GLOBAL RESEARCH IMMERSION PROGRAM
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4D Printing of Liquid Crystal Elastomer

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Research Objective

To design and build a 3D printing system that utilizes temperature as a control parameter for aligning liquid crystal elastomers, with the aim of fabricating structures exhibiting spatially varying properties.

Background

Liquid crystal elastomers (LCEs) are unique materials capable of significant shape changes in response to external stimuli. Their potential for creating dynamic structures has attracted considerable interest. 4D printing, an extension of 3D printing that introduces the element of time, enables the creation of objects that can change shape over time. This research explores the integration of LCEs into 4D printing to fabricate objects with programmable shape transformations. By combining the precision of 3D printing with the responsive nature of LCEs, we aim to develop innovative applications for this promising technology. Initial experiments focus on simple structures to demonstrate the feasibility of temperature-induced shape changes. Future work will explore more complex geometries and stimuli-responsive behaviors.

Experimental Section

Materials

4-(3-Acryloyloxypropyloxy)-benzoesure 2-methyl-1, 4-phenylester (RM257), 4 '-Pentyl-4-cyanobiphenyl (5CB), EDDET, 2-benzyl-2-(dimethylamino)-4'-morpholino-butyroph (Irgacure 369), dipropylamine (Figure 1).

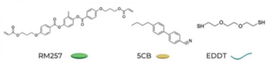

Figure 1. Schematic illustrations of the chemical structures of ink.

LCE ink was prepared by combining 0.5415 grams of RM257 with 50 wt% 5CB and heating the mixture to 80°C for 2 minutes. The resulting mixture was stirred vigorously at 800 rpm for 5 minutes. Concurrently, a crosslinking agent solution was prepared by mixing 0.1524 grams of EDDET, 0.0035 grams of Irgacure 184, and 0.0162 grams of DPA. Upon combining the two solutions, the mixture was stirred vigorously at 800 rpm until a viscous consistency was achieved. The final LCE ink was stored in a dark environment.

Method

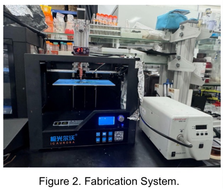
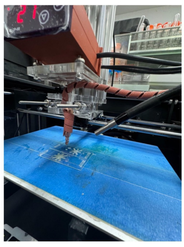
Figure 2. Fabrication System.

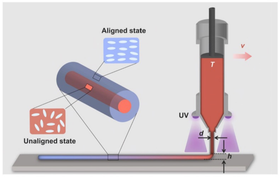

Figure 3. Schematic illustrations of the printing process.

The printing ink is prepared through the Michael addition reaction between liquid crystal mesogen RM257 and chain extender 2,2 '-(ethylenedioxy) diethanethiol (EDDET) with acrylate moieties as the end groups (fig. S1). At room temperature, because of the shear stress generated through the extrusion process, liquid crystal mesogens are aligned along the printing path. Because the shear stress is greater at the edge of the needle, the liquid crystal in the outer shell is better aligned, while the internal orientation is not obvious and presents a multi-domain state. By heating the needle mouth, we can obtain non-oriented LCE filaments. The extruded LCE filaments are further crosslinked under ultraviolet (UV) illumination to permanently fix the mesogen alignment.

Result and Discussion

As reported in previous studies, liquid crystal mesogens align under shear and extensional flow along the printing path. The liquid crystal mesogens are nearly uniformly aligned in the outer shell, while the mesogen orientation in the core is random in a macroscopic scale, namely, in a polydomain state.

Figure 4. Polarized optical microscope (POM) images of LCE filaments printed at room temperatures.

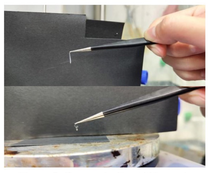
Figure 5. Demonstration of filament contraction under heat conditions.

The assumption described above is further validated with experiments. Using polarized optical microscopy (POM), we can examine the mesogen alignment in printed LCE filaments. An extruded, single LCE filament is observed between crossed polarizers, as shown in Fig. 4. For a filament printed at room tempreture, the outer shell of the filament is dark, and the inner core is bright under POM when the axial direction of the filament is parallel to the polarizer or the analyzer. The brightness of the outer shell increases greatly when the axial direction of the filament is 45° with respect to the polarizer(Picture C), indicating that the liquid crystal mesogens in the shell align along the printing path. However, the brightness of the inner core does not vary much when we rotate the filament, confirming that the inner core of the filament is in a polydomain state . A free-standing LCE filament generates thermal contraction only when the liquid crystal mesogens are aligned in a macroscopic scale. Therefore, for one filament, the aligned shell is the only contributor to its actuation behavior.

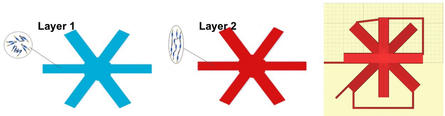

Figure 6. Two layers of LCE elastomer with different structures and print trajectory preview.

We designed a double-layer structure with six petals to mimic a blooming flower. When the printed petals were placed on a hot plate at 120°C, the first layer of unoriented LCE did not deform, while the second layer of oriented LCE curled up. Macroscopically, each petal curled up evenly due to the mismatch in contraction between the two layers.

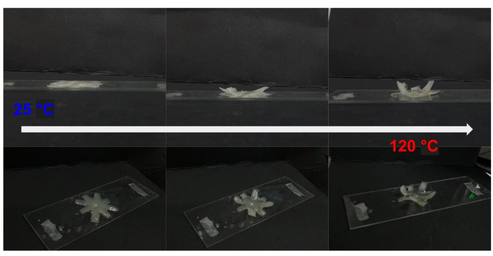

Figure 7. Demonstration of petal contraction under heat conditions, inculde top view and 45 degrees view.

Conclusion

This study demonstrates the potential of direct ink writing (DIW) for fabricating complex liquid crystal elastomer (LCE) structures. By precisely controlling the arrangement of liquid crystal molecules within the material, we achieved diverse actuation behaviors. This approach aligns with the concept of 4D printing, where 3D printed structures incorporating active materials can undergo programmed shape changes in response to stimuli. This technology holds promise for applications in soft robotics and biomedical devices.

Reference

1. Martinez, A. P., Ng, A., Nah, S. H., & Yang, S. (2024). Active-Textile Yarns and Embroidery Enabled by Wet-Spun Liquid Crystalline Elastomer Filaments. *Advanced Functional Materials*, 2400742.
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