**Polymers at the Interface with Biology**

Timothy J. Deming,1,\* Harm-Anton Klok,2,\* Steven P. Armes,3 Matthew L. Becker,4 Julie A. Champion,5 Eugene Y.-X. Chen,6 Sarah C. Heilshorn,7 Jan C. M. van Hest,8 Darrell J. Irvine,9 Jeremiah A. Johnson,10 Laura L. Kiessling,11 Heather D. Maynard,1,12 Monica Olvera de la Cruz,13 Millicent O. Sullivan,14 Matthew V. Tirrell,15 Kristi S. Anseth,16 Sebastien Lecommandoux,17 Simona Percec,18 Zhiyuan Zhong,19 Ann-Christine Albertsson20

1 Departments of Bioengineering, Chemistry and Biochemistry, University of California, Los Angeles, California 90095-1600, USA.

2 École Polytechnique Fédérale de Lausanne (EPFL), Institut des Matériaux and Institut des Sciences et Ingénierie Chimiques, Laboratoire des Polymères, Bâtiment MXD, Station 12, CH-1015 Lausanne, Switzerland.

3 Dainton Building, Department of Chemistry, University of Sheffield, Brook Hill, Sheffield, S3 7HF, South Yorkshire, UK.

4 Department of Polymer Science, The University of Akron, Akron, OH 44325-3909, USA.

5 School of Chemical & Biomolecular Engineering, Georgia Institute of Technology, Atlanta, Georgia 30332-2000, USA.

6 Department of Chemistry, Colorado State University, Fort Collins, Colorado 80523-1872, USA.

7 Department of Materials Science & Engineering, Stanford University, Stanford, CA 94305, USA.

8 Department of Biomedical Engineering & Department of Chemical Engineering and Chemistry, Eindhoven University of Technology, P.O. Box 513, 5600 MB Eindhoven, The Netherlands.

9 Koch Institute for Integrative Cancer Research, Department of Biological Engineering, Department of Materials Science & Engineering, Massachusetts Institute of Technology, Cambridge, MA 02139, USA.

10 Department of Chemistry, Program in Polymers and Soft Matter, and Koch Institute for Integrative Cancer Research, Massachusetts Institute of Technology, Cambridge, MA 02139, USA.

11 Department of Chemistry, Massachusetts Institute of Technology, 77 Massachusetts Ave., Cambridge, MA 02139, USA.

12 California NanoSystems Institute, University of California, Los Angeles, 570 Westwood Plaza, Los Angeles, California 90095-1569, USA

13 Departments of Materials Science and Engineering, Chemistry, Chemical and Biological Engineering and Physics and Astronomy, Northwestern University, Evanston, Illinois 60208, USA.

14 Department of Chemical & Biomolecular Engineering, University of Delaware, Newark, Delaware 19716, USA.

15 Institute for Molecular Engineering, University of Chicago, 5640 South Ellis Avenue, Chicago, IL 60637, USA

16 Department of Chemical and Biological Engineering and the BioFrontiers Institute, University of Colorado Boulder, Boulder, Colorado 80309, USA.

17 Laboratoire de Chimie des Polymères Organiques, LCPO, Université de Bordeaux, CNRS, Bordeaux INP, UMR 5629, 16 Avenue Pey Berland F-33600 Pessac, France.

18 Department of Chemistry, Temple University, Philadelphia, PA 19122, USA.

19 Biomedical Polymers Laboratory, and Jiangsu Key Laboratory of Advanced Functional Polymer Design and Application, College of Chemistry, Chemical Engineering and Materials Science, Soochow University, Suzhou, 215123, P. R. China.

20 Fibre and Polymer Technology, Royal Institute of Technology, Teknikringen 56-58, SE-100 44 Stockholm, Sweden.

Biology in its broadest sense is an important model and inspiration for science and technology. In relation to polymers, biology uses a variety of complex macromolecules to accomplish myriad functions in living systems. These biopolymers incorporate many unique features that have inspired the polymer community, including sequence specificity, renewable feedstocks, catalytic activity, self-replication, and specific recognition. Bioinspired synthetic and biologically derived polymers are critical components of many innovative solutions aimed at addressing some of the most pressing problems related to human health and the environment. Challenges and opportunities for the polymer science community at large include both developing synthetic strategies towards such materials as well as studying and developing a fundamental understanding of their interactions with biological systems. Since its inception in 2000, *Biomacromolecules* has strived to become the leading forum for the dissemination of cutting-edge research at the interface of polymer science and biology. Articles published in *Biomacromolecules* contain strong elements of innovation in terms of macromolecular design, synthesis and characterization, or in new applications of polymers to biology and medicine.

The aims of this Editorial are to review the evolution of research at the interface of polymer science and biology, and to present a forward-looking view of this field. We do this by highlighting some areas of research that have been prominently featured in *Biomacromolecules* over the past years, and by presenting some emerging topics that we consider of great relevance and interest to the polymer science community and the readership of *Biomacromolecules*. This Editorial is partly based on a symposium entitled “*Polymers at the Interface with Biology*” and an associated “round-table” discussion that took place during the 2017 American Chemical Society (ACS) Fall Meeting in Washington DC. The participants of this discussion included the Editor-in-Chief and Associate Editors of *Biomacromolecules* as well as a group of 13 invited experts.

