VIDEO ARTICLE

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3D Printing of Functional Hydrogels

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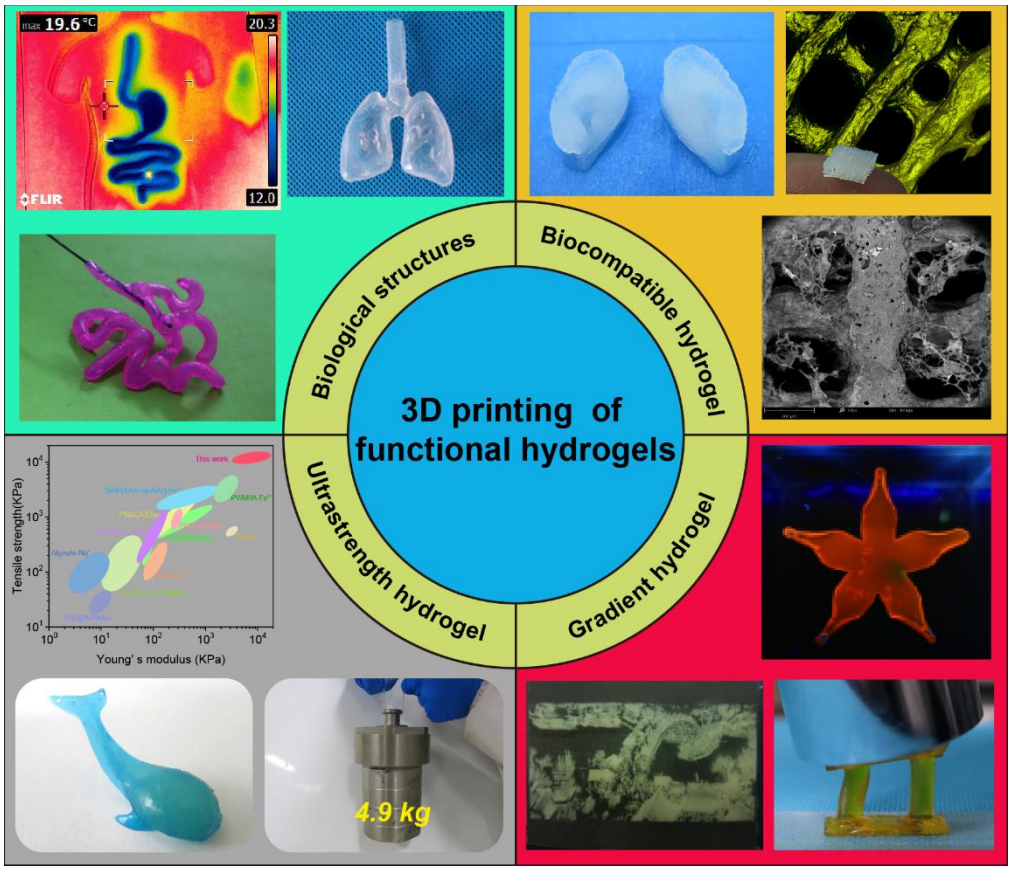
Graphical Abstract

Three-dimensional (3D) printing of functional hydrogels has been widely used in biomedicine and many other fields. Here, we proposed a series of new functional hydrogels, such as hollow hydrogels with biological geometries, biocompatible hydrogel used for cell culture, ultra-strength hydrogel for artificial cartilage, and the gradient hydrogel for hydrogel patterning and actuators. The resultant hydrogels possess excellent comprehensive performance including tailored structures, good mechanical properties, biocompatibility. These 3D printed functional hydrogels are believed to provide emerging opportunities in biomedicine, smart devices and soft machines, etc.



Highlights

- 3D printed thermoreversible hydrogel templates realized biological hollow-hydrogel structures with high complexity.
- 3D printed dual physical crosslinking hydrogel achieved good biocompatibility and ultrahigh strength and toughness.
- Grayscale image projection realized 3D printed hydrogels with coding shape deformation and patterning.

