Robust and Versatile Physical Hydrogels Mediated by Metal Coordination

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In the past decades, many efforts have been devoted to improve hydrogels' mechanical properties by introducing an effective energy dissipation mechanism, such as double-network hydrogels. However, the tough hydrogels usually have a permanent network, leading to the poor processibility, self-healing properties. To solve these problems, chemically crosslinking points are supposed to be placed with the non-covalent interactions, such as ionic bonds, hydrogen bonds, hydrophobic associations, metal-ligand coordinations, which are dynamic and recoverable after being broken. Herein, we report meta-coordination mediated physical hydrogels with high toughness, self-recovery, self-healing, and shape memory properties. In addition, a stick-slip phenomenon is found during the tearing of these gels.

Gel synthesis

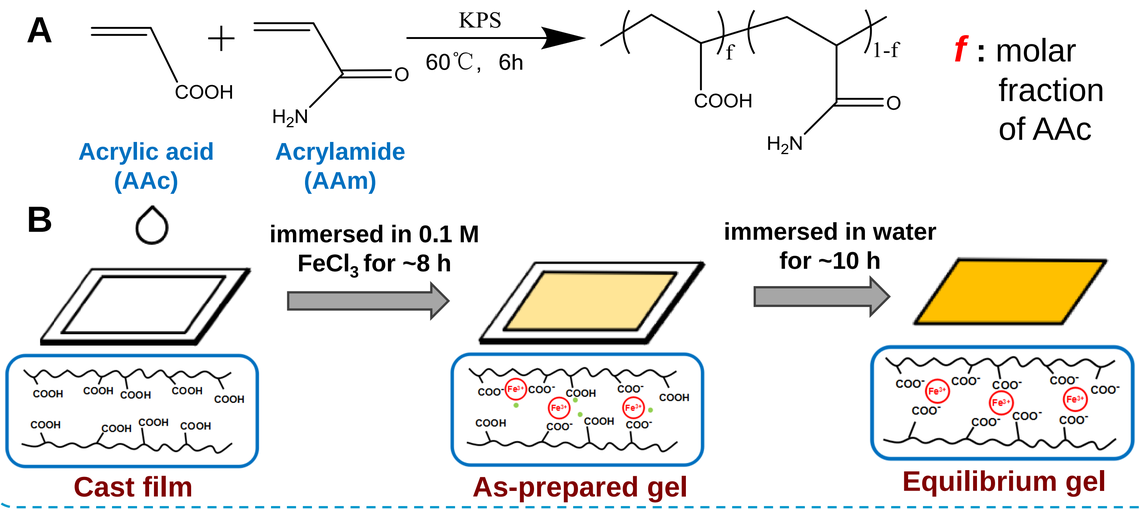


Figure 1. (A) Synthesis of the copolymer. (B) The scheme for the preparation of the gel films. (C) Water content of the as-prepared and equilibrated gels.

