical functionality, and complex macromolecular architecture. Combining such sophisticated polymerization protocols with the ability for regulation over growth of the polymer chain via an external stimulus has recently proven valuable, with light-driven photochemistry a particularly appealing approach. We have demonstrated that under visible light, typically used RAFT agents can be photochemically excited to induce a polymerization that can yield very well-defined, “living”, polymers.<sup>1</sup> Although the UV-activation of thiocarbonyl compounds in fact represents one of the earliest successes in control over a radical polymerization (i.e., the ‘photoiniferter’ process<sup>2</sup>), the activation of these compounds by visible light (**Fig. 1**) – a lower energy, safer, and more naturally abundant wavelength of irradiation – had not been reported until very recently.<sup>1,3</sup> In this talk the effect of employed wavelength, irradiation intensity, and choice of monomer/RAFT agent will be critically discussed with regards to their ability to afford well-defined polymers in a visible light photopolymerization system.<sup>4</sup> Some associated benefits of such a system devoid of exogenous (photo)initiators or (photo)catalysts will also be highlighted.

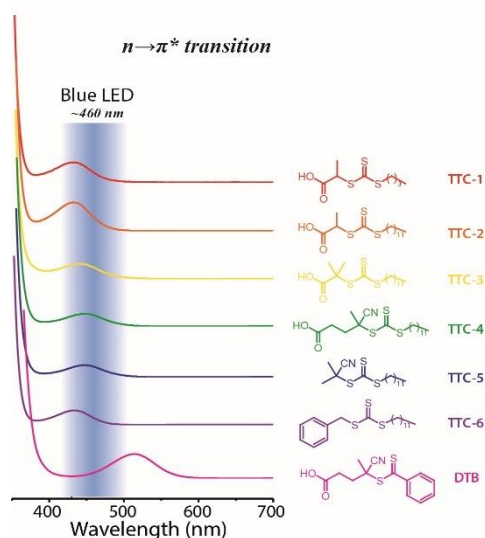


**Figure 1.** Emission-absorption overlap of several conventional RAFT agents with blue LED light source shows potential for visible light photoexcitation.<sup>4</sup>

<sup>1</sup>McKenzie, T. G. et al., *Macromolecules*. **2015**, *48*, 3864.

<sup>2</sup>Otsu, T., *J. Polym. Sci. A. Polym. Chem.* **2000**, *38*, 2121.

<sup>3</sup>Xu, J. et al., in *Controlled Radical Polymerization: Mechanisms*, American Chemical Society **2015**, *1187*, 247.

<sup>4</sup>McKenzie, T. G.. et al., *Polym. Chem.* **2016**, DOI: 10.1039/c6py00808a.

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Personal History:

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| Sep 2012- Mar 2016                                                                 | PhD candidate, Polymer Science Group, University of Melbourne |
| Mar 2016 – present                                                                 | Research Fellow, University of Melbourne                      |
| Research interests: radical polymerization, photochemistry, macromolecular design. |                                                               |

# Spontaneous Zwitterionic Copolymerization of Cyclic Imino Ethers: First Steps to Biodegradable and Biocompatible 2-Oxazoline Polymers?

Kristian Kempe

ARC Centre of Excellence in Convergent Bio-Nano Science and Technology,  
Monash Institute of Pharmaceutical Sciences, Monash University,  
381 Royal Parade, Parkville, VIC 3052, Australia

Five and six membered cyclic imino ethers, also better known as oxazolines and oxazines are highly functional compounds which have been proven to be potent monomers in cationic ring-opening polymerisations (CROP) to yield poly(2-oxazoline)s and poly(2-oxazine)s, respectively.<sup>1</sup> In particular poly(2-methyl-2-oxazoline) (PMeOx) and poly(2-ethyl-2-oxazoline) (PEtOx) have attracted significant attention in the last decades owing to their excellent biocompatibility and antifouling and stealth properties,<sup>2</sup> which has made them one of the top alternatives for the 'gold standard' poly(ethylene glycol) (PEG).

An alternative approach to prepare 2-oxazoline based polymer systems is the spontaneous zwitterionic copolymerisation (SZWIP). This polymerization is believed to occur via a zwitterionic intermediate which is formed upon the reaction between an electrophilic ( $M_E$ ) and nucleophilic monomer ( $M_N$ ) without the addition of an additional catalyst or initiator. Cyclic imino ethers act as  $M_N$  and when combined with a  $M_E$  containing acidic hydrogens they form poly(ester amide)s or more specific N-acylated poly(aminoester)s (NPAE).<sup>3,4</sup>

In this presentation the potential of cyclic imino ethers for the preparation of functional biocompatible and biodegradable materials will be discussed. Particular focus will lie on the synthesis and modularity of functional NPAE based macromonomers and the combination of CROP/SZWIP and reversible-deactivation radical polymerization techniques for the design of responsive comb/brush polymers.

<sup>1</sup> R. Hoogenboom, *Angew. Chem. Int. Ed.* **2009**, *48*, 7978-7994.

<sup>2</sup> L. Tauhardt, K. Kempe, M. Gottschaldt, U. S. Schubert, *Chem. Soc. Rev.* **2013**, *42*, 7998-8011.

<sup>3</sup> K. Kempe, P. A. J. M. de Jongh, A. Anastasaki, P. Wilson, D. M. Haddleton, *Chem. Commun.* **2015**, *51*, 16213-16216.

<sup>4</sup> K. Kempe, S. L. Ng, S. T. Gunawan, K. F. Noi, F. Caruso, *Adv. Funct. Mat.* **2014**, *24*, 6187-6194.

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2007 – 2011 PhD (FSU Jena, Germany; University of California, Santa Barbara, USA)

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2014 – 2016 Senior Research Fellow (University of Warwick, UK)

Since 2016 NHMRC-ARC Dementia Research Development Fellow (Monash University, Australia)

Research interests: stimuli-responsive materials, protein-repellent surfaces, polymer-peptide/protein conjugations, glycopolymers, defined macromolecular architectures: comb/star-shaped polymers, poly(2-oxazoline)s, poly(ester amides), nanoparticles, drug delivery, brain delivery, AD

# Peptide Decorated Polymeric Dendrimers and Their Biological Applications

Zhongfan Jia<sup>1</sup>, Derong Lu<sup>1</sup>, Michael, J. Monteiro<sup>1</sup>

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'Click' chemistry provides a method for designing and synthesising of highly branched macromolecules, such as polymeric dendrimers, from polymer building blocks.<sup>1</sup> These polymeric dendrimers contained both very different chemical composition and multiple functionalities. Moreover, with less generation, polymeric dendrimers showed similar or larger size to those small-molecular based dendrimers. Further modification of these dendrimers with amino acids or (oligo)peptides to the peripheral layer represented one form of peptidomimetics (i.e. a hybrid peptide/dendrimer construct) that has found use in biological applications.

Herein, we presented a pair of copper catalysed orthogonal 'click' reactions, copper catalysed alkyne-azide cycloaddition (CuAAC) reaction and single electron transfer-nitroxide radical coupling (SET-NRC) reaction.<sup>2</sup> This 'click' reaction pair showed rapid and highly efficient synthesis of G3 polymeric dendrimers.<sup>3, 4</sup> Attaching functional peptides and amino acid to the periphery of the dendrimers led to the applications of these peptidomimetics as therapeutic agents, such as vaccine<sup>5</sup> and ion channel block agents (Figure 1).<sup>6</sup>

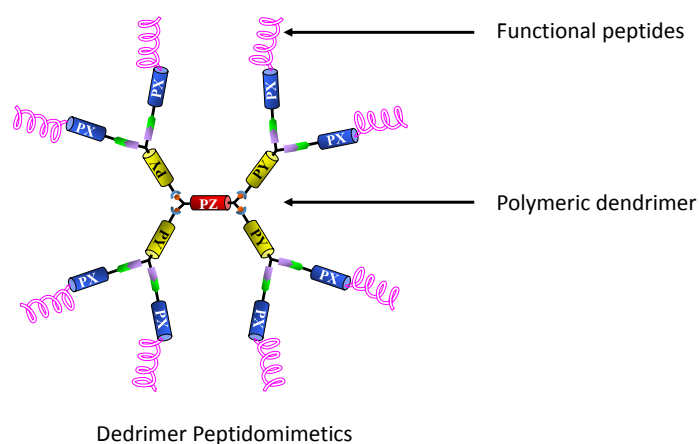


**Figure 1.** Synthesis of polymeric dendrimers as peptidomimetics for biological applications.

1. P. Wu, M. Malkoch, J. N. Hunt, R. Vestberg, E. Kaltgrad, M. G. Finn, V. V. Fokin, K. B. Sharpless and C. J. Hawker, *Chemical Communications*, 2005, DOI: 10.1039/B512021G, 5775-5777.
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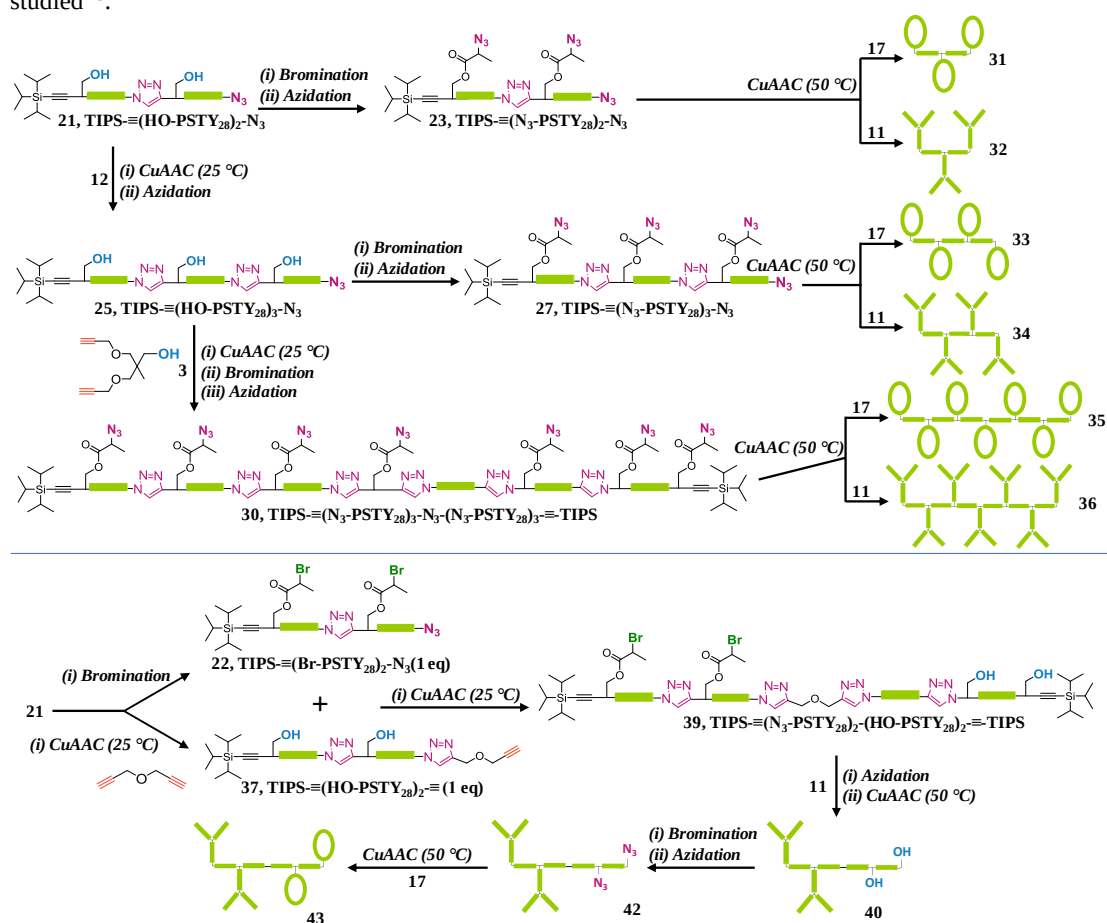
Research interests: Living Radical Polymerization, Complex Polymer Architecture and Polymer Nanostructure, Drug and siRNA Delivery, Organic Radical Polymer, Electrochemical Energy Storage

# Synthesis of Symmetrical and Asymmetrical Complex Polymer Architectures via Convergent “Graft to” Approach

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Complex polymer architectures including symmetrical cyclic brush and dendronized polymers and an asymmetrical cyclic and wedge dendron tailored architecture have been synthesized. The key synthetic strategy in this work involves the retention of the bromine group/s during CuAAC click reactions at room temperature (25°C) and at elevated temperature (50°C). This was achieved by modulating the activity of Cu(I) species to ensure faster coupling of azide and alkyne moieties and no bromine abstraction<sup>1</sup>. This provided absolute control over polymer building blocks, which lead to the construction of precisely controlled linear backbones containing equally spaced two three and seven pendent side-chain hydroxyl groups. The hydroxyl groups were azidated and alkyne functionalized cyclic and wedge dendron moieties were coupled to these azide groups via convergent “graft to” approach to afford cyclic brush and dendronized architectures. A linear backbone containing two equally spaced hydroxyl groups on one side and two azide groups on the other side was also synthesized. Sequential coupling of alkyne-functionalized wedge dendron and cyclic moieties to this backbone produced a mechanistically asymmetrical architecture. All these architecture were synthesized in high yield and good coupling efficiencies. The effects of these topologies on glass transition temperature  $T_g$  have been studied<sup>1-3</sup>.



Scheme 1: Synthesis of complex polymer architectures.

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# Engineering shape and core of polymeric nanoparticles via emulsion polymerization and temperature-induced morphological transformation

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Emulsion polymerization is an environmentally friendly technique for the synthesis of polymeric nanoparticles for a wide range of applications such as paint, surface coating, sensor technology, catalysis, and nanomedicine.<sup>1</sup> However, the control over both shape and core of polymeric nanoparticles via emulsion polymerization remains challenging. In this contribution to APS36, we present our recent works on using emulsion polymerization followed by temperature-induced morphological transformation (TIMT, see Fig. 1) to engineering both the shape and the core of polymeric nanoparticles.<sup>2,3</sup> These techniques pave the way for the preparation of a large library of nanoparticles with different shapes (sphere, cylinder, vesicle, and lamella) and various cores (e.g., polystyrene, poly(methyl methacrylate), poly(ethyl methacrylate), poly(propyl methacrylate), etc.) from novel biocompatible thermoresponsive polymers. The cloud point temperature of these thermoresponsive polymers could be predictably tuned by changing the hydrophilic contents. The novel thermoresponsive polymer is well-tolerated and suitable for the RAFT-mediated emulsion polymerization of styrene and methacrylates. The formed diblock copolymers have unimodal molecular weight distributions, low dispersities even at high conversion, and the ability to form different shapes with various cores. The ability to produce nanoparticles from low-toxicity materials with both spherical and non-spherical shapes opens the door to many novel applications including nanomedicine.

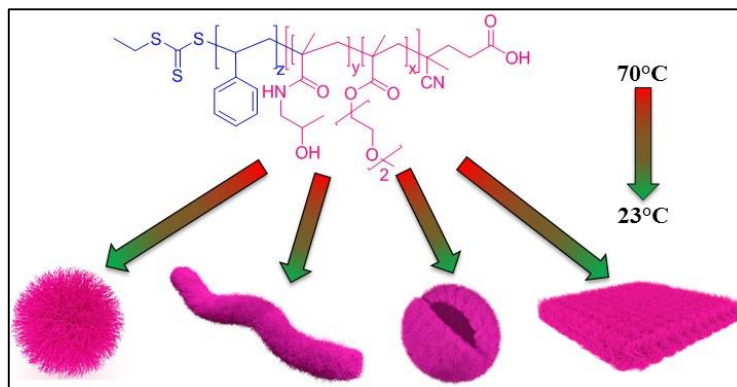


**Figure 1.** Temperature-induced morphology transformation allows the synthesis of polymeric nanoparticles with tunable shapes and cores.<sup>2</sup>

<sup>1</sup> P. B. Zetterlund, S. C. Thickett, S. Perrier, E. Bourgeat-Lami and M. Lansalot, *Chem. Rev.*, **2015**, 115, 9745.

<sup>2</sup> N. P. Truong, M. R. Whittaker, A. Anastasaki, D. M. Haddleton, J. F. Quinn and T. P. Davis, *Polym Chem-Uk*, **2016**, 7, 430.

<sup>3</sup> N. P. Truong, J. F. Quinn, A. Anastasaki, D. M. Haddleton, M. R. Whittaker and T. P. Davis, *Chem. Commun.*, **2016**, 52, 449.

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# Hyperbranched polymers by thiol-yne chemistry: Control of molecular weight and branch point distribution

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Highly branched and three dimensional macromolecular structures, or dendritic polymers, have become an important class of materials over recent decades. An efficient route to hyperbranched polymers with very high degrees of branching was developed by the Perrier group in 2009 using thiol-yne chemistry.<sup>1</sup> We propose a new and versatile approach to the production of hyperbranched polymers with high degrees of branching and low dispersity values (*D*), involving slow monomer addition of a thiol/yne monomer to multifunctional core molecules in the presence of photoinitiator and under UV irradiation.<sup>2</sup>

Introduction of small molecule monomer by slow addition to a multifunctional alkyne core or alkene core was found to lower dispersity (compared to equivalent batch polymerisation) at varying monomer concentrations (**Fig.1**). Degrees of branching were determined by <sup>1</sup>H NMR spectroscopy to be greater than 0.8 in most cases. Increasing the fraction of core molecule was found to decrease dispersity to values as low as 1.26 and 1.38 for the alkene core and alkyne core respectively. The Kuhn-Mark-Houwink-Sakurada  $\alpha$  parameter decreased from 0.35 for the batch process to values as low as 0.16, indicating that the thiol-yne structures became more globular and dense with the slow monomer addition strategy.

This slow monomer addition method has recently been expanded to include hyperbranched polymers with controlled molecular weight distributions from poly(2-ethyl oxazoline) thiol-yne macromonomers. This is an exciting new development which highlights the diversity of materials able to be synthesised, with control over molecular weight, macromonomer functionality, and branch point distribution.

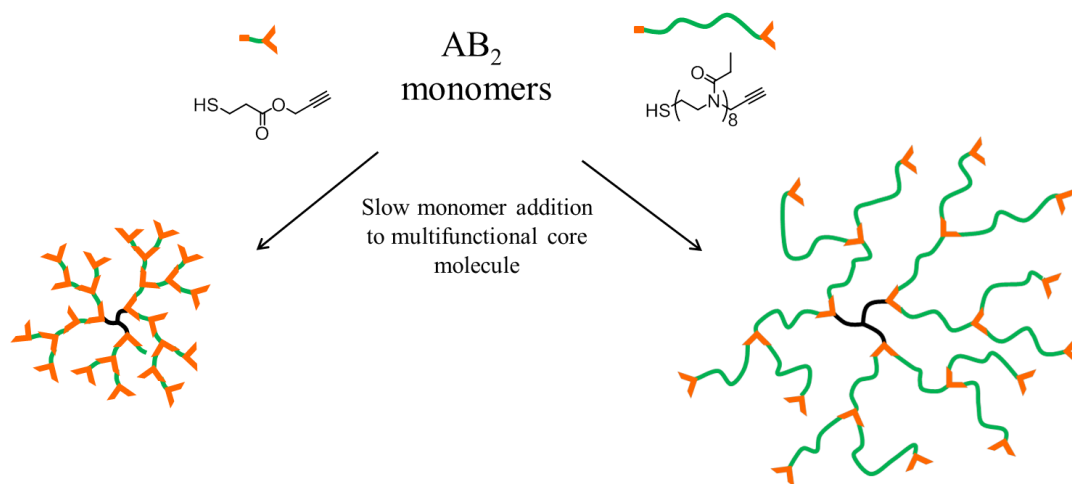


**Figure 1.** Routes to thiol-yne hyperbranched polymers by slow monomer addition strategy.

<sup>1</sup> D. Konkolewicz, A. Gray-Weale, S. Perrier, *J. Am. Chem. Soc.*, **2009**, *131* (50), 18075–18077.

<sup>2</sup> A.B. Cook, R. Barbey, J.A. Burns, S. Perrier, *Macromolecules*, **2016**, *49* (4), 1296–1304.

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# Functional Nanomaterials *via* Click Chemistry: From Polymeric Nanoparticles to Magneto-Induced Drug-Delivery Systems

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This presentation will describe our work to design and to prepare new functional organic (polymer-derived) and hybrid (magnetic iron oxide) nanoparticles by using orthogonal click chemistry reactions with emphasis on our most recent results using azlactone, alkyne, and furan as ‘clickable’ moieties.

The azlactone (oxazolone) ring reacts under mild conditions with primary amines without the formation of a byproduct, a reaction that can be regarded as proceeding with the advantages associated with conventional ‘click’-type reactions.<sup>1</sup> We have investigated azlactone-functionalized polymers as a platform for the design of new reactive thermoresponsive nanoparticles<sup>2</sup> and hybrid functional magnetite nanoparticles.<sup>3</sup>

By using a versatile methodology that employs a combination of orthogonal click reactions, namely copper-catalyzed azide-alkyne cycloaddition (CuAAC) and thermoreversible Diels-Alder reaction, we designed a new class of functional iron oxide magnetic nanoparticles showing unprecedented, active control over drug release using hyperthermia effects. Upon alternating magnetic field exposure, the retro-Diels-Alder reaction enables the release of a conjugated drug without any significant heating of the medium. This method has the potential to improve hyperthermia therapies by expanding the range of polymers and molecules (drugs, dyes, etc) that can be used. The so-obtained functional nanoparticles may become important tools for preparing the next generation of controlled-release devices and nanomedicines for *in vitro* and *in vivo* applications.

*Co-workers and students who have been contributing to this work are gratefully acknowledged.*

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# Controlled polymer architectures for probing cell uptake mechanism

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Chemists are remarkably proficient at directing the synthesis of small molecules, but fine-tuning the structures of large molecules, such as those found in polymers, is far more taxing. Despite many years of research, the field of macromolecular engineering i.e. the preparation of large molecules with strict control over their size and chemical groups has many mountainous challenges yet to overcome. Nature provides endless examples of precisely engineered macromolecules; proteins, for instance, which contain amino-acid side-chains that are accurately positioned, often in a way that determines the proteins' roles. Synthetic chemists have tried to recreate nature's exceptional control over macromolecules, and in so doing they have designed new materials with precisely defined structures, for use in applications ranging from materials to medicine. The lecture will describe our use of complex polymeric architectures designed by controlled polymerisation techniques to probe cellular mechanisms. In particular, we are interested in the design of drug delivery vectors to optimise their uptake by cells.

## Selected Publications

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- ## Recent Reviews
- Zetterlund, P.; Thickett, S.; Perrier, S.; Bourgeat-Lami, E.; Lansalot, M. *Chem. Rev.*, **2015**, DOI: cr-2014-00625k.R2
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# Dihydroxyanthraquinone Derivatives: Do Positions of Hydroxyl Substituents affect their Photoinitiating Ability of Photopolymerization under Blue LED Bulb?

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Photoinitiator (PI) is one of the most important components in photoinitiating system (PIS) as PI absorbs certain wavelengths of light (and interacts with relevant additives through electro transfer or energy transfer reactions) to generate active species (*e.g.* radicals or cations) for the initiation of various types of photopolymerization reactions.<sup>1</sup> The performance of photoinitiators is particularly important as it not only affects the photopolymerization efficiency of various monomers or oligomers but also influences the final properties of the produced polymeric materials. For instance, natural dyes may act as ideal photoinitiators and exhibit potential advantages for the elaboration of biocompatible polymeric materials as they are derived from natural systems. 9,10-anthraquinone derivatives (AQs) are one type of natural dyes from plants or biological sources.<sup>2</sup> To the best of our knowledge, no study has been reported of the use of dihydroxyanthraquinone derivative-based PISs for high efficiency of both free radical and cationic photopolymerization under the irradiation of household blue LED bulb. Therefore, it is interesting and valuable to investigate different dihydroxyanthraquinone (DHAQ) derivative-based PISs and the effect of chemical structures on their photoinitiation efficiency.

This research demonstrated that the positions of hydroxyl substituents in the anthraquinone molecule played an important role on their photoinitiation efficiency when combined with different additives (Figure 1). Among all the studied dihydroxyanthraquinone derivative-based photoinitiating systems, 18-DHAQ-based systems exhibited the highest photoinitiating ability for both free radical and cationic photopolymerization under the irradiation of blue LED bulb while 12-DHAQ-based systems were inefficient due to its low solubility. Interestingly, 14-DHAQ/Iod/NVK or 18-DHAQ/Iod/NVK system could initiate the free radical photopolymerization upon exposure to the household green LED bulb, and 14-DHAQ based combination was more efficient than 18-DHAQ based system due to the more overlap between the light absorption of 14-DHAQ and the emission spectrum of the green LED bulb.

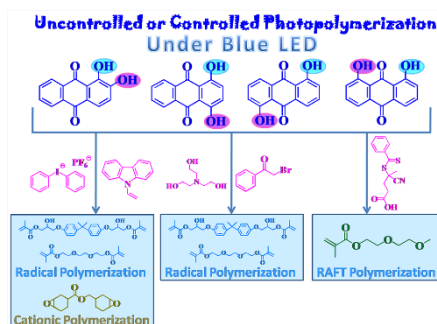


**Figure 1.** Dihydroxyanthraquinones as versatile photoinitiators of various photopolymerization reactions.

1. P. Xiao, J. Zhang, F. Dumur, M. A. Tehfe, F. Morlet-Savary, B. Graff, D. Gigmes, J. P. Fouassier and J. Lalevée, *Prog. Polym. Sci.*, 2015, **41**, 32-66.
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## Selective Photo-Activation for Precision Polymer Synthesis

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The ability of plants to convert solar energy into chemical energy via photoredox processes (natural photosynthesis) has inspired generations of chemists to try to reproduce such systems. Recently, the use of visible light photoredox catalysis in organic chemistry has enabled the synthesis of known chemical compounds through novel synthetic routes, limiting the formation of side products and eliminating complex purification procedures.<sup>1</sup> In addition to the high efficiency of these reactions, which require catalyst concentrations in ppm amounts, a number of photoredox catalysts have demonstrated additional remarkable properties, including compatibility and selectivity. Taking advantage of its compatibility with a number of common catalytic processes, recent development in the field of visible light photocatalysis has been dedicated to merging various catalysts to perform complex organic transformation in a single pot. In addition to this compatibility, photoredox catalysts are also able to selectively activate specific substrates. Such properties may play an essential role in organic syntheses for the production of added-value chemicals.<sup>1</sup>

More recently, the successful implementation of photoredox catalysis in polymer chemistry has led to the development of novel light mediated polymerizations, in living polymerizations.<sup>2</sup> We have developed a living radical polymerization technique involving the reversible deactivation of thiocarbonylthio compounds by photoredox catalysts via a photoinduced electron transfer (PET) process or energy transfer process. Recently, we reported the first exploitation of selective activation of trithiocarbonates in the presence of zinc tetraphenylporphine (ZnTPP), leading to the fast and controlled polymerization of acrylates, even under open air conditions.<sup>2e</sup>

In this abstract, we employ the unique property of selective activation for controlled radical polymerizations,<sup>3</sup> specifically PET-RAFT polymerization, to produce precise polymer architectures (**Figure 1**): (A) single unit monomer insertion to RAFT agents for functional polymers with precise location of functionalities; (B) controlling polymerization sequences under high degree of control.

