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Supporting Information

Controlled Shape Deformation of Bilayer Film with Tough Adhesion between Nanocomposite Hydrogel and Polymer Substrate

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Uniaxial tensile tests of as-prepared dumbbell-shaped samples (50 mm in length, 4 mm in width, and 2 mm in thickness) were performed with commercial testing machine with a 100 N load cell with a crosshead speed of 100 mm min^{-1}. The tensile strength (σ) was defined as $\sigma_f = F/A_0$, where F is the load force at the sample fracture, and A_0 is the original specimen cross-sectional area. Tensile strain (ε_f) was determined as $\varepsilon_f = \Delta l/l_0$ (mm/mm), where Δl is the elongate length and l_0 is the initial length. Elastic modulus (E) of the gels was calculated by fitting the initial linear regime of stress-strain curve. Fracture energy (W) was calculated from the area under the stress-strain curve of the tensile test.

As shown in Figures S2a, S2b, and Table S1, P2 hydrogel had weak tensile properties (i.e. high tensile strength of 0.14 MPa and elastic modulus of 0.02 MPa), while the as prepared bilayer film with P2 hydrogel and PET film exhibited excellent tensile properties (i.e. high tensile strength of 3.44 MPa and large elastic modulus of 10.61 MPa).

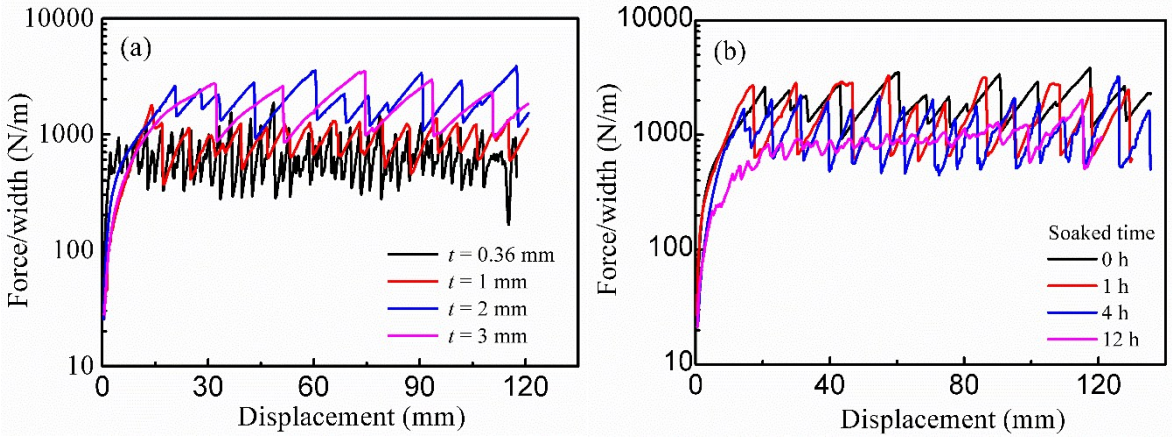

Figure S1. (a) Peeling curves of force per width of hydrogel sheet versus displacement for the bilayer films with different hydrogel thickness. (b) Peeling curves of force per width of hydrogel sheet versus displacement for the P2 bilayer film soaked in water.

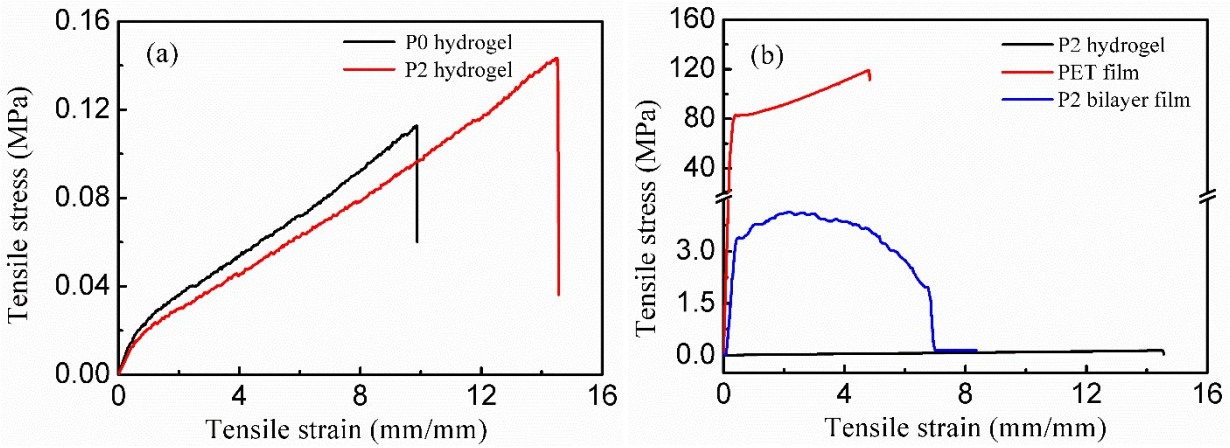

Figure S2. Tensile stress-tensile strain curves of (a) P0 and P2 hydrogels and (b) P2 hydrogel, PET film and bilayer film.

