

## Support information

### A Facile Preparation of Liquid Crystalline Elastomer with Self-healing Performance for Light-Responsive Soft Actuator

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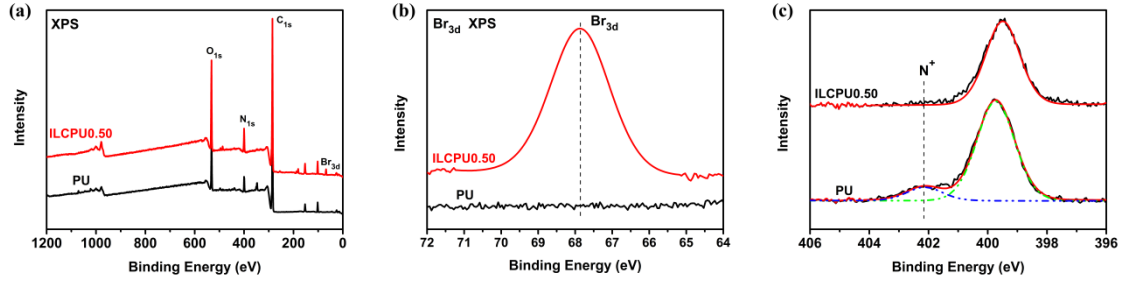
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**Figure S1.** (a) XPS, (b) Br<sub>3d</sub>, and N<sub>1s</sub> spectra of PU and ILCPU0.50.

The number-average molecular weight of polymers was evaluated by Gel permeation chromatography (GPC). The model of the GPC is Beijing Wenfen LC98IIRI equipped with two gel columns (Shodex, KD-803 and KD-806), and the refractive index detector is RI-201H, using DMF as the mobile phase. The test temperature is 40 °C and the standard sample is polystyrene.

**Table S1.** The GPC results of PU and ILCPU samples.

Sample	M <sub>n</sub> (g/mol)	M <sub>w</sub> (g/mol)	Polymer Dispersity Index
PU	2.17 × 10 <sup>4</sup>	3.47 × 10 <sup>4</sup>	1.60
ILCPU0.25	2.51 × 10 <sup>4</sup>	3.56 × 10 <sup>4</sup>	1.42
ILCPU0.50	2.92 × 10 <sup>4</sup>	3.94 × 10 <sup>4</sup>	1.35
ILCPU0.75	3.17 × 10 <sup>4</sup>	4.09 × 10 <sup>4</sup>	1.29
ILCPU1.00	3.45 × 10 <sup>4</sup>	4.17 × 10 <sup>4</sup>	1.21

The calculus of the grafting ratios of the liquid crystalline units:

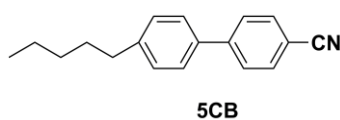
$$G = 100 \times \frac{M_{n,ILCPU} - M_{n,PU}}{M_{Br-C6-CB}} \bigg/ \frac{M_{n,PU} \times \omega}{M_{MDEA}}$$

where, G denotes the grafting ratios of the liquid crystalline units of the samples;

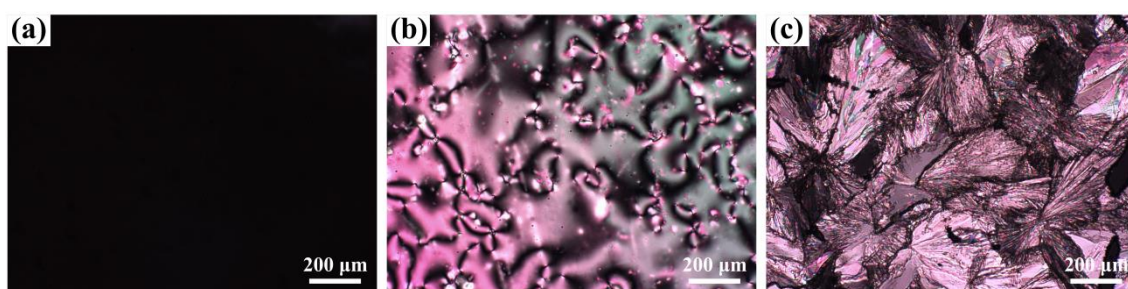
$M_{n,ILCPU}$  and  $M_{n,PU}$  denote the number-average molecular weight of ILCPU and PU,

