ADVANCED FUNCTIONAL MATERIALS

Supporting Information

for *Adv. Funct. Mater.,* DOI: 10.1002/adfm.201901721

Printing Hydrogels and Elastomers in Arbitrary Sequence with Strong Adhesion

Hang Yang, Chenghai Li, Meng Yang, Yudong Pan, Qianfeng Yin, Jingda Tang, Hang Jerry Qi, and Zhigang Suo**

Supporting Information

Printing Hydrogels and Elastomers in Arbitrary Sequence with Strong Adhesion

Hang Yang, Chenghai Li, Meng Yang, Yudong Pan, Qianfeng Yin, Jingda Tang Hang Jerry Qi and Zhigang Suo**

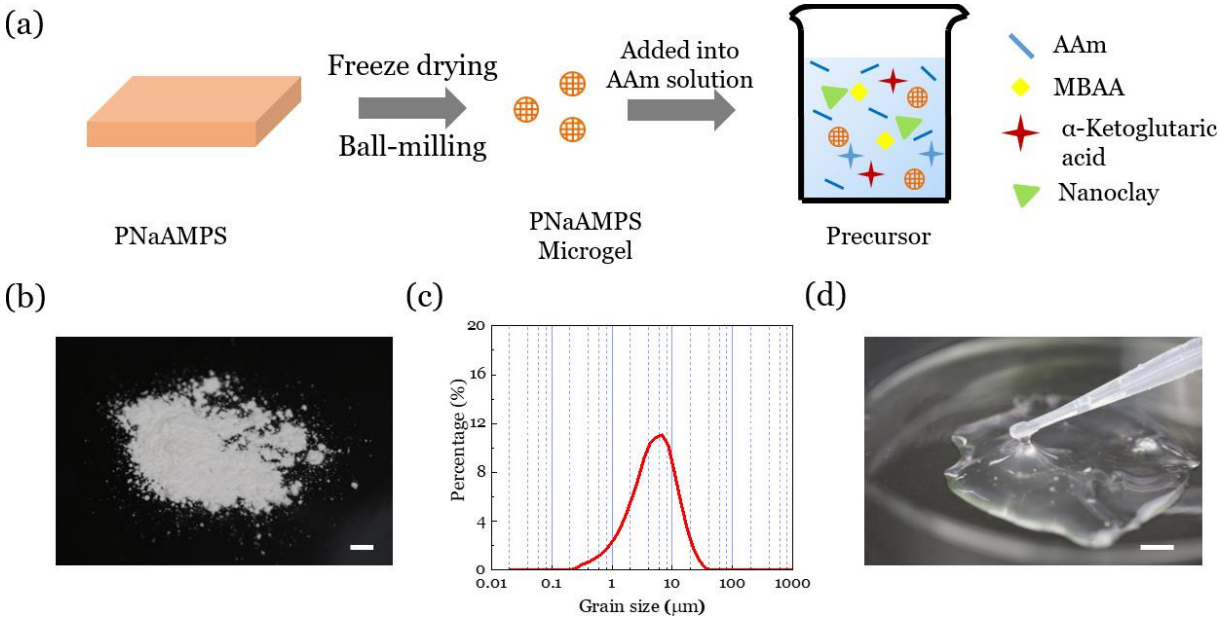

Figure S1. Preparation of the hydrogel ink. (a) The PNaAMPS hydrogel is synthesized by radical polymerization, and is freeze-dried and ball-milled into fine powders of microgel. The microgel, along with a rheological modifier (nanoclay), is mixed into the precursor of the polyacrylamide hydrogel. (b) An image of the microgel powders. (c) The distribution of grain size of the microgel powders. (d) The hydrogel ink has the rheology of a plastic liquid, stable against gravity and capillarity. The scale bars are all 5 mm.