Research at the intersection of polymer science and biology has significantly evolved over the past two decades. To illustrate this, **Table 1** provides a collection of the most highly cited manuscripts published in *Biomacromolecules* since the start of the journal in the year 2000. **Tables 1** only includes original research papers, i.e. no review articles.. The table illustrates how the focus of many of the most cited papers in the field has gradually shifted over time. The focus of highly cited papers that appeared between 2000 and 2006 was heavily influenced by natural biopolymers (e.g. cellulose, silk) as well as electrospinning of polymer fibers. Other highly cited manuscripts that were published in *Biomacromolecules* during this period include seminal work on the development of reduction-sensitive block copolymer micelles12, the use of “click-type” conjugate addition reactions9,19 or DOPA chemistry17 to produce and functional hydrogel materials as well as the design of antibacterial surfaces films using surface-initiated controlled radical polymerization methods.30 By comparison, many highly cited papers published in *Biomacromolecules* between 2007 and 2013 report on the preparation, characterization, or use of cellulose nanofibers. In addition, this era features work on the preparation of non-fouling polymer coatings,51 and also shows an increased interest in the design of pH and/or reduction sensitive polymer nanocarriers for intracellular drug delivery47,64,82,84 and further examples to explore the utilization of DOPA-based chemistries for the preparation of polymer nanoparticles68 microcapsules73 and hydrogels.80 Highly cited work from the most recent period (2014 – present day) includes a number of articles focused on the development of surfaces or scaffolds designed to enhance tissue regeneration or for cell culture.88,89,91 Other examples include self-healing materials,105 mussel-inspired pH responsive hydrogels,87 investigation of how adsorbed proteins influence cellular uptake of nanoparticles,92 and several studies on pH, redox, temperature, and light responsive polymer particles designed to facilitate intracellular or intratumoral drug release.86,90,94,100 While the highly cited papers listed in **Table 1** and highlighted above reflect topics that have generated significant interest, it is also important to recognize that they, of course, are not exclusively representative of the content published in *Biomacromolecules*. As is evident also from **Table 1,** research fields continuously evolve and *Biomacromolecules* aims to capture new and exciting work at the forefront of the field.

