Supporting information for

Three-Dimensional Thermochromic LCE Structures with Reversible Shape-Morphing and Color-Changing Capabilities for Soft Robotics

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Supplementary Note 1

Complementary to the measurements reported in Figure 2, an Atomic Force Microscope was utilized to statistically assess the local modulus over a range of temperatures (Asylum Research MFP-3d with Polymer Heater accessory and a BudgetSensors Tap150Al-G cantilever). Arrays of 10x10 indentations were acquired to a peak load of 200 nN over an area of 54 µm 54 µm on a slide (the resulting 6 µm period ensures noninteracting measurement volumes to best statistically assess the surfaces). The modulus was calculated for each of the resulting 100 force-distance curves based on the conventional Hertz contact mechanics model, assuming a conical indenter ("Sneddon approximation") with a half angle of 19° according to the silicon probe specifications. Such measurements were then repeated for a range of sample temperatures, spanning 25 °C (room temperature) to 95 °C in steps of 10 °C with 15 minutes of equilibration time at each temperature step. The data normalized by the mean signal at room temperature is the most illustrative for demonstrating the temperature dependent mechanical properties (**Figure S3**). Specifically for the pristine (a) and red-colorless (c) specimens, a nearly uniform decrease in the log of these relative moduli (i.e. the average contrast) is clearly apparent. However, there is one key exception for the red-colorless specimen at 45 °C and 55 °C (rectangular overlay), where the relative modulus appears to be similar instead of markedly decreasing with temperature as it does for all other temperature steps. The mean of each 10x10 array of normalized measurements per specimen and temperature step (b), with error bars depicting the standard error (which is smaller than the data points for all but 25 and 35 °C), confirms the generally exponential decrease in modulus vs. temperature as expected. But again, compared to the pristine sample, there is a pronounced deviation for the red-colorless (r-c) specimen exactly at the transition temperature (oval overlay). Note that the modulus results are all presented with the same log scale relative to the mean signal at room temperature, and over identical length scales, as indicated for the highest temperature figure panels (lower right of a and c).

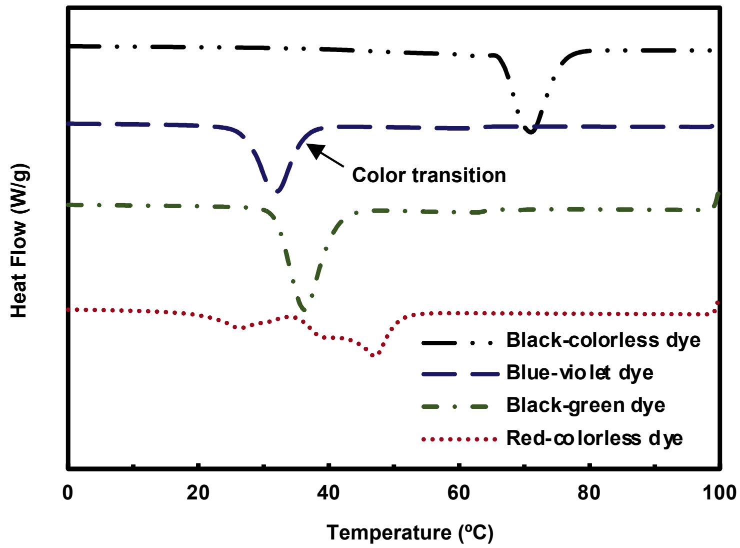

Figure S1. DSC curves of pure thermochromic dyes (black-colorless, blue-violet, black-green, and red-colorless).

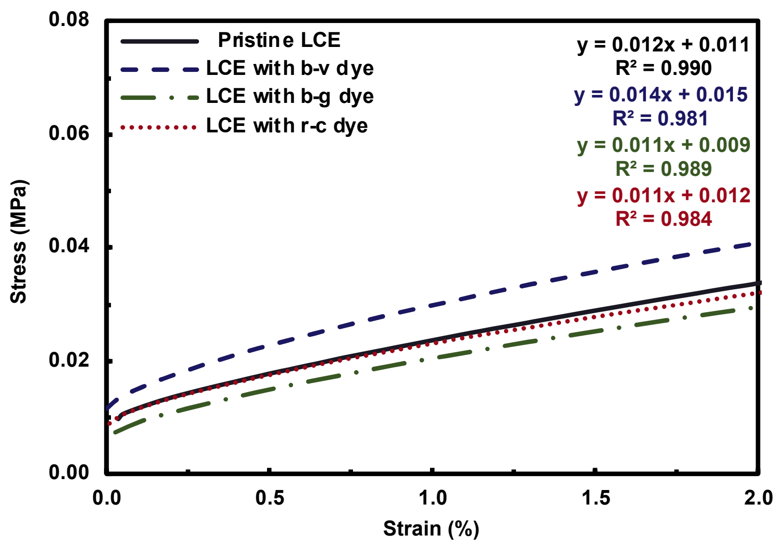

Figure S2. The linear regime of the stress-strain curves (from 0 to 2% of strain).