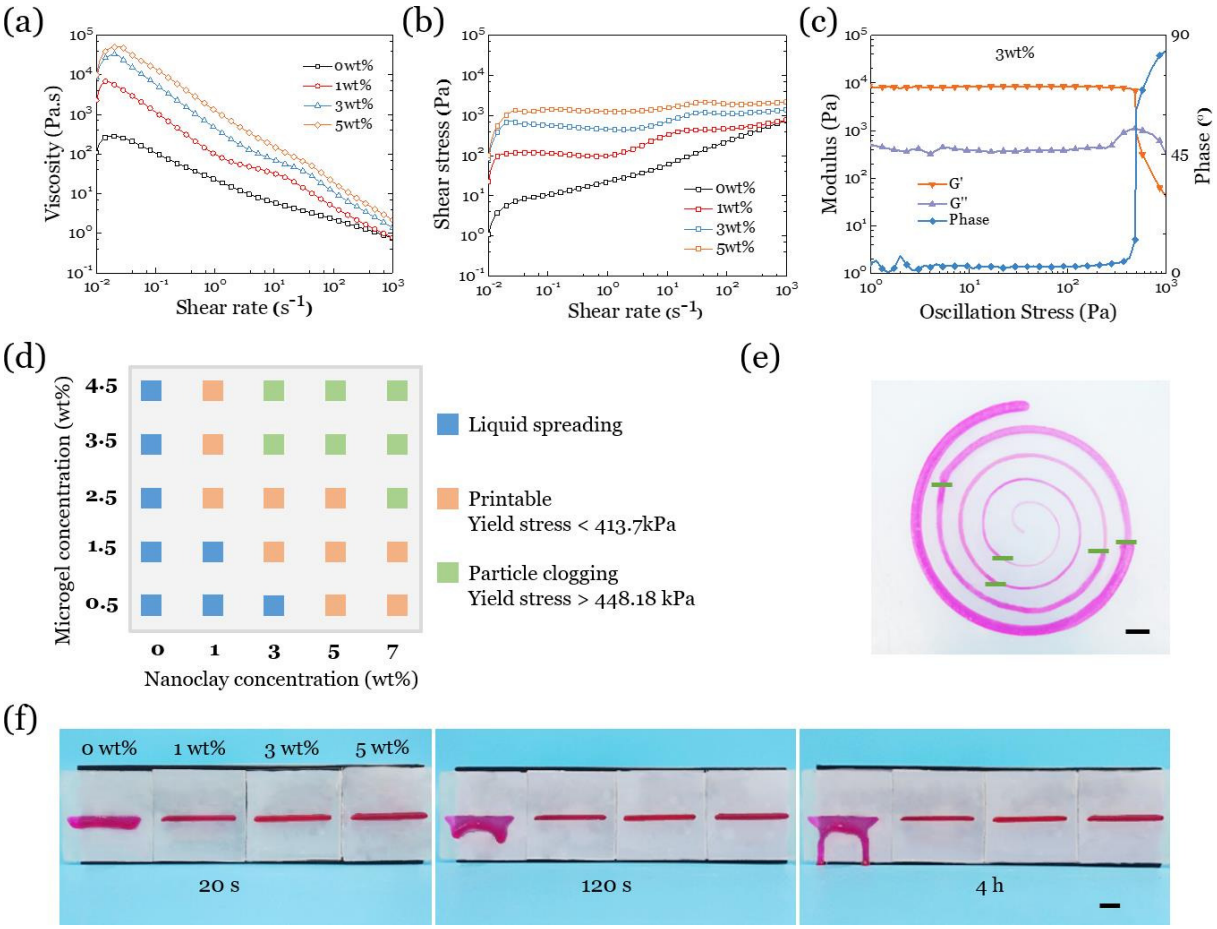

Figure S2. Rheology of the hydrogel inks. (a) Viscosity and (b) shear stress of the hydrogel inks with different concentrations of nanoclay in a range of shear rates. (c) Oscillatory rheology results showing shear storage (G') and loss (G") moduli evolution of hydrogel inks over increasing shear stress (2.5 wt% of microgel and 1 wt% of nanoclay relative to the total mass). (d) The ink is printable for certain combinations of the concentrations of the microgel and nanoclay. (e) A print using nozzles of several diameters (0.26 mm, 0.41 mm, 0.60 mm, 0.84 mm, 1.20 mm and 1.54mm). Different parts are distinguished by short green lines. The scale bar is 2 mm. (f) Hydrogel inks with different concentrations of nanoclay (0 wt%, 1 wt%, 3 wt% and 5 wt %) were printed on the elastomer. The hydrogel inks containing nanoclay self-support on the elastomer after standing vertically for four hours. The scale bar is 5 cm.

