

## Polymers from Nature and for Nature

The field of polymer science has intimate historical connections with nature. It is interesting to note that polymers are actually indispensable to all living organisms. For example, cellulose, known as the most abundant organic material on Earth, is a main structural constituent for plants. Starch is a major component in foods like potatoes and cereal grains. Polyhydroxyalkanoates (PHAs) are produced in nature by various bacteria such as *Cupriavidus necator* and *Alcaligenes latus*. Natural rubbers are harvested from the *Hevea brasiliensis* trees. Chitin constitutes the exoskeletons of shellfish and insects. Silk is produced by the mulberry silkworms. Proteins, essential to all organisms including human being, are involved in basically every process in all living cells, and nucleic acids, carrying the genetic information in all life forms on Earth, are present in the cell nuclei. It is of great scientific interest to study the structures, properties, and biological functions of natural polymers not only to unveil the secrets of nature but also to find novel uses including biomedical and pharmaceutical applications for different natural polymers.

Ever since the early days of polymer research, the focus has been on how to design, synthesize, and apply novel polymers, in addition to natural polymers, for nature. For instance, biodegradable plastic bags and films have been developed to avoid soil pollution. Resorbable sutures and fixation devices are devised to replace nondegradable ones for better wound healing as well as to circumvent a second surgery to remove them. Poly(ethylene glycol) has been utilized to prolong the circulation time and significantly improve the treatment efficacy of therapeutic proteins in patients. Biodegradable micro-particles and implants are developed for sustained release of chemical drugs and proteins over a long period of time from weeks and months to years. Polymeric nanoparticles have been designed and investigated for targeted cancer therapy and early diagnosis. Thermosensitive polymer films have been applied for elegantly harvesting cell sheets that can help to restore functions of human organs. Three-dimensional porous scaffolds have been constructed to recruit live cells and/or support/guide cell growth to engineer or regenerate functional tissues. Cationic polymers and nanoparticles have been studied as nonviral vectors, which are safer, inexpensive, and can be repeatedly administered compared with viruses for gene therapy. It should be noted that the design and development of many of these synthetic polymers has been actually inspired by nature.

The examples provided above signify the interplay between polymer science and nature, which has grown quickly and diversified over the past years. *Biomacromolecules* aims to publish the most exciting work on polymers “from nature” and “for nature”. Examples of research in polymer science “from nature” include, for example, conceptual advances in the study and characterization or novel applications of natural polymers as well as novel biosynthesis of monomers and polymers. Polymers “for nature” encompass innovative synthetic approaches for biomedical and environmental polymers as well as design and development of new polymers for biological,

medical, or environmental applications. In recent years, particular focus has been on the development of polymers to combat diseases and to aid in the regeneration or repair of tissues.

The research on polymers “from nature” and “for nature” is very actively pursued in Asia. The objective of this Special Issue is to highlight some of the most exciting research that is being carried out in Asian countries such as China, India, Japan, Korea, and Singapore. Notably, this Special Issue contains a collection of 55 papers, which include 2 perspective articles, 10 review papers, and 43 research articles. For polymers “from nature”, Yen Wei et al. have given a comprehensive overview on the recent advances of melanin-like materials for various biomedical applications including bioimaging, photothermal therapy, and drug delivery.<sup>1</sup> Samir K. Maji et al. outlined recent progress of amyloid fibrils for cell adhesion and tissue engineering.<sup>2</sup> The groups of Zhengzhong Shao and Keiji Numata investigated the mechanical properties and structural transitions of *Antheraea pernyi* silk fiber induced by its contraction and combination of amorphous silk fiber spinning and postspinning crystallization for tough regenerated silk fibers, respectively.<sup>3,4</sup> Jian Ji et al. employed hemoglobin as a natural pH-sensitive nanocarrier for the delivery of the near-infrared dye IR780.<sup>5</sup> The groups of Changsheng Zhao, Nathaniel Hwang, and Mei-Chin Chen studied natural polysaccharides including carrageenan, heparin, hyaluronic acid, and chitosan-based systems as self-anticoagulant hemoperfusion adsorbents and for growth factor and vaccine delivery, respectively.<sup>6–8</sup> Ben Zhong Tang and Wang Zhang Yuan et al. disclosed clustering-triggered emission and persistent room temperature phosphorescence of sodium alginate.<sup>9</sup> Yongjun Zhang et al. reported that gossypol, a polyphenol isolated from cottonseed oil, followed a zero-order release profile from gossypol/PEG layer-by-layer films, leading to a significantly improved antifertility effect.<sup>10</sup>

