

Synergistic Triple-Stimuli Responsive Ferrogels: From Microstructural Control to Multi-Modal Soft Actuation

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Polymeric hydrogels are soft, deformable, and highly hydrated materials that serve as versatile platforms for tissue engineering, soft robotics, and sensors. Their inherent biocompatibility makes them particularly suitable for biomedical devices. A defining characteristic of these "smart" systems is their ability to respond to external stimuli such as pH, light, temperature, and magnetic fields [1]. While magnetic actuation is prized for being fast, remote, reversible, and biologically safe, most current actuators are limited by single-stimulus responsiveness. This work addresses the need for multi-stimuli responsive hydrogels to achieve complex, reconfigurable deformation.

In this work, we report novel triple-stimuli responsive hydrogels (pH, temperature, and magnetic field) synthesized via a sequential fabrication approach. The matrix consists of a pH-responsive chitosan network coordinated by Ag ions, sequentially interpenetrated with agarose to impart thermal responsiveness. The integration of iron micro-particles confers precise magnetic field responsiveness. We conducted a thorough characterization of the hydrogels' physical and chemical properties, focusing on the interplay between microstructure, mechanical behavior, and swelling kinetics across a range of pH and temperature conditions.

Our results show that the swelling behavior strongly depends on pH, reaching a maximum under slightly acidic conditions, while contraction is observed at basic pH due to deprotonation of the chitosan chains. The mechanical properties vary significantly with composition and environmental triggers: while pure chitosan hydrogels exhibit limited stability in acidic media, the hybrid network remains stable and gel-like across a broader pH range. This synergy between the biopolymer blend and the magnetic phase enables controlled multi-stimuli actuation.

Finally, a proof-of-concept application was designed to demonstrate the material's potential as a soft robotic gripper. The system utilizes a magnetic field to achieve macro-positioning (claw-like configuration), while pH or temperature changes maintain the new configuration even after the magnetic field is switched off. This dual-action mechanism enables the magnetically guided transport and subsequent release of cargo, highlighting the potential of these triple-responsive ferrogels for advanced controlled manipulation and soft robotics.

References:

[1] Xinyue Liu, Ji Liu, Shaoting Lin, Xuanhe Zhao, *Materials Today*, 36 (2020) 102.

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