

Active Pillar Array Structure by Solvent-free Liquid Crystal Elastomer Resin

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 Electronic Supplementary Information

Abstract Bioinspired active pillar structures, known for their large surface area, mechanical compliance, and diverse deformation modes, have garnered extensive research interest. Among various active pillar structures, liquid crystal elastomer (LCE) pillar arrays are capable of exhibiting significant and reversible anisotropic deformation under cyclic heating and cooling, showing great potential in tunable adhesion, soft robots, and biomedical devices. However, scaling up LCE pillar manufacturing remains challenging, limiting its practical applications. In this work, a solvent-free LCE resin is developed with unique features including simple operating procedure, short fabrication time, and tunable responsive temperature, enabling rapid and large-scale production of LCE pillar arrays. The LCE resin allows for the preparation of complex 3D shapes in addition to film or specimen. The fabrication time can be as short as 4 h, without the need to evaporate solvent. Moreover, the LCE resins can be adjusted with a variable phase transition temperature range from 49.4 °C to 97.7 °C by incorporating non-liquid crystal acrylate chains. The resulting active pillar array structure can undertake sequential actuation upon heating with the tunable actuation temperature. Finally, the application of these pillar arrays in multi-level information encryption is demonstrated. The LCE pillar structure introduced here offers a new strategy for constructing advanced active LCE structures with tunable responsive behavior.

Keywords Pillar array; Liquid crystal elastomer; Solvent-free resin; Multi-level encryption

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INTRODUCTION

Various creatures have evolved microstructural surfaces with specialized functions for self-cleaning, dry adhesion, and structural coloration.^[1] For instance, lotus leaves exhibit superhydrophobicity and exceptional self-cleaning abilities because of their microscale papillae structure and hydrophobic epidermal wax on their surface.^[2] Geckos possess remarkable climbing ability owing to the micro- and nanostructures on their toe surfaces, which allow them to attach to and detach from nearly any surface *via* van der Waals forces.^[3,4] Inspired by nature, researchers have developed a variety of bioinspired active pillar structures capable of performing complex and precise tasks through dynamic and reversible deformation in response to external stimuli, highlighting their broad application prospects in fields such as haptic sensors, tunable adhesion, and biomedical devices.^[5–9] These active pillar array structures can be fabricated from various soft active materials, including shape memory

polymers (SMPs),^[10] magnetic soft materials (MSMs),^[11] hydrogels,^[12,13] and liquid crystal elastomers (LCEs).^[14–22]

Among these active materials, LCEs have garnered significant interest owing to their large reversible actuation, anisotropic deformation, and versatile programmable actuation modes.^[23–29] LCEs consist of liquid crystal mesogens and polymer networks, combining the anisotropic properties of mesogens with the elasticity of the polymer networks.^[30–37] When heated above the nematic-isotropic phase transition temperature (T_i), the LCEs undergo macroscopic and anisotropic deformation.^[38–44] The actuation is fully reversible. Once the heat is removed, the LCEs return to their original shapes.^[45–52] Cui *et al.* developed an LCE pillar structure using a pair of permanent magnets to align mesogens along the pillar direction.^[53] The pillar arrays can switch reversibly between adhesive and non-adhesive states in response to temperature changes. However, the size of the platform between magnets with a high field strength is limited, making the preparation of large arrays of pillar structures significantly challenging. Additionally, mechanical stretching typically requires removal of the solvent after the initial polymerization step, which makes it difficult to maintain the geometry of the pillar.^[54–57]

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In this work, we fabricate LCE pillar array structures using solvent-free LCE resins with a simple operating procedure, short fabrication time, and tunable response temperature.^[58–60] Compared to conventional two-step polymerization methods, the fabrication time in this work can be as short as 4 h (Table S1 in the electronic supplementary information, ESI). This is because the solvent-free fabrication process allows the pillar arrays to retain their as-prepared shapes without the need for solvent evaporation, thereby significantly shortening the production time. The active pillar structures show tunable phase transition temperature T_i with a range from 49.4 °C to 97.7 °C with the incorporation of non-liquid crystal acrylate chains. The T_i of the LCEs is found to be highly dependent on the molecular weight and content of the flexible acrylate backbones. The resulting structures can exhibit multiple actuation modes through different programming methods and undergo sequential and multiple shape changes upon heating. Finally, we integrate LCE pillar arrays with varying T_i for information encrypting, allowing decryption at specific temperatures and providing multi-level protection of information security.

EXPERIMENTAL

Materials

1,4-Bis-[4-(3-acryloyloxypropyloxy)benzoyloxy]-2-methylbenzene (RM257, 98%), poly(ethylene glycol) diacrylate (PEGDA, Aladdin, average molecular weight ~200, 400, and 600, contains 400–600 ppm 4-methoxyphenol as an inhibitor), 2,2'-(ethylene-dioxy) diethanethiol (EDDET, Aladdin, 97%), pentaerythritol tetra (3-mercaptopropionate) (PETMP, Innochem, 90%), dipropylamine (DPA, Alfa, 99%), (2-hydroxyethoxy)-2-methylpropionophenone (Irgacure 2959, Aladdin, 98%), and methylene chloride (CH_2Cl_2) were used as received without further purification.

Synthesis of Solvent-free LCE Resin

Component A: A solvent-free LCE resin containing PEGDA with a molecular weight of 200 and molar ratio of 5% was used as an example. First, liquid crystal mesogen RM257 (13.42 g, 22.8 mmol), non-liquid crystal PEGDA200 (0.24 g, 1.2 mmol), and extender EDDET (2.19 g, 12 mmol) are dissolved in CH_2Cl_2 (20 mL). Then, catalyst DPA (0.06 g, 0.6 mmol) is added into the mixture dropwisely. The resulting solution was stirred overnight at room temperature. Finally, the mixture was placed in an oven at 80 °C for 24 h to allow the complete evaporation of the solvent. Component A is in an opaque polydomain state at room temperature.

