### Supplementary information

## **Imparting Conformational Memory for Material Adhesion**

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### Supplementary video legends

Supplementary Video 1: Stretching of a calcium alginate hydrogel bonded on a strip of PDMS coated with and without conformed polydopamine coating.

Supplementary Video 2: Lifting of a glass jar with the hybrid hydrogel and polydopamine as a joint in water.

Supplementary Video 3: Lifting of a soft drink bottle with an artificial mussel byssus formed by hydrogels.

#### **Supplementary figures**



Supplementary Fig. 1: Measured water content of polydopamine aggregates in equilibrium with relative humidity. The normalized water content extrapolated to 100% relative humidity is equivalent to the water content of polydopamine in a fully hydrated environment. The error bars represent one standard deviation ( $n \ge 3$ ).



Supplementary Fig. 2: Chemical characterization of polydopamine. a, Raman spectrum of polydopamine aggregates. b, Fouriertransform infrared spectrum of polydopamine aggregates. c,d, X-ray photoelectron survey (c) and high-resolution spectrums of C1s, O1s, and Fe2p (d) of polydopamine coatings coated on gold.



Supplementary Fig. 3: Physical characterization of polydopamine. a, Atomic force microscopy scans on polydopamine coatings coated on PDMS; the cracks were caused by the drying of polydopamine and the large difference in stiffness between the coating and the underlying substrate, where the pattern of cracks reflects the physical structures of polydopamine. b,c, Advancing (b) and receding (c) contact angles of water on polydopamine coatings coated on PDMS. The error bars represent one standard deviation ( $n \ge 3$ ).



Supplementary Fig. 4: Apparent work of adhesion between polydopamine (+Fe) coating and alginate polymers. The polydopamine (+Fe) coating was formed by depositing ferric ions (0.2 mg/mL) together with polydopamine (1-step) or as a second stage after the deposition of pure polydopamine (2-step). The error bars represent one standard deviation ( $n \ge 3$ ).



Supplementary Fig. 5: Apparent work of adhesion between +MBAA and polyacrylamide polymers. +MBAA was formed by depositing MBAA (0.2 mg/mL) with polydopamine. The polyacrylamide polymers were pre-polymerized and ultraviolet irradiation was not used during indentation. The error bars represent one standard deviation ( $n \ge 3$ ).



Supplementary Fig. 6: Apparent work of adhesion between +Fe and polyacrylamide polymers. +Fe was formed by depositing Fe (0.2 mg/mL) with polydopamine. The error bars represent one standard deviation ( $n \ge 3$ ).



Supplementary Fig. 7: Apparent work of adhesion between polydopamine deposited with isopropanol (+isoOH) or methanol (+meOH) and polyacrylamide polymers at different alcohol concentrations. The error bars represent one standard deviation ( $n \ge 2$ ).