

Polarization-dependent and mechanochromic dual-encryption composite textile enabled by robust and resilient cholesteric liquid crystal elastomer

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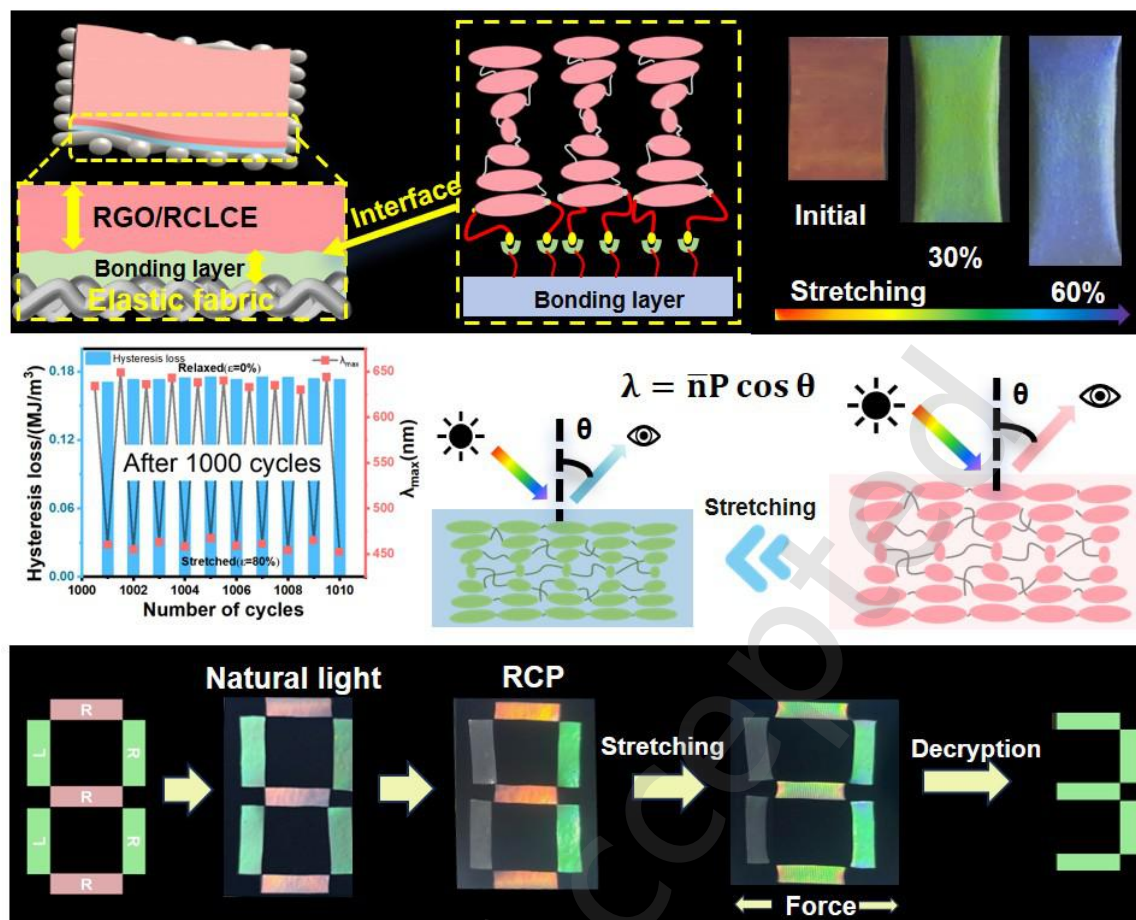
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Liquid crystal elastomer composite films with excellent color uniformity, good mechanochromic sensitivity, and remarkable mechanical properties were fabricated via a synergistic strategy integrating shear-induced alignment and RGO doping. The circular polarization characteristics and mechanochromic performance of the composite textile were further exploited to explore the applications in information-encrypted textiles.

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ABSTRACT: Mechanochromic textile has attracted growing interest for applications such as limb motion monitoring, sensing and information encryption. Cholesteric liquid crystal elastomer (CLCE) is a promising candidate for mechanochromic textile due to its flexibility and ease of adhesion to textile substrates. However, synergistically improving the mechanochromic sensitivity, color uniformity, and reversible stability of CLCE remains a major challenge. Herein, we leveraged the density compatibility and intermolecular interaction between reduced graphene oxide (RGO), dichloromethane (DCM) and CLCE prepolymer to achieve uniform and stable RGO dispersion. Scrapping shear force was applied to induce alignment of both liquid crystal mesogens and RGO sheets. Subsequent solvent evaporation facilitated the helical self-assembly of cholesteric mesogens, producing RGO/CLCE composite films with high mechanochromic sensitivity, excellent color uniformity, and superior mechanical performance. These films were then integrated with elastic fabrics through thermal bonding, forming a composite textile with both physical and chemical adhesion. Notably, the resulting composite textile exhibited stable resilience and reversible mechanochromic behavior over 1000 stretching-recovery cycles. Furthermore, leveraging the circular polarization and mechanochromic properties, the composite textile enabled a dual-mode information encryption strategy. Thus, this work deepens the understanding of the interplay between microstructure, mechanical deformation, and optical properties in CLCEs, paving the way for advanced functional textile and multifunctional device.

KEYWORDS: cholesteric liquid crystal elastomer, mechanochromic, composite textile, sensitivity, information encryption

1 Introduction

Structural color materials generate vivid and reversible colors through the interaction between light and micro-nano periodic structures [1], and they exhibit irreplaceable application value in frontier fields such as intelligent sensing [2-4], information encryption [5-7], flexible electronics [8], and bionic devices [9, 10]. Recently, structural color materials have also received increasing attention compared to pigment color materials [11-13]. As a type of intelligent material that integrates structural color properties and the mechanical advantages of elastomers, cholesteric liquid crystal elastomers (CLCEs) possess a unique helical superstructure that can be altered by external stimuli (e.g., force, heat), enabling dynamic color switching. This makes CLCEs a core candidate material for fabricating high-performance responsive optical devices [14-16]. In particular, the selective reflection of polarized light by CLCEs endows them with great potential in application scenarios such as information security [6]. However, traditional CLCEs are still constrained by multiple core bottlenecks: precise regulation of liquid crystal molecular orientation is highly

challenging, directly leading to poor color uniformity and insufficient mechanochromic response sensitivity [17, 18]; the low mechanical modulus of CLCEs makes it difficult to meet structural support requirements in practical application scenarios [19].

To achieve the controllable fabrication of CLCEs, researchers have developed techniques including the direct ink writing [20-22], template assistance [23-27], spray coating [15], and dry spinning [28], etc. However, direct ink writing (DIW) method requires strict control of parameters such as printing temperature, speed, and pressure; moreover, only unidirectional printing paths can ensure uniformity, and mesogen misorientation tends to occur in large-area or alternating-path printing [20]. Li et al. prepared cholesteric helical structures by spray-depositing CLC coatings: Marangoni flow induced by solvent evaporation drives liquid crystal molecules to self-assemble and orient along flow channels, followed by photocuring. However, it is limited to toluene as the optimal solvent. Solvent with excessively high polarity (e.g., acetone, DMF) and low boiling points (e.g., dichloromethane) cannot achieve effective orientation, resulting in poor solvent compatibility [15]. Geng et al. realized the directional

arrangement of cholesteric helical structures in CLCE fibers via the synergistic effect of stretching-induced pre-orientation and solvent evaporation-induced orientation; however, this approach leads to surface orientation defects: a “skin layer” forms on the fiber surface due to rapid solvent evaporation, causing local helical orientation to deviate from the vertical direction and generating scattered light spots [28]. Although these techniques address orientation regulation issues to some extent, the optical uniformity of the material surface and the mechanical properties still need further improvement, which severely restricts the performance breakthrough and application expansion of CLCE materials.

Inorganic powders have been used in CLCEs for modification, synergistically enhancing the mechanical strength and responsive performance [29]. However, existing preparation strategies have significant common defects: in the ways of solvent evaporation induction or solution casting, nanoscale fillers tend to undergo spontaneous sedimentation and aggregation due to factors like high density and insufficient interfacial bonding with the matrix, eventually forming gradient structures or Janus structures with filler enrichment at the bottom of the film [30]. To address these issues, modifications of inorganic powders or CLCE matrices have been utilized to enhance interfacial interactions. For example, carbon nanotubes (CNTs) modified with polyvinylpyrrolidone (PVP), were doped into bistable CLCEs via ultrasonic dispersion and UV crosslinking technology, preparing liquid crystal films that can be selectively erased by electricity, heat, or near-infrared light. However, CNTs were still prone to spontaneous aggregation and large-scale agglomeration occurred at high concentrations, leading to reduced uniformity of liquid crystal textures and increased defects, which impaired performance stability [31]. Furthermore, isocyanate groups were introduced onto CLCE surfaces by grafting 2-ICEMA, which covalently connected with MXene, preparing MXene-CLCEs with visible-infrared-radar multispectral camouflage. However, MXene layer has insufficient tensile resistance, tends to delaminate from the matrix, and causes surface cracks in the composite [32]. Therefore, it remains challenging to simultaneously achieve precise orientation regulation of liquid crystal molecules and uniform dispersion of nanoscale fillers, as well as synergistically improve mechanochromic sensitivity and mechanical performance.

Herein, this work firstly dispersed reduced graphene oxide (RGO) in dichloromethane (DCM) to achieve a stable and uniform suspension. The suspension was then dispersed into CLCE prepolymers, and the viscosity matching between the dispersion and prepolymers was regulated to ensure the preparation of a homogeneous RGO/CLCE mixture. RGO-doped cholesteric liquid crystal elastomer composite films were prepared using a scratching shearing-solvent evaporation self-assembly method, which synergistically improves mechanochromic sensitivity, mechanical strength, and surface color uniformity. Furthermore, the films are compounded with elastic fabrics to explore their applications in visual sensing, information encryption, and camouflage. This work not only solves the key problems of

nanoscale filler agglomeration and bottom enrichment in CLCEs but also provides a simple and efficient preparation method for orientation regulation and performance optimization of liquid crystal-based intelligent optical materials, laying an important foundation for expanding the applications of multifunctional intelligent soft materials.

2 Experimental Section

2.1 Materials

1,4-Bis-[4-(3-acryloyloxypropoxy) benzoyloxy]-2-methylbenzene (RM257, purity: 98%) and pentaerythritol tetra(3-mercaptopropionate) (PETMP, purity: 95%) were purchased from Bidepharm. (3R,3aS,6aS)-Hexahydrofuro[3,2-b] furan-3,6-diyl bis(4-(4-(4-(acryloyloxy) butoxy) carbonyloxy) benzoyloxy) benzoate) (LC756, purity: 90%) was purchased from Adamas-beta. 5,6-Dihydro-5-(trans-4-propylcyclohexyl)-4H-dinaphtho(2,1-F:1,2-h) (1,5) dioxacyclononatetraene (S5011, purity: 99%) was purchased from Rhawn. 2,2'-(Ethylenedioxy) diethanethiol (EDDET, purity: 97%), 2,2-dimethoxy-2-phenylacetophenone (I-651, purity: 99%) and dipropylamine (DPA, purity: 99%) were purchased from Aladdin. Dichloromethane (DCM, AR) was purchased from Sinopharm.

