Electrically Controlled Soft Actuators with Multiple and Reprogrammable Actuation Modes

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New forms of soft actuators have enabled the construction of novel soft robots with various functionalities. Though previous researches have successfully fabricated soft actuators with diverse actuation modes, their actuation capabilities are often fixed once their fabrication is completed. Herein, an electrically controlled soft actuator with multiple and reprogrammable actuation modes is designed and fabricated. The soft actuator is composed of two layers of disulfide liquid crystal elastomer (ss-LCE) film with embedded resistive heating wires of serpentine shape. The actuation mode of the actuator can be programmed by introducing a certain alignment of liquid crystal mesogens in the ss-LCE film, which is achieved by the controlled deformation of the actuator at room temperature and the rearrangement of the polymer network through disulfide exchange reaction in the material. The actuation mode of the actuator can be easily erased by heating it up to 180 °C, and a new mode of actuation can be introduced by deforming the actuator to a new shape at room temperature. With the reprogrammable and multiple actuation modes, a reusable and general-purpose soft actuator is demonstrated herein which can meet various requirements in constructing new soft active devices.

Soft actuators are of great interest in diverse applications where muscle-like actuations are desired.[1] Advances in soft actuators have enabled the design and fabrication of novel soft robotics,[2–4] biomimetic devices,[5,6] and wearable devices.[7] To meet the requirements of various applications, soft actuators with different actuation modes have been developed. For example, polymer composite fibers[8,9] and twisted fishing line artificial muscles[10] can generate large uniaxial contractions. LCE tubular actuators[11] and self-sensing paper-based actuators[12] can exhibit bending actuation. Shear-vacuum-actuated machines[13] show active shear deformation. However, most previously developed soft actuators can only have one actuation mode, which is fixed once it is fabricated. In this work, by embedding stretchable heating wires into disulfide liquid crystal elastomers (ss-LCEs),[14,15] we have successfully designed and fabricated an electrically controlled soft actuator with multiple and reprogrammable actuation modes. We believe that the ss-LCE-based soft actuator can find its wide applications in constructing different soft active structures and devices.

Thermally driven LCE has been widely recognized as a promising soft actuating material with many unique features such as large actuation stress, strain, and work density.[16–21] The actuation of an LCE originates from the phase transition of the liquid crystal mesogens in the material. Once the temperature of an LCE is increased above the nematic-to-isotropic phase transition temperature, aligned mesogens in the nematic phase transform to isotropic phase, accompanied with significant shape change, as shown in Figure 1a. Such shape change is reversible once the temperature is decreased down to room temperature with mesogens transiting from isotropic phase back to nematic phase. Previous work has demonstrated that various actuation modes of LCE can be achieved by controlling different alignment of mesogens.[22–27] Moreover, our recent work has shown that by introducing disulfide bonds into LCE, its actuation mode can be easily programmed at room temperature through dynamic bond exchange reaction.[15] In this work, we choose to use ss-LCE (see its chemical structure in Figure S1a, Supporting Information) to construct a reprogrammable soft actuator.

As shown in Figure 1a, with the help of disulfide exchange reaction (Figure S1b, Supporting Information), actuation mode of an ss-LCE film can be easily reprogrammed. The sample has to be first heated up to 180 °C for 20 mins and then cooled down to room temperature. After the temperature is lowered to room temperature, mesogens in the LCE are in a polydomain state, and thus its preexisting actuation mode (or alignment of mesogens) is erased. Subsequently, by deforming the ss-LCE into another shape and holding the deformation for 24 h, the new alignment of mesogens can be introduced; thus, the ss-LCE is reprogrammed (Figure S2, Supporting Information). With the heating time of 20 mins at 180 °C and deformation holding time of 24 h, the new alignment of mesogens can be fixed (Figure S3, Supporting Information). No degradation of the actuation performance of a reprogrammed ss-LCE is detected during the reprogramming process (Figure S4, Supporting Information). In addition, the mechanical properties of ss-LCEs at room

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temperature, 100 and 140 °C, can be found in Figure S5, Supporting Information.

