Self-Assembly: From Chemistry to Applications

The concept of self-assembly—the spontaneous organization of smaller building blocks into larger structures—is remarkably diverse in breadth. Electrons localize in atomic orbitals, atoms bond together to form molecules, and molecules arrange themselves into (often complex) solids such as crystals. All of this chemical "magic" happens routinely when the right ingredients are combined under appropriate conditions, for example, using solution- or solid-phase synthetic techniques. Clearly, a topic this broad in scope transcends specific scientific disciplines and brings together researchers spanning the gamut of expertise, from chemistry to physics, materials, math, and beyond.

In the spirit of such interdisciplinarity, this special issue of *Journal of Polymer Science* focuses on a topic in the realm of self-assembly that attracts far-reaching interest among researchers and society alike: polymers. As will become clear by perusing the articles within, self-assembling polymers play a key role in a variety of familiar and upcoming applications, where fundamental insights into spontaneous organization across a wide range of length scales drive the design of new soft materials with unique performance.

Jin Kon Kim and coworkers (pol.20230292) start things off with a review on block copolymers—a well-known class of self-assembling polymers that has attracted significant attention because the structures they form can be controlled on the nanometer length scale. Their review focuses on unconventional morphologies such as asymmetric lamellae and highly swollen spheres that can be achieved with unusual but tractable molecular designs. **Joona Bang and coworkers** (pol.20230491) build on these concepts with a perspective on how to harness block copolymers for advanced patterning, both in the context of lithography, where small features of order 5 nanometers are desired, and optics, where nanostructures with periodicities closer to the wavelength of light promote useful phenomena such as photonic bandgaps.

Chunhua Cai and Jiaping Lin (pol.20220748) provide a complementary perspective on self-assembly in a very different but equally important context: solutions. Much like proteins in living organisms, synthetic proteins known as polypeptides that are also constructed from amino acids self-assemble in water to form rods, disks, plates, and other interesting nanostructures. Their perspective highlights state-of-the-art strategies to design and prepare self-assembling polypeptides, including insights into new types of driving forces that operate efficiently in solution. Surely inspired by these ideas but in a very different system, **Tim Long and coworkers** (pol.20230320) report original research that uses nucleobases (the building blocks of DNA), not amino acids, to construct supramolecular networks through blending. The morphology and resulting mechanical properties are sensitive to the efficiency of hydrogen bonding between nucleobase pairs attached to different polyacrylates, which hints at the role played by a polymer matrix in facilitating the formation of supramolecular interactions (or not) that yield unique mechanical properties. **Yoan Simon and coworkers** (pol.20230028) demonstrate the power of coupling these concepts in solution self-assembly with photoresponsivity using bottlebrush copolymers as a platform to control the aggregation of hierarchically assembled nanomaterials.

Blends are central to a number of other articles in this special issue as well. Returning to the topic of block copolymers, **An-Chang Shi and coworkers** (pol.20230306) simulate the impact of blending two distinct diblocks—AB and CD—on the resulting morphology with a focus on the sphere-forming region of phase space. Carefully tailoring the relevant Flory–Huggins interaction parameters (χ) and degrees of polymerization (N) results in microphase separation into different sphere packings similar to well-known (hard) alloys. Many of these predictions remain to be tested experimentally. **Cyrille Boyer and coworkers** (pol.20230634) also study blends but for a very different reason, namely 3D printing. Here, the polymerization of a typical formulation that includes a macro–chain-transfer-agent (macroCTA), monomer, and crosslinker results in a competition between microphase separation and solidification, leading to complex and aperiodic morphologies more reminiscent of macroscopic nucleation and growth or spinodal decomposition. By using blends of macroCTAs with different molecular weights, this work demonstrates an improved ability to control the average length scale of these interesting and potentially useful materials. Leveraging the topic of blending but again in a very different context, **Robert Macfarlane and coworkers** (pol.20230355) report a novel design strategy for tuning the performance of waterborne pressure-sensitive adhesives through blending with inorganic polymer-grafted nanoparticles. With clever molecular design, these organic–inorganic blends provide a mechanism to fill void space left upon the evaporation of water during casting, which would negatively impact performance. **Christopher Ellison and coworkers** (pol.20230404) study a different type of molecular architecture effect in the compatibilization of poly(ethylene terephthalate) and linear low-density polyethylene by adding small amounts of the homologous block copolymer poly(ethylene terephthalate)-*block*-polyethylene having either a triblock or multiblock sequence. These results have significant implications in resolving the plastic waste calamity, a topic that was featured as another recent special issue of *J. Poly. Sci.* (2022, 60, 3253–3255). Finally, bringing everything together, **Chungryong Choi and coworkers** (pol.20230293) provide a nice perspective on the role of sustainability in the design of next-generation thermoplastic elastomers. This timely subject brings the field full circle, as styrenic thermoplastic elastomers were one of the first commercial success stories for block copolymers produced by the Shell Oil Company (now Kraton) starting in 1955.

On behalf of the editors of *J. Poly. Sci.*, we would like to thank all of the reviewers and contributors who participated in this special issue. We hope readers find that it represents an interesting and useful intersection of disciplines in the area of self-assembly.

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