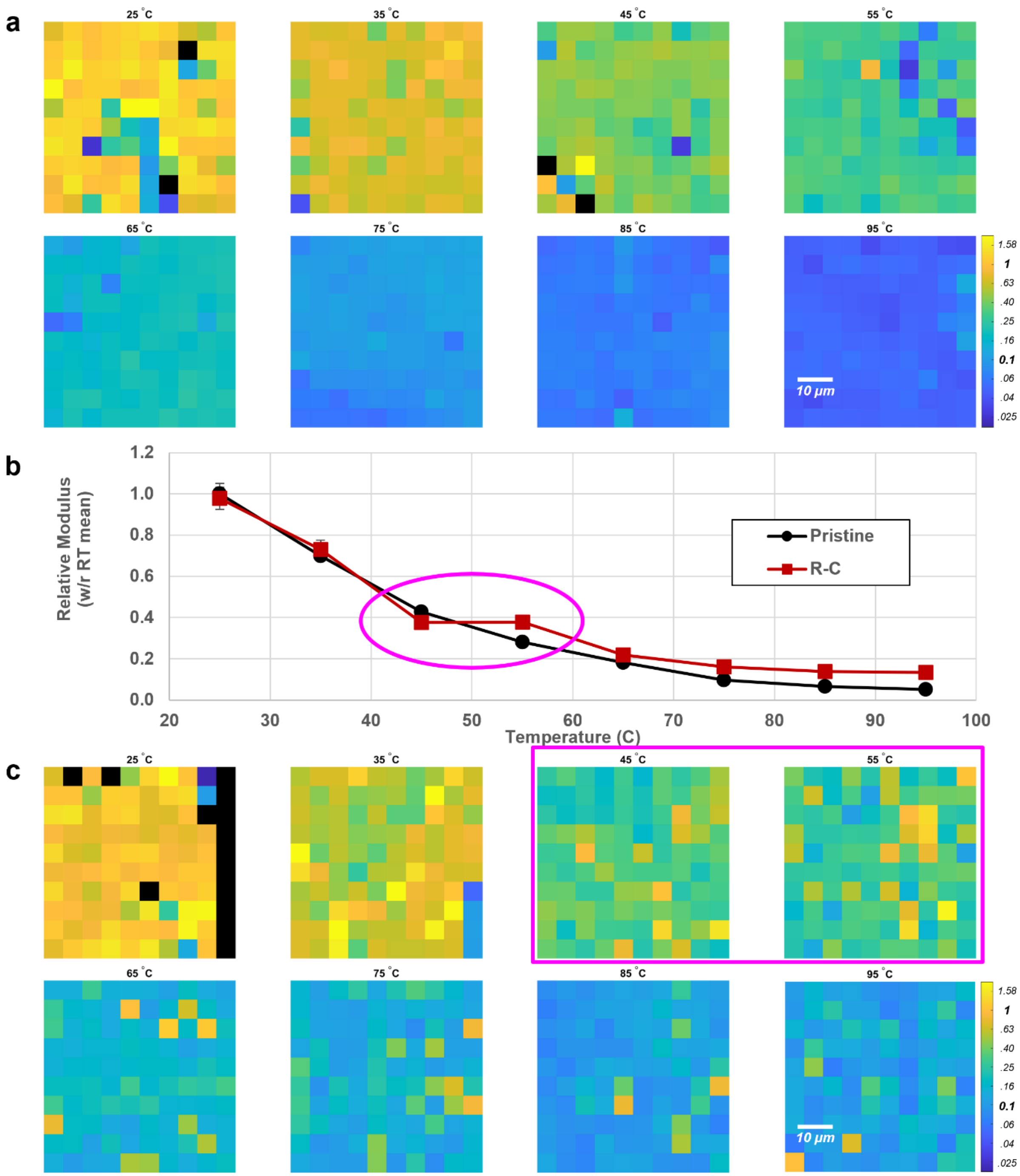

Figure S3. (a) Modulus map for pristine LCEs at various temperatures. (b) Relative modulus as a function of temperature for pristine LCEs and LCEs embedded with red-colorless dye. (c) Modulus map for LCEs embedded with the red-colorless dye. Please note that the modulus value is normalized by the mean modulus at room temperature.