In addition to the actuating material, how to apply external stimuli to trigger the actuation is crucial for an actuator in its applications. In the past, different strategies have been adopted to trigger the actuation of an LCE as thermally responsive material. One commonly adopted way to change the temperature of LCE is through controlling the environmental temperature.[28,29] Photothermal effects have also been used to realize remote control of local temperature change in LCE.[10,31] Compared with those heating strategies, Joule heating generated by resistive heating wires embedded in the material has shown...
several advantages including simple control, low cost, and easy integration.\textsuperscript{31} To construct an electronically controllable actuator, in this work, we integrate resistive heating wires of carefully selected shape with ss-LCE films.

To make a soft actuator with great stretchability along an arbitrary direction, we embedded heating wires with half-and-half Peano shape\textsuperscript{32} into the actuator (Figure 1b). The heating wires were fabricated through standard photolithography, as described in the Supporting Information and Figure S6 and S7, Supporting Information. To further enhance the stretchability of the heating wires, we attached the heating wires to an initially biaxially pre-stretched LCE film, and the heating wires were under biaxial compression after the prestretch in the LCE films was removed. Detailed fabrication steps are shown in Figure 1b and also in Experimental Section. With the fabricated actuator, we can program or reprogram it with various actuation modes, as discussed in the following paragraphs.

We first programmed the soft actuator with the contraction mode, which is one of the most important actuation modes for an actuator. The programming process can be found in the Experimental Section. To quantitatively characterize the actuation performance determined by Joule heating of the heating wires, we performed isotonic (constant load) and isometric (constant displacement) tests to measure the actuation strain and stress, as shown in Figure 2a. We first measured the surface temperatures of the actuator with different applied voltages (2, 3, and 4 V for 120 s) using infrared (IR) camera (FLIR E75-42). Figure 2b shows the maximum temperature on the surface of the actuator as a function of time. Once the voltage was turned on, the surface temperature started increasing and then reached a plateau value. The plateau value of the maximal surface temperature increased from 83 to 140 and to 191 °C, when we successively increased the voltage from 2 to 3 and to 4 V. When we turned off the voltage, the surface temperature gradually decreased to room temperature within 180 s. Although a higher voltage resulted in a relatively higher surface temperature with a shorter period, we had to make sure that the actuating temperature of the actuator is below 180 °C to prevent erasing the programmed actuation.

To measure the actuation strain of the actuator, we applied three different levels of voltages (2, 3, and 4 V) for 120 s. The actuator was subjected to 0.196 N axial load (20 g weight) to keep it straight, as shown in Figure 2a. The actuation strain can be defined as \( e = \frac{l - l_0}{l_0} \times 100\% \), where \( l \) is the length of the actuator in the initial state, whereas \( l_0 \) is the length in the actuated state. As shown in Figure 2c, the actuation strain increased and reached the maximal value after 60 s for all three different voltages. Specifically, when the voltage was 3 V, 23% actuation strain could be generated within 45 s. In addition, the response of the soft actuator was faster as we increased the applied voltage. An average strain rate of 0.15%, 0.75%, and 1.5% s\(^{-1}\) could be realized as the applied voltage was 2, 3, and 4 V, respectively. Furthermore, the actuation stress could be measured by fixing the length of the actuator (Figure 2a) while applying different levels of voltage (2, 3, and 4 V), as shown in Figure 2d. Similarly, for a given value of voltage, the actuation stress increased from zero to the maximum value after the voltage was turned on and gradually dropped to zero when the voltage was turned off. Specifically, a maximum actuation stress of 0.2 MPa could be produced by the actuator when the voltage is 3 V. The actuator could also generate cyclic actuation when the voltage was turned on and off cyclically, as shown in Figure S8, Supporting Information. The actuation stress generated by the actuator depended on the magnitude as well as the duration of the voltage when the voltage was turned on.

We next demonstrated the reprogrammability of the actuator. The actuator was first programmed with uniaxial contraction mode along X direction, as shown in Figure 2e. We then heated the actuator to 180 °C in an oven for 20 mins to erase its contractive actuation in X direction and reprogrammed it with contractive actuation along Y direction. We further erased the contractive actuation in Y direction and repeated the aforementioned reprogramming and erasing processes. The active contraction along two orthogonal directions can be alternatively introduced into the actuator, as shown in Figure 2f. Before reprogramming, the actuator programmed for the first time could generate an actuation stress of 0.17 MPa in isometric condition with an applied voltage of 3 V, and its actuation strain under the isotonic condition could reach 28% after the voltage was turned on for 120 s. After each reprogramming, both actuation strain and stress of the actuator maintained almost an unchanged state, indicating its very robust reprogrammability.

