

# Supporting Information

## One-pot and One-step Fabrication of Salt Responsive Bilayer Hydrogels with 2D and 3D Shape Transformations

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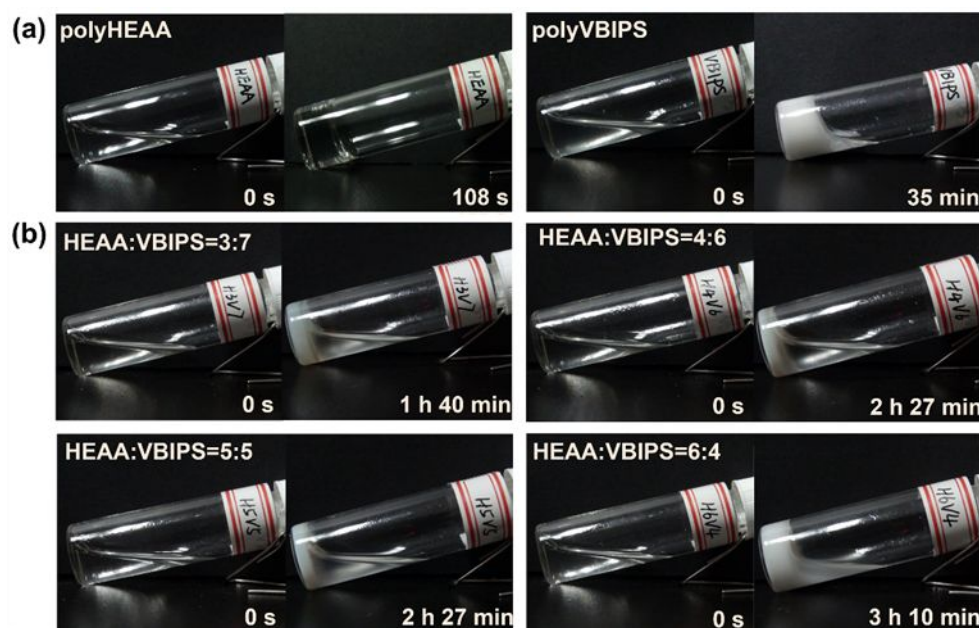


Figure S1. (a) Gel formation of the pristine polyHEAA and polyVBIPS hydrogels and (b) first layer gel formation of polyHEAA/polyVBIPS bilayer hydrogels with different monomer ratios.

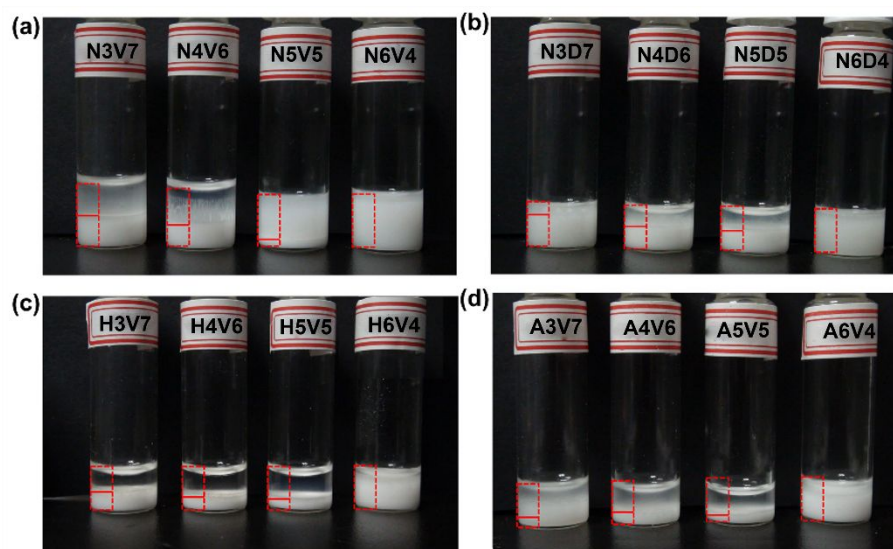
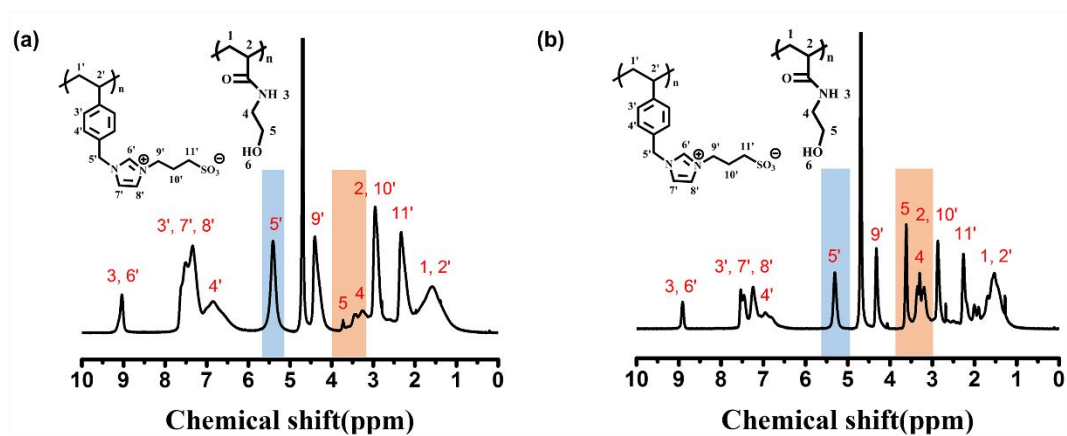