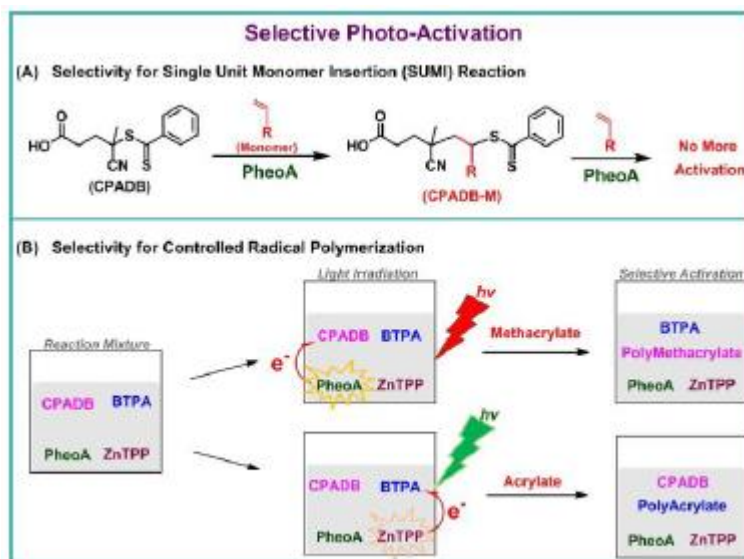


**Figure 1.** Selective Photo-Activation of PET-RAFT polymerization.

<sup>1</sup> (a) J. M. R. Narayanam and C. R. J. Stephenson, *Chem. Soc. Rev.*, **2011**, *40*, 102–113; (b) C. K. Prier, D. A. Rankic and D. W. C. MacMillan, *Chem. Rev.*, **2013**, *113*, 5322–5363. (c) N. Corrigan, S. Shanmugam, J. Xu, C. Boyer, *Chem. Soc. Rev.* **2016**, DOI:10.1039/C6CS00185H.

<sup>2</sup> (a) J. Xu, K. Jung, A. Atme, S. Shanmugam and C. Boyer, *J. Am. Chem. Soc.*, **2014**, *136*, 5508–5519; (b) J. Xu, K. Jung and C. Boyer, *Macromolecules*, **2014**, *47*, 4217–4229; (c) J. Xu, K. Jung, N. A. Corrigan and C. Boyer, *Chem. Sci.*, **2014**, *5*, 3568–3575; (d) J. Xu, S. Shanmugam, H. T. Duong and C. Boyer, *Polym. Chem.*, **2015**, *6*, 5615–5624. (e) S. Shanmugam, J. Xu and C. Boyer, *J. Am. Chem. Soc.* **2015**, *137*, 9174–9185.

<sup>3</sup> J. Xu, S. Shanmugam, C. Fu, K. Aguey-Zinsou and C. Boyer, *J. Am. Chem. Soc.* **2016**, *138*, 3094–3106.

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## New Multifunctional “Switchable” RAFT Agents for Accessing Triblock Copolymers with Less Active and More Active Segments

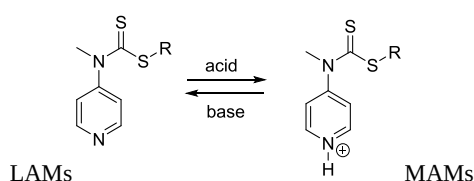
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RAFT agents are selected based on the reactivity of the monomer being polymerized, with monomers characterised into the two broad categories of more active monomers (MAMs) and less active monomers (LAMs). Conventional RAFT agents are generally suitable to polymerize one class of monomers; trithiocarbonates and dithioesters give control over MAMs while better control for LAMs is achieved with xanthates and dithiocarbamates.<sup>1</sup> Acid/base switchable *N*-pyridinyl dithiocarbamate RAFT agents (**Scheme 1**) have since been developed to overcome this and provided a synthetic route blocks consisting of both LAMs and MAMs.<sup>2-5</sup>



**Scheme 2:** Structure of acid/base switchable *N*-methyl-*N*-pyridyldithiocarbamate RAFT agents

We are currently exploring the development of new multifunctional switchable RAFT agents, the thermal end group removal of a variety of MAMs and LAMs, as well as the copolymerization of LAMs and MAMs with both the standard switchable RAFT agents and bis-switchable RAFT agents. The focus of this work is on the synthesis of new triblocks consisting of the more active monomer *N*-isopropylacrylamide and the less active *N*-vinylpyrrolidone. Recent results obtained in these areas will be presented.

<sup>1</sup> D. J. Keddie, *Chem Soc Rev.* **2014**, 43, 496-505

<sup>2</sup> G. Moad, E. Rizzardo, S. H Thang, *Chem. Asian J.* **2013**, 8, 1634-44

<sup>3</sup> M. Benaglia, M. Chen, Y. K. Chong, G. Moad, E. Rizzardo, S. H. Thang, *Macromolecules*, **2009**, 42, 9384–9386

<sup>4</sup> D. J. Keddie, C. Guerrero-Sanchez, G. Moad, E. Rizzardo, S. H. Thang, *Macromolecules*, **2011**, 44, 6738–6745.

<sup>5</sup> S. J. Stace, G. Moad, C. M. Fellows, D. J. Keddie, *Polym. Chem*, **2015**, 6, 7119-7126

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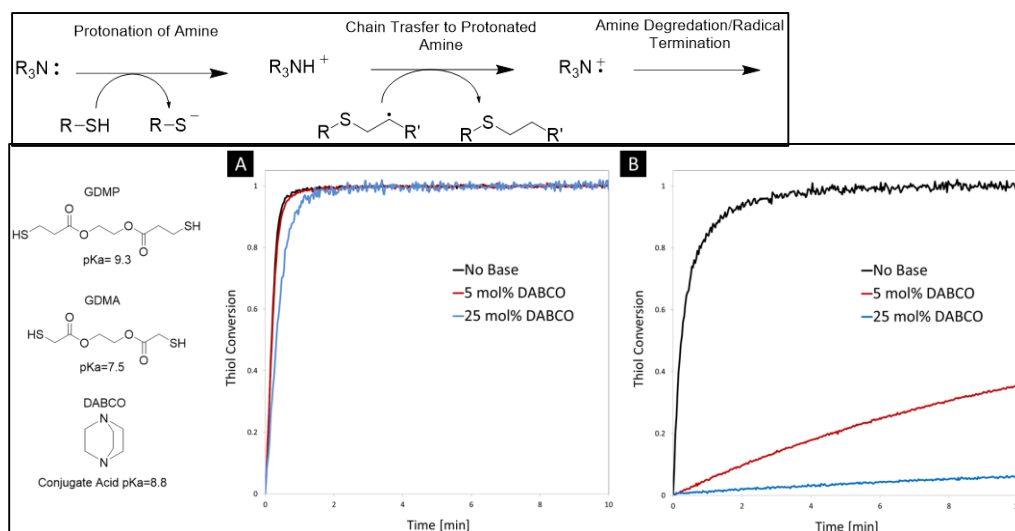
Research interests: RAFT, RDRP, Thermolysis, Emulsion Polymerization

## Amine Retardation of Radical Mediated Thiol-ene

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Owing to mild reaction conditions, versatile reactant design, lack of side products, and potential for spatial and temporal control, radically initiated thiol-ene has become an important tool in organic synthesis in general,<sup>1</sup> and polymer chemistry particularly.<sup>2</sup> Although well known to be side-group tolerant, our investigations of thiol-ene kinetics have shown that amines can significantly retard radical thiol-ene through a radical scavenging pathway. We hypothesize that the amine in the conjugate acid form is undergoing a chain-transfer event (**Scheme 1**), in which a carbon centered radical abstracts a hydrogen from the amine generating a radical cation centered on the nitrogen, leading to radical termination and amine degradation, as evidenced by NMR. The magnitude of retardation depends primarily on the acid-base properties of the thiol and amine (**Fig.1**); we observe that when the amine's conjugate acid pKa is lower than that of the thiol the reaction proceeds relatively unaffected. Inversely, when the pKa of the base is higher than the thiol, significant retardation is observed at amine loading as low as 1 mol% equivalence, indicating that the amine must be in the protonated form in order to retard the reaction. Furthermore, in aqueous thiol-ene systems with amines present, polymerization rates for hydrogel formation exhibit a significant negative correlation to small increases in pH when the pH is similar to the pKa of the amine.



**Scheme 1.** Proposed mechanism for amine retardation of radical thiol-ene

**Figure 1.** Thiol conversion versus time for a thiol-ene polymerization of two dithiols, (A) GDMP and (B) GDMA, with a diallyl ether and varying molar equivalence of DABCO. Reactions were formulated with 1 mol% DMPA photoinitiator and irradiated at 365 nm, 2.5 mW/cm<sup>2</sup>, under ambient conditions.

Our results have implications for bulk thiol-ene polymerizations in the presence of small quantities of amines as well as for dual cured polymer networks where one stage is a base catalyzed, thiol-Michael, thiol-epoxy, thiol-isocyanate, or thiol-halide cure. Additionally, there may be utility in our observations for stabilizing thiol-ene formulations through the incorporation of amines in low concentrations.

<sup>1</sup> F. Denes, M. Pichowicz, G. Povie, P. Renaud, *Chem. Rev.* **2014**, *114*, 2587-2693

<sup>2</sup> C. Hoyle, C. Bowman, *Angew. Chem. Int. Ed.* **2010**, *49*, 1540-1573

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## Protein delivery via self-assembling peptide scaffolds: supporting human embryonic stem cell-derived transplants in stroke

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Brain injury results in neuronal loss and disruption of the brain parenchyma. Therefore, to achieve functional recovery post insult, it is necessary to promote the long-term survival of neurones and circuitry reconstruction. This is a complicated process, which will not only involve attenuating secondary injury, but to also concomitantly provide neuroprotection within the penumbra immediately following tissue insult. Ultimately, repair will depend on: (1) supporting the penumbra surround the lesion; (2) transplanting cells into the injury site; (c) providing a suitable microenvironment for transplanted cells to interegrate with the host and (d) to support surviving cells within the penumbra.

Focusing on this, we engineered a brain tissue-specific self-assembling peptide (SAP) based on the IKVAV sequence in laminin<sup>1</sup>, and the growth factor brain derived neurotrophic factor (BDNF); important in neuronal survival, differentiation and proliferation. Hydrogels were formed via a pH switch at a concentration of 10 mg/ml and were extensively characterised<sup>2</sup>. BDNF was loaded into these hydrogels at a concentration of 30ng/mL. The stroke model was achieved by stereotactically inject human ET-1 peptide into the frontal, motor cortex of rats. We then undertook neural patterning, induction and differentiation of GFP+ human embryonic stem cells (hESCs) into forebrain cortical progenitors. These were subsequently grafted into the cystic lesion cavity. The grafts were characterised by assessing behaviour, cortical atrophy and using immunocytochemistry.

Here, we have shown that the transplanting hESC derived forebrain cortical progenitors concomitantly with our Fmoc-DDIKVAV hydrogel across the necrotic lesion cavity, provided both physical and trophic support to transplanted cells, as well as for host cells within the penumbra. We showed that when BDNF was released from our scaffolds the initial grafted cells number and graft size was significantly enhanced (Fig. 1). The SAP hydrogels were shown to significantly extend the delivery of BDNF from minutes to days, which improves the survival and integration of the transplanted cells over a 9 month period. While the focus of this study was on repairing the injured brain, our biologically relevant hydrogels have broader implications for exploitation in areas such as cancer therapy, muscle and bone regeneration.

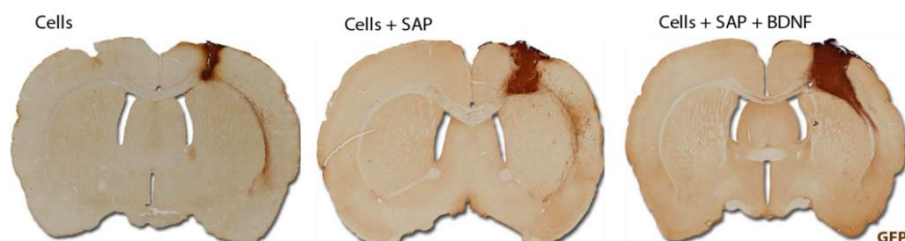


Figure 1: Coronal sections of a rat brain with hESC derived forebrain cortical progenitor cells transplanted within an ET-1 cystic cavity. GFP+ imaging was taken 28days post implantation for a cells alone control, cells + SAP hydrogel and cells + SAP + BDNF.

1. Rodriguez AL, Wang TY, Bruggeman KF, Horgan CC, Li R, Williams RJ, Parish CL, Nisbet DR, J Mater Chem B; 2: 7771-7778; 2014.
1. AL Rodriguez, T Wang, K Bruggeman, R Li, R Williams, C Parish, Nisbet D, Nano Research; 9(3): 674-684; 2016.



## Polypeptide block co-polymers as versatile platform for drug delivery

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Synthetic polypeptides are advantageous for biomedical applications due to their structural similarities to natural proteins and peptides. These materials can address the challenges of biocompatibility and biodegradability associated with fully synthetic polymers. Moreover, they adopt stable secondary structures like helices, sheets and random coil through non-covalent interactions leading to novel self-assembled supramolecular structures.<sup>1</sup> Additionally, these polypeptides can reveal a transition in response to the change in local environment like temperature, pH, salt etc. depending on the side chain functionality of constituting amino acids.<sup>2</sup> Polypeptides of desired molecular weight possessing tunable properties, can be synthesized from amino acid building blocks in a controlled manner using ring-opening polymerization (ROP) of N-carboxyanhydride (NCA) monomers.<sup>3,4</sup> In this study, synthetic block co-polypeptides consisting of varying ratios of hydrophobic and hydrophilic components have been synthesized. The hydrophobic core of self-assemblies of these amphiphilic polypeptides were loaded with drug of choice by hydrophobic-hydrophobic interaction. When monitored, these nano-assemblies showed prolonged controlled release of drug payload at physiological conditions.

<sup>1</sup>H. Lu., *et al.*, *Chem. Commun.*, **2014**, 50, 139.

<sup>2</sup>J.Huang, A. Heise, *Chem. Soc. Rev.*, **2013**, 42, 7373.

<sup>3</sup>G. J. M. Habraken, *et al.*, *Polym. Chem.*, 2011, 2, 1322

<sup>4</sup>T. Stukenkemper, *et al. Macromol. Biosci.*, **2015**, 15, 138.

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# The electrokinetic properties of lubricin anti-adhesive protein

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Lubricin is a glycoprotein found in articular joints which has long been recognized as being an important biological boundary lubricant molecule and, more recently, an impressive antiadhesive that readily self-assembles into a well ordered, polymer brush layer on virtually any substrate. The lubricin molecule possesses an overabundance of anionic charge, a property that is atypical among antiadhesive molecules, that enables its use as a coating for applications involving electrokinetic processes such as electrophoresis and electroosmosis. Coating the surfaces of silica and polymeric microfluidic devices with self-assembled lubricin coatings affords a unique combination of excellent fouling resistance and high charge density that enables notoriously "sticky" biomolecules such as proteins to be used and controlled electrokinetically in the device without complications arising from nonspecific adsorption. Using capillary electrophoresis, we characterized the stability, uniformity, and electrokinetic properties of lubricin coatings applied to silica and PTFE capillaries over a range of run buffer pHs and when exposed to concentrated solutions of protein. In addition, we demonstrate the effectiveness of lubricin as a coating to minimize nonspecific protein adsorption in an electrokinetically controlled polydimethylsiloxane/silica microfluidic device.

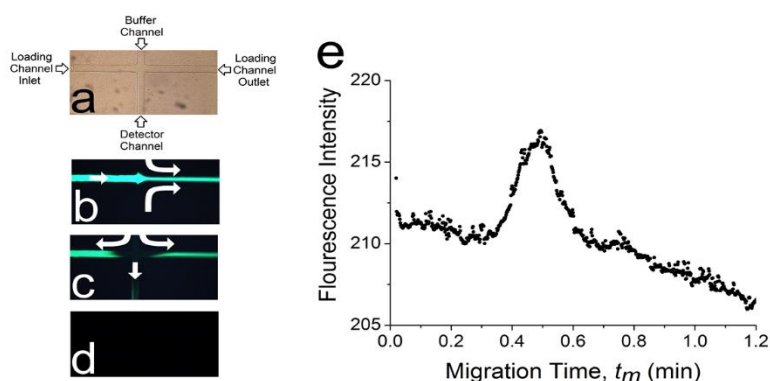


**Figure 1.** The electrokinetic loading, injection, and fluorescence detection of a 0.5 mg/ml solution of Alexa Fluor® 488 labelled IgG in MOPS buffer (pH 7.2) performed in a LUB coated microfluidic PDMS/silica electrophoresis chip using the applied electrical potentials described in Fig. 1. (a) White field optical image showing the injection point and configuration of the individual channels in the cross-channel chip. (b) Fluorescence image showing the loading of the anti-rabbit IgG with white arrows indicating the directions of fluid flow in chip channels. (c) Fluorescence image showing the moment of injection (i.e. the change in voltages) of the anti-rabbit IgG with white arrows indicating the directions of fluid flow in chip channels. (d) Fluorescence image of the same region of the chip shown in (a-c) approximately 10s after the injection shown in (c) indicating no discernible residual fluorescence due to non-specifically adsorbed anti-rabbit IgG. (e) Plot of the fluorescence intensity vs. the migration time indicating the detection of the injected anti-rabbit IgG.

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# Controlled attachment of *Pseudomonas aeruginosa* with colloidal crystal based topographies

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Bacterial biofilm development on medical implants resultant infections are a major problem for health care. It has been shown that surface micro- and nanopatterns can interrupt bacterial adhesion to surfaces and interrupting the development of biofilms. There are numerous techniques that have been used to create micro- and Nano topographies with most of them requiring very expensive, sophisticated and often time consuming fabrication processes. However, less focus has been on creating easier ways to generate low cost micro- and nanopatterns. In these work binary colloidal crystals (BCC) self-assembly in a confined area is used to pattern surfaces with highly ordered crystal patterns (**Fig.1**) of Si and PMMA form the micro- to the nanoscale<sup>1</sup>. AAMpp was then deposited on all binary particles to create a uniform chemistry on all binary surfaces thus overcoming dual chemistry effects and investigating the role of topography only on bacterial adhesion. *P. aeruginosa* was chosen to study the bacterial interactions with these binary colloidal crystal patterned surfaces. Scanning electron microscope (SEM), x-ray photoelectron spectroscopy (XPS), and Epi-fluorescence microscopy were used for pattern characterization, surface chemical analysis, and imaging of attached bacteria. XPS results confirm the successful attachment of AAMpp surfaces and homogenous, smooth AAMpp surfaces had high levels of bacterial attachment because of electrostatic attractions. However, the results for the AAMpp coated crystals showed that micron sized binary assemblies can be used as a surface for delaying bacterial attachment by selectively attracting the bacteria to the small particle regions of the patterns. These types of patterns are easy to fabricate and could be useful in further applications in biomedical research.

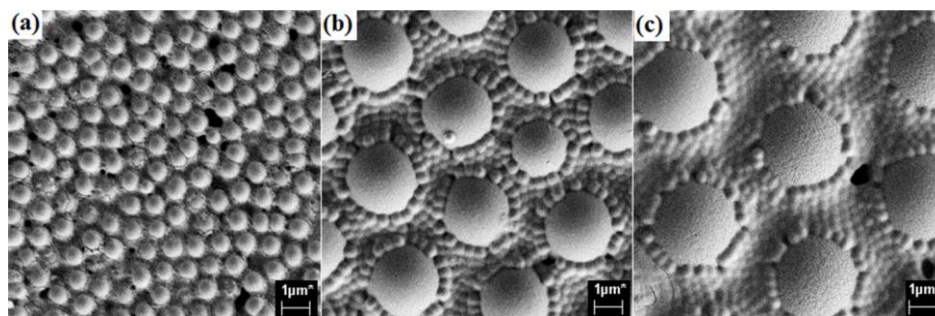


**Figure 1.** SEM images of Si/PMMA BCC layers generated on a glass substrate. (a) Si1.1/PMMA0.11, (b) Si3.7/PMMA0.4, (c) Si4.9/PMMA0.4

<sup>1</sup> H. Pingle, PY. Wang, Biointerphases, 2015. 10(4): p.04A309.

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# Polymeric Nano-Objects of Various Morphologies via Block Copolymer Self-Assembly Using Microporous Membranes

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Polymeric nano-objects of non-spherical morphology (*e.g.* rods, vesicles) have a range of potential applications, and it is thus of great interest to develop synthetic approaches that enable large scale production as well as fine-tuning of the morphology. Polymeric nano-objects based on self-assembly are traditionally prepared via post-polymerization methods in dilute aqueous solutions (*e.g.* using dialysis), thus making large scale preparation of these materials a challenge.<sup>1</sup> The relatively recently developed technique of polymerization induced self-assembly (PISA)<sup>2</sup> represents a very significant step forward in that it enables direct one-pot preparation of polymeric nano-objects of various morphologies in concentrated solutions.

In the present work, we have developed an alternative approach to prepare polymeric nano-objects of various morphologies based on self-assembly of diblock copolymers in tandem with microporous membrane technology (Shirasu Porous Glass (SPG) membranes).<sup>3</sup> Membrane emulsification, which involves generation of a liquid dispersed phase by passage through the pores of a microporous membrane directly into the continuous phase, entails the use of two immiscible liquids for preparation of oil-in-water or water-in-oil emulsions of well-defined droplet size. In this work, this approach has been adapted to self-assembly of amphiphilic diblock copolymers. Importantly, the two liquids in the present work are miscible, *i.e.* an emulsion is not generated. Diblock copolymers poly(oligoethylene glycol acrylate)-*b*-poly(styrene) (POEGA-*b*-PSt) comprising different block lengths were prepared by RAFT polymerization. Self-assembly using SPG membrane technology was subsequently conducted by first dissolving the polymer in tetrahydrofuran, followed by continuous passage of this copolymer solution through the membrane pores into the aqueous phase. The results indicate that it is possible to conveniently tune the nano-object morphology via the process parameters of pressure and pore size for a given formulation (**Fig. 1**). The technique also holds promise with regards to high throughput, in particular in the case of vesicles.

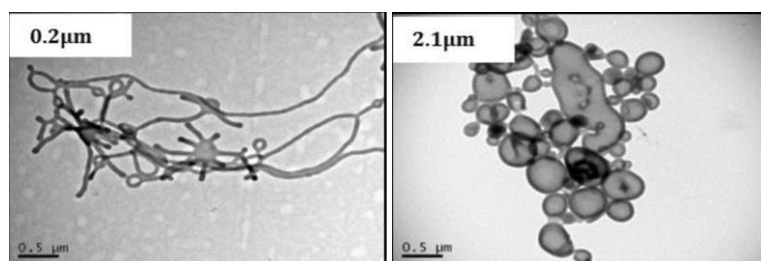


**Figure 1.** Nano-object morphologies obtained using POEGA<sub>17</sub>-*b*-PSt<sub>290</sub> (THF/water system) for different SPG membrane pore sizes as indicated (keeping all other parameters constant).

<sup>1</sup> Y. Mai and A. Eisenberg, *Chem. Soc. Rev.*, **2012**, *41*, 5969-5985

<sup>2</sup> N. J. Warren and S. P. Armes, *J. Am. Chem. Soc.*, **2014**, *136*, 10174-10185

<sup>3</sup> K. Kandori, K. Kishi and T. Ishikawa, *Colloids and Surfaces*, **1991**, *55*, 73-78

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## Nanoscale Patterning of Inherently Conducting Polymers for Regenerative Therapy

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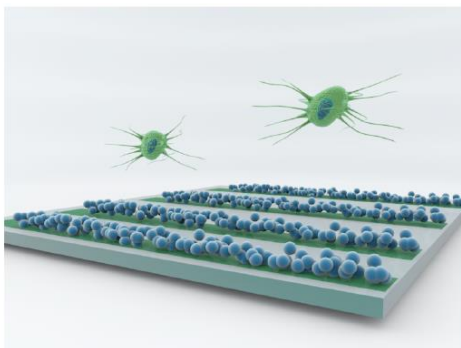
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The use of programmed electrical signals to influence biological events has been a widely accepted clinical methodology for neurostimulation. Traditionally metallic electrodes and more recently carbon fibers have been effectively employed in bionic devices. However, stiff metal electrodes suffer a major drawback of eliciting tissue damage over long-term implantation. An optimal biocompatible platform for neural activation efficiently transfers electrical signals across the electrode–cell interface and also incorporates large-area neural guidance conduits. Inherently conducting polymers (ICPs) have emerged as frontrunners as soft biocompatible alternatives to traditionally used metal electrodes, which are highly invasive and elicit tissue damage over long-term implantation. However, fabrication techniques for the ICPs suffer a major bottleneck, which limits their usability and medical translation. In this talk I will demonstrate that these limitations can be overcome using colloidal chemistry to fabricate multimodal conducting polymer nanoparticles. Furthermore, I will demonstrate that these polymer nanoparticles can be precisely assembled into large-area linear conduits using surface chemistry. Finally, I will present data to validate that this platform can act as guidance conduits for neurostimulation, whereby the presence of electrical current induces remarkable dendritic axonal sprouting of cells.



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Prof Swaminatha Iyer obtained a Bachelor of Engineering in Polymer Science and Technology at the University of Mysore in India (1999). He then obtained a Masters in Polymer Science, at Clemson University in South Carolina in the USA (2002), where he also completed his PhD in Materials Science and Engineering (2004). Prof Iyer then undertook a year of post doctoral research in Biophysics at Clarkson University in New York before taking up an ARC Research Fellowship at UWA in 2006 where he leads an internationally recognized research program in the field of bionanotechnology, in The School of Chemistry and Biochemistry at The University of Western Australia. His research focuses on the use fundamental concepts in chemistry to engineer innovative polymer nanoformulations which are designed for the treatment of untreatable medical emergencies like traumatic brain injuries, cardiovascular diseases, placental related disorders in pregnancy and cancers.

