EDITORIAL

Introduction to advancements in synthesis of high-performance materials from Nature's building blocks

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The growing environmental concerns associated with traditional petroleum-based polymers have intensified the search for sustainable alternatives. This themed collection highlights innovative approaches in polymer science that prioritize sustainability without compromising performance. The featured articles encompass a spectrum of strategies, from the utilization of bio-derived monomers and biopolymers to the engineering of dynamic, self-healing materials.

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Bio-derived monomers for high-performance polymers

Harnessing renewable resources to create polymers that rival or surpass their synthetic counterparts is a focal point in sustainable materials research. In this context, Mahajan *et al.* (https://doi.org/10.1039/D4MA00039K) report the development of non-isocyanate polyurethanes (NIPUs) derived from lignin-based precursors, specifically bisguaiacols and bissyringols. By varying the number of methoxy groups on the aromatic rings, they successfully tuned the rheological, thermal, and mechanical properties of

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Eleftheria Roumeli is an assistant professor in the Materials Science & Engineering department of the University of Washington. With a focus on developing and understanding sustainable materials, her research group explores new families of bioplastics, biocomposites, and environmentally friendly construction materials derived from biological building blocks, and specifically from biopolymers. The group investigates the relationships between structure, processing, mechanical properties and life cycle impacts in these novel classes of sustainable materials. Prior to joining UW, Eleftheria completed her postdoctoral training at the California Institute of Technology (2017–2020) and ETH Zurich (2015–2017) – both in Departments of Mechanical Engineering. She earned her BS (2009) and PhD in Physics (2014) from the Aristotle University of Thessaloniki in Greece, where her research focused on understanding the structure–property relationships in synthetic polymer nanocomposite materials.

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Samantha Kristufek is an assistant professor at Texas Tech University. Her group began in 2021, where they work on employing the chemical toolbox to transform natural product-based starting materials into building blocks for the synthesis of modular, sustainable materials as well as upcycling commodity plastics. Prior to the start of her independent career, she completed postdoctoral training at Massachusetts Institute of Technology (chemistry, drug delivery) and the University of Melbourne, Australia (chemical engineering, nano-objects). She earned her PhD from Texas A&M University, where she worked on the synthesis and characterization of quercetin-based materials. She started her academic journey at Penn State Behrend where she earned her BS in chemistry.

the polymers. Notably, polymers with higher methoxy content exhibited increased hydrogen bonding, leading to enhanced elongation at break (up to 210%) and toughness (62 MJ m^{-3}). These bio-based NIPUs demonstrate mechanical performance on par with, or even superior to, petroleum-derived analogs, opening avenues for their application in packaging, automotive parts, and medical devices. Complementing this work, Wong and Korley (**https://doi.org/10.1039/D4MA00159A**) focused on designing bio-derived acrylate thermosets using lignin-based vanillyl alcohol diacrylate and bisguaiacol F diacrylate, mixed with bio-based *n*-butyl acrylate. By adjusting the aromatic content and diacrylate composition, they achieved significant tunability in storage modulus, Young's modulus, and thermal relaxation modes. These thermosets exhibit properties suitable for high-performance applications such as composites and damping materials, underscoring the potential of engineering polymers from bio-derived monomers.

Utilization of biopolymers in advanced materials

Moving beyond monomers, several studies explore the direct use of biopolymers to create materials with tailored properties. Subramani *et al.* (**https://doi.org/10.1039/D3MA01056B**) present a variety of protein-based materials that are made by blending recombinant amyloid silk proteins with distinct molecular features to produce composite fibers with customizable mechanical properties. By adjusting the ratio of proteins with different molecular weights, charges, and intrinsic mechanical characteristics, they demonstrated the ability to fine-tune fiber properties. This method holds promise for applications in textiles, biomedicine, and material engineering, where specific mechanical attributes are desired. In another innovative approach, García *et al.* (**https://doi.org/10.1039/D4MA00112E**) enhance the functionality of protein-based materials by integrating conductive nanoparticles.

They developed conductive and printable biocomposites by incorporating MXene Ti$_3$C$_2$T$_x$ into curli-fiber hydrogels. By loading 25–75 wt% MXene and optimizing the incorporation method, they achieved hydrogels with adjustable conductivity and strain tolerance ranging from 7% to 84%. The ability to retain conductivity upon storage further advances the applicability of these materials in flexible electronics and wearable devices.

