Supporting Information

Highly stretchable and tough pH-sensitive hydrogels with reversible swelling and recoverable deformation

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Tensile Properties of Agar/P(AM-co-AA) IPN hydrogels

Figure S4 demonstrated the tensile properties of the Agar/P(AM-co-AA) IPN hydrogels with different concentrations of agar monomers. Under the same crosslinker dosage, P(AM-co-AA) hydrogels showed very weak properties, whereas increasing agar content tremendously enhanced the mechanical properties of Agar/P(AM-co-AA) hydrogels. In the selected range of agar monomer concentrations, the fracture stress, the fracture strain and the work showed an increasing trend due to the increased agar helix bundles crosslinking density. This result indicated that incorporation of agar could remarkably improve the mechanical strength of the hydrogels.

Subsequently, we investigated the effect of PEGDA6K concentration on the tensile properties of the Agar/P(AM-co-AA) hydrogels as shown in Figure S5. It was obvious that introducing an appropriate amount of PEGDA6K would sharply increase the tensile strength. The fracture strain and work showed a decreasing trend with the PEGDA6K increasing and the highest fracture stress (280KPa) was obtained when the concentration of PEGDA6K was 0.07mol%. It had been found that the crosslinking density was critical for the mechanical strength of hydrogels [1]. When the covalent crosslinking density was too high, each individual chain between neighboring crosslinking points was short. As a result, highly crosslinked hydrogels were tend to be stiff (high elastic modulus), but they were generally quite brittle and had low stain at break.

Reference

1 B. H. Cipriano, S. J. Banik, R. Sharma, D. Rumore, W. Hwang, R. M. Briber and S. R. Raghavan, Macromolecules, 2014, 47, 4445-4452.

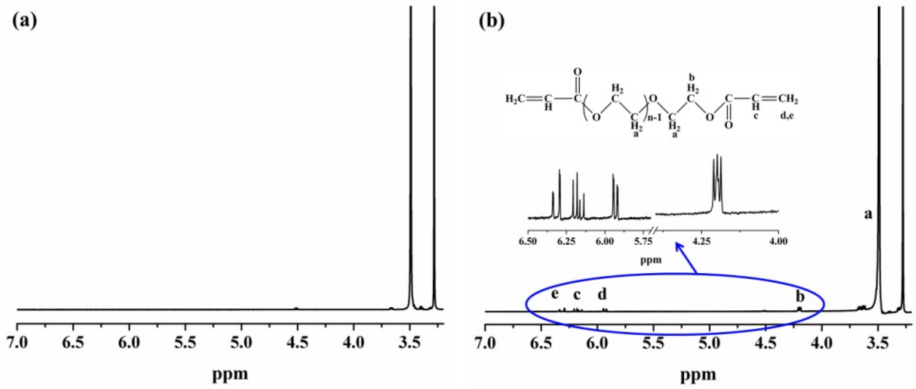

Figure S1 ^{1}H NMR spectra of PEG6K (a) and PEGDA6K (b).

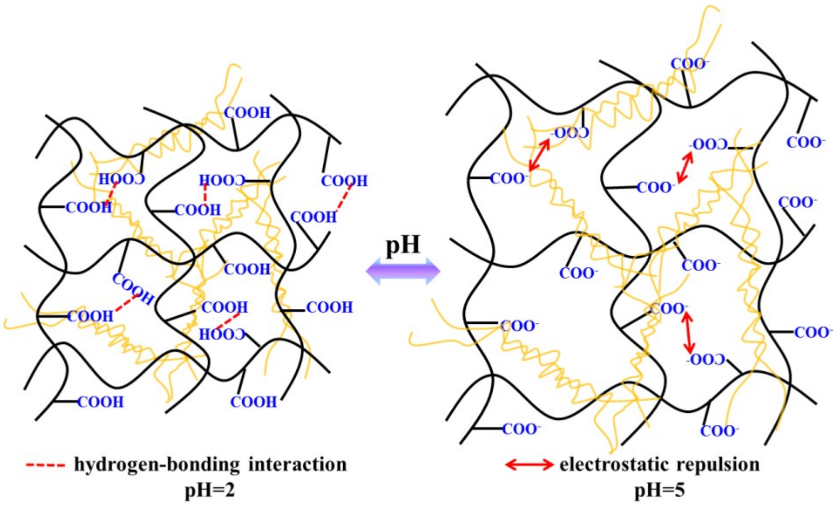

Figure S2 Schematic diagram of pH-sensitive Agar/P(AM-co-AA) IPN hydrogels.

