

Porous, Biomimetic Hydrogels Demonstrating Structurally-Dependent Mechanical Properties

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Abstract: Although materials mimicking the microstructure and properties of natural tissues are highly sought after in biomedical research, current approaches fixate on replicating tissue properties in bulk materials without the necessary microstructure to support cell proliferation. Here, we demonstrate a new method of producing hydrogels with biomimetic, porous structures resulting from the self-assembly of amphiphilic triblock copolymers. Most notably, we show that introducing biomimetic microstructures significantly impacts the mechanical properties, resulting in reversible, elastic deformation mechanisms reminiscent of natural tissues.

Poly(styrene-*b*-ethylene oxide-*b*-styrene) block copolymers self-assemble on contact with water, forming a network composed of hydrophobic micelle cores bridged by hydrophilic mid-blocks. Current methods of producing hydrogels from these block copolymers involve hydrating the polymer directly. In contrast, if the polymer is first dissolved in a water-miscible solvent and injected into a water bath, the diffusion of the water-miscible solvent will produce micron-sized pores in the micelle network.

Remarkably, the porous hydrogels exhibit dramatically increased elasticity and high reversibility, as well as a strain-hardening response similar to that of natural tissue. The mechanical properties of the hydrogels are closely tied to pore morphology, which can be tuned via the solvent and concentration used in the initial solution. Additionally, the micelle network can be modified by varying the size and relative composition of the block copolymer itself, allowing additional avenues to control the resulting mechanical properties. Furthermore, if gels are dried under strain, the aligned crystallization of the mid-blocks produces strong and repeatable actuation triggered by melting the crystals via heat or water. This exciting class of materials presents both diverse potential applications and an opportunity to further explore the complex physics of block copolymer self-assembly.