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Supplementary Materials for

Hydrogels of arrested phase separation simultaneously achieve high strength and low hysteresis

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Other Supplementary Material for this manuscript includes the following:

Movies S1 and S2

Supplementary Text 1

For the PEA used in this work, the entanglements greatly outnumber crosslinks, so that chain length determined by the entanglements instead of the chemical crosslinks is used to predict the rupture stretch. The entanglement molecular weight of PEA is ~ 12,000 g mol^{-1}, and the molecular weight of its monomer is ~ 100 g mol^{-1}, which gives a chain length, n ~ 120 monomers (*21*). We adopt a freely-jointed model to predict the rupture stretch (*13*). In the undeformed state, the mean-square-root distance between the two ends of the polymer chain is $\sqrt{n}a$, where a is the length of a repeating unit. In the state right before rupture, the polymer chain is fully extended and its length becomes na. Consequently, the predicted rupture stretch is $\sqrt{n}$. Using the representative value of n ~ 120 gives a predicted rupture stretch ~ 11.

Supplementary Text 2

A stage III PEA-PAAc hydrogel contains 15.6 wt% PEA, 56.3 wt% PAAc, and 28.1 wt% water. The corresponding stage IV sample contains 5.3 wt% PEA, 19.1 wt% PAAc, and 75.6 wt% water. We use these mass fractions to estimate the rule of mixture of the strength of the PEA-PAAc hydrogel.

The fractocohesive length is estimated as Γ/W_f, where Γ is the toughness and W_f is the work of fracture (*26*). The work of fracture is obtained from the area under the loading curves. For pure PEA, Γ ~ 1,400 J m^{-2} and W_f ~ 12.04 MJ m^{-3}, which gives a Γ/W_f ~ 116 μm. For PEA-PAAc hydrogel, Γ ~ 1650 J m^{-2} and W_f ~ 6.6 MJ m^{-3}, which gives a Γ/W_f ~ 262 μm.

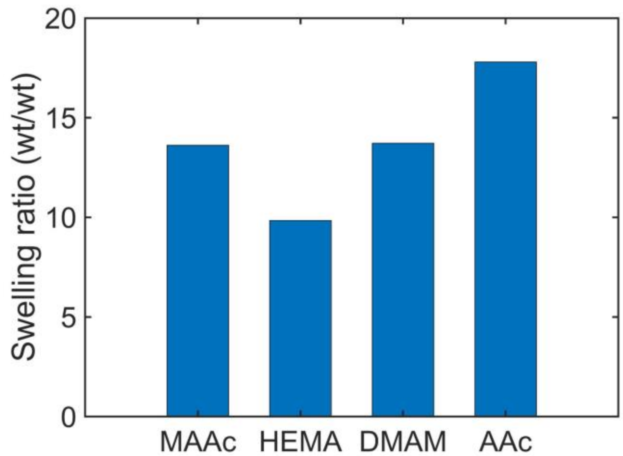

Fig. S1. PEA elastomer swells to equilibrium in several liquid monomers of hydrogels. PEA elastomers are submerged in the liquid monomers to swell to equilibrium. The swelling ratio is defined by the weight ratio of the PEA elastomer after swelling to equilibrium and the pristine PEA elastomer. MAAc: methacrylic acid; HEMA: (hydroxyethyl)methacrylate; DMAM: *N,N*-dimethylacrylamide; AAc: acrylic acid.

Table S1. The modulus, strength, and hysteresis of the PEA-PAAc hydrogel, PAAc hydrogel, and PEA elastomer.

	Modulus (kPa)		Strength (MPa)		Hysteresis (%)	
	Average	STDEV	Average	STDEV	Average	STDEV
PEA-PAAc hydrogel	440.8	19.6	6.7	0.2	16.9	0.2
PAAc hydrogel	160.7	5.7	0.40	0.07	2.1	0.2
PEA elastomer	609.1	23.1	2.6	0.2	6.7	0.1

The test for each composition is repeated using five samples.