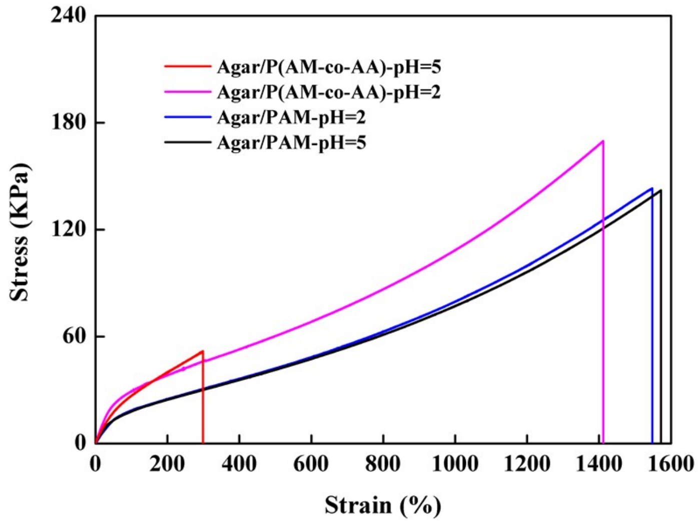

Figure S3 The effect of pH on the tensile properties of the Agar/P(AM-co-AA) and Agar/PAM hydrogels.