Component B: The crosslinker PETMP (1.02 g, 2.1 mmol), extender EDDET (0.88 g, 4.8 mmol), catalyst DPA (0.04 g, 0.4 mmol) and photoinitiator Irgacure 2959 (0.09 g, 0.4 mmol) are mixed at 80 °C to form a homogeneous solution.

Preparation of Pillar Arrays Using Solvent-free LCE Resin

First, components A and B of the LCE resin were mixed at 80 °C, and the mixture was degassed under vacuum and poured into a customized mold. The mixture was then filled into the holes of the mold under vacuum. After curing at 80 °C for 3 h, the polydomain LCE pillar arrays were obtained. Finally, the polydomain LCE pillars were programmed into different shapes under exter-

nal forces and further fixed under 365 nm UV light for 30 min.

Characterizations

Thermogravimetric analysis (TGA) was performed on a TG-DSC 3+ (Mettler) instrument in a nitrogen atmosphere. The temperature range of the TGA measurements was from 25 °C to 800 °C, and the heating rate was set at 5 °C·min⁻¹. ¹H nuclear magnetic resonance (NMR) spectroscopy measurements were conducted on an AV600M spectrometer at room temperature using chloroform-d (CDCl_3) as the solvent. Differential scanning calorimetry (DSC) measurements were performed using a DSC250 calorimeter (TA Instruments) in a nitrogen atmosphere. The temperature range of the DSC measurements was from -50 °C to 150 °C, and the heating and cooling rates were set at 5 °C·min⁻¹. The DSC curve of the second heating run was used to remove the thermal history of the samples. The dynamic modulus and tan delta of the LCEs were recorded using a DMA Q800 (TA Instruments) in frequency-strain mode. The dimensions of the measured samples were 10 mm × 3 mm × 1 mm. The temperature was increased from -50 °C to 150 °C, and the heating rate was set to 5 °C·min⁻¹. The other test parameters were selected as 0.01 N preload force, an oscillating strain of 0.2% at 1 Hz, and a force track of 125%. The temperature dependence of the length changes of the LCEs was measured using a DMA Q800 (TA Instruments). A sample size of 10 mm × 3 mm × 0.5 mm is used. The temperature was increased from -50 °C to 150 °C at a heating rate of 5 °C·min⁻¹. Uniaxial tensile tests of the LCEs at room temperature were performed on a Universal Mechanical Testing System (WANCE) with a 100 N loading cell. The LCEs were cut into rectangular shapes with dimensions of 30 mm × 10 mm × 0.5 mm. The strain rate was varied from 0.2 min⁻¹. Optical images were captured using a digital camera (Nikon Z5).

RESULTS AND DISCUSSION

Preparation of Pillar Arrays Using Solvent-free LCE Resin

Fig. 1(a) schematically depicts the pillar array preparation process using a solvent-free LCE resin. Component A consists of liquid crystal oligomers with acrylate moieties as the end groups, synthesized through a thiol-acrylate Michael addition reaction involving a liquid crystal mesogen (1,4-bis-[4-(3-acryloyloxypropyloxy)benzoyloxy]-2-methylbenzene) (RM257), non-liquid crystal poly(ethylene glycol) diacrylate (PEGDA), and the chain extender 2,2'-(ethylenedioxy) diethanethiol (EDDET). The molar ratio of acrylate to thiol groups is set as 2:1. The mesogen RM257 is partially replaced by flexible PEGDA, allowing the tuning of the phase transition temperatures T_i in the resulting LCEs. Component B contains the crosslinker pentaerythritol tetra (3-mercaptopropionate) (PETMP), extender EDDET, catalyst dipropylamine (DPA), and photoinitiator Irgacure 2959 (Fig. 1b). Components A and B of the LCE resin are mixed and poured into a customized mold. After curing at 80 °C for 3 h, a polydomain LCE pillar array (Shape A) is formed via a first-crosslinking reaction. Subsequently, the polydomain LCE pillars can be programmed into various shapes under three types of external forces (mechanical stretching, compressing, or bending) without the need for solvent evaporation, thereby significantly shortening the production time. The alignment of mesogens in these programmed shapes is further fixed by a second-