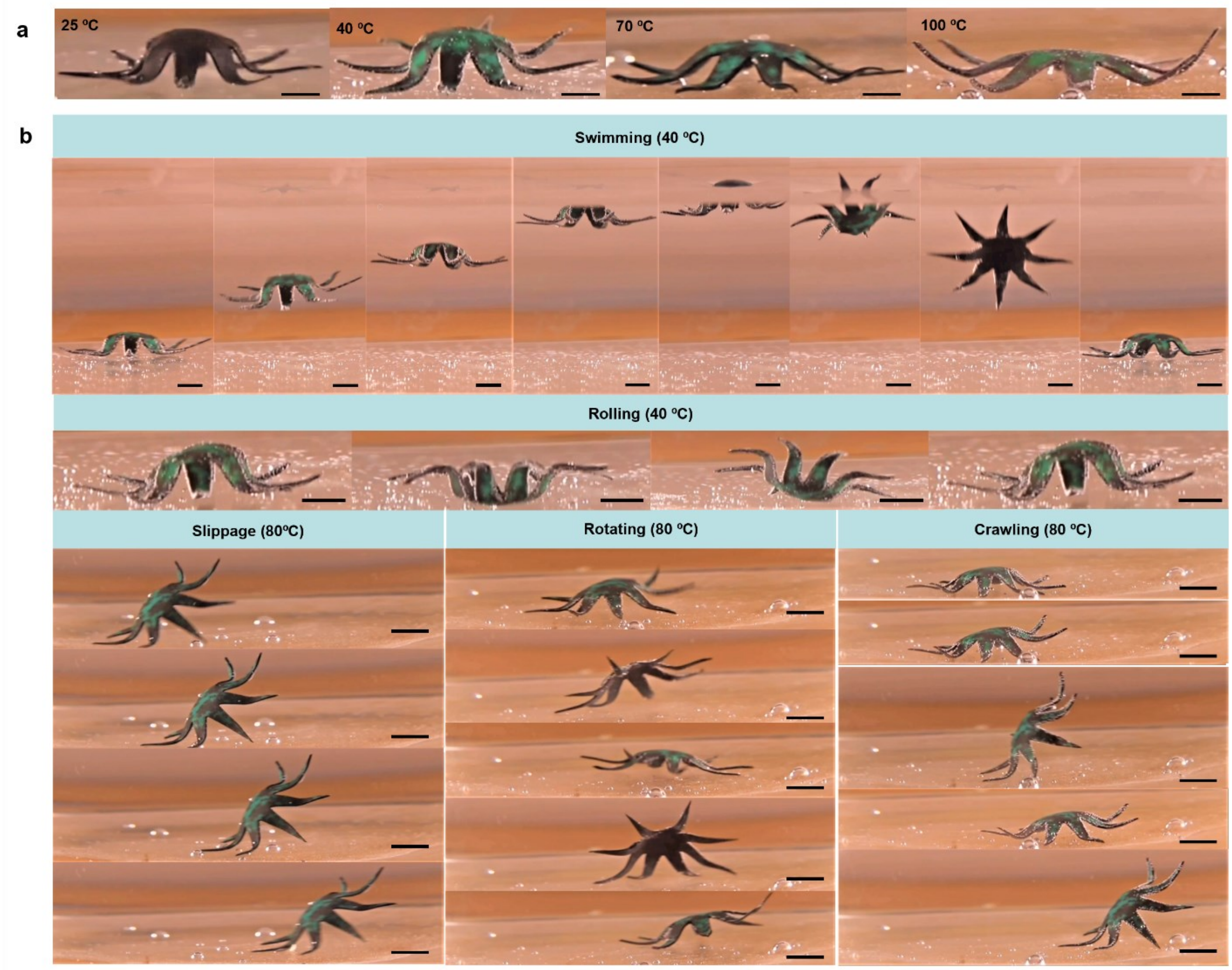

Figure S4. Ferromagnetic shape-morphing and color-changing thermochromic LCE robot for underwater applications. (a) The reversible shape-switching and color-changing behaviors of the robot as the underwater temperature increases. Scale bars: 3 mm. (b) Diverse motion modes of the robot include swimming, rolling, slippage, rotating, and crawling, under thermal and magnetic actuation. Scale bars: 3 mm.

Captions for supplementary movies

Supplementary movie S1: Reversible shape-switching and color-changing behaviors of thermochromic LCE structures that resemble a flower, butterfly, and chameleon under thermal stimulation.

Supplementary movie S2: Reversible shape-switching and color-changing behaviors of a bilayer thermochromic and ferromagnetic LCE structure under thermal stimulation.

Supplementary movie S3: Magnetic actuation of a bilayer “octopus” structure underwater at two different temperatures of 25 ºC and 85 ºC.

Supplementary movie S4: Adaptive motion of the bilayer “octopus” structure at two different temperatures of 25 ºC and 85 ºC.

Supplementary movie S5: Thermo-magnetic dual responsiveness of the bilayer “octopus” structure for various motion modes and camouflage behaviors.