respectively;  $M_{Br-C6-CB}$  and  $M_{MDEA}$  donate the molecular weight of Br-C6-CB and MDEA, respectively;  $\omega$  denotes the mass fraction of MDEA in the materials which were used to prepare PU.

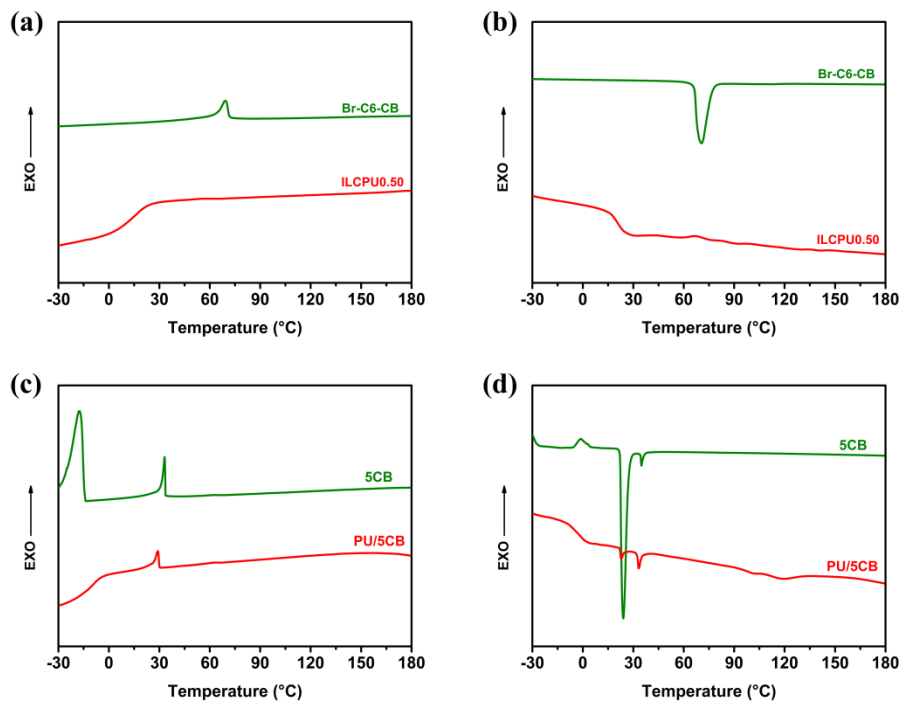
The calculated grafting ratios (G) were 12.55%, 27.69%, 36.91%, and 47.25%, corresponding to ILCPU0.25, ILCPU0.50, ILCPU0.75, and ILCPU1.00, respectively.