For polymers “for nature”, the Xiquan Jiang and Lichen Yin groups have given overviews on translatable high drug-loading drug delivery systems based on biocompatible polymer nanocarriers and photoresponsive drug/gene delivery systems, respectively.<sup>11,12</sup> In-Kyu Park et al. reviewed recent advances of phototriggered nanoparticles for cancer immunotherapy.<sup>13</sup> Yanjing Wang and Chi Wu highlighted recent work on site-specific conjugation of polymers to proteins.<sup>14</sup> Nikhil Jana et al. wrote a perspective paper on anti-amyloidogenic chemical/biochemical-based nanoparticles as artificial chaperones for the efficient inhibition of protein aggregation.<sup>15</sup> Changyou Gao et al. gave an overview on the design and applications of cell-selective surfaces and interfaces.<sup>16</sup> The Bin Liu and Peter Ma groups summarized recent advances in conducting polymers for biomedical applications and tissue engineering, respectively.<sup>17,18</sup> Jayanta Haldar et al. reported recent progress in polymer research to tackle infections and antimicrobial resistance.<sup>19</sup>

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Jianzhong Du et al. wrote a perspective article on synthesis, self-assembly, and biomedical applications of antimicrobial peptide–polymer conjugates.<sup>20</sup> The groups of Byeongmoon Jeong, Xian Jun Loh, Manickam Jayakannan, Doo Sung Lee, Shiyong Liu, Sangyong Jon, Youqing Shen, Kazunori Kataoka, Yunching Chen, Linqi Shi, Zhongwei Gu, and Zhishen Ge designed and developed different polymeric nanosystems including thermo, pH, reduction, photo, ROS, and/or enzyme-sensitive systems for the targeted delivery of anticancer drugs,<sup>21–26</sup> nucleic acids,<sup>27–29</sup> photosensitizers,<sup>30,31</sup> and Fenton reactors,<sup>32</sup> respectively, for the treatment of diverse malignancies. The groups of Xintao Shuai, Yiyun Cheng, Jintao Zhu, Won Jong Kim, Zi-Chen Li, and Xiangyang Shi reported novel polymeric nanoassemblies for combination therapy of different tumors, which include the combination of chemotherapy and gene therapy,<sup>33,34</sup> chemotherapy and photothermal/dynamic therapy,<sup>35–37</sup> and radiotherapy and photothermal therapy.<sup>38</sup> The Guosong Chen and Lintao Cai groups reported glycolyx-mimicking nanoparticles for improved anti-PD-L1 cancer immunotherapy through reversion of tumor-associated macrophages and ROS-inducing micelles sensitizing tumor-associated macrophages to TLR3 stimulation for potent immunotherapy, respectively.<sup>39,40</sup> The Xuesi Chen and Nobuhiko Yui groups reported pH- and amylase-responsive carboxymethyl starch/poly(2-isobutyl-acrylic acid) hybrid microgels as effective enteric carriers for oral insulin delivery and pH-responsive coacervate droplets formed from acid-labile methylated polyrotaxanes as an injectable protein carrier, respectively.<sup>41,42</sup> Xianzheng Zhang et al. disclosed a universal approach to render nanomedicine with biological identity derived from cell membranes.<sup>43</sup> The Haeshin Lee and Li Ren groups developed boronic acid-alginate hydrogels with stretchable, self-healing, stimuli-responsive, remoldable, and adhesive properties and self-healable and bioadhesive hybrid polymeric hydrogels with mineralization-active functions, respectively.<sup>44,45</sup> Jianshu Li et al. studied multifunctional biomaterial coatings based on bioinspired polyphosphate and lysozyme supramolecular nanofilm.<sup>46</sup> En-Tang Kang and Mary B. Chan-Park et al. reported chitosan-based peptidopolysaccharides as cationic antimicrobial agents and antibacterial coatings.<sup>47</sup> The groups of Atsushi Maruyama and Priyadarsi De investigated cationic copolymers for allosteric control of peroxidase-mimicking DNAzyme activity and a recyclable thermoresponsive polymer– $\beta$ -glucosidase conjugate with intact hydrolysis activity, respectively.<sup>48,49</sup> Fujian Xu et al. reported a series of in situ photoinduced polymer graftings for sensitive detection of protein biomarkers via cascade amplification of liquid crystal signals.<sup>50</sup> Shu Wang et al. developed a supramolecular strategy based on conjugated polymers for discrimination of virus and pathogens.<sup>51</sup> The Jing Sun and Hua Lu groups studied charge-determined LCST/UCST behavior in ionic polypeptoids and salt- and pH-triggered helix–coil transitions of ionic polypeptides under physiology conditions, respectively.<sup>52,53</sup> Meng et al. investigated the impact of organocatalysts on the copolymerization kinetics and copolymer microstructure for organocatalytic ring-opening copolymerization of trimethylene carbonate and dithiolane trimethylene carbonate.<sup>54</sup> Hong Tan et al. reported simultaneous improvement of oxidative and hydrolytic resistance of polycarbonate urethanes based on polydimethylsiloxane/poly(hexamethylene carbonate)-mixed macrodiols.<sup>55</sup>

This Special Issue provides a sense of up-to-date research and development of polymers “from nature” and “for nature” in Asia,

which also nicely reflects hot research topics in the field of biomacromolecules. We envision that polymers “from nature” and “for nature” will attract continuously growing interest in the next decade.

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### Notes

Views expressed in this editorial are those of the authors and not necessarily the views of the ACS.

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