Cellulose, the most abundant biopolymer, also features prominently in sustainable material development. Roberts *et al.* (**https://doi.org/10.1039/D3MA00936J**) addressed the challenge of dispersing cellulose nanocrystals (CNCs) by modifying their surfaces with oligosaccharides. Their paper tackled uncharged CNCs produced using HCl hydrolysis, which are highly crystalline but difficult to disperse as individual nanoparticles due to their lack of surface charge. The addition of oligosaccharides significantly improved the dispersibility and colloidal stability of CNCs in aqueous media. Interestingly, the charge content of the modified CNCs did not increase substantially with higher oligosaccharide content, suggesting that steric stabilization (rather than electrostatic repulsion) was the primary mechanism behind the improved dispersion. This process offers a sustainable and scalable method for enhancing the performance of CNCs without harsh chemical treatments, which are typically used to introduce surface charges.

Haridevan *et al.* (**https://doi.org/10.1039/D3MA00865G**) explore the incorporation of cellulose nanofibers (CNFs) into rigid polyurethane foams (RPUFs) to enhance thermal insulation and mechanical properties. Minimal additions as low as 0.01% w/w CNF, resulted in a 20% reduction in thermal conductivity. The study reported a 25% and 56% improvement in specific compressive strength and modulus, respectively, at low CNF loadings, particularly for low-density foams. This improvement is attributed to the good dispersion of CNFs, which acted as nucleating agents during foaming.

Biodegradable polymer blends and composites

The development of biodegradable polymers that do not sacrifice mechanical performance is crucial for implementation in everyday applications, which will reduce environmental impact. Wahbi *et al.* (**https://doi.org/10.1039/D4MA00233D**) designed a series of biodegradable polymer blends suitable for 3D printing by combining polylactic acid (PLA) and poly(butylene adipate-*co*-terephthalate) (PBAT), compatibilized with epoxidized canola oil (ECO). The addition of ECO improved printability and self-healing properties, resulting in significant increases in impact strength—62% for an 80/20 PLA/PBAT blend and 106% for a 70/30 blend compared to non-compatibilized blends. Enhanced fusion between printed strands reduced voids and improved ductility, demonstrating the potential of these blends for durable and biodegradable printed parts.

In their perspective, Clarke *et al.* (**https://doi.org/10.1039/D4MA00411F**) examined the potential of, and opportunities with, the emerging class of polyhydroxyalkanoates (PHA), which have been gaining momentum in the sustainable polymers landscape. They emphasize the importance of integrating PHA production with recycling technologies rather than relying solely on biodegradability. This approach addressed plastic waste more effectively by promoting a circular economy model where materials are continuously repurposed.

Dynamic polymers and molecular design for sustainability

Dynamic polymers that can self-heal or be reprocessed contribute significantly to sustainability by extending material lifespans and reducing waste. Rokni *et al.* (**https://doi.org/10.1039/D4MA00297K**) focus on enhancing the sustainability of commercial silicones by incorporating dynamic sulfur cross-links into polydimethylsiloxane (PDMS). By partially intercepting the traditional curing

process with sulfur vulcanization, they introduced reversible S–S bonds, creating a vitrimeric version of PDMS capable of self-repair. The modified silicones maintained comparable mechanical strength at lower sulfur concentrations and demonstrated self-healing properties, with full recovery achieved within 24–30 hours at 120 °C. However, the modified PDMS demonstrated lower thermal stability compared to the unmodified analogue, especially at higher sulfur content (15%), where signs of depolymerization were observed within 24 hours.

Understanding the molecular design rules that govern dynamic behavior is essential for advancing such materials. Khare *et al.* (**https://doi.org/10.1039/D3MA00799E**) investigated how the energy landscape of cross-linkers impacts the dynamic mechanical properties of polymer hydrogels using coarse-grained molecular dynamics simulations. The study focuses on ideal polymer networks with reversible cross-linkers. The model simulates a tetrahedral polymer network based on 4-arm PEG molecules cross-linked with metal ions (ex. Ni^{2+}), where the cross-linker chemistry significantly influences network mechanics. The results reveal that the energy potential of the cross-linker has a direct influence on network viscosity. Stronger cross-linker potentials lead to higher viscosities, and the relationship is more pronounced at lower shear rates, indicating that cross-linking chemistry can be used to fine-tune the mechanical behavior of hydrogels for various applications. The network's viscosity increases with the number of defects, such as dangling bonds or bond clusters, which develop during deformation. The defect rate is influenced by the interaction potential of the cross-linker, with weaker potentials leading to more defects and lower network connectivity. These findings contribute to the understanding of how molecular-level cross-linker interactions dictate macroscopic material properties. This is valuable for designing polymer hydrogels with specific mechanical performance for use in bioengineering, structural materials, and other advanced applications.

The collective efforts showcased in this themed collection underscore the multifaceted approaches being pursued to advance sustainable polymers. From the engineering of bio-derived monomers and the innovative use of biopolymers to the strategic design of dynamic, repairable materials, these studies contribute to a growing body of knowledge aimed at reducing environmental impact while meeting the demands of modern applications. By integrating renewable resources, enhancing biodegradability, and developing materials with extended lifespans, the polymer science community is making significant strides toward a more sustainable future.