# Engineering of Functional Nanocapsules by Nanoprecipitation

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There has been a growing interest in the preparation of synthetic capsules owing to their numerous applications.<sup>1</sup> Routes to polymeric colloids in the submicronic scale usually require soft matter technologies (self-organization of amphiphilic block copolymers, LbL deposition on sacrificial template particles, polymerization-induced self-assembly or interfacial polymerization in miniemulsion conditions). Another means of generating nano-objects relies on the nanoprecipitation technique or “Ouzo effect”, the principle of which lies on the supersaturation of whatever hydrophobic solute (oil, polymer, solids) dissolved in a hydrophilic solvent by adding a far excess of the non-solvent, water. When conditions are set up in a narrow range of the ternary phase diagrams, one can reproducibly generate colloids. Herein we present a general undemanding strategy to build multi-tagged, oil-filled nanocapsules with controlled morphologies.<sup>2</sup>

We show that the exploitation of phase diagrams for polymer/water/solvent and oil/water/solvent systems allows to define experimental conditions in which polymer chains preferentially stick at the interface of the oil droplets to generate polymeric nanocapsules. The resulting nanocapsules can be cross-linked, shell-functionalized with molecules of biological interest and/or loaded with actives in a rapid one-pot procedure.

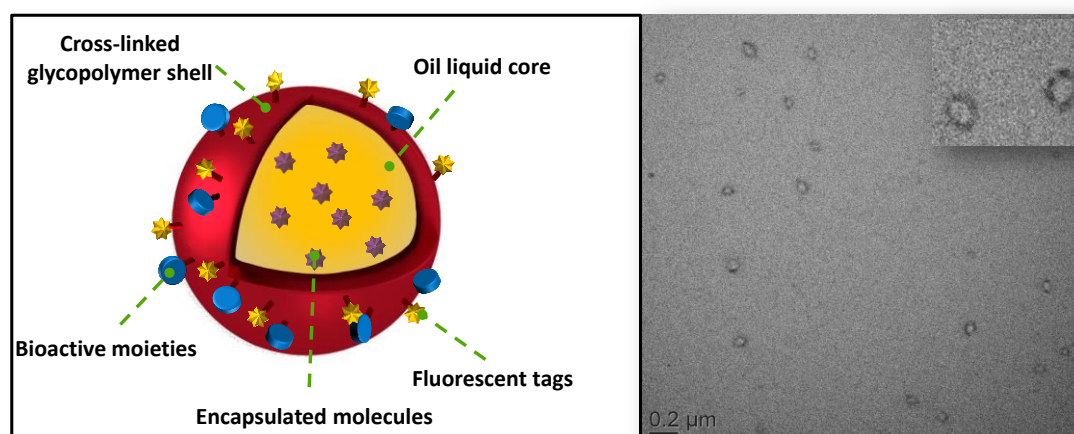


Figure 1. Left: General sketch of the functional nanocapsules. Right: TEM picture of the nanocapsules

2. Reddy, L. H.; Arias, J. L.; Nicolas, J.; Couvreur, P. *Chem. Rev.* **2012**, *112*, 5818-5878.
3. a) Yan, X.; Delgado, M.; Fu, A.; Katz, J.; Gouin, S.; Fleury, E.; Ganachaud, F.; Bernard J. *Angew. Chem. Int. Ed.* **2014**, *53*, 6910-6913. b) Yan, X.; Sivignon, A.; Alcouffe, P.; Burdin, B.; Favre-Bonté, S.; Bilyy, R.; Barnich, N.; Fleury, E.; Ganachaud, F.; Bernard, J. *Chem. Commun* **2015**, *51*, 13193-13196

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## Cationic Polymers based on Fructose and Galactose Moieties for Nucleic Acids Delivery

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Cationic polymers and glycopolymers were synthesised using the RAFT technique. Combining cationic polymers with glycopolymers has great potential in targeted nucleic acid delivery.<sup>1,2</sup> However, many obstacles prevent the use of cationic glycopolymers as vectors including low success in nucleic acid delivery and high toxicity of the cationic polymer. This project aims to investigate RAFT synthesis of cationic glycopolymers with galactose or fructose carbohydrates, their binding ability with their specific lectins and with negatively charged nucleic acids.

The cationic polymer poly(2-(dimethylamino)ethyl methacrylate) (PDMAEMA) was synthesised using RAFT polymerisation. The galactose monomer, 2-(2',3',4',6'-tetra-O-acetyl- $\beta$ -D-galactosyloxy) ethyl methacrylate (AcGalEMA), and the fructose monomer, 1-O-methacryloyl-2,3:4,5-di-O-isopropylidene- $\beta$ -D-fructopyranose (1-O-MAiPFru)<sub>3</sub>, were polymerised with PDMAEMA to form cationic glycopolymers. Chain extension was confirmed using proton nuclear magnetic spectroscopy (NMR) and gel permeation chromatography (GPC). Gel permeation chromatography was also performed to determine the polydispersity index (uniformity) of the polymers.

The protected glycopolymer blocks were modified by deacetylation of the galactose block and acid deprotection of the fructose block. Characterisation of the modified cationic glycopolymers was achieved using proton nuclear magnetic spectroscopy for confirmation of deacetylation/deprotection, and dynamic light scattering to determine the sizes of the diblock copolymers. The zeta potential (ionic charge) of the diblock copolymers was recorded.

Aggregation assays between the cationic glycopolymers and plant lectins were assessed. The galactose-containing glycopolymers were conjugated with peanut agglutinin lectin and the fructose-containing glycopolymers were conjugated with lectin from *Ulex europaeus*. The assays were analysed using dynamic light spectroscopy and ultraviolet-visible spectroscopy. Complexation of the cationic glycopolymer with small interfering RNA (siRNA) was accomplished. The size of the resulting polyplex was recorded with dynamic light spectroscopy. The zeta potential was measured and compared to the zeta potential measurement before complexation with siRNA.

Results indicated that RAFT polymerisation was successful in producing diblock polymers of controlled weight and uniform size. The cationic glycopolymers were partially successful in deacetylation/deprotection and highly successful in binding to their specific lectins. The cationic glycopolymer complexed with siRNA; however, further research into the appropriate N:Pratio is necessary. In conclusion, RAFT polymerisation is a suitable technique for the synthesis of cationic glycopolymers for use in nucleic acid delivery. The cationic block of the polymer is able to complex with nucleic acids while the glycopolymer block is able to bind to specific lectins. Further research into carbohydrates specific binding and further modifications to increase nucleic acid delivery efficiency would be beneficial.

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<sup>3</sup>J. Zhao, K. Babiuch, H. Lu, A. Dag, M. Gottschaldt and M. H. Stenzel, *Chem. Comm.*, **2014**, 50, 159228.

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Research interests: Reversible deactivation radical polymerisation





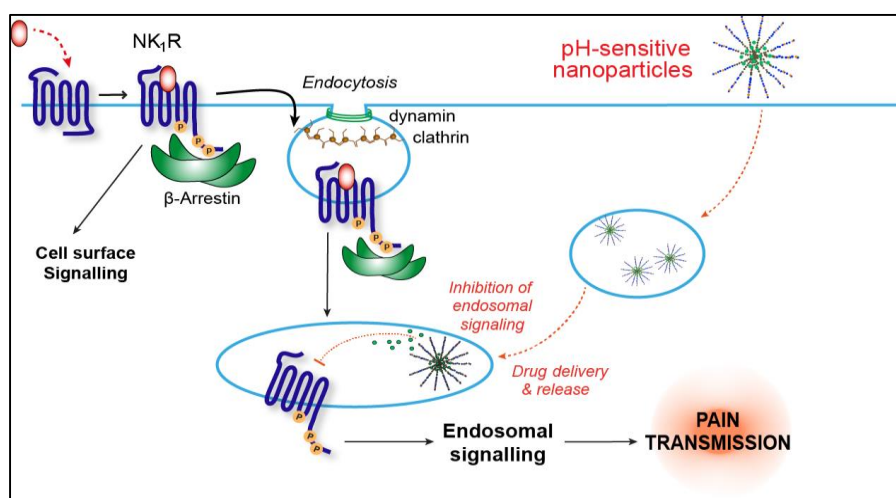
## Using Adaptive Nanoparticles to Target Endosomal Signalling

Nicholas Veldhuis<sup>1</sup>, Michael R. Whittaker<sup>1</sup>, Paulina Ramirez-Garcia<sup>1</sup>, Quynh N. Mai<sup>1</sup>, An Huu Tran-Doan<sup>1</sup>, TinaMarie Lieu<sup>1</sup>, Nghia P. Truong<sup>1</sup>, John F. Quinn<sup>1</sup>, Thomas P. Davis<sup>1</sup> and Nigel W. Bunnett<sup>1</sup>

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Many chronic conditions of global relevance could benefit from targeted delivery of therapeutics to modulate cellular processes. While chemotherapeutic nanoparticle approaches are designed to induce cell death by “endosomal escape” of a cytotoxic agent, nanoparticle delivery systems that enhance cellular drug uptake while maintaining cell viability may also be therapeutically advantageous. We have recently determined that a G Protein-Coupled Receptor known as the Neurokinin 1 Receptor (NK<sub>1</sub>R) stimulates transient signalling at the cell surface and sustained cell signalling from endosomes. Importantly, the NK<sub>1</sub>R endosomal signalling promotes sustained spinal neuron excitability and central pain transmission. To explore the potential to develop novel analgesic agents that target endosomal pain transmission, we have designed and tested pH-responsive nanoparticles that are rapidly taken up into cells and selectively release cargo (NK<sub>1</sub>R antagonists) into endosomes, without disrupting cellular function (**Fig 1**).

**Figure 1.**  
Proposed mechanism for drug delivery and release into endosomes for inhibition of receptor-mediated pain transmission.



The acidic environment within the lumen of early and late endosomes (~pH 6.5-5.9) was exploited as a trigger for nanoparticle disassembly and cargo release. Inspired by the work of Gao et al.<sup>1</sup>, a library of nanoparticles was generated comprising block copolymers synthesized via RAFT polymerisation. The particles comprised a nonfouling hydrophilic shell incorporating poly(ethylene glycol) methacrylate (PEGMA) and 2-(dimethylamino)ethyl methacrylate (DMAEMA) units. The hydrophobic core incorporated units derived from either 2-(diethylamino)ethyl methacrylate (DEAEMA, pK<sub>a</sub> ~7.1) or 2-(diisopropylamino)ethyl methacrylate (DiPAEMA, pK<sub>a</sub> ~6.1). While the particles were stable at physiological pH, with endosomal uptake the reduced pH promotes monomer protonation, triggering a hydrophobic-hydrophilic solubility switch that facilitates nanoparticle disassembly and concomitant release of the nanoparticle cargo in the endosomes. As a strategy for increasing cell viability of these pH-responsive particles, we have also explored the potential for introduction of diethylene glycol methacrylate (DEGMA) units into the core-forming block. Outcomes of cell toxicity, drug loading, intracellular uptake and trafficking, and *in vitro* and *in vivo* responses will be discussed.

<sup>1</sup>K. Zhou, Y. Wang, X. Huang, K. Luby-Phelps, B.D. Sumer, J. Gao, Angew. Chem. Int. Ed. **2011**, 50, 6109-114

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Research interests: Endosomal signalling, pain transmission, neuroscience, Pharmacology of Lipidated probes, G Protein-Coupled Receptors, TRP ion Channels

## Cyclic peptide-polymer conjugates: self-assembling vectors for drug delivery

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The use of functional drug carrier systems is an effective method for improving the potency of drugs and reducing the occurrence of side effects.<sup>1</sup> Among the variety of possible delivery vectors, organic nanotubes, especially those formed via the hydrogen bonding-directed self-assembly of cyclic peptides-polymer conjugates are of particular interest due to their biocompatibility and anisotropic shape.<sup>2</sup> These nanotubes are built around a core made of  $\beta$ -sheet forming cyclic peptides (CPs).<sup>3</sup> CPs presenting an even number of alternating D- and L-amino acids can adopt a flat conformation in which the alignment of amide functionalities allows the formation of hydrogen bonds between cyclic peptides, promoting their stacking into tubular assemblies. The conjugation of polymers to these CPs has proven to be an effective way of circumventing issues relative to their poor solubility and the limited control over nanotube length, as well as a convenient way of introducing a wide range of functionalities.<sup>4,5</sup>

Here, the synthesis, characterisation and early *in vitro* tests of cyclic peptide-polymer nanotubes are described (**Figure 1**). Cyclic peptide-poly(2-hydroxypropyl methacrylate) (pHPMA) conjugates were synthesized, loaded with an anticancer drug, and assembled in solution. A comonomer was used to provide a reversible ligation handle for the drug. The self-assembled structures were characterized by small angle neutron scattering (SANS) which proved an elongated cylindrical shape. *In vitro* testing of the nanotubes on the ovarian cancer cell line A2780 reveals that the conjugates themselves were non-toxic, but the drug-loaded nanotubes are more potent than both the free drug and the drug-loaded polymers.

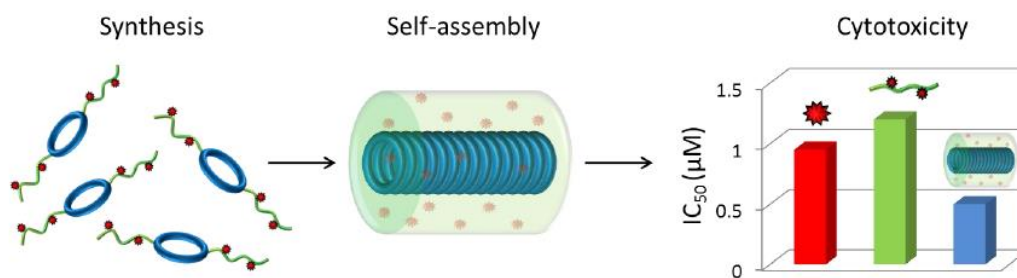


**Figure 1.** Schematic representation of the conjugates and their self-assembly.

<sup>1</sup>A. Z. Wang, R. Langer, O. C. Farokhzad, *Annual Review of Medicine* **2012**, 63(1), 185-198.

<sup>2</sup>Y. Geng, P. Dalhaimer, S. S. Cai, R. Tsai, M. Tewari, T. Minko, D. E. Discher, *Nat. Nanotechnol.* **2007**, 2(4), 249-255.

<sup>3</sup>M. R. Ghadiri, J. R. Granja, R. A. Milligain, D. E. McRee, N. Khazanovich, *Nature* **1993**, 366 (6453), 324-327.

<sup>4</sup>R. Chapman, M. Danial, M. L. Koh, K. A. Jolliffe, S. Perrier, *Chem. Soc. Rev.* **2012**, 41 (18), 6023.

<sup>5</sup>S. C. Larnaudie, J. C. Brendel, K. A. Jolliffe, S. Perrier, *J. Polym. Sci., Part A: Polym. Chem.* **2015**, 54, 1003-1011.

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## **$^{19}\text{F}$ Magnetic Resonance Imaging switches: Advanced drug delivery and monitoring**

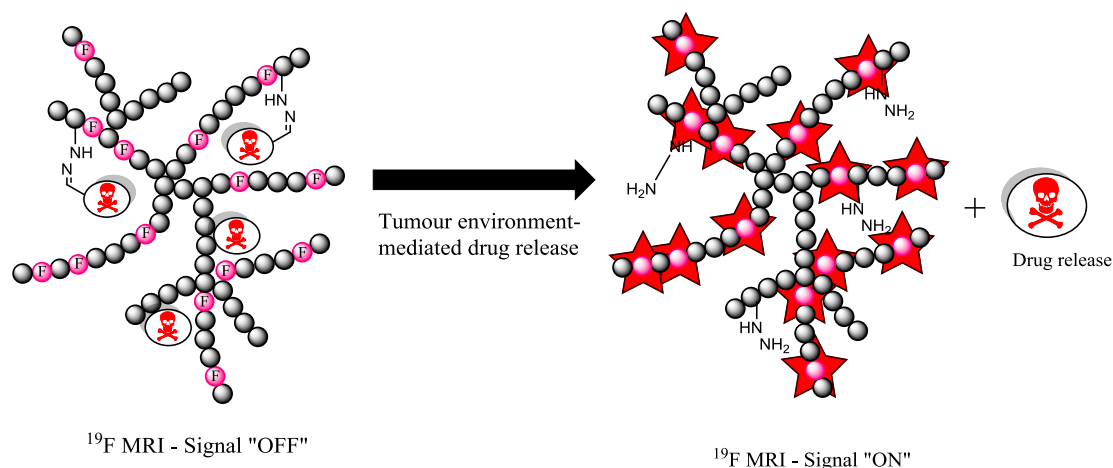
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Development of “personalised medicine” is one of the major challenges of the modern field of nanomedicine. This means that not only does a device need to be capable of delivering a therapeutic dose to the correct site within the body, but there must also be a mechanism for online diagnosis, monitoring of disease progression and visualisation of drug delivery, release and efficacy of treatment. Such a concept will require the combination of imaging modalities with unique “switching” capabilities in order to monitor cell-driven release of drugs.

The current work focusses on the development of a  $^{19}\text{F}$  MRI theranostic vehicle entailing stimuli-responsive therapeutic release. Through changes in structure-property relationships of the vehicle, the simultaneous change in  $^{19}\text{F}$  relaxivity provides the ability to image and qualify and/or quantify this delivery.<sup>1,2</sup> This was achieved through the release of a hydrophobic therapeutic from the hyperbranched polymer under the simulated decreased pH of the tumour micro-environment. The switch in hydrophobicity caused an increase in the swelling and solubility parameters of the PEG-based polymer, which in turn effected a change in the  $^{19}\text{F}$  relaxivity (**Scheme 1**). The relaxivity change was investigated by  $^{19}\text{F}$  NMR as a measure of release of the therapeutic.



**Scheme 1.** The  $^{19}\text{F}$  switch resulting from the structure-property relationship change of a hyperbranched polymer as a therapeutic is released.

<sup>1</sup> B.E. Rolfe, I. Blakey, O. Squires, H. Peng, N.R. Boase, C. Alexander, P.G. Parsons, G.M. Boyle, A.K. Whittaker, K.J. Thurecht, *J. Am. Chem. Soc.* **2014**, 136, 2413-2419.

<sup>2</sup> K. Wang, H. Peng, K. J. Thurecht, S. Puttick, A. K. Whittaker, *Polym. Chem.*, **2014**, 5, 1760-1771.

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## Engaging the sharks in a tumour attack – single domain antibodies for specific targeting of nanomedicines

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Using antibodies (Abs) as active targeting agents for nanomedicine is a developing field in cancer therapy giving the possibility to deliver drugs to selected sites with high specificity. Several Abs and conjugates thereof are already approved by US FDA for drug delivery<sup>1</sup>. However, the relatively large size of Abs (~150 kDa), can be a disadvantage as potential drug conjugates increases dramatically in size when Abs are attached, consequently limiting tumour penetration and altering biodistribution<sup>2</sup>. Downsizing these molecules and utilizing only the antigen recognition region, so-called single chain variable fragments (scFvs), is one advantageous strategy to accommodate this problem, resulting in ~27 kDa molecules<sup>3</sup>.

An even smaller alternative at half the size of scFvs are single domain antibodies (sdAbs), derived from heavy chain only Abs (HCAs) found in camelids and cartilaginous fishes (e.g. sharks)<sup>4</sup>. In addition to their smaller size several advantages exist using sdAbs over scFvs; production costs are significantly lower, upscaling is straightforward, and they show high stability at various temperatures and pH<sup>5,6</sup>, making sdAbs useful in various biomedical applications.

In order to exploit the promising properties of sdAbs for drug delivery purposes, we intend to use shark variant sdAbs (vNARs) to actively target tumour tissues with polymer nanoparticles already developed at the Centre for Advanced Imaging<sup>7</sup>. By attaching cancer marker specific vNARs to polymers holding therapeutic drugs and imaging agents (**Fig. 1**) sophisticated theranostic nanomedicines can be prepared. A phage library has been acquired from *Orectolobus ornatus* (ornate wobbegong shark) immunised with human cancer cell membranes. Panning this library, it has been possible to identify and isolate HCAs with affinity for specific cancer markers. From the isolated HCAs, the vNAR coding DNA could be derived for expression. With this promising new targeting strategy, we are confident that vNARs in conjugation with nanomaterials have great potential in theranostics.

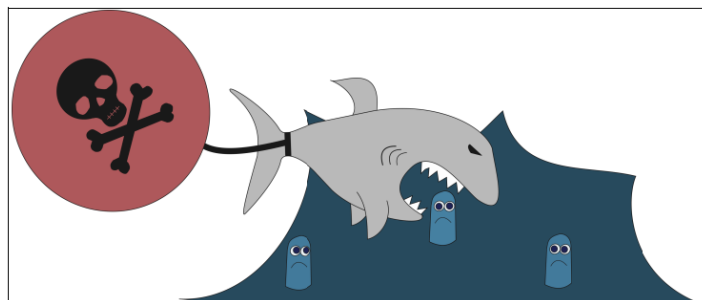


**Figure 1.** Illustration of vNAR delivering anti-cancer drug to tumour tissue. vNAR (shark) carrying nanomedicine (red bubble) binds to specific surface protein (blue rod) of cancer cell (petrol).

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<sup>6</sup> Griffiths, K.; Dolezal, O.; Parisi, K.; Angerosa, J.; Dogovski, C.; Barraclough, M.; Sanalla, A.; Casey, J.; González, I.; Perugini, M.; Nuttall, S.; Foley, M. *Antibodies* **2013**, 2, 66.

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# Stimuli-Responsive Smart Soft Materials

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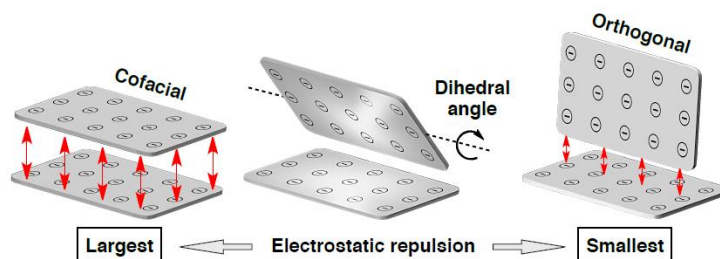
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Machine technology frequently puts magnetic or electrostatic repulsive forces to practical use, as in maglev trains, vehicle suspensions or non-contact bearings. In contrast, materials design overwhelmingly focuses on attractive interactions, such as in the many advanced polymer-based composites, where

inorganic fillers interact with a polymer matrix to improve mechanical properties. However, articular cartilage strikingly illustrates how electrostatic repulsion can be harnessed to achieve unparalleled functional efficiency: it permits virtually frictionless mechanical motion within joints, even under high compression. Here we describe a composite hydrogel with anisotropic mechanical properties dominated by electrostatic repulsion between negatively charged unilamellar titanate nanosheets embedded within it. Crucial to the behaviour of this hydrogel is the serendipitous discovery of cofacial nanosheet alignment in aqueous colloidal dispersions subjected to a strong magnetic field, which maximizes electrostatic repulsion<sup>6</sup> and thereby induces a quasi-crystalline structural ordering over macroscopic length scales and with uniformly large face-to-face nanosheet separation. We fix this transiently induced structural order by transforming the dispersion into a hydrogel using light-triggered *in situ* vinyl polymerization. The resultant hydrogel, containing charged inorganic structures that align cofacially in a magnetic flux, deforms easily under shear forces applied parallel to the embedded nanosheets yet resists compressive forces applied orthogonally. We anticipate that the concept of embedding anisotropic repulsive electrostatics within a composite material, inspired by articular cartilage, will open up new possibilities for developing soft materials with unusual functions.

More recently, we reported a novel carbon nitride polymer, which shows anomalous mechanical responses to minute fluctuations in the ambient humidity.



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- [3] M. Liu, Y. Ishida, Y. Ebina, T. Sasaki, T. Hikima, M. Takata, and T. Aida, *Nature* **2015**, 517, 68–72.
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**A**

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**B**

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| Fellows, C        | 109, 226          | Forsythe, J | 92, 94, 95, 96, 169, 195 |
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| Finnveden, M      | 114               | Fox, P      | 60                       |
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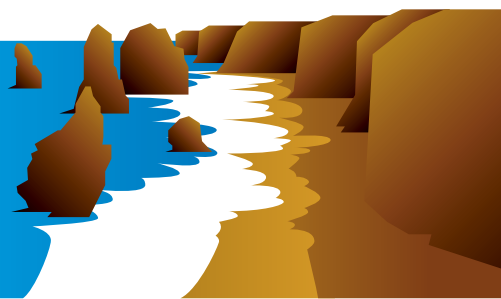
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# POSTER ABSTRACTS



# Alginate Based Core-Shell Nanoparticles for Encapsulation

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Alginate is an abundant natural polysaccharide which has been thoroughly investigated in the realm of drug & cell delivery as well as tissue engineering, primarily because it's structural conformation closely resembles extra-cellular matrices and it demonstrates little to no immune response with a high biocompatibility. Alginate cross-links readily in the presence of divalent cations (eg.  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{Ba}^{2+}$ ) – a function which has been long exploited by the personal care, medical and culinary industries in encapsulation strategies. However, the true potential of these polymers have not been realized due to limited modification options.<sup>1</sup> There have been no reported instances where alginate has been modified while still affording it's cross-linking characteristics.

This paper seeks to demonstrate how alginates can be polymer functionalized via either the 'click' approach or by controlled radical polymerization, thereby affording access to previously unreported comb copolymers which retain alginate's natural ability to crosslink. In the case of hydrophilic grafted polymer chains such as PEG, it allows the copolymer the ability to self-assemble into core-crosslinked nanoparticles upon the addition of calcium ions (**Fig. 1**). The PEG chains stabilize the polymer and afford a directed self-assembly and do so once a critical amount calcium is added (**Fig. 1b**).

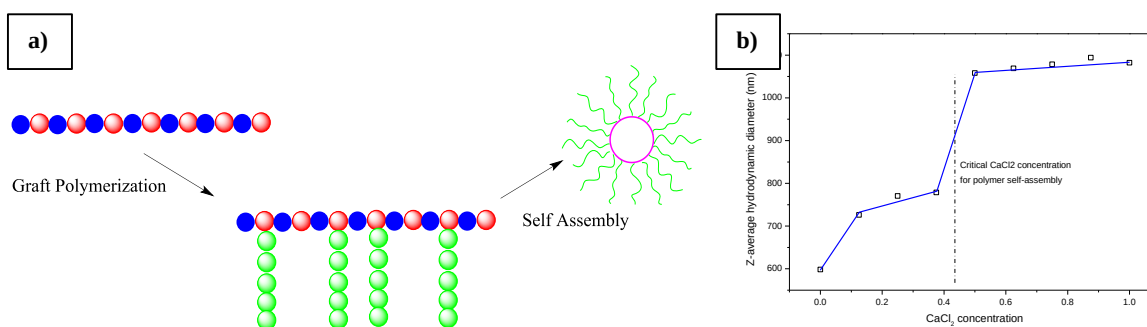


**Figure 1.** (a) Access to alginate based comb copolymers which self-assemble into core-crosslinked nanocapsules upon the introduction of a divalent cation; (b) z-average hydrodynamic diameter of alginate-comb-poly(polyethylene glycol methacrylate) with respect to  $\text{CaCl}_2$ , showing a large jump in particle size once the concentration exceeds 0.4M.