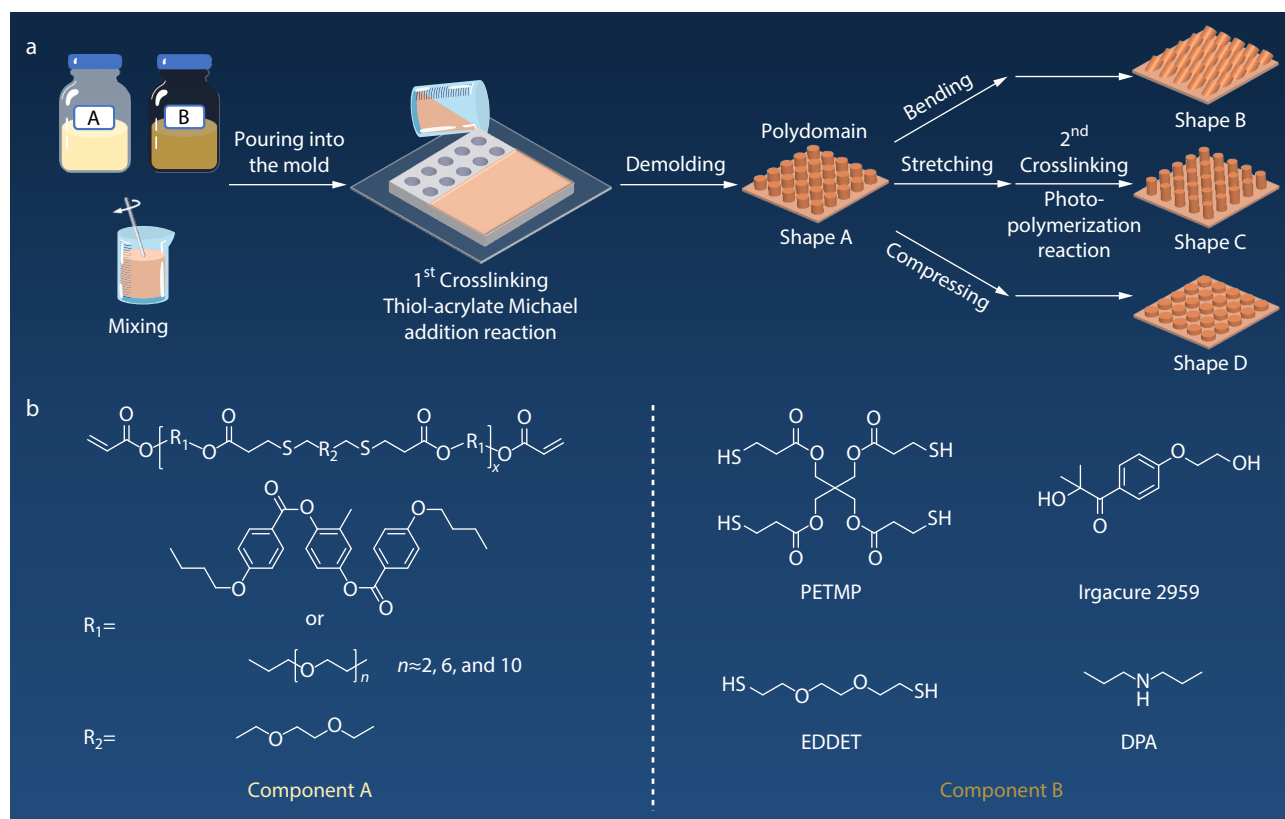


Fig. 1 Preparation of active pillar arrays using solvent-free LCE resin. (a) Schematic illustration of the process of preparing pillar array structure by solvent-free LCE resin. (b) The structures of chemicals in component A and component B.

crosslinking reaction under ultraviolet (UV) illumination. Detailed preparation procedures are provided in the electronic supplementary information. The aligned LCE pillars exhibit three distinct actuation modes when subjected to cyclic heating and cooling: contraction, elongation, and bending.

The thermo-responsive properties of LCE resins can be finely adjusted by replacing RM257 with non-liquid crystal acrylate chains. In the following section, we study the influence of the molecular weight and content of PEGDA on the phase transition temperature of the obtained LCEs.

Influence of the Molecular Weight of PEGDA on the T_i of LCEs

Replacing mesogen RM257 with non-mesogenic PEGDA segments effectively lowers the isotropic temperature (T_i) of the LCEs. We investigate the impact of the molecular weight and content of PEGDA segments on the phase transition temperature of the LCEs in detail. First, we prepare a solvent-free LCE resin using PEGDA with a molecular weight of 200 and molar ratio of 5%, defined as $n(\text{PEGDA})/(n(\text{PEGDA}) + n(\text{RM257}))$. Component A appears as opaque at room temperature, indicating that it is in a polydomain state (Fig. S1 in ESI). The degree of polymerization (DP) of the oligomer in component A is characterized by $^1\text{H-NMR}$ spectroscopy (Fig. S2 in ESI). The DP can be calculated as 1.07, which is consistent with the feed ratio between the acrylate groups and thiol groups. The thermal decomposition temperature (T_d) at 5% weight loss is 329 °C, indicating that component A remains stable in the temperature range of our experiment, which is below 200 °C (Fig. S3 in ESI). The glass transition temperature T_g and phase transition temperature T_i of the

oligomer, determined by differential scanning calorimetry (DSC), are -23.0 and 77.6 °C, respectively (Fig. S4 in ESI). TGA measurement shows that the mass loss of the oligomer is less than 1% when the temperature rises to 100 °C (Fig. S3 in ESI), confirming that component A is solvent-free. Detailed synthetic procedures are provided in the ESI.

Using solvent-free resins, polydomain LCEs with residual reactive groups are obtained through a first-crosslinking reaction. Photographs of component A, component B, and the mixture before and after curing are shown in Fig. S5 (in ESI). The influence of the molecular weight of PEGDA on T_i is explored using LCEs with a PEGDA molar ratio of 5%. Three types of LCEs containing PEGDAs with molecular weights of 200, 400, and 600 are denoted as LCE-200, LCE-400, and LCE-600, respectively. The T_i and T_g values of the polydomain LCEs are characterized using DSC, as shown in Fig. 2(a). The results indicate that both T_i and T_g decrease when increasing the molecular weight of PEGDA (Fig. 2b). Specifically, T_i decreases from 62.9 °C for LCE-200 to 47.0 °C for LCE-600. This reduction is attributed to the increased length of the flexible acrylate backbones in the polymer network, which facilitates chain segment mobility, thereby lowering T_i . Correspondingly, all LCEs exhibit a soft elasticity plateau in their stress-strain curves at room temperature, indicating their polydomain state (Fig. 2c).

Additionally, the polydomain LCEs are stretched and fixed into monodomain states through a second-crosslinking reaction of the residual reactive groups. The T_i and T_g of the monodomain LCEs are characterized by dynamic mechanical anal-

