# Electronic Supplementary Information 

## 3D Printing Programmable Liquid Crystal Elastomer Soft Pneumatic Actuators

Wei, Liao and Zhongqiang Yang*a, b
${ }^{\text {a }}$ Key Laboratory of Organic Optoelectronics \& Molecular Engineering of the Ministry of Education, Department of Chemistry, Tsinghua University, Beijing 100084, China.
${ }^{\mathrm{b}}$ Laboratory of Flexible Electronics Technology, Tsinghua University, Beijing 100084, China.

E-mail: zyang@tsinghua.edu.cn

This file includes:
Fig. S1 to S19
Legends for Movie S1 to S6

## Figures



Fig. S1. The length deformation ( $L / L o$ ) of LCE tubes obtained by different printing speeds at room temperature $(L o)$ and at $140^{\circ} \mathrm{C}(L)$.


Fig. S2. A) 2D-WAXD patterns of LCE tubes obtained at different printing speeds. B) The relationship between order parameters $(f)$ of the LCE tubes and their printing speeds.

The order parameter $(f)$ along the longitudinal direction was calculated based on the HermansStein orientation distribution function and shown below (Equation 1 and 2).

$$
\begin{aligned}
& f=\frac{\left.3<\cos ^{2} \varphi\right\rangle-1}{2} \\
&<\cos ^{2} \varphi>= \int_{0}^{\frac{\pi}{2}} I(\varphi) \sin \varphi \cos ^{2} \varphi d \varphi \\
& \int_{0}^{\frac{\pi}{2}} I(\varphi) \sin \varphi d \varphi \text {. } \\
& \text { (Equation 1) } \\
& \text { (Equ) }
\end{aligned}
$$

Where $I(\varphi)$ was a gaussian function to fit the curve of XRD intensity (I) and azimuthal angle ( $\varphi)$.


Fig. S3. POM images of LCE obtained by different printing paths. Scale bar $500 \mu \mathrm{~m}$.


Fig. S4. POM images of boundaries of LCE with three different alignments. The cartoon shows the alignment of the LCE and the position of the photos. The arrow lines in the cartoons indicate the distribution of the alignments. Scale bar 1 cm .


Fig. S5. A) A schematic of preparing samples. The red dashed line boxes indicate the crosssection shapes of the samples. B, C) The displacement-force curves of LCE samples for tensile testing. The cartoon represents the stretching direction and the alignment. The details of the sample size are shown beside the cartoon.

As shown below, Equations 3 and 4 were applied to obtain the strain ( $\Delta L / L \mathrm{o}$ ) and stress, respectively.

$$
\begin{gathered}
\text { Strain }=\frac{\frac{\text { Displacement }}{\text { Length }}}{}(\text { Equation 3) } \\
\text { Stress }=\overline{\text { Crorce }} \text {-section Area } \\
\text { (Equation 4) }
\end{gathered}
$$

As for the sample (Fig. S5B), stretching in the direction perpendicular to the alignment, its cross-section was a rectangular shape. Therefore, its cross-section area $\left(A_{\perp}\right)$ can be calculated from the equation below.
$A_{\perp}=W \times T$
In which $W$ and $T$ refer to the width and thickness of the sample, respectively.

Then, $A_{\perp}=2.57 \mathrm{~mm} \times 0.101 \mathrm{~mm}=0.25957 \mathrm{~mm}^{2}$

As for the sample (Fig. S5C), stretching in the direction parallel to the alignment, its crosssection was a fan shape. Therefore, its cross-section area $\left(A_{\|}\right)$can be calculated from the equation below.
$A_{\|}=A_{\text {out }}-A_{\text {in }}=\frac{1}{2} \theta R_{\text {out }}^{2}-\frac{1}{2} \theta R_{\text {in }}^{2}$
In which $A_{\text {out }}$ and $A_{\text {in }}$ indicate the cross-section area of the outer and inner sectors; $R_{\text {out }}$ and $R_{\text {in }}$ are the radii of the outer and inner sectors; $\theta$ is the angle of the sectors.
As the ink was printed on a cylinder substrate to form the LCE tube, the $R_{\text {in }}$ was equal to half of the diameter of the substrate $\left(D_{\mathrm{o}}\right), R_{\mathrm{in}}=D \mathrm{o} / 2$. Therefore, $R_{\mathrm{out}}=R_{\mathrm{in}}+T$ ( $T$ - thickness of the sample). The arc length of the outer sector ( $L_{\text {out }}$ ) was measured to be the width of the sample ( $W$ ), so
$\theta=\frac{L_{\text {out }}}{R_{\text {out }}}=\frac{W}{\frac{D_{o}}{2}+T}$
Then,

