Supplementary Information

Tough and Super Resilient Hydrogels Synthesized by Using Peroxidized Polymer Chains as Polyfunctional Initiating and Cross-linking Centers

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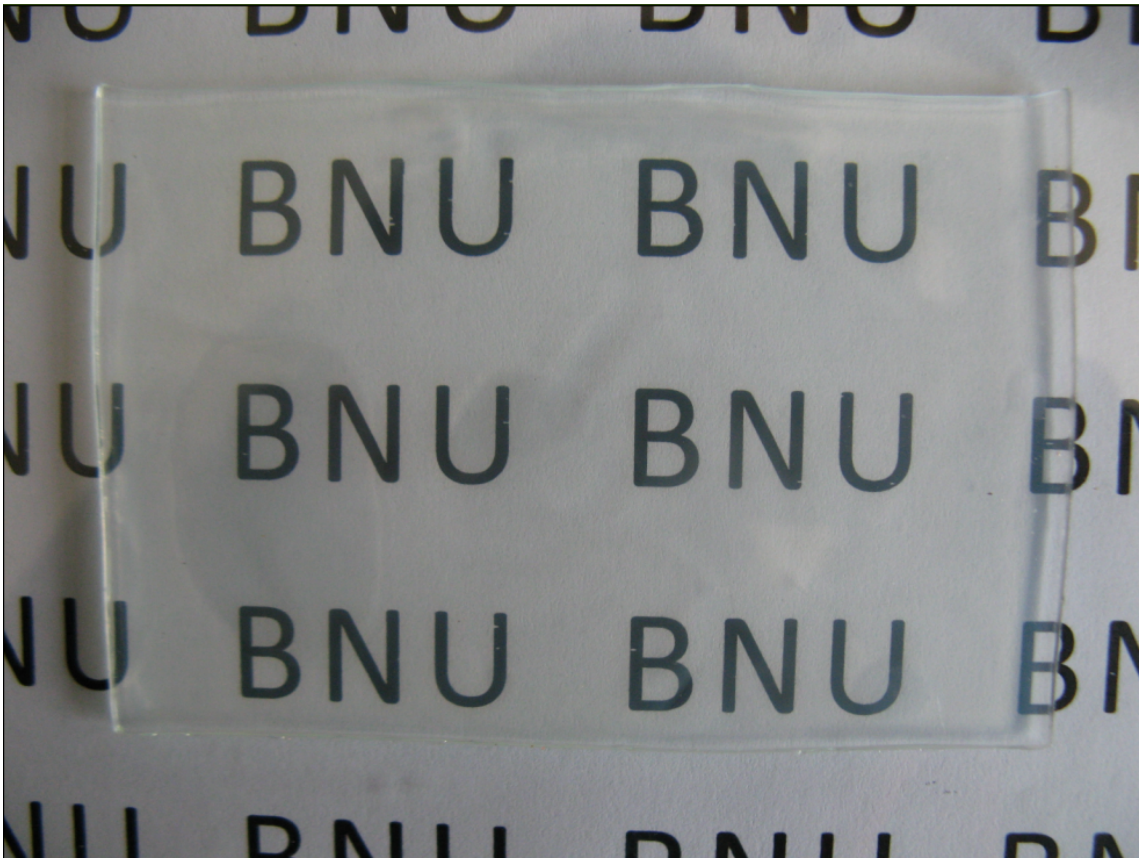

Fig. S1 Photograph showing the high transparency of the gel.

