

Supplementary Materials for

A readily programmable, fully reversible shape-switching material

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Published 24 August 2018, *Sci. Adv.* **4**, eaat4634 (2018)
DOI: 10.1126/sciadv.aat4634

The PDF file includes:

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Other Supplementary Material for this manuscript includes the following:

(available at advances.sciencemag.org/cgi/content/full/4/8/eaat4634/DC1)

- Movie S1 (.mp4 format). A programmed LCE folds and unfolds during thermal cycling.
- Movie S2 (.mp4 format). A square peg falling through a circular hole.

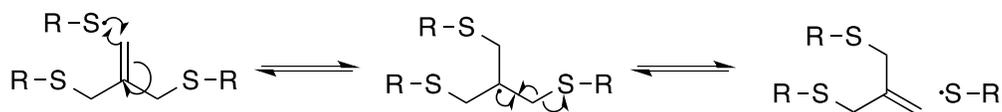


Fig. S1. Mechanism of allyl sulfide exchange. The process is initiated by radicals generated by photolytic processes.

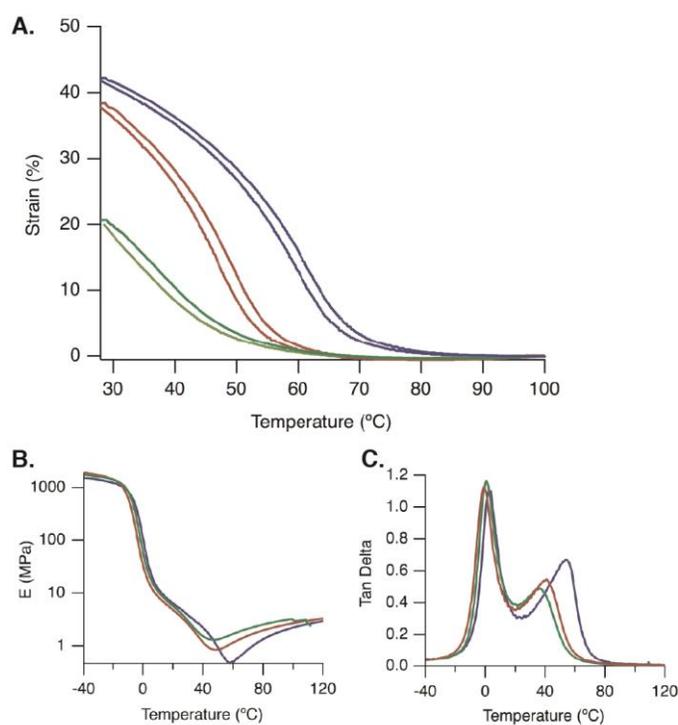


Fig. S2. Thermomechanical analysis of LCEs. A) Thermal cycling of programmed monodomain LCEs with varying amounts of neopentyl glycol diacrylate. Equivalents of RM82, NPGDA, and Allyl dithiol shown as follows; 1: 0.5: 2.35 (blue), 1: 0.75: 1.575 (red), and 1: 0.9: 1.71 (green). B and C) Dynamic mechanical analysis of the same unprogrammed, polydomain LCE. Storage modulus (B) and tan delta (C) were measured at 1 Hz and 3°C/ min.

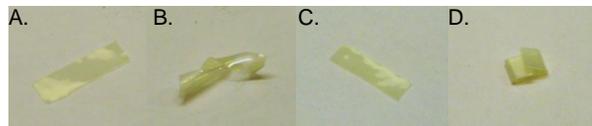


Fig. S3. Reprogramming of the LC shape. Beginning with a flat polydomain LCE (A), AFT programmed the twisted structure in (B). This was programmed by molding into the twisted shape and irradiating with $50\text{-}100\text{ mW/cm}^2$ of $320\text{-}500\text{ nm}$ light for 10 s . Heating to 100°C coupled with a second 10 s light exposure reprogrammed the flat shape (C). A second, folded shape was then programmed with light exposure ($50\text{-}100\text{ mW/cm}^2$ of $320\text{-}500\text{ nm}$ light for 60 s) demonstrated the reconfigurability from Fig 1B in more complex shapes.

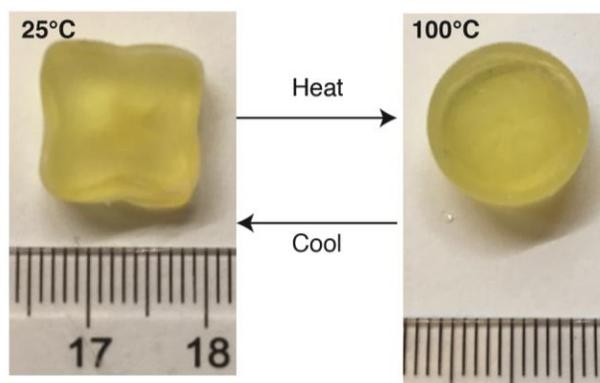


Fig. S4. A square peg morphs into a round peg when heated into the isotropic phase. A 3 mm thick round peg was photopolymerized in a round vial at 120°C . After removal, the round peg was molded by hand into a square peg and programmed with $320\text{-}500\text{ nm}$ light for 10 minutes (30 mW cm^{-1}) with occasional flipping. After, programming the programmed LCE repeated switched between shapes during thermal cycling. Movie S2 demonstrates this sample falling through a circular hole.