$$
\begin{aligned}
A_{\|} & =\frac{1 W}{\frac{2 D_{o}}{2}+T}\left(\frac{D_{o}}{2}+T\right)^{2}-\frac{1 W}{\frac{2 D_{o}}{2}+T}\left(\frac{D_{o}}{2}\right)^{2} \\
& =\frac{1}{2} W\left(\frac{D_{o}}{2}+T\right)-\frac{1 W}{\frac{W D_{o}}{2}+T}\left(\frac{D_{o}}{2}\right)^{2} \\
& =\frac{1}{2} \times 2.40 \mathrm{~mm} \times\left(\frac{3.00 \mathrm{~mm}}{2}+0.099 \mathrm{~mm}\right)-\frac{1}{2} \times \frac{2.40 \mathrm{~mm}}{\frac{3.00 \mathrm{~mm}}{2}+0.099 \mathrm{~mm}} \times\left(\frac{3.00 \mathrm{~mm}}{2}\right)^{2}
\end{aligned}
$$

$$
\approx 1.91880 \mathrm{~mm}^{2}-1.68856 \mathrm{~mm}^{2}=0.23024 \mathrm{~mm}^{2}
$$

Increasing Pressure


Fig. S6. Images of a contracting LCE-SPA with longitudinal alignment inflated at different pressures.


Fig. S7. Images of an elongating LCE-SPA with circumferential alignment inflated at different pressures.


Fig. S8. Images of a bending LCE-SPA with combined longitudinal and circumferential alignment inflated at different pressures.

## Bending



Figure S9. The define of bending angle ( $\alpha$ ).


Fig. S10. The distortion of LCE tubes with different alignments before and after removing from the substrate.

Two red points was marked at the two sides of LCE tubes for observation.

After removing from the substrate, the length of LCE tubes with longitudinal alignment increased ( $+1.5 \pm 0.4 \%$ ).
After removing from the substrate, the length of LCE tubes with circumferential alignment decreased ( $-1.6 \pm 0.2 \%$ ).


Fig. S11. Images of a twisting LCE-SPA with torsional alignment inflated at different pressures.

## Twisting


$\beta$


Fig. S12. The define schematic of twisting angle $(\beta)$.


Fig. S13. The soft gripper. A) Schematic of the gripper. B) The gripper performed grabbing function before and after inflation at 80 kPa .


Fig. S14. The twisting angle of an LCE-SPA with torsional alignment upon inflation and deflation. 80 kPa inflating pressure was applied to actuate the actuator at the frequency of 0.5 Hz.


Fig. S15. Thermal deformation of LCE-SPAs shown in Figure 2. The arrow lines in the cartoons indicate the distribution of the alignments. Scale bar 1 cm .


Fig. S16. Thermal deformation of LCE-SPAs shown in Figure 3. The arrow lines in the cartoons indicate the distribution of the alignments. The scale bar 1 cm .


Fig. S17. The relationship between the shear viscosity and the shear rate of the LCE ink at 65 ${ }^{\circ} \mathrm{C}$.


Fig. S18. The printing path of different alignments. The diameter $(D)$ of the rotating substrate is 3 mm , and $\pi D$ indicate the circumference of the rotating substrate.


Fig. S19. Scheme of preparing LCE-SPAs.

## Legends for Movies

Movie S1. The deformation process of LCE-SPAs with the alignment of bending and twisting motions at the top view.

Movie S2. Lifting and lowering loads by a contracting LCE-SPA after inflation and deflation.

Movie S3. Lowering and lifting loads by an elongating LCE-SPA after inflation and deflation.

Movie S4. Grabbing and lifting a basket containing cereals by a soft gripper.
Movie S5. Mixing water and ink in the beaker by a mixer.
Movie S6. Printing process of different paths.

