Electronic Supplementary Material (ESI) for Soft Matter.

Supplementary Information for

Design of Stiff, Tough and Stretchy Hydrogel Composites via Nanoscale Hybrid Crosslinking and Macroscale Fiber Reinforcement

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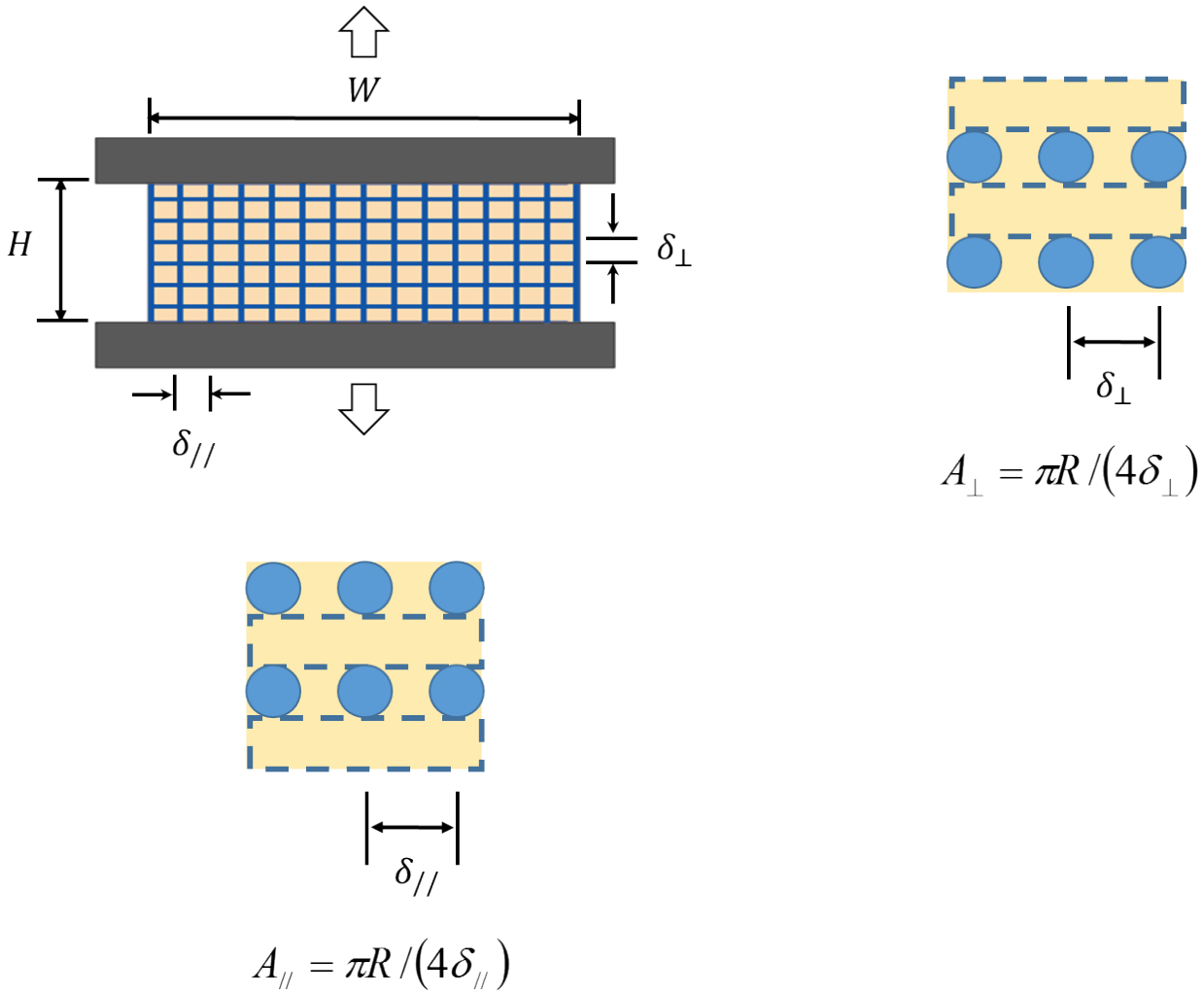

Figure S1. Illustration of the area fractions of fibers along the applied force $A_{//}$ and perpendicular to the applied force $A_\perp$, and the distances between adjacent fibers along the applied force $\delta_{//}$ and perpendicular to the applied force $\delta_\perp$.