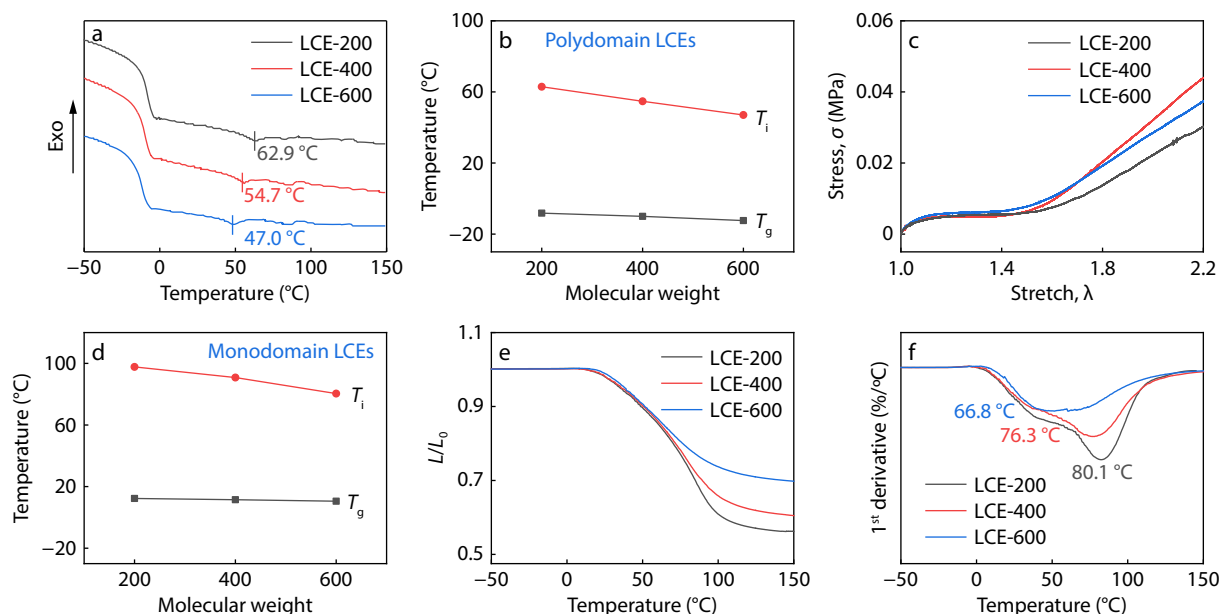


Fig. 2 The influence of the molecular weight of PEGDA on the phase transition temperature T_i of LCEs. (a) DSC curves of three kinds of polydomain LCEs containing PEGDAs with different molecular weights of 200, 400, and 600. (b) The phase transition temperature T_i and glass transition temperature T_g of the polydomain LCEs as a function of the molecular weight of PEGDA. (c) The uniaxial tensile curves of LCE-200, LCE-400, and LCE-600. The strain rate is 0.2 min⁻¹. (d) The T_i and T_g of the monodomain LCEs as a function of the molecular weight of PEGDA. (e) The relative length changes (L/L_0) versus temperature curves of LCE-200, LCE-400, and LCE-600. (f) Actuation rate of three kinds of LCEs determined by the first derivative of the length change with respect to temperature.

ysis (DMA) (Figs. S6–S8 in ESI). T_g is determined as the temperature at which the $\tan\delta$ value reaches its maximum, whereas T_i is determined where the storage modulus (E') has a minimum value. As the molecular weight of PEGDA increases, both T_i and T_g decrease (Fig. 2d). We further investigate the actuation behavior of monodomain LCEs by measuring the length changes upon heating. The contraction ratio is calculated as $(L_0 - L_i)/L_0$, where L_0 and L_i represent the lengths of the LCE specimen at -50 and 150 °C, respectively. LCE-200 presents a maximum contraction ratio of 43.8% in the ranges from -50 °C to 150 °C. The change in length is defined as L/L_0 , where L and L_0 correspond to the length of the LCE specimen at a given temperature and -50 °C, respectively. As the molecular weight of PEGDA increases, the length change (L/L_0) of the LCEs decreases, which is attributed to the relative reduction in the liquid crystal mesogen content (Fig. 2e). The actuation performance of the LCEs is stable, cycling 20 times between -15 and 150 °C without any loss in length change (Figs. S9–S11 in ESI). We calculated the actuation rate by taking the first derivative of the length change with respect to the temperature. Notably, the temperature at which the maximum deformation rate occurs is 15 °C lower than T_i , owing to their different testing and calculation methods, yet showing similar trends to those observed for T_i (Fig. 2f).

Influence of PEGDA Content on the T_i of LCEs

Given the excellent actuation performance of the LCE containing PEGDA200, we investigate the effect of PEGDA content on T_i by preparing a series of LCEs with varying PEGDA molar ratios, referring to as LCE-200- x , where x represents the PEGDA molar ratio. As shown in Fig. 3(a), the T_i and T_g values of the polydomain LCEs are measured by DSC. T_i decreases significantly from 62.9 °C for LCE-200-5% to 29.1 °C for LCE-200-20%, primarily due

to the reduced content of liquid crystal mesogens in LCEs (Fig. 3b). In the stress-strain diagram at room temperature, as the PEGDA content increases, the span of the stress plateau of the LCEs notably shortens, reaching almost zero for LCE-200-20% (Fig. 3c). Upon gradual heating from room temperature to 65 °C, the LCE sheets with varying T_i exhibit a sequential transition from opaque polydomain states to transparent isotropic states, consistent with the experimental results mentioned above (Fig. 3d).

The T_i and T_g values of the monodomain LCEs are further characterized by DMA (Figs. S12–S15 in ESI). As the PEGDA content increases from 5% to 20%, T_i decreases from 97.7 °C to 49.4 °C, attributed to the decrease of liquid crystal mesogen content (Fig. 3e). All monodomain LCEs with varying PEGDA contents exhibit contraction along the longitudinal axis upon heating, with length changes (L/L_0) decreasing as the PEGDA content increases (Fig. 3f). Additionally, the temperature at which the maximum actuation rate occurs also declines with increasing PEGDA content (Fig. 3g). The LCEs demonstrate excellent reversible actuation behavior, maintaining a stable actuation performance after 20 cycles between -15 and 150 °C (Figs. S16–S19 in ESI). As the temperature gradually increases from room temperature to 100 °C, the monodomain LCE specimens sequentially achieve maximum contraction. The contraction ratio decreases from 41% for LCE-200-5% to 26% for LCE-200-20% (Fig. 3h).