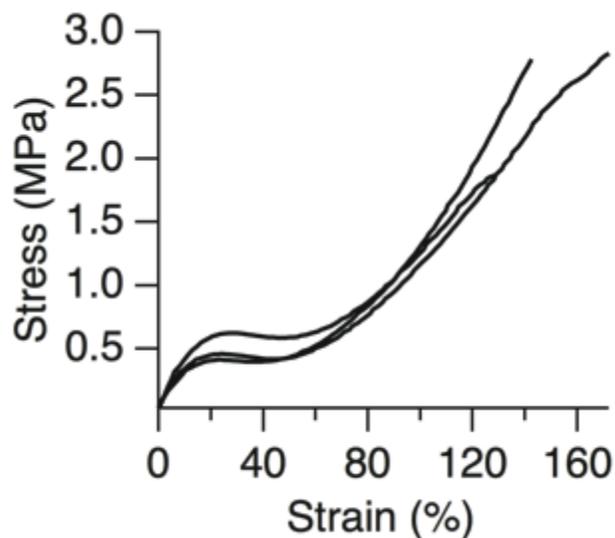


Fig. S5. Strain to break experiments for 1:0.5:1.35 RM82/NPGDA/allyl dithiol. Conditions: $3.14 \times 0.25 \times 6$ mm was stretched at 0.1 mm/s until slip or break on a TA RSA-G2. Experiment was run in triplicate.



Fig. S6. Polarized optical microscopy of unprogrammed and programmed LCEs. A) Polydomain, “as polymerized” LCE observed through POM. B) AFT programmed sample observed at 45° . Programming was down with 50 mW/cm^2 of 320-500 nm light for 100s after stretching of a 250 micron thick film to a monodomain by hand. C) Same monodomain sample shown at a 0° angle to crosspolarizer.

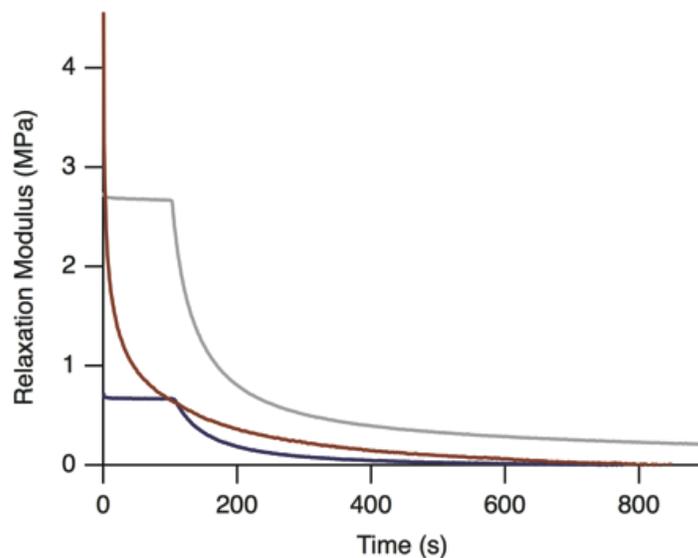


Fig. S7. Stress relaxation behavior of LCE at 120°C (gray), 67°C (blue), and 25°C (red). Conditions: 10% strain, light on at 100 s (30 mW cm^{-2} , 320-500 nm).

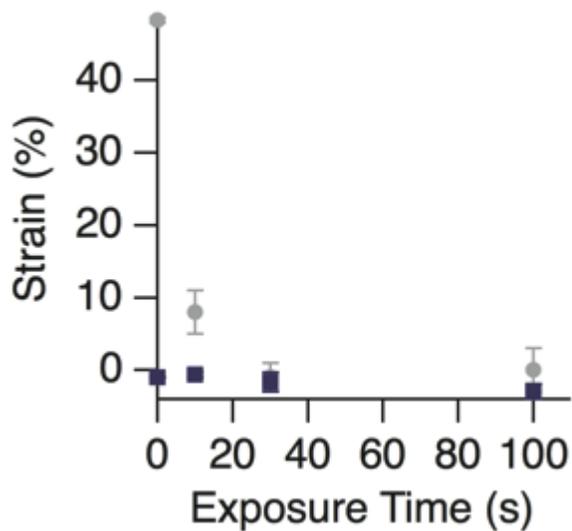


Fig. S8. Heat coupled with light-erased programmed LC strain. Samples were programmed with 100 s, 365 nm, 30 mW cm^{-2} . Programmed samples were erased by heating to 100°C and irradiated with 320-500 nm 30 mW cm^{-2} . The LC (●) and isotropic (■) phase shape was then measured as described above