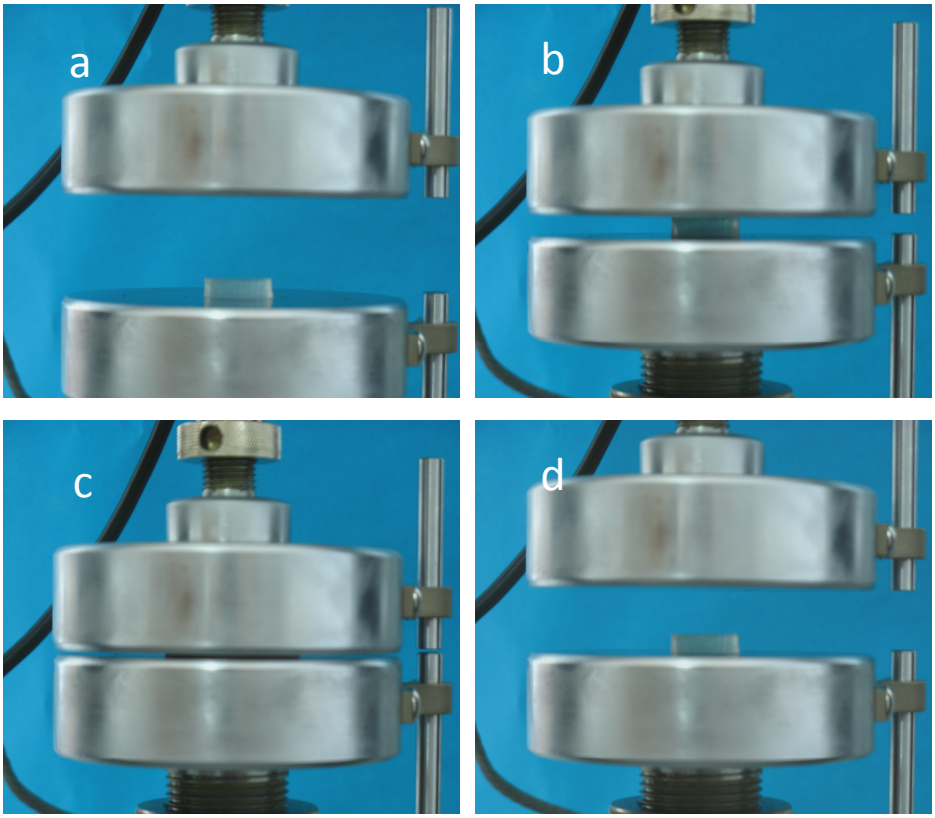

Fig. S2 Photographs showing a gel specimen (without MBA) during the compression test. (a): Before compression; (b): at the beginning of the compression; (c): being compressed to a high compressive strain; (d): after the test. (C_M =5.0 M; water content of the gel: 80 *wt.*%; crosshead speed: 800% min^{-1})