Abstract

Three-dimensional (3D) printing of hydrogels with the desirable structures and functions has been widely used in many fields, including biomedicine, soft robotics, flexible electronics, smart actuators, and so on. However, the current hydrogels still remain the fundamental challenges on the structural complexity, weak mechanical properties, uncontrollable physical-chemical properties for biomedical and engineering purposes. Herein, we propose novel strategies to 3D printing of functional hydrogels with the complex architectures, good mechanical performance and gradient heterogeneity in physical-chemical properties. A series of physical crosslinking hydrogels were 3D printed to achieve the hydrogels with the biological geometries, outstanding biocompatibility, swelling-resistance, and the ultrahigh strength and toughness. Specifically, the printed thermoreversible gelatine/carrageenan hydrogel templates combined with the surface supramolecular assembly process were used to construct the biological hollow-hydrogel structures, such as branched vascular replicas for simulating the guidewire navigation, digestive system for in vitro drug delivery, distal lung subunit, and glomerulus with outstanding mechanical robustness and favorable physiological environments. Moreover, direct ink writing of (polyvinyl alcohol (PVA) and κ-carrageenan) hybrid hydrogel ink with outstanding rheology and thixotropy achieved the pure physical crosslinking hydrogel scaffolds and biological objects with good mechanical properties, biocompatibility, as well as bioactivity, in which cells not only attach well to the surface of the hydrogels, but also stretch into the spaces in the grid architectures, providing appropriate microenvironments for cell culture. In fact, 3D printing of hydrogels with high intrinsic mechanical performance is of significant engineering applications in artificial cartilage, soft robotics and impact-resistant devices yet has been proven to be a fundamental challenge. We propose a novel strategy of dual physical crosslinking (DPC) networks based on PVA and chitosan (CS) to realize the 3D printing of ultrahigh strength hydrogel. The cyclic freezing-thawing followed by sodium citrate solution soaking realize the first network of PVA crystallization and the second one of CS ionic interaction between amino and carboxyl groups, respectively. The printed DPC hydrogels display the optimized tensile strength of 12.71 ± 1.32 MPa at tensile strain of 302.27 ± 15.70%, Young's modulus of 14.01 ± 1.35 MPa, elongation at break of 22.10 ± 2.36 MJ m^{-3} and toughness of 9.92 ± 1.05 kJ m^{-2} due to the efficient energy dissipation of stiff chitosan ionic network. This robust hydrogel 3D printing can readily realize not only hydrogel structures such as lattices, honeycombs and springs, but also secondary shaping hydrogel objects including whale, octopus and butterfly via the local DPC strategy. Finally, we provided the protocol, named as grayscale digital light process (G-DLP), to prepare the gradient hydrogel in three dimensions. The grayscale projection causes the difference of crosslinking density of hydrogel spatially, which resulted in the gradient heterogeneity in mechanical performance, swelling, wettability, transmittance, etc. As results, the bioinspired hydrogel actuators with the coding shape deformation and the hydrogel patterning with the high-definition and reversibility were realized. We believe that the 3D printing of functional hydrogels will pave the way towards various fields for extensive potentials including soft machine, biomedicine, tissue engineering, and bionic engineering.

Keywords: Hydrogel; 3D printing; organ replicas; actuation; ultrahigh strength and toughness.

Acknowledgements

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Biography of Presenting Author

Xiaolong Wang is a full Professor at the Lanzhou Institute of Chemical Physics (LICP), Chinese Academy of Sciences (CAS). He joined the LICP in 2007 when he got his PhD in the major of Polymer Chemistry and physics at Lanzhou University. During his stay at the LICP, he spent a year (2010–2011) in Hong Kong Polytechnic University as a Research Associate and then two years (2011–2013) at the University of Western Ontario (Canada) as an Adjunct Professor. He was promoted to a full professor at LICP in 2014. From then on, he is leading a 15+ members research group at the State Key Laboratory of Solid Lubrication named as 3D printing Tribology Devices. His research interests focus on 3D printing, biomimetic lubrication, functional soft materials, and flexible electronics and his achievements have been published in more than 80 peer-reviewed papers with a current h-index of 36, and issued 20+ patents by the China National Intellectual Property Administration.

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