Supporting Information

for

Synergy of Hofmeister Effect and Ligand Crosslinking Enabled the Facile Fabrication of Super-strong, Pre-stretching-enhanced Gelatin-based Hydrogel

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Optical photos showing rapid curing and volume shrinkage

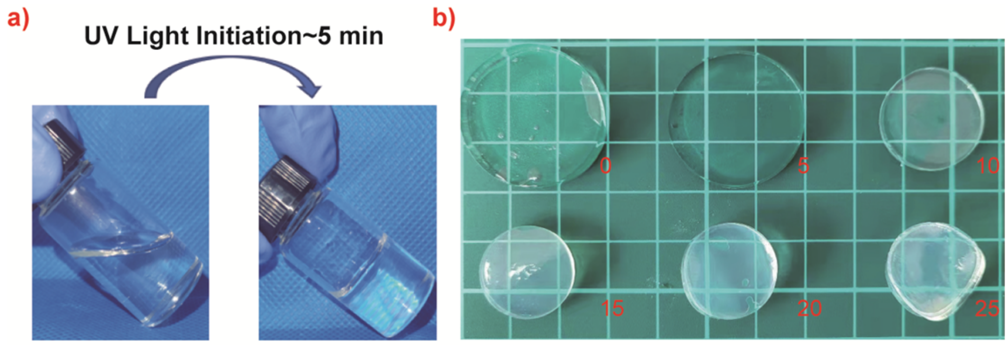

Figure. s1 (a)Hydrogels can be cured quickly in five minutes under UV light. (b) Optical photographs show the volume shrinkage of pure gelatin hydrogels treated with different concentrations of AS solutions (5-25 wt%).

Effect of AAc/AAm ratio on the mechanical properties of hydrogels

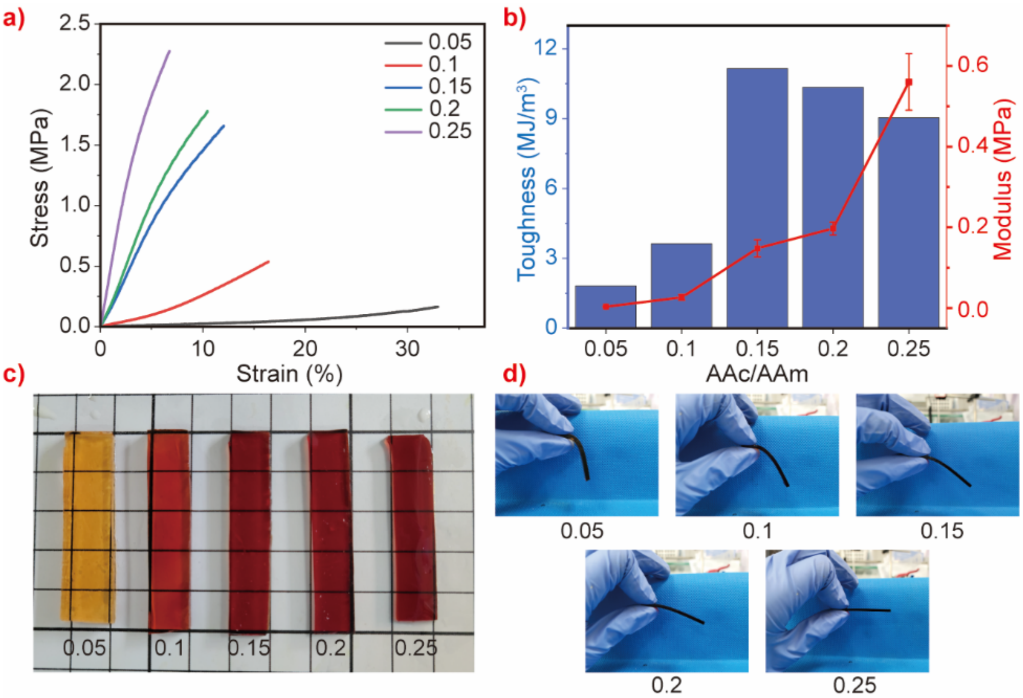

Figure. s2 (a) Stress-strain curves and(b) Toughness vs. elastic modulus for DN/Fe hydrogels with different AAc/AAm ratios. (c) Optical photographs showing the color and (d) softness of DN/Fe hydrogels with different AAc/AAm ratios.