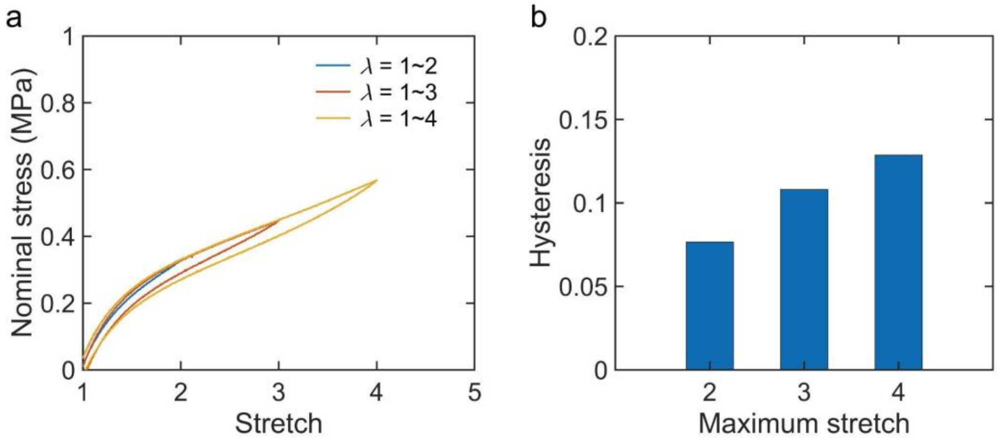

Fig. S2. Pure PEA. a, Stress-stretch curves under loading and unloading of pure PEA with increasing maximum stretches. Each sample is tested for one cycle. **b**, The hysteresis of pure PEA at different maximum stretches.

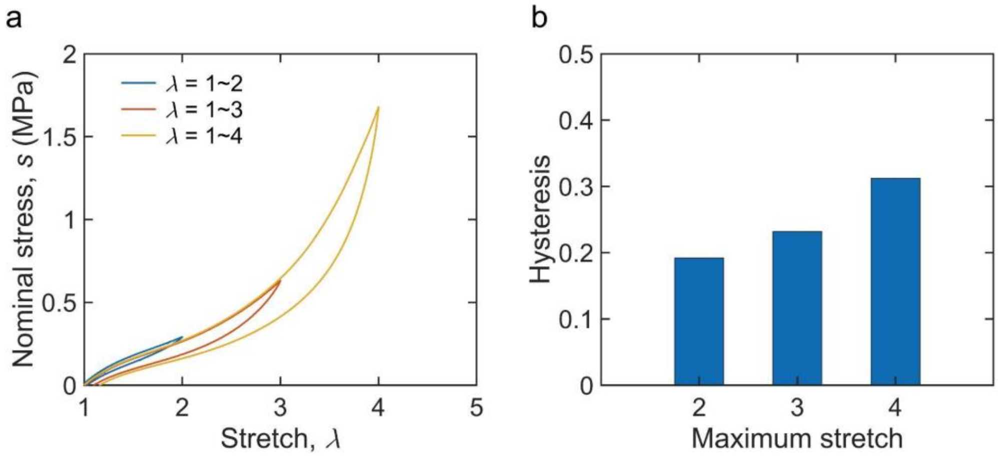

Fig. S3. PEA-PAAc hydrogel. a, Stress-stretch curves under loading and unloading of the PEA-PAAc hydrogel with increasing maximum stretches. Each sample is tested for one cycle. **b**, The hysteresis of the PEA-PAAc hydrogel at different maximum stretches.

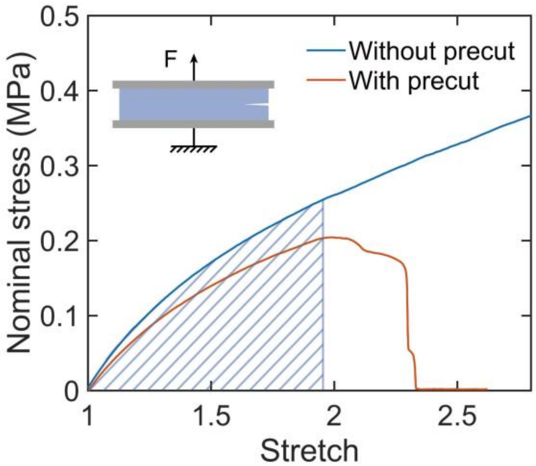

Fig. S4. The toughness of the PEA-PAAc hydrogel. Two samples of the PEA-PAAc hydrogels are prepared, one with precut and the other without precut. Both samples are monotonically stretched until rupture, and the stress and stretch are recorded. The toughness is calculated as the area under the curve of the uncut sample between stretch 1 and λ_{max}, where λ_{max} is the stretch when the crack starts to propagate in the precut sample.