2.2 Precursor synthesis and fabrication of CLCE

mechanochromic films

Precursor synthesis. RM257, LC756 or S5011 were dissolved in DCM. Then, PETMP, EDDET and I-651 were added into the solution. DPA (diluted with DCM of 1: 50 v/v ratio) was added as a catalyst for the first-stage Michael addition reaction to turn the system into precursor with low crosslinking density. The reaction time was controlled for 30 minutes at room temperature. Subsequently, this precursor was rapidly used to prepare films.

Preparation of CLCE Films. Using a glass plate as the substrate, the precursor solution was uniformly coated on the glass plate by a film applicator (KTQ- II adjustable film applicator) in a scratching manner. Then, it was placed at 30 °C for the solvent evaporation self-assembly for 4h. After the solvent evaporated, the film was irradiated with ultraviolet light (365 nm, 50 mW·cm⁻², 5 min) for cross-linking. The CLCE films doped with reduced graphene oxide (RGO) were prepared by adding RGO/DCM dispersion (0.1 g/mL, m/v, 400-600 μL) to the above precursor solution. The fabrication process was following as above. For right-handed chiral CLCE (RCLCE) films with red, yellow, green and blue colors, the weight ratios (k₁) of the chiral monomer LC756 to RM257 were 0.045, 0.050, 0.055 and 0.060, respectively, and the formulations are shown in Table S1. For left-handed chiral CLCE (LCLCE) films with red, yellow, green and blue colors, the weight ratios (k₂) of the chiral monomer S5011 to LC756 were 0.893, 0.942, 0.975 and 1, respectively, and the formulations are shown in Table S2.

2.3 Characterization

The water contact angles were investigated with a drop-shaped analyzer (DSA 100, Kruss). POM images were taken with a Zeiss Primotech for microscopic observation. 2D WAXS experiment was conducted on a Xenocs Xeuss 2.0 Discover diffractometer in transmission mode. The reflection spectra were recorded using an ideaoptics PG-2000 spectrometer. Then, the color trends of the samples were plotted in the CIE 1931 chromaticity diagram. Scanning electron microscope (SEM) images were taken on a Hitachi SU5000 scanning electron microscope analyzer. Mechanical stress-strain experiments were conducted using an INSTRON 5967 instrument, and CLCE film samples were stretched at a straining rate of 50 mm/min. The films were cycled 1000 times at the same rate $\epsilon_{xx} = 0-0.8-0$, and held for 180 seconds after each cycle. The average refractive index (n_{avg}) experiments were carried out using a Horoba UVISEL PLUS instrument. Differential scanning calorimetry (DSC, NETZSCH DSC 204F1) was performed with heat from -40 to 240 °C at a scanning rate of 10 °C/min. Circular dichroism spectra were measured using a JASCO1700 circular dichroism spectrometer. The friction resistance test was carried out using an SDLATLAS M2359A instrument, and the dynamic thermomechanical analysis test was completed using a Hitachi DMA 7100 instrument.

3 Results and discussion

3.1 Fabrication strategy of CLCE films

Figure 1 systematically demonstrates the process of design, preparation, orientation, and self-assembly for cholesteric liquid crystal elastomers (CLCE) and RGO/CLCE composites, along with the characterization results of surface wettability on the top and bottom surfaces of the film. As shown in Figure 1A, this work constructs intelligent elastomers with photo-crosslinked liquid crystal elements as the core component. The main monomer RM257 provides the basis for ordered liquid crystal phase arrangement, while the terminal acrylate groups serve as active sites for thiol-ene click crosslinking. The crosslinking system employs a blend of PETMP and EDDT, which ensures network stability and confers reversible deformation capability. The chiral dopant LC756 and S5011 were used to control the pitch and helix direction of the liquid crystal. Figure 1B illustrates the fabrication process of mechanochromic chiral liquid crystal elastomer (CLCE) films and the schematic diagram of the mechanochromic mechanism. The RGO morphology is displayed in Figure S1. Figure 1C shows the prepared RGO/RCLCE precursor and RGO/DCM suspension. It demonstrates that the suspension prepared by dispersing RGO in RCLCE precursor exhibits superior stability, remaining homogeneous and stable even after 30 minutes of incubation. Powdered reduced graphene oxide (RGO) has a density of 1.8-2.1 g/cm³, compared with 1.325 g/cm³ for dichloromethane at 25 °C and 3.96 g/cm³ (measured by ourselves) for the precursor solution. Accordingly, RGO is capable of forming a stable suspension within the precursor

solution. Furthermore, RGO achieves stable dispersion in dichloromethane solution of CLCE precursors, constituting a representative solvent-solute-additive ternary synergistic system. The amphiphilic architecture of RGO enables interfacial interactions with solvent and precursor. The lipophilic unoxidized graphitic regions of RGO exhibit affinity toward dichloromethane and aromatic moieties within CLCEs through π - π stacking, whereas the hydrophilic residual oxygenated functional groups interact with solvent and precursor molecules via dipole-dipole coupling and hydrogen bonding [33]. CLCE precursor macromolecules are capable of adsorbing onto RGO nanosheet surfaces. The considerable molecular dimension of these precursors introduces prominent steric repulsion, efficiently suppressing van der Waals-induced restacking and aggregation of RGO laminates.

The shear-induced preorientation and evaporation-induced self-assembly (EISA) were used to control the self-assembly of liquid crystal for mechanochromic cholesteric liquid crystal elastomer films with regular helical nanostructures. The film surface immediately exhibits vivid structural colors, followed by ultraviolet (UV) irradiation photo-crosslinking. The film thickness can be controlled by adjusting the viscosity of the precursor and the coating thickness. In this study, the film thickness was about 100 μ m in order to optimize the mechanical properties and the mechanochromic sensitivity of the film. Secondary alignment control of liquid crystal elements is achieved during external stretching of the film, and the surface color of the film changes by adjusting the pitch. CLCEs can exhibit selective reflection at a specific wavelength (λ) that varies with the pitch, in accordance with Bragg's law [34]:

$$\lambda = n_{avg} P \cos \theta \quad (1)$$

where θ is the angle of incident light and n_{avg} is the average refractive index of CLCE.

Figure 1D displays the static water contact angles of the right-handed chiral CLCE (RCLCE) and RGO/RCLCE composite films. The RCLCE film exhibits an angle of 95° on the top surface and 81° on the bottom surface, resulting from the top and bottom surface composition and morphological differences induced by the scrapping-induced alignment of liquid crystal mesogens. After introducing RGO, the water contact angle on the top surface decreases to 86° and on the bottom surface to 72°, indicating improved hydrophilicity on both surfaces. This enhancement is attributed to the oxygen-containing functional groups on the RGO surface, which increase hydrophilicity, and the synergistic effect of surface roughness between the top and bottom surfaces due to the dispersion of RGO within the liquid crystal elastomer [35]. Moreover, as shown in Figures S2, RGO is uniformly dispersed within the CLCE matrix. Both the top and bottom surfaces of the composite film exhibit roughness. This is consistent with the above analysis, demonstrating that RGO can be uniformly suspended in CLCE precursors. After the scrapping process, RGO remains evenly dispersed in the matrix, ensuring both surfaces contain RGO. This is different from the reported Janus structure after adding inorganic additives to CLCE [30].

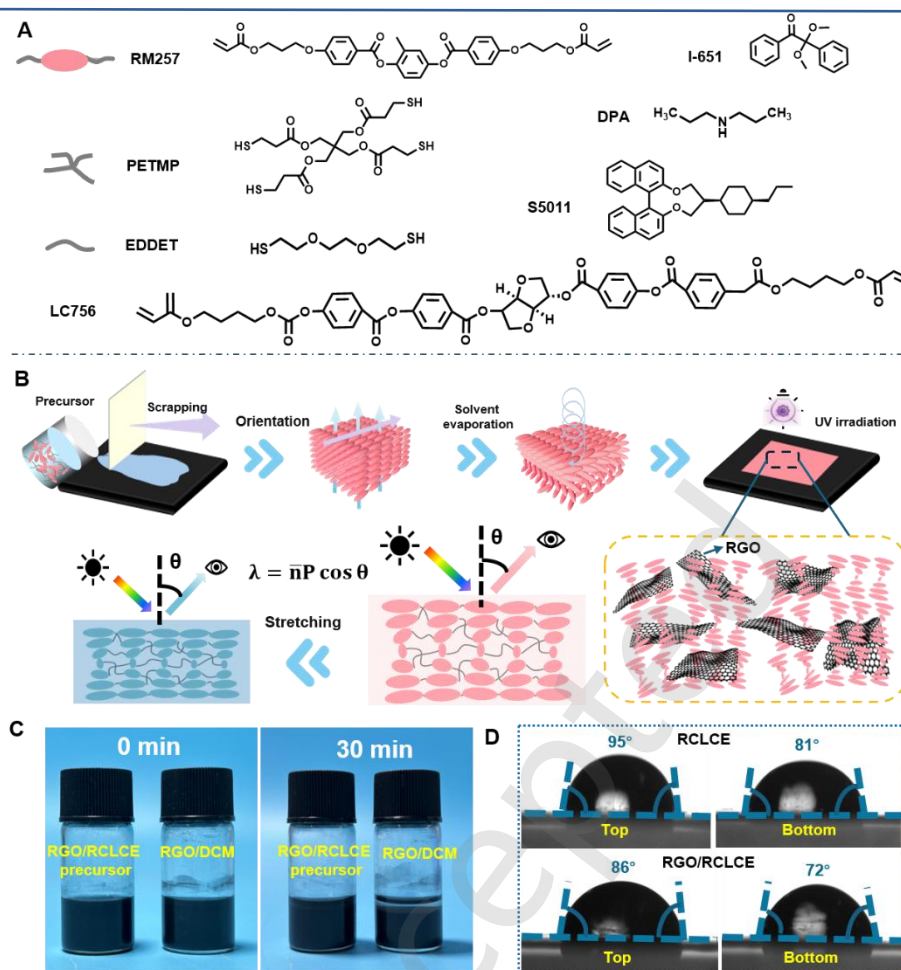


Figure 1 (A) Chemical structures of monomers, chiral dopants, cross-linking agent, catalyst and initiator used in the study. (B) Schematic illustration of fabricating color-tunable RGO/CLCE composite film and mechanochromic mechanism. (C) The suspension of RGO/RCLCE precursor and RGO/DCM. (D) The water contact angles of the top and bottom surface of the RCLCE film and RGO/RCLCE composite film.