**Table 1:** Overview of highly cited original research papers published in *Biomacromolecules* since 2000.

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| **Manuscript title** | **Authors** | **Year** | **Ref.** |
| Structural and Rheological Properties of Methacrylamide Modified Gelatin Hydrogels | Van den Bulcke, AI; Bogdanov, B; De Rooze, N; Schacht, EH; Cornelissen, M; Berghmans, H | 2000 | 1 |
| Quaternary Ammonium Functionalized Poly(propylene imine) Dendrimers as Effective Antimicrobials: Structure-Activity Studies | Chen, CZ; Beck-Tan, NC; Dhurjati, P; van Dyk, TK; LaRossa, RA; Cooper, SL | 2000 | 2 |
| New Insight Into Agarose Gel Mechanical Properties | Normand, V; Lootens, DL; Amici, E; Plucknett, KP; Aymard, P | 2000 | 3 |
| Multifunctional Epoxy Supports: A New Tool To Improve the Covalent Immobilization of Proteins. The Promotion of Physical Adsorptions of Proteins on the Supports before Their Covalent Linkage | Mateo, C; Fernández-Lorente, G; Abian, O; Fernández-Lafuente, R; Guisán, JM | 2000 | 4 |
| Periodate Oxidation of Crystalline Cellulose | Kim, UJ; Kuga, S; Wada, M; Okano, T; Kondo, T | 2000 | 5 |
| *Candida antartica* Lipase B Catalyzed Polycaprolactone Synthesis: Effects of Organic Media and Temperature | Kumar, A; Gross, RA | 2000 | 6 |
| Molecular Basis of Ca2+-Induced Gelation in Alginates and Pectins: The Egg-Box Model Revisited | Braccini, I; Pérez, S | 2001 | 7 |
| Relation between the Degree of Acetylation and the Electrostatic Properties of Chitin and Chitosan | Sorlier, P; Denuzière, A; Viton, C; Domard, A | 2001 | 8 |
| Conjugate Addition Reactions Combined with Free-Radical Cross-Linking for the Design of Materials for Tissue Engineering | Elbert, DL; Hubbell, JA | 2001 | 9 |
| X-ray Structure of Mercerized Cellulose II at 1 Å Resolution | Langan, P; Nishiyama, Y; Chanzy, H | 2001 | 10 |
| Mechanisms and Kinetics of Thermal Degradation of Poly(ε-caprolactone) | Persenaire, O; Alexandre, M; Degée, P; Dubois, P | 2001 | 11 |
| Glutathione-Sensitive Stabilization of Block Copolymer Micelles Composed of Antisense DNA and Thiolated Poly(ethylene glycol)-block-poly(L-lysine) | Kakizawa, Y; Harada, A; Kataoka, K | 2001 | 12 |
| Electrospinning of Collagen Nanofibers | Matthews, JA; Wnek, GE; Simpson, DG; Bowlin, GL | 2002 | 13 |
| Electrospinning *Bombyx mori* Silk with Poly(ethylene oxide) | Jin, HJ; Fridrikh, SV; Rutledge, GC; Kaplan, DL | 2002 | 14 |
| Disulfide Cross-Linked Hyaluronan Hydrogels | Shu, XZ; Liu, Y; Luo, Y; Roberts, MC; Prestwich, GD | 2002 | 15 |
| Genetically Encoded Synthesis of Protein-Based Polymers with Precisely Specified Molecular Weight and Sequence by Recursive Directional Ligation:  Examples from the Elastin-like Polypeptide System | Meyer, DE; Chilkoti, A | 2002 | 16 |
| Synthesis and Gelation of DOPA-Modified Poly(ethylene glycol) Hydrogels | Lee, BP; Dalsin, JL; Messersmith, PB | 2002 | 17 |
| Surface Modification of Polycaprolactone Membrane via Aminolysis and Biomacromolecule Immobilization for Promoting Cytocompatibility of Human Endothelial Cells | Zhu, Y; Gao, C; Liu, X; Shen, J | 2002 | 18 |
| Synthesis and Physicochemical Characterization of End-Linked Poly(ethylene glycol)-*co*-peptide Hydrogels Formed by Michael-Type Addition. | Lutolf, MP; Hubbell, JA | 2003 | 19 |
| Rational Design of Cytophilic and Cytophobic Polyelectrolyte Multilayer Thin Films | Mendelsohn, JD; Yang, SY; Hiller, J; Hochbaum, AI; Rubner, MF | 2003 | 20 |
| Complex Coacervation of Whey Proteins and Gum Arabic | Weinbreck, F; de Vries, R; Schrooyen, P; de Kruif, CG | 2003 | 21 |
| Ionic Strength Dependence of Protein-Polyelectrolyte Interactions | Seyrek, E; Dubin, PL; Tribet, C; Gamble, EA | 2003 | 22 |
| Bioactive Coatings of Endovascular Stents Based on Polyelectrolyte Multilayers | Thierry, B; Winnik, FM; Merhi, Y; Silver, J; Tabrizian, M | 2003 | 23 |
| Synthesis and Characterization of Injectable Poly(*N*-isopropylacrylamide-*co*-acrylic acid) Hydrogels with Proteolytically Degradable Cross-Links | Kim, S; Healy, KE | 2003 | 24 |
| Porous 3-D Scaffolds from Regenerated Silk Fibroin. | Nazarov, R; Jin, HJ; Kaplan, DL | 2004 | 25 |
| Effect of Sulfate Groups from Sulfuric Acid Hydrolysis on the Thermal Degradation Behavior of Bacterial Cellulose | Roman, M; Winter, WT | 2004 | 26 |
| Homogeneous Acetylation of Cellulose in a New Ionic Liquid | Wu, J; Zhang, J; Zhang, H; He, J; Ren, Q; Guo, M | 2004 | 27 |
| TEMPO-Mediated Oxidation of Native Cellulose. The Effect of Oxidation Conditions on Chemical and Crystal Structures of the Water-Insoluble Fractions | Saito, T; Isogai, A | 2004 | 28 |
| Structure and Properties of Silk Hydrogels | Kim, UJ; Park, JY; Li, C; Jin, HJ; Valluzzi, R; Kaplan, DL | 2004 | 29 |
| Permanent, Nonleaching Antibacterial Surfaces. 1. Synthesis by Atom Transfer Radical Polymerization | Lee, SB; Koepsel, RR; Morley, SW; Matyjaszewski, K; Sun, Y; Russell, AJ | 2004 | 30 |
| Effect of Reaction Conditions on the Properties and Behavior of Wood Cellulose Nanocrystal Suspensions | Beck-Candanedo, S; Roman, M; Gray, DG | 2005 | 31 |
| Elastic Modulus and Stress-Transfer Properties of Tunicate Cellulose Whiskers | Šturcová, A; Davies, GR; Eichhorn, SJ | 2005 | 32 |
| Sustained Release of Proteins from Electrospun Biodegradable Fibers | Chew, SY; Wen, J; Yim, EKF; Leong, KW | 2005 | 33 |
| Controlled Degradation and Mechanical Behavior of Photopolymerized Hyaluronic Acid Networks | Burdick, JA; Chung, C; Jia, X; Randolph, MA; Langer, R | 2005 | 34 |
| Preparation and Mechanical Properties of Chitosan/Carbon Nanotubes Composites | Wang, SF; Shen, L; Zhang, WD; Tong, YJ | 2005 | 35 |
| Characterization of the Surface Biocompatibility of the Electrospun PCL-Collagen Nanofibers Using Fibroblasts | Zhang, YZ; Venugopal, J; Huang, ZM; Lim, CT; Ramakrishna, S | 2005 | 36 |
| Homogeneous Suspensions of Individualized Microfibrils from TEMPO-Catalyzed Oxidation of Native Cellulose | Saito, T; Nishiyama, Y; Putaux, JL; Vignon, M; Isogai, A | 2006 | 37 |
| Electrospun Poly(ε-caprolactone) Microfiber and Multilayer Nanofiber/Microfiber Scaffolds:  Characterization of Scaffolds and Measurement of Cellular Infiltration | Pham, QP; Sharma, U; Mikos, AG | 2006 | 38 |
| Study of Biodegradable Polylactide/Poly(butylene adipate-*co*-terephthalate) Blends | Jiang, L; Wolcott, MP; Zhang, J | 2006 | 39 |
