

Supporting Information

for

Spin-Coating-Assisted Fabrication of Ultrathin Physical Hydrogel Films with High Toughness and Fast Response

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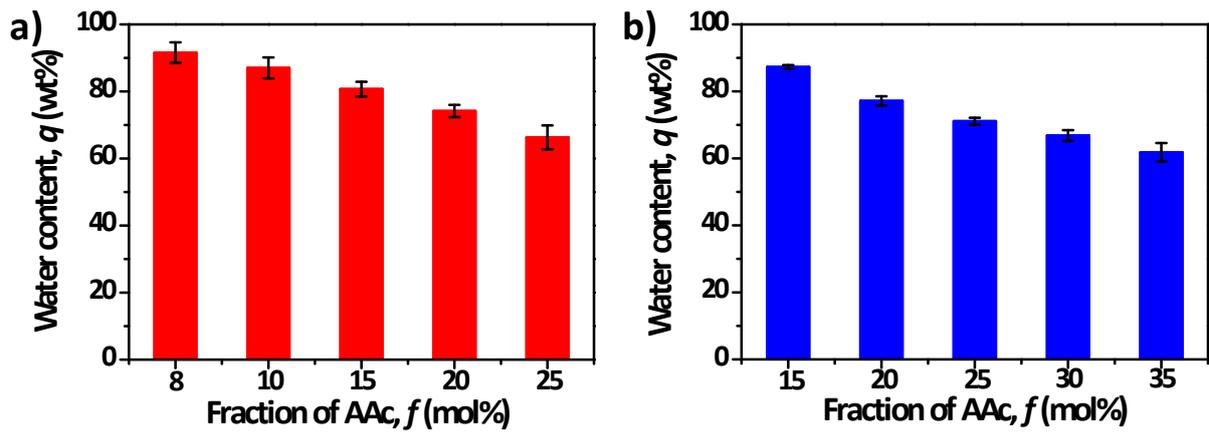


Figure S1. Water content of the P(AAc-co-AAm) gel film (a) and P(AAc-co-NIPAm) gel film (b) with different copolymer composition.

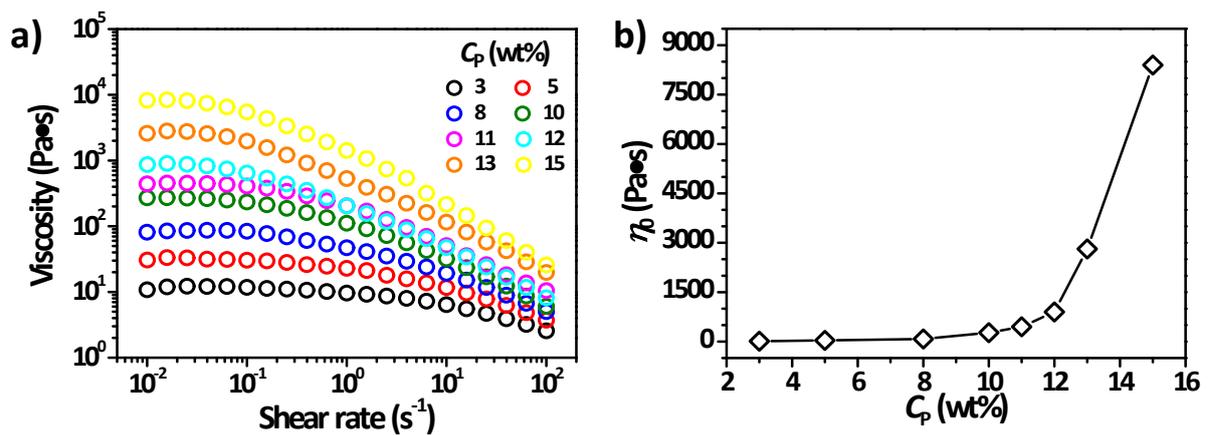


Figure S2. (a) Steady flow test of the P(AAc-co-AAm) solutions ($f = 15$ mol%) with different concentration. (b) Zero shear rate viscosity of the solution, η_0 , as a function of polymer concentration, C_p .