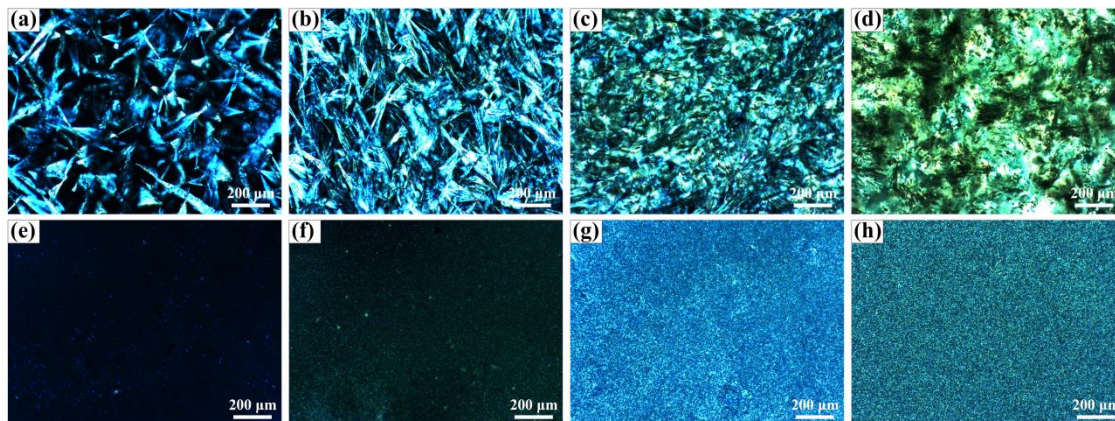
**Figure S2.** The chemical structure of 5CB.



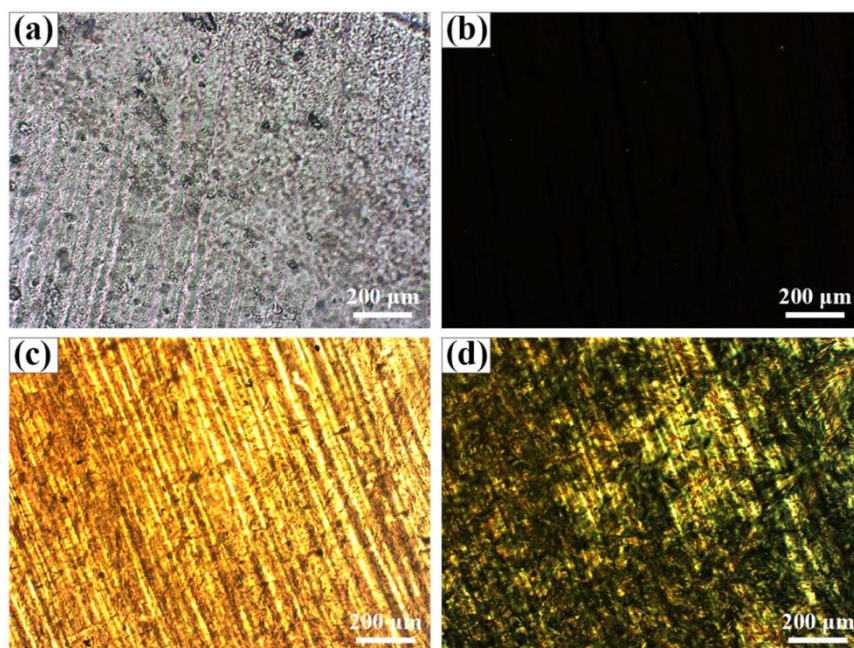
**Figure S3.** POM images of Br-C6-CB at (a) 80 °C, (b) 55 °C, and (c) 0 °C on cooling process.



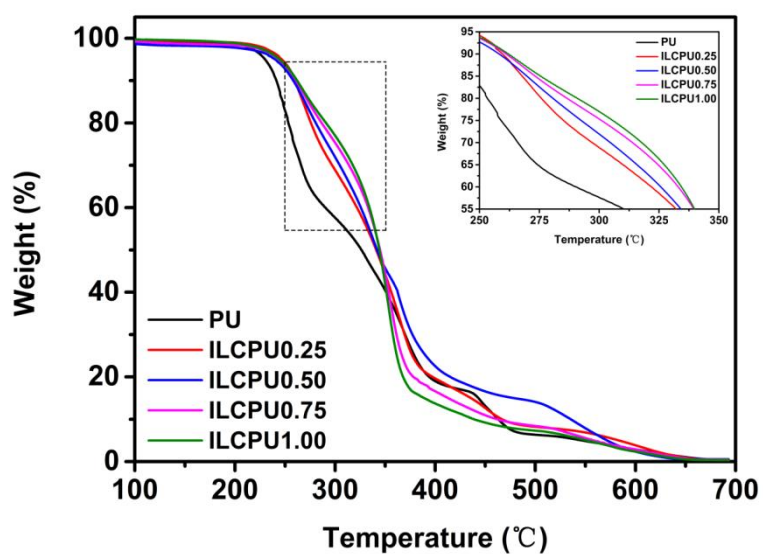
**Figure S4.** DSC curves of ILCPU0.50 on (a) first cooling process and (b) second heating process. DSC curves of PU/5CB on (c) first cooling process and (d) second heating process.



