Materials LAB Perspective

Liquid Crystal Elastomers for Soft Actuators

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Zhijian Wang[1*], Yang Wang[2], Shengqiang Cai[2,3], and Jiping Yang[1*]

[1] *Key Laboratory of Aerospace Advanced Materials and Performance, Ministry of Education, School of Materials Science and Engineering, Beihang University, Beijing 100191, China*
[2] *Materials Science and Engineering Program, University of California, San Diego, La Jolla, California 92093, United States*
[3] *Department of Mechanical and Aerospace Engineering, University of California, San Diego, La Jolla, California 92093, United States*
* Corresponding author, E-mail: zhijianw@buaa.edu.cn; jyang@buaa.edu.cn

Abstract

Liquid crystal elastomers (LCEs) are a type of responsive materials combining liquid crystal mesogens with polymer networks. The LCEs exhibit outstanding actuation performance responsive to multiple external stimuli and show great potential as soft actuators. However, compared with conventional soft actuators, the LCEs need to be carefully synthesized and a few fabrication methods have been developed. Herein, we highlight the strategies for the material design and manufacturing techniques. Several recent studies on the mechanical design for LCE actuators are overviewed. We further discuss the challenges and future perspectives of the LCE based actuators for soft robots.

Key words: Liquid crystal elastomers; Soft actuators; Soft robots; 3D printing

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Main text

Soft actuators are key components in the soft robotic system, transducing the input energy into force and driving the robotic systems.[1,2] Compared with conventional rigid motors, the soft actuators are compliant, stretchable, and exhibit a continuous deformation with a large number of degrees of freedom (DOFs).[3] They show multiple deformation modes when interacting with the environments, such as bending, twisting or adapting their shapes in a confined space. Recently, researchers have developed numerous types of soft actuators using polymeric materials, such as pneumatic actuators,[4,5] dielectric elastomer actuators (DEAs),[6,7] responsive gels,[8,9] liquid crystal polymers,[10,11] etc. Among those smart materials and structures, liquid crystal elastomers (LCEs) have attracted lots of interests for their large reversible actuation strain and stress.

The LCEs are the integration of liquid crystal molecules and crosslinked polymer networks, possessing the elastic properties of polymer networks as well as the anisotropic properties of liquid crystals.[12–14] Different from the liquid crystal polymers that are linear polymers with high melting temperatures and large moduli, the LCEs are soft and compliant, with a glass transition temperature (T_g) below room temperature and a modulus from hundreds of kilopascals to several megapascals. Notably, the LCEs exhibit a large deformation in response to external stimuli. When the LCEs are subjected to the external stimuli such as heat or light illumination, the liquid crystal mesogens in LCEs undergo liquid crystal-isotropic phase transition, leading to a macroscopic contraction in the alignment direction of the mesogens (Fig. 1a). The actuation is reversible. After removing external stimuli, the LCEs recover to their initial shapes. The reversible actuation strain could be as large as 40% and the actuation stress could reach 10 MPa.[15] The LCEs can be classified into the thermally responsive LCEs and photo responsive LCEs according to the stimulus conditions. The photo-responsive LCEs could exhibit different behaviors, depending on the illumination angles, alignment of the liquid crystal mesogens and the geometry of the sample. For example, Li et al. demonstrated a photo-responsive LCE micropost array, in which the microposts exhibited a variety of self-regulated deformation patterns due to the interaction between the illumination angle and the alignment of liquid crystal mesogens.[16] The thermally responsive LCEs could be integrated with the heating elements, such as Joule heating materials[17] and photo-thermal materials,[18] etc. Yang and coworkers embedded photo-thermal NIR dye into the thermally responsive LCE Mobius strip and used the light to drive the motion of the strip.[19] These unique features of LCEs make them a great material for soft actuators.

However, the study of LCEs has been limited in the community of chemistry for a long time because of the complex preparation process. The synthesis of LCEs is difficult because of the following reasons: first, most liquid crystal monomers for the LCEs are not commercially available and these monomers need to be synthesized in a chemistry lab with multiple steps. Up to now, only few reactive liquid crystal monomers, such as RM257 and RM82, can be purchased and

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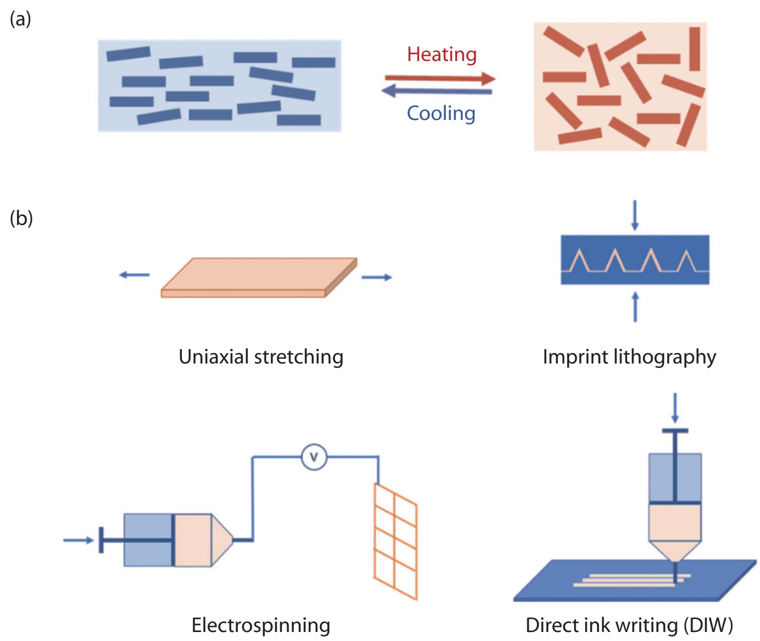