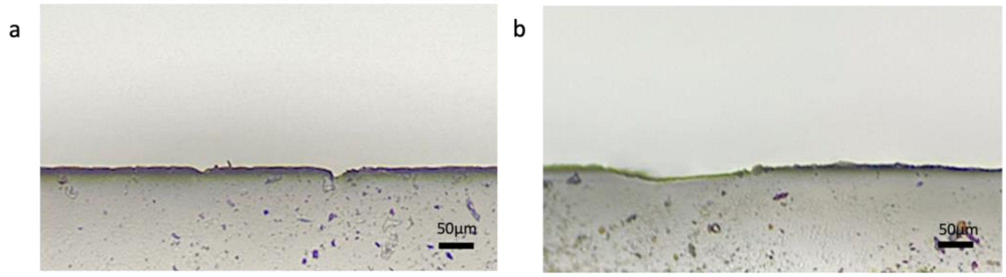

Fig. S5. The microscopic images of the samples cut by the dumbbell shape cutter. Flaws with size ~ 10 μm are observed on the edge of the samples.

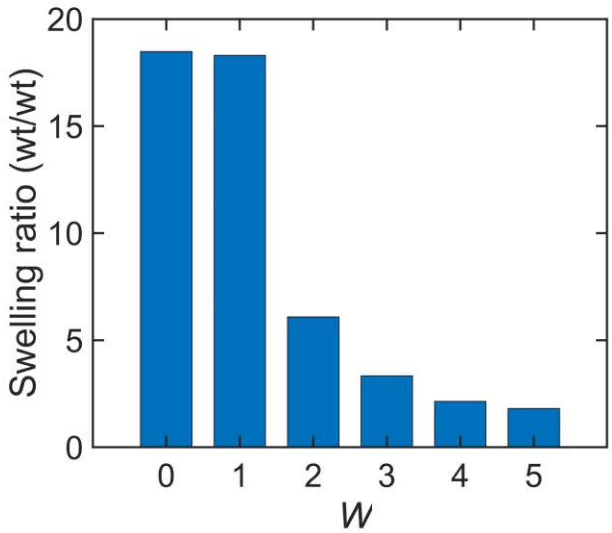

Fig. S6. The PEA elastomer swells differently in AAc-water solution with different *W*. PEA elastomers are submerged in AAc-water solutions with different *W* to swell to the equilibrium. The swelling ratio increases as *W* decreases. When $W \leq 1$, the swelling ratio of PEA is nearly constant.

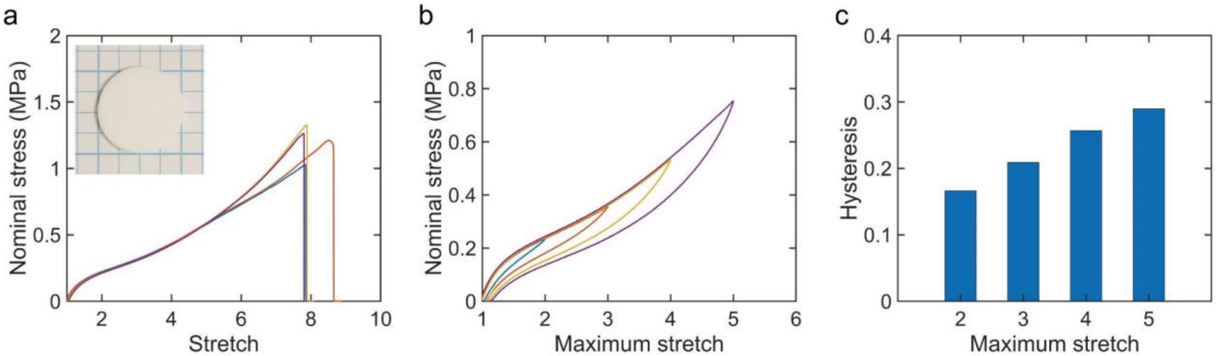

Fig. S7. The PEA-PAAc hydrogel of *W* = 4. a, The PEA-PAAc hydrogel of *W* = 4 is monotonically stretched until rupture. The inset is the optical image of the hydrogel. **b**, Stress-stretch curves under loading and unloading with increasing maximum stretches. Each sample is tested for one cycle. **c**, The hysteresis at different maximum stretches.