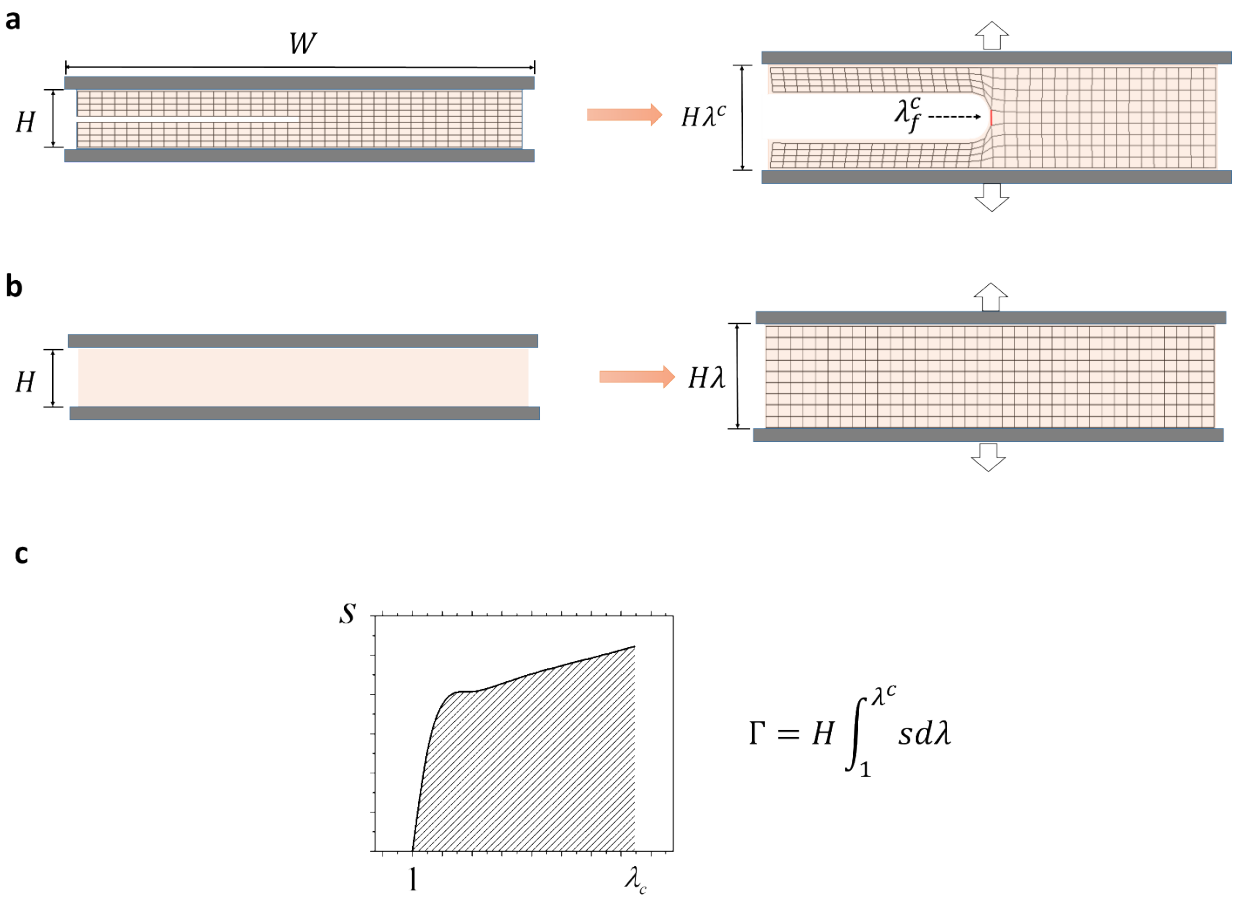

Figure S2. Schematic illustration of the pure-shear test for measuring fracture energy of the fiber-reinforced hydrogel composites. (a) A piece of a fiber-reinforced hydrogel with a notch was stretched to a critical stretch of λ^c until the crack propagates. At the critical stretch, the fiber at the crack tip was stretched to its fracture stretch of λ_f^c. **(b)** The same piece of sample but without notch was stretched to λ^c with the nominal stress s recorded as a function of the stretch λ. **(c)** The fracture energy of the fiber-reinforced hydrogel composite can be calculated as $\Gamma = H\int_1^{\lambda^c} s d\lambda$.

