Supporting Materials for:

## Phase Transition in Amphiphilic Poly(N-isopropylacrylamide): Controlled Gelation

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**Figure S1.** (a) FT-IR characterization of p(NIPAM-co-BA). (b) <sup>1</sup>H NMR spectrum (400M, CDCl<sub>3</sub>) of p(NIPAM-co-BA).



**Figure S2.** GPC traces of p(NIPAM-co-BA) synthesized with varying the feeding ratio of comonomers and chain transfer agent (CTA). (a) Different CTA feeding ratios to monomers at a fixed BA/NIPAM ratio of 0.1. (b-d) Different BA feeding ratios at a fixed CTA/NIPAM ratio.



**Figure S3.** Temperature dependence of the light scattering intensity (a) and hydrodynamic diameters (b) for p(NIPAM-co-BA) solutions with different molecular weights and BA feeding ratios. Measurements were conducted with 5 mg mL<sup>-1</sup> polymer solutions and a temperature increment of 1 °C.



**Figure S4.** Phase diagram of aqueous p(NIPAM-co-BA) solutions with different molecular weights and BA feeding ratios. TS: transparent solution phase; OS: opaque solution phase; TG: transparent gel phase; OG: opaque gel phase; DG: dehydrated gel phase.



**Figure S5.** Summary of the temperature ramp curves of 20 w/w% aqueous p(NIPAM-co-BA) solutions at the regions with  $G' \ge G''$ . Measurements were performed at a constant strain of 1% and frequency of 6.3 rad s<sup>-1</sup>.



**Figure S6.** Frequency sweep of 20 w/w% aqueous  $P_{10}(33K)$  solution at the gel region determined by the oscillatory temperature ramp. Tan  $\delta$ =1 was used as the boundary to distinguish different rheological behaviors, the gel was predominantly elastic at tan  $\delta$  <1 regime, and predominantly viscous at tan  $\delta$  >1 regime. Measurements were performed at a constant strain of 0.5%.



**Figure S7.** Rheological reversibility of p(NIPAM-BA) between different phase states. (a) 5 °C (transparent solution) and 25 °C (opaque gel). (b) 5 °C (transparent solution) and 32 °C (dehydrated gel). The aqueous polymer solution was made of  $P_{10}(33K)$  at the concentration of 20 w/w%.



**Figure S8.** Rheological hysteresis curves of 20 w/w% aqueous  $P_{10}(33K)$  solution in a heating and cooling cycle between different phase states. (a) 10 °C (transparent solution) and 25 °C (opaque gel). (b) 10 °C (transparent solution) and 40 °C (dehydrated gel).



**Figure S9.** Long-term rheological stability of p(NIPAM-co-BA) in an oscillatory time sweep at different phase states. Blue square: 25 °C (opaque gel); olive square: 32 °C (dehydrated gel). The polymer solution was made of  $P_{10}(33K)$  at the concentration of 20 w/w%, and the measurements were performed at a constant strain of 1% and frequency of 6.3 rad s<sup>-1</sup>.



**Figure S10.** Photographs showing the self-healing process of P(NIPAM-co-BA) hydrogels with two different Mw at constant temperature (18 °C). The colorless gel was made of 30 w/w% aqueous  $P_{10}(33K)$  solution, and the red gel was made of 30 w/w% aqueous  $P_{10}(29K)$  solution stained with rhodamine B to facilitate visualization.