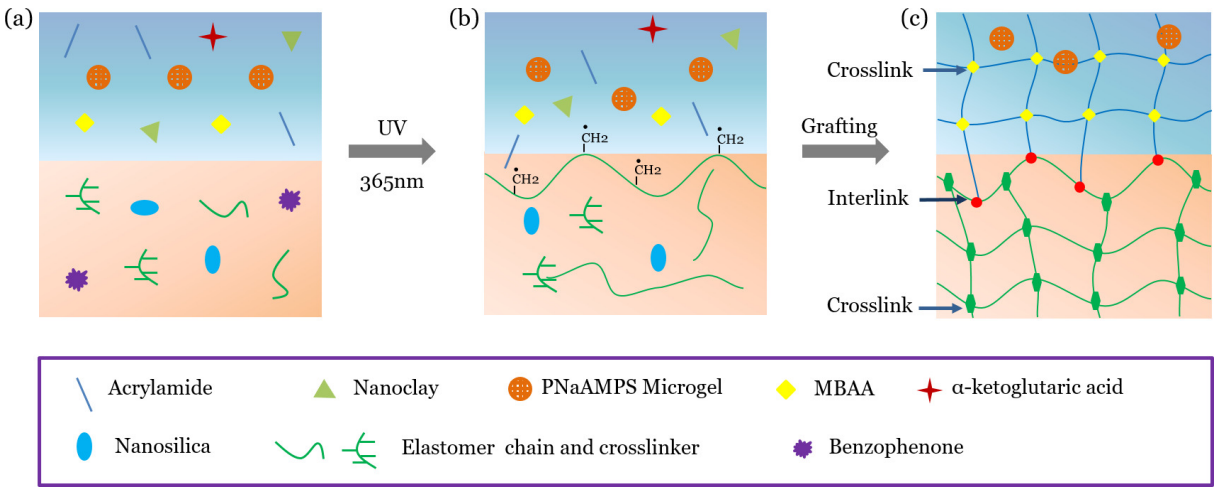

Figure S3. The chemistry of cure. (a) During print, the hydrogel ink and the elastomer ink are in contact. (b) During cure, the elastomer forms a covalent network. The interlink initiator (benzophenone) abstracts hydrogen atoms from the C-H bonds on the elastomer chains and create reactive sites, which will graft polymer chains of the hydrogel. (c) After cure, covalent crosslinks form within the hydrogel network and the elastomer network, and covalent interlinks form between the two networks.

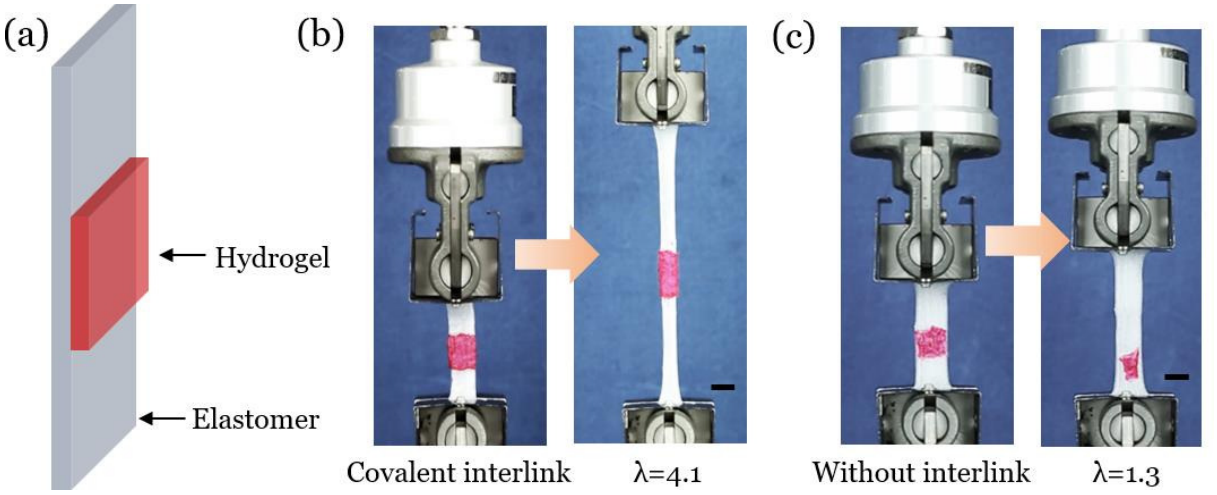

Figure S4. Hydrogel-elastomer bilayer under tension. (a) The schematic of the bilayer. (b) The hydrogel remains bonded to the benzophenone-modified elastomer at a large stretch of 4.1. (c) The hydrogel debonds from the unmodified elastomer at a small stretch of 1.3. The scale bars are all 1cm.