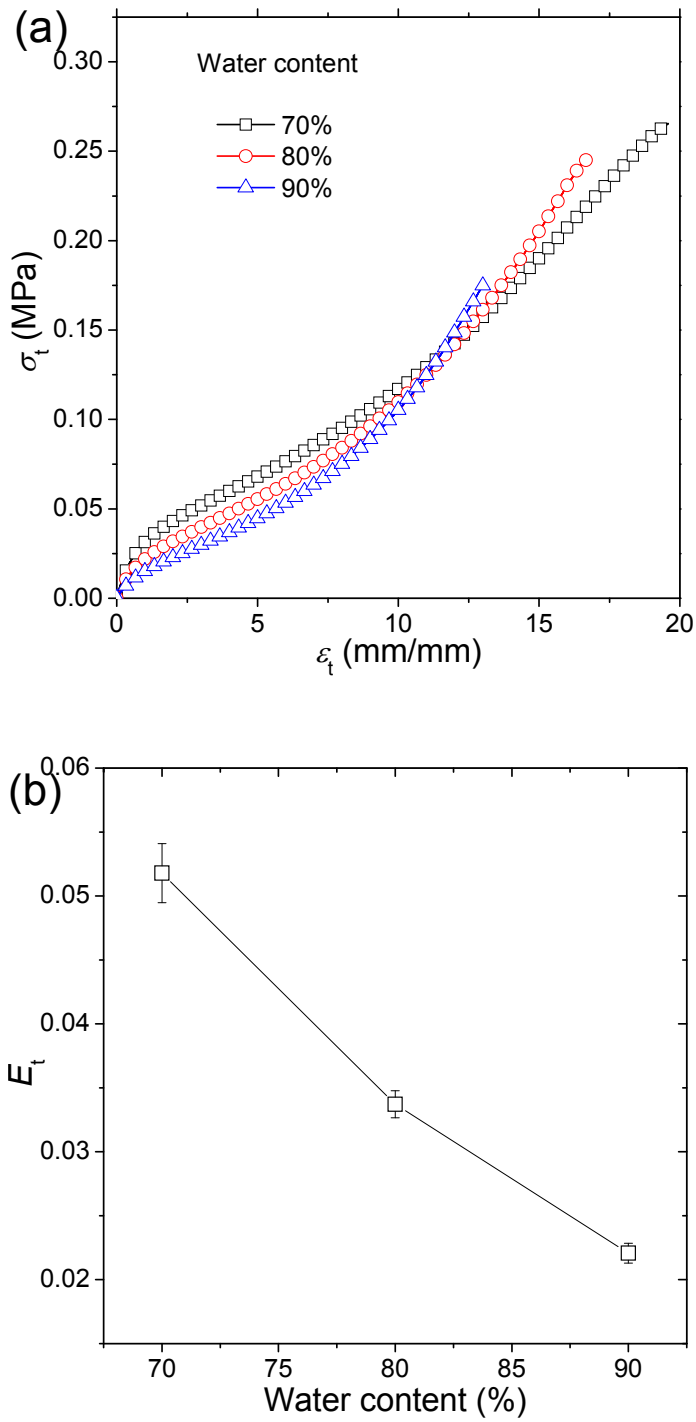

Fig. S3. The tensile mechanical properties of the hydrogel samples with different water contents. The hydrogel was fabricated with 3 M AAm and 0.05% MBA. (a) The stress-strain (σ_t-ε_t) curves of the hydrogels; and (b) the moduli of the hydrogels.

The tensile strength, fracture stain and the modulus of the gel decrease with increasing water content.

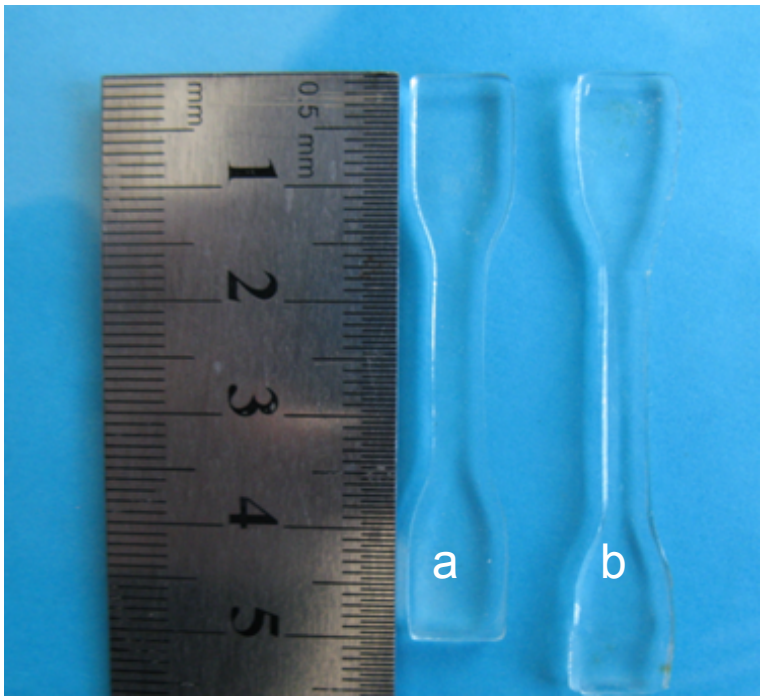

Fig. S4 Photo showing the appearance of the gel specimens without MBA before (a) and after (b) the cyclic tensile test to a ε_{max} of 1600% for 10 cycles. The monomer concentration was 3.0 M, and the water content of the samples was about 90 *wt*.%.