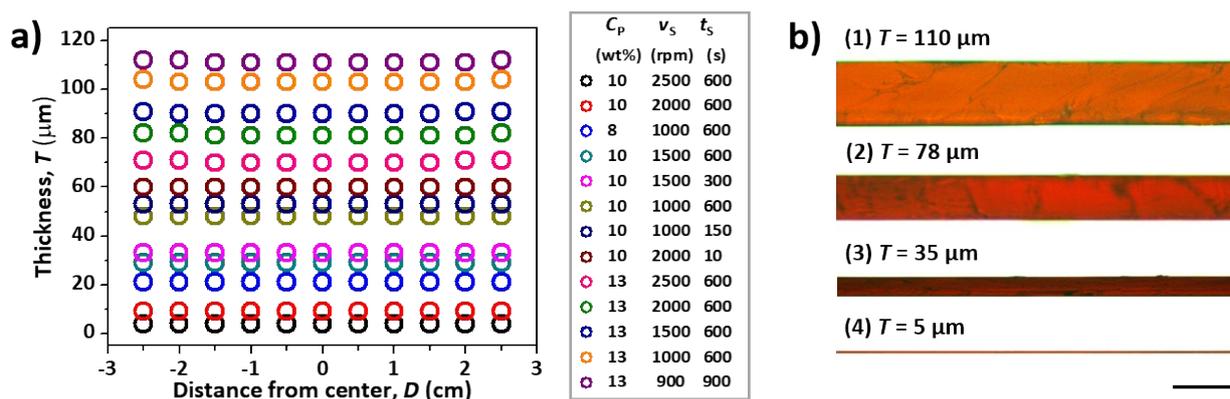


Figure S3. (a) Thickness profile of uniform P(AAc-co-AAm) hydrogel films prepared with different spin speed v_s , spin time t_s , and polymer concentration C_p ($f = 15$ mol%). (b) Representative micrographs of the cross-section of gel films with different thickness. (1) $C_p = 13$ wt%, $v_s = 900$ rpm, $t_s = 900$ s; (2) $C_p = 13$ wt%, $v_s = 2000$ rpm, $t_s = 600$ s; (3) $C_p = 10$ wt%, $v_s = 1500$ rpm, $t_s = 300$ s; (4) $C_p = 10$ wt%, $v_s = 2500$ rpm, $t_s = 600$ s. Scale bar: 100 μm .

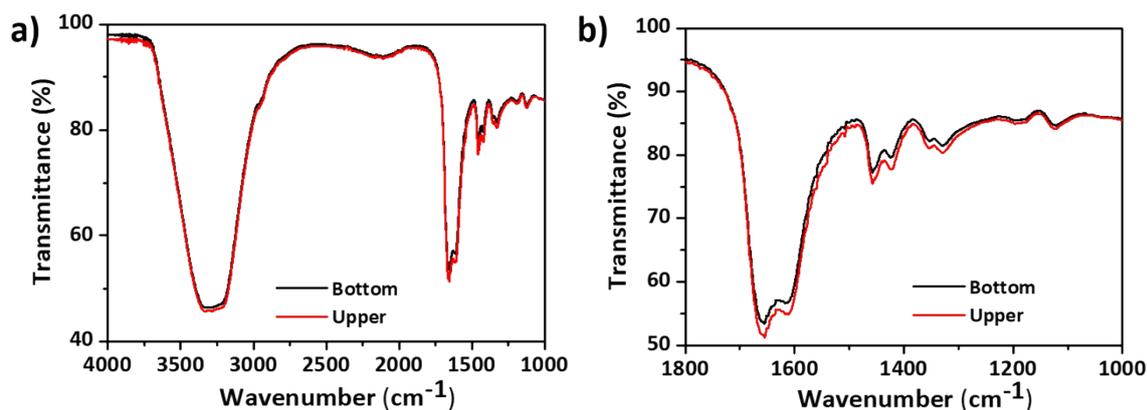


Figure S4. ATR-FTIR spectra of the upper and bottom surfaces of the P(AAc-co-AAm) hydrogel film. $f = 15$ mol%, $T = 100$ μm . The peaks at 1600-1700 cm^{-1} and 1400-1500 cm^{-1} correspond to the asymmetric and symmetric stretching vibrations of carbonyl groups of P(AAc-co-AAm). The upper surface (where the gelation takes place first) has relatively low transmittance, indicating that the upper surface has higher polymer content than the bottom one.

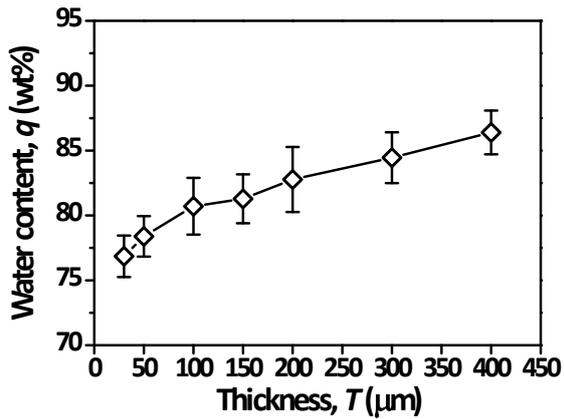


Figure S5. Water content of the P(AAc-co-AAm) hydrogel films with different thickness. $f=15$ mol%, $C_P = 10$ wt%.