As shown in Figure s2(a)(b), the tensile strength and elastic modulus of DN/Fe hydrogels increased gradually with the increase of the AAc/AAm ratio, and the tensile strength increased significantly at the ratio of 0.15. This is because, with the increase of the AAc/AAm ratio, Fe^{3+}, which can cross-link with the carboxyl group on acrylic acid, has more cross-linking sites, which makes the cross-linking density increase and the hydrogel change from "soft and tough" to "hard and brittle". The maximum value of toughness of DN/Fe hydrogel is achieved at AAc/AAm=0.15. The maximum toughness of DN/Fe hydrogel was obtained at AAc/AAm=0.15, and we chose the DN/Fe hydrogel with AAc/AAm=0.2 to continue the experiment. Figure s2(c)(d) shows the optical photographs of DN/Fe hydrogels with different AAc/AAm ratios, and it can be seen that the color of DN/Fe hydrogels gradually deepens and the hardness increases with the increase of AAc/AAm ratio.

DN/Fe-AS hydrogel treated with saturated ammonium sulfate solution

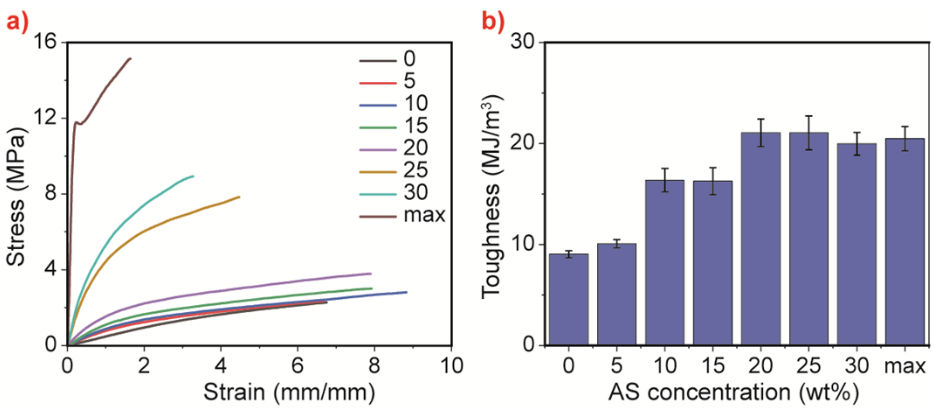

Figure. s3 (a) Stress-strain curves of DN/Fe-AS hydrogels treated in different concentrations of AS solutions (0-saturated wt%) with (b) toughness.

Figure s3 shows the stress-strain curves and toughness of DN/Fe-AS hydrogels treated in different AS solutions. It can be seen that the DN/Fe-AS hydrogels treated in saturated AS solutions have a large difference in mechanical properties from those treated at other concentrations. This is because the highly convoluted gelatin molecular chains inside the hydrogel, combined with the large external osmotic pressure lead to a large water loss in the hydrogel, the DN/Fe-ASmax hydrogel resembled a partially anhydrous polymer. gels.

