

## Supporting Information

### **Crack Blunting and Advancing Behaviors of Tough and Self-healing Polyampholyte Hydrogel**

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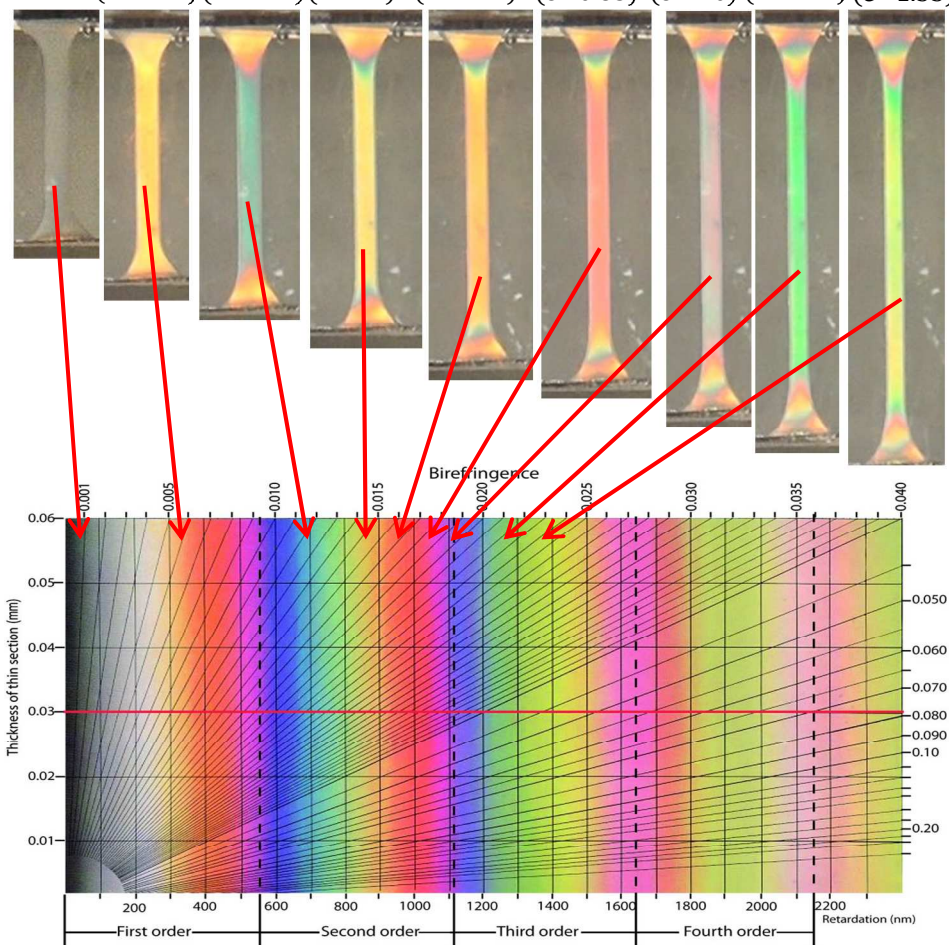
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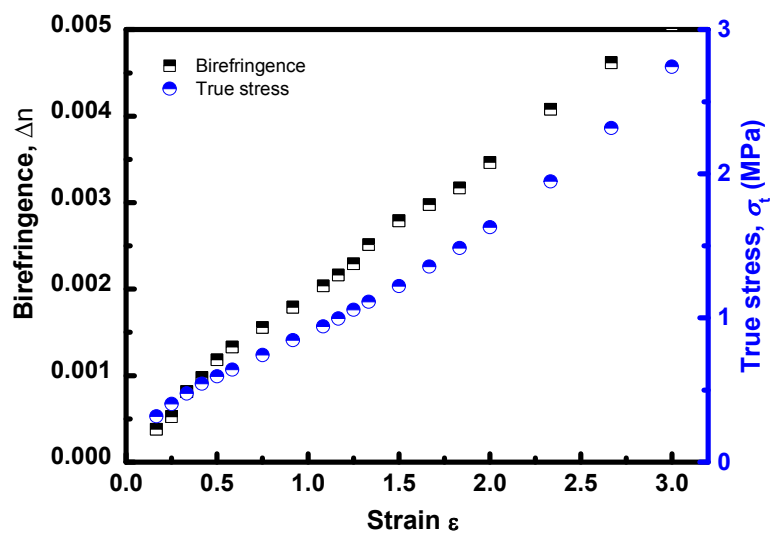
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(a) ( $\epsilon \sim 0$ ) ( $\epsilon \sim 0.17$ ) ( $\epsilon \sim 0.33$ ) ( $\epsilon \sim 0.5$ ) ( $\epsilon \sim 0.67$ ) ( $\epsilon \sim 0.83$ ) ( $\epsilon \sim 1.0$ ) ( $\epsilon \sim 1.17$ ) ( $\epsilon \sim 1.33$ )