Multimodal Actuation of LCE Pillar Arrays

The LCE pillar array structures can be programmed using various actuation modes. First, we prepare polydomain LCE-200-5% pillar arrays containing reactive acrylate groups. Each pillar has a dimension of 3.5 mm in height and 2.0 mm in diameter. These pillars can be programmed into different shapes *via* mechanical

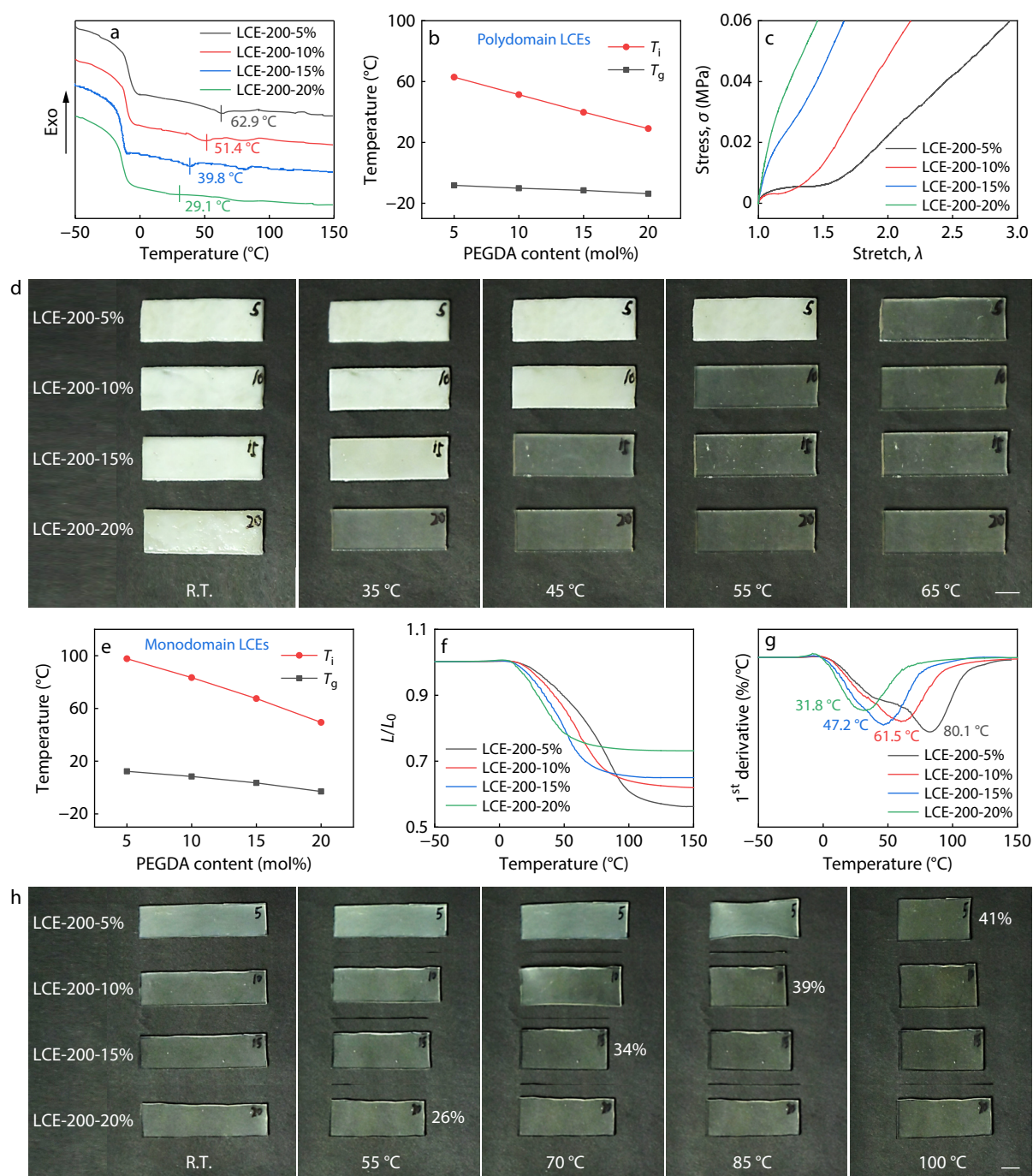


Fig. 3 The influence of the PEGDA content on the T_i of LCEs. (a) DSC curves of polydomain LCEs with different PEGDA contents. (b) The T_i and T_g of the polydomain LCEs as a function of the content of PEGDA. (c) The stress-stretch curves of four kinds of LCEs. The strain rate is 0.2 min^{-1} . (d) The photographs of four kinds of polydomain LCE sheets at different temperatures. (e) The T_i and T_g of the monodomain LCEs as a function of the content of PEGDA. (f) The relative length changes (L/L_0) versus temperature curves of monodomain LCEs with different PEGDA contents. (g) The actuation rate of four kinds of LCEs by performing the first derivative of the length change over temperature. (h) The contraction of monodomain LCE specimens with different T_i at different temperatures. Scale bars: 6 mm.

stretching, compressing, or bending. They exhibit distinct actuation modes during the heating and cooling cycles following a second step curing process.