This approach demonstrates a strategy towards well defined, biocompatible encapsulation materials that could afford a high degree of ease in self-assembly, while ensuring the production of well-defined nanoparticles of controlled size and morphology – something unprecedented in present polymer technology.

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# Synthesis of Renewable Bio-based Acrylic Homo- and Co-polymers

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Substitution of petrochemical-sourced monomers by bio or renewable monomers is of much interested form environment reasons. Almost all acrylic monomers used by industry are produced from non-renewable resources. Bio-acrylics are considered to be promising materials because of their carbon neutrality and durability. In this work, the synthesis of fully bio-derived acrylic homo and co-polymers are reported. A new furanone obtained from a bio-source is converted into acrylic monomers and is successfully homo-polymerized and co-polymerized with other acrylic monomers (also obtained from bio-sources) using free-radical solution polymerization. The structure of the derived monomer and subsequent polymers are confirmed by a range of chemical, spectroscopic and physical techniques, and their materials engineering properties determined.

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## Graphene coated scaffold for brain stimulation and recording

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A novel electrode for brain stimulation and recording was engineered by combining an electrospun poly-ε-caprolactone (PCL) scaffold and graphene. Graphene has received a lot of attention due to its electrical and mechanical properties. In this case, it is used as a source of electrical conductivity for the non-conductive PCL scaffold. Graphene oxide, a non-conductive material, was reduced to its conductive form using vitamin C. It is hypothesised that the PCL scaffold coated with reduced graphene oxide (rGO) will enhance axonal extension, support the formation of neurite network and synaptic communication of primary cortical neurons. The scaffold is fabricated via an electrospinning technique which allows the properties of scaffold such as fibre diameter, inter-fibre spacing and thickness to be tailored to match the desired application.

The fibrous scaffold has a uniform fibre diameter of  $\sim 1\ \mu\text{m}$  with a random fibre orientation (Figure 1). PCL is chosen due to its biodegradable and biocompatible nature. Reduced graphene oxide is coated on to the scaffold via a filtration technique<sup>1</sup>. The live/dead assay and MAP-2 (axon marker) immunostaining (Figure 2) results showed that rGO coated electrospun scaffold was biocompatible. It was able to provide a platform for the attachment, proliferation and axonal extension of primary neurons. Calcium imaging was used to identify the formation of neurite network and synaptic communication.

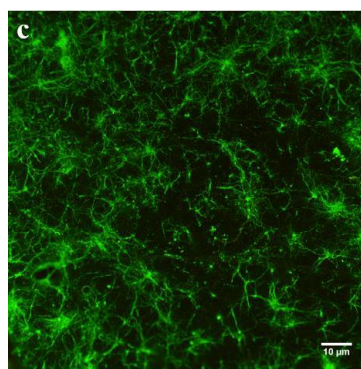
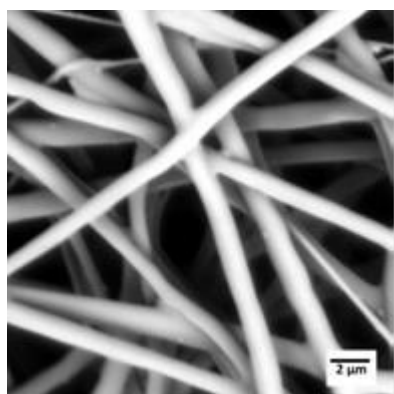


Figure 2. Immunostaining of dendrites by MAP-2 (green) on primary cortical neurons cultured for 9 days on rGO scaffold.

Figure 1. SEM image of randomly orientated electrospun PCL fibres.

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1 ZQ, Feng. T, Wang. B, Zhao. J, Li. L, Jin. *Advanced Materials*. **2015**, 27, 6462-6466

## Polymer-Inorganic Hybrid Nanoparticles via Polymerization-Induced Self Assembly

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Polymerization-induced self-assembly (PISA) represents a new method of preparing polymer nanoparticles with variety of morphologies, such as spheres, rods and vesicles<sup>[1]</sup>. The PISA approach uses controlled radical polymerization to grow polymer chains in a controlled manner with minimal termination of polymer chains, where unique particle morphologies can be targeted based on the length of the polymer. This process is illustrated as Figure 1. An interesting class of monomers that find application for the preparation of polymer-inorganic hybrid materials are silane-containing monomers, such as 3-methacryloxypropyltrimethoxysilane (MPS). This monomer contains a specific functional group to enable the preparation of silica nanoparticles by a sol-gel process, and has been previously used to make polymer-silica nanoparticles by emulsion polymerization<sup>[2, 3]</sup>.

In this work, silane-containing monomers such as MPS are utilised to produce nanoparticles of controllable morphology via Reversible Addition-Fragmentation chain Transfer (RAFT) and the PISA process. Si-containing monomers are used as the solvophilic block in the PISA process for the first time, followed by chain extension to form an insoluble polymer in a given solvent (e.g. benzyl methacrylate in ethanol). These nanoparticles serve as a template to create new classes of silica nanospheres, rods or vesicles via the controlled growth of the silica shell at the particle interface. Experimental factors (e.g. the length of the silane-containing block, solvent, temperature and co-monomer type) are demonstrated in how they influence the ability to control nanoparticle architecture. In comparison to traditional methods for the preparation of silica nanoparticles (e.g. the Stöber method)<sup>[4]</sup>, this approach enables access to normally inaccessible hybrid nanoparticle architectures.

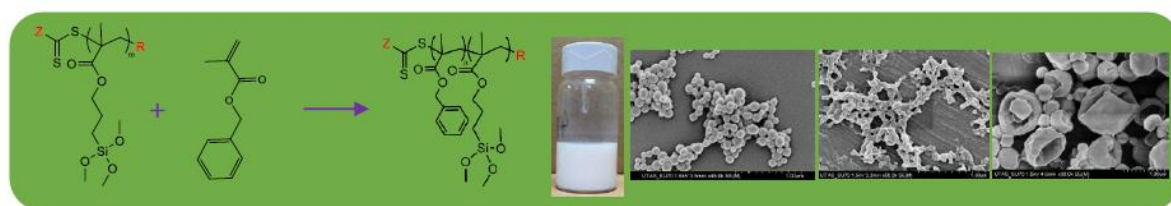


**Figure 1:** Pathway to form different morphology through RAFT and PISA process

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# Soft, porous, polymeric material as scaffolds for brain tissue engineering using human pluripotent stem cells

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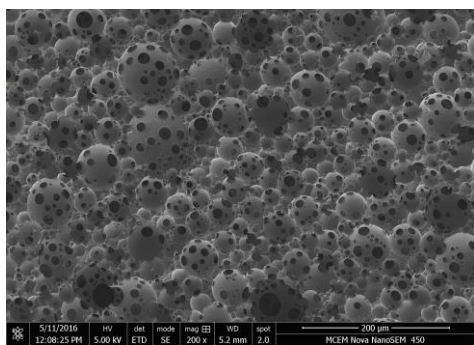
Cells cultured in flat, stiff 2D environments results in cultures with irregular cell morphology and unnatural cell-cell interactions<sup>1</sup>. Cells cultured in 3D, typically with aid of a scaffold, have shown improved physiological relevance in term of important cell functions. 3D cell culture and tissue engineering aim to bridge the gap between traditional 2D cell culture and natural cellular physiology.

Tissue engineering scaffolds come in many formats with the most popular being hydrogels, electrospun fibers and porous polymers. Whilst all systems have distinct advantages, porous polymers can be easily manufactured, stored over long term, allow more natural cell migration and give easy access for nutrient/waste diffusion.

Porous polymers prepared by the emulsion templating technique allow for the fabrication of interconnected porous-network polymers, in a simple and versatile manner<sup>2</sup> (**Fig. 1**). The process allow for fine control over porosity and pore size distribution. Porous polymers produced via 'click chemistry' reactions allow for rapid, highly efficient polymerization which can be particularly beneficial when preparing emulsions of low stability. The 'thiol-ene' click reaction is one such example<sup>3</sup>.

When engineering tissue, it is ideal for the tissue culture scaffold to resemble the natural extracellular matrix as much possible. The human brain extracellular matrix (ECM) is composed mainly of the proteins laminin and fibronectin, and has an elastic modulus ranging from 0.1-16kPa<sup>4</sup>. It is therefore ideal for the scaffold to mimic this modulus.

The work presented here aims at producing an emulsion templated porous polymer with a modulus in the range of 0.1-16kPa. Significant challenges exist in preserving the intricate microstructure of the material at such low moduli. By experimenting with crosslinking density though long chain multifunctional monomers (**Fig. 2**) it is hypothesized that the material will become softer, closer resembling that of the natural human brain ECM.



<sup>1</sup>Carletti,

**Figure 1.** SEM image of porous polymer

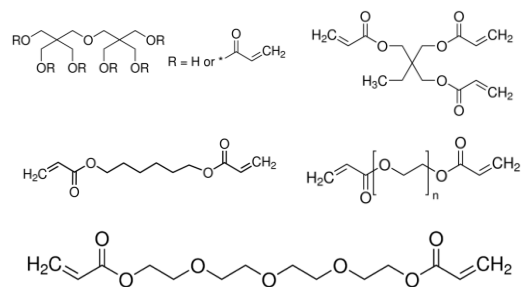


**Figure 2.** Multifunctional acrylate monomers investigated for the synthesis of soft porous polymers

E., A. Motta, and C. Migliaresi, *Scaffolds for Tissue Engineering and 3D Cell Culture*, in *3D Cell Culture*, J.W. Haycock, Editor. 2011, Humana Press. p. 17-39

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# Synthesis of Iron Oxide Nanoparticles Grafted with Polyethylene Glycol and Poly(2-ethyl-2-oxazoline)s Brushes for Protein Avoidance

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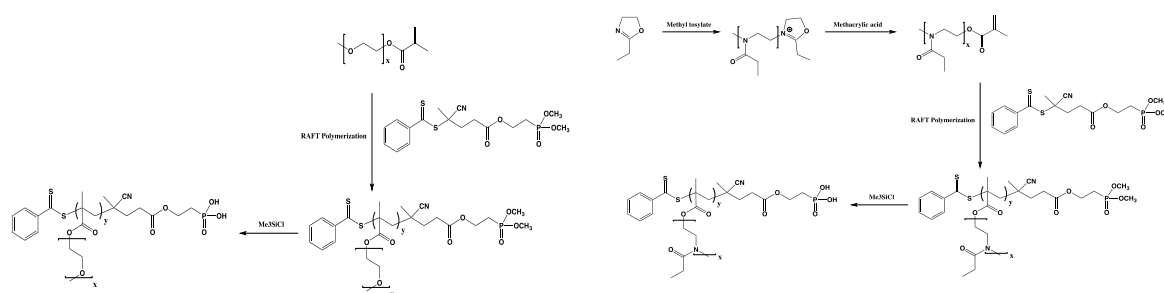
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PEGylation is a common grafting strategy in ascribing the stealth effect to engineered nanostructures against immune responses.<sup>1</sup> This strategy utilises the steric repulsion and hydrogen bonding capacities of polyethylene glycol (PEG) to confer a resistance to the adsorption of plasma proteins.<sup>2</sup> However, chronic administration of PEG renders accumulation of the polymer *in vivo* with unknown consequences. In addition, repeated dosing of PEG gives rise to accelerated blood clearance,<sup>3</sup> compromising the circulation of PEGylated drugs and carriers.

Poly(2-oxazoline)s (POx) and zwitterionic polymers are viable alternatives to PEG in terms of biocompatibility and stealth behavior.<sup>4,5</sup> POx, in particular, are an emerging polymer class which have attracted significant attention during the last decades as they provide straightforward access to functional polymers with tuneable properties as opposed to PEG.

Superparamagnetic iron oxide nanoparticles (IONPs) possess tailorable biodistribution and pharmacokinetic properties.<sup>1</sup> Consequently, IONPs have found broad applications in magnetic resonance imaging, thermoablation therapy, sensing, and targeted drug delivery. We have synthesised IONPs grafted with PEG and POx brushes (Scheme). The synthesis of PEG and POx was performed by reversible addition-fragmentation chain-transfer (RAFT) polymerisation and cationic ring opening polymerization, respectively. The IONPs grafted with PEG and POx were then characterised by NMR, GPC, DLS, TEM and TGA.



**Scheme 1.** Synthesis of PEG and POx brushes grafting on IONPs

In addition to synthesis, we will present our results on the antifouling behaviours of the IONPs against plasma proteins.

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<sup>3</sup> N. J. Butcher, G. M. Mortimer and R. F. Minchin, *Nature Nanotechnol.* **2016**, *11*, 310-311.

<sup>4</sup> T. Blin, A. Kaminen, E. H. Pilkington, A. Ivask, F. Ding, J. F. Quinn, M. R. Whittaker, P. C. Ke and T. P. Davis, *Polym. Chem.* **2016**, *7*, 1931-1944.

<sup>5</sup> L. Tauhardt, K. Kempe, M. Gottschaldt and U. S. Schubert, *Chem. Soc. Rev.* **2013**, *42*, 7998-8011.

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## Novel Film Forming Polymer Latexes

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One method of obtaining chemically prepared polymer latexes is through the process of emulsion polymerisation. Emulsions formed at the initial stage are thermodynamically unstable and stabilisation against coalescence is achieved through additives that absorb on to the liquid-liquid interface and provide electrostatic, steric and rheological resistance that prevents recombination or droplet collision. This is achieved primarily through the use of surfactants which are a common emulsion stabiliser. Other stabilisers include the use of different types of polymers or colloids. Colloids can be utilized at a liquid-liquid interface forming Pickering emulsions that are stable against coalescence for a wider range of coverage of the droplet surface and facilitate adsorption at the interface<sup>1</sup>. These particle stabilised emulsions retain the basic properties of emulsions stabilised by surfactants<sup>2</sup> and can therefore, effectively replace surfactants<sup>3</sup>. Pickering emulsions have a significant advantage over the use of surfactants wherein they have a high stability against coalescence brought about by the solid nanoparticles. The smaller solid stabilising particles allow stabilisation of both micro-droplets and larger droplets as well which, like surfactants, takes place at the surface of emulsion droplets through the adsorption of solid particles. Bon et.al have reported emulsion polymerization where colloidal stability is provided by nanosized particles for polymer latexes<sup>4</sup> which can be processed for the production of paint. The result is a polymer latex surrounded by a layer of nanoparticles shown in Figure 1. The use of polymer latexes that have morphologies that are multilayered or are armored in nature, can be applied to waterborne adhesives and coatings.

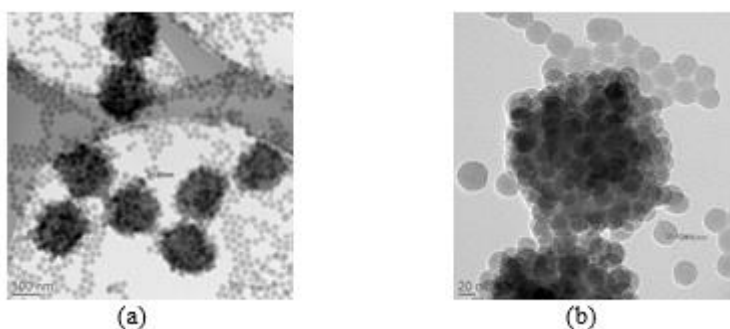


Figure 1(a): TEM image of Latex armoured with nanoparticle at a scale of 100 nm (b) TEM image of Latex armoured with nanoparticle at a scale of 20 nm

- 1 Pawar, A.B., et al., Arrested coalescence in Pickering emulsions. *Soft Matter*, 2011. 7(17): p. 7710.
- 2 Chevalier, Y. and M.-A. Bolzinger, Emulsions stabilized with solid nanoparticles: Pickering emulsions. *Colloids and Surfaces A: Physicochemical and Engineering Aspects*, 2013. 439: p. 23-34.
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# Injectable Composite Biomaterials for Cartilage Tissue Engineering

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Injectable materials and hydrogels in particular have become a promising technique for tissue repair[1]. However, weak properties of some these materials limit their application. In this study, composite materials have been fabricated from blend of hydrogels, which are polyethylene glycol diacrylate (PEGDA) and methacrylated hyaluronic acid (Me-HA) in one system and PEGDA and methacrylated alginate Me-A in the other, and porous particles of polyHIPE which has appropriate properties to be used in applications of tissue regeneration[2]. These materials together may combat the drawbacks of using hydrogels alone especially in terms of poor mechanical properties and also provides porous particles which can promote cell proliferation and migration within this composite system. Rheological and other experiments have been carried out to investigate the achievability of using this system and materials for this purpose. The dynamic results have illustrated that storage modulus increased with increasing polyHIPE particles and it has been shown to be tuneable according to the ratios of materials used. It is expected that these injectable biomaterials will supply an effective contribution to solve some problems related to tissue regeneration.

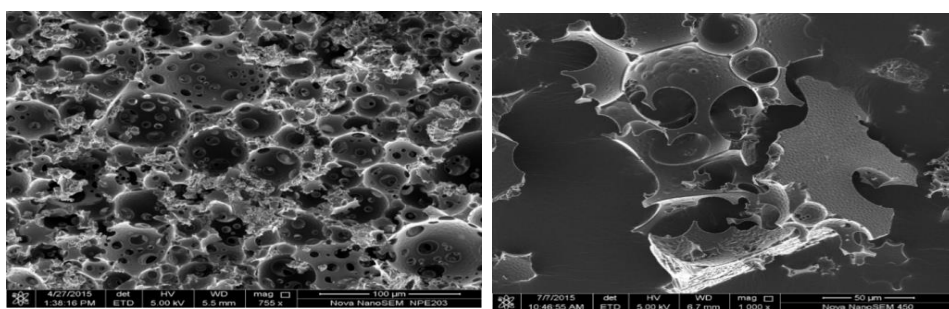


Fig.1. SEM images of a) polyHIPE sheet b) polyHIPE particles.

Fig.2. Effect of polyHIPE concentration on storage modulus of PEGDA hydrogels

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# The development of injectable hydrogels with human amnion epithelial cells (hAECs) for the treatment of ischaemic stroke

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Ischaemic stroke refers to the sudden disruption of blood flow to regions of the brain, leading to neuronal cell death, transient functional loss and potentially permanent neurological disability. Current treatments primarily focus on early intervention by promoting reperfusion using pharmacological agents or surgical means but these have critical temporal limitations. Unfortunately, there is also a lack of viable long-term therapies available to patients to reverse any resultant cellular damage and reinnervate impaired neuronal connections. To address this, substantial attention has been directed to the growing field of stem cell therapy. Human amnion epithelial cells (hAECs), sourced from the placenta, are a promising candidate<sup>1</sup>. Previously, they have been shown to be effective in treating intracerebral haemorrhage in rats<sup>2</sup> and reducing foetal brain injury induced by intrauterine inflammation in sheep<sup>3</sup>.

To help promote the survivability and effectiveness of the hAECs once implanted in the stroke brain, they can be injected along with a hydrogel that will gelate *in situ* to encapsulate and protect the cells. A hyaluronic acid-PEG hydrogel that is crosslinked via a biorthogonal click reaction based on the nitrile oxide-norbornene cycloaddition<sup>4</sup> is being developed for this purpose. The hydrogels are injectable and can be tuned to possess a modulus that is akin to cerebral tissue which allows them to simulate the 3D extracellular matrix environment that is more conducive to cellular growth. The gelation of the hyaluronic acid based hydrogels were characterised using rheology and swelling measurements and the efficacy of the hydrogel assessed by 3D culture of hAECs.

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<sup>2</sup> W. Dong, H. Chen, X. Yang, L. Guo, G. Hui, *Cell Biol Int.* **2010**, 34, 573-577

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## Investigation of Osmosis Driven Burst Release Systems for Vaccine Delivery

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Vaccination schedules often require administration of a secondary dose of antigen in order to elicit an adequate immune response. For treatment of wild animals, repeat administration requires recapture, which can be logistically challenging and stressful for the animal.<sup>1</sup> There is a need for an autonomously triggered delivery device, that will provide a delayed, controlled release of vaccine at a physiologically relevant time. This research investigates the use of osmosis triggered release of a payload from a polymer capsule, implanted under the skin, for the administration of a chlamydia vaccine to koalas. The concept involves a polymer capsule, made from hydrophobic and hydrophilic monomers, to allow for uptake of water from the surrounding tissue, filled with the vaccine and an osmotic agent. As the capsule swells the pressure on the polymer shell would increase and eventually rupture, releasing the antigen (**Fig 1**).

Polycaprolactone was fabricated into tube shaped capsules using a mould. Burst pressure and release characteristics of the capsules were assessed and physico-chemical properties of polymers examined. Osmosis driven burst tests demonstrated that the rupture of these capsules can be delayed by a number of weeks before the payload is released. Development of a controlled-release, drug delivery system capable of producing a delayed, pulse-like dose of vaccine could allow for single administration vaccination of wild animals and also provide a means of vaccine delivery to isolated human populations, which could minimise the problem of poor patient compliance.

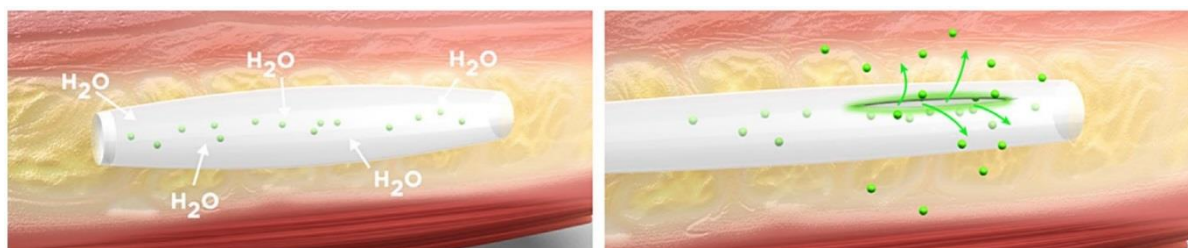


**Figure 1.** Schematic diagram of a polymeric capsule implanted under the skin, the semipermeable polymer capsule allows uptake of water by osmosis (left). The capsule swells until it ruptures and the vaccine is released (right).<sup>2</sup>

A. Kollipara, C. George, J. Hanger, J. Loader, A. Polkinghorne, K. Beagley, P. Timms, *Vaccine* **2012**, *30*, 1875-1885

<sup>2</sup> F.P.W. Melchels, I. Fehr, A.S. Reitz, U. Dunker, K.W. Beagley, T.R. Dargaville, D.W. Hutmacher, *Biotechnol. Bioeng.* **2015**, *112*, 1927-1935

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# Porous polymer scaffolds for 3D culture and maintenance of pluripotent stem cells

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While pluripotent stem cells have the potential to overcome many problems in areas such as tissue engineering and cell replacement therapies, their use is hindered by the difficulty of producing the large amounts of high quality cells required. Current culture methods are labour intensive, low throughput, and may rely on animal derived factors. Therefore, there is a strong requirement for a method that can produce high quantities of reliable pluripotent stem cells under fully defined conditions.

3D cell culture offers a viable method for scaled up production. 3D scaffolds need to provide structural support, allow cell migration and maintain pluripotency. PolyHIPEs are a potential 3D scaffold. They are produced by polymerising a high internal phase emulsion (HIPE) to produce a porous foam (polyHIPE). They possess an interconnected structure that is suitable for cell culture as it allows cell migration, nutrient delivery, and waste removal. The pore size of polyHIPEs can be tuned easily via heating, shear rate, or addition of destabilising agents<sup>1</sup>.

PolyHIPEs have been shown to support cell growth<sup>2</sup>, and are open to modifications that may allow them to support the unique requirements of pluripotent cells. Addition of ECM peptides, for example, can mimic the stem cell niche and help maintain pluripotency.

PolyHIPEs are produced via a photo-initiated reaction between a thiol and a multi-functional acrylate in emulsion with water. The resulting emulsion is cast in a mould and exposed to high intensity UV light. This method allows the polyHIPE to be made in any shape up to 5cm thickness. The final shapes can be cut using a microtome.

Morphology is determined using SEM. Pore size is calculated by SEM image analysis using ImageJ processing software.

After confirming the void morphology is suitable, the surface is functionalised. Currently, maleimide is being attached via a Michael reaction with residual surface thiols. This process is being optimised with the aim towards functionalising the polyHIPE with molecules which promote cell attachment, growth and pluripotency.

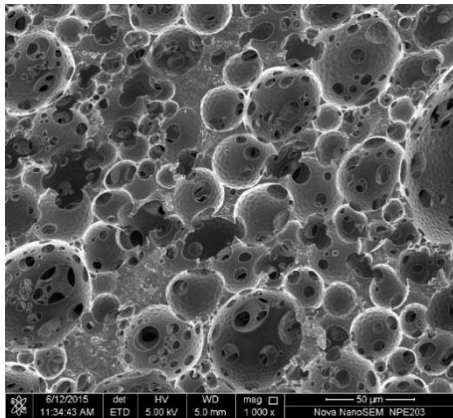


**Figure 2:** SEM image of as-synthesised polyHIPE

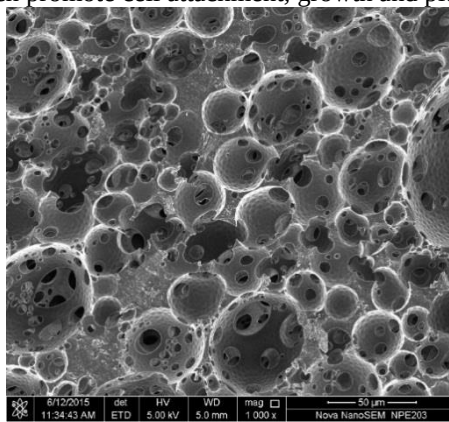


**Figure 2** shows the typical morphology of the thiol-acrylate polyHIPE. The larger cavities are referred to as ‘voids’ and the smaller connecting holes are known as ‘windows’. The average void size is  $38 \pm 16 \mu\text{m}$ . The large pores and interconnectivity of the scaffold are suited for cell

attachment and migration.

Raman spectroscopy was used to track the surface functionalisation with maleimide and showed decrease in the scaffold thiol peak as the maleimide ratio WAS increased. This trend was confirmed using XPS, and quantified using Ellman’s assays.

The polymer scaffolds investigated in this project have already proven suitable for 3D cell culture, and with further modification could be tailored to better mimic the niche environment of pluripotent stem cells. The ability to culture such cells in 3D while maintaining pluripotency would have a strong downstream impact on tissue engineering, cell replacement, toxicology screening and other technologies that make use of pluripotent stem cells.