(b)



(c)

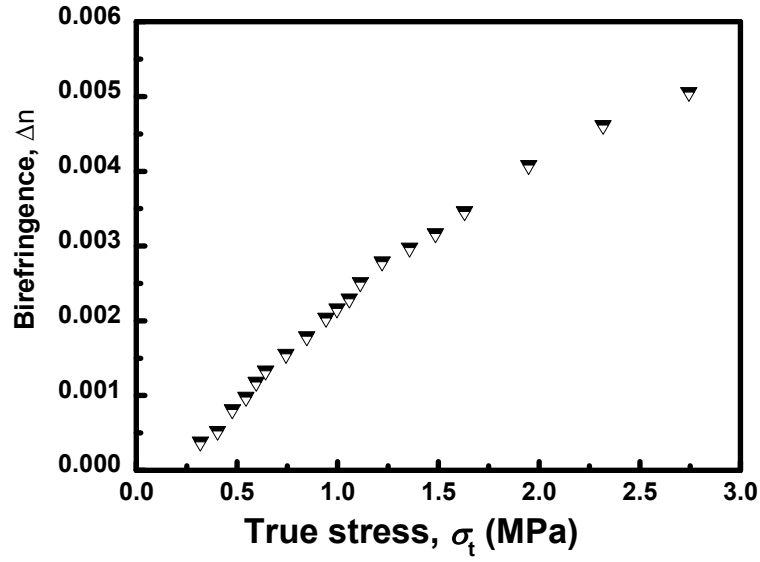


Figure S1. (a) Typical photographs taken under circular polarizer at selected strains for the polyampholyte hydrogel stretched at  $\dot{\epsilon} = 0.14 \text{ s}^{-1}$ , the bottom is the Michel-Levy chart; (b) The birefringence ( $\Delta n$ ) and true stress ( $\sigma_t$ ) as a function of strain ( $\epsilon$ ) for the sample stretched at  $\dot{\epsilon} = 0.14 \text{ s}^{-1}$ ; (c) The birefringence ( $\Delta n$ ) as a function of true stress ( $\sigma_t$ ) for the sample stretched at  $\dot{\epsilon} = 0.14 \text{ s}^{-1}$ .