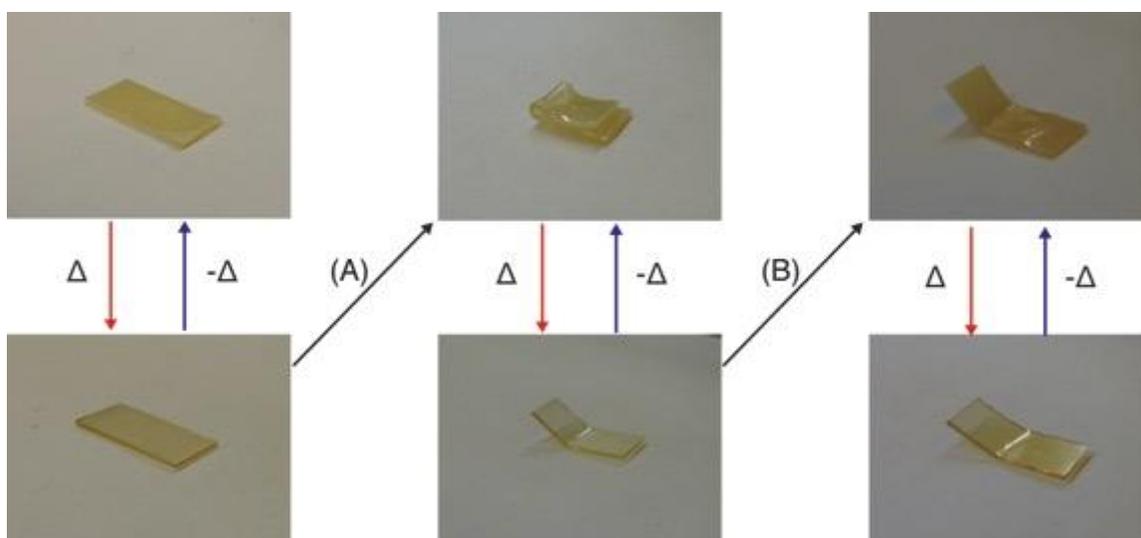


Fig. S9. Stress relaxation in the isotropic shape with and without load. (A) Sample was fixed in folded state and programmed (365 nm, 50 mW cm⁻², 50 s). (B) Same sample was heated unconstrained to 120°C and reprogrammed (365 nm, 50 mW cm⁻², 150 s).

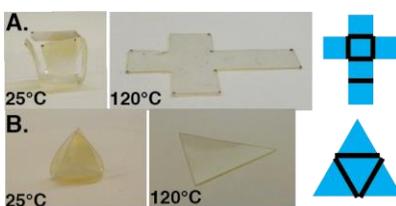


Fig. S10. Folded structure developed by programming each face separately. A box (A) and pyramid (B) programmed by stretching the LCE perpendicular to the fold (20% strain) followed by irradiation in the corresponding patterns shown on the right. The sample was irradiated at room temperature (365 nm, 50 mW/cm²) on one face through the depicted photomask then irradiated without the photomask on the opposite face.

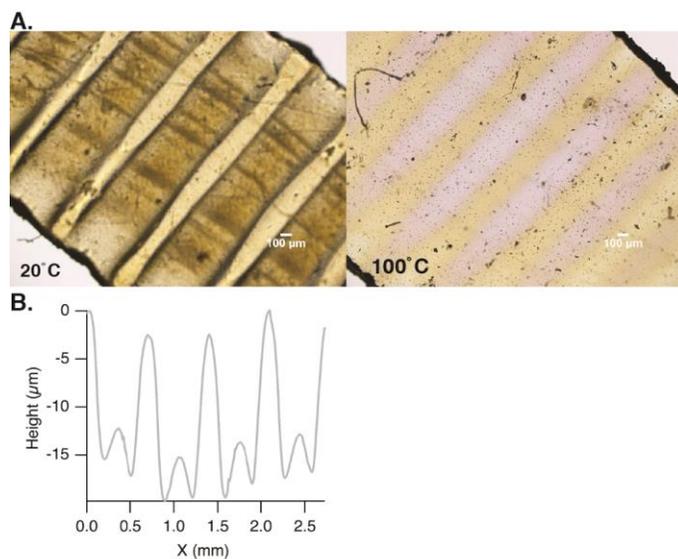


Fig. S11. Patterned alignment results in surface topography. A) Patterned alignment seen through microscopy at ambient temperature and 100°C. Scale bar is 100 microns. B) Surface profile of the patterned alignment at ambient temperature. The complex stresses that develop between the aligned and polydomain regions manifest as buckling on the surface.

Movie S1. A programmed LCE folds and unfolds during thermal cycling. When placed under a heat lamp, the miura unfolded and folded during cooling after removal of heat source. Speed 4x.

Movie S2. A square peg falling through a circular hole. The square, polymerized as a cylinder, was programmed by hand into a square peg followed by irradiation with UV light for 10 minutes. During heating, the square peg morphs into a cylinder allowing it to simply fall through the hole and after cooling, returns to a square. Speed: 8x.