<sup>1</sup> C. Langford, D. Johnson, N. Cameron, Polymer Chemistry, 2014. 5(21), 6200-6206

<sup>2</sup> S.Caldwell, et al., Soft Matter, 2012. 8(40), 10344-10351

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## pH responsive nanoparticles to target pain

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The Neurokinin 1 receptor (NK<sub>1</sub>R) is a G protein-coupled receptor that is stimulated by the neuropeptide substance P to mediate inflammation and central pain transmission<sup>1</sup>. With stimulation at the cell surface, the NK<sub>1</sub>R mediates plasma membrane signaling events and is then rapidly internalised into endosomes. Recent evidence from our laboratory indicates that NK<sub>1</sub>R can form unique signalosome complexes within endosomes, initiating signaling events that are spatially and temporally distinct from those initiated at the cell surface. Furthermore, we have demonstrated that signaling events mediated by endosomal NK<sub>1</sub>R are consistent with pain transmission. Selectively delivering drugs to endosomes to inhibit intracellular NK<sub>1</sub>R signaling therefore offers a novel approach for regulation of pain. The acidified environment of the endosomes can be exploited to deliver an NK<sub>1</sub>R antagonist exclusively into the endosome using pH responsive nanoparticles that disassemble under acidic conditions. Most importantly, this selective delivery of a NK<sub>1</sub>R antagonist must occur without endosomal disruption or loss of cell viability.

Nanoparticles loaded with the NK<sub>1</sub>R antagonist aprepitant were co-assembled using a hydrophobic block incorporating 2-(diisopropylamino)ethyl methacrylate (DiPAEMA) units with a pK<sub>a</sub> of 6.1 (responsible for the pH responsive disassembly), and a hydrophilic block incorporating di(ethylene glycol) methyl ether methacrylate (DEGMA), 2-(dimethylamino)ethyl methacrylate (DMAEMA) and poly(ethylene glycol) monomethyl ether methacrylate (PEGMA) units to increase cell viability, cellular uptake and long circulating capabilities, respectively. Dynamic light scattering results showed that the nanoparticles exhibited a size of 22.5nm at physiological pH. This size decreases to 6.4nm at pH 5, correlating with the pH-triggered disassembly of the DiPAEMA block. This is caused by protonation and a hydrophobic-hydrophilic solubility switch of the DiPAEMA units within the core of the particle. MTT cell viability assays were also performed to test the toxicity of the nanoparticles compared to the toxicity of the free aprepitant. Finally, we used FRET biosensors to measure compartmentalised signaling events that are initiated at the cell surface (cytoplasmic ERK activity) or from endosomes (nuclear ERK activity). The targeting and toxicity of the nanoparticles and their ability to regulate endosomal signaling events are discussed.

1. M. S. Steinhoff, B. von Mentzer, P. Geppetti, C. Pothoulakis, N. W. Bunnett. *Physiol Rev.* **2014**, 94, 265-301

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# Topochemical Polymerization of bis-thyminyl monomer in Microemulsion

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Topochemical polymerization is a special crystal-to-crystal reaction. Recently, we have developed a new monomer from alkyl-bis thymine that undergoes topochemical polymerization by the photo-reversible  $[2\pi + 2\pi]$ -cycloaddition of thymine units (Scheme 1).<sup>1</sup>

In the crystal structure of bis-thyminyl monomers, the molecules are stacked with thymine rings into tight layers from neighbouring molecules stacking on top of each other (Fig.2). The polymerization was conducted by irradiating the stacked thymine rings with UV light  $>270$  nm, the cyclobutane ring is formed which connects the stacked molecules into the polymer chains (Scheme 2). The successful of this polymerization due to the proper monomer alignment in the crystal.<sup>2</sup> This reaction done in the solid state, so it does not need initiator or addition of monomer for the reaction.

In this study, dimerization and polymerization of bis-thyminyl monomers were conducted in different types of microemulsions. The size of microemulsions was measured by dynamic light scattering (DLS) and small angle neutron scattering (SANS). GPC was used to determine the molecular weight and the molecular weight distribution of products from the microemulsion polymerization. Polymerization results will be discussed in detail.

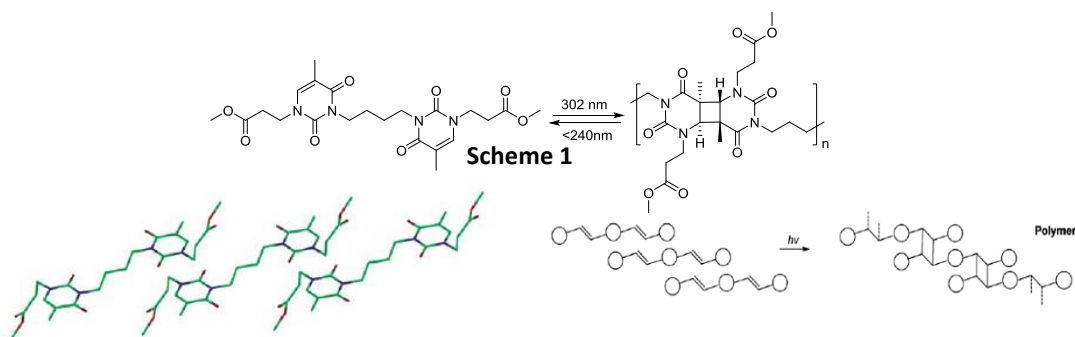


**Figure 2.** Molecular stacking of alkyl linked Bis-thymine in Crystal

**Scheme 2**

<sup>1</sup> P. Johnston, C. Braybrook and K. Saito, *Chem.Sci.*, **2012**, 3, 2301-2306.

<sup>2</sup> G. Kaur, P. Johnston and K. Saito, *Polym.Chem.*, **2014**, 5, 2171-2186

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# Effect of ZnO and TiO<sub>2</sub> particle size reduction on the dielectric properties of natural rubber composites

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Natural rubber (NR) is mostly used in the form of solid vulcanized rubber, which is employed in the production of thin-walled products such as surgical gloves, balloons, tires, and other applications<sup>1</sup>. In addition to the vulcanization additives and accelerators of the vulcanization process, many other compounding ingredients are present in the commercial product, including fillers, pigments, plasticizers, reinforcing resins, and others<sup>2</sup>. In this investigation we evaluated the influence of the reduction of particle size, such as zinc oxide and titanium oxide, in the dielectric properties of vulcanized rubber films. Fig. 1a plots the leakage current versus voltage until breakdown conditions. The addition of nano-sized filler in the latex formulation decreased the leakage current and increased the breakdown voltage values when the solid particles were sonicated for 10 min. Therefore, the addition of nano-filler particles favours the reduction of defects inside the composite material and also improves the local electric field dissipation.

**Figure 1.** a) AC polarisation curves obtained from breakdown strength measurements for nanocomposite rubber and b) Optical micrographs from the hole obtained after breakdown experiments (circles are related to agglomerated particles inside polymer film).

1. M. Wang , L. Mi , Y. Wu , J. Zhang and Q. Fang, J. Therm. Anal. Calorim., **2014**, 115:1173–118.
2. M. Mariano, N. Kissi and A. Dufresne, Carbohydrate Polymer, **2016**, 137: 174–183.

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**Research interests:** Natural Rubber, nanocomposite, polymer synthesis and characterization

# Preparation of natural rubber nanocomposites containing functionalized BaTiO<sub>3</sub> nanoparticles for electrical insulating applications

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Green nanocomposites for electrical insulating applications can be manufactured by combining a natural polymer, such as the natural rubber (NR), with ferroelectric ceramics, like barium titanate (BaTiO<sub>3</sub>). This filler has been intensively investigated due to its high dielectric constant<sup>1</sup>. However, BaTiO<sub>3</sub> has a poor compatibility with polymer matrices, and also its high surface energy promotes nanoparticles agglomeration. Therefore, in order to improve their dispersion into polymer matrices there are several surface modification methods, like: hydroxylation, amination, sulphonation, and grafting polymers<sup>2</sup>. The aim of the present work is to improve the compatibility of NR polymer matrix with BaTiO<sub>3</sub> nanoparticles. For this purpose, the BaTiO<sub>3</sub> particles were chemically modified with H<sub>2</sub>O<sub>2</sub> to obtain OH groups on their surface (Fig. 1a). The hydroxylation process was carried out by the reaction of 10 g of BaTiO<sub>3</sub> nanoparticles and 80 ml of H<sub>2</sub>O<sub>2</sub> aqueous solution (30 wt%) and refluxed at 105 °C for 24 h<sup>3</sup>. Figure 1b shows the plot of the leakage current versus voltage until breakdown. It can be observed that the addition of BaTiO<sub>3</sub> nanofillers 0.25wt %, 0.5 wt %, 1 wt %, 5 wt % and 10wt %) in the latex formulation increases the breakdown voltage. Therefore, the dielectric strength of the material has been improved. Chemical impedance analysis results also indicated that the composite film is highly insulating due to the better compatibility of the NR with the ceramic particles, thus decreasing the ionic conductivity inside the polymer matrix (Fig. 1c).

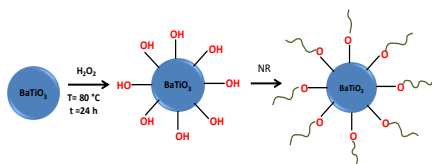


**Figure 1.** a) Functionalization of BaTiO<sub>3</sub> with H<sub>2</sub>O<sub>2</sub>, b) AC curves obtained from breakdown strength analysis and c) Nyquist plots for pristine NR and NR with different concentrations of BaTiO<sub>3</sub> chemically modified.

1. N. Gonzalez, M. Custal, S. Lalaouna, J.R. Riba, E. Armelin, Eur. Polym. J., 75, 210–222, 2015.
2. H. Tang; P. Wang; P. Zheng and L. Xiaobo, Compos. Sci. Technol., 2016, 123, 134–142.
3. Z. Tao, Z. Jun-Wei, C. Rui-Yao and D. Zhi-Min, ACS Appl. Mater. Interfaces, 2011, 3, 2184–2188.

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**Research interests:** Natural Rubber, nanocomposite, polymer synthesis and characterizatio

## Complex nanoprobes for highly selective multimodal imaging

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Biomedical imaging is an important diagnostic tool that provides basic physiological, anatomical and molecular information to detect diseases. The challenge of biomedical imaging is that none of the current techniques offer a universal solution to detect, diagnose and monitor disease; instead a combination of imaging techniques is often used together.<sup>1</sup> This highlights the need for novel complex multimodal nanoprobes. This project aims to develop and assess a novel theranostic nanoparticle for application as a multimodal imaging agent in magnetic resonance imaging (MRI) and positron emission tomography (PET).

MRI is an imaging technique that produces 3D tomography images with anatomical and physiological information.<sup>2</sup> Contrast agents, such as gadolinium chelates, are used to improve the contrast between different tissues. Despite the use of contrast agents, there is no selectivity towards tumorous tissue to help facilitate the detection of disease. To enable MRI to indicate tumorous tissue, a nanoprobe that is responsive to the tumour microenvironment is of interest. The external environment of cancerous cell is slightly more acid than healthy tissue, hence a nanoprobe that has pH responsive component can exploit the different in pH to selectively image tumour tissue.

In this work, nanoprobes incorporating a pH responsive polymer are used to moderate the activity of gadolinium chelates in order to manipulate the MRI signal. The pH responsive polymer contains tertiary amine based monomers and a hydrophilic comonomer (e.g., PEG-acrylates). Tertiary amine based polymers have been used in nanomedicine to control drug release or fluorescent dye release based on pH.<sup>3,4</sup> At high pH, the tertiary amine based polymer is hydrophobic; at low pH, the tertiary amine protonates to form an ammonium and becomes hydrophilic. This hydrophobic/hydrophilic switch facilitates changes to the conformation of the nanoprobe, allowing for manipulation of the MRI signal.

In this presentation, the synthesis and characterisation of a pH responsive MRI nanoprobe is reported. Ultimately, a nanoprobe that has the ability to act as a MRI contrast agent, a PET tracer and a drug carrier will be synthesised for the application in theranostics (**Fig.1**).

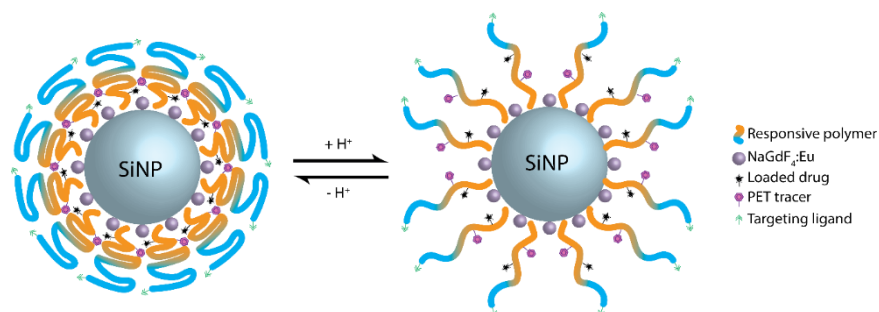


**Figure 1.** Representation of a silica nanoparticle based nanoprobe that has MRI, PET and drug delivery capabilities. Manipulation of the MRI signal can be achieved by using a pH switch to turn the MRI sign ‘on’.

J. Key, JF. Leary, *Int. J. Nanomedicine*. **2014**, 9, 711-725

<sup>2</sup> SC. Bushong, *Magnetic Resonance Imaging: Physical and Biological Principles*, **2003**

<sup>3</sup> JT. Sun, CY. Hong, CY. Pan, *J. Phys. Chem.* **2010**, 114, 12481-12486

<sup>4</sup> Y. Wang, J. Gao, et al *Nat. Mat.* **2014**, 13, 204-211

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# A Highly Efficient Photo-triggered Post-polymerisation Functionalization Method via Thioaldehyde Tapping.

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Postpolymerization functionalization is a versatile synthetic approach in obtaining functional materials. [1,2] Various methods for postpolymerization functionalization have been developed since Staudinger, who looked into the hydrogenation of rubber [3] and styrene. [4] More recently, a number of advanced postpolymerisation techniques have emerged, such as the highly efficient click chemistry, [5] thiol-ene and thiol-yne addition. [6,7] Of particular interest are activated ester polymers [8] and their thiol analogues. This presentation describes the synthesis and kinetics of photo-responsive functional thiolaldehyde-based copolymers by RAFT polymerisation. After activation by UV irradiation, they are capable of post-polymerisation functionalization via the fast trapping of a range of primary amines (**Fig 1**). The ability for the choice of amine allows for the selection of specific properties and structures for the polymers with broad application in synthetic chemistry and industry.

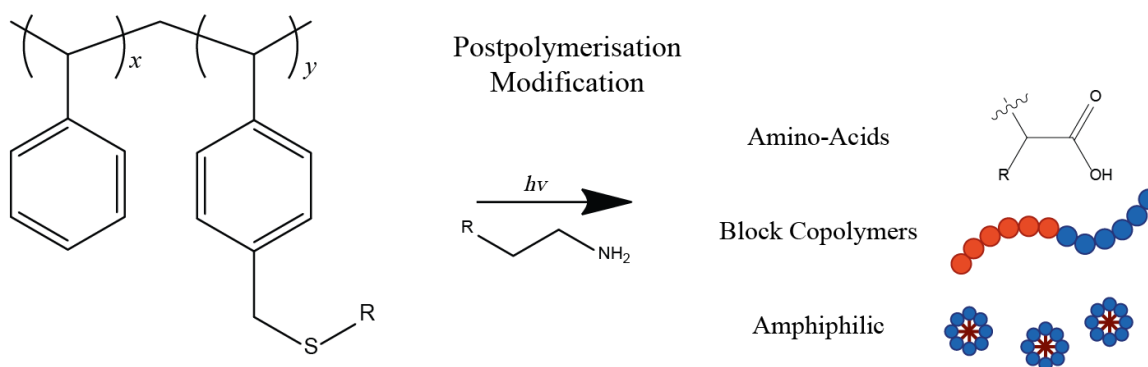


**Figure 1.** Postpolymerisation modification technique: after irradiation of the polymer, the reactive thiol group is able to trap a range of amines to create specific polymers with specific functionality and structure

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- [2] K. Günay, P. Teatro, H. Klok, *Journal of Polymer Science Part A: Polymer Chemistry*. **2013**, 51, 1-28.
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- [6] A. Lowe, C. Hoyle, C. Bowman, *Journal of Materials Chemistry*. **2010**, 23, 4345-4750.
- [7] N. Singha, M. Gibson, B. Koory *et al*, *Biomacromolecules*. **2011**, 12, 2908-2913.
- [8] L. He, K. Szameit, H. Zhao *et al*, *Biomacromolecules*. **2014**, 15, 3197-3205.

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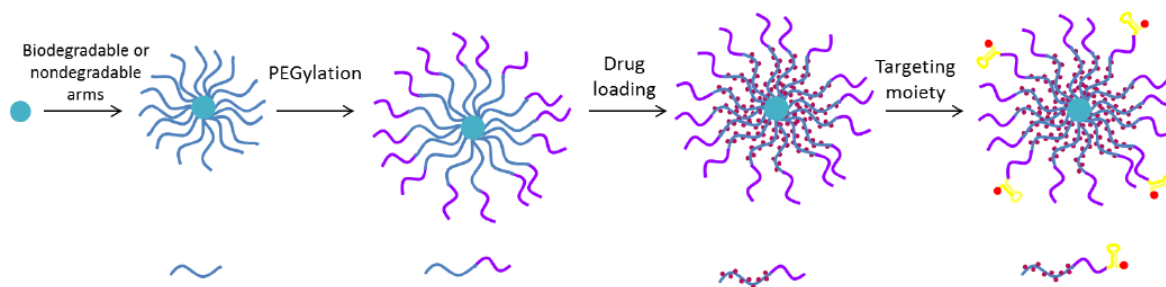
## Doxorubicin-loaded polymeric nanoparticles for cancer therapy

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Delivery of drug-loaded nanoparticles for cancer therapy is a popular concept due to the unique advantages of nano-sized particles. Tumour vasculature is often disorganised, leading to “leaky” blood vessels with gaps between endothelial cells <sup>i</sup>. Nanoparticles can extravasate into the tumour through these gaps, a process known as the enhanced permeation and retention (EPR) effect <sup>ii</sup>. Nanoparticles can accumulate in the tumour, reducing adverse effects when compared to traditional chemotherapy due to the lower systemic concentration of free chemotherapeutic <sup>iii</sup>. Nanocarriers can also improve the solubility of poorly water soluble chemotherapeutics <sup>iv</sup>. Drug is commonly conjugated to polymer nanoparticles via covalent stimuli-responsive (acid, enzyme, redox labile) linkers, enabling drug release inside the cell. For non-biodegradable polymers, such as poly(*N*-2-(hydroxypropyl methacrylamide), drug conjugation strategies such as these must be employed. The accumulation of these nonbiodegradable particles also poses potential long term toxicity issues <sup>v</sup>. Biodegradable polymers, such as poly (lactic-co-glycolic acid) and polypeptides, offer an alternate mechanism for drug release by utilising the degradation of the polymer backbone itself to effect drug release. The degradation of the polymer to excretable metabolites also prevents accumulation, thereby avoiding potential long term toxicity. To date, little has been done to compare the efficacy of biodegradable and nondegradable polymer nanocarriers with similar characteristics and their direct effect on drug release. In this work, nanoparticle internalization, degradation, drug release, and toxicity of biodegradable and nonbiodegradable nanocarriers of comparable size, charge, and drug loading were investigated *in vitro* using multiple cancer cell lines. Several architectures were studied, including star and linear polymers, as well as the incorporation of targeting ligands.



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<sup>iv</sup>A. Eatemadi, *Artif Cells Nanomed Biotechnol.* **2015**, 44, 1008-1017

<sup>v</sup>R. Duncan, *Nat Rev Drug Discov.* **2003**, 2, 347-360

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# Advanced biomaterials to promote endothelialization

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Cardiovascular disease remains a leading cause of death worldwide. The development of blood compatible biomaterials could relieve this burden by enabling the development of improved cardiovascular devices such as small diameter vascular grafts, stents, and artificial heart valves<sup>1</sup>. An attractive strategy for improving blood compatibility is to generate biomaterials that foster a confluent and functioning endothelial cell layer. Several strategies have been explored to improve endothelialization, but no blood compatible biomaterials have obtained commercial success as of yet. The lack of a blood compatible interface is one of the most pressing challenges in the biomaterials field<sup>2</sup>.

Key cellular functions including survival, migration, differentiation, and cell division are regulated by cell adhesion to the extracellular matrix (ECM)<sup>3</sup>. One class of cell-ECM interactions are triggered by binding of the extracellular domain of integrin receptors to peptide ligands present in surrounding biomolecules. However, integrin binding is insufficient to elicit a full adhesion response. In addition to an integrin being occupied by a ligand, the receptors must also be clustered to form adhesion complexes<sup>3</sup>. A recent breakthrough in the field is the synthesis and fabrication of materials that presents peptide ligands in nanoscale clusters, and these advanced biomaterials promote the clustering of integrins, formation of adhesion complexes, and improvement of various cellular functions such as adhesion, migration, adhesion strength, differentiation, and phenotype<sup>4</sup>. For instance, several researchers have taken advantage of the nanometer size of the individual polymers to achieve the nanoscale clustering of the RGD peptide ligand<sup>4</sup>.

In this study, we test the hypothesis that biomaterials that present nanoscale clusters of integrin-binding ligands can be used to improve endothelialization and thus blood compatibility of cardiovascular devices. In this work, a random copolymer of methyl methacrylate and poly(ethylene oxide) methacrylate was synthesized via raft polymerization with high molecular weight and low polydispersity (Fig.1a). This polymeric system can be readily functionalized via RGD peptides and blended with nonfunctionalized polymers to enable the nanoscale organization of RGD peptides to improve cellular functions of endothelial cells. As a model system, the adhesion and proliferation of NIH/3T3 model fibroblast cells were investigated to determine the potential of this biofunctionalized polymeric materials as a next generation biomaterial platform that can promote endothelialization (Fig.1b).

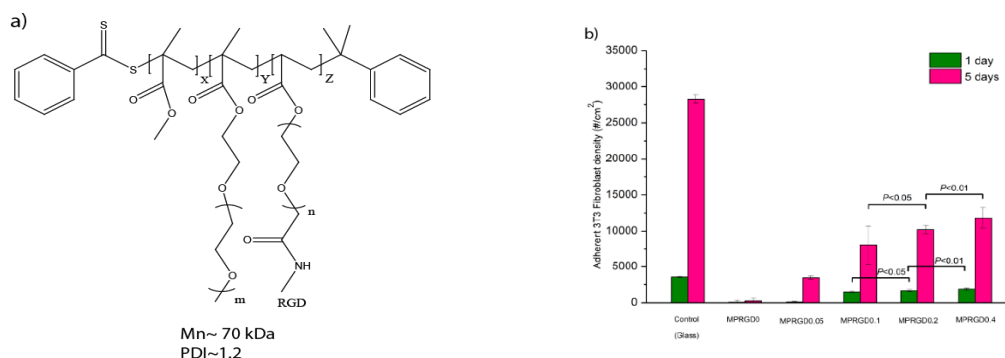


**Figure 1.** a) Synthesized polymer via raft polymerization b) Adhesion and growth of NIH/3T3 fibroblast cells on the surface of polymers containing various RGD percentages from 0-0.4 mol%

<sup>1</sup> B. Ratner, *Biomaterials*. **2007**, 28, 5144

<sup>2</sup> V. Catto, *ISRN Vascular Medicin*. **2014**, 2014

<sup>3</sup> J. Ivaska, *Nature cell biology*. **2012**, 14, 981

<sup>4</sup> B. Ekerdt, *Biotechnology. J.* **2013**, 8, 1411

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## **BM1565: a very versatile RAFT agent**

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The Reversible Addition-Fragmentation chain Transfer (RAFT) polymerisation process<sup>1</sup> is one of the most versatile methods of Controlled/living free Radical Polymerisation. The process is mediated by thiocarbonylthio compounds, such as thiocarbamates, dithioesters and xanthates, which act as Chain Transfer Agents (RAFT agents). A broad range of monomer systems can be used, including styrenes, acrylonitriles, (meth)acrylates, (meth)acrylamides, dienes, and vinyl monomers.<sup>2</sup>

RAFT agents are selected for polymerization depending on the reactivity of monomer that is to be polymerised; monomers can be classified into more-activated monomers (MAM) and less-activated monomers (LAM) toward RAFT polymerisation. There are not many examples in the literature where a RAFT agent is used to co-polymerise a mixture of LAM/MAM; also, the preparation of block copolymers by RAFT polymerisation that are derived from LAM and MAM has proven to be tedious with the use of switchable RAFT agents.<sup>3</sup>

Our group synthesised a novel class of dithiocarbamate RAFT agents which are capable of producing polymers with predetermined molecular weight and narrow molecular weight distribution, or low dispersity ( $\bar{D}$ ), using both types of monomers, LAM and MAM, without the need to be structurally modified or “switched” in any way.<sup>4</sup> The latest RAFT agent made commercially available: BM1565 (2-cyanobutan-2-yl 4-chloro-3,5-dimethyl-1Hpyrazole-1-carbodithioate) has proven to be the most versatile. Firstly, using BM1565, RAFT homopolymers containing MAM monomers with  $\bar{D} \sim 1.1$  were obtained; using the same RAFT agent, homopolymers containing LAM monomers were obtained with  $\bar{D} \sim 1.2$ . Secondly, random copolymers containing mixtures of MAM monomers and MAM/LAM with  $\bar{D} \sim 1.2$  were synthesised. Finally, RAFT block copolymers have been obtained with MAM-MAM and MAM-LAM with  $\bar{D} \sim 1.2$ . In addition, the thermally initiated RAFT polymerisation of Styrene with BM1565 (i.e. without solvent or initiator) produced polystyrene with  $\bar{D} < 1.1$ . Random copolymers with a mixture of MAM monomers with  $\bar{D} \sim 1.1$  have also been obtained with the thermal initiation of a mixture of the monomers and BM1565.

BM1565 has been developed in our group in collaboration with Boron Molecular Pty Ltd and is now available in commercial quantities, thus allowing the large scale production of RAFT copolymers or block copolymers with a new range of properties and applications.

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- <sup>2</sup> G. Moad, E. Rizzardo, S. H. Thang, *Acc. Chem. Res.* **2008**, *41*, 1133.
- <sup>3</sup> D. Keddie, C. Guerrero-Sanchez, G. Moad, E. Rizzardo, S. H. Thang, *Macromolecules*, **2011**, *44*, 6738.
- <sup>4</sup> J. Gardiner, I. Martinez-Botella, J. Tsanaktsidis, G. Moad, *Polym. Chem.*, **2016**, *7*, 481-492.