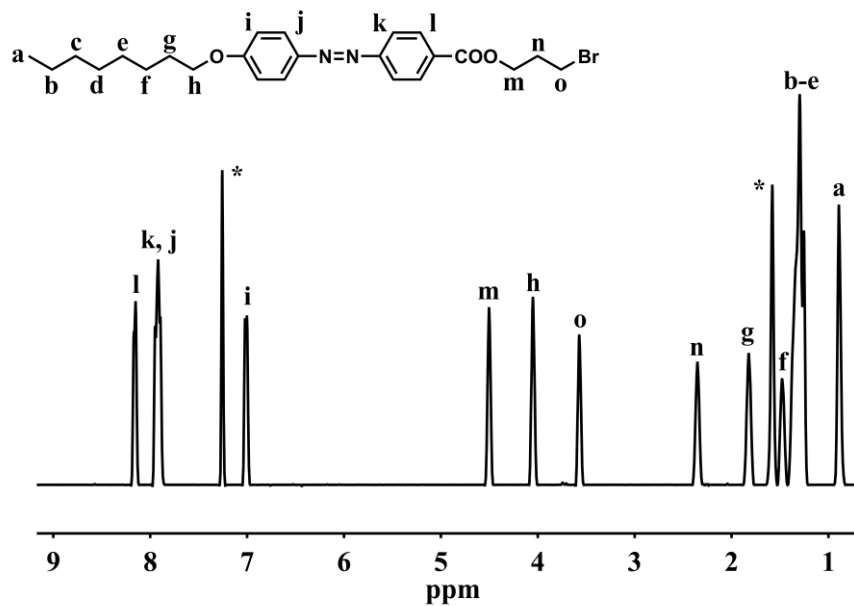
**Figure S5.** POM images of (a) ILCPU0.25, (b) ILCPU 0.50, (c) ILCPU0.75, and (d) ILCPU1.00 at room temperature. POM images of (e) ILCPU0.25, (f) ILCPU 0.50, (g) ILCPU0.75, and (h) ILCPU1.00 at room temperature after cooling from 120 °C.



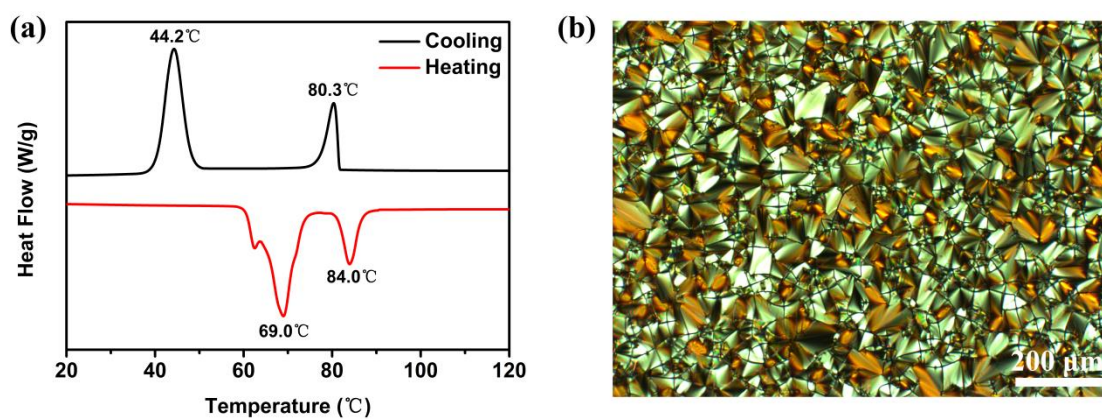
**Figure S6.** Photomicrographs of PU at room temperature under POM with (a) plane-polarized light and (b) cross-polarized light. Photographs of ILCPU0.50 at room temperature under POM with (c) plane-polarized light and (d) cross-polarized light.