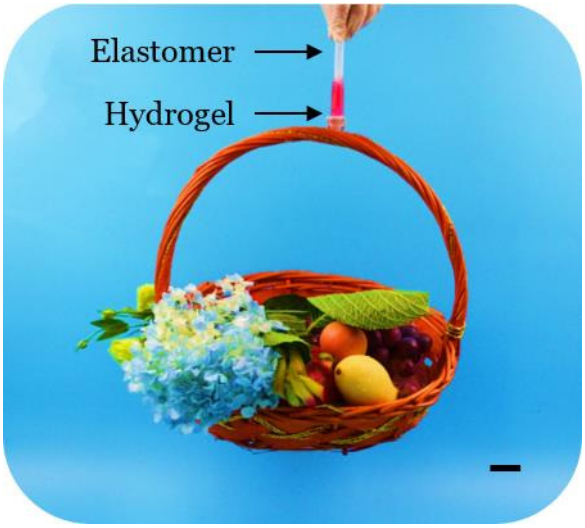

Figure S5. The hydrogel-elastomer hybrid cylinder can bear the weight of a basket full of flowers and fruits. The estimated tensile stress in the hydrogel-elastomer is 70 kPa. The scale bar is 5cm.

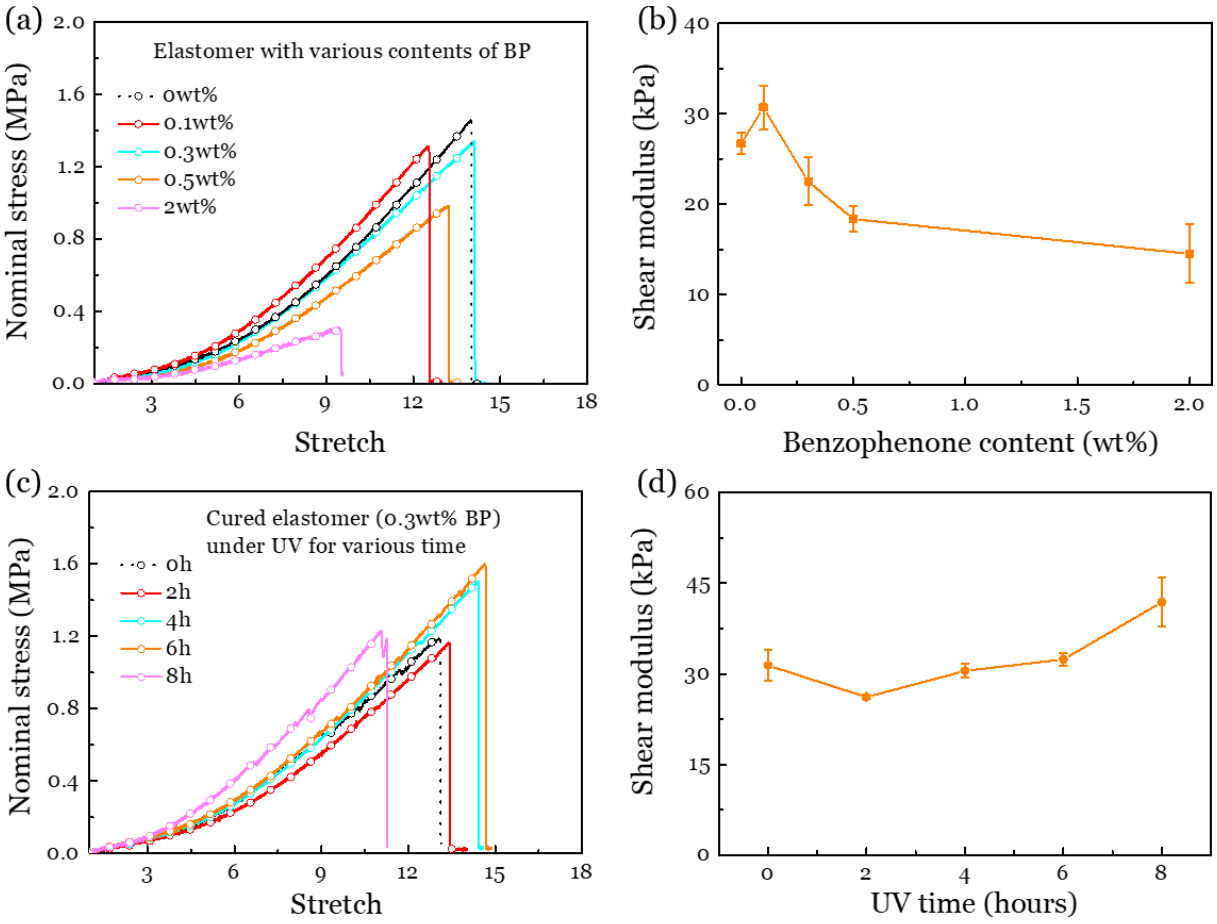

Figure S6. The interlink initiator in the elastomer ink affects the stress-stretch curves of the cured elastomer. (a) Stress-stretch curves and (b) shear moduli for the elastomers with various concentrations of the interlink initiator. The elastomers are cured under UV (365 nm 2.5 mW cm^{-2}) for 8 hours then stored in oven for 2 hours at 80°C. (c) Stress-stretch curves and (d) shear moduli for pre-cured under UV treatment for various time. The elastomers contain 0.3wt% of the interlink initiator and stored without light overnight before UV treatment.

