Enhancing Stiffness and Toughness of Hydrogel by Internal Structure Remodeling

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Efforts for mechanical reinforcement of hydrogels are mostly focused on enhancing rigidity. Generally, increase in rigidity degrades extensibility, and resulting scarce increase or even decrease in toughness. Here we report a method that can greatly enhance rigidity and toughness of preformed hydrogel simultaneously by internal structure remodeling. The remodeling of hydrogel is carried out by unidirectional stretching and solvent exchange, and the remodeled structure is subsequently fixed by ionic crosslinking. The solvent exchange step makes aligned polymer chains in the stretched hydrogel much closer. The resulting densification and enhanced alignment of polymer chains allow dense ionic crosslinking of alginate chains. In addition, the solvent exchange induces macroscopic rigidity gradient and phase separation that make the hydrogel tougher by complementary interaction between mechanically contrasted regions. As an appropriate poor solvent could be applied to any types of hydrogels to induce polymer-polymer self-interactions, a similar structure remodeling of hydrogels to modulate their mechanical property by solvent exchange will be broadly applicable to other hydrogel systems.