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Supplementary Materials for

Programming a crystalline shape memory polymer network with thermo- and photo-reversible bonds toward a single-component soft robot

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- table S1. Photo-programming parameters for locally defined reversible shape memory materials (white area represents the exposed area of the sample).

Other Supplementary Material for this manuscript includes the following:

(available at advances.sciencemag.org/cgi/content/full/4/1/eaao3865/DC1)

• movie S1 (.mp4 format). Cyclic reversible actuation of the 3D crane (accelerated by 15).



fig. S1. ¹**H NMR spectra of the polymer precursors.** (**A**) Four-armed PCL, the number of repeat unit (n) was calculated from the peak ratio between b and e'. (**B**) Nitro-cinnamate end functionalized PCL, R_c was calculated based on the peak ratio between a and e'. All spectra were collected using 500M (Bruker, Avance III) and CDCl₃ as the solvent.



fig. S2. Differential scanning calorimeter curve of the polyurethane network. The test was carried on a TA Q200 machine with a scan speed of 10 °C/min.



fig. S3. Cyclic photo-programming and photo-erasing performance. (sample $R_c=0.47$, pre-stretch strain=400%). The programming was conducted using 312 nm light (75 mW/cm², 2 min) and the erasing process was performed using 254 nm light (1.3 mW/cm², 5 hours).



fig. S4. Scheme for spatio-selective photo-defining reversible actuation onto a 3D shape. The green and blue areas were active (with actuation) and inactive (without actuation) areas, respectively.

table S1. Photo-programming parameters for locally defined reversible shape memory materials (white area represents the exposed area of the sample).

	Original size	Pre-strain	Exposure time	Photo Patten
Eight-peta lled flower	8 mm ۳ ۳ ۵	100 %	3 min	(three strips, each width is 0.5 mm)
3D "M"	10 mm uu g:2	400%	2 min	25 mm
3D crane	10 mm	300 %	2 min	(three strips, each width is 0.5 mm)
3D elephant	10 mm	300 %	2 min	(three strips, each width is 0.5 mm)