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Design stiff, tough and stretchy hydrogels via nanoscale hybrid crosslinking and macroscale fiber reinforcement

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ABSTRACT

Hydrogels' applications are limited by their weak mechanical properties. The toughness, modulus, and strength of conventional hydrogels (single network gels) are, respectively, $<10 \text{ J m}^{-2}$, $<100 \text{ kPa}$, and $<10 \text{ kPa}$, which fail to provide sufficient mechanical properties in large quantities of applications. Here, we designed highly stretchable, tough, yet stiff hydrogels via nanoscale hybrid crosslinking and macroscale fiber reinforcement. We used 3D printing technology to fabricate 3D patterned fibrous structures. Hydrogel composites were constructed by impregnating the PLA fiber mesh with highly stretchable and tough PAAM-alginate hydrogels. Synthetic gels can reach fracture energies of $\sim 9000 \text{ J m}^{-2}$. However, modulus of these tough hydrogels is only $\sim 100 \text{ kPa}$. Here, we designed fiber reinforced hydrogels, which can reach fracture energy of about $30\,000 \text{ J m}^{-2}$ and modulus of $\sim 6 \text{ MPa}$. The enhancement of toughness is due to multiscale toughening mechanism which spans over multiple length scales ranging from nanometers to millimeters. This design of fiber reinforced hydrogel composites can serve as a model to expand the application of hydrogels in both biomedical and robotic areas.