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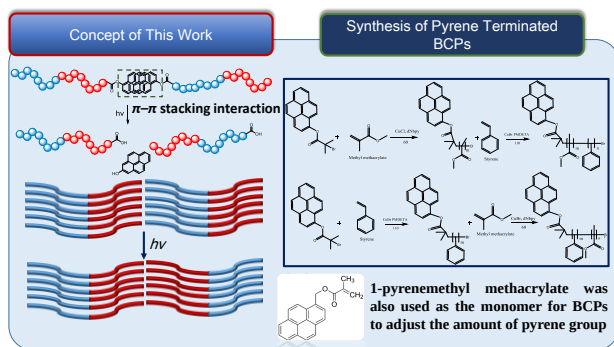
# Tuning Nano-structures in Photo-responsive Block Copolymers with $\pi$ - $\pi$ Stacking Interaction

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Materials with nano-structure embedded have attracted extensively interest as their potential applications in energy conversion and storage devices, photon detection, drug delivery and tissue engineering, and electrochemistry etc. Numerous techniques have been utilized for fabricating the nano-structured materials. Block copolymers (BCP) is a hot spot of research for generating a rich diversity of nano-morphologies by controlling the chemical composition and sizes of building blocks.<sup>1</sup> In addition, photo-responsive block copolymers, one of the most popular candidates for controlling nano-structures to give precise spatial control without disrupting structures, has been well studied. However, using these polymers for morphological transitions in the solid state or controlling structural hierarchies with other inter/intra molecular interactions remains limited. In this work,  $\pi$ - $\pi$  stacking interactions (using pyrene groups) and polymer-polymer miscibility was combined to control the morphologies of BCPs.<sup>2,3</sup>



Living radical polymerization (LRP) is one of the most powerful techniques that enables us to synthesize photo-responsive polymers due to its inherent robustness and versatility. Pyrene terminated BCPs were synthesized via atom transfer radical polymerization (ATRP) using an ATRP-initiator containing a pyrene substituent.<sup>4</sup> (**Scheme 1**) The procedure termed “halogen exchange” provides a tool that allows methyl methacrylate monomer to alter the order of addition of monomers (methyl methacrylate and styrene) to yield a sequential block

**Scheme 1.** Concept of this work and the synthesis procedures of pyrene terminated block copolymers of PMMA-b-PS and PS-b-PMMA.

copolymerization from that dictated by monomer reactivity thereby allowing increased freedom in designing polymer architecture.<sup>5</sup> This series of BCPs were prepared and which allowed the morphology to be tuned through photo-responsiveness. The characterisation of the materials in bulk and in thin films through techniques such as differential scanning calorimetry, small angle x-ray scattering, ellipsometry, scanning electron microscopy was investigated.

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# Multi-responsive injectable bioactive delivery system based upon Pluronic

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Stimuli-sensitive polymers, especially thermosensitive polymers have attracted increasing interest over the two past decades, particularly in the fields of drug delivery and tissue engineering.<sup>1</sup> [ENREF 1](#) Due to their inherent thermal-reversible nature, Pluronics (PEO-PPO-PEO triblock copolymers) have a great potential to serve as injectable drug reservoirs. Typically at room temperature Pluronics form a solution in water and can be used to inject drugs into the desired tissue site by a syringe, and then quickly form into hydrogel *in situ* at body temperature encapsulating the drug.<sup>2</sup> However, the resulting self-assembled hydrogels are typically unstable and easily revert into a liquid state when diluted in body fluids.<sup>3</sup> [ENREF 3](#) [ENREF 3](#) [ENREF 3](#) [ENREF 3](#) Here we report the synthesis and characterisation of covalently stabilised Pluronics hydrogels which have potential to enable slow release of therapeutic agents.

Photo-initiated crosslink between self-assembled Pluronics hydrogels was investigated via the synthesis of diacrylate-terminated Pluronics, followed by their thermos-responsive gelation study to *in situ* encapsulate fluorescent protein as a therapeutic model. After injection, the solution quickly change into hydrogel, and then was exposed to UV light for up to 2 minutes to generate a chemically crosslinked, more stable hydrogel (**Fig. 1**).

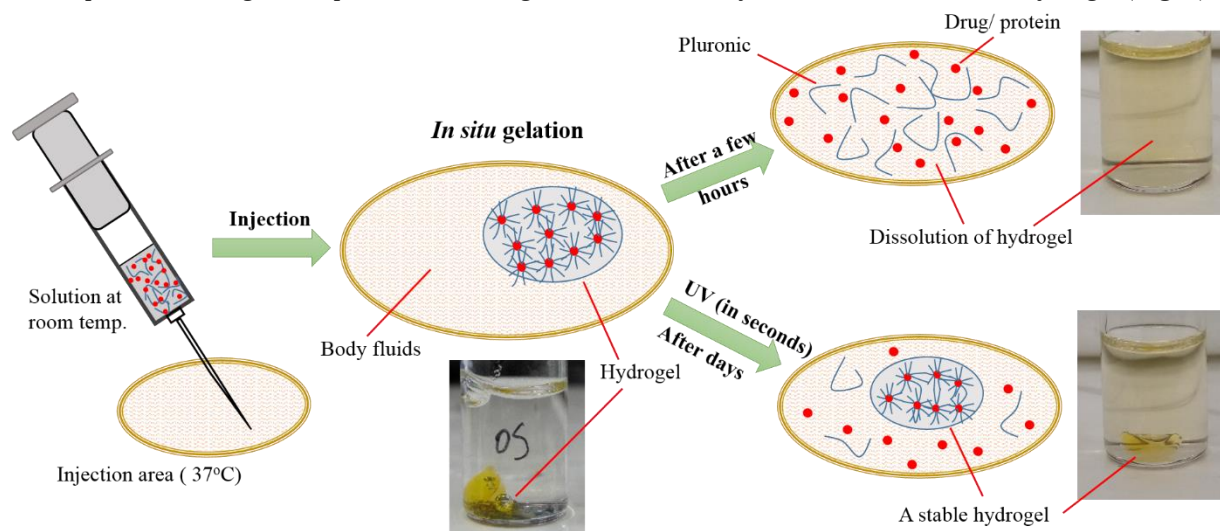


**Figure 1.** Scheme of photo-crosslinkable and thermo-sensitive Pluronics for injectable drugs delivery, and the release pictures of FITC-BSA from Pluronic hydrogel into buffer with and without UV crosslinking.

Covalently cross-linked hydrogels exhibits higher stability over simple physically assembled ones upon dilution. Structure-property relationships of the hydrogels were investigated using Pluronics of different hydrophilicity (PEO/PPO block length) and concentration, *e.g.* F-127, F-108, F-87, P-105, and P123. The correlation between hydrogel physical properties and drug release kinetics were investigated using rheology, SAXS, DLS analysis, and UV spectroscopy. Pluronic-based hydrogels are promising drug delivery systems and have potential to expand the biomedical applications of these FDA-approved polymers and provide a versatility to deliver different types of therapeutics.

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<sup>2</sup> H. Lee, T. G. Park, *Journal of Biomedical Materials Research Part A* **2009**, 88 (3), 797-806.

<sup>3</sup> M. Di Biase, P. de Leonardis, V. Castelletto, I. W. Hamley, et al., *Soft Matter* **2011**, 7 (10), 4928-4937.

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# Synthesis of Miktoarm Star Polymers By Raft Polymerisation For Drug Delivery Across Biological Barriers

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Brain cancers such as glioblastoma are some of the most dangerous cancers, with many treatment options but an alarmingly high mortality rate. It is reported to have lowest survival rates of all cancers at less than 20% over 5 years<sup>1</sup>. The main reason for this is the ineffective delivery of the anticancer drug to the tumour location, which is due to the presence of certain barriers in the brain (for example, the blood brain barrier, BBB). These barriers not only protect the brain from harmful material, but also restrict the entry of useful therapeutic molecules<sup>2</sup>. Thus, there is a need for these barriers to be targeted by non-invasive delivery agents which can pass through without damaging their integrity, thus delivering the drug molecules to the site of action.

The present project deals with one such non-invasive approach, involving miktoarm star polymer synthesis using radical addition fragmentation chain transfer (RAFT) polymerisation process. RAFT is one of the most versatile living radical polymerisation techniques for synthesising complex molecular architectures in a controlled fashion<sup>3</sup>. The project involves the synthesis of a range of polymers with desirable properties such as hydrophobicity/hydrophilicity, 'stealth' properties and ability to conjugate targeting ligands and drug molecules. These are cross-linked to form miktoarm star polymers which are optimised for transport across BBB and delivering drug payload at the site of cancer tissue.

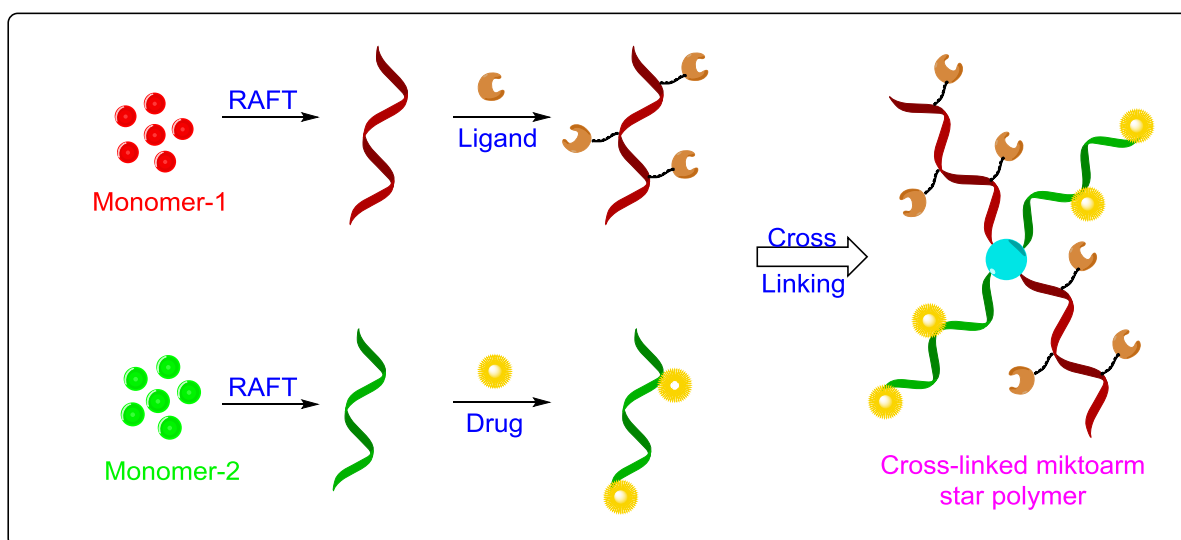


**Figure 1.** Schematic representation of preparation of RAFT polymers for drug delivery

<sup>1</sup>Siegel, R. L.; Miller, K. D.; Jemal, A., *CA: a cancer journal for clinicians* **2015**, 65 (1), 5-29.

<sup>2</sup>Pardridge, W. M., *J Cereb Blood Flow Metab* **2012**, 32 (11), 1959-72

<sup>3</sup>Gregory, A.; Stenzel, M. H., *Progress in Polymer Science* **2012**, 37 (1), 38-105.

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# Engineering Functional Biological Properties into Polymeric Materials

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Advanced Materials have the potential to combat the major challenges of the 21<sup>st</sup> Century, with substantial progress being made in areas ranging from medicine right through to industry. Research into the use of polymeric materials as vectors for therapeutics has increased within the last decade, providing a platform for materials scientists to engineer the next generation of therapeutic carriers. Two vital aspects of nanoparticle-based therapeutic delivery include active targeting and controlled cargo release. Directing the nanoparticle to specific cells is regularly achieved via a targeting moiety (biomaterial such as antibody) whereas sub-cellular targeting and cargo release are often attained by tuning chemical properties into the synthetic polymer. Conjugating biomaterials to synthetic polymers has numerous advantages, as the useful properties from both material types can be harnessed in the one system. Hybrid materials can possess functional properties such as cellular specificity from the biomaterial component, whilst maintaining cargo release to specific cellular compartments from the synthetic component.

Herein, we report of a hybrid carrier based on Poly[(2-(diethylamino) ethyl methacrylate)-co-(2-(diisopropylamino) ethyl methacrylate)] and Poly[(2-(diethylamino) ethyl methacrylate)-b-poly(ethylene glycol)], which were conjugated to model targeting proteins. Nanobodies were used as they are smaller than conventional antibodies, consisting of only a single protein domain. Bioconjugate chains were then assembled into nanoparticles through the nanoprecipitation method (Figure 1). Selective incorporation of ionisable monomeric functional groups correspondingly permitted the particles to become responsive to pH. The polymeric carrier was shown to be responsive in the biologically relevant pH range, as seen through the induction of swelling upon acidification. pH induced nanoparticle disassembly in the cell's endosomal compartments (pH 5-7.2) would promote effective endosomal escape, a necessary trait of any carrier intending to deliver its therapeutic payload to the cytosol. *In vitro* experiments revealed promising results, highlighting that the proposed system could enter specific cells and escape from the endosomal compartments.

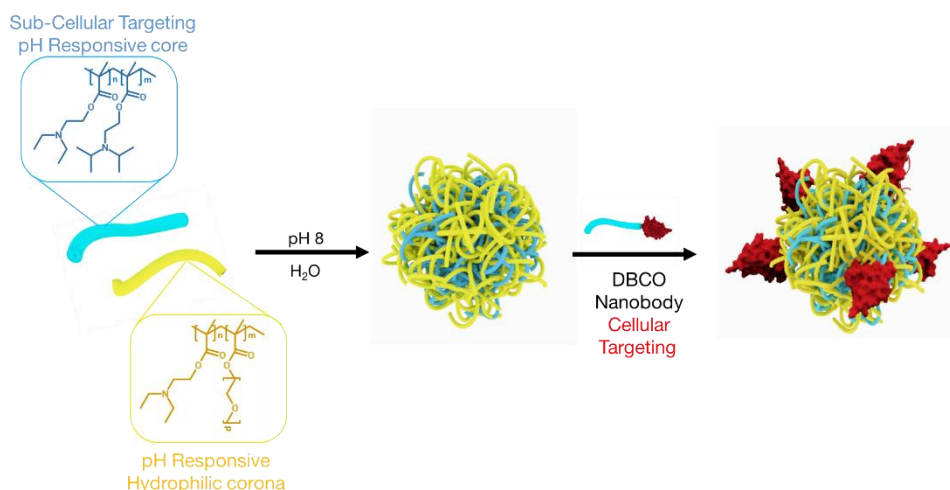


**Figure 1.** Biologically functional polymers assemble into therapeutic nano-carriers.

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## Evaluation of two epoxy systems under accelerated degradation

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Epoxy resins are structural adhesives used in a wide range of industrial and civil engineering applications such as long-term protective coating systems, electrical engineering and industrial pipes for outdoor conditions. In outdoor conditions, epoxy resins are exposed to various factors such as ultra-violet irradiation, moisture, temperature extremes, etc. which are mainly responsible for the degradation. Photodegradation of epoxy is one of the principal factors that lead to a loss of the structural and mechanical properties <sup>1, 2, 3</sup>.

This aim of this work to evaluate two epoxy systems, using diglycidyl ether of bisphenol A (DGEBA), and hydrogenated diglycidyl ether of bisphenol (HDGEBA) cured with an aliphatic amine trimethyl hexa di- amine (TMDA) to artificial accelerated weathering (AW) as a baseline for development of epoxy composite formulations more suitable for outdoor timber applications. The cast DGEBA and HDGEBA epoxy samples were exposed to cyclic ultraviolet irradiation, using UVA chamber that contains ultraviolet fluorescent lamps (UVA-340nm) for different times (0, 168, 336, 504, 720, 1008 h) and then followed exposure with the same times to moisture condition in moisture chamber (50°C±5). Unweathered and weathered samples were evaluated using FTIR spectroscopy. Mechanical tests, DSC, TGA, and SEM. Typical tensile test profiles (Figure 1) and a summary of the stress at break results for HDGEBA and DGEBA-based epoxies (Figure 2) are shown below. In order to relate the structural properties to processes on the molecular scale, we are extracting the weathered epoxy samples and analysing degradation products as a function of weathering time by NMR and GC-MS.

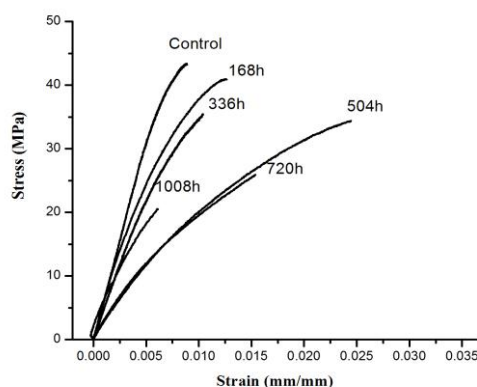


Figure 1. Tensile strength of DGEBA/TMDA epoxy for different accelerated weathering times.

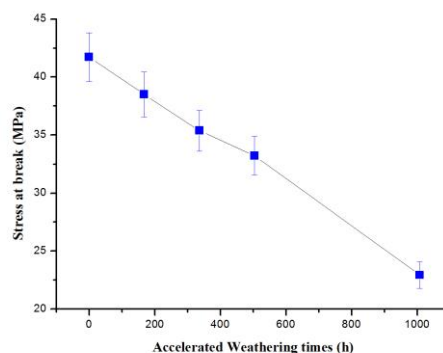


Figure 2. Stress at break of DGEBA/TMDA epoxy for different accelerated weathering times.

### References

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- <sup>2</sup> F. Liu, M. Yin, B. Xiong, F. Zheng, W. Mao, Z. Chen, C. He, X. Zhao, P. Fang, *JJAP Conf. Proc. Japan*, **2014**.
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# The Effects of Molecular Weight on the Performance of Electrochromic Polymers

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Electrochromic conjugated polymers belong to an exciting class of materials as they can reveal optical and color changes following a redox process under an electrical stimulus. Their color-availability and tunability, coupled with large color contrasts, fast switching speeds and low power consumptions render them suitable in a huge variety of applications such as smart windows, electronic displays, ophthalmic eyewear and camouflage cloths.<sup>1</sup> Recently, our group reported numerous diketopyrrolopyrrole (DPP)-based electrochromic polymers, focusing on color-tuning<sup>2</sup> and structure-property relationship studies (e.g. effect of chain lengths and donor-acceptor ratio).<sup>3,4</sup> In this work, we direct our attention towards the study of the effects of molecular weight on the optical, colorimetric, electrochemical, morphological and electrochromic properties on a DPP-based donor-acceptor type polymer (**Fig. 1a**). The polymer with a higher molecular weight (DPP<sub>H</sub>) reveals a stronger optical absorptivity and more reversible redox activity than the lower molecular weight counterpart (DPP<sub>L</sub>). While both polymers exhibit green-to-grey electrochromic switching (**Fig. 1b**), performance parameters in terms of optical contrasts, switching speeds, coloration efficiencies and stabilities are enhanced in DPP<sub>H</sub>. All the above results demonstrate the important role of molecular weight in determining the performance of polymeric electrochromic devices.

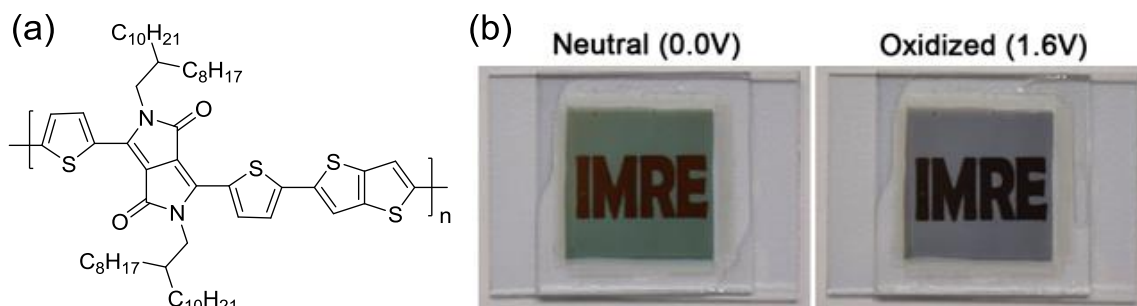


**Figure 1.** (a) Chemical structures DPP-based polymers studied in this work and (b) green-to-grey electrochromic switching of fabricated devices.

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<sup>2</sup> W. T. Neo, C. M. Cho, Z. Shi, S. -J. Chua, J. Xu, *J. Mater. Chem. C.* **2016**, *4*, 28-32.

<sup>3</sup> J. Z. Low, W. T. Neo, Q. Ye, W. J. Ong, I. H. K. Wong, T. T. Lin, J. Xu, *J. Polym. Sci. Part A: Polym. Chem.* **2015**, *53*, 1287-1295.

<sup>4</sup> W. T. Neo, Z. Shi, C. M. Cho, S. -J. Chua, J. Xu, *ChemPlusChem*, **2015**, *80*, 1298-1305.

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# Dielectric probing of Novel Thermoplastic Ionic Elastomer based on Natural Rubber:- An alternative to Vulcanized Rubber

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Incorporation of small amounts of ionic groups in hydrocarbon polymers exerts a profound effect on their mechanical properties<sup>1-3</sup>. These ionic groups neutralized with suitable metal ions act as physical cross-links within the polymer matrix. The ionic associations can be thermally relaxed to permit sufficient melt flow at the processing temperature. The ion-containing polymer thus behaves as a thermoplastic elastomer having the unique ability to behave as cross-linked elastomer at ambient temperatures, and to melt and flow at elevated temperatures like thermoplastics<sup>4</sup>.

Chemical modification of rubber is an endeavor to improve its mechanical properties. Grafting and vulcanization are some of the viable techniques for modifying the properties of natural rubber. The incorporation of reactive aromatic molecules like styrene into natural rubber backbone, through grafting produces natural rubber with improved mechanical properties. The grafting of monomers such as styrene to the natural rubber backbone has been reported<sup>5</sup>. The subsequent modification of grafted natural rubber into ionomers would yield materials with improved mechanical properties compared to plain rubber. To date, many types of ionomers have been developed, and used for special applications. Among them ionomers based on EPDM, nitrile rubber, polychloroprene rubber, and ethylene methacrylic acid copolymers are very important<sup>1, 6</sup>.

This paper reports the pioneering efforts to: (a) introduce ionic groups into styrene grafted natural rubber to produce zinc sulfonated styrene grafted natural rubber (ZnS-SG NR) of different ionic concentrations, (b) characterize the zinc sulfonated styrene grafted natural rubber using spectral and thermal methods, (c) evaluate the effects of ionic groups on the mechanical properties of the zinc sulfonated styrene grafted natural rubber (d) study the reprocessability and (e) probe the dielectric behavior of the of the newly synthesized ionic polymer.

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## Self-assembly of amphiphilic diblock co-oligomers in aqueous solution

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Amphiphilic diblock co-oligomers are short block copolymers that maintain some of the desirable features of conventional surfactants such as water solubility and short equilibration times. The aqueous solubility of amphiphilic diblock co-oligomers is significantly better than long block copolymers, while their surface activities are similar to conventional surfactants. Amphiphilic diblock co-oligomers composed of a common acrylic acid hydrophilic block (AA) (5 to 10 units of acrylic acid) and a hydrophobic block (n-butyl acrylate, tert-butyl acrylate or ethyl acrylate) were synthesized by RAFT polymerization.<sup>1</sup> Self-assembly of BA<sub>5</sub>AA<sub>5</sub>, tBA<sub>x</sub>AA<sub>5</sub> (x = 5, 7 or 10) and EA<sub>y</sub>AA<sub>5</sub> (y = 5, 10 or 20) co-oligomers into micelles and liquid crystals, i.e. hexagonal phase (H<sub>1</sub>), cubic (I<sub>1</sub>) and lamellar phase (L<sub>α</sub>), was studied using polarizing optical microscopy, small angle x-ray scattering (SAXS), pyrene fluorescence and small angle neutron scattering (SANS). Self-assembled nanostructures and lyotropic phase stability of these systems, in both concentrated and dilute aqueous solutions, are highly responsive to the degree of ionization of acrylic acid block, the length and structure of the hydrophobic block. By controlling these parameters, the self-assembly of the co-oligomers can be varied and even be switched from insoluble to soluble in water. As compared to a previous system<sup>2</sup>, hexagonal nanostructure was observed in BA<sub>5</sub>AA<sub>5</sub> system. The self-assembly of these co-oligomers has promise for potential applications in industry, e.g. surfactants, detergents and drug delivery agents.

G. Moad, E. Rizzaroda, S.H. Thang, *Aust. J. Chem.* **2009**, 62, 1402-1472

<sup>2</sup> J. Heinen, A. Blom, B. Hawzett, G. Warr, *J. Phys. Chem. B* **2013**, 117, 3005-3018

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## Injectable cyclodextrin-based hydrogel as hydrophobic drug delivery platform

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Conventional drug delivery methods such as injection and oral delivery often experience difficulty in succeeding steady-state drug concentrations and pharmacokinetic profiles<sup>1</sup>. Thus, alternative methods of administration have been studied to control the rate of drug release, prevent undesirable side effects and increase drug bioavailability<sup>2</sup>. Hydrogels are the most investigated drug delivery platforms as specific design principles can be applied to these networks in order to fit the complex nature of biological systems and reach long term release through safe mechanisms<sup>3</sup>. These three-dimensional networks of cross-linked polymers closely mimic living tissues and due to their high water content often exhibit high biocompatibility<sup>4</sup>. However, current delivery systems based on hydrogels encounter difficulties with the loading of lipophilic drugs inside an aqueous gel network. Due to increasing the internal hydrophobicity, the hydrogel network might collapse and the hydrophobic molecules just diffused out meaning no sustained or controlled release. One way to address this issue is using cyclodextrins (CDs) as solubilizing agents. CDs with hydrophobic interior can form inclusion complexes with hydrophobic guest molecules. Thus, Incorporation of CD moieties into hydrogel networks can facilitate the entrapment and controlled release of lipophilic molecules<sup>5</sup>.

Herein, a novel injectable hydrogel has been developed for delivery of a hydrophobic drug, nifedipine (NIC). CD-functionalized polyhydrazine with PEG pendant (PG) was synthesized as the first polymer involved in hydrogel formation. Oxidized dextran (DA) was the second polymer used in formation of the matrix. Two sets of *in-situ* formed hydrogels with different cross-linking densities were prepared by mixing aqueous solutions of DA and PG. Gelation occurred rapidly *in-situ* via hydrazone linkages. These injectable networks successfully encapsulated NIC via their CD moieties and release profile of NIC from the gels exhibited a long term controllable release. Fig 1 illustrates hydrogel formation in this study.

Synthetic and characterization methods of the polymers and the hydrogels along with the release mechanism will be discussed.