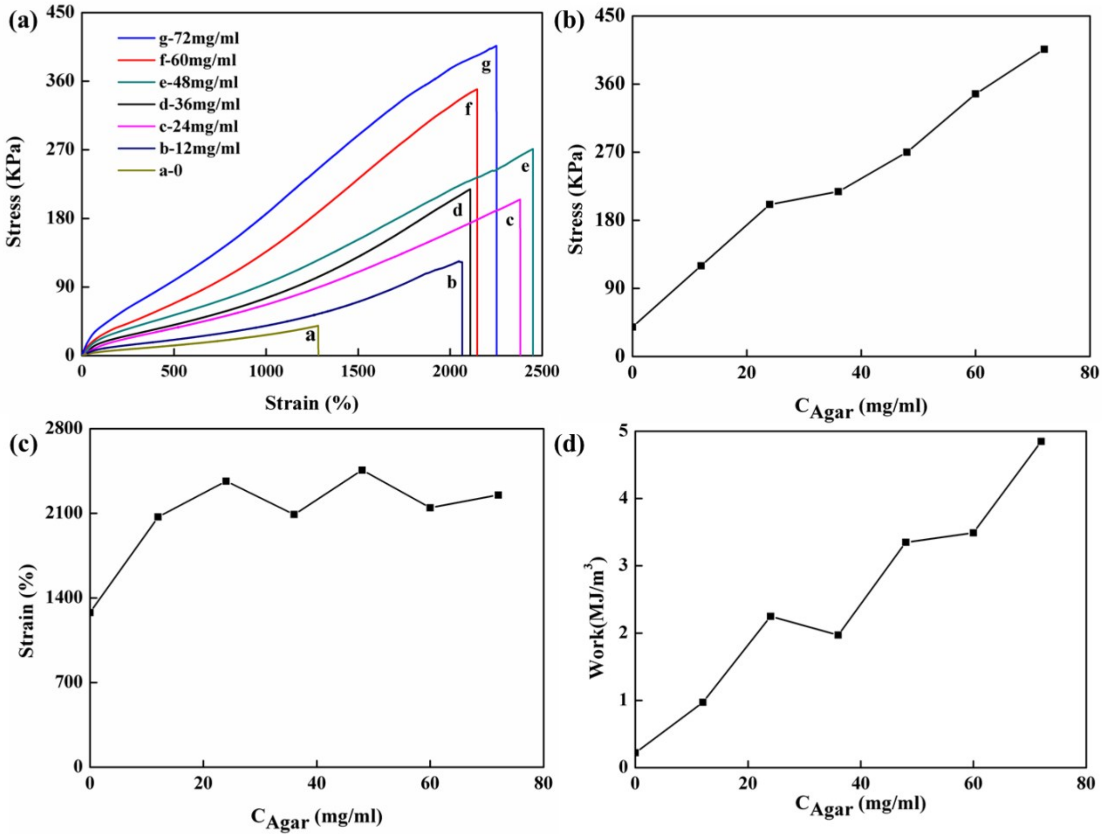

Figure S4 The effect of Agar concentration on the tensile properties of the Agar/P(AM-co-AA) IPN hydrogels.

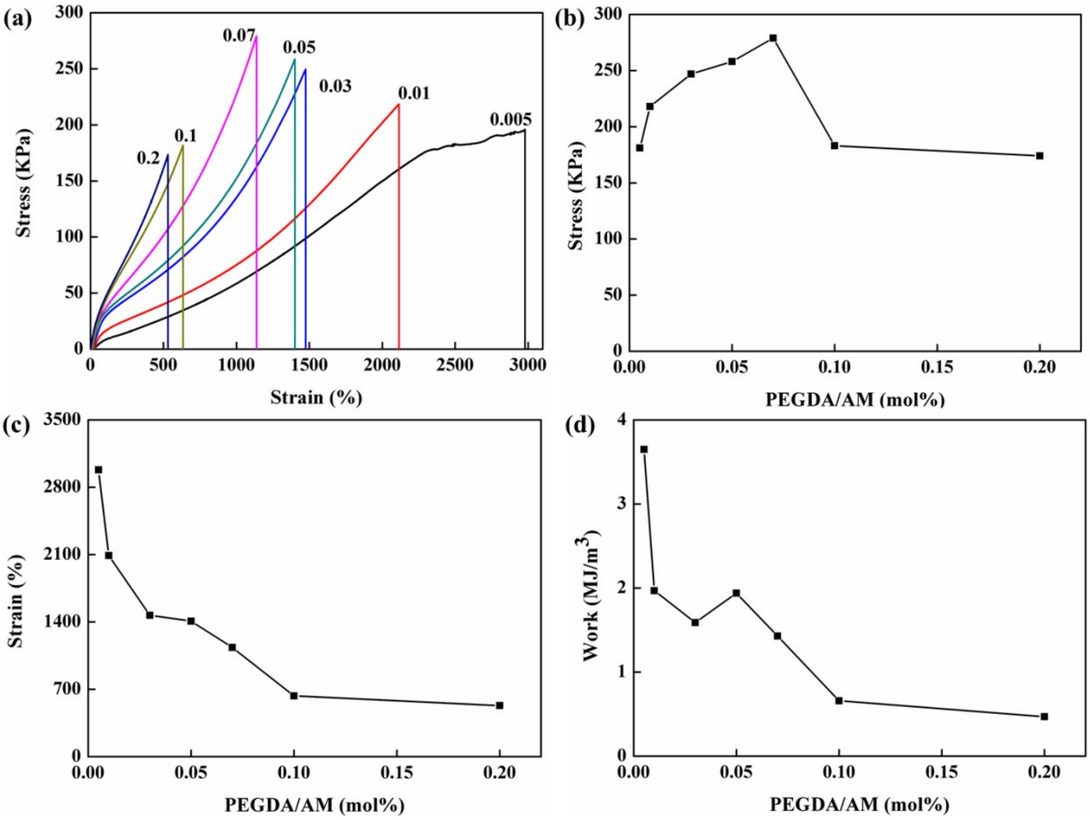

Figure S5 The effect of PEGDA6K concentration on the tensile properties of the Agar/P(AM-co-AA) IPN hydrogels.