| PAMAM Dendrimer-Based Multifunctional Conjugate for Cancer Therapy:  Synthesis, Characterization, and Functionality | Majoros, IJ; Myc, A; Thomas, T; Mehta, CB; Baker, JR | 2006 | 40 |
| Coaxial Electrospinning of (Fluorescein Isothiocyanate-Conjugated Bovine Serum Albumin)-Encapsulated Poly(ε-caprolactone) Nanofibers for Sustained Release | Zhang, YZ; Wang, X; Feng, Y; Li, J; Lim, CT; Ramakrishna, S | 2006 | 41 |
| Superior Solubility of Polysaccharides in Low Viscosity, Polar, and Halogen-Free 1,3-Dialkylimidazolium Formates | Fukaya, Y; Sugimoto, A; Ohno, H | 2006 | 42 |
| Enzymatic Hydrolysis Combined with Mechanical Shearing and High-Pressure Homogenization for Nanoscale Cellulose Fibrils and Strong Gels | Pääkö, M; Ankerfors, M; Kosonen, H; Nykänen, A; Ahola, S; Österberg, M; Ruokolainen, J; Laine, J; Larsson, PT; Ikkala, O; Lindström, T | 2007 | 43 |
| Cellulose Nanofibers Prepared by TEMPO-Mediated Oxidation of Native Cellulose | Saito, T; Kimura, S; Nishiyama, Y; Isogai, A | 2007 | 44 |
| Interactions between Alginate and Chitosan Biopolymers Characterized Using FTIR and XPS | Lawrie, G; Keen, I; Drew, B; Chandler-Temple, A; Rintoul, L; Fredericks, P; Grøndahl, L | 2007 | 45 |
| Obtaining Cellulose Nanofibers with a Uniform Width of 15 nm from Wood | Abe, K; Iwamoto, S; Yano, H | 2007 | 46 |
| PEG-SS-PPS:  Reduction-Sensitive Disulfide Block Copolymer Vesicles for Intracellular Drug Delivery | Cerritelli, S; Velluto, D; Hubbell, JA | 2007 | 47 |
| New Nanocomposite Materials Reinforced with Flax Cellulose Nanocrystals in Waterborne Polyurethane | Cao, X; Dong, H; Li, CM | 2007 | 48 |
| Cellulose Nanopaper Structures of High Toughness | Henriksson, M; Berglund, LA; Isaksson, P; Lindström, T; Nishino, T | 2008 | 49 |
| The Shape and Size Distribution of Crystalline Nanoparticles Prepared by Acid Hydrolysis of Native Cellulose | Elazzouzi-Hafraoui, S; Nishiyama, Y; Putaux, JL; Heux, L; Dubreuil, F; Rochas, C | 2008 | 50 |
| Zwitterionic Polymers Exhibiting High Resistance to Nonspecific Protein Adsorption from Human Serum and Plasma | Ladd, J; Zhang, Z; Chen, S; Hower, JC; Jiang, S | 2008 | 51 |
| Fluorescence Study of the Curcumin-Casein Micelle Complexation and Its Application as a Drug Nanocarrier to Cancer Cells | Sahu, A; Kasoju, N; Bora, U | 2008 | 52 |
| Interaction of β-Lactoglobulin with Resveratrol and its Biological Implications | Liang, L; Tajmir-Riahi, HA; Subirade, M | 2008 | 53 |
| The Effect of Hemicelluloses on Wood Pulp Nanofibrillation and Nanofiber Network Characteristics | Iwamoto, S; Abe, K; Yano, H | 2008 | 54 |
| Transparent and High Gas Barrier Films of Cellulose Nanofibers Prepared by TEMPO-Mediated Oxidation | Fukuzumi, H; Saito, T; Iwata, T; Kumamoto, Y; Isogai, A | 2009 | 55 |
| Cellulose Whiskers versus Microfibrils: Influence of the Nature of the Nanoparticle and its Surface Functionalization on the Thermal and Mechanical Properties of Nanocomposites | Siqueira, G; Bras, J; Dufresne, A | 2009 | 56 |
| Individualization of Nano-Sized Plant Cellulose Fibrils by Direct Surface Carboxylation Using TEMPO Catalyst under Neutral Conditions | Saito, T; Hirota, M; Tamura, N; Kimura, S; Fukuzumi, H; Heux, L; Isogai, A | 2009 | 57 |
| Elastic Modulus of Single Cellulose Microfibrils from Tunicate Measured by Atomic Force Microscopy | Iwamoto, S; Kai, W; Isogai, A; Iwata, T | 2009 | 58 |
| Investigation of the Interaction between Berberine and Human Serum Albumin | Hu, YJ; Liu, Y; Xiao, XH | 2009 | 59 |
| Non-cytotoxic Silver Nanoparticle-Polysaccharide Nanocomposites with Antimicrobial Activity | Travan, A; Pelillo, C; Donati, I; Marsich, E; Benincasa, M; Scarpa, T; Semeraro, S; Turco, G; Gennaro, R; Paoletti, S | 2009 | 60 |
| Fabrication, Mechanical Properties, and Biocompatibility of Graphene-Reinforced Chitosan Composites | Fan, H; Wang, L; Zhao, K; Li, N; Shi, Z; Ge, Z; Jin, Z | 2010 | 61 |
| Cytocompatibility and Uptake of Halloysite Clay Nanotubes | Vergaro, V; Abdullayev, E; Lvov, YM; Zeitoun, A; Cingolani, R; Rinaldi, R; Leporatti, S | 2010 | 62 |
| Nanofiber Composites of Polyvinyl Alcohol and Cellulose Nanocrystals: Manufacture and Characterization | Peresin, MS; Habibi, Y; Zoppe, JO; Pawlak, JJ; Rojas, OJ | 2010 | 63 |
| Shell-Sheddable Micelles Based on Dextran-SS-Poly(ε-caprolactone) Diblock Copolymer for Efficient Intracellular Release of Doxorubicin | Sun, H; Guo, B; Li, X; Cheng, R; Meng, F; Liu, H; Zhong, Z | 2010 | 64 |
| Fast Preparation Procedure for Large, Flat Cellulose and Cellulose/Inorganic Nanopaper Structures | Sehaqui, H; Liu, A; Zhou, Q; Berglund, LA | 2010 | 65 |
| Entire Surface Oxidation of Various Cellulose Microfibrils by TEMPO-Mediated Oxidation | Okita, Y; Saito, T; Isogai, A | 2010 | 66 |
| Transition of Cellulose Crystalline Structure and Surface Morphology of Biomass as a Function of Ionic Liquid Pretreatment and Its Relation to Enzymatic Hydrolysis | Cheng, G; Varanasi, P; Li, C; Liu, H; Menichenko, YB; Simmons, BA; Kent, MS; Singh, S | 2011 | 67 |
| Bioinspired Polymerization of Dopamine to Generate Melanin-Like Nanoparticles Having an Excellent Free-Radical-Scavenging Property | Ju, KY; Lee, Y; Lee, S; Park, SB; Lee, JK | 2011 | 68 |
| Strong and Tough Cellulose Nanopaper with High Specific Surface Area and Porosity | Sehaqui, H; Zhou, Q; Ikkala, O; Berglund, LA | 2011 | 69 |
| Synthesis of Multiresponsive and Dynamic Chitosan-Based Hydrogels for Controlled Release of Bioactive Molecules | Zhang, Y; Tao, L; Li, S; Wei, Y | 2011 | 70 |
| Surface Charge Affects Cellular Uptake and Intracellular Trafficking of Chitosan-Based Nanoparticles | Yue, ZG; Wei, W; Lv, PP; Yue, H; Wang, LY; Su, ZG; Ma, GH | 2011 | 71 |
| Clay Nanopaper with Tough Cellulose Nanofiber Matrix for Fire Retardancy and Gas Barrier Functions | Liu, A; Walther, A; Ikkala, O; Belova, L; Berglund, LA | 2011 | 72 |
| Immobilization and Intracellular Delivery of an Anticancer Drug Using Mussel-Inspired Polydopamine Capsules | Cui, J; Yan, Y; Such, GK; Liang, K; Ochs, CJ; Postma, A; Caruso, F | 2012 | 73 |
| Relationship between Length and Degree of Polymerization of TEMPO-Oxidized Cellulose Nanofibrils | Shinoda, R; Saito, T; Okita, Y; Isogai, A | 2012 | 74 |