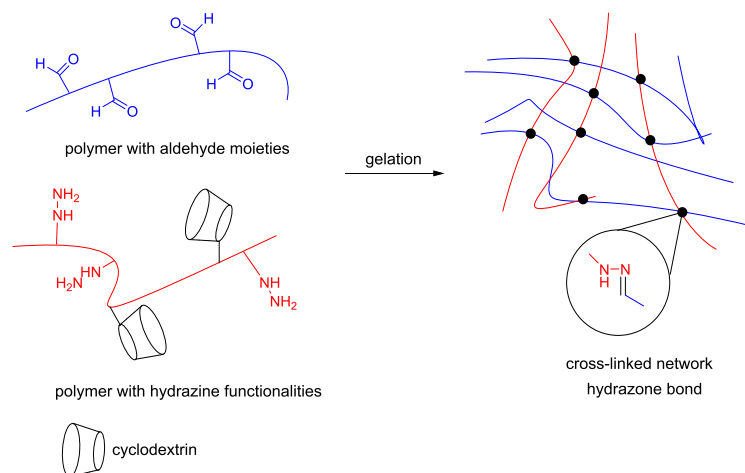


Fig 3: Hydrogel formation via hydrozone bonds.

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# Biologically-inspired polymer synthesis via biocatalyst-mediated radical polymerization

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In the last few decades, reversible deactivation radical polymerization (RDRP) techniques have paved the way for great progress in the field of polymer synthesis by providing the opportunity for synthesizing polymers of well-defined molecular weight, composition, and structure. With the continued development of RDRP methodologies, such as atom transfer radical polymerization (ATRP), nitroxide-mediated polymerization (NMP), and reversible addition-fragmentation chain transfer (RAFT), there has recently been a great deal of research effort to control these reactions via different physical and chemical stimuli.<sup>i</sup>

Many studies have employed transition metal catalysts - generally complexes of iron or copper - to activate ATRP and RAFT polymerizations of vinyl monomers. Although these transition metal-based catalysts have shown good performance in various polymerization systems, they bring inherent concerns of toxicity, especially for the production of polymers intended for use in biomedical applications. Additionally, these complexes may increase the cost of production since the techniques required to purify the polymers are usually quite expensive.<sup>ii</sup>

Recently, the principles of “green” chemistry have attracted the attention of scientists, including those in the field of polymer synthesis.<sup>iii</sup> As a subset of this field, the use of biocatalysts (i.e., catalysts derived from biological sources) for synthesizing well-defined polymers remains an underexplored area of polymer science. In the present work, the ability of various biocatalysts to mediate RDRP reactions is investigated. The mechanism of biocatalyst activation towards the polymerization is assessed for the synthesis of linear polymers (**Fig. 1**), with a view towards the potential for these materials to be utilized in biologically-relevant applications.

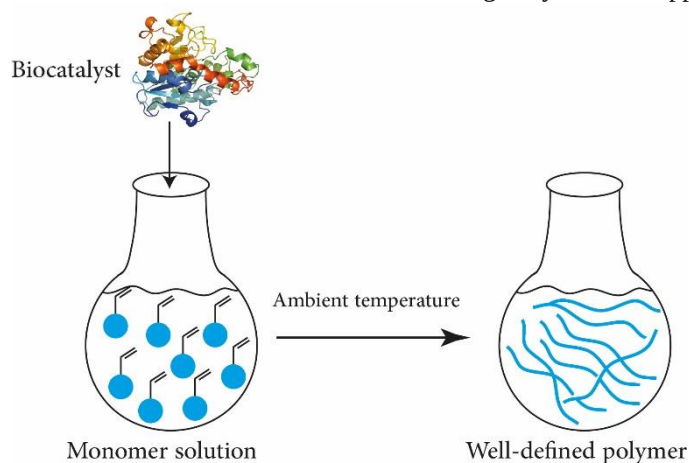


**Figure 1.** Basic schematic of biocatalyst-mediated polymer synthesis.

<sup>i</sup> F. A. Leibfarth, K. M. Mattson, B. P. Fors, H. A. Collins, C. J. Wawker, *Angew. Chem. Int. Ed.* **2013**, 52, 199-210.

<sup>ii</sup> P. Maximiano, P. V. Mendonça, J. O. R. Costa, N. L. Haworth, A. N. C. Serra, T. Guliashvili, M. L. Coote, J. F. Coelho, *Macromolecules* **2016**, 49, 1597-1604.

<sup>iii</sup> K. Schroder, K. Matyjaszewski, K. J. Noonan, R. T. Mathers, *Green Chem.* 2014, 16, 1673-1686.

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## Synthesis of well-defined polystyrene diblock copolymers with interchangeable surface end groups and core-confined Cyanine 5 by RAFT-mediated emulsion polymerisation

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While en route to their respective target/s, nanoparticles encounter a complex milieu of blood-bound materials. Consequently, nanoparticles associate with different blood components, thus influencing their biological activities. There is a rapidly increasing body of evidence that nanoparticle-blood component association is significantly influenced by nanoparticle physiochemical properties (size, surface charge, morphology etc.). In order to further elucidate nanoparticle-blood interactions, we developed fluorescent polystyrene nanoparticles with interchangeable surface end groups and tuneable size. Well-defined polystyrene diblock copolymers were synthesised by using a RAFT-mediated emulsion polymerisation technique with a novel water-soluble, biocompatible macromolecular chain transfer agent (macro-CTA)<sup>1</sup>. This new technique provides access to polystyrene nanoparticles with narrow distribution of molecular weights and particle size, tuneable particle size, and excellent stability. To track nanoparticle interactions *in vitro*, highly fluorescent cyanine 5 (Cy5) was incorporated into the styrenic core by RAFT polymerisation of a large excess of styrene and a discrete molar quantity of Cy5 maleimide. To explore the effect of surface terminal end group on blood component association, the chain transfer agent (CTA) Z group was modified. The CTA, 4-cyano-4-(ethylthiocarbonothioylthio)pentanoic acid (ECT), was modified by esterification of the carboxylate end group using dicyclohexylcarbodiimide (DCC) in combination with various alcohols to yield CTA with different Z groups. Subsequent RAFT-mediated emulsion polymerisation yielded well-defined Cy5 labelled polystyrene copolymers with different surface end groups.

<sup>1</sup>Truong, N.P, Dussert, M.V, Whittaker, M.R, Quinn, J.F, Davis, T.P, Rapid synthesis of ultrahigh molecular weight and low polydispersity polystyrene diblock copolymers by RAFT-mediated emulsion polymerization. *Polymer Chemistry* **2015**, 6, 3865 - 3874

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# Functional Substituents and Heteroatoms Effects on the Redox Behavior of Nitroxide Radicals

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Nitroxide radicals have been found a variety of applications such as in organic catalytic reactions, ‘living’ radical polymerization, biomedical probe imaging, electrochemical energy storage etc.<sup>1-4</sup>

Comparing to other application areas, nitroxide radicals as electrochemical energy storage materials only emerged in last decade. This application relies on the reversible redox behaviour of nitroxide radicals. In fact, the redox behaviour of nitroxide radicals is determined by their chemical structures, mainly their substitution groups and hetero atoms adjacent to the nitroxide radicals. Despite the computational simulation by molecular orbital theory and density functional theory (DFT) revealing the distinct effect,<sup>5</sup> very few examples have shown the functional substituents and heteroatoms effects on the redox behaviour of nitroxide radicals.<sup>6</sup>

Herein, we have synthesized twenty different nitroxide structures with different substitutions and heteroatoms on the nitroxide radicals. Systematic studies on their electron spin resonance and electrochemical redox reaction were carried out and the results demonstrated the inherent correlation between the redox potentials and nitroxide radical structures (Figure 1). These results were significant for further design the nitroxide radical polymers as electrode materials for virtual plastic battery.

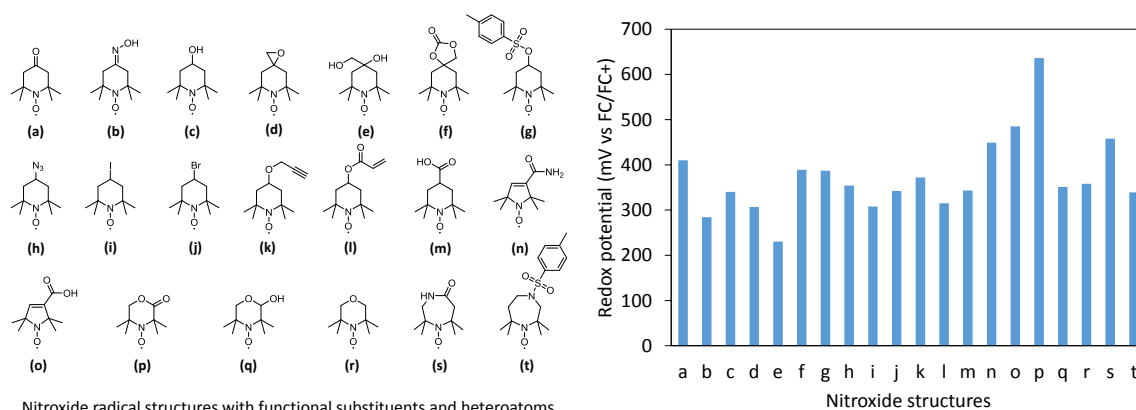


**Figure 1.** Nitroxide radical structures (a-t) and their redox potentials

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## Polymeric Ciprofloxacin prodrug carriers for the treatment of respiratory Melioidosis

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*Burkholderia pseudomallei* is the causative agent of Melioidosis, a bacterial infection with a mortality rate of 40-50% when untreated and a recurrence rate of 10-20% even with antibiotic treatment<sup>1</sup>. Much of the lethality of this disease is due to the ability of *B. pseudomallei* to reside within the alveolar macrophage following inhalation, where they evade the immune system and become difficult to combat with traditional antibiotic therapies. Furthermore, this bacterium is aerosolizable and easily weaponized for biowarfare; as such the DHHS, DCD, and USDA consider *B. pseudomallei* a tier 1 agent<sup>3</sup>. Current care is limited, requiring weeks of intravenous or oral antibiotic delivery. This reliance on systemic delivery is due to multiple challenges to the delivery of free antibiotics and leaves much room for improvement.

In order address these challenges and improve the treatment of pulmonary Melioidosis, various polymeric prodrug carriers were developed that improve drug solubility and stability, that are aerosolizable for direct delivery to the lungs to improve drug bioavailability and eliminate systemic off-target effects, and that use various chemistries and architectures to provide multiple controlled and sustained drug release profiles. Ciprofloxacin (Cipro) was modified to include a methacrylate functional group and hydrolysable linker to allow for direct polymerization using RAFT and to control drug release kinetics. The Cipro monomer (CTM) was copolymerized with carboxybetaine (CBM) in various formulations to maximize drug loading by improving drug solubility and biocompatibility. These formulations were shown to have highly tunable drug release profiles (Fig. 1), be highly efficacious within an in vitro intracellular infection model (Fig. 2) while remaining nontoxic to mammalian cells, and show excellent in vivo biocompatibility (in vivo activity data not yet available). In addition to these novel prodrug polymeric carriers, an antimicrobial peptide monomer is also under development, for eventual incorporation into the carboxybetaine-Cipro systems to boost activity of the codelivered Cipro and to address the growing problem of drug resistant *B. pseudomallei* strains<sup>3</sup>.

<sup>1</sup> A.C. Cheng, B.J. Currie. *Clin. Microbiol. Rev.* **2005**, 18, 383-416.

<sup>2</sup> DHHS, CDC, USDA. **2014**. <http://www.selectagents.gov/SelectAgentsandToxinsList.html>

<sup>3</sup> H.P. Schweizer. *Future Microbiol.* **2012**. 12, 1389-1399.

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# Preserving the Activity of Immobilised Proteins by Polymer Brushes

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Surface modification techniques have played an important role in biological and medical fields for different purposes, such as preventing fouling by cells and proteins<sup>1</sup>. This project is aimed at the immobilisation of biomolecules on silicon wafers modified by polymers that are grafted by 'grafting to' technique. Specific polymer is chosen in this work, which is poly(ethylene glycol) (PEG) (**Fig.1**). Polymer brushes systems can act as stimuli responsive materials, such as in friction switching surfaces<sup>2</sup> or surfaces that change wettability<sup>3</sup>. Advanced analytical techniques have been exploited to characterise the modified surfaces, including X-ray photoelectron spectroscopy (XPS), spectroscopic ellipsometry (SE), atomic force microscopy (AFM) and water contact angle measurements. The nanoroughness of substrate changes the structure and functionality of adsorbed proteins, and the nanotopography can alter biological responses, although no studies have been performed that change, the local chemistry of nanofeatures using polymer layers to preserve biomolecule activity. The PEG graft density was varied by increasing the salt content ( $K_2SO_4$ ) and the temperature of the solution. XPS showed that under these specific conditions, 1%, 2% and 4% of (3-Amino propyl)triethoxysilane (APTS) in the presence of 0.6M  $K_2SO_4$  at 60°C produce grafted PEG thicker than other conditions, and the optimal density of PEG was found at 2% APTS in the presence of cloud point condition (0.6M  $K_2SO_4$  at 60°C) (**Fig.2**). The water contact angle measurements confirmed the hydrophilicity of modified surface. Moreover, all these grafted layers confirmed by SE and AFM analysis. Mixed polymeric, short and long chain molecular layers will be synthesised, the goal is to trap proteins in a layer of low density polymer chains and utilise under layer for covalent attachment of the protein thus improving stability.

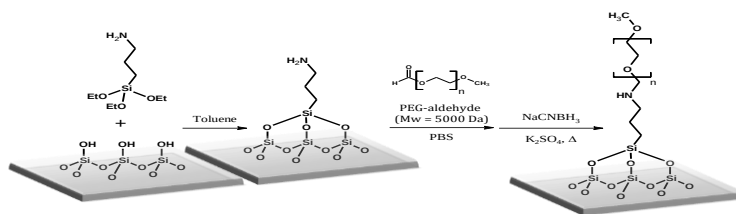


**Figure 1.** PEG-aldehyde grafting onto APTS modified silicon wafers.



**Figure 2.** The XPS C 1 s spectra of (A) PEG grafted at the 'cloud point' onto 2% APTS/0M  $K_2SO_4$ /60°C and (B) PEG grafted at the 'cloud point' onto 2% APTS /0.6M  $K_2SO_4$ /60°C.

<sup>1</sup> X. Gao, et al. *J. Am. Chem. Soc.*, **2009**, 25, 10271, <sup>2</sup> J. Zhang, et al, *Chem. Soc. Rev.*, **2010**, 39, 676-693, <sup>3</sup> M. Vyas, et al, *Soft Matter*, **2008**, 4, 1024-1032.

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## **Self-healing polymers**

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Intrinsically self-healing polymers (SHPs) have the ability to heal damage by the use of an appropriate stimulus. This healing can restore the mechanical properties of a damaged polymer allowing for return to service without the need for replacement of the polymer. When used as a coating, a SHP can ensure the continued protection of the underlying structure after damage, with minimal downtime of the component and no replacement of the coating necessary. Synthesis of polymers with the desired self-healing will be confirmed by IR & UV-vis and the polymers characterised by DSC, DMA and TGA.

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## Polypeptide-based brush polymers for antimicrobial therapies

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Antibiotic resistance is a growing cause for concern worldwide, with several prominent bacterial strains having recently developed plasmid mutations to survive treatment with commonly used antibiotics.<sup>1,2</sup> It is imperative to develop new antimicrobial agents that can have a delayed resistance response, for instance, polymers able to attack bacterial cell walls through non-specific modes of action. Synthetic mimics of antimicrobial peptides (AMPs) are attractive alternatives to traditional antibiotics because they are easily and inexpensively synthesized and allow for functionalization, yet they retain the most important features of AMPs, namely amphiphilicity.<sup>3</sup> This careful balance between cationic charges and hydrophobic moieties allows the synthetic AMPs to come in contact with the membrane of bacteria, with the hydrophobic parts exerting their cell membrane penetration effect and ultimately killing the bacteria.

Brush polymers are attractive architectures in the quest for antimicrobial efficacy because their degree of polymerization dictates their morphology, which can range from stars to rigid cylinders. The choice of monomer, polymerization technique, and side-chain population can lead to vastly different constructs, some of them with remarkable potency.<sup>4</sup>

In this study, we report the development of polypeptide-based norbornene bottlebrush polymers via ring opening metathesis polymerization (ROMP) as potential antimicrobial agents. Two synthetic approaches were explored, grafting from and grafting through (Fig. 1), with *N*-carboxyanhydride ring-opening polymerization (NCA-ROP) used to form polypeptide brush-arms. The antimicrobial activity and biocompatibility of the bottlebrush polymers were then evaluated against model bacterial and mammalian cell lines, respectively.

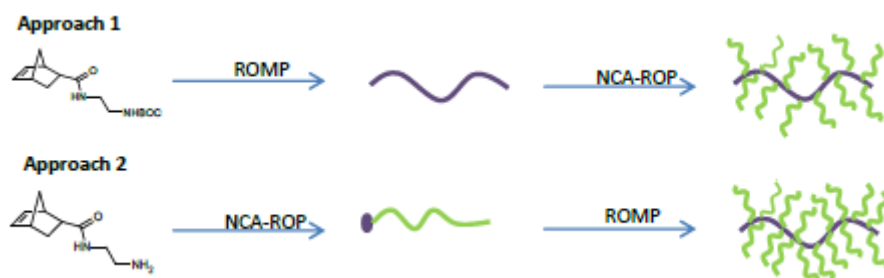


**Figure 1.** Synthetic routes for the formation of polypeptide-based bottlebrushes

<sup>1</sup>K. Kumarasamy. *Lancet Infect. Dis.* **2010**, 10, 597

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<sup>3</sup>K. Lienkamp. *Chem. Eur. J.* **2009**, 15, 11784

<sup>4</sup>K. Lienkamp. *J. Amer. Chem. Soc.* **2008**, 130, 9836

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## Comparison of suspension and solution polymerization for industrial carbon fiber production

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In 2007 a chemical regulation order was adopted in Europe and China, to protect the environment and human beings from hazardous substances in consumer goods and their working environment. It is a topic of interest for the rest of the world, as well. Some substances are banned by law from industrial application. The organic solvents Dimethylformamide (DMF) and Dimethylacetamide (DMAc) are candidates for prohibition. To be prepared, the producers of carbon fibers, hollow fibers and wet spun textile products are looking for alternative solvents for their production processes and try to gain according process Know-How. Aqueous solutions of inorganic salts are the most promising alternative.

In this context the polymerization method for the production of the polyacrylonitrile (PAN) polymer becomes more important. The use of inorganic salts allows solution polymerization, which has several advantages regarding production costs and the achievable molecular weight and weight distribution of the polymer.<sup>1,2</sup> Within the paper, dispersion polymerization (which is the common method for organic solvents) and solution polymerization are compared in regard to industrial carbon fiber production. In this context both processes are described on molecular level. It is shown, which polymerization method is best suited for which type of solvent and how the best fiber quality and most cost effective polymerization for carbon fiber production can be achieved.

<sup>1</sup>A.K. Gupta, D.K. Paliwal, P. Bajaj, *J. of Macromol. Sc.*, **1991**, *Polymer Reviews* 31, p. 1-89

<sup>2</sup>A.T. Derkov, G.A. Budnitskii, *Fiber Chemistry*, **1993**, Vol. 25, p. 3-7

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## Artificial Lipases

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Enzymes are nature's exquisitely efficient catalysts, enabling the reaction of even the least reactive molecules in ambient conditions. They are extensively used in a range of applications in food and dairy industries, leather and textile processing and in the manufacture of fine chemicals, detergents and pharmaceuticals, lending their exceptional specificity, high yields and rapid rates of reaction. Enzymes, being evolved proteins, are unstable outside their physiological environment and are extremely sensitive to variations in pH, temperature, solvents, and presence of foreign elements. These disadvantages along with the ever-rising demand for enzymes in industries, brings into question their economic feasibility. Hence, the search for artificial enzymes that can imitate native enzymes can be justified. Various studies on artificial enzymes have been developed but the turnover and catalytic activity still remain not up to that of the natural enzymes like lipases.<sup>1</sup> Lipases hydrolyse triacylglycerols (TAGs) to glycerol and fatty acids; and its use has been steadily increasing in diverse applications viz. detergents, medicines, for processes involving hydrolysis, esterification and transesterification in oil and fat industry. The syntheses of artificial lipases are reasonably economical as compared to the expensive industrial manufacturing processes of these enzymes. The primary aspect considered while imitating nature's enzymes is the positioning of the catalytic residues in the microenvironment of the enzyme mimic.<sup>2</sup> The simple artificial lipase mimic delivers hydrolysis reminiscing the action of the catalytic triad in natural lipases.

1. Motherwell WB, Bingham MJ, Six Y. Recent progress in the design and synthesis of artificial enzymes. *Tetrahedron*. **2001**;57(569):4663-4686.
2. Kuah E, Toh S, Yee J, Ma Q, Gao Z. Enzyme Mimics : Advances and Applications. *Chem A Eur J*. **2016**;22:1-28. doi:10.1002/chem.201504394.

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## Thixotropic supramolecular pectin-poly(ethylene glycol) methacrylate (PEGMA) hydrogels

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Pectin is an anionic, water-soluble polymer mostly consisting of covalently 1,4-linked  $\alpha$ -D-galacturonic acid units. This naturally occurring, renewable and biodegradable polymer is often underutilized in polymer science due to its insolubility in organic solvents, which renders conventional grafting techniques impossible. To circumvent this problem, cerium-initiated radical polymerization was utilized to graft methoxy-poly(ethylene glycol) methacrylate (mPEGMA) onto pectin in water. mPEGMA is soluble in organic solvents and so the resultant co-polymers exhibited increased organic solubility. The co-polymers were characterized by <sup>1</sup>H nuclear magnetic resonance spectroscopy, thermogravimetric analyses and differential scanning calorimetry. They were used in the formation of supramolecular hydrogel systems through the addition of  $\alpha$ -cyclodextrin ( $\alpha$ -CD) molecules to induce crosslinking. The hydrogels formed possessed thixotropic properties, where they could be shear-thinned into liquids upon agitation, but settled into gels at rest (**Figure 1**). Compared to most of the other hydrogels produced through the use of PEG-grafted polymers<sup>1, 2, 3, 4</sup>, the pectin-PEGMA/ $\alpha$ -CD hydrogels were unique in being unaffected by temperature changes.

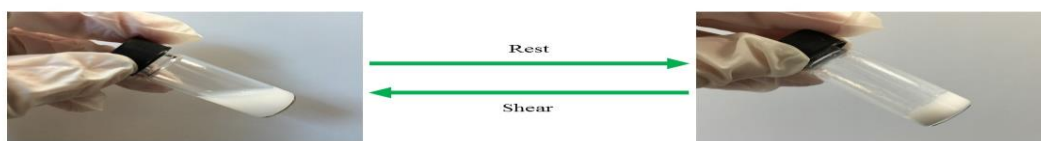


**Figure 1.** The liquid becomes gel when it is settled at rest and could be shear-thinned into liquids upon agitation.

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- <sup>2</sup>Huh, K. M., Cho, Y. W., Chung, H., Kwon, I. C., Jeong, S. Y., Ooya, T., Lee, W. K., Sasaki, S., & Yui, N. (2004). Supramolecular Hydrogel Formation Based on Inclusion Complexation Between Poly(ethylene glycol)-Modified Chitosan and  $\alpha$ -Cyclodextrin. *Macromolecular Bioscience*, 4 (2), 92-99.
- <sup>3</sup>Ren L., He, L., Sun, T., Dong, X., Chen, Y., Huang, J., & Wang, C. (2009). Dual-Responsive Supramolecular Hydrogels from Water-Soluble PEG-Grafted Copolymers and Cyclodextrin. *Macromolecular Bioscience*, 9 (9), 902-910.
- <sup>4</sup>Abdul Karim, A., & Loh, X. J. (2015). Design of a micellized  $\alpha$ -cyclodextrin based supramolecular hydrogel system. *Soft Matter*, 11 (27), 5425-5434.

## Prostaglandin Analogue Hydrogels for the Treatment of Glaucoma

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Glaucoma, a progressive optic neuropathy, is a leading cause of blindness worldwide. The most common treatment involves daily instillation of eye drops. However, less than 10 % of the instilled drug reaches aqueous humor and patient compliance to daily administration of drops is poor.<sup>1,2</sup> In order to overcome these barriers, efforts have been recently concentrated on developing new ocular drug delivery systems such as ocular implants.<sup>1,3</sup>

Here we present the synthesis and characterisation of poly(ethylene glycol)-based hydrogels covalently conjugated to prostaglandin analogues which can be fabricated in the form of an ocular implant and under physiological conditions release the prostaglandin analogue by means of hydrolysis of the degradable link between the drug and the hydrogel polymer. Both *in vitro* drug release and *in vivo* animal efficacy studies have been investigated (**Fig. 1**).

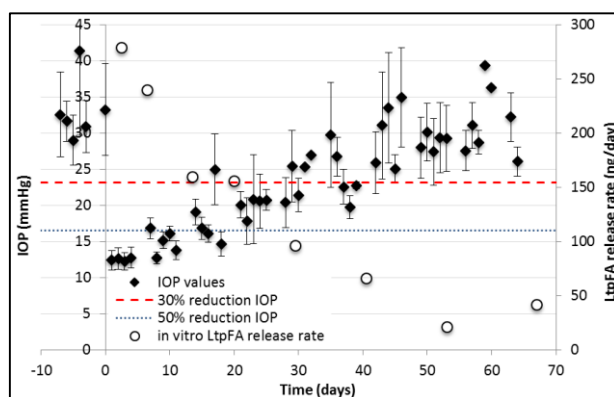


**Figure 1.** Average IOP (mmHg)  $\pm$  standard error of the mean *versus* time. The LtpFA implants were administered at day 0 into one eye of each of 4 dogs. IOP measurements were taken from Monday to Friday, inclusive, of each week. Baseline measurements were conducted for 7 days prior to administration of the implant.

<sup>1</sup> I. Januleviciene, L. Siaudvytyte, R. Barsauskaite, *Pharmaceutics*, **2012**, 4, 243-251.

<sup>2</sup> H.H. Chan, T.T. Wong, E. Lamoureux, S. Perera, *J. Glaucoma*, **2015**, 24 (7), 485-492.

<sup>3</sup> D. Ghatge, H.F. Edelhauser, *J. Glaucoma*, **2008**, 17 (2), 147-156.

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# Investigation of the structure-activity relationship of well-defined acrylamide copolymers as antimicrobial agents

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As more research focuses on the role of antimicrobial peptides (AMPs) as a way to tackle the development of resistance against antibiotics, synthetic versions of AMPs have recently attracted increasing interest<sup>1</sup>. Polymeric materials have predominantly appeared as the ideal candidates because their properties can be tuned easily and can be much cheaper to produce on industrial scale than peptides. Various systems were explored within the last decade with a wide range of backbones<sup>2</sup>, architecture of monomers or even sequence of monomers<sup>3</sup>. Polyacrylamides have yet to be investigated in terms of antimicrobial potency, as they could offer materials with greater stability towards hydrolysis. The design of these polymers was based on the composition of AMPs, which includes both hydrophobic and cationic amino-acids. A lysine-like acrylamide monomer was synthesised and copolymerised in different ratios with more hydrophobic monomers. Furthermore, the sequence of monomers was investigated through multiblock copolymers as it should alter the self-assembling behaviour of the chains, thus modify the interactions with the bacterial membranes (Fig. 1).