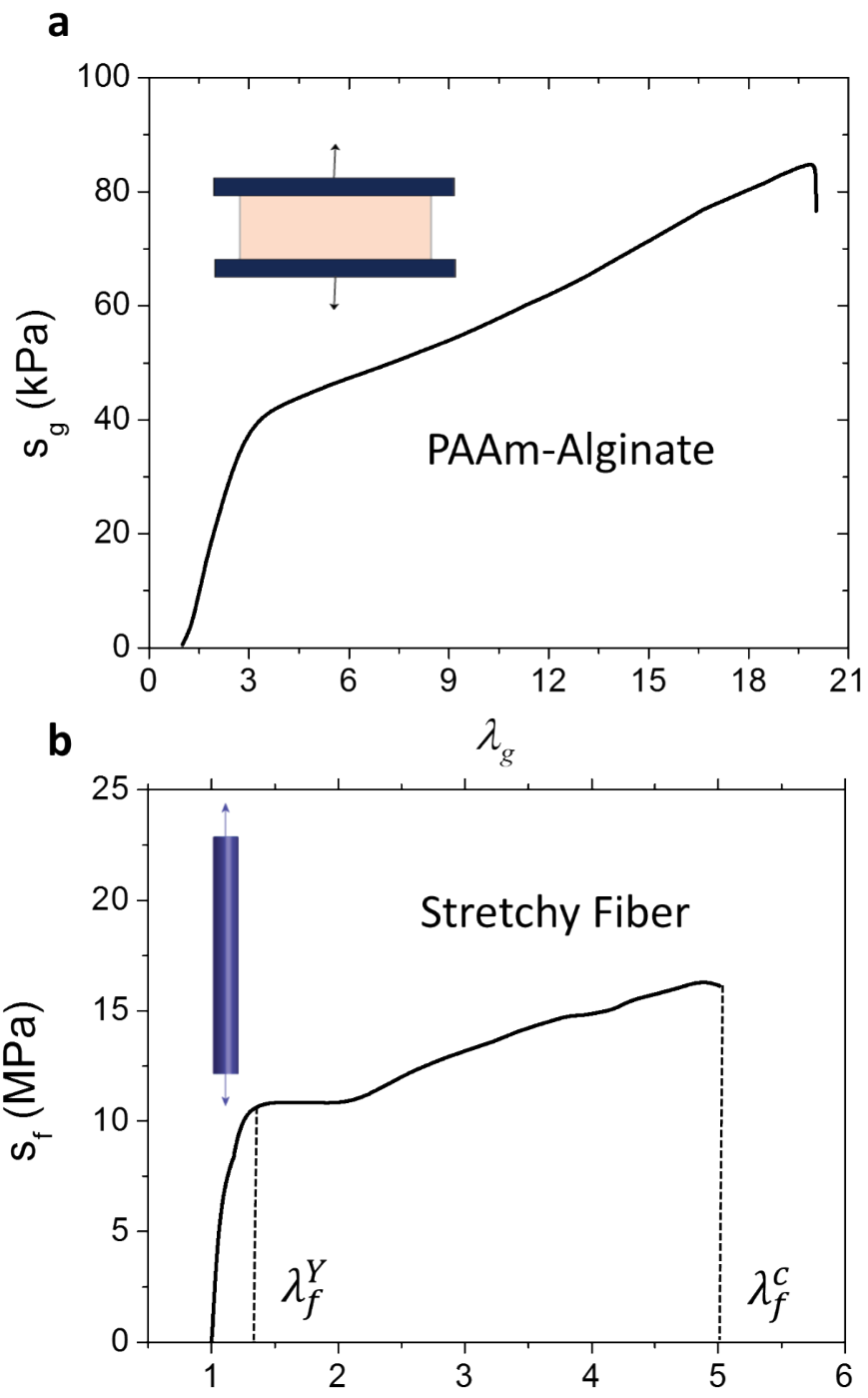

Figure S3. The stress vs. strain curves of (a) a pure PAAm-Alginate gel and (b) a single thermoplastic-elastomer PLA fiber.

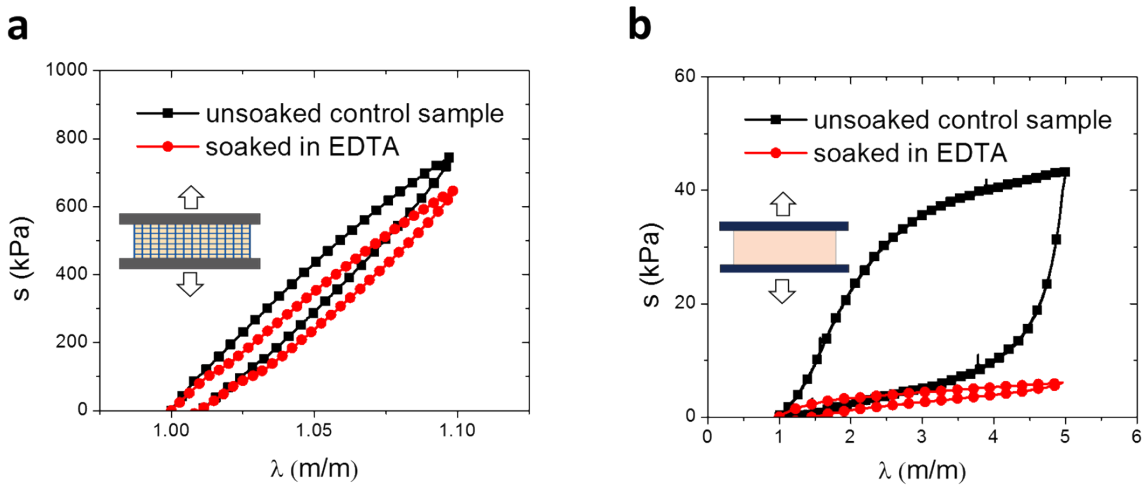

Figure S4. Comparison of stress-stretch hysteresis loops between samples soaked in EDTA for 8 hours (red curves) and un-soaked control samples (black curves): (a) The fiber-reinforced PAAm-Alginate hydrogels ($A_{//} = 7.69\%$). **(b)** Pure PAAm-Alginate hydrogels.