Fig. 1 (a) Schematic illustration of the phase transition of LCEs. The LCEs can generate large and reversible actuation based on the liquid crystal-isotropic phase transition. (b) Schematic illustration of the manufacturing techniques of LCEs.

directly used. Second, the fabrication process of actuatable LCEs are complex. The actuation performance of LCEs comes from the anisotropic orientation of the liquid crystal mesogens. Nevertheless, the LCEs are usually obtained in the polydomain state without additional treatment in the preparation process, in which the orientations of mesogens in each domain are random. The polydomain LCEs could not show any macroscopic deformation in response to external stimuli. To achieve the actuating ability, the liquid crystal mesogens should be aligned and crosslinked, which is also denoted as monodomain LCEs. Third, it is difficult to prepare the LCE actuators in arbitrary shapes with desired alignment patterns of mesogens in a large scale, preventing their further uses in special mechanical designs.

In the past decades, enormous efforts have been made to develop strategies to prepare the free-standing LCE samples with reversible and programmable actuation performance. They can be classified into three categories based on the formation of relaxed polymer networks: one-step polymerization, two-step polymerization and dynamic covalent chemistry strategies.[20] For the one-step polymerization strategy, the polymerizable liquid crystal meosgens are aligned first with the assistance of electric/magnetic fields or surface template alignment techniques. Further crosslinking would form a new polymer network to fix the alignment of the liquid crystal mesogens. The alignment of the LCEs could be determined by the electric/magnetic fields or the surface templates. However, the thickness of the alignment layer is restricted in a scale of around hundreds of micrometers. Later, Finkelmann and coworkers developed the two-step polymerization method to fabricate a monodomain LCE specimen in a size of several centimeters.[21] They first obtained a loosely crosslinked polymer network containing liquid crystal mesogens, which was stretchable. The liquid crystal mesogens were further aligned by stretching and fixed through the second crosslinking with residue reactive functional groups. Yakacki and coworkers employed a commercially available liquid crystal mesogen RM257 in the two-step polymerization strategy, greatly improving the simplicity and reproducibility of the LCE preparation.[22] After that, the liquid crystal mesogen RM257 has been widely used in the preparation of the LCEs, especially by engineers. However, the grand challenge still exists because the LCEs prepared by one-step or two-step polymerization method is thermoset, whose actuation behavior could not be reprogrammed. In order to endow the reprogrammability to LCEs, Ji and coworkers reported a new LCE preparation route by introducing the dynamic covalent bonds into the polymer network.[23] The dynamic covalent bonds could be triggered by light or heat, inducing the rearrangement of the polymer network. The newly obtained LCEs showed reprogrammable, modular and reprocessable properties. This seminal work inspired the development of dynamic LCEs, including the incorporation of Diels-Alder adducts,[24,25] disulfide bonds,[26–29] boronic esters,[30] etc. It is noted that the polymer network should be stable in the actuation conditions to avoid losing the actuation performance. Thus, in the thermally responsive dynamic LCEs, the reprogramming temperatures are usually very high, far above the phase transition temperature. An exception is the LCEs with disulfide bonds, which could be directly processed in the ambient temperature below its phase transition temperature.[31] It is because the bimolecular termination process of the active thiyl free radicals is very slow, providing enough time for the rearrangement of the polymer network. In recent years, the three different strategies have begun to be integrated.

The actuation behavior of the LCEs highly depends on their shapes and alignment of liquid crystal mesogens. However, in the conventional fabrication techniques, the LCEs are usually

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obtained as thin sheets or rods. With the better understanding in the material design strategies of LCEs, multiple advanced manufacturing techniques have been utilized to fabricate the LCEs in desired shapes and alignment patterns, such as 3D printing and electrospinning. 3D printing is a process fabricating objects in an additive manner with the help of computer-aided design (CAD). Compared with isotropic printing materials, the alignment accessories need to be incorporated into the device for printing aligned LCE materials. Several 3D printing techniques, including direct ink writing (DIW), digital light processing (DLP) and direct laser writing (DLW), have been reported to print LCE with 3D shapes.[32–36] Among them, DIW technique has a low cost and is easy to be adjusted. The actuation mode and performance of the printed LCE actuators can be well controlled by the printing parameters, such as printing path, the height between nozzle and substrate, the temperature of ink reservoir, and the temperature of substrate.[37,38] The printed LCE actuators could also be functionalized by modifying the ink or redesigning the print device. Kotikian et al. printed a coaxial filament composed of liquid metal core and an LCE shell. The LCE composite actuator could not only contract under Joule heating but also self-sense the deformation by the resistive feedback of liquid metal.[39] The printed LCE structure could be regulated with closed-loop regardless of different bias loading conditions. The development of LCE 3D printing techniques would provide more possibilities for the intelligent LCE actuators.