We then demonstrated the multimode programmability of the actuator. In the experiment, we programmed an actuator with three different actuation modes: contraction mode, bending mode, and shear mode. The detailed steps of programming an ss-LCE actuator are shown in the Experimental Section. As shown in Figure 3a, the actuator with the contraction mode could lift a weight of 60 g up by 6 mm (80 times of its own weight) with an applied voltage of 3 V. Figure 3b shows its actuation strain as a function of time. It took 45 s for the actuator to reach the actuation strain of 25%, which was comparable with the case shown in Figure 2b with the weight of 20 g.

As shown in Figure 3c, the actuator with bending actuation can be achieved by attaching a strain-limiting layer (Kapton tape, 3M Company) to a contractive actuator. As the voltage was applied to the actuator, bending actuation could be produced, as shown in Figure 3c. The bending angle shown in Figure 3d was measured as a function of time. The actuator was not straight as it was fabricated and had an initial bending angle of 20°. When we applied a voltage of 3 V, the bending angle of the actuator increased from 20° to 160° within 40 s. After we turned off the voltage, its bending angle gradually reduced to its initial value within 180 s.

We also studied the actuator with the shear mode, as shown in Figure 3e. In the experiment, two acrylic plates were attached to the two parallel edges of the actuator. One of the plates was fixed, and the other one was allowed to move horizontally along the glass slider. When the voltage was turned on, large shear deformation could be generated by the actuator (Figure 3e). When the voltage was turned off, the actuator gradually recovered back to its initial shape. In Figure 3f, we show the shear angle of the actuator as a function of time when the voltage was first turned on for 90 s and then turned off for 150 s.

Finally, we showed the possibility of transforming the actuator from one actuation mode to another. We first programmed a pair of actuators with shear mode and then combined them into one actuator to generate bi shear actuation, as shown in Figure 3g.
Figure 2. Characterizations of ss-LCE-based soft actuator. a) Schematics of the experiment setup for the characterization of actuation strain and actuation stress of an ss-LCE actuator. b) Maximum surface temperature of the actuator as a function of time during contraction (voltage on) and recovery (voltage off), the inset is the IR image showing the surface temperature distribution of the actuator when the applied voltage is 3 V at 120 s. The dashed lines in the inset show the area of the actuator. c) Actuation strain of the actuator as a function of time under three different levels of applied voltages: 2, 3, and 4 V. The actuator was subjected to 0.198 N load (20 g weight) to keep the actuator straight. d) Actuation stress of the actuator as a function of time for three different levels of applied voltages: 2, 3, and 4 V. e) Schematics of reprogramming the actuator. Uniaxial contraction along X or Y direction was alternately reprogrammed and erased four times in the same actuator. f) Actuation strain and actuation stress along either X or Y direction as a function of time after the actuator was programmed or reprogrammed.
Figure 3. Multiple actuation modes can be programmed in the actuator. a) Contraction mode of ss-LCE actuator. The actuator could lift a weight of 60 g when a voltage of 3 V was applied to the actuator. b) The actuation strain of the actuator as a function of time. c) Bending mode can be realized by attaching strain-limiting layer (Kapton tape) to a contractive actuator. d) Bending angle (defined in the inset) as a function of time with the applied voltage of 3 V for 40 s. e) Shear mode of the actuator. f) Shear angle (defined in the inset) as a function of time with the applied voltage of 3 V for 90 s. g) A pair of actuators with shear mode could be combined to one actuator to generate the bishear actuation. The actuator with shear mode actuation could be erased and then reprogrammed with contraction mode. This newly generated contractive actuator could lift up a skeleton arm.
The free end of the new actuator was able to pull a spring by 4.8 mm with a voltage of 3 V applied to both two actuators for 90 s. We next took one of the actuators out and reprogrammed it with contraction mode. As shown in Figure 3g, after reprogramming, the actuator could lift up a skeleton arm within 60 s with applied voltage of 3 V. Such significant transformation of actuation mode in an actuator can be very useful in many engineering applications.