3.2 Control of the structural coloration and characterization of CLCE films

As mentioned above, the initial color of the CLCE film can be controlled by adjusting the type and content of chiral reagents by this fabrication method. Figure 2 presents the optical properties, microstructure, and application demonstration of the RGO/RCLCE composite films. Figure 2A shows that the composite films exhibit red, yellow, green, and blue colors under natural light. Under right-circularly polarized (RCP) light, it displays the same colors as natural light, while under left-circularly polarized (LCP) light, it appears nearly black. It directly reflects the circularly polarized selective reflection property of cholesteric liquid crystals, which only reflect circularly polarized light with the same helical orientation [36]. For comparison, Figure S3 displays optical images of RCLCE and LCLCE films prepared without RGO. Under circularly polarized light, the observed patterns are consistent with the aforementioned results. Subsequently, according to the mathematical transformation of the CIE 1976 Lab color space [37], we used the reflectance spectral data to calculate the L^* , a^* , and b^* values of RCLCE and RGO/RCLCE films, respectively. The reflectance spectra of different five points were measured on the same film. As quantified in Table S3, L^* mean value of RCLCE and

RGO/RCLCE films is 84.02 and 89.59, respectively, and coefficient of variation (CV%) values of L^* measured 1.12 for RCLCE and 0.85 for RGO/RCLCE film. Therefore, comparative analysis reveals that the addition of RGO enhances the color saturation and lightness of the film, which is ascribed to that the black RGO effectively absorbs scattered light. RGO/RCLCE film exhibits superior color uniformity.

As shown in Figure 2B and Figure S4, circular dichroism (CD) spectra demonstrate the different chiral properties of RCLCE, LCLCE and RGO/RCLCE [38]. The absorption spectra in Figure 2C show that RGO/RCLCE films exhibit two characteristic absorption peaks at approximately 260 nm and 300 nm. The former is attributed to the π - π^* electron transition of sp^2 hybridized carbon atoms in the RGO layers [39, 40]. In contrast, pure RCLCE exhibits only a broad absorption peak in 260-300 nm, which corresponds to the electronic transition overlap of characteristic functional groups (e.g., benzene rings, acryloyl groups) in the liquid crystal monomer. It is worth emphasizing that after RGO doping, the unique broad absorption peak of pure RCLCE disappears, and RGO/RCLCE exhibits two distinct characteristic peaks. This phenomenon indicates the interaction between RGO and liquid crystal mesogens, with the mechanism being that the π - π stacking effect between RGO and liquid crystal mesogens regulates the electronic transition environment of the liquid crystal mesogens.

Simultaneously, it also demonstrates that RGO is uniformly dispersed in the RCLCE matrix without significant aggregation. Figure 2D shows weak linear dichroism (LD) signals, further confirming that the film's color originates from Bragg reflection rather than light absorption. In the visible light region (400-700 nm), both samples maintain low absorbance (<0.1) with stable baselines, indicating that RGO doping does not disrupt the inherent selective

reflection properties of the cholesteric liquid crystal phase [41]. Therefore, while preserving the helical structure integrity of RCLCE to maintain its mechanochromic property, RGO enhances the orderliness of liquid crystal mesogens through its lamellar orientation-induced effect. This provides both experimental evidence and theoretical support for improving the mechanical properties and functional integration of RCLCE-based composites.

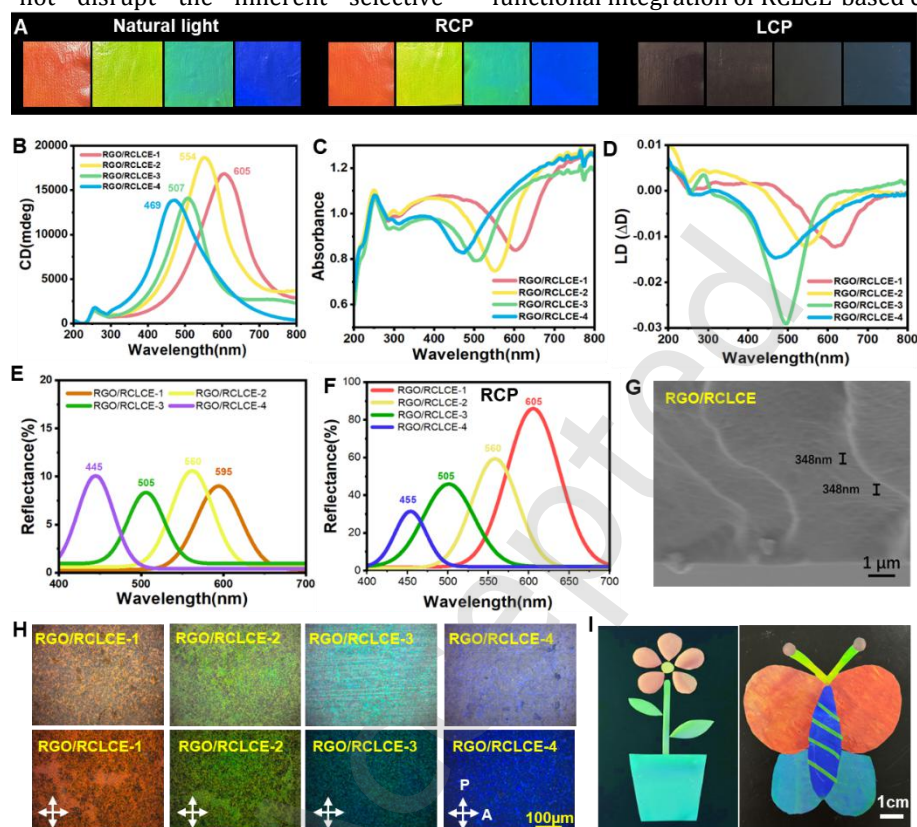


Figure 2 (A) RGO/RCLCE composite films with different colors under natural light, right-circularly polarized (RCP) light and left-circularly polarized (LCP) light. (B, C, D) Circular dichroism spectra of RGO/RCLCE composite films with different colors. The selectively reflected light spectra corresponding to RGO/RCLCE composite films (E) without and (F) with right-handed circular polarizer. (G) SEM image of RGO/RCLCE composite film with original red color. (H) The microscopy photos and reflected POM images of the top surface of RGO/RCLCE composite films with different original colors. (I) Multi-colored flower and butterfly patterns fabricated with RGO/RCLCE composite films

In addition, the characteristic peak positions (469 nm, 507 nm, 555 nm, 605 nm) of circular dichroism (CD) spectra in Figure 2B correspond to the apparent color and the reflective wavelength in Figure 2E. Compared with the spectra of RCLCE in Figure S4 and Figure S5, no additional CD impurity peaks are observed across the full wavelength range. However, the film colors exhibit a slight blue shift after RGO doping, which further confirmed the uniform dispersion of RGO in the matrix. It is displayed from Figure 2F and Figure S5C that the reflectivity of the CLCE films is improved by the polarizer, which verifies the optical anisotropy and polarization response characteristics of RGO/RCLCE and RCLCE film [42, 43]. In Figure 2G and Figure 2E, the pitches of RGO/RCLCE and RCLCE with the same initial color were measured by SEM. The refractive index of the films is 1.68 and 1.66, respectively, which were measured by an ellipsometer. According to Bragg's law in Eq. (1), the calculated reflective wavelength of RGO/RCLCE and RCLCE are ~ 585 nm and ~ 606 nm, respectively, which are consistent with the results in Figure 2E (595 nm) and Figure S6B (645 nm). As can be seen, the addition of RGO causes a

blue shift of the film's color, which is consistent with the optical images of the film previously displayed. Optical photographs, POM images and reflection spectra of the RGO/RCLCE composite film's bottom surface were measured, as shown in Figure S7 and Figure S8. The bottom surface displays a paler color and a noticeable red shift compared with the top surface, which is consistent with the reflected spectra: the characteristic reflection wavelengths show a moderate red shift relative to Figure 2E. This phenomenon can be attributed to the restricted solvent evaporation at the bottom surface of the film during the assembly process. This is also consistent with the aforementioned differences in contact angles between the top and bottom surfaces of the composite film.

Furthermore, the micro-structural morphology and liquid crystal texture of RGO/RCLCE composite films and RCLCE can also be further obtained from Figure 2H and Figure S9. In Figure S9, reflective optical micrographs (top row) and cross-polarized optical microscopy (POM) images (bottom row) of neat cholesteric liquid crystalline elastomer films (RCLCE-1 to RCLCE-4) exhibit tunable structural color

gradually shifting from orange-red to blue, which originates from the continuous reduction of cholesteric helical pitch induced by enhanced helical twisting power. Under orthogonal polarizer (P) and analyzer (A), all samples display uniform dense fingerprint texture characteristic of chiral nematic (N^*) liquid crystal without large-scale oily streak networks [44, 45]. In Figure 2H, tiny dark speckles in RGO/RCLCE groups correspond to some RGO aggregates rather than liquid crystalline topological defects. Therefore, we can get the liquid crystal elastomer film with full visible spectrum color by regulating the composition and fabrication method, so that a variety of gorgeous patterns can be got easily.

3.3 Mechanical and mechanochromic properties

As previously mentioned, the incorporation of RGO is expected to enhance the mechanical strength of liquid crystal elastomers. Figure 3A presents the stress-strain curves of the films. The results demonstrate that the composite films exhibit a significant increase in fracture

strength after RGO addition. Young's modulus of RGO/RCLCE reaches 0.103 MPa, significantly higher than the 0.029 MPa of RCLCE, which directly demonstrates the mechanical reinforcement effect of RGO on elastomers. It is consistent with the reported results [46, 47]. Comparing the results in Figure 3B and Figure S10, the content of RGO in the film is approximately 7%, which is consistent with the fabrication formular. The XRD spectra in Figure 3C and Figure S11 show that the characteristic peak of RGO at approximately 26.4° , confirming that RGO retains its characteristic properties in the matrix and is well dispersed. This provides structural support for the ordered structure regulation and performance synergy of the composite film. For Comparison, we adjusted the addition amount to 5% and examined changes in film color, reflected spectrum, and mechanical properties, as shown in Figures S12 and Figures S13. It is observed that under fixed other component proportions, lower RGO content results in lower color brightness and saturation, red-shifted reflection wavelengths, and poorer mechanical performance relative to the composite film with 7 wt% RGO.