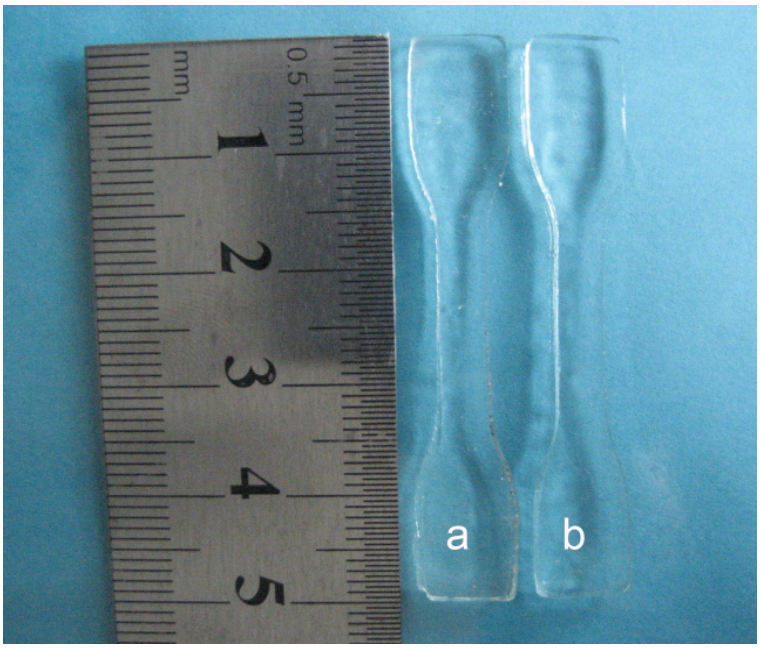

Fig. S5 Photo showing the appearance of the gel specimens with 0.05 *wt*.% MBA before (a) and after (b) the cyclic tensile test to a ε_{max} of 800% for 10 cycles. The monomer concentration was 3.0 M, and the water content of the samples was about 90 *wt*.%.