In conclusion, we have successfully constructed an ss-LCE-based soft actuator, which exhibits various actuation modes after programming or reprogramming. We have also characterized different actuation modes such as contraction, bending, and shear of the ss-LCE actuator. Most existing soft actuators have fixed actuation modes after they fabricated. In contrast, we have shown that both erasing and reprogramming an actuation mode in the newly developed soft actuator are facile. Moreover, the actuation of the soft actuator can be electrically controlled with a low voltage, which enables its simple integration with most existing control systems with low costs. We hope the newly developed reprogrammable soft actuator can find its wide applications in constructing diverse soft active structures.

Experimental Section

Fabrication of ss-LCE Actuator: We first fabricated heating wires of half-and-half Peano shape on a glass substrate using photolithography (Figure S6, Supporting Information, and Supporting Information for fabrication details). Meanwhile, by following our previous work,[15] we synthesized two ss-LCE thin films with the thickness of ≈0.25 mm and then cut them into a square shape of 4 cm × 4 cm. Details of synthesizing an ss-LCE film can be found in Supporting Information.

We then stretched both ss-LCE films equal biaxially with the stretch ratio ≈1.3 and fixed the stretch by adhering the four edges of each film to an acrylate plate using VHB tapes (4910, 3M company). To attach the heating wires onto the surface of one of the ss-LCE films, we first transferred the heating wires from the glass substrate to a water-soluble tape (Arc-Zone) and then attached the tape onto the ss-LCE film using stick glue. The ss-LCE film together with the water-soluble tape and heating wires was left untouched for 1 day. We next cut out the central area of the ss-LCE film and took it away from the acrylate plate. We then dissolved the water-soluble tape on the surface of the ss-LCE film using deionized (DI) water, while the heating wires still attached on the surface of the ss-LCE film. After the entire film was fully dried, we covered it by the other stretched ss-LCE film on top to obtain a sandwich structure. We then heated up the sandwich structure using a hotplate with a temperature of 120 °C for 5 mins, which caused the biaxial contraction of the entire film and thus increased stretchability of heating wires. After that, we compressed the sandwich structure using two glass slides and multiple clips and then put it in an oven of 180 °C for 20 mins to form bonding between two ss-LCE films. Finally, we took the entire structure out from the oven and cooled it down to room temperature. To connect the actuator with external power supply, we carefully peeled off small pieces of the LCE film to expose the metal contact pads (Figure S7, Supporting Information) of the heating wires and then soldered electrical wirings onto them for the connection of external power supply. The final actuator was of 3 cm × 3 cm with a thickness of ≈0.3 mm. We can then program the actuator as discussed in the following paragraphs.

Programming of ss-LCE Actuator into Different Modes: The programming of actuators with different modes shares a similar procedure: first, the actuator was heated to 180 °C for 20 mins and cooled down to room temperature; then, a certain deformation was applied to the actuator and maintained for 24 h. The heating time of 20 mins and the time for holding the deformation (24 h) were determined through parametric studies (Figure S3, Supporting Information). Detailed information on different modes of programming is described as follows. 1) Contraction mode: we first heated the actuator up to 180 °C and cooled it down. Then, we fixed one end of the actuator using Kapton tape (3M company) and manually applied a uniaxial stretch with the stretch ratio of 1.5 to the actuator. We then fixed the other end using Kapton tape to hold the stretch for 24 h. 2) Bending mode: We first programmed the actuator with a uniaxial stretch of 1.5, as described earlier. Then, we adhered Kapton tape with the width of 8 mm and length of 3 cm to one surface of the actuator to form a bilayer structure. 3) Shear mode: We first heated the actuator up to 180 °C and cooled it down. Then we sheared an actuator with a shear angle of 30° using a simple customized device, as shown in Figure S9, Supporting Information. We then held the shear deformation in the actuator for 24 h.

Characterization of ss-LCE Actuator: We used an external power supply (Keysight E3642A) to apply voltage to the actuator. We measured the surface temperature of the actuator during its actuation using Advanced Thermal Imaging Camera (FLIR E75-42). We measured the actuation strain of the actuator by processing the photos taken by the digital camera (Canon 80D) using ImageJ. We measured the actuation stress of the actuator using the Instron Universal Testing System (5965 Dual Column Testing Systems; Instron) with 10 N loading cell.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

liquid crystal elastomers, multiple actuation modes, reprogrammable actuators, soft actuators

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