Self-recovery

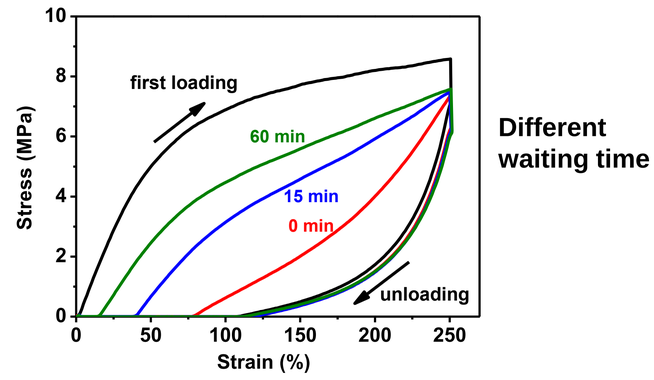
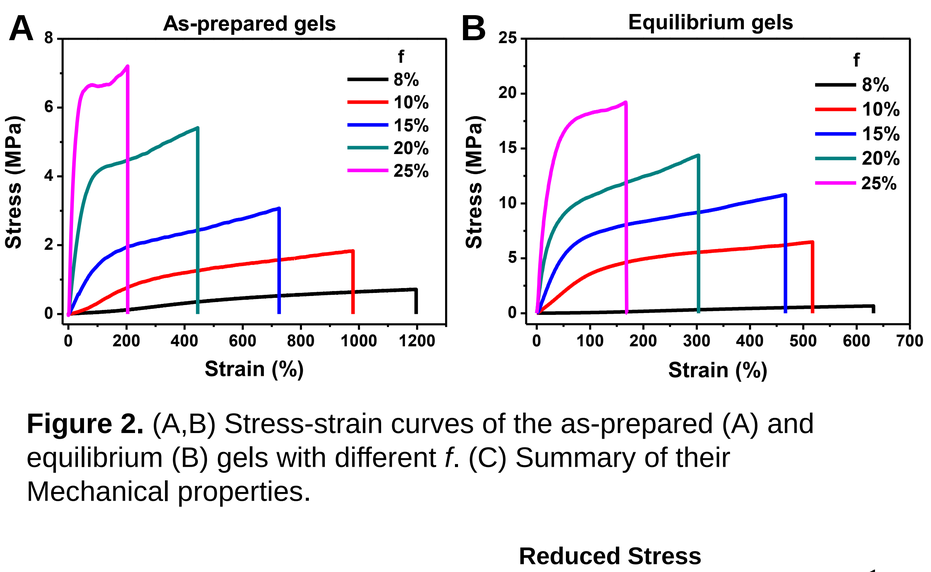


Figure 5. Cyclic tensile stress-strain curves of the equilibrated gel (*f* = 15%) after different waiting time.

Tensile tests



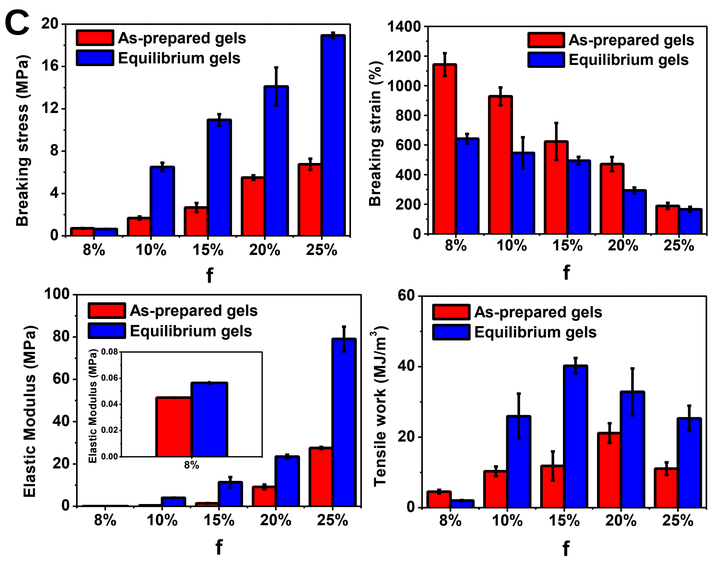
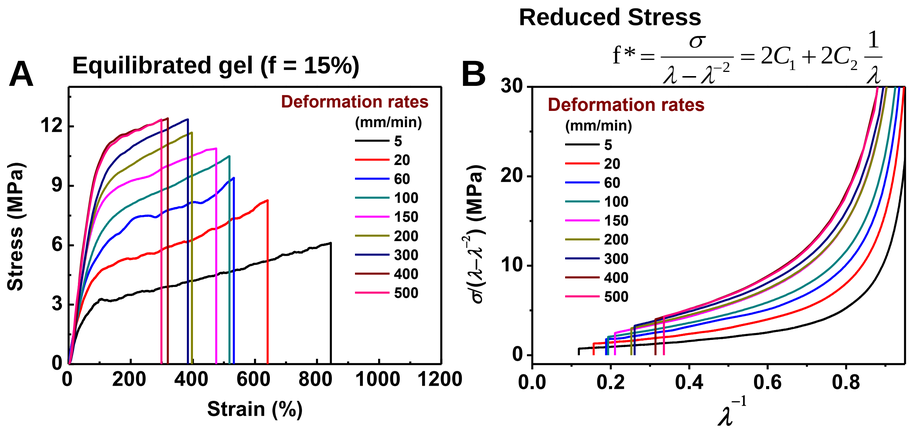


Figure 2. (A,B) Stress-strain curves of the as-prepared (A) and equilibrium (B) gels with different *f*. (C) Summary of their Mechanical properties.