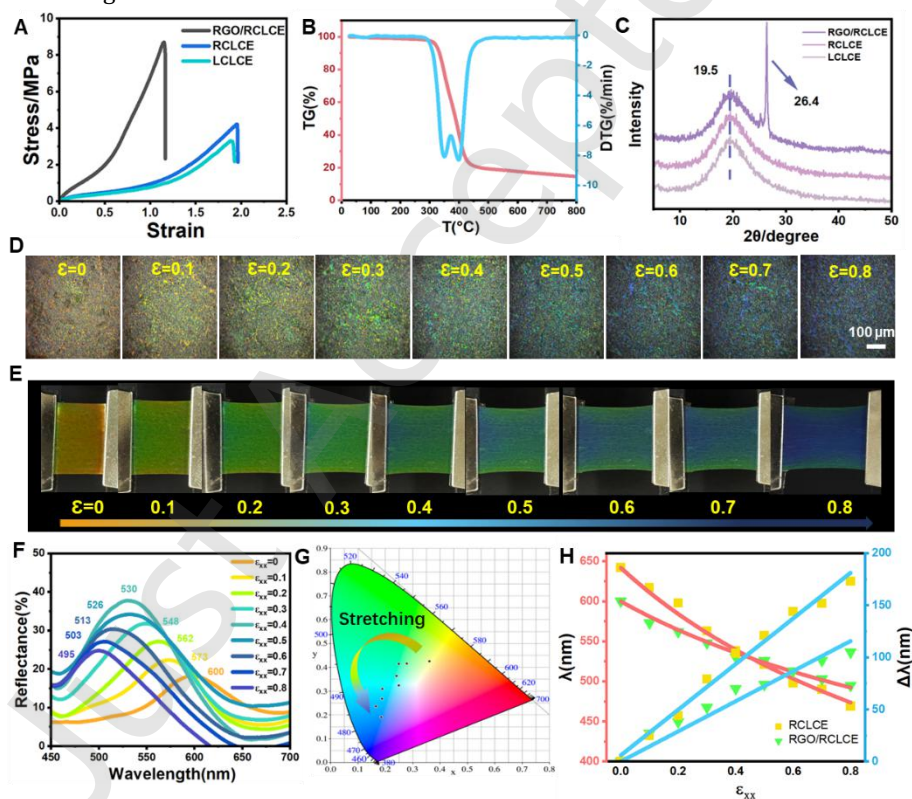


Figure 3 (A) The stress-strain curves of RCLCE, LCLCE and RGO/RCLCE films. (B) TGA and DTG curves of RGO/RCLCE film. (C) XRD curves of RCLCE, LCLCE and RGO/RCLCE films. (D, E) Microphotographs and optical photographs of initially red-reflecting RGO/RCLCE film under uniaxial mechanical stretching strain from 0.0 to 0.8. (F) Reflection spectra of RGO/RCLCE film at length strain from 0.0 to 0.8, respectively. (G) Change in the reflective color under a length strain of 0.00 to 0.8 on the CIE 1931 color space during stretching. (H) Reflection wavelength λ versus the uniaxial strain (ϵ_{xx}). The black line is the best fit for Equation (2).

The mechanochromic properties are investigated by stretching the RGO/RCLCE composite film (Figure 3D, Figure 3E and Video S1). They demonstrate the internal texture evolution of the composite film and its continuous color transition from yellow to green to blue during stretching. The reflective spectra in Figure 3F further reveal a distinct blue shift in the film's surface color as strain increases. The CIE 1931 coordinate diagram in Figure 3G further confirms the color change, which is attributed to that optical property

reconfiguration is achieved by altering the helix pitch of the cholesteric liquid crystal under stretching. Moreover, the mechanochromic response of RGO/RCLCE and RCLCE is analyzed by considering a Poisson's ratio (ν) of the films. According to the reported work [23], we can thus fit a function

$$\lambda = \lambda_0 (1 + \epsilon_{xx})^{-\nu} \quad (2)$$

to obtain the Poisson's ratio (ν) from the mechanochromic

reflective spectra, assuming constant v . By fitting Eq. (2) to the mechanochromic reflection spectra, the curves are shown in Figure 3H, and v of the RGO/RCLCE and RCLCE is ~ 0.34 and ~ 0.5 , respectively. The Poisson's ratio (ν) of RGO/RCLCE and RCLCE measured by the actual dimension change during stretching is ~ 0.45 and ~ 0.5 . As is well known, the Poisson's ratio of rubber materials approaches 0.5, exhibiting optimal elasticity. Consequently, the addition of RGO results in a certain degree of elasticity reduction of liquid crystal elastomer films. We plot the wavelength shift ($\Delta\lambda$) versus uniaxial stretching strain ($\Delta\lambda \sim \epsilon_{xx}$), as shown in Figure 3H. The curve slopes show that the RCLCE has higher mechanochromic sensitivity when the film is stretched to different strain. In addition, when the strain is below 0.5, both the initial and post-strain colors of RGO/RCLCE exhibit a blue shift. Conversely, when the strain exceeds 0.5, RGO/RCLCE shows a red shift compared to RCLCE, as evidenced by the photo comparison between Figure 3E, Figure S14 and Figure S15. It is because the addition of RGO increases the film's modulus and reduces its Poisson's ratio. Moreover, RGO is interwoven within the liquid crystal helix axis, which impedes the reduction of pitch when the strain becomes sufficiently large. We define the mechanochromic sensitivity as the wavelength shift per one percent of strain ($\text{nm}\cdot\%^{-1}$). Therefore, the slope of the $\Delta\lambda \sim \epsilon_{xx}$ fitting curve in Figure 3H indicates that the mechanochromic sensitivity of RCLCE is $2.29 \text{ nm}\cdot\%^{-1}$, while the mechanochromic sensitivity of RGO/RCLCE is $1.44 \text{ nm}\cdot\%^{-1}$. Although the addition of RGO reduced the mechanochromic sensitivity of RGO/RCLCE, the shear orientation effect during scrapping resulted in a higher sensitivity for the composite film compared to that reported in the literature [26, 30, 32, 47]. In addition, we fabricated CLCE films of varying thicknesses with an initial red color and measured their reflectance spectra under different strains ($\epsilon_{xx} = 0 \sim 0.8$), as shown in Figure S16. The results indicate that as film thickness increases, the shift in reflectance spectra under the same strain becomes less pronounced, suggesting reduced mechanochromic sensitivity. Additionally, the peak width of the reflectance spectrum widens with increasing thickness, indicating diminished color uniformity across the film surface.

3.4 Mechanochromic reversibility

The elastic recovery and mechanochromic reversibility of the films were investigated by cyclic stretching. Figure 4A schematically illustrates the structural evolution of the liquid crystal pitch during the "stretching-recovery" process, intuitively demonstrating its reversible control over structure and morphology. The optical photographs and 2D WAXS spectra of the initial state and the state with 80% strain are shown in Figure 4B. 2D WAXS patterns change from a ring to a pair of arcs along the transverse direction, indicating the polydomain structure in the initial state and the monodomain structure in the stretched state, which further confirms that the liquid crystal director predominantly aligns along the direction of mechanical stretching [24, 26, 48]. According to reported method [49], the diffraction angle (2θ) was calculated from the outer ring diffraction radius to be approximately 26.37° . This angle

closely matches the position of the characteristic (002) crystal plane diffraction peak of RGO in XRD ($2\theta = 26.4^\circ$), indicating that the outer ring diffraction peak corresponds to the stacking structure of RGO layers. For comparison, the optical photographs of the initial, 80% strained and recovered RCLCE film as well as the corresponding 2D WAXS patterns are displayed in Figure S17, which demonstrates the similar result. Furthermore, based on the 2D-WAXS profile (Figure 4C and Figure S18) at the original state, the order parameter (S) of RGO/RCLCE and RCLCE films is 0.19 and 0.18, respectively. After stretching at strain $\epsilon = 80\%$, the half-peak width of the azimuth curve is 62° , and the order parameter (S) of RGO/RCLCE is calculated to be 0.22 while S is 0.24 for RCLCE film [50]. When the strain is recovered, the diffraction curve also returns to the initial state, which indicates that the liquid crystal helix structure in the film has excellent reversible recovery.

After repeated stretching of the film, the color can maintain stable change, which is also related to the mechanical properties of the film, especially its resilience. We perform cyclic tensile tests to evaluate the resilience of the film. Firstly, cyclic tensile tests of the RGO/RCLCE film are performed with gradually larger strains from 0.2 to 0.8. In these sequential cyclic tensile tests (Figure 4D and Figure S19), there is no waiting time between two consecutive loadings. Clearly, the film possesses good resilience owing to the small hysteresis loop. The mechanochromic reversibility is investigated by fiber stretching and recovery. As shown in Videos S1, the response is immediate, fully reversible, and visible to the naked eye. To evaluate the long-term durability under realistic usage conditions, the RCLCE film was measured by cyclic stretching for strain up to 0.8 and then subjected to 1000 cycles. It can be seen from Figure 4D that the RCLCE film shows the same reflectance wavelength at the same strain either during stretching or recovery. For the RGO/RCLCE composite film, three cycles were conducted at strain levels of 0.2, 0.4, 0.6, and 0.8, followed by ten additional cycles at a strain level of 0.8. The results can be seen in Figure 4E and Figure S20. It indicates that the pitch in film can recover immediately if the strain recovers according to Bragg's law [51]. The mechanochromic response is practically intact and reversible. The stress relaxation performance of RCLCE and RGO/RCLCE with $\epsilon = 80\%$ was also investigated and the result shows that both materials exhibit the stress relaxation behavior characteristic of crosslinked polymers (Figure S21). However, the stress decay rate of RGO/RCLCE is slightly higher, which also indicates that its elastic recovery performance is slightly inferior compared to that of RCLCE. Furthermore, we analyzed the thermomechanical properties and strain recovery capability of the liquid crystal elastomer using DMA and creep curve. The results (Figure 4F, Figure 4G and Figure S22) demonstrate that the liquid crystal elastomer exhibits excellent resilience at both room temperature and low-temperature environments, which is consistent with its superior reversible color-changing properties. The creep curve (Figure 4H) indicates that the strain of the film can recover quickly to the initial state after being stretched to 50% strain. All the results fully demonstrate that RCLCE and RGO/RCLCE exhibit excellent

mechanochromic reversibility and mechanical-optical stability, providing performance support for their

application in flexible intelligent devices.

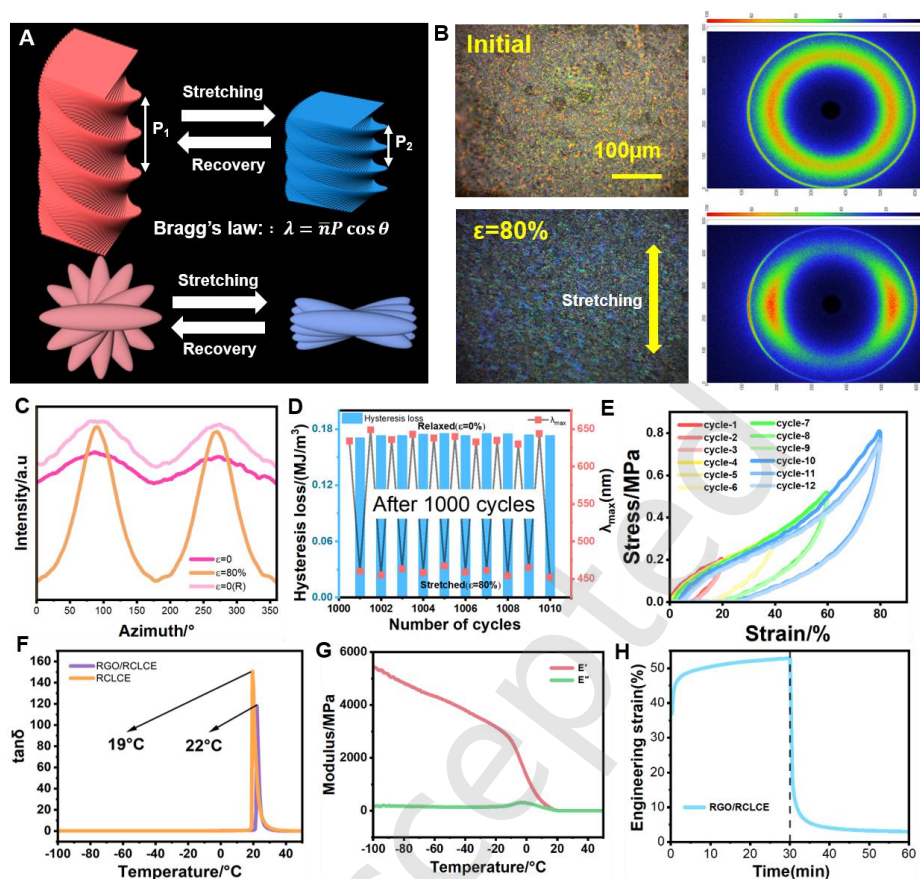


Figure 4 (A) Schematic depictions of strain-induced deformation and eventually unwinding of the cholesteric helix viewed parallel or perpendicular to the helix axis. (B) Optical photographs and the corresponding 2D WAXS patterns of the initial and 80% strained RGO/RCLCE film subjected to the stretching. (C) The scattering intensity as a function of the azimuthal angle obtained from the 2D WAXS pattern. (D) Changes of reflectance wavelength and hysteresis loss of RCLCE during 1000 times stretching-recovery cycles under a uniaxial strain of 0.0 to 0.8. (E) Hysteresis curves of RGO/RCLCE film under different strains of 0.2, 0.4, 0.6 and 0.8. (F) $\tan \delta$ versus temperature curve of RCLCE and RGO/RCLCE films. (G) Storage modulus (E') and loss modulus (E'') of RGO/RCLCE film versus temperature. (H) Creep curve of RGO/RCLCE film.