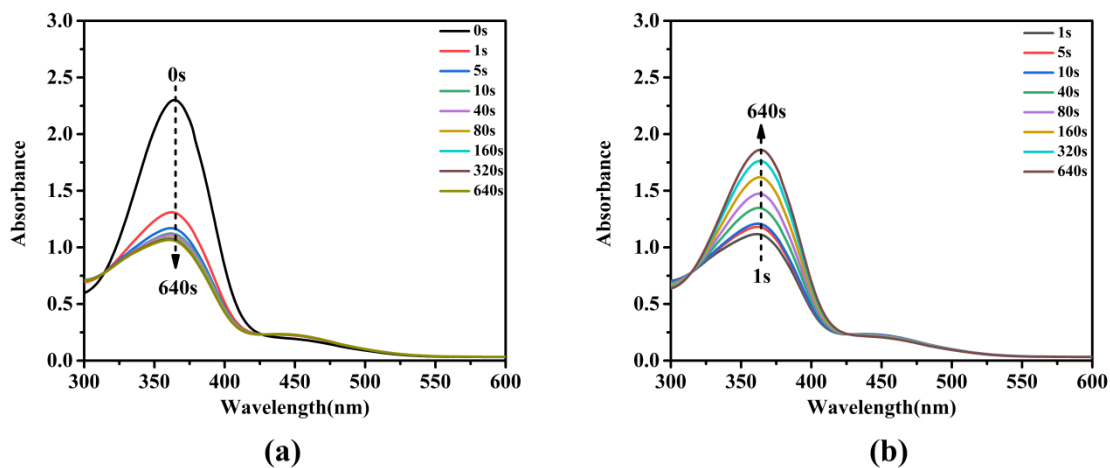
**Figure S7.** TGA curves of PU and ILCPU<sub>x</sub>.



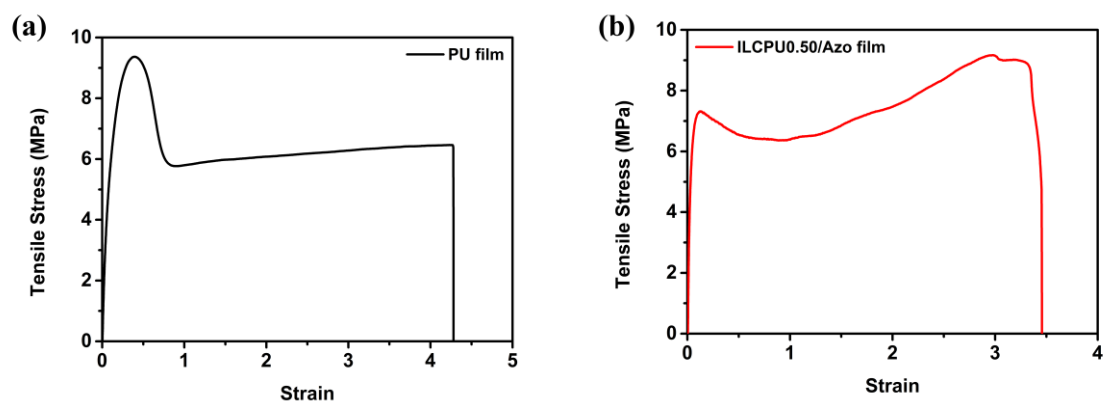
**Figure S8.**  $^1\text{H}$  NMR spectrum (600 MHz,  $\text{CDCl}_3$ , 298 K) recorded for Azo-Br.