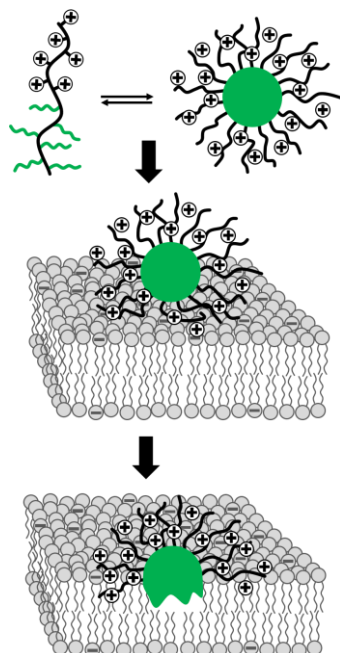


**Figure 1.** Schematic representation of the antimicrobial activity of block copolymers

1 Zasloff, M. *Nature* 2002, 415, 389.

2 Ganewatta, M. S.; Tang, C. *Polymer* 2015, 63, A1.

3 Locock, K. E. S.; Michl, T. D.; Stevens, N.; Hayball, J. D.; Vasilev, K.; Postma, A.; Griesser, H. J.; Meagher, L.; Haeussler, M. *ACS Macro Letters* 2014, 3, 319.

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# Understanding The In Vivo Response Of Self-Assembled Bioconjugates By Balancing The Stealth Versus Targeting Components

Amal Jayakumar Sivaram<sup>1,2</sup>, Adrian Fuchs<sup>1,2</sup>, Kristofer Thurecht<sup>1,2</sup>

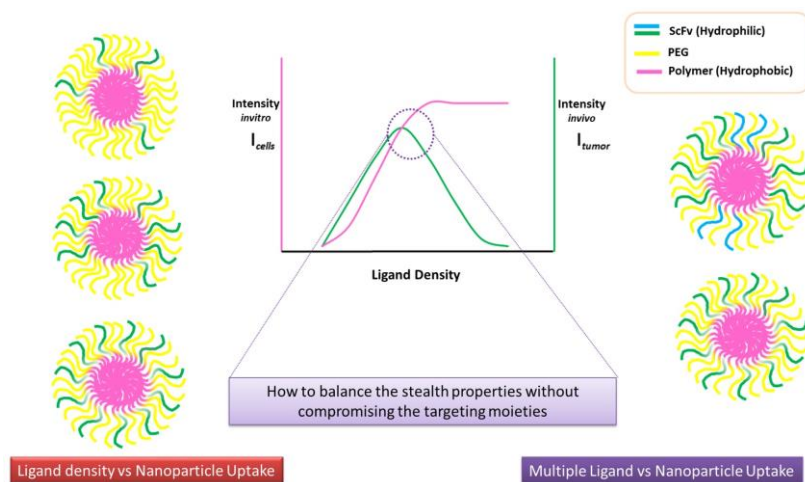
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The concept of the use of nanoparticle based receptor targeting receives much attention due to its capability for delivering drugs directly to the tumour site.<sup>1,2</sup> Research to date has established some general rules for designing new nanomaterials for drug delivery, with respect to the size, surface moieties etc. However, these generalised rules seldom transfer between the *in vitro* to *in vivo* and there is no clear methodology for optimising targeting approaches for drug delivery vehicles.

In this presentation we report on the development of amphiphilic protein-polymer conjugates for assembly into targeted micellar structures. By controlling the protein density of mixed micelles, we show how we can address key questions relating to optimisation of their function: 1) the effect of ligand density; and 2) the effect of multiple ligand binding to surface receptors. This approach gives fundamental insight into how these different parameters affect the rate and mechanistic pathways into cells, as well as a method to probe the intricate interplay between increased targeting efficiency (ligand density) versus the subsequent immune response (through protein recognition).

In summary, we have developed a novel class of targeted nanomedicines where ligand density is precisely controlled through chemistry and self-assembly. *In vitro* and *in vivo* studies allow us to investigate the parameters that dictate biological fate of nanomedicines.



**Scheme 1.** Development of micellar nanoparticles with defined targeting ligand density, followed by an investigation into both *in vitro* and *in vivo* behaviour to critically assess how structure affects drug delivery function. The box describes our hypothesis that an optimum density is key to balancing the targeting vs stealth effects.

S Xu, B.Z. Olenyuk, C.T. Okamoto, S. F. Hamm-Alvarez, *Advanced drug delivery reviews*, **2013**, 65(1), 121-138.

<sup>2</sup> A. V. Fuchs, B. W. C. Tse, A. K. Pearce, M.-C. Yeh, N. L. Fletcher, S. S. Huang, W. D. Heston, A. K. Whittaker, P. J. Russell and K. J. Thurecht, *Biomacromolecules*, **2015**, 16, 3235-3247.

# Incorporation of RAFT Synthesised Hyperbranched Polymers into Solid Networks by Photoinitiated Thiol-ene Reactions.

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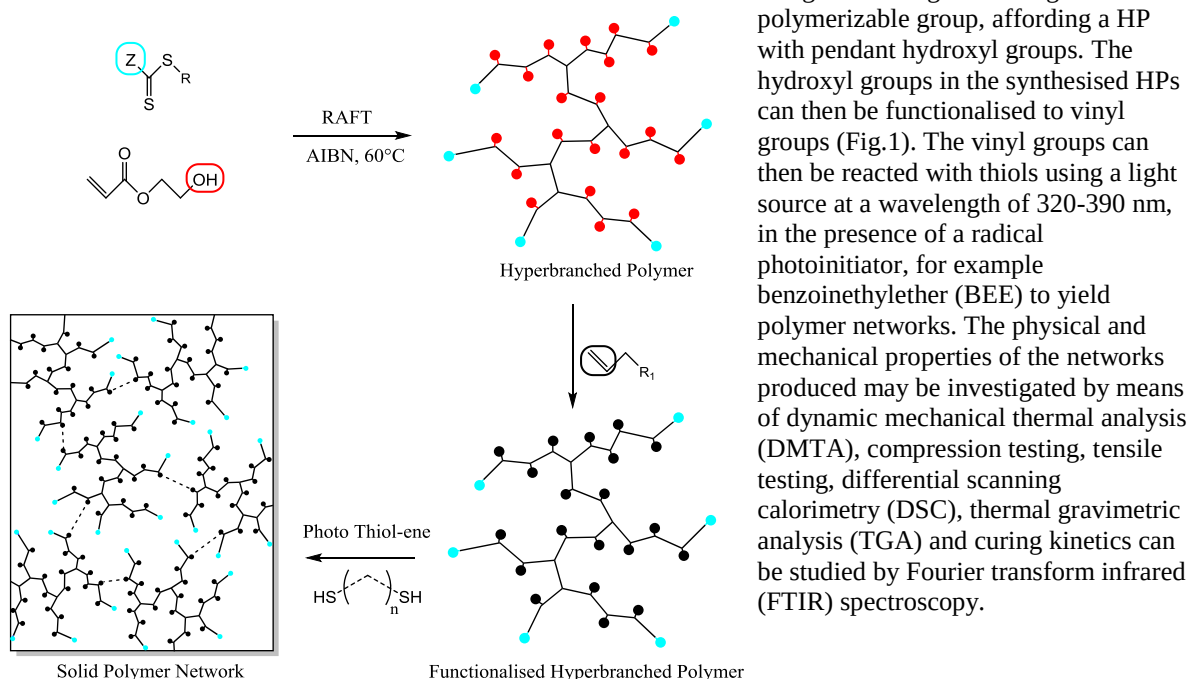
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There is a need for control over the structure of polymer networks for specific end applications in areas such as biomaterials, 3D printing and coatings.<sup>6,2</sup> Currently, polymer networks produced are largely restricted to the commercial availability of monomers. Additionally the desired purity of these monomers is often lacking. This is a shortcoming that drastically limits the structural composition of networks formed and in turn restricts the physical properties. With control of polymer network structures through tailored synthetic design of the monomers, designer networks can be produced with desired physical properties. Our aim is to create designer photo curable hyperbranched based resins for 3D printing application, and ultimately use a printing rig fitted with a monochromatic light source to cure complex structures layer by layer.

Hyperbranched polymers (HPs) can be synthesised by RAFT polymerisation of hydroxy-functional acrylates



using a RAFT agent bearing a polymerizable group, affording a HP with pendant hydroxyl groups. The hydroxyl groups in the synthesised HPs can then be functionalised to vinyl groups (Fig.1). The vinyl groups can then be reacted with thiols using a light source at a wavelength of 320-390 nm, in the presence of a radical photoinitiator, for example benzoinethylether (BEE) to yield polymer networks. The physical and mechanical properties of the networks produced may be investigated by means of dynamic mechanical thermal analysis (DMTA), compression testing, tensile testing, differential scanning calorimetry (DSC), thermal gravimetric analysis (TGA) and curing kinetics can be studied by Fourier transform infrared (FTIR) spectroscopy.

**Figure 1.** Scheme showing the step by step synthesis of functionalised RAFT synthesised hyperbranched polymers being incorporated into solid networks by thiol-ene reaction with dithiol molecules.

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# Structural characterization of multiscale starch-based cellulose composites

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Starch can be easily processed to a natural and biodegradable polymeric matrix. The thermoplastic starch (TPS) was introduced as a starch that had been heated to a high enough temperature and for enough time) [1]. Under these conditions, the starch granules are melted and plasticized, forming a material that represent a thermoplastic behaviour. However, one of the major problem associated with TPS films are high water sensitivity [2]. To address these issues, TPS can be reinforced by natural fibers. Their unique nanocellulosic structure can be isolated by a top-down approach and incorporated into polymeric matrices by a bottom-up approach to produce multi-functional, value-added materials that are sustainable, renewable, recyclable, and environmentally friendly.

In this research pulverized water retted kenaf bast fibers (0.5  $\mu\text{m}$ ) and nanofibers (7 nm) isolated by mechanical shearing of alkali treated fibers were used to reinforce a TPS matrix. Thin films with various loadings (0-10 wt%) were processed by casting and evaporation. The mixtures of aqueous suspensions of fibers, starch and glycerol, underwent gelatinization at the same time. The effects of filler type and loading on structural characteristics of prepared materials were studied using transmission and scanning electron microscopies (TEM and SEM), X-ray diffractometry (XRD), Fourier transform infrared spectroscopy (FTIR), thermogravimetric analysis (TGA), differential scanning calorimetry (DSC) and moisture absorption analysis.

SEM and TEM micrographs of the cryo-fractured surfaces of materials displayed homogeneous dispersion of nanofibers within the TPS matrix, indicating strong association between matrix and nanofiller. XRD results revealed that the diffractograms of composite films are only superimpositions of the diffractograms of the two components; starch and cellulose, and with increasing the nanofiber content the magnitude of the XRD peaks became more significant. FTIR graphs of the fabricated materials displayed similar features in the whole spectral range, regardless of the fiber content. TGA studies showed that thermal decomposition of the starchy matrix occurs in three main steps, and the  $T_{\text{max}}$  values of composite films shifted toward higher temperatures upon nanofibers addition. The TPS reinforced with 8% nanofibers was the most thermally stable film over all. According to DSC studies no significant effect on the melting point of composite films with reference to the unfilled matrix was observed. Moreover, addition of the nanoreinforcements effectively decreased the moisture sensitivity of the TPS film. A 21 % decrease in moisture content at equilibrium was observed with addition of 10 wt% nanofiber while this value was only 5.7% for the respective raw fiber reinforced film.

Figure 1. A brief illustration of results; from left to right: image of neat TPS, TPS-RF 6%, TPS-NF 6% films fracture surface from SEM, and moisture uptake of the TPS film and biocomposites conditioned at 98 % RH as a function of time.

<sup>1</sup> Lay G, Rehm J, Stepto RF et al., *US Patent 5,095,054*. **1992**, US Patent and Trademark Office, Washington, DC

<sup>2</sup> Kumar AP, Singh RP, *Bioresour Technol* . **2008**, 99:8803–8809

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## Thin film nanofiltration by advance polymeric material for water purification using Na<sub>2</sub>SO<sub>4</sub> draw solution

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Thin film nanofiltration (TFNF) membranes, were fabricated using polyamide (PA) /asymmetric polysulfone substrates via dope formulations. The membrane permeation and Na<sub>2</sub>SO<sub>4</sub> salt rejection performance was evaluated by using indigenously fabricated NF assembly. TFNF 4 was considered as an optimal membrane that exhibited a much higher permeation flux (35.4 L/m<sup>2</sup>.h) and salt rejection (95.6%) compared to the PSf membrane. Membranes were characterized by Fourier transform infra-red spectroscopy (ATR-FTIR), Scanning electron microscope (SEM), atomic force microscopy (AFM) and X-ray diffraction (XRD) analysis. These characterizations determine the successful interactions between polymer chains and other components and morphology. It was found that the membrane surface became smoother and more hydrophilic membrane. The SEM micrograph analysis showed average pore size < 100nm in TFNF4 membrane without any mottled surface.

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## Study and modeling of the coalescence of two different polymer particles PVDF-PMMA

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<sup>2</sup> Arts et Métiers ParisTech, Chambéry, France.

The present work focuses on the study of coalescence phenomenon of two different miscible polymers the PolyVinylidene Fluoride (PVDF), and Poly (methyl methacrylate) PMMA [ref miscibility]. First the blend has been characterized by thermal and thermomechanical methods; its rheological characteristics have been studied at molten state. Then the coalescence tests have been performed on the grains of these polymers using a polarized light optical microscope combined by a hot stage, video camera and video capture software. Different tests were carried out on different pairs of PVDF/PVDF, PMMA/PMMA and PVD/PMMA. The effect of different parameters like temperature, particle size on the rate of coalescence was investigated. In order to follow the effect of the neighboring grains on the kinetic of coalescence of two grains a third grain has been put in contact with these two grains in different positions. These tests showed clearly the miscibility of PMMA and PVDF.

The second part of this study concerns the modeling of coalescence of the grains of different polymers. In previous works, our group studied the effect of viscosity, surface tension and relaxation time on coalescence phenomenon and used the bellhumeur's <sup>1</sup> model in the case of two particles of the same polymer. We use the same model for the pair of PVDF/PMMA grains. In this case it is necessary to follow the effect of viscosity, surface tension and relaxation of each grain and also their miscibility on kinetic model.

<sup>1</sup> C. Bellehumeur, M. Kontopoulou, J. Vlachopoulou. Particle Coalescence and Bubble Dissolution in Rotational Molding. *Progress and Trends in Rheology*. **1998**, V, 37, 270-278.

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# Stochastic analysis of fracture under tension in crystalline polymeric solids

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The ductile fracture behavior under uniaxial tension of isotactic polypropylene (iPP), which is a typical plastic material, was investigated from the statistical point of views. Each tensile test was performed at room temperature more than one hundred times and the statistic data of break points, where the time to break, the ultimate stress (fracture strength), and the tensile toughness determined from the area under the stress-strain curve from the origin to the break point, were obtained from each tensile condition over one hundred tensile tests.

It was found that all the probability density curves of the fracture time, the ultimate stress and the tensile toughness approximately follow symmetric Gaussian statistics which is given by

$$p(x) = \frac{1}{\sqrt{2\pi}\Delta} \exp\left[-\frac{(x-\bar{x})^2}{2\Delta^2}\right] \quad (1)$$

Where  $x$  is the probability of fracture time, strength and toughness herein with the average  $\bar{x}$  separately, and the distribution variance  $\Delta$ . [1]

According to the fracture time result, as shown in fig.1, As increasing the elongation speed, the average fracture time tend to be shorter and the distribution variance of the time also tend to be narrower. Furthermore, there is a linear relationship between the elongation speed and no matter the average fracture time but also the distribution variance.

Consequently, we found a stochastic (namely Kolmogorov type) equation for fracture time of iPP to predict of mean and deviation of distribution of fracture time. The stochastic equation for the time to break can provide an explanation of the statistical properties of the strain at break. [2]

Also, the fracture toughness was an intrinsic characteristic, being independent of the elongation speed and gauge length, suggesting that the toughness, i.e. stored mechanical energy, has a possibility of being a criterion for tensile fracture.

In addition, as shown in fig. 2, we found a simple constitutive relation, where the stress is proportional to the elongation time around the break points (eq.2). Consequently, we can apply a static Kalman filter system to the present fracture data to determine a conditional probability density function. Also, this application made it possible to predict the mean and deviation of tensile strength of iPP under any tensile condition.

(2)

Where  $\sigma_B$  is ultimate strength,  $C_p$  is slope of the curve,  $t_B$  is fracture time,  $w_p$  is noise.

<sup>1</sup>C. Li, K. Nitta, *J. Macromol. Sci.*, **2015**, Part B: Phys., B54. 1053

<sup>2</sup>C. Li, K. Nitta, *J.phys.,Conference Ser.*,**2015**,622,012018

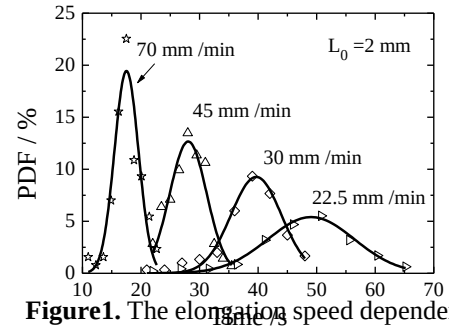


**Figure1.** The elongation speed dependence of the fracture time distribution of the iPP specimens with the gauge length of 2 mm.

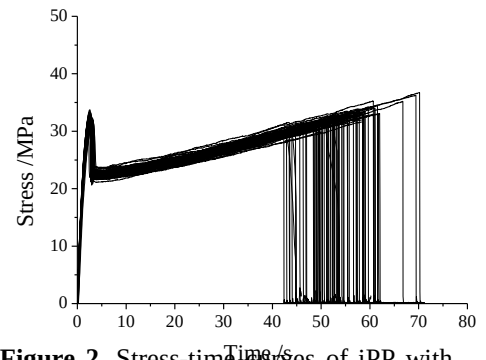


**Figure 2.** Stress-time curves of iPP with thickness of 200 mm, crystallinity of 59%, gauge length of 4mm, measured at 15 mm/min.

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# Property of CaCO<sub>3</sub> in situ treated bamboo pulp fiber reinforced composites by vacuum-assisted resin transfer molding

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The aim of this study is to investigate the properties of CaCO<sub>3</sub> in situ treated bamboo pulp fiber (BPF) composites that have been reinforced with epoxy resin by means of vacuum-assisted resin transfer molding (VARTM). Un-treated and treated BPF were processed at a pressure of 0.24 MPa into BPF preforms. VARTM was used to infuse epoxy resin through the BPF preforms to make BPF composites. The mechanical and thermal properties of the BPF composites were analyzed. CaCO<sub>3</sub> with the loading of 30% in situ modification has an effect on the performance of composite. The flexural strength did not decrease and modulus of the treated BPF composites increased by 9.38% while the impact strength decreased by 50.98%, compared to the control sample. Dynamic mechanical analysis revealed the maximum elastic moduli of the treated specimens increased by 1.67 times in the temperature range of -20-120°C. The thermal decomposition temperature of the composites was influenced by the effect of the crystal field and size effect of CaCO<sub>3</sub>.

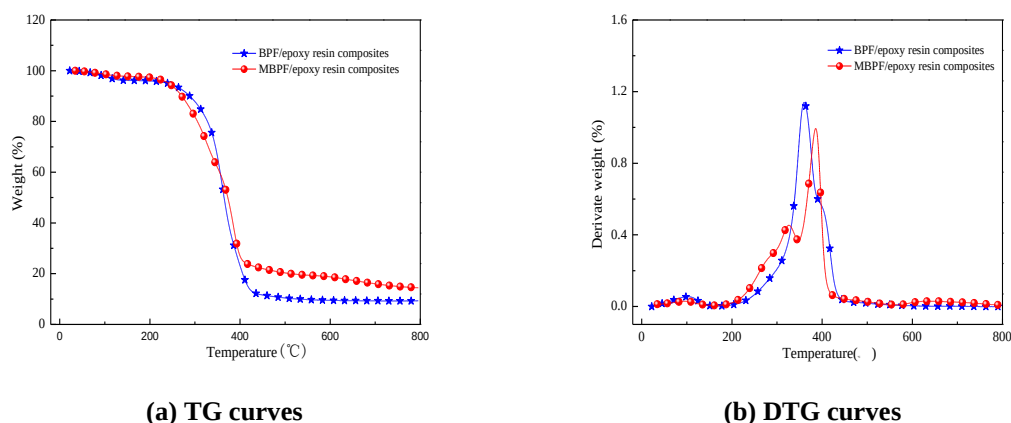


**Fig.5.** TG (a) and DTG (b) curves of composites manufactured by VARTM

<sup>1</sup> S.Q. Shi, D.J. Gardner, *Compos Part A Appl Sci and Manuf.* **2006**, 37, 1276-1285.

<sup>2</sup> H.T. Cheng, J. Gao, G. Wang, S.Q. Shi, S.B. Zhang, L.P. Cai, *Holzforchung.* **2015**, 69, 215-221.

<sup>3</sup> C.L Xia, S.Q. Shi, L.P. Cai, *Compos Part B En.* **2015**, 78, 138-143.

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# Experimental And Simulative Analysis Of The Thermal Behavior Of Polyacrylonitrile During The Stabilization

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Carbon fibres combine excellent mechanicals properties with low density and therefore they are very often used in lightweight constructions. The production of carbon fibers is divided into three parts. First part is the production of the raw material (precursor). The second is the thermal conversion to stabilize the fiber and prepare it for the final step. In the last part, the carbonization, almost all but carbon atoms are eliminated, resulting in a fibre material containing over 90% of carbon<sup>2</sup>.

Even though carbon fibre has been used since the late 1950s in aerospace application, there is still no general method available to find the optimal production parameters and the trial-and-error approach is most often the only resolution. Therefore, the stabilization process is analyzed experimentally and by simulation<sup>3</sup>.

The simulative approach is an enhancement on a previously developed simulation model for the exothermic main reactions (dehydrogenization, oxidation and cyclization) in a homogenous fibre bundle. Besides an improved investigation of the input parameters for the model, a multi-stage temperature profile during simulation is developed. With this simulation model, it is possible to perform a complete simulation of the fibre undergoing all zones of stabilization<sup>4</sup>.

Experiments were conducted to improve the input parameters for the simulation model and to validate the simulation results of the fibre temperature during stabilization. From DSC and TGA measurements the material parameters for the simulation are determined. The experiments for the validation were conducted on a pilot scale stabilization oven. To measure the fibre bundle temperature a new measuring method is developed.

The simulation model allows the calculation of the concentration of the main reaction products and the temperature of the fibre bundle for any position in the fibre bundle. (Fig.1). show the resulting temperature in the centre of the fibre bundle for a typical temperature profile.

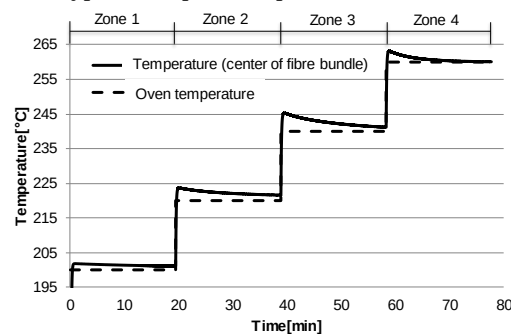


**Figure 1.** Simulated temperature in the centre of the fibre bundle

The graph shows, that the fibre bundle is heated up by the exothermic reactions. The temperature in the fibre bundle reaches the highest temperature after a few minutes and falls asymptotically afterwards. This effect is more pronounced in the higher Zones. Moreover, the average differences between the temperature of the fibre bundle and the oven temperature grow in the Zones 1 – 3. In the fourth Zone the difference drops again.

<sup>1</sup> E. Hau, Windkraftanlagen: Grundlagen, Technik, Einsatz, Wirtschaftlichkeit, 6. Auflage – Springer, 2008

<sup>2</sup> P. Morgan: Carbon fibres and their composites, Taylor & Francis, Boca Raton, 2005

<sup>3</sup> M.G. Dunham, D.D. Edie: Model of Stabilization for PAN-Based Carbon Fibre Precursor Bundles, Pergamon Press, Clemson, USA, Clemson University, 1992

<sup>4</sup> M. Akdere, F. Pohlkemper, G. Seide, T. Gries: Simulation Model for Stabilization of Carbon Fibres, Marmara Journal of Pure and Applied Sciences 2015, Special Issue-1: 19-22, DOI:10.7240/mufbed.62638

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# Click crosslinkable gelatin based hydrogels for the culture of OP9-DL4 cells: An initial step towards *in vitro* generation of T Cells

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Hydrogels are three dimensional hydrophilic polymeric networks which can absorb large amounts of water. Biocompatible hydrogels can better mimic the natural extracellular matrix and provide a suitable environment for cell adhesion and growth. In this study we have utilised a bioorthogonal strain-promoted alkyne-azide cycloaddition (SPAAC) click chemistry approach for the synthesis of gelatin based hydrogels<sup>1</sup>. Gelatin is a natural polymer derived from collagen, which retains the cell recognition RGD sequence. However, gelatin lacks mechanical stability to support cells. In this study we have functionalised gelatin with an azide group which can be crosslinked *in situ* with polyethylene glycol linker via dibenzylcyclooctyne (DBCO) - azide click reaction (Fig. 1).

T cell development occurs in the thymus through a complex process involving various developmental checkpoints, TCR gene rearrangement and positive/negative selection. In this process Notch signalling is an indispensable pathway that controls multiple cell fate decisions. Here we used OP9-DL4 cells which are bone marrow stromal cells transduced to express the Notch ligand Delta-like-4<sup>2</sup>. OP9-DL4 cells are capable to induce T cell differentiation *in vitro* in the absence of thymus. In the current study we have aimed to culture OP9-DL4 cells within the 3D hydrogel. Hydrogel properties such as mechanical strength, swelling and degradation were analysed.

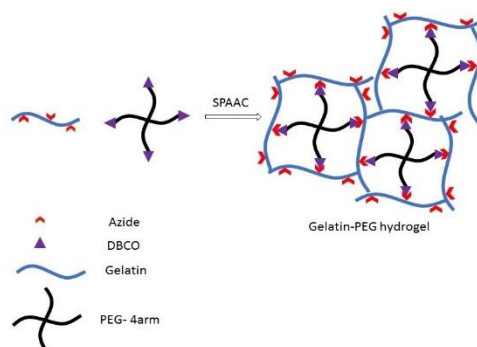


Figure 1: Representative scheme for the formation of gelatin-PEG hydrogels by SPAAC click chemistry

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