| Modulation of Cellulose Nanocrystals Amphiphilic Properties to Stabilize Oil/Water Interface | Kalashnikova, I; Bizot, H; Cathala, B; Capron, I | 2012 | 75 |
| Ultrastrong and High Gas-Barrier Nanocellulose/Clay-Layered Composites | Wu, CN; Saito, T; Fujisawa, S; Fukuzumi, H; Isogai, A | 2012 | 76 |
| Photoresponsive Poly(S-(o-nitrobenzyl)-L-cysteine)-b-PEO from a L-Cysteine N-Carboxyanhydride Monomer: Synthesis, Self-Assembly, and Phototriggered Drug Release | Liu, G; Dong, CM | 2012 | 77 |
| Transparent Films Based on PLA and Montmorillonite with Tunable Oxygen Barrier Properties | Svagan, AJ; Akesson, A; Cárdenas, M; Bulut, S; Knudsen, JC; Risbo, J; Plackett, D | 2012 | 78 |
| An Ultrastrong Nanofibrillar Biomaterial: The Strength of Single Cellulose Nanofibrils Revealed via Sonication-Induced Fragmentation | Saito, T; Kuramae, R; Wohlert, J; Berglund, LA; Isogai, A | 2013 | 79 |
| Self-Healing Mussel-Inspired Multi-pH-Responsive Hydrogels | Krogsgaard, M; Behrens, MA; Pedersen, JS; Birkedal, H | 2013 | 80 |
| Self-Assembling Behavior of Cellulose Nanoparticles during Freeze-Drying: Effect of Suspension Concentration, Particle Size, Crystal Structure, and Surface Charge | Han, J; Zhou, C; Wu, Y; Liu, F; Wu, Q | 2013 | 81 |
| Redox-Responsive, Core-Cross-Linked Micelles Capable of On-Demand, Concurrent Drug Release and Structure Disassembly | Wang, H; Tang, L; Tu, C; Song, Z; Yin, Q; Yin, L; Zhang, Z; Cheng, J | 2013 | 82 |
| Isolation of Thermally Stable Cellulose Nanocrystals by Phosphoric Acid Hydrolysis | Espinosa, SC; Kuhnt, T; Foster, EJ; Weder, C | 2013 | 83 |
| pH-Triggered Charge-Reversal Polypeptide Nanoparticles for Cisplatin Delivery: Preparation and In Vitro Evaluation | Huang, Y; Tang, Z; Zhang, X; Yu, H; Sun, H; Pang, X; Chen, X | 2013 | 84 |
| Dual Responsive Pickering Emulsion Stabilized by Poly[2-(dimethylamino)ethyl methacrylate] Grafted Cellulose Nanocrystals | Tang, J; Lee, MFX; Zhang, W; Zhao, B; Berry, RM; Tam, KC | 2014 | 85 |
| Light-Responsive Micelles of Spiropyran Initiated Hyperbranched Polyglycerol for Smart Drug Delivery | Son, S; Shin, E; Kim, BS | 2014 | 86 |
| Mussel-Mimetic Protein-Based Adhesive Hydrogel | Kim, BJ; Oh, DX; Kim, S; Seo, JH; Hwang, DS; Masic, A; Han, DK; Cha, HJ | 2014 | 87 |
| Electrically Conductive Chitosan/Carbon Scaffolds for Cardiac Tissue Engineering | Martins, AM; Eng, G; Caridade, SG; Mano, JF; Reis, RL; Vunjak-Novakovic, G | 2014 | 88 |
| Aerogel Microspheres from Natural Cellulose Nanofibrils and Their Application as Cell Culture Scaffold | Cai, H; Sharma, S; Liu, W; Mu, W; Liu, W; Zhang, X; Deng, Y | 2014 | 89 |
| PEG-b-PCL Copolymer Micelles with the Ability of pH-Controlled Negative-to-Positive Charge Reversal for Intracellular Delivery of Doxorubicin | Deng, H; Liu, J; Zhao, X; Zhang, Y; Liu, J; Xu, S; Deng, L; Dong, A; Zhang, J | 2014 | 90 |
| 3D Bioprinting Human Chondrocytes with Nanocellulose-Alginate Bioink for Cartilage Tissue Engineering Applications | Markstedt, K; Mantas, A; Tournier, I; Ávila, HM; Hägg, D; Gatenholm, P | 2015 | 91 |
| Protein Corona of Nanoparticles: Distinct Proteins Regulate the Cellular Uptake | Ritz, S; Schöttler, S; Kotman, N; Baier, G; Musyanovych, A; Kuharev, J; Landfester, K; Schild, H; Jahn, O; Tenzer, S; Mailänder, V | 2015 | 92 |
| Thermogelling Polymer-Platinum(IV) Conjugates for Long-Term Delivery of Cisplatin | Shen, W; Luan, J; Cao, L; Sun, J; Yu, L; Ding, J | 2015 | 93 |
| Bioreducible Shell-Cross-Linked Hyaluronic Acid Nanoparticles for Tumor-Targeted Drug Delivery | Han, HS; Thambi, T; Choi, KY; Son, S; Ko, H; Lee, MC; Jo, DG; Chae, YS; Kang, YM; Lee, JY; Park, JH | 2015 | 94 |
| Tea Stains-Inspired Initiator Primer for Surface Grafting of Antifouling and Antimicrobial Polymer Brush Coatings | Pranantyo, D; Xu, LQ; Neoh, KG; Kang, ET; Ng, YX; Teo, SLM | 2015 | 95 |
| Grafting of Bacterial Polyhydroxybutyrate (PHB) onto Cellulose via In Situ Reactive Extrusion with Dicumyl Peroxide | Wei, L; McDonald, AG; Stark, NM | 2015 | 96 |
| In Situ Synthesis of Antimicrobial Silver Nanoparticles within Antifouling Zwitterionic Hydrogels by Catecholic Redox Chemistry for Wound Healing Application | GhavamiNejad, A; Park, CH; Kim, CS | 2016 | 97 |
| Halloysite Clay Nanotubes for Enzyme Immobilization | Tully, J; Yendluri, R; Lvov, Y | 2016 | 98 |
| Structural Description of the Interface of Pickering Emulsions Stabilized by Cellulose Nanocrystals | Cherhal, F; Cousin, F; Capron, I | 2016 | 99 |
| Facile Construction of pH- and Redox-Responsive Micelles from a Biodegradable Poly(β-hydroxyl amine) for Drug Delivery | Li, D; Bu, Y; Zhang, L; Wang, X; Yang, Y; Zhuang, Y; Yang, F; Shen, H; Wu, D | 2016 | 100 |
| Enhanced Mechanical Properties in Cellulose Nanocrystal–Poly(oligoethylene glycol methacrylate) Injectable Nanocomposite Hydrogels through Control of Physical and Chemical Cross-Linking | De France, KJ; Chan, KJW; Cranston, ED; Hoare, T | 2016 | 101 |
| Optically Transparent Wood from a Nanoporous Cellulosic Template: Combining Functional and Structural Performance | Li, Y; Fu, Q; Yu, S; Yan, M; Berglund, L | 2016 | 102 |
| Highly Efficient Supramolecular Aggregation-Induced Emission-Active Pseudorotaxane Luminogen for Functional Bioimaging | Liow, SS; Zhou, H; Sugiarto, S; Guo, S; Chalasani, MLS; Verma, NK; Xu, J; Loh, XJ | 2017 | 103 |
| Amphiphilic and Hydrophilic Block Copolymers from Aliphatic *N*-Substituted 8-Membered Cyclic Carbonates: A Versatile Macromolecular Platform for Biomedical Applications | Venkataraman, S; Tan, JPK; Ng, VWL; Tan, EWP; Hedrick, JL; Yang, YY | 2017 | 104 |
| Facile Access to Multisensitive and Self-Healing Hydrogels with Reversible and Dynamic Boronic Ester and Disulfide Linkages | Guo, R; Su, Q; Zhang, J; Dong, A; Lin, C; Zhang; J | 2017 | 105 |
| Controlling Self-Assembling Peptide Hydrogel Properties through Network Topology | Gao, J; Tang, C; Elsawy, MA; Smith, AM; Miller, AF; Saiani, A | 2017 | 106 |
| Polyvalent Folate-Dendrimer-Coated Iron Oxide Theranostic Nanoparticles for Simultaneous Magnetic Resonance Imaging and Precise Cancer Cell Targeting | Luong, D; Sau, S; Kesharwani, P; Iyer, AK | 2017 | 107 |
| Effects of Xylan Side-Chain Substitutions on Xylan–Cellulose Interactions and Implications for Thermal Pretreatment of Cellulosic Biomass | Pereira, CS; Silveira, RL; Dupree, P; Skaf, MS | 2017 | 108 |

