**Supplement: 3D printed reversible shape changing soft actuator assisted by liquid crystal elastomer**

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S1. Fabrication procedure of the ‘hands-free’ monodomain LCEs

The following steps follow the chemistry to a previous study$^1$, while video demonstration of the synthesis procedure has also been published$^2$. Overall, the chemistry relies on an excess of acrylate functional groups to allow for stretching and photo-crosslinking after the initial polydomain LCE has been synthesized. For this chemistry, a 1.15:1 ratio of acrylate to thiol functional groups is used. Within the thiol groups, 13% of the thiol groups belong to the tetra-functional crosslinker to enable an elastomeric material at room temperature with high degree of failure strain and actuation$^3$.

1. 8g of diacrylate mesogen powder RM257 is dissolved in 2.48g (31 wt%) toluene and heated to 80°C in the oven. The RM257 powder can be fully dissolved in ~10 min.

2. Cool the dissolved RM257 solution to room temperature (20°C). 1.92g (24 wt%) of dithiol flexible spacer 2,2’-(ethylenedioxy) diethanethiol (EDDET) and 0.384g (0.048 wt%) of tetra-functional thiol crosslinker pentaerythritol tetrakis (3-mercaptopropionate) (PETMP) are in turn added into the dissolved RM257 solution. After this step, recrystallization of RM257 may usually occur. In this circumstance, we need to place the solution back to the 80°C oven until the RM257 is fully dissolved again. Then we cool the solution to room temperature before the next step.

3. Add 0.048g of photoinitiator (2-hydroxyethoxy)-2-methylpropiophenone (HHMP) into the above solution.

4. Prepare a separate solution where the catalyst DPA is diluted in the toluene at a weight ratio of 1:15. Add 0.356g (0.0445 wt%) diluted catalyst solution into the above monomer solution. Mix the solution on a Vortex mixer for 1 min.

5. Place the mixed solution in the vacuum chamber at 20°C and 508mmHg for 2 min to remove all the air bubbles.

6. Pour the solution into the pre-prepared rectangular mold and leave it at room temperature. The solution is found to be cured in about 1 hour.

7. Place the cured LCE in the vacuum chamber oven at 80°C and 508mmHg for 12 hr to evaporate the toluene. Once this step is done, a polydomain LCE film with an opaque appearance is obtained.
8. Cut the polydomain LCE film into thin strips (75mm × 6mm × 2mm) and stretch these strips on the MTS machine to achieve a 150% strain.

9. Irradiate the stretched LCE strips in the UV light (wavelength of 300-500 nm and light intensity of 5mW·cm\(^{-2}\)) for 10 min by using a spot UV curing lamp (OmniCure S2000, Lumen Dynamics, Ontario, Canada). Once this step is done, monodomain LCE strips are obtained with the director oriented in the direction of stretching.

S2. DMA results of inkjet printed materials

![DMA results of inkjet polymer materials](image)

__Fig.S1 DMA results of the inkjet polymer materials (Tangoblack and Verowhite).__

Dynamic mechanical analysis (DMA) tests were performed on a dynamic mechanical analyzer (Model Q800, TA Instruments, New Castle, DE, USA) in the uniaxial tension mode. A preload of 1mN was applied on the sample (dimension 10mm×3mm×1mm) and the strain was oscillated at a frequency of 1Hz with a peak-to-peak amplitude of 0.1%. Samples were first heated to 90 °C (60 °C for the Tangoblack) and stabilized for 10 min to reach thermal equilibrium. The temperature was then decreased from 90 °C (60 °C for the Tangoblack) to -30 °C (-50 °C for the matrix material) at a rate of 2 °C/min. The temperature dependence of the storage modulus and tan\(\delta\) are shown in Fig. S1. The glass transition temperature (\(T_g\)) is identified as the temperature corresponding to the peak of the tan\(\delta\) curve. The \(T_g\)s of the fiber and the matrix material are ~60 °C and ~0 °C respectively.

S3 SEM image of the cross section of active hinge
To inspect the printing quality of the conductive ink, field emission scanning electron microscope (SEM) images of the interface between the cured silver ink and Tangoblack substrate were taken using a Hitachi FE-SEM machine (SU8010, Hitachi Ltd, Chiyoda, Tokyo, Japan). It is seen that the cured ink and substrate are well connected and no damage is created in the substrate.

**S4 Cyclic test of active hinge**

To ensure the repeatability of the active hinge, resistance variation of conductive wire within 100 bending cycles was tested on a universal material testing machine (MTS CriterionTM Model 41, MTS Systems Corp., Eden Prairie, MN, USA) in a displacement-controlled mode with a 50N load cell. The top view of the sample is shown in Fig. S3a, which assembles the hinge shown in Fig. 3b except that no LCE strip is bonded on the substrate. The sample was clamped on the fixtures at both ends and connected to a programmable multimeter (Keithley2100, Keithley Instruments, Cleveland, OH, USA) controlled by a LabVIEW data acquisition program to measure the resistance variation. In
the experiment, a cyclic displacement with an amplitude of 8mm and frequency of 0.5Hz was applied on the sample to mimic the cyclic bending of the active hinge. The initial and deformed configurations of the hinge are respectively shown in Fig. S3b and S3c. It is seen from Fig. S3c that when the maximum displacement of 8mm was applied, a ~120° bending angle was achieved by the hinge. The displacement and corresponding resistance variation within 100 cycles were measured and shown in Fig. S3d. Only very slight resistance variation was observed, which verifies that the repeatability of the hinge is promising within hundreds of cycles.

