3D Printing Method for Tough Multifunctional Particle-Based Double-Network Hydrogels

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3D printing of hydrogels finds widespread applications in biomedicine and engineering. Practical examples like artificial cartilage and heart valves, soft robots require high mechanical performance of complex structures. While recently many tough hydrogels have been developed, complicated synthesis processes hinder their fabrication in 3D printing. Here, a strategy is proposed to formulate hydrogel inks, which can be printed into various strong and tough particle-based double-network (P-DN) hydrogels of arbitrary shapes without any rheological modifiers. This hydrogel ink consists of microgels and a hydrogel precursor. The microgels are individual highly crosslinked networks. They are prepared by swelling dried microparticles in the hydrogel precursor that consists of monomers, intiators, and crosslinkers. Microgels regulate the rheological properties of the hydrogel ink and enable the direct printing. After printing and curing, the precursor forms a sparsely crosslinked network that integrates the microgels, leading to a P-DN hydrogel. (**Fig. 1**) It is a universal strategy that applies to a variety of hydrogel systems, including common hydrogels, stimuli-responsive hydrogels, biocompatible hydrogels, hydrogel composites with high mechanical performance and strong adhesion to diverse materials. This strategy will open new avenues to fabricate multifunctional devices in tissue engineering and soft robotics.

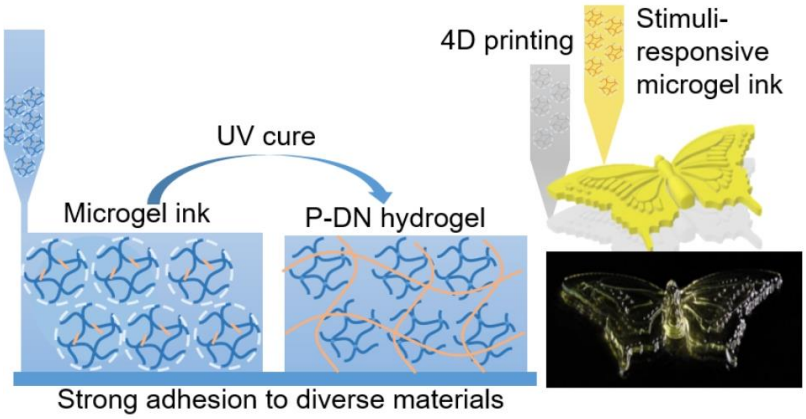

Fig. 1. Principle of 3D printing of P-DN hydrogel. The hydrogel ink consists of microgels and a hydrogel precursor. The microgels regulate the rheolgoy during printing. After curing, the monomers polymerize into a second network, integrating the microgels to form the P-DN hydrogel.

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