One important objective of the “*Polymers at the Interface with Biology*” symposium was to develop a forward-looking view of the field and highlight emerging topics that are of particular interest to the readership of *Biomacromolecules*. Many interesting topics relevant to this theme were presented by the speakers at the symposium in Washington DC. One example is the diverse field of biorelated synthetic polymers, which includes those based on natural biopolymers, such as polypeptides, polynucleic acids, and polysaccharides, as well as those that mimic nature, including polypeptoids and other peptidomimetics, polymers from biological feedstocks, and sequence controlled polymers.

For the field of bio-sourced sustainable polymers, Prof. Eugene Chen discussed and emphasized the importance of enhancing the thermal and mechanical properties of bio-derived synthetic polyesters and also realizing the potential to chemically recycle these polymers back to their constituent monomers. He reported catalytic systems capable of preparing such polyesters with enhanced properties via ring-opening polymerization of γ-butyrolactone and its derivatives, as well as the methodology that permits their complete depolymerization back to the original building blocks.109-111 Polymers containing functional side-chains and possessing the ability to respond to different stimuli continue to be developed as functional and structural mimics of biological polymers and assemblies. Related to this theme, Prof. Steven Armes presented the synthesis of pH responsive triblock copolymers designed to self-assemble into framboidal vesicles that were capable of mimicking the structural features and pH-triggered morphological transitions of certain viruses, e.g. the Dengue virus.