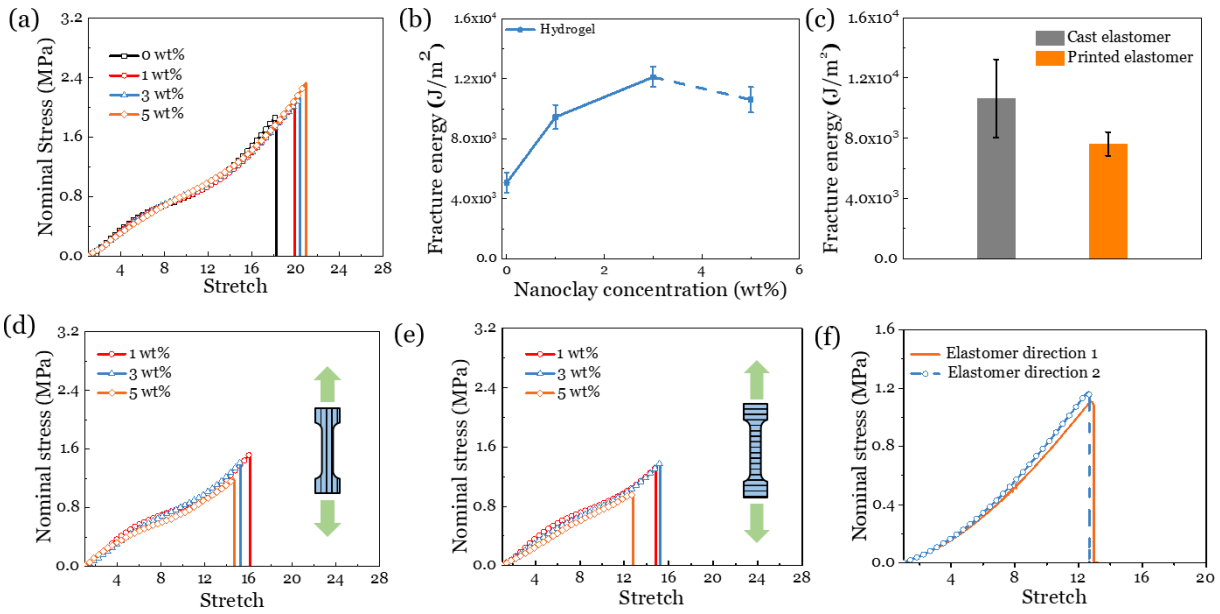

Figure S7. Mechanical properties of casts and prints. (a) Stress-stretch curves and (b) fracture energy of cast hydrogels with different concentrations of nanoclay. (c) Fracture energy of cast and printed elastomer. (d) Stress-stretch curves of printed hydrogels along the print direction (direction 1) and (e) vertical to the print direction (direction 2). (f) Stress-stretch curves of printed elastomer in two directions. Unless otherwise noted, the elastomer was added with 4 wt% of nanosilica.

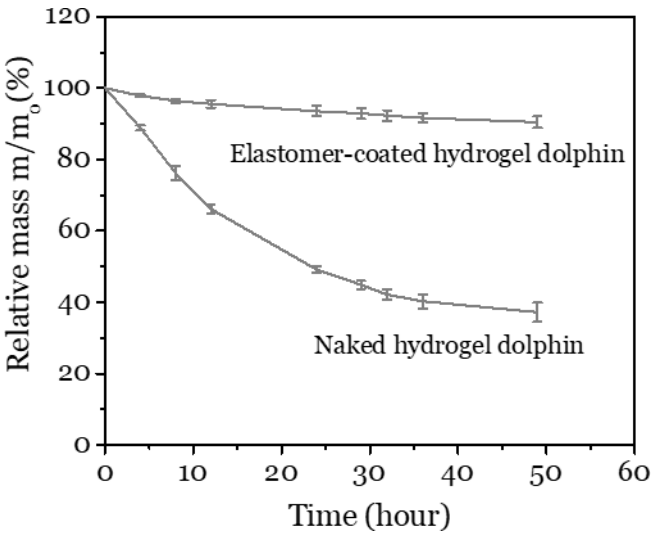

Figure S8. The dehydration of hydrogel dolphins with or without elastomer coating in the open air. The naked hydrogel dolphin loses much weight. The elastomer-coated hydrogel dolphin loses a small amount of weight.