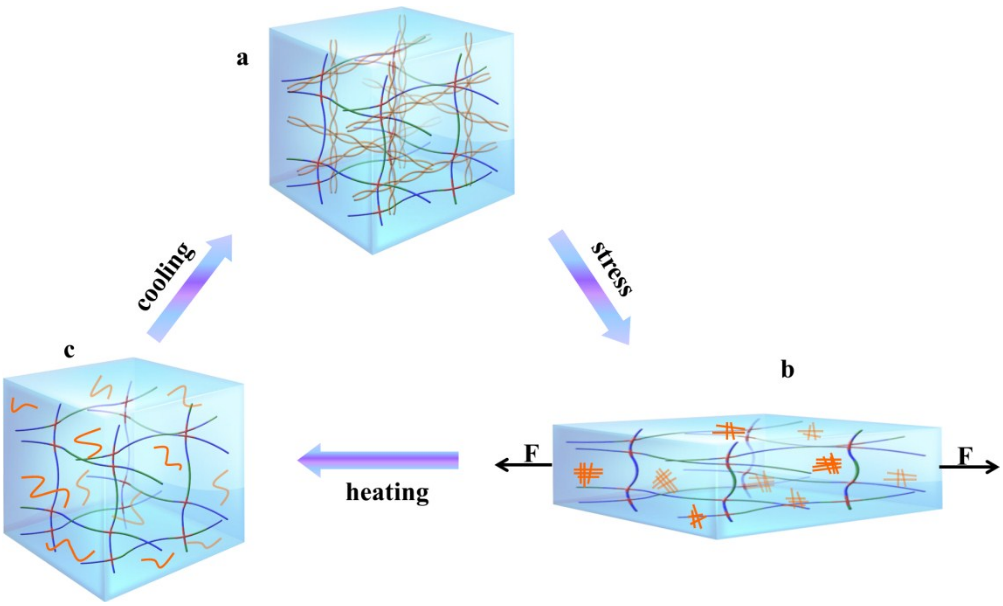

Figure S6 Schematic illustration of Agar/P(AM-co-AA) IPN hydrogel recovery.

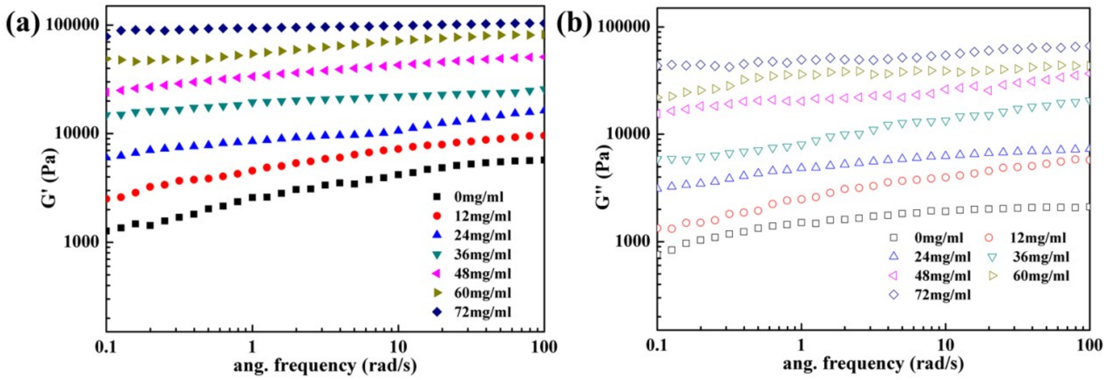

Figure S7 Rheological properties as a function of frequency at strain of 0.5% for the hydrogels with different agar contents at 25ºC: (a) storage modulus G'; (b) loss modulus G''.