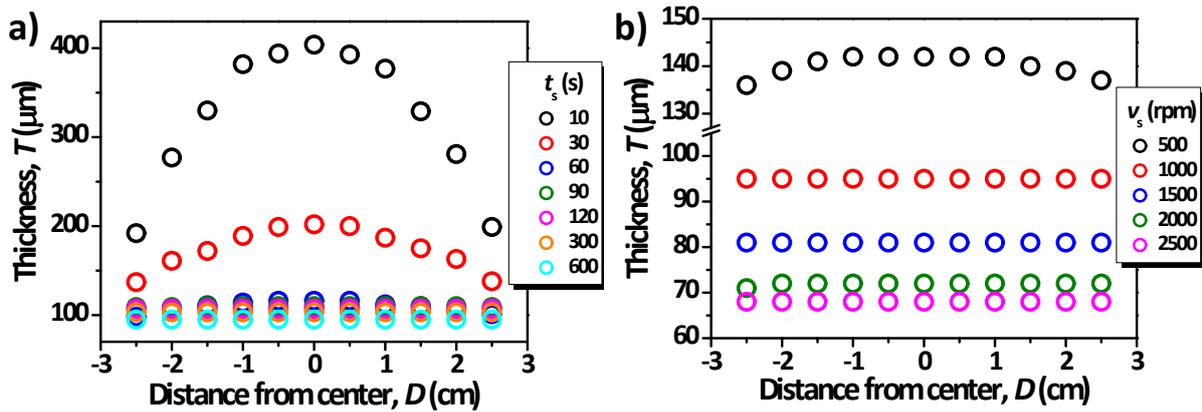
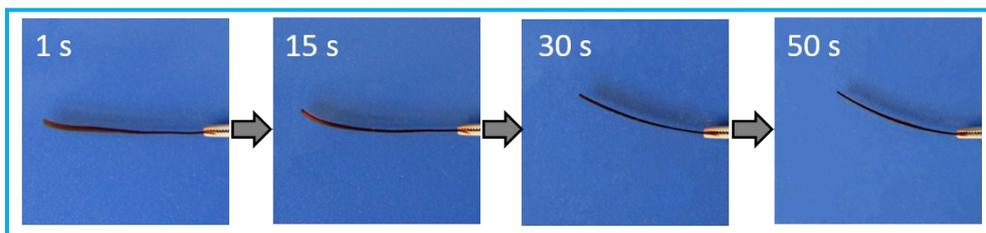


Figure S6. Thickness profile of the equilibrated P(AAc-co-NIPAm) hydrogel films prepared with different spin time, t_s (a) and spin speed, v_s (b); $f=20$ mol%, $C_P = 10$ wt%. (a) $v_s = 1000$ rpm; (b) $t_s = 600$ s.

$C_{\text{NaCl}} = 3 \text{ M}$



Pure water

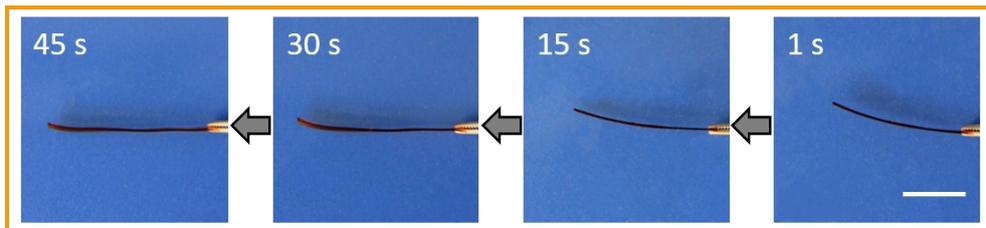


Figure S7. Deformation of P(AAc-*co*-NIPAm) hydrogel film after being transferred from pure water to 3 M saline solution (the upper photos) and then back to pure water (the bottom photos). The flat gel film bends in the direction of bottom surface after being incubated in saline solution, indicating that the upper surface has more compact gel matrix than the bottom one. $f = 20 \text{ mol}\%$; gel dimensions: $30 \text{ mm} \times 5 \text{ mm} \times 100 \text{ }\mu\text{m}$. Scale bar: 10 mm.