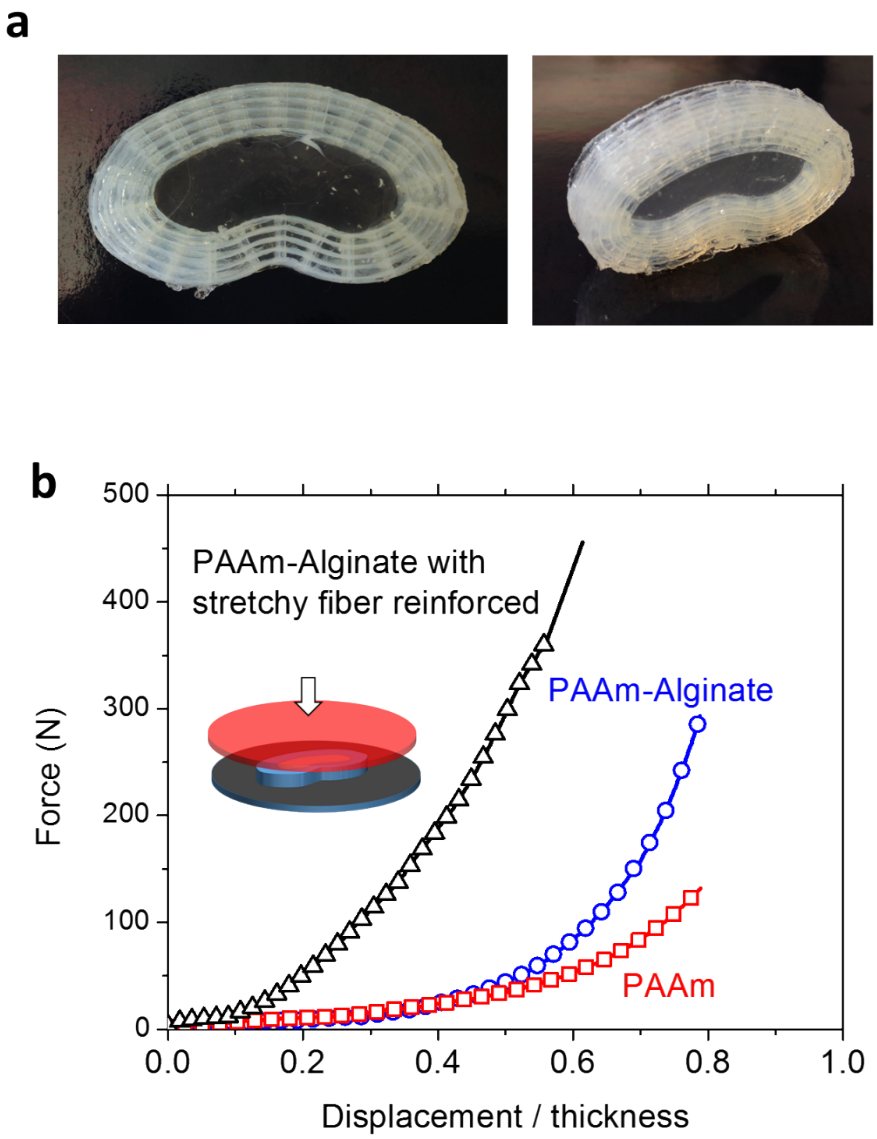

Figure S5. Compression tests on the artificial intervertebral discs with different constitutes: PAAm-Alginate hydrogel as nucleus pulposus surrounded by stretchy-fiber-reinforced PAAm-alginate hydrogel composite as annulus fibrosus (Black line with triangles); pure PAAm-Alginate hydrogel (Blue line with round circles); and pure PAAm hydrogel (Red line with squares, 5.5 ml of 18.7% acrylamide with 377 μL of 0.2g per 100ml N,N-methylenebisacrylamide as the crosslinker, 102 μL of 0.2 M ammonium persulphate as the photo initiator and 8.2 μL N,N,N',N'-tetramethylethylenediamine as the crosslinking accelerator).

Video S1. Crack propagation in a notched sample of the stretchy fiber-reinforced PAAm-Alginate hydrogel composite under pure-shear test.