Initially, the polydomain pillar arrays with Shape A are deformed into Shape B and fixed by a second crosslinking under UV illumination (Fig. S20a in ESI). When heated to 100 °C,

Shape B transitions into Shape A accompanied by a transparent isotropic state. This deformation process is fully reversible. Upon cooling to ambient condition, the pillar arrays revert to bending shape B (Fig. 4a). In addition to bending, using tape to adhere the pillar structure to the top and bottom glass sheets, the polydomain LCE pillars can also be stretched

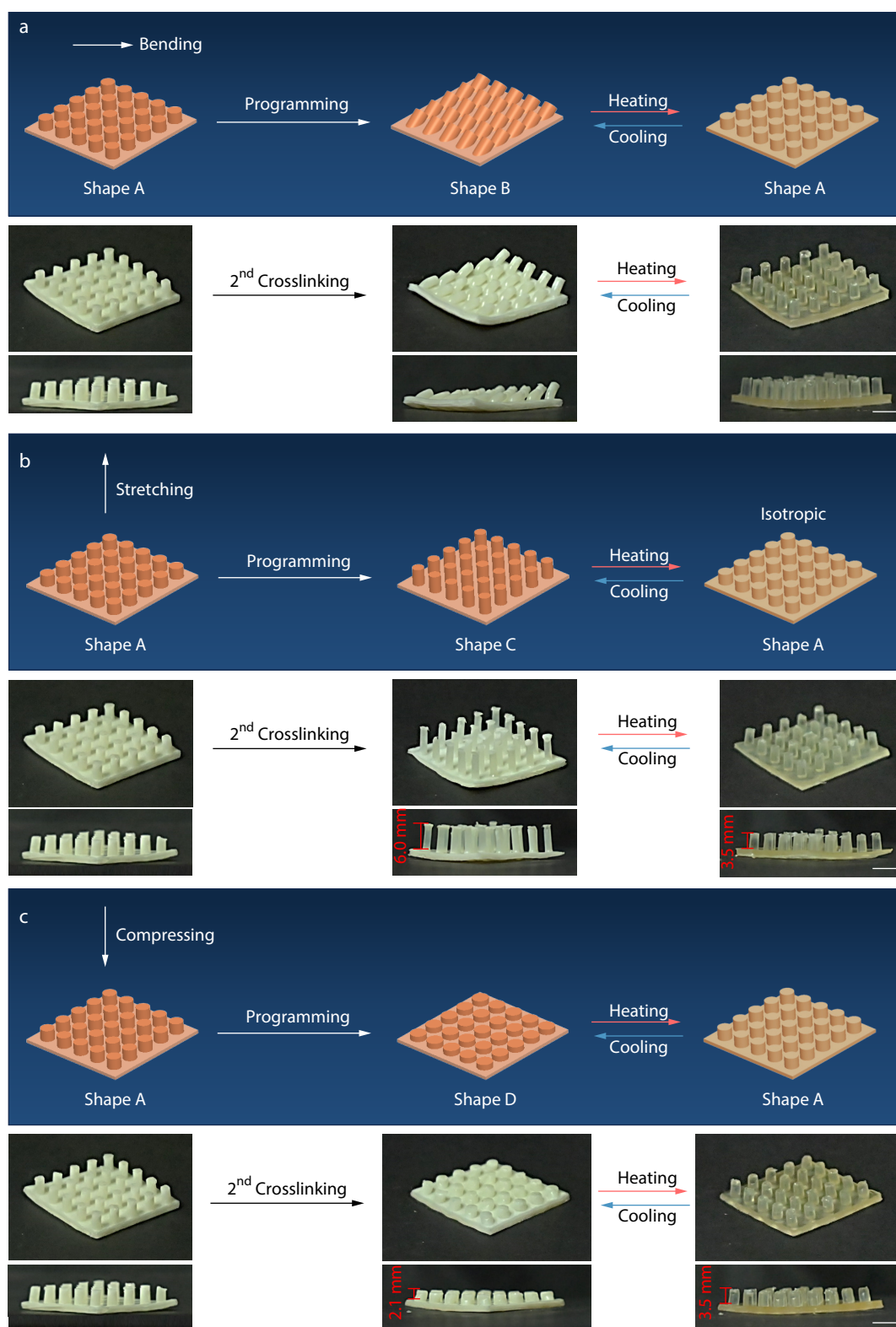


Fig. 4 Multimodal actuation of LCE pillar arrays. (a) The polydomain pillar arrays with Shape A are deformed and fixed into Shape B. The programmed pillar array structure deforms between Shape A and Shape B under heating and cooling cycles. (b) The polydomain LCE pillars are stretched along the longitudinal axis and fixed into Shape C under external force. After being heated at 100 °C, the heights of the pillars contract from 6.0 mm to 3.5 mm, while their diameters increase from 1.4 mm to 2.0 mm. This deformation process is reversible. After cooling to ambient condition, pillar arrays return to shape C. (c) The polydomain Shape A is compressed into Shape D along the longitudinal direction. The dimensions of pillars undergo reversible deformation between 2.1 mm in height $\times$ 2.6 mm in diameter and 3.5 mm in height $\times$ 2.0 mm in diameter under 25 °C and 100 °C cycles. Scale bars: 6 mm.

along the longitudinal axis and fixed into Shape C under an external force (Fig. S20b in ESI). Upon heating to 100 °C, the heights of the pillars contract from 6.0 mm to 3.5 mm, while their diameters increase from 1.4 mm to 2.0 mm, as illustrated in Fig. 4(b). Furthermore, polydomain Shape A can be compressed into Shape D along the longitudinal direction (Fig. S20c in ESI). The dimensions of the pillars undergo reversible deformation, transitioning between 2.1 mm in height $\times$ 2.6 mm in diameter and 3.5 mm in height $\times$ 2.0 mm in diameter during cycles between 25 °C and 100 °C (Fig. 4c).

LCE Pillar Arrays with Different T_i for Multi-level Encryption

In addition to various actuation modes, LCE pillar array structures can also demonstrate temperature-dependent sequential actuation. The T_i values of monodomain LCE-200-5%, LCE-200-10%, and LCE-200-15% are 97.7, 83.4 and 67.5 °C, respectively. We fabricate three pieces of LCE material with different actuation temperatures in Shape B and place them in the same heating environment. At room temperature, all three LCEs remain below their T_i , maintaining their Shape B configurations (Fig. 5a). As the temperature rises to 70 °C, the LCE-200-15% pillar array

structure first transitions into isotropic Shape A. The LCE-200-10% shows a slight shape change owing to its broader temperature response range, while LCE-200-5% exhibits minimal deformation at this temperature (Fig. 5b). As the temperature increases further to 85 °C, the LCE-200-10% pillar array structure also shifts from Shape B to Shape A (Fig. 5c). Finally, at 100 °C, all the LCE pillar structures transform into Shape A, achieving transparent isotropic states, as shown in Fig. 5(d). This deformation process is fully reversible. Upon cooling to ambient condition, all pillar arrays return to their bent shape B.