3.5 Thermochromic ability of RCLCE and RGO/RCLCE films

Figure 5 presents the thermochromic ability of RCLCE and RGO/RCLCE films. Figure 5A shows the visual color changes of RGO/RCLCE and RCLCE at different temperatures. The RGO/RCLCE film exhibits a distinct color gradient from bright green to brownish to dark as temperature rises (from low to high), while RCLCE shows a relatively gradual color change. This clearly demonstrates that the introduction of RGO significantly enhances the contrast in temperature-induced optical property modulation. Figure 5B displays the temperature-dependent changes in the green (G) component of the color for RGO/RCLCE and RCLCE films. The data clearly shows that the green component of RGO/RCLCE drops sharply as temperature rises (25–102 °C), while RCLCE exhibits a significantly slower reduction rate. This quantitative analysis demonstrates that RGO enhances the material's optical response sensitivity to temperature variations. In Figure 5C, with plant leaves as the background, the RGO/RCLCE film appears green at 25 °C, matching the

background seamlessly. When the background leaves' color changes to yellow-green, the film can be irradiated with 808 nm NIR irradiation for 10s or shrined, and the film can change to a yellow-green color like the background, to achieve the purpose of camouflage. It demonstrates the potential application value of the film's thermochromic ability in camouflage. DSC curves indicate that RGO/RCLCE and RCLCE films possess LC-to-isotropic transition temperature (T_i) at 42 °C and 56 °C (Figure S23), respectively. Benefiting from the high thermal conductivity of RGO, the nanosheets serve as abundant internal heat hotspots, which rapidly raise the interior temperature of the film and create local temperatures higher than the surrounding environment, further facilitating the liquid crystal phase transition. Moreover, RGO was intercalated between the molecular chains of the liquid crystal elastomer, dilution effect arising from the layered structure accounts for the substantial reduction of the clearing temperature [52]. These factors reduced the LC-to-isotropic transition temperature from 56 °C of pristine RCLCE to 42 °C for the RGO/RCLCE composite. In addition, as documented in Figure 5D, RGO/RCLCE films with different initial colors can change color with temperature variations, exhibiting

thermochromic abilities.

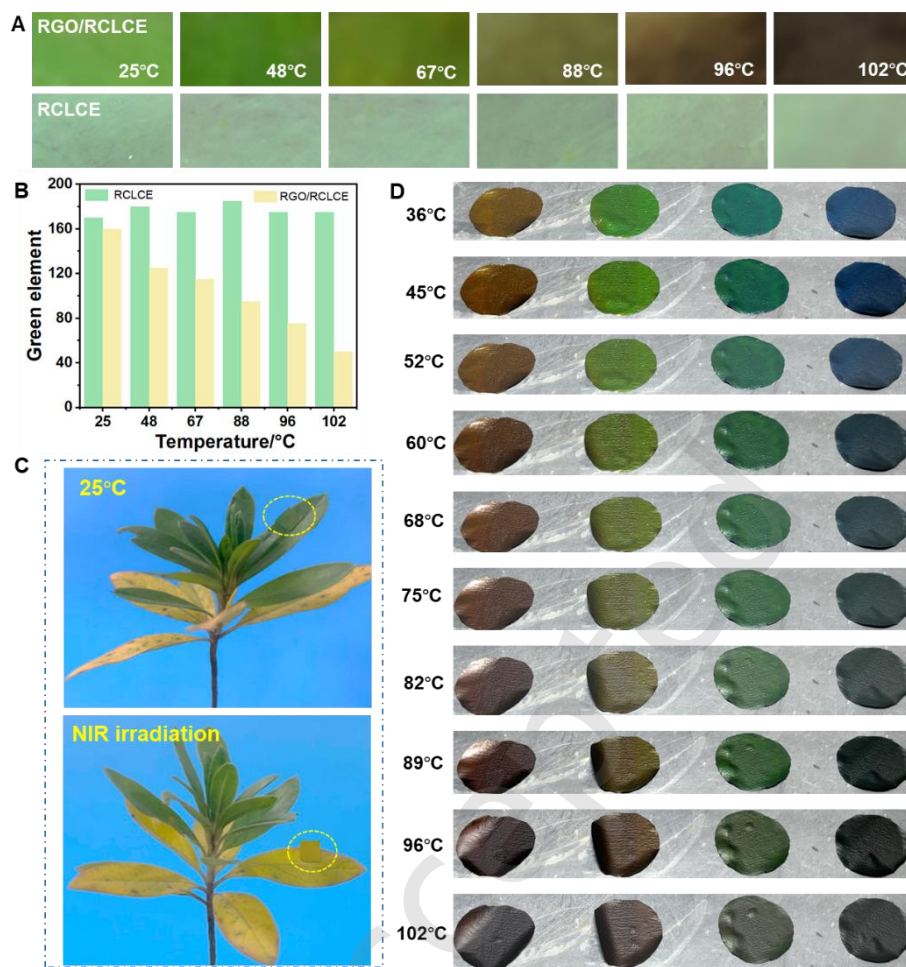


Figure 5 (A) Optical photographs of color changing of RCLCE and RGO/RCLCE films versus temperature. (B) Changes of a green element in the CIE xy chromaticity diagram during temperature increasing. (C) The color changes of RGO/RCLCE film on plant leaves with temperature variations. (D) The color changes of RGO/RCLCE films with different original colors during temperature increasing.

3.6 Fabrication of composite textile and properties

We integrated composite films with elastic fabrics to investigate the application value in smart textile. Figure 6 systematically demonstrates the structural design and mechanical-optical properties of RGO/RCLCE composite textile, illustrating the material's progression from structural construction to practical application scenarios. Figure 6A demonstrates a sandwich composite structure design comprising RGO/RLCE films, a bonding layer, and spandex elastic fabric. The thermoplastic polyurethane (PU) hot-melt adhesive serves as the flexible bonding layer, which not only enhances the system's mechanical load-bearing capacity but also ensures the material's flexibility and stretchability. The RGO/RCLCE film contains incompletely reacted double bonds (Figure S24A), while PU hot-melt adhesive contains amino groups (Figure S24B). Through thermal pressing, the double bonds undergo Michael addition reactions with the amino groups, covalently linking the mechanochromic layer to the bonding layer (Figure S25). Meanwhile, due to the uneven surface of the elastic fabric, during hot pressing, the hot melt adhesive can penetrate into the gaps of the elastic fabric. Moreover, both the hot melt adhesive and the elastic fabric are made of polyurethane, which exhibits good compatibility. Therefore, the interfacial force between the

bonding layer and the substrate fabric is enhanced through both physical and chemical interactions. The abrasive resistance was also investigated, and it can be seen in Figure S26 that the surface characteristics of the composite textile remained unchanged after undergoing 40,000 friction tests. Therefore, the interfacial forces between the mechanochromic layer, the bonding layer and the fabric layer are significantly enhanced, which can realize the good mechanical properties of the composite textile and avoid the delamination of the mechanochromic layer [7, 53, 54]. Figure 6B displays the cross-sectional morphology of the composite textile, showing that the three layers are tightly bonded together.

In Figure 6C, the composite textile exhibits a continuous color transition from red to green to blue during the stretching process (strain: 0→30%→60%), demonstrating sensitive color-changing capability under tensile stress. Figure 6D presents the DMA curve of the composite textile. The results demonstrate that, similar to the composite RGO/RCLCE film, the composite textile exhibits excellent elasticity at room temperature. It confirms that the hot-press bonding process did not compromise the inherent properties of the RGO/RCLCE film. Figure 6E demonstrates the reflective wavelength variation of composite textile during repeated 1000 stretching-recovery cycles (strain:

0→80%→0). As can be observed from the figure, as the tensile-relaxation cycles increase, the reflected wavelength on the composite textile surface does not change significantly; this also demonstrates the mechanochromic stability of the composite textile. Figure 6F and Video S2 present the composite textile fabricated in this study, while

Figure 6G and Video S3 illustrate the color changes of the composite textile on the elastic glove when fingers are flexed from 0° to 90°. These findings indicate that the composite textile can serve as a visual indicator for assessing finger joint mobility.