Effect of bulk water treatment on DN/Fe-AS hydrogels

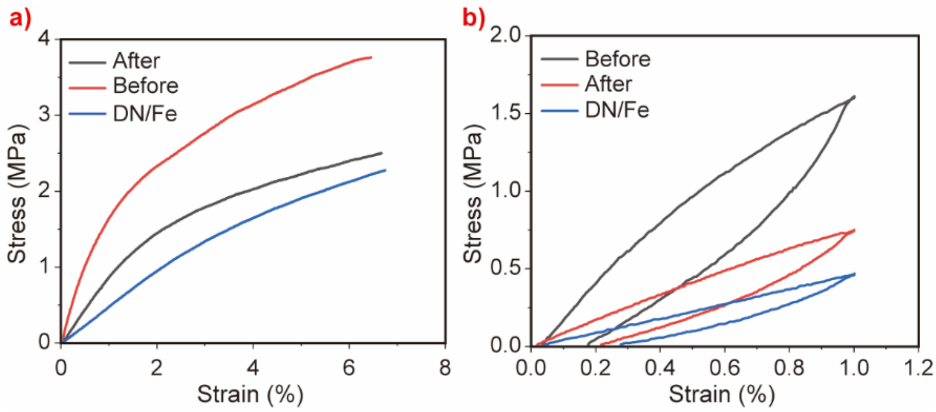

Figure. s4 (a) Stress-strain curve and (b) Load-unload test curve of DN/Fe, DN/Fe-AS, and DN/Fe-AS hydrogel after treatment in a large amount of ultrapure water.

As shown in Figure s4 (a) (b), the DN/Fe-AS hydrogel showed a significant decrease in tensile strength, and the area enclosed by the loading-unloading curve was reduced after treatment with a large amount of ultrapure water, but it was still higher than that of the DN/Fe hydrogel. This indicates that the entanglement of gelatin molecular chains disappeared substantially because of the removal of ammonium sulfate, but some of the cross-linked structure was still retained, making the mechanical properties of the hydrogels still superior to those of DN/Fe hydrogels.

Rheological properties of DN, DN/Fe, and DN/Fe-AS hydrogels

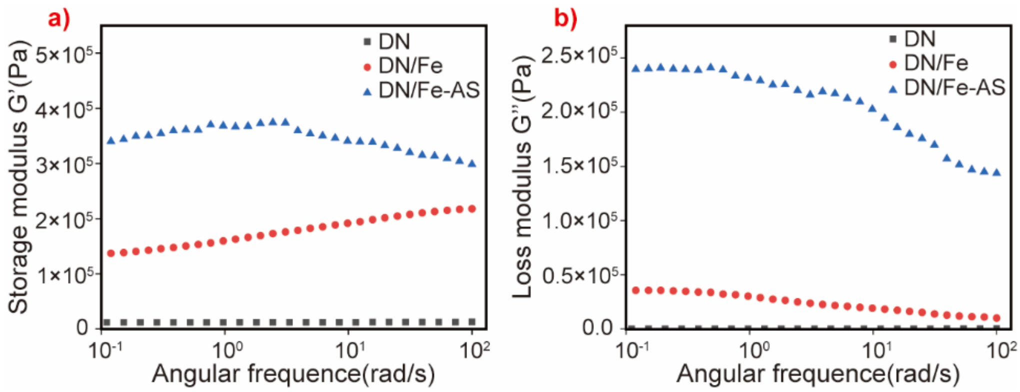

Figure. s5 Frequency sensitivity of (a) elastic modulus and (b) loss modulus of DN, DN/Fe, and DN/Fe-AS hydrogels.

The dynamic mechanical properties of DN, DN/Fe, and DN/Fe-AS hydrogels were then analyzed by rheological tests to evaluate the adhesive enhancement effect of aggregated gelatin chains. As shown in Figure s5(a)(b), the elastic modulus and loss modulus of DN/Fe-AS hydrogels were significantly higher than those of DN and DN/Fe hydrogels and were in the same order of magnitude as those calculated by stress-strain curves. the G' of both DN/Fe and DN/Fe-AS hydrogels exhibited a clear frequency dependence from low to high frequencies, indicating that the gels were viscoelasticity. The higher modulus indicates the formation of dense cross-linked regions with superior energy dissipation ability.