Table S1. Mechanical properties of P0 hydrogel, P2 hydrogel, PET film and bilayer film.

Samples	σ_f (MPa)	ε_f (mm/mm)	E (MPa)	W (MJ/m^3)
P0 hydrogel	0.11±0.009	9.53±1.221	0.03±0.003	0.62±0.100
P2 hydrogel	0.14±0.004	14.95±0.672	0.02±0.0002	1.12±0.044
PET film	120.12±1.653	4.37±0.619	252.02±4.971	422.22±43.932
bilayer film	3.44±0.208	5.04±0.557	10.61±1.804	21.21±2.589

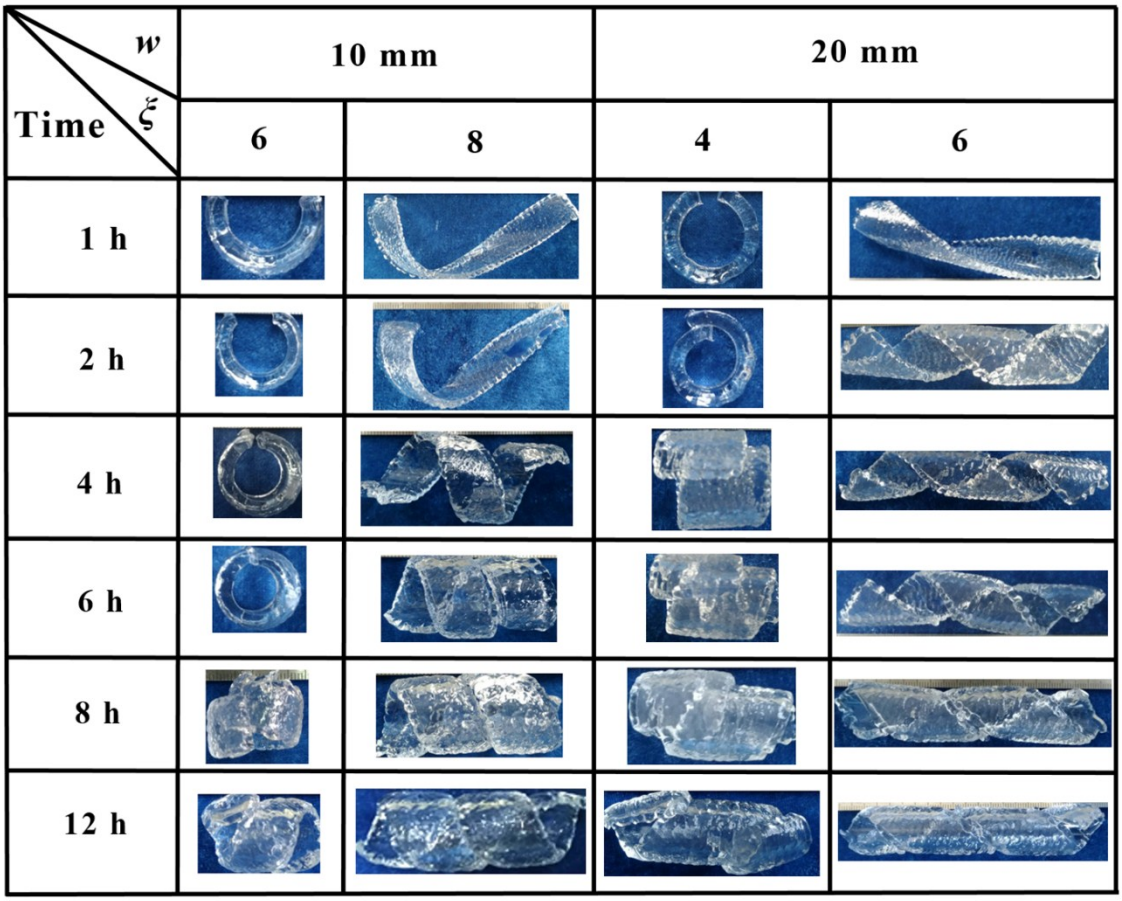

Figure S3. Shape-shifting process of the bilayer film with different sizes.