## Supporting Information

## Anisotropic Hydrogels with High Mechanical Strength by Stretching-Induced Oriented Crystallization and Drying

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**Figure S1.** Thickness change of anisotropic PAM-CS hydrogels sheets under different prestretching strains in re-swollen state (CS mass concentration, 7.5%; treatment time in alkaline solution, 1 h).



Figure S2. Tensile curves for different kinds of PAM-CS hydrogels. "a" means alkaline solution: "200%" is the pre-stretching strain.



**Figure S3**. IR of PAM-CS hydrogels with soaking into NaOH solution for different time of 0, 0.5, 1, 2 and 5 h. (The IR of PAM-CS hydrogels with soaking into NaOH solution for different time were detected. It can be seen from the figure that 3290 cm<sup>-1</sup> ~ 3346 cm<sup>-1</sup> is the stretching vibration absorption peak of intermolecular hydrogen bond O-H, and 1646 cm<sup>-1</sup> ~ 1659 cm<sup>-1</sup> is the stretching vibration absorption peak of C=O in amide. However, the absorption peak of C=O in the carboxyl group is 1720 cm<sup>-1</sup> ~ 1706 cm<sup>-1</sup>, and there is a wide and strong stretching absorption peak of O-H at 3300 cm<sup>-1</sup> ~ 2500 cm<sup>-1</sup>. Then it is believed that polyacrylamide is not hydrolyzed to poly acrylic acid.)



**Figure S4**. The elastic modulus of hydrogels with different mass concentration of CS. The elastic modulus increases both in parallel and vertical directions with rising the CS mass concentration (pre-stretching strain, 200%; treatment time in alkaline solution, 1 h).