Different from pneumatic actuators and DEAs, most of the LCE actuators are triggered by cyclic heating and cooling. The responsive speed of LCE actuators depends on the thermal diffusion time. One way to shorten the time for the heat transfer is to introduce the convection. For instance, by embedding the microfluidic channel into the LCEs, we can alternatively inject hot and cold fluids into the internal channel of LCEs and realize the fast actuation as well as recovery.[40,41] The actuator can generate the contractive strain rate of 20%/s, which is close to that of human muscle. Another approach to shorten the heat transfer time is to reduce the size of LCEs, which could greatly enhance the response speed of LCEs. He et al. reported the fabrication of LCE microfibers using electrospinning technique.[42] In the electrospinning technique, a highly concentrated solution of LCE precursor is ejected under a high-voltage electric field. The liquid crystal mesogens are aligned along the axial direction of microfibers under the stretching force in the electric field and the alignment were further fixed by UV curing. It is found that the electrospun LCE fibers demonstrate the faster response speed (300%/s) and higher power density (400 W/kg) than those of the bulk LCE samples. This small scale LCE microfiber actuator show great potential in the applications of the micro robotic systems and micro devices.

With appropriate mechanical design, the LCE actuators could realize bending and twisting actuation modes as well as contraction mode to drive the motion of the soft robots. For example, the LCE tubular actuator constructed with three embedded heating wires showed multimodal actuation behavior such as multi-directional bending and homogeneous contracting by selectively heating different wires.[43] In this work, the LCEs were prepared through two-step polymerization method with careful treatment. Compared with that of monodomain LCEs, the preparation of polydomain LCEs can bypass the alignment procedures and is much simpler. With an external loading, the mesogens in the polydomain LCEs are aligned and can exhibit reversible actuation performance. The prestretched LCE would contract, upon heating above the phase transition temperature and the contracted sample would recover to the initial state under external force after removing the heating. Cai and coworkers employ the prestretched polydomain LCEs as cables to construct a six-rod tensegrity structure, in which the cables were in the tensile state and the rods were in the compressive state.[44] The LCE cable would contract under NIR irradiation through photothermal effect, driving the shape change and rolling of the tensegrity structure. This work provides a representative example of how the mechanical design would affect the material preparation methodology. The soft robots are usually composed of actuators, sensors, computation, and power sources. The material preparation and fabrication procedures should be adjusted based on the mechanical designs of the soft robots accordingly.

The past decade has seen great advancements of LCE actuators. However, challenges still exist in developing superior LCE actuators to meet the requirements of versatile soft robots. First, it is of emergence to make commercially available LCE materials with easy handling and high reproducibility, which could be used directly by mechanicians and engineers. Then, we need to establish standard protocol to evaluate actuation properties of the LCE actuators, such as the actuation strain, actuation stress, response speed, strength and fatigue in the cyclic heating and cooling. Furthermore, with the better understanding of the physical behavior of LCEs, the LCEs could be designed with enhanced actuation performance and simplified processing procedures. In addition, the liquid crystal-isotropic transition temperature of LCEs is relatively high (>60 °C), restricting its potential applications in biomedical field. Reducing the phase transition temperature requires substitution of rigid liquid crystal mesogens by using more flexible backbone. As a result, the LCEs-based soft actuator can potentially replace the damaged muscle in human body, both of which could show the contracting modes.[40] Nevertheless, it is still challenging to incorporate the control and energy source components in the LCE artificial muscle for the actuation. Finally, due to the thermally induced phase transition, the LCEs-based soft actuators have extremely low energy efficiency (<1%), which is not desired for the soft robotic system. Developing alternative strategies (pressurized air or electricity) to trigger the actuation of LCEs-based soft actuator is highly desired.[45,46]

Beyond the actuation performance, the soft actuators need to be intelligent in the future, which can sense, compute, and actuate by itself. So, what could an intelligent LCE actuator look like? How could we integrate the actuation behavior and feedback elements with the computer-aided fabrication techniques to make the LCE actuators intelligent? It still has a long way to go for the development of LCE actuators, which needs interdisciplinary and long-term research efforts from physicists, chemists, mechanicians, and engineers. It is expected that a wide variety of LCE actuators can be employed in constructing collaborative soft robots that can work closely with human in the near future.

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Conflict of interest

The authors declare no conflict of interest.

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Biography

Zhijian Wang is currently a professor in the School of Materials Science and Engineering at the Beihang University. He obtained his bachelor and PhD degree from Peking University in 2010 and 2015. Then, he worked in the Department of Mechanical and Aerospace Engineering at the University of California, San Diego (UCSD) as a postdoctoral researcher between 2015 and 2020. He joined Beihang University in 2020. His research is mainly focused on the synthesis and manufacturing of soft intelligent materials, mechanics of soft materials, and the design and fabrication of soft robots.