The sequential actuation of the LCE enables the multi-level encryption of information. To demonstrate this concept, we prepare polydomain pillar array structures by using LCE-200-5% and LCE-200-15%. Each pillar has a height of 1.5 mm and a diameter of 2.0 mm. The two types of polydomain pillar array structures are compressed along the longitudinal axis and fixed into flat sheet shapes under external force (Fig. S21 in ESI). The LCE-200-15% sheets can transform into pillar array structures when heated to 70 °C, whereas the LCE-200-5% sheets require heating to 100 °C to undergo shape changes. Additionally, we prepare flat polydomain LCE sheets by removing the pillars, which do not exhibit any deformation up-

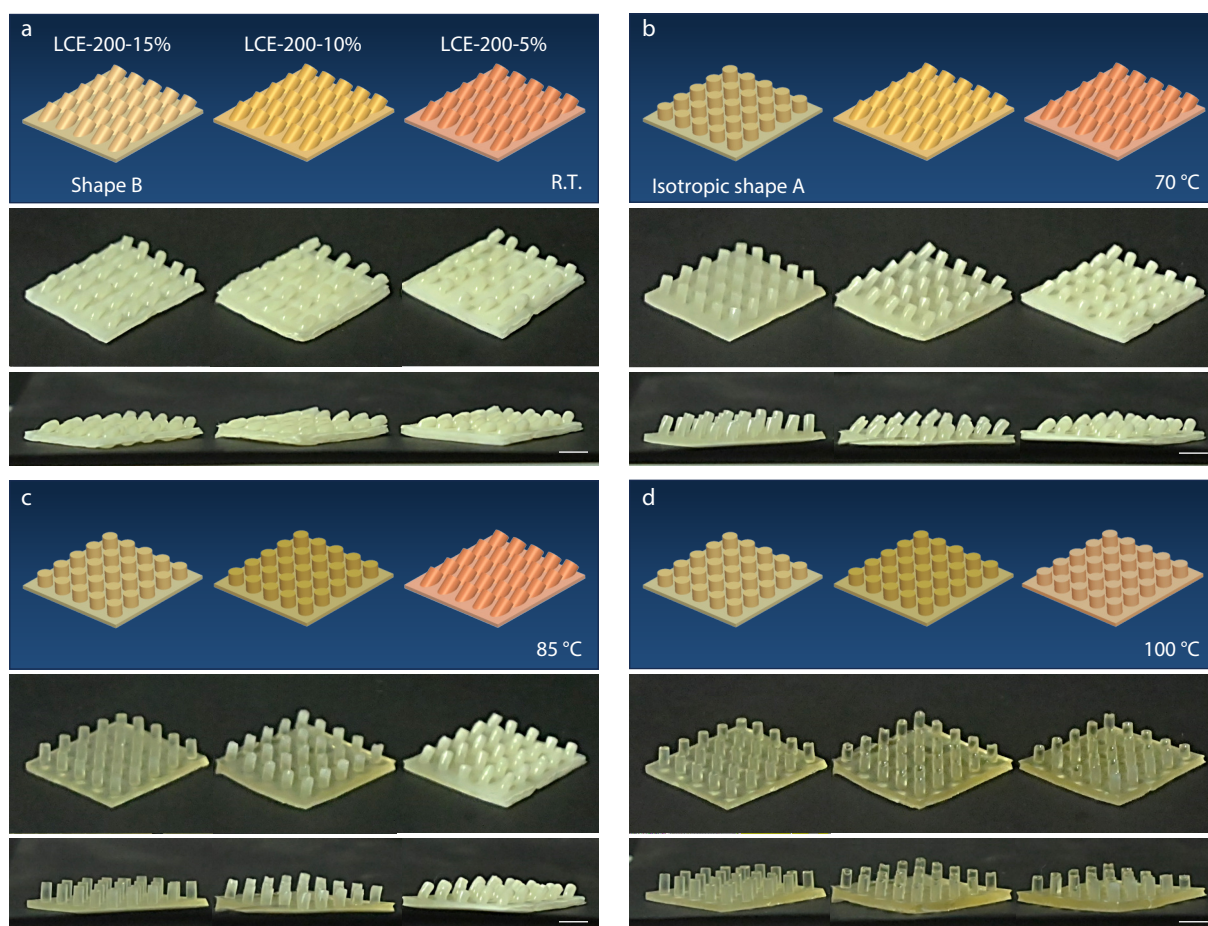


Fig. 5 Temperature-dependent sequential actuation of LCE pillar arrays. (a) Three kinds of pillar structures with different T_i maintain shape B at room temperature. (b) The photographs of three kinds of pillar structures at 70 °C. LCE-200-15% pillar array structure deforms into isotropic Shape A. The LCE-200-10% exhibits slight shape change due to the wide temperature response range of LCEs, while the LCE-200-5% has almost no deformation. (c) When the temperature further increases to 85 °C, LCE-200-10% pillar array structure also transits into isotropic Shape A. (d) All LCE pillar structures deform into Shape A with a transparent isotropic state at 100 °C. Scale bars: 6 mm.