Engineered biorelated polymers, such as recombinant proteins and polymers produced using biocatalysis, are another core area for *Biomacromolecules*. In the symposium, Prof. Jan van Hest described engineered chimeric proteins composed of elastin like segments and cowpea chlorotic mottle virus subunits and their assembly into nanostructures that form biomimetic structures capable of responding to pH, temperature and salt. These assemblies take advantage of the stimuli responsive properties of elastin sequences, and the precision subunit assembly features of viral proteins.112-114 Related to this theme, Prof. Julie Champion discussed the design and preparation of protein constructs containing segments composed of coiled-coil and antibody binding motifs that enable them to assemble into well-defined nanostructures capable of binding and presenting antibodies. These nanocarriers are being evaluated for the intracellular delivery of therapeutic antibodies.115

Beyond synthesis and structure, understanding the properties and dynamics of biorelated polymer assemblies is also central to the scope of *Biomacromolecules*. Prof. Monica Olvera de la Cruz studies how multivalent ions and polymers can interact with amphiphilic molecules to form different morphologies with diverse chemical functionality.116-119 Modeling of such systems can lead to the discovery of new functional structures that can mimic biological functions. In studies aimed at mimicking coacervate formation observed with intrinsically disordered proteins in membraneless organelles within cells, Prof. Matthew Tirrell presented studies on complex coacervation of oppositely charged synthetic polyelectrolytes where a variety of features, including polymer stereochemistry, polymer chain length, and solution ionic strength were found to influence polyelectrolyte complex phase separation.120 These insights into protein/polyelectrolyte complexation show promise for the design of new biomimetic materials.121

An obstacle that presents a significant hurdle towards the clinical implementation of polymers and polymer-based nanomaterials for drug delivery applications is a lack of reproducible and scalable synthetic protocols. Polymer nanoparticles, as an example, are typically obtained via multiple formulation and modification steps. To overcome these challenges, Prof. Jeremiah Johnson described the preparation of macromolecular prodrugs starting from complex, small building blocks, which are accessible via organic synthesis.122-125 These small building blocks are then assembled together, for example, using ring-opening metathesis polymerization (ROMP), into the desired nanomaterial, thereby decoupling synthetic complexity and scalability.

