Supporting Information

Title Water-retaining, tough and self-healing hydrogels and their uses as fire-resistant materials

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Figure S1. Hydrogels prepared by adding LiCl into the prepolymerization solution directly.

As **Figure S1** shows, solution stratification is obviously seen when C_{LiCl} is larger than 2 mol/L if adding LiCl into the prepolymerization solution directly. Without the constraints of the P(AAm-co-SMA) network, the Li-alginate chains will precipitate at the bottom owing to the reducing of hydrophilic in high C_{LiCl} solution. Thus, the Li-alginate/P(AAm-co-SMA) should be prepared by synthesis the P(AAm-co-SMA) network previously to obtain a homogeneous IPN gel network.



Figure S2. Micelle size in LiCl solution with different concentrations.



Figure S3. Scanning electron microscopy pictures of Li-alginate/P(AAm-co-SMA) hydrogels with C_{LiCl} of 2 mol/L (a) and 4 mol/L (b).

The scanning electron microscopy (SEM) was performed using Hitachi S3400N scanning electron microscopy. The gel specimens were prepared by freeze-drying and then cryogenically fractured in liquid nitrogen. The fractured surface was coated with a thin layer of nano-gold and then applied to SEM observation. Figure S3 shows the SEM pictures of Li-alginate/P(AAmco-SMA) (2) (**Figure S3 a**) and Li-alginate/P(AAm-co-SMA) (4) (**Figure S3 b**) hydrogels. The gel network changes from homogeneous to porous structure with different pore size. This is probably due to the phase separation of Lialginate and P(AAm-co-SMA) networks. Because the LiCl has a significant effect on the hydrophilicity of alginate polyelectrolyte. So, the higher C_{LiCl} (4 mol/L) in hydrogels causes greater difference in hydrophilicity between Lialginate and P(AAm-co-SMA) networks. As a result, the morphology of hydrogels networks changes along with phase separation.



Figure S4. Force-displacement curves of hydrogel specimens. The unnotched and notched specimens as depicted are applied to stretch to obtain the critical distance when the notch begins to expand. Then fracture energy is obtained by the area with the horizontal lines divided by the cross-sectional area of gel specimens.

The fracture energy of hydrogels was tested and calculated as follows. In brief, two specimens (a notched and an unnotched) with the same width a_0 and thickness b_0 were prepared. Then the unnotched and notched specimens were stretched to obtain the force-displacement curves. Critical displacement was also been recorded from the force-displacement of notched specimen before the notch began to expand (the red dot in **Figure 3S**). The area beneath the force-displacement of unnotched specimen gave the work (W_c) done by force before the sample ruptured. Finally, to quantify the crack resistance property, the fracture energy was calculated according to **Equation S1** as below:

$$W_{\rm c}/(a_0 \times b_0)$$
 (S1)