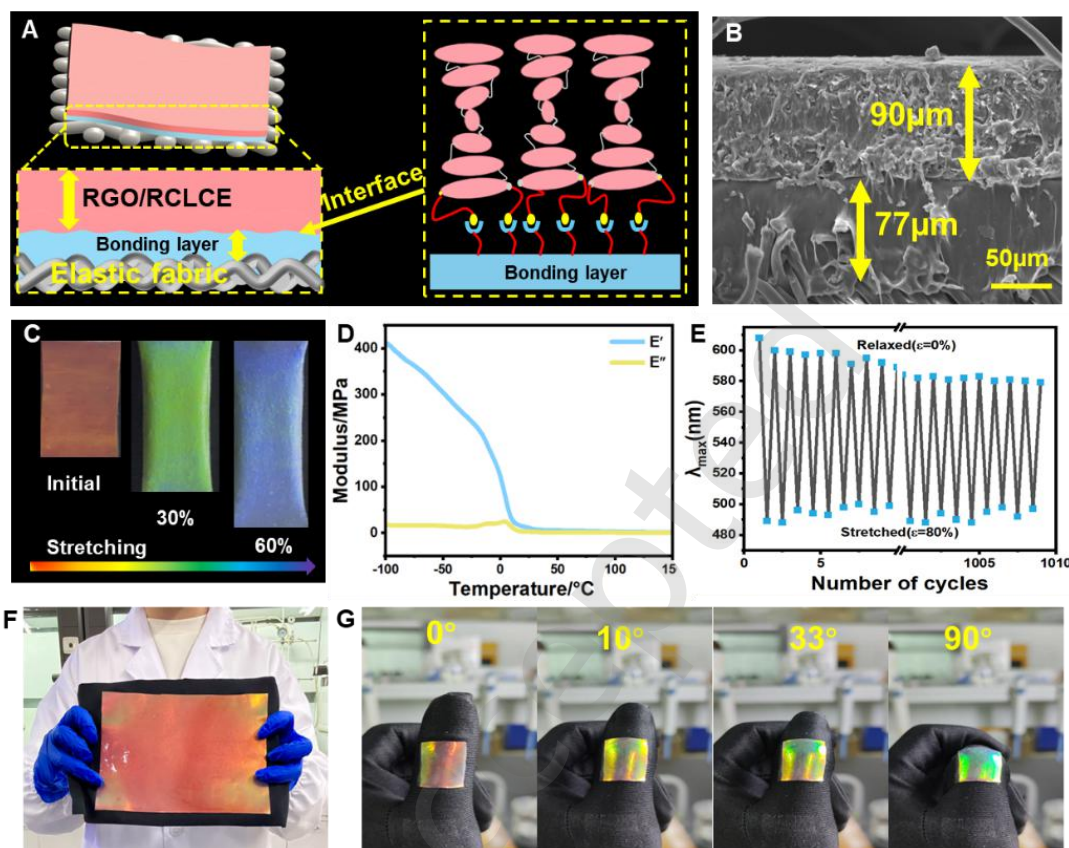


Figure 6 (A) Schematic illustration of composite textile. (B) SEM image of composite textile. (C) Optical photographs of initially red-reflecting composite textile under uniaxial mechanical stretching strain from 0 to 60%. (D) Storage modulus (E') and loss modulus (E'') of composite textile versus temperature. (E) Changes of reflectance wavelength of composite textile during 1000 times stretching-recovery cycles under a uniaxial strain of 0.0 to 0.8. (F) The photograph of composite textile. (G) Composite textile for monitoring finger joint activity angle.

3.7 Composite textile for information encryption

In addition to its mechanochromic property, cholesteric liquid crystals exhibit a distinctive circular polarization characteristic [51], as illustrated in Figures 2B-2D. Therefore, we explore the application of composite textile in the field of information encryption by using the circular polarization characteristics of cholesteric liquid crystal. In Figure 7A, a composite liquid crystal elastomer film with varying optical properties is integrated with elastic fabric while encoding information. The composite textile displays a uniform blue pattern under natural light, showing distinct QR codes under left-handed (LCP) and right-handed (RCP) polarizers (Video S4). Notably, the QR code under RCP polarizer can be scanned by a smartphone to reveal the “WTU” information, demonstrating the material’s potential for circularly polarized selective reflection in information encryption and controlled reading. Similarly, we applied liquid crystal elastomer composite textile to display the logo of Wuhan Textile University (Figure 7B). It further verifies the application value of the textile composite in the field of polarization-dependent anti-counterfeiting mark and

dynamic display. Figure 7C utilizes the circular polarization-selective reflection property to decrypt the initially designed “88” into the digital “17” through polarization modulation by natural light, RCP, and LCP, achieving polarization-dependent controllable information reading (Video S5). Figure 7D integrates the dual-responsive characteristics of “polarization” and “stretching” in liquid crystal elastomers. Under RCP, after stretching deformation, the initial digital “8” deciphers into “3”, demonstrating the multi-stimulus synergistic information modulation capability (Video S6). Moreover, the initial information is pre-embedded into the composite textile, which can display Morse code patterns triggered by mechanical force, ultimately decoding into “MESSAGE” (Figure 7E). This expands the application scenarios of force-triggered information encryption in composite textile and provides a design basis for advanced textile anti-counterfeiting. Therefore, these results not only validate the superior circular polarization and force-responsive optical properties of CLCE, but also demonstrate their potential applications in multi-stimulus information encryption/decryption and anti-counterfeiting. It provides a practical approach for transforming smart-responsive materials into information devices.

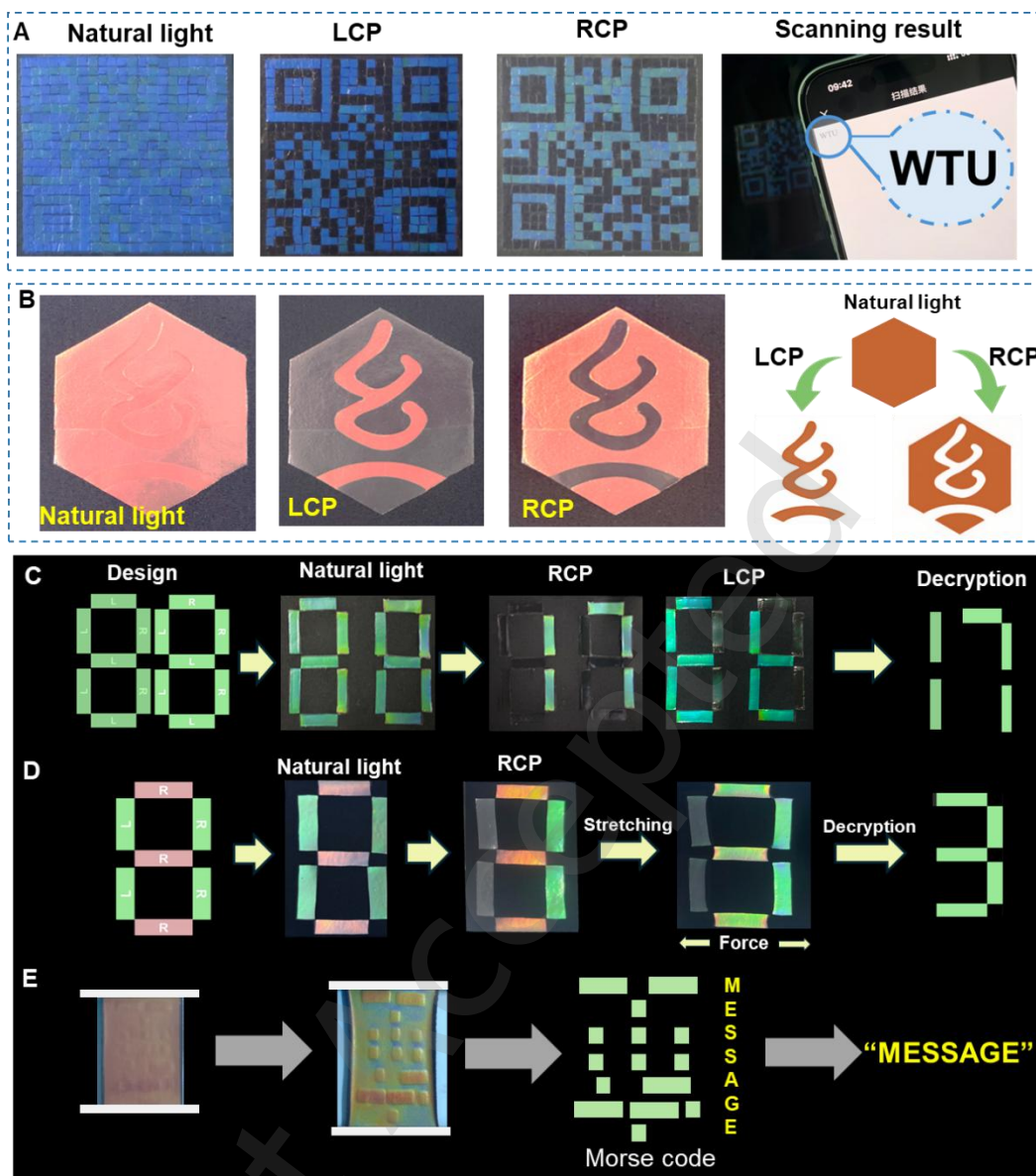


Figure 7 (A) Photographs of composite textile with QR code prepared by RGO/RCLCE and RGO/LCLCE film, and display of coding information under different polarizers. (B) Wuhan Textile University badge prepared by RGO/RCLCE and RGO/LCLCE film, and display under different polarizers. (C, D) Digital information encryption and information display under different polarization and stretching. (E) Morse code encryption and information display under stretching.

4 Conclusions

In summary, this study successfully fabricated RGO-doped liquid crystal elastomer composite films with excellent color uniformity, good mechanochromic sensitivity, and remarkable mechanical properties via a synergistic strategy integrating shear-induced alignment and RGO doping. During the preparation process, both the initial color of the films and the circular polarization properties of the liquid crystal elastomers can be effectively regulated. Additionally, the composite films exhibit sensitive thermochromic performance. Furthermore, when thermally bonded to elastic fabrics, interfacial physical and chemical interactions reinforce interfacial adhesion, realizing the synergistic enhancement of both mechanochromic reversibility and resilience of the composite textile. The circular polarization characteristics and mechanochromic performance of the

composite textile were further exploited to explore their potential applications in information-encrypted textiles. This work not only addresses a fundamental basis for achieving favorable dispersion of two-dimensional nanofillers in CLCEs, but also provides a simple and efficient approach for alignment control and assembly regularity optimization of liquid crystal mesogens in liquid crystal elastomers. It achieves the simultaneous enhancement of internal assembly structure, mechanical properties, and optical performance of liquid crystal elastomers, laying a crucial foundation for the application of multifunctional smart textiles in fields such as intelligent sensing, information encryption, and camouflage.

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Declaration of competing interest

There is no competing interest.

Use of AI statement

None.

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Electronic Supplementary Material

Polarization-dependent and mechanochromic dual-encryption composite textile enabled by robust and resilient cholesteric liquid crystal elastomer

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Table S1 Chemical formulations and monomers molar ratio for RCLCE film with different original reflecting color.

Samples	Mass ratio (k1) of LC756 to RM257	RM257 (mg)	LC756 (mg)	PETMP (mg)	EDDET (mg)	I-651 (mg)	DPA (μl)
RCLCE-1	0.045	268.8	12.1	17.92	69.88	2.1	65
RCLCE-2	0.050	268.8	13.4	17.92	69.88	2.1	65
RCLCE-3	0.055	268.8	14.8	17.92	69.88	2.1	65
RCLCE-4	0.065	268.8	16.3	17.92	69.88	2.1	65

Table S2 Chemical formulations and monomers molar ratio for LCLCE film with different original reflecting color.

Samples	Mass ratio (k2) of S5011 to LC756	RM257 (mg)	LC756 (mg)	S5011 (mg)	PETMP (mg)	EDDET (mg)
LCLCE-1	0.893	268.8	12.1	10.8	17.92	69.88
LCLCE-2	0.942	268.8	12.1	11.4	17.92	69.88
LCLCE-3	0.975	268.8	12.1	11.8	17.92	69.88
LCLCE-4	1	268.8	12.1	12.1	17.92	69.88

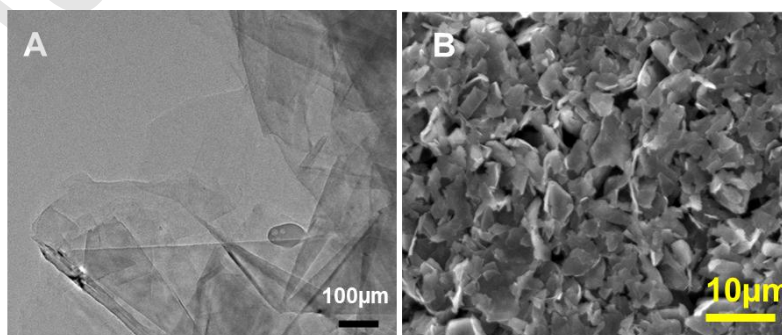


Figure S1 (A) TEM and (B) SEM image of RGO.