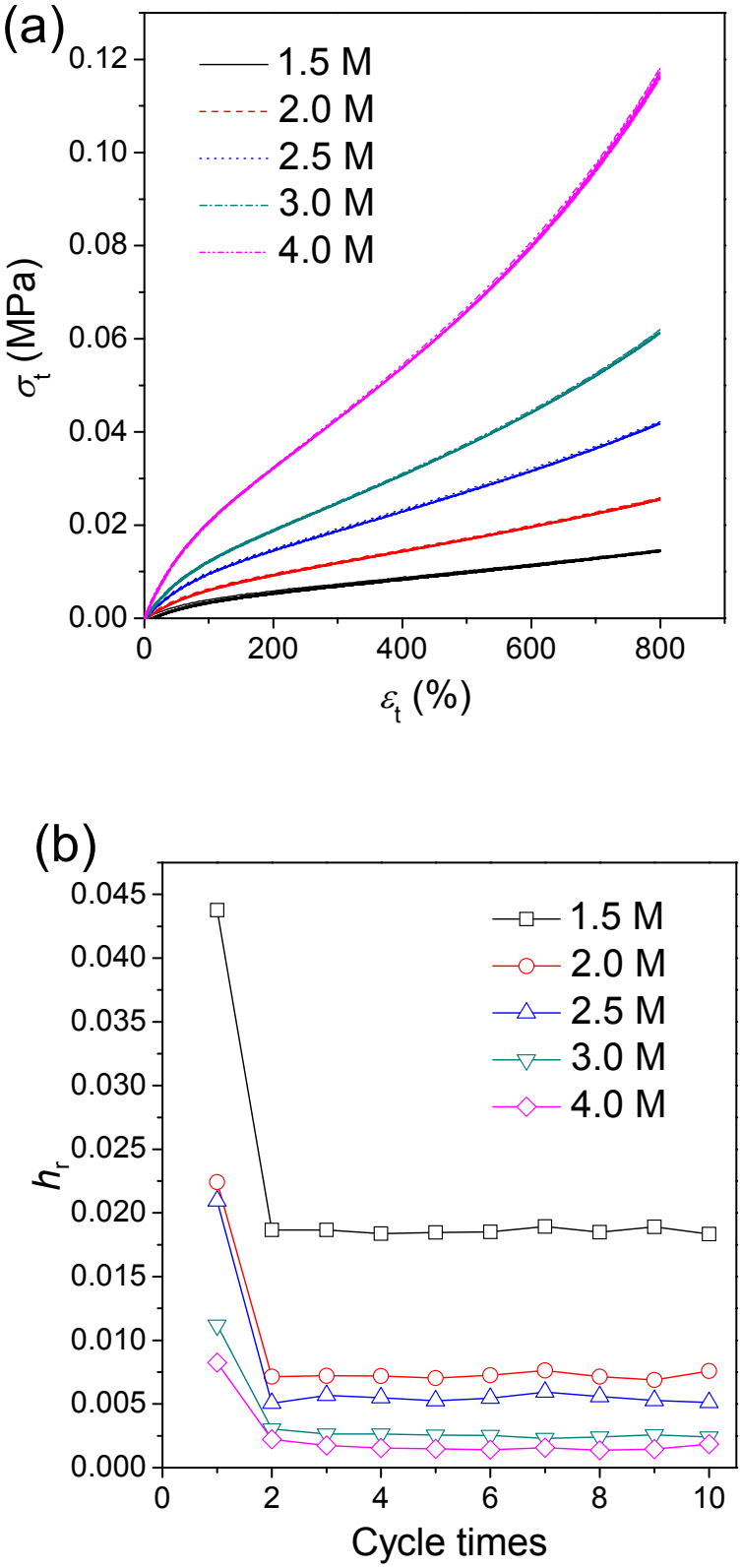

Fig. S6 (a): The cyclic tensile loading-unloading curves for the hydrogels synthesized with different C_M and with MBA to the strain of 800% for 10 runs; (b): the corresponding hysteresis ratio (h_r) of each run cycle.

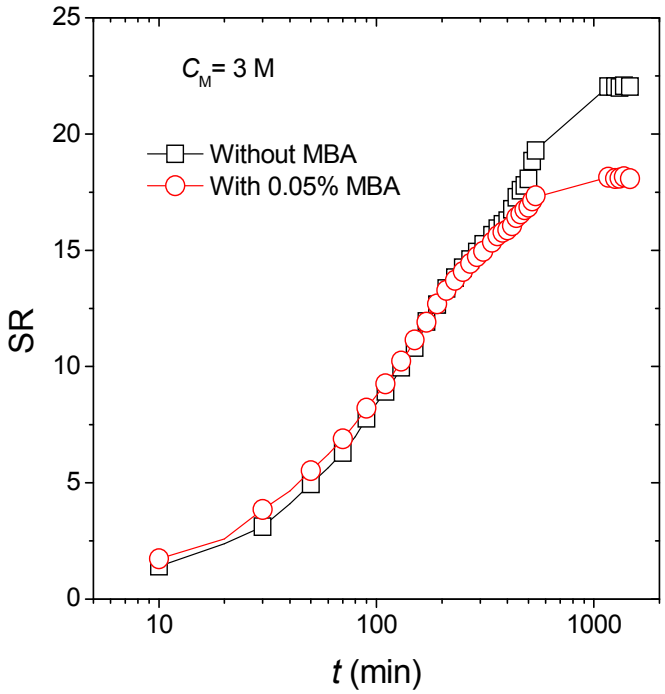

Fig. S7. Swelling kinetics of the hydrogels fabricated without and with MBA.

The equilibrium swelling ratios of the hydrogels have been measured. Fig. S7 shows the swelling of the hydrogels fabricated with C_M= 3 M, and with or without MBA. There are some differences between the MBA-containing and MBA-free hydrogels, the MBA-containing hydrogels usually show lower equilibrium swelling ratios. However, the difference is not very significant, since the content of crosslinker and hence the increase of crosslinking density is very small. As shown in Fig. S7, the gel with MBA exhibited an equilibrium swelling ratio of 18.8 and the gel without MBA 22.