Figure S2. Images of the precipitation layer formed from copolymerization systems of (a) NIPAM/VBIPS, (b) NIPAM/DVBAPS, (c) HEAA/VBIPS, and (d) AAm/VBIPS with different monomer ratios. The systems have the same recipes with those of bilayer hydrogels but without crosslinking agent.



FigureS3.  $^1\text{H}$ -NMR spectra of copolymers formed in first step from HEAA:VBIPS= (a) 3:7 and (b) 6:4.

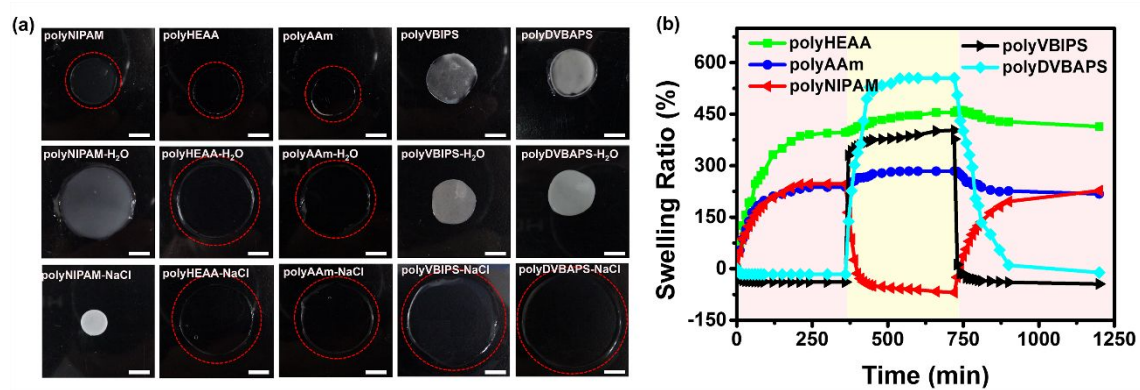


Figure S4. (a) Images and (b) swelling/deswelling curves of pristine polyNIPAM, polyHEAA, polyAAM, polyVBIPS and polyDVBAPS hydrogels.