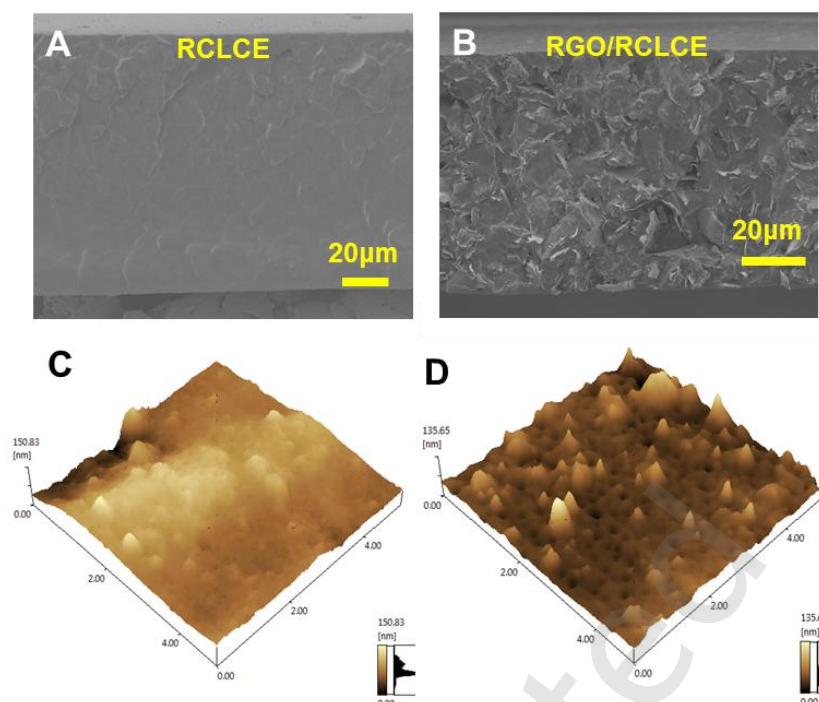


Figure S2 (A, B) The SEM cross-section of RCLCE and RGO/RCLCE films. (C, D) The AFM images for morphologies of the top and down surface of RGO/RCLCE film.

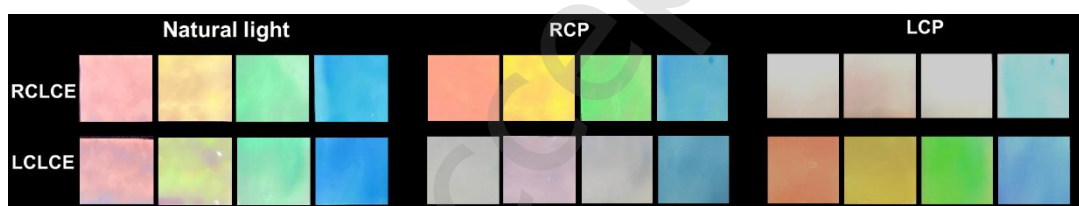


Figure S3 The photos of RCLCE and LCLCE with different initial colors.

Table S3 Comparison of L^* , a^* , b^* values of RCLCE and RGO/RCLCE films.

Samples	Parameter	Mean	Standard Deviation (SD)	Coefficient of Variation (CV%)	Intra-group Range (max-min)
RCLCE	L^*	84.02	0.94	1.12	1.89 (86.01-84.12)
	a^*	14.14	0.59	4.25	1.5 (15.23-13.71)
	b^*	19.35	0.81	4.19	2.15 (20.51-18.36)
	CIE-x	0.49	0.005	1.02	0.01 (0.498-0.488)
	CIE-y	0.386	0.004	1.04	0.01 (0.39-0.38)
	L^*	89.59	0.76	0.85	2.15 (90.68-88.53)
RGO/RCLCE	a^*	13.58	0.79	5.83	1.98 (14.65-12.67)
	b^*	15.33	0.65	4.24	1.78 (16.15-14.37)
	CIE-x	0.489	0.006	1.23	0.016 (0.498-0.482)
	CIE-y	0.359	0.004	1.12	0.012 (0.369-0.357)

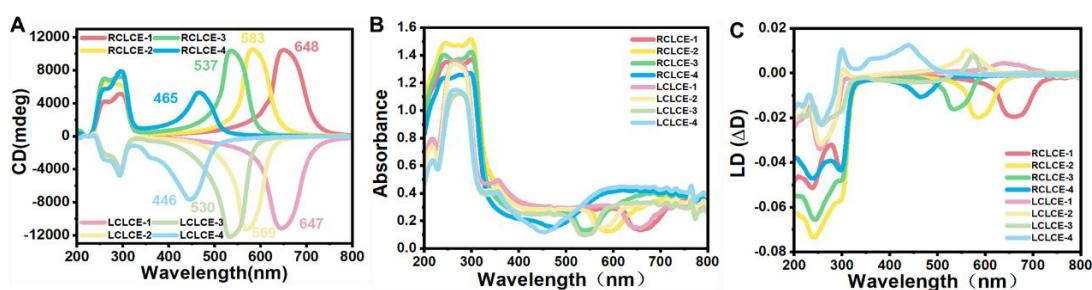


Figure S4 Circular dichroism spectra of RCLCE and LCLCE films with different colors.

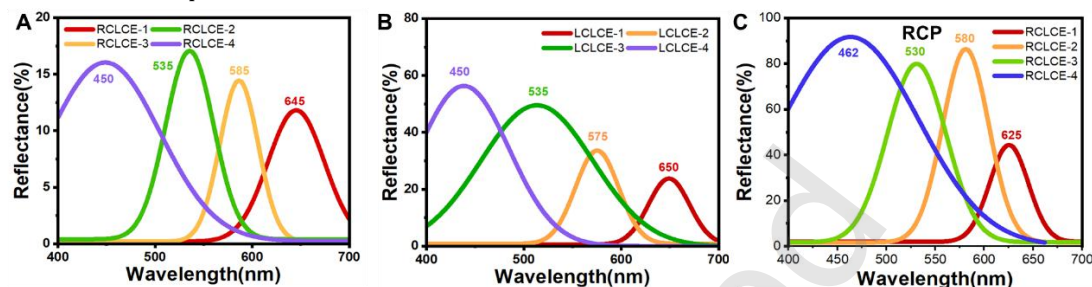


Figure S5 (A, B) The selectively reflected light spectra of RCLCE, and LCLCE films. (C) The selectively reflected light spectra of RCLCE films with right-handed circular polarizer.

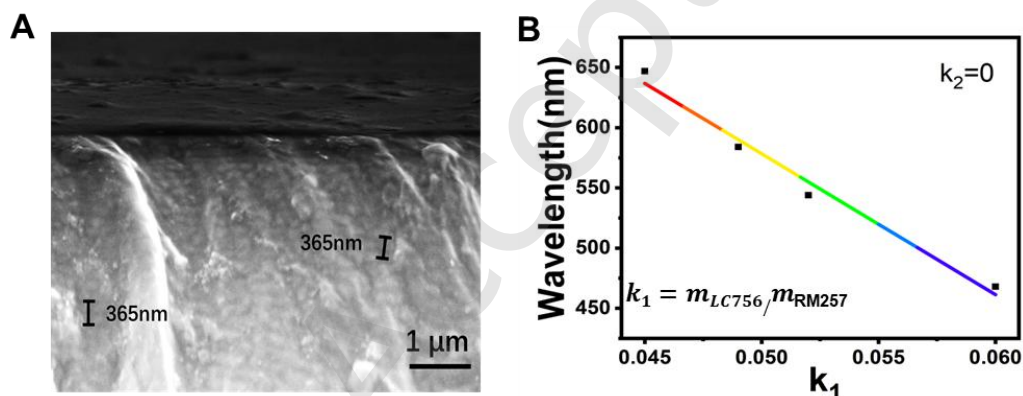


Figure S6 (A) The SEM cross-section of RCLCE film with red initial color. (B) The reflective wavelength of the RCLCE film versus the mass ratio (k_1) of LC756 to RM257.

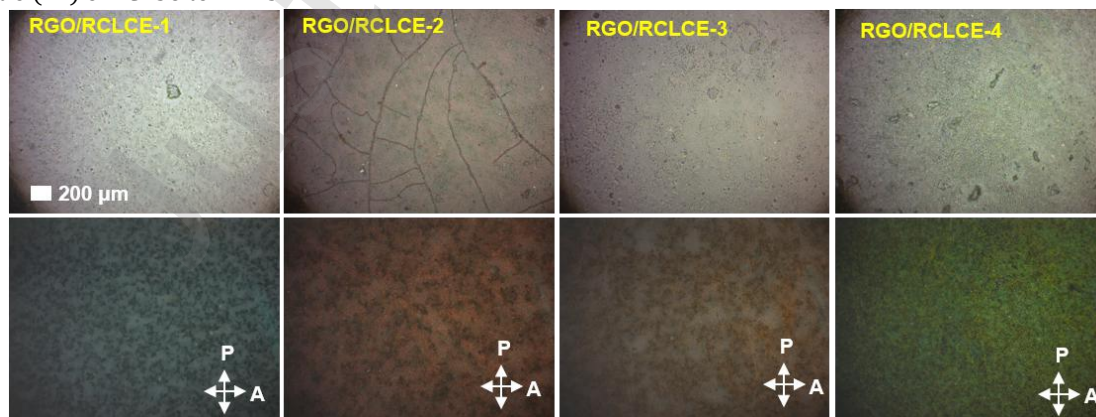


Figure S7 The microscopy photos and reflected POM images of the bottom surface of RGO/RCLCE composite films with different original colors.

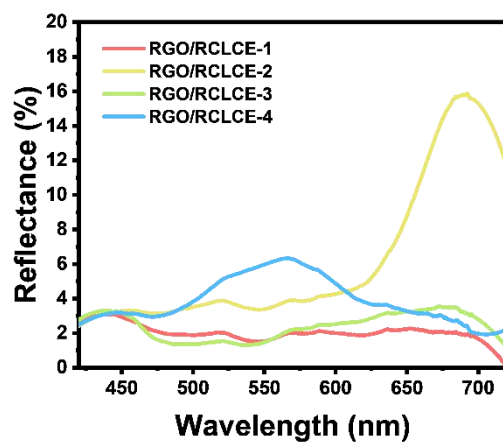


Figure S8 The selectively reflected light spectra of the bottom surface corresponding to RGO/RCLCE composite films.