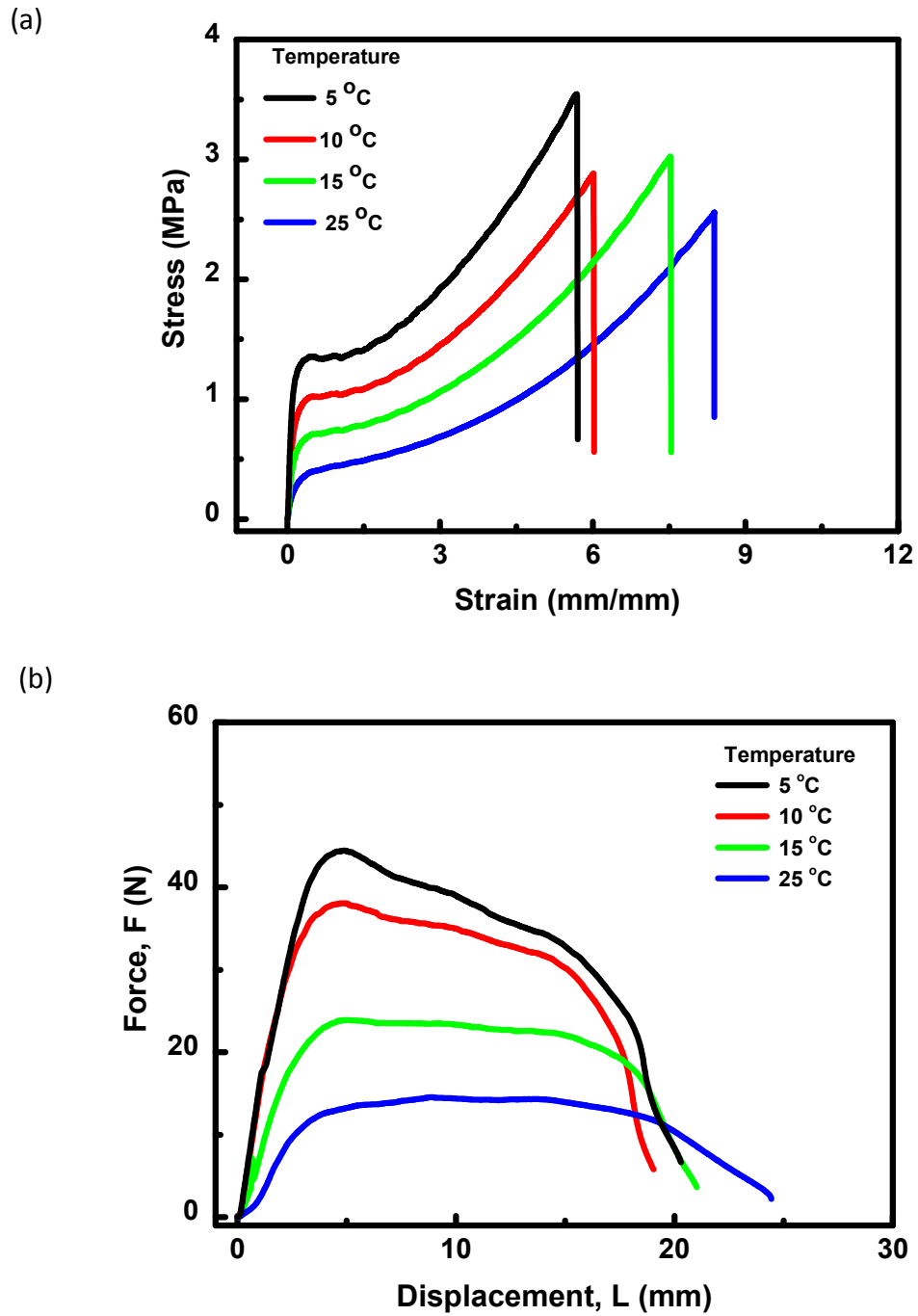


Figure S2. Behaviors of the polyampholyte hydrogel at various temperatures and at a fixed stretch rate of  $0.2 \text{ s}^{-1}$ . (a) Tensile stress–strain curves; (b) Force vs displacement curves for the single edge notched sample.