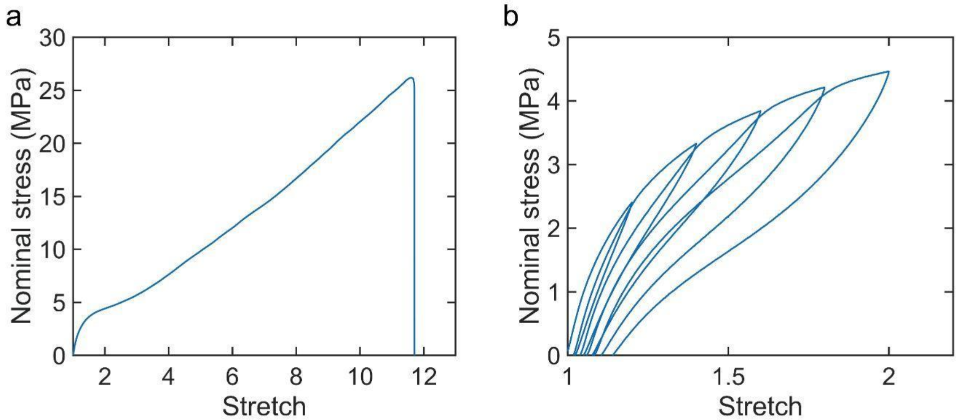

Fig. S8. The mechanical properties of PEBAX elastomer. a, The PEBAX elastomer is monotonically stretched until rupture. **b**, The stress-stretch curves of one PEBAX during cyclic loading with increasing maximum stretches. The same PEBAX sample is tested continuously throughout the test.

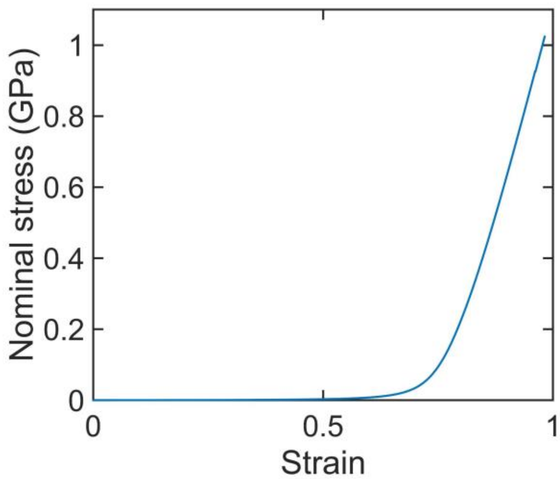

Fig. S9. The compressive test of the PEBAX-PAAc hydrogel. A disk of the PEBAX-PAAc hydrogel (diameter ~ 3 mm) is compressed monotonically. The maximum stress that can be applied is limited by the Instron machine, and the sample does not rupture after the compression.

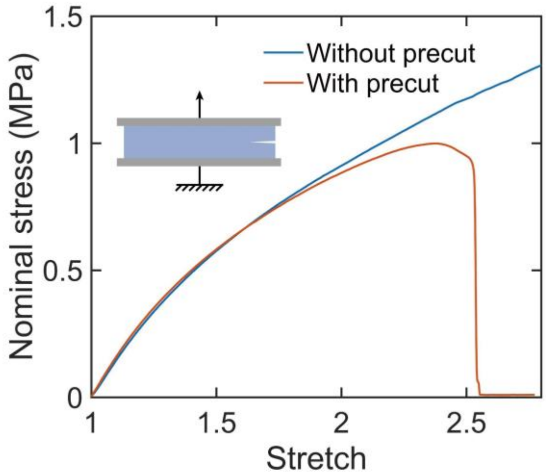

Fig. S10. The toughness test of the PEBAX-PAAc hydrogel. Two samples of the PEBAX-PAAc hydrogels are prepared, one with precut and the other without precut. Both samples are monotonically stretched until rupture, and the stress and stretch are recorded. The toughness is obtained from the area under the curve of the uncut sample between stretch 1 and λ_{max}, where λ_{max} is the stretch when the crack starts to propagate in the precut sample.

Movie S1. The PEA-PAAc hydrogel is stretched and then released by hand.
Movie S2. The PEA-PAAc hydrogel is used to lift a gallon (3.8 liters) of water.