Polymers and polymer nanoparticles are widely acknowledged for their ability to prolong the blood circulation time of therapeutics and to facilitate targeted delivery (e.g. to a tumor in cancer therapy). In addition to controlling plasma half-life and enabling targeted delivery of therapeutics, another pressing problem, in particular for biologics (such as peptide, protein or nucleotide based actives), is their stability during shipping and storage.126 This is a fundamental research problem, yet one with enormous impact in those parts of the world where an effective and reliable cold chain from the manufacturer to the patient is absent. Addressing this challenge, Prof. Heather Maynard emphasized the need for improved polymers for protein stabilization, especially for prolonged storage, and presented functional polymer designs, some also degradable, that enabled protein protection to heat and mechanical agitation. The polymers could be conjugated to proteins and peptides, added as excipients or surround the biologic as a nanoparticle for potential use in medicine.127-134

Considerable research over the past few decades has accumulated an increasingly robust understanding of the behavior of polymers and polymer nanoparticles in blood circulation as well as mechanisms for their cellular uptake. Fundamental design principles to control properties such as plasma half-life or to promote cellular internalization have been established for the preparation of more effective polymer nanomedicines. However, because the target for many active compounds is a specific cellular organelle, understanding and controlling the behavior of polymer nanomedicines at the sub-cellular level remains an important challenge. Aimed at addressing this issue, Prof. Millicent Sullivan and coworkers developed light-sensitive mPEG-b-poly(5-(3-(amino)propoxy)-2-nitrobenzyl methacrylate) polymers to deploy nucleic acids into cells with ‘on/off’ control over the timing and amount of delivery, and spatial control at cellular length scales.135-139

In addition to their utility for drug delivery, polymers and polymer assemblies also possess great potential for use in the broad realm of immunotherapy, including the targeted delivery of immunomodulatory drugs or vaccines to lymphoid organs or tumors. Prof. Darrell Irvine presented the use of polymer-based amphiphiles to increase the safety and potency of immunotherapies. Initially, these polymer amphiphiles were used to bind antigens and adjuvants to albumin.140,141 Next, these amphiphiles were designed to associate with a STING (Stimulator of interferon genes) agonist and assemble into nanofibers or nanodiscs that may be administered locally or systemically. Another strategy that underlines the potential of polymer science to advance immunotherapy was presented by Prof. Laura Kiessling, who described polymers that target antigens to dendritic cells.142 These polymers exploit the features of lectins, which are important for the recognition, uptake, and processing of antigens.143 She reported that the fate of glycosylated antigens in dendritic cells is affected by their physical properties (e.g., size, length), which can be altered using controlled polymerization techniques. These parameters define how polymers can be used to deliver antigens to dendritic cells to avoid immune detection or to promote immunity.

In addition to the diagnosis and treatment of human diseases, another important medical application for polymer-based materials is in the repair or regeneration of damaged or lost tissue. A particularly challenging problem in this context is bone defect generation, since it requires polymers that are exceptionally strong and at the same time can also degrade at designed intervals. Prof. Matthew Becker presented a class of α-amino acid based poly(ester urea)s (PEUs) that were designed for this purpose.144 One of the keys to the successful development of these materials were optimized step polymerization protocols and functionalization strategies, which afforded high molecular weight materials and provided great synthetic flexibility.145 In sheep segmental tibia defect models, the use of scaffolds fabricated from these PEU polymers allowed near to complete defect healing within 16 weeks.

Minimally invasive soft tissue regeneration demands hydrogels that provide mechanical protection to cells during injection and are also able to adapt to accommodate local cell remodeling of the polymer network.146,147 One approach towards such materials was presented by Prof. Sarah Heilshorn who described a new class of double-network hydrogels. Prior to injection, these materials are crosslinked *ex situ* by the formation of dynamic covalent hydrazone bonds that result from mixing a hydrazine modified elastin-like polypeptide (ELP) and an aldehyde modified hyaluronic acid. *In situ*, after injection, thermoresponsive aggregation of the ELP reinforces the network resulting in a hydrogel matrix that possesses viscoelastic stress-relaxation behavior.148

These topics presented in Washington DC highlight some of the research directions at the forefront of polymer science and biology, and represent areas and communities *Biomacromolecules* aims to serve. These fields are dynamic: new synthetic methodologies are being developed, more accurate characterization tools become available, and biology moves to smaller and smaller length-scales and becomes more quantitative. With these changes, and as new important societal challenges arise, *Biomacromolecules* endeavors to adapt to include emerging themes and scientific breakthroughs at the interface of polymer science and biology.

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