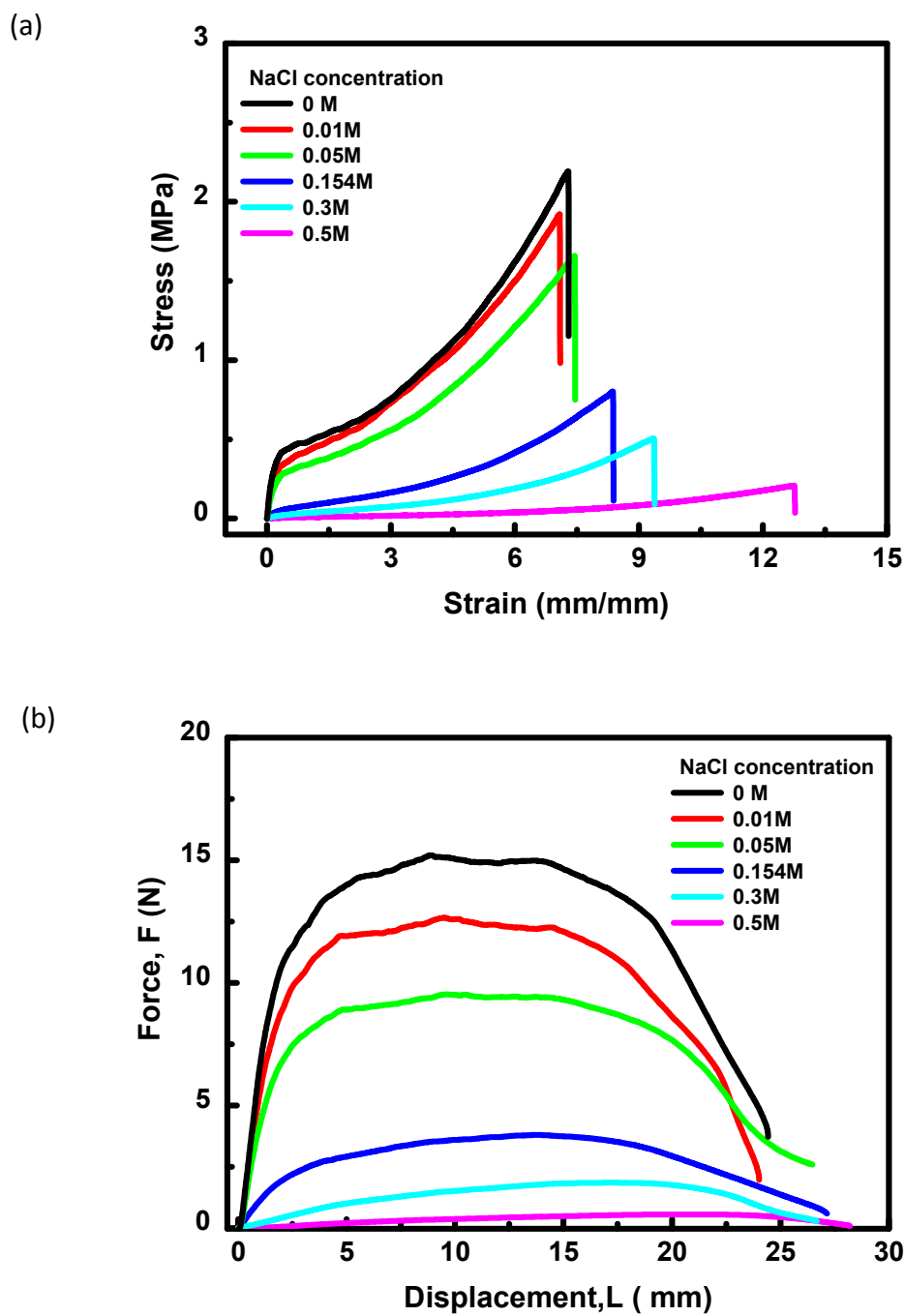


Figure S3. Behaviors of the polyampholyte hydrogel in various NaCl concentrations and at a fixed stretch rate of  $0.2 \text{ s}^{-1}$ . (a) Tensile stress–strain curves; (b) Force vs displacement curves for the single edge notched sample.