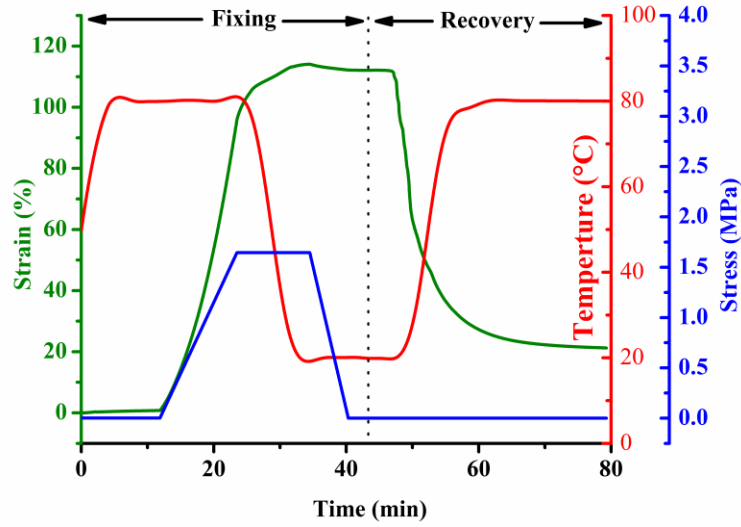
**Figure S9.** (a) DSC curves of Azo-Br on first cooling process and second heating process. (b) POM image of Azo-Br at 75  $^{\circ}\text{C}$  on cooling process.



**Figure S10.** UV-vis absorption spectra of Azo-Br in DMF solution ( $2.0 \times 10^{-6}$  mol/L) upon irradiation with (a) UV light (365 nm) and (b) visible light (450 nm).



**Figure S11.** The stress-strain curves of (a) PU film and (b) ILCPU0.50/Azo film measured at room temperature.



**Figure S12.** Quantitative shape memory curve of ILCPU0.50/Azo film obtained by DMA.

Dynamic mechanical analysis (DMA) was used to quantitatively analyze the shape memory effect of ILCPU0.50/Azo film, which was measured on a DMA Q800 (TA Instruments, USA) under controlled-force mode.

The calculus of the shape fixity ratio ( $R_f$ ):

$$R_f = \frac{\varepsilon_d}{\varepsilon_{load}}$$

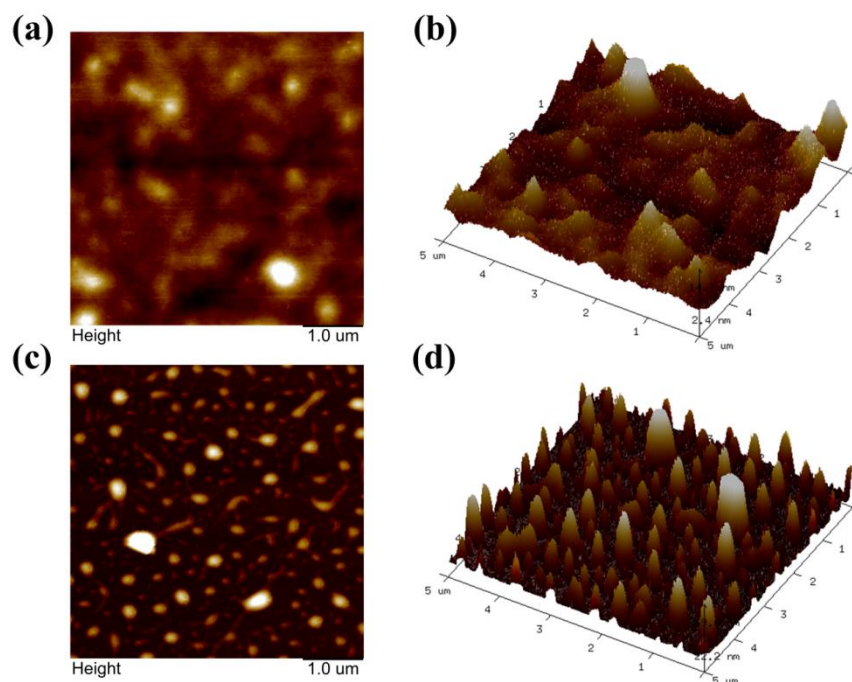
The calculus of the shape recovery ratio ( $R_r$ ):