S5. Theoretical model

In order to assist the structural design, a theoretical model that accounted for the thermal equilibrium and shrinkage-induced bending is developed. For the thermal part, energy equilibrium relation is constructed by assuming that the energy absorbed by the sample is equal to the Joule heat produced by the conductive wire subtracting the dissipated energy induced by the convection between the sample and the ambient environment. The governing equation and initial condition of the heating process are given by

\[
\rho cV \frac{dT}{dt} = I^2 R - h(T - T_{\text{amb}})S_{\text{conv,ctio}}, \quad 0 < t < t_1 \\
T(0) = T_{\text{amb}}
\]

where \( \rho \) is the density of the sample, \( c \) is the specific heat, \( V \) is the volume, \( T \) is the temperature, \( t \) is the time, \( I \) is the input current, \( R \) is the resistance, \( h \) is the convection coefficient between sample surface and ambient air, \( T_{\text{amb}} \) is the ambient temperature, \( S_{\text{conv,ctio}} \) is the convection surface area, \( t_1 \) is the time duration of applied current. By solving Eq. (1), the temperature of the sample can be obtained by

\[
T(t) = T_{\text{amb}} + \frac{I^2 R}{hS_{\text{conv,ctio}}} \left(1 - e^{-\frac{t}{\tau}}\right), \quad 0 < t < t_1
\]

where \( \tau = \frac{\rho cV}{hS_{\text{conv,ctio}}} \)

Following the similar procedures, the temperature variation during the cooling process can be obtained by

\[
T(t) = T_{\text{amb}} + (T_1 - T_{\text{amb}}) e^{-\frac{t}{\tau}}, \quad t_1 < t < t_2
\]
The above described temperature variation will correspondingly induce the length change of LCE incorporated in the active hinge. The temperature-dependent shrinkage strain of LCE was measured on the DMA tester from 20°C to 100°C under the heating rate of 5°C/min. In our experiment, three different monodomain LCE samples were used to measure the actuation strain. Since the test results were very close, one curve was selected to fit the three parameters. To describe the relationship between the shrinkage strain and temperature in the model, an exponential-type function

\[ \varepsilon_{\text{shrinkage}} = \frac{A}{1 + \exp[-B(T - T_r)]} \]  

is employed to fit the experiment curve, where A, B and T are the fitting parameters. For the fitting curve shown in Fig.S4, we have A=0.452 ± 0.00105, B=0.0865 ± 0.000446 and T =74.6 ± 0.0793.

The length change of LCE will then induce the overall bending of the active hinge. In our model, the hinge is simplified as a bi-layer laminate where an LCE strip is bonded onto a Tangoblack substrate. Based on the beam theory, the bending curvature \( \kappa \) is determined by the modulus of LCE and Tangoblack substrate \( (E_{\text{LCE}} \text{ and } E_{\text{Tangoblack}}) \), the thicknesses of the two layers \( (H_{\text{LCE}} \text{ and } H_{\text{Tangoblack}}) \) and the shrinkage strain of LCE, \( \varepsilon_{\text{shrinkage}} \). The analytical solution is given by

\[ \kappa = \frac{6\varepsilon_{\text{shrinkage}}(1+m)^2}{(H_{\text{LCE}} + H_{\text{Tangoblack}})\left[3(1+m)^2 + (1+mn)\left(m^2 + \frac{1}{mn}\right)\right]} \]  

where \( m = H_{\text{LCE}} / H_{\text{Tangoblack}} \) and \( n = E_{\text{LCE}} / E_{\text{Tangoblack}} \).

Once the bending curvature is obtained, the bending angle \( \theta \) of the hinge can be calculated as

\[ \theta = \kappa L \]
where $L$ is the hinge length.

**S6 Comparison of the silver wire layout**

![Images of conductive wire layouts and thermal images](image1)

**Fig.S5** Effect of the conductive wire layout on the heat generation. (a) Top view of the crawler with conductive wire of straight line design. (b) Top view of the crawler conductive wire of serpentine design. (c) Thermal infrared image of the crawler with conductive wire of straight line design. (d) Thermal infrared image of the crawler with conductive wire of serpentine design.

Serpentine design of conductive wires was printed and compared with straight line design. High quality thermal infrared images of different conductive wire designs were taken by using the infrared camera (FLIR i5, FLIR systems Inc., Portland, USA) to measure the temperature field under a given applied power of 2.25W. It is seen that both of the two patterns will generate a relatively uniform temperature field since the spacing of adjacent wires is small. We can imagine that the wire pattern will have an influence on the temperature field when the wire spacing is large enough.

**References**