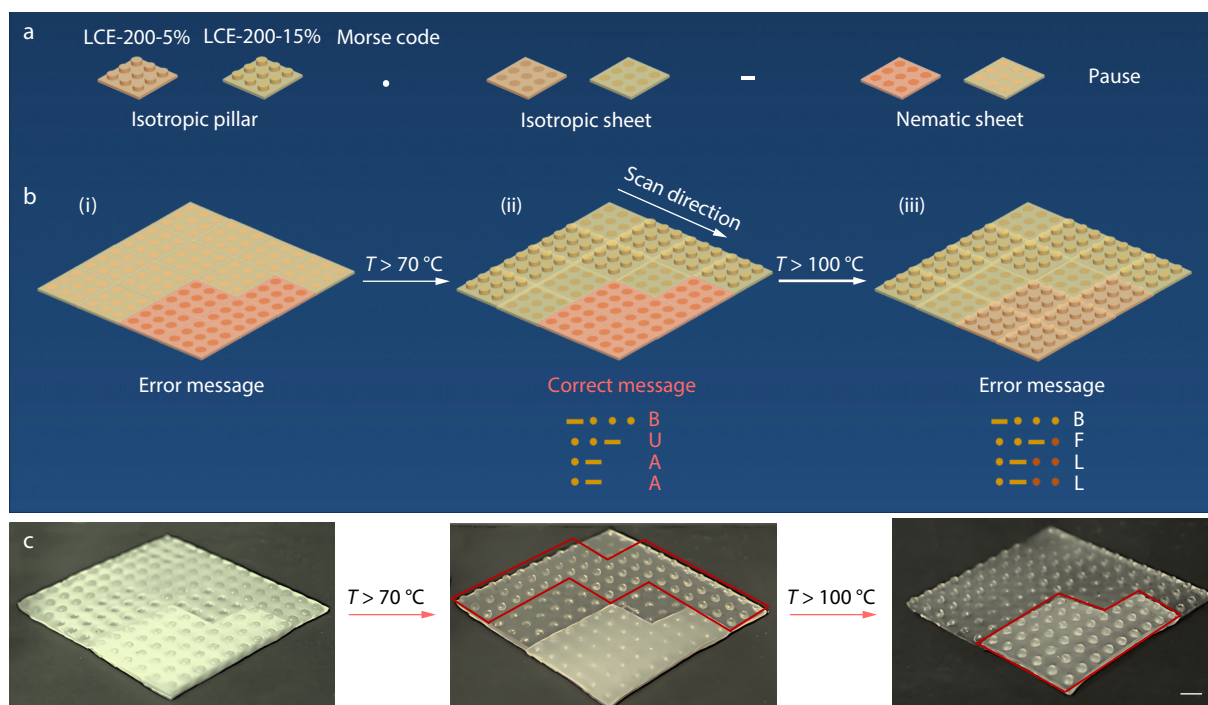


Fig. 6 LCE pillar arrays with different T_i for multi-level encryption of information. (a) The transparent isotropic pillar and sheet units at high temperature represent the long signal “—” and the short signal “•” in Morse code, respectively. While the opaque nematic LCE sheets at room temperature represent the pauses between each letter (Fig. 6a). A large plate consisting of 4×4 different LCE units is fabricated. At room temperature, all the units appear as opaque flat sheets, resulting in no information being displayed (Fig. 6b(i)). As the temperature rises to 70 °C, the LCE-200-15% sheet units transform into transparent isotropic states and partially deform into pillar arrays, whereas the LCE-200-5% units remain unchanged in transparency and shapes. At this temperature, the correct message “BUAA” is revealed (Fig. 6b(ii)). If the temperature increases further to 100 °C, the LCE-200-5% sheet units will also deform into pillar arrays with transparent isotropic states and reveal the incorrect information “BFL”, effectively protecting the secure information (Fig. 6b(iii)). Photographs showing the changes in the LCE plate upon heating are shown in Fig. 6(c). The deformation process is shown in movie S1 (in ESI). It is worth mentioning that the changes in shape and transparency of LCE units are fully reversible. When the temperature returns to room temperature, the LCE plate recovers to its initial state and redisplay the erroneous message.

on heating. In this setup, we use transparent isotropic pillar and sheet units to represent the long signal “—” and the short signal “•” in Morse code, respectively. The opaque nematic LCE sheets at room temperature represent the pauses between letters (Fig. 6a). A large plate consisting of 4×4 different LCE units is fabricated. At room temperature, all the units appear as opaque flat sheets, resulting in no information being displayed (Fig. 6b(i)). As the temperature rises to 70 °C, the LCE-200-15% sheet units transform into transparent isotropic states and partially deform into pillar arrays, whereas the LCE-200-5% units remain unchanged in transparency and shapes. At this temperature, the correct message “BUAA” is revealed (Fig. 6b(ii)). If the temperature increases further to 100 °C, the LCE-200-5% sheet units will also deform into pillar arrays with transparent isotropic states and reveal the incorrect information “BFL”, effectively protecting the secure information (Fig. 6b(iii)). Photographs showing the changes in the LCE plate upon heating are shown in Fig. 6(c). The deformation process is shown in movie S1 (in ESI). It is worth mentioning that the changes in shape and transparency of LCE units are fully reversible. When the temperature returns to room temperature, the LCE plate recovers to its initial state and redisplay the erroneous message.

CONCLUSIONS

In summary, we present a novel method for the simple and rapid preparation of pillar array structures using a solvent-free LCE resin. The fabrication time of the active LCE can be shortened to 4 h without the need to evaporate the solvent. By ad-

justing the molecular weight and content of the flexible non-liquid crystal acrylate backbones, we can finely tune the phase transition temperature T_i of the LCE resin. The resulting LCE pillar array structures not only demonstrate various deformation modes, but also exhibit temperature-dependent sequential actuation. Furthermore, we fabricate a large plate containing LCE pillar units with different T_i , enabling the display of various patterns at different temperatures and achieving multi-level information encryption. The correct message can be decrypted at a specific temperature. This LCE pillar array offers a new strategy and extensive design potential for stimuli-responsive materials and active structures.

Conflict of Interests

The authors declare no interest conflict.

Electronic Supplementary Information

Electronic supplementary information (ESI) is available free of charge in the online version of this article at <http://doi.org/10.1007/s10118-025-3322-x>.

Data Availability Statement

The data supporting the findings of this study are available from the corresponding author upon reasonable request. Author

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