## **Electronic Supplementary Information**

## Dual physically crosslinked hydrogels based on the synergistic effects

## of electrostatic and dipole-dipole interactions

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| Sample ID  | $c_{ m Ch}$ | $\mathcal{C}_{AAc}$ | $c_{\rm AN}$ | Fracture stress | Fracture strain |
|------------|-------------|---------------------|--------------|-----------------|-----------------|
|            | (wt%)       | (wt%)               | (wt%)        | (MPa)           | (%)             |
| DPC 3-15-5 | 3           | 15                  | 5            | 0.07            | 136             |
| DPC 3-20-5 | 3           | 20                  | 5            | 0.80            | 753             |
| DPC 3-25-5 | 3           | 25                  | 5            | 0.60            | 777             |
| DPC 3-30-5 | 3           | 30                  | 5            | 0.43            | 660             |
| DPC 3-35-5 | 3           | 35                  | 5            | 0.32            | 555             |
| DPC 3-40-5 | 3           | 40                  | 5            | 0.17            | 378             |

**Table S1** Preparation and mechanical properties of the DPC hydrogels.



**Fig. S1** The mechanical behavior of the as-prepared, water-equilibrated, acid-treated and DMSO-treated DPC 2-20-7 hydrogels.



**Fig. S2** The volume change ratio (VCR) of the (a) DPC 2-20-*z*, (b) DPC *x*-20-5 and (c) DPC 2-*y*-5 hydrogels.



Fig. S3 (a) The volume swelling ratio (VSR) of the DPC hydrogels in DMSO/H<sub>2</sub>O. (b) Photograph of the DPC hydrogels immersed in DMSO. The hydrogel on the left has a sufficiently Ch content so that it swelled and maintained its shape in DMSO, whereas the hydrogel on the right without any Ch completely dissolved in DMSO.



**Fig. S4** The calculated total toughness and the dissipated energy of (a) DPC 2-20-5, (b) DPC 2-20-8, (d) DPC 0-20-5 and (e) DPC 3-20-5 hydrogels during the loading-unloading tests under different strains.



Fig. S5 Compressive stress-strain curves of the DPC hydrogels.



Fig. S6 Dissipated energy of DPC 2-20-5 during ten successive loading-unloading cycles.



Fig. S7 The time-dependent recovery of the hysteresis loops of DPC 2-20-5.



**Fig. S8** The mechanical properties of the O-DPC hydrogels compared with the corresponding DPC hydrogels.