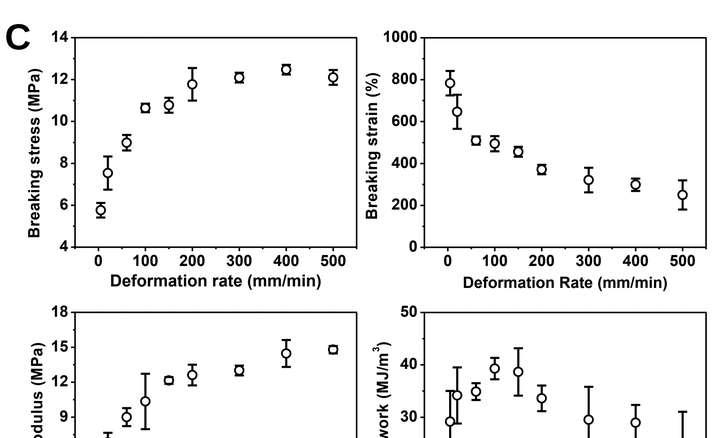


Figure 3. (A, B) Stress-strain curves of the equilibrium gels (*f* = 15%) at different initial deformation rates (A) and their reduced stress σ* as a function of λ^{-1} (B). (C) Summary of their mechanical parameters.

Self-healing

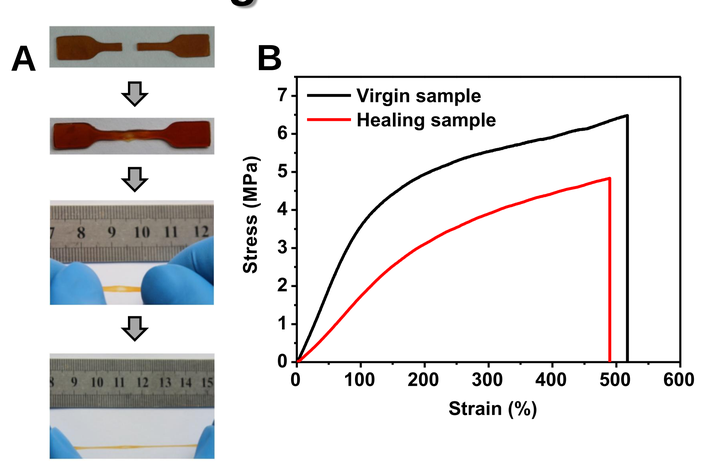


Figure 6. (A) Self-healing behaviour of the equilibrated gels (*f* = 10%) assisted by HCl and $FeCl_3$ solutions. (B) Stress-strain curves of the virgin and healing samples.

Shape memory

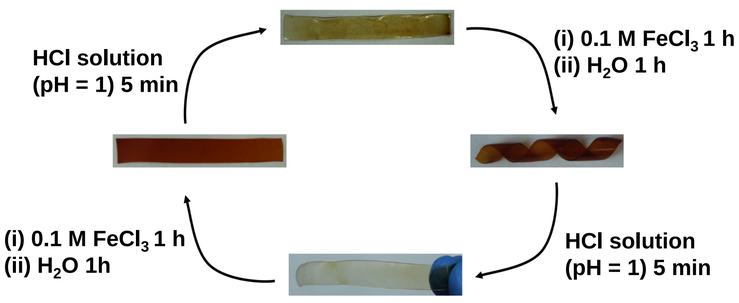


Figure 7. Shape memory behaviour of the equilibrated gels (f = 15%).