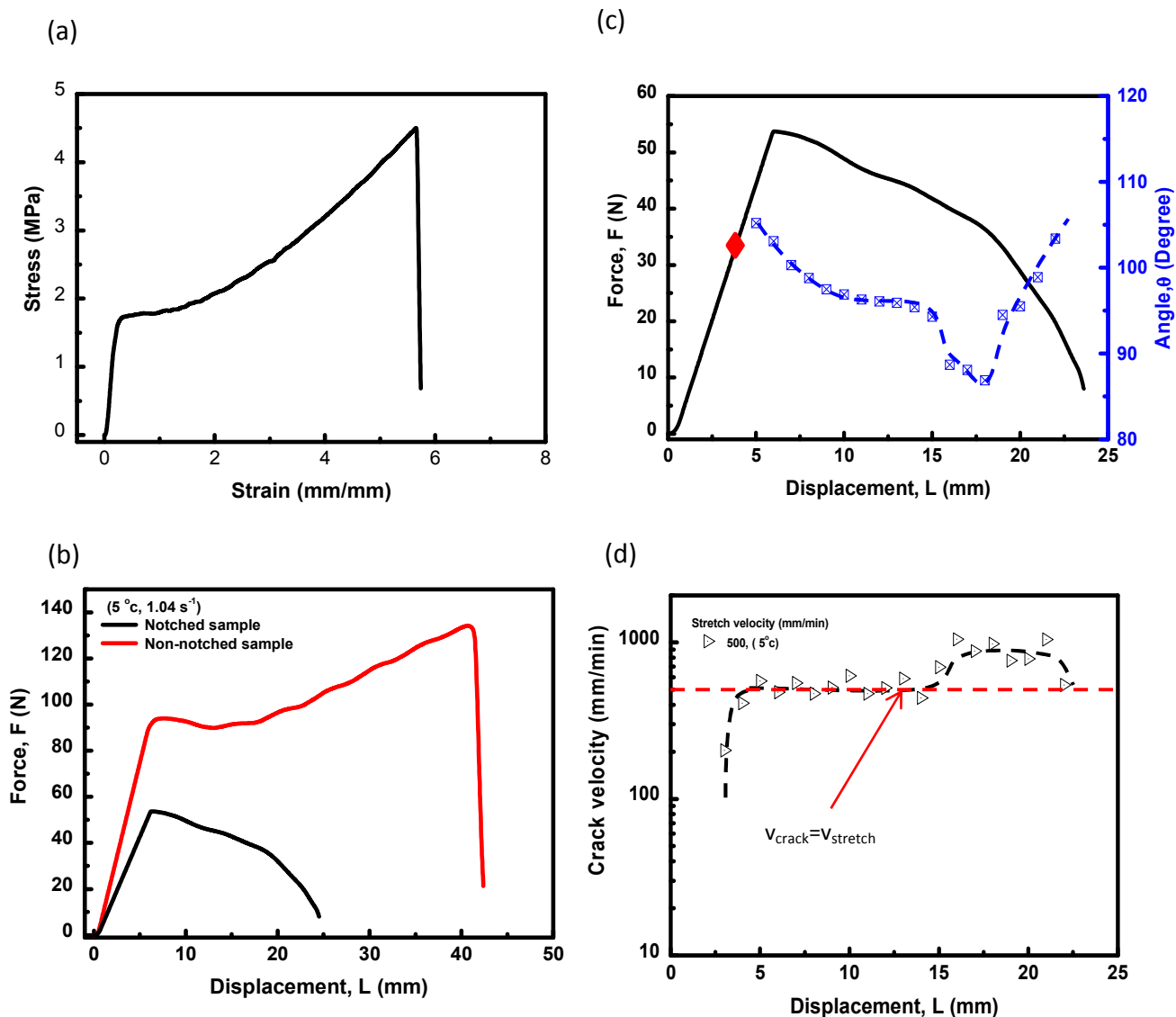


Figure S4. Behaviors of the polyampholyte hydrogel tested at 500mm/min (stretch rates  $1.04 \text{ s}^{-1}$ ) and  $5^\circ\text{C}$ . (a) Tensile stress–strain curves; (b) Force vs displacement curves for both notched and non-notched samples; (c) The force curve of the notched sample and the angle  $\theta$  of crack tip during crack advance. The critical point for the starting of crack advancing is marked on the curve by the diamond square; (d) the crack advancing velocity during crack growth. The red dashed line corresponds to the stretch velocity 500mm/min.

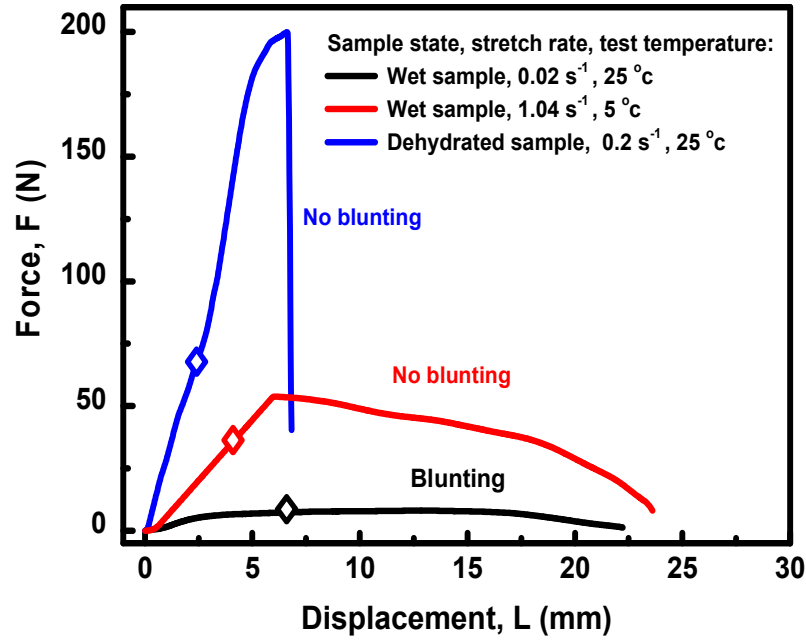


Figure S5. Comparison of force-displacement curves for notched sample in various states. Blue curve: in rigid and brittle state (dehydrated to 26 wt% water content, stretch rate  $0.2 \text{ s}^{-1}$  and temperature  $25^\circ\text{C}$ ); Red curve: In rigid and ductile state (52 wt% water content, stretch rate  $1.04 \text{ s}^{-1}$  and temperature  $5^\circ\text{C}$ ); Black curve: In soft and ductile state (52 wt% water content, stretch rate  $0.02 \text{ s}^{-1}$  and temperature  $25^\circ\text{C}$ ). The black curve is the same curve in **Figure 4a**. The critical points of crack growth are marked on each curve by the diamond square.

**Movie S1.** *In-situ* recording video during tensile test of the polyampholyte hydrogel at  $\dot{\epsilon} = 0.14 \text{ s}^{-1}$  ( $v=100 \text{ mm/min}$ ).

**Movie S2.** *In-situ* recording video during the pure shear test of the single-edge-notched polyampholyte hydrogel at  $\dot{\epsilon} = 0.02 \text{ s}^{-1}$  ( $v=10 \text{ mm/min}$ ). Video is replayed five times of the real speed.

**Movie S3.** *In-situ* recording video during pure shear test of the single-edge-notched polyampholyte hydrogel at high stretch rate ( $\dot{\epsilon} = 1.04 \text{ s}^{-1}$ ) and low temperature ( $5^{\circ}\text{C}$ ). No blunting occurred at this condition. Video is replayed 1/8 times of the real speed.

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