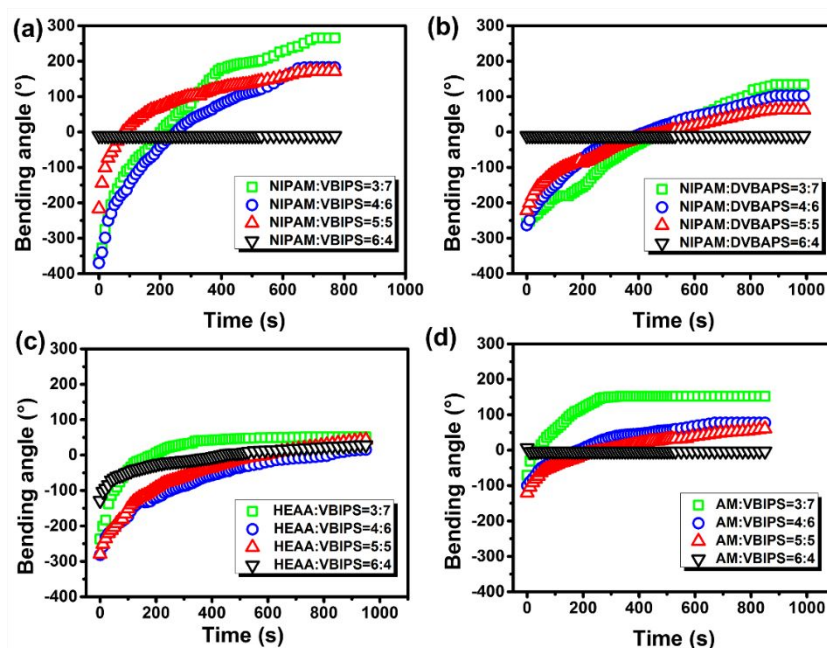


Figure S5. Bending kinetics of (a) polyNIPAM/polyVBIPS, (b) polyNIPAM/polyDVBAPS, (c) polyHEAA/polyVBIPS and (d) polyAAM/polyVBIPS bilayer hydrogels, prepared at different monomer ratios, in response to the environmental changes from water to 1.0 M NaCl solution.

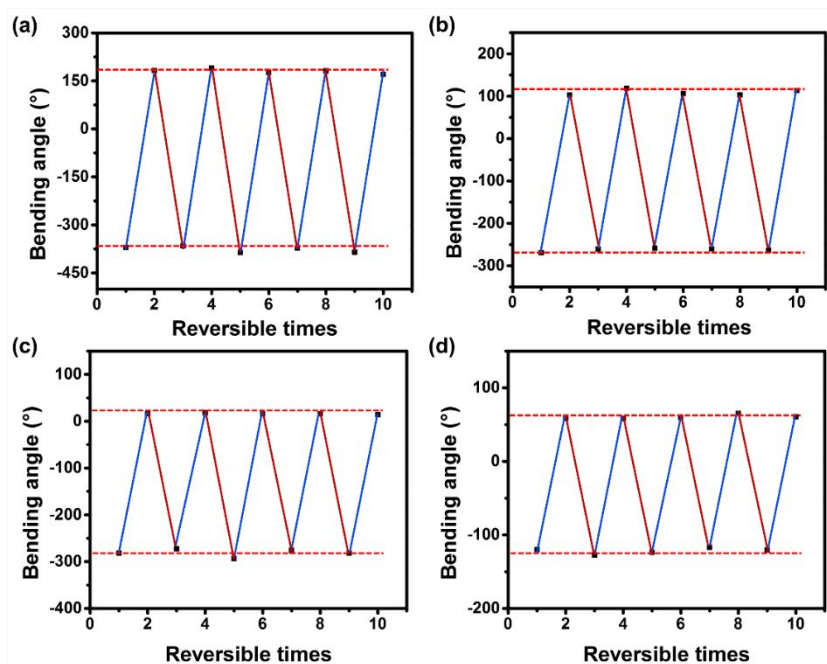


Figure S6. Reversible bending of (a) polyNIPAM/polyVBIPS (N4V6), (b) polyNIPAM/polyDVBAPS (N4D6), (c) polyHEAA/polyVBIPS (H4V6) and (d) polyAAM/polyVBIPS (A5V5) bilayer hydrogels with 1.0 mm thickness with the environments switched between in water and in 1.0 M NaCl solutions.