$$R_r = \frac{\varepsilon_d - \varepsilon_{rec}}{\varepsilon_d}$$

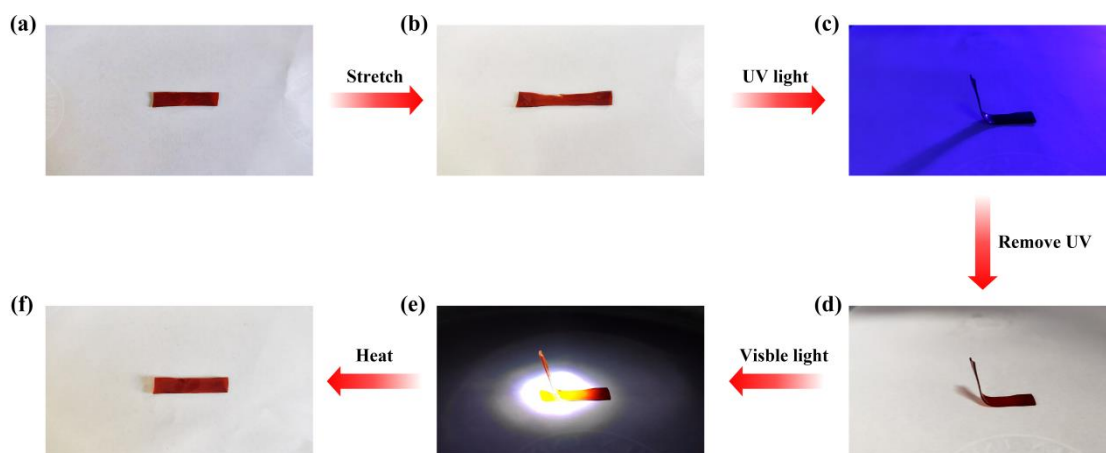
where  $\varepsilon_{load}$  represents the maximum strain under load,  $\varepsilon_{rec}$  is the recovered strain, and  $\varepsilon_d$  is the fixed strain after cooling and restriction removal.

According to the above equations, the shape recovery ratio is about 81.0% and the shape fixity ratio is about 98.2%.





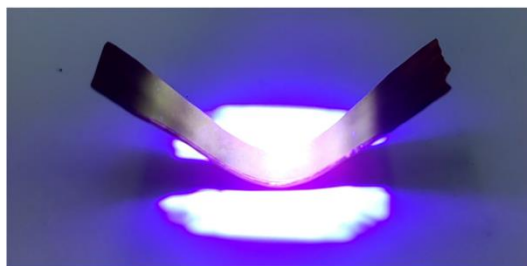
**Figure S13.** 2D (a) and (b)3D AFM phase images of PU film. 2D (c) and (d) 3D AFM phase images of ILCPU0.50/Azo film.



**Figure S14.** Photo-induced triple shape memory effect of ILCPU0.50/Azo film at different stages, (a) original state, (b) stretched state, (c) bent state upon UV light, (d) bent state after removing UV light, (e) bent state upon visible light for ten minutes, and (f) flat state after heating to 80 °C for some minutes.



**(a)**



**(b)**

**Figure S15.** Photographs of the film after recycled and re-formed via mechanical stretching treatment before (a) and after (b) irradiation with UV light (365 nm).