Tearing tests: stick-slip behavior

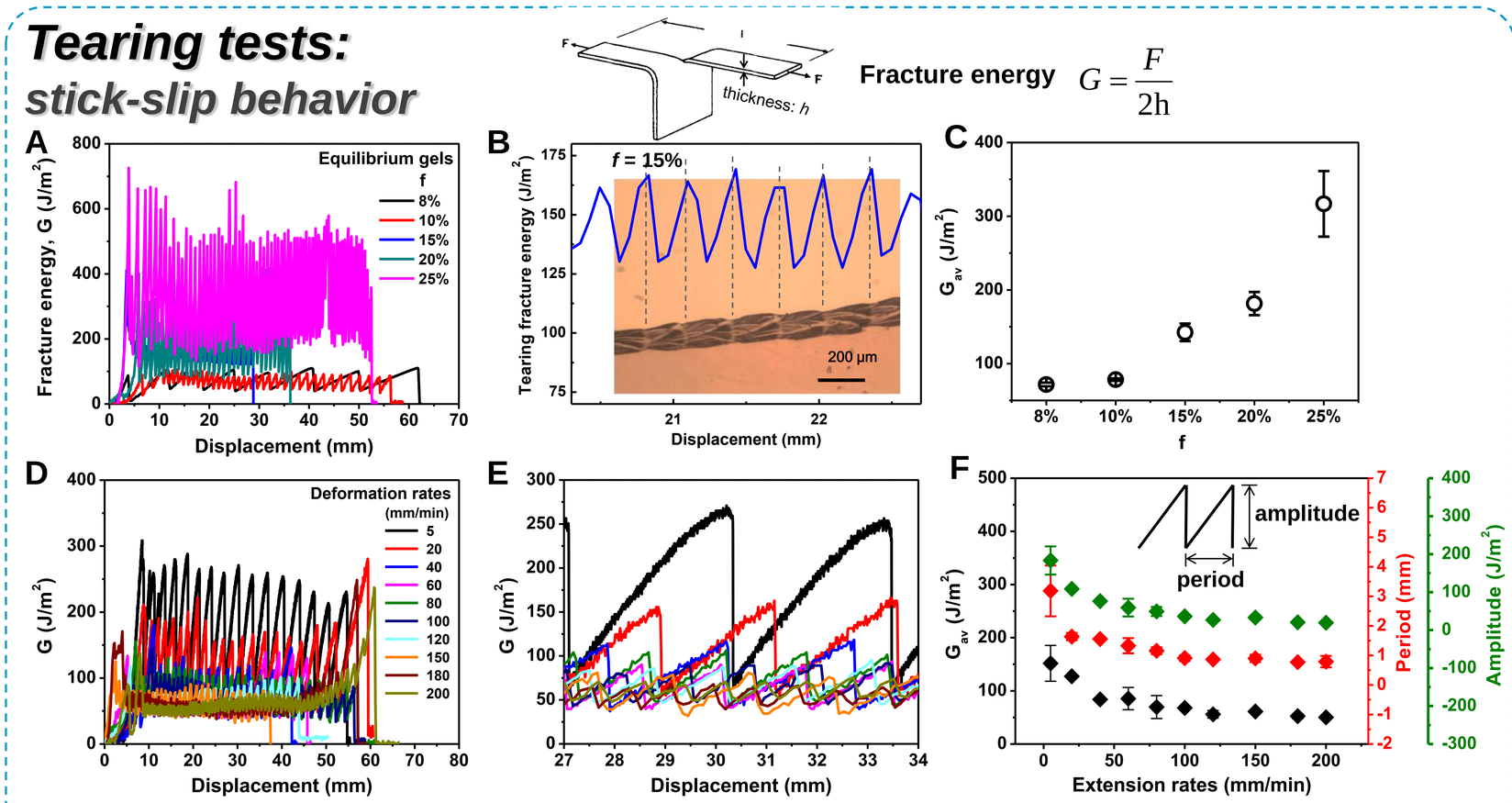


Figure 4. Tearing energy-displacements curves of the equilibrated gel films with different *f* (A-C) and extension rates of the gel with *f* = 10% (D-F). (C) The image in (B) is the fracture surface of the gel with *f* = 15%.

Conclusions

In summary, we have developed a new class of super tough physical hydrogels with strong metal-coordination. A stick-slip instability is found during the tearing. Owing to the reversible and dynamic noncovalent bonds, these gels show self-recovery, stimuli-triggered healing and shape memory behaviors.

Acknowledgement

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References

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