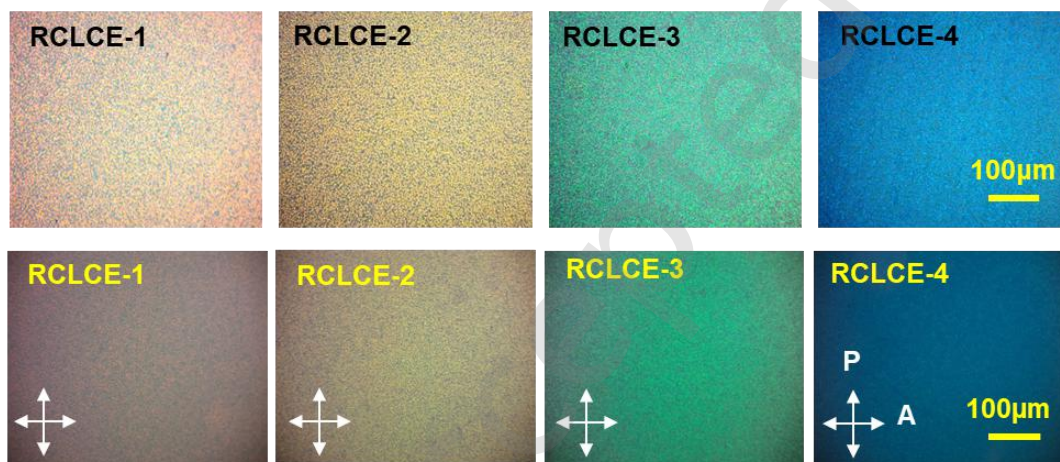


Figure S9 The microscopy photos and reflected POM images of RCLCE composite films with different original colors.

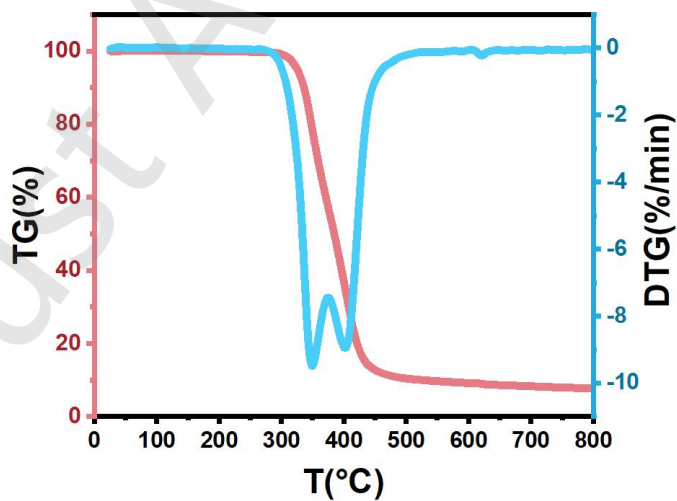


Figure S10 TGA and DTG curves of RCLCE film.

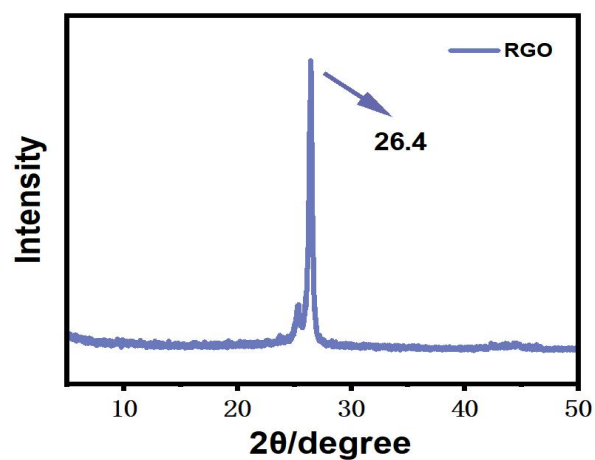


Figure S11 XRD curve of RGO.

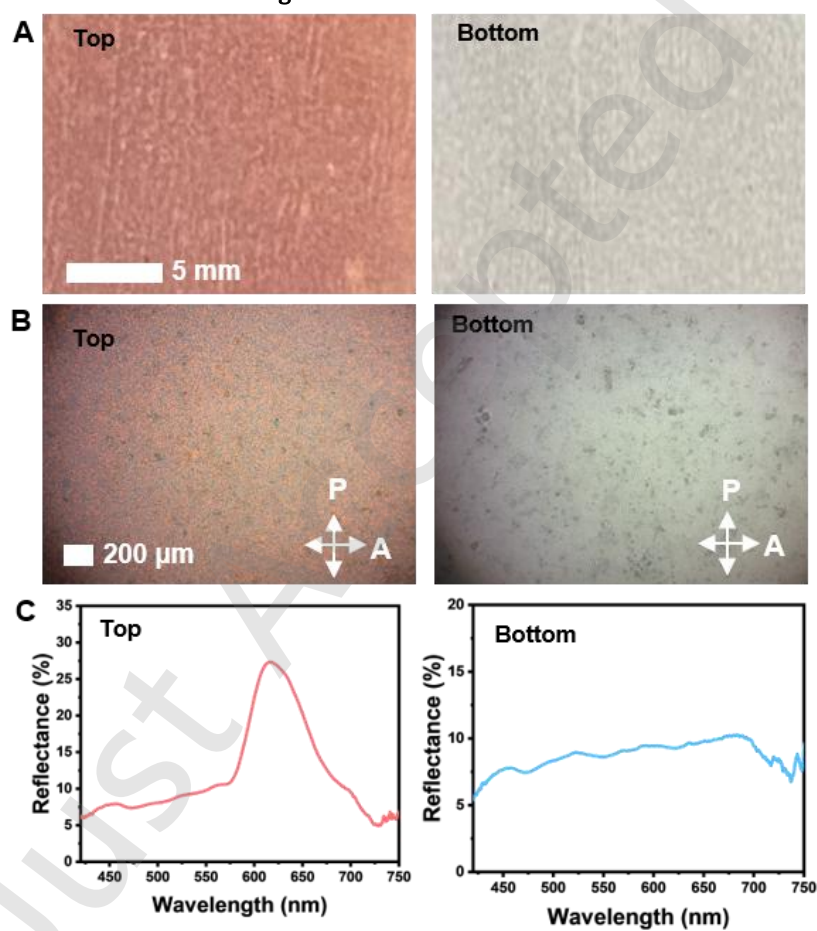


Figure S12 (A) Optical photographs, (B) reflected POM images and (C) selectively reflected light spectra corresponding to the top and bottom surface of RGO/RCLCE composite film with RGO content of 5%.

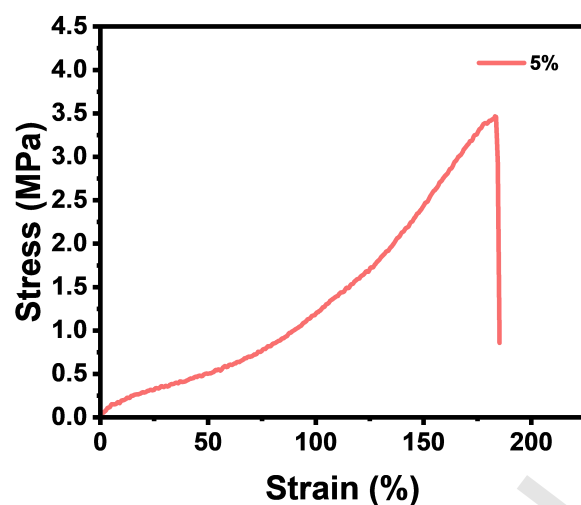


Figure S13 The stress-strain curve of RGO/RCLCE film with RGO content of 5%.

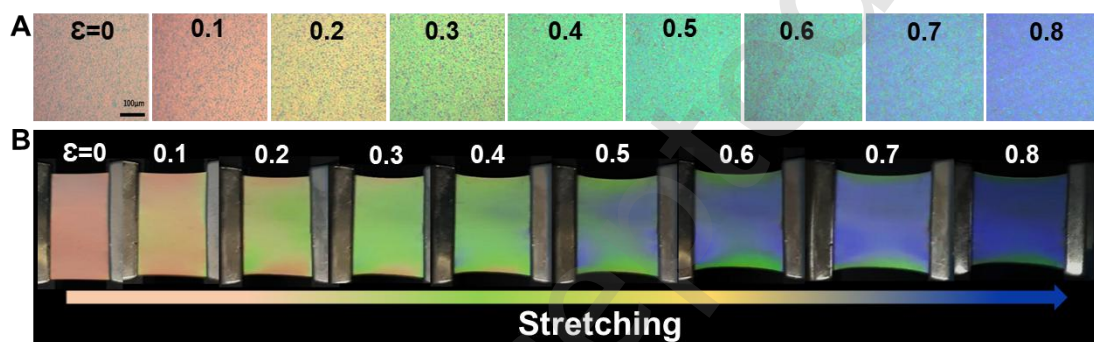


Figure S14 (A, B) Microphotographs and optical photographs of initially red-reflecting RCLCE film under uniaxial mechanical stretching strain from 0.0 to 0.8.

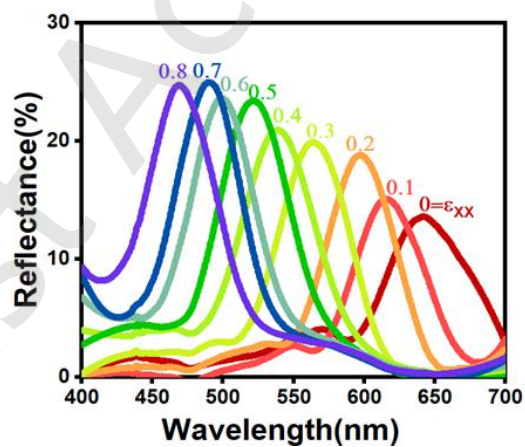


Figure S15 Reflection spectra of RCLCE film at length strain from 0.0 to 0.8, respectively.

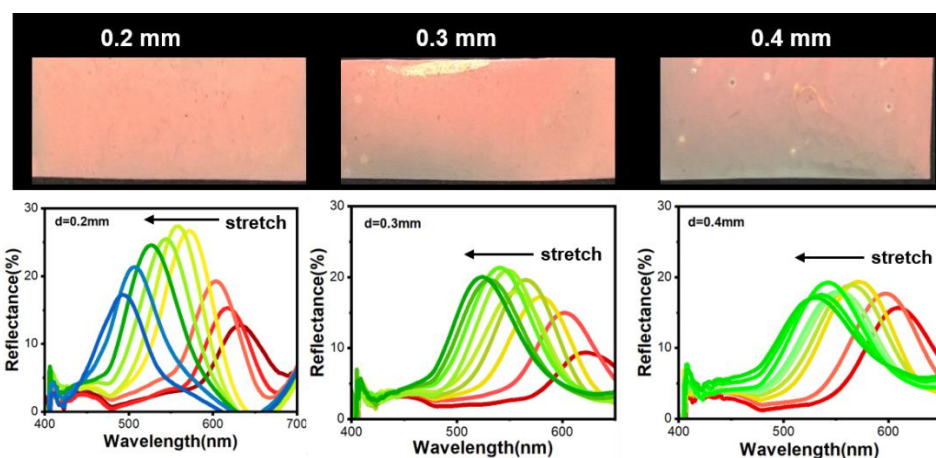


Figure S16 Optical photographs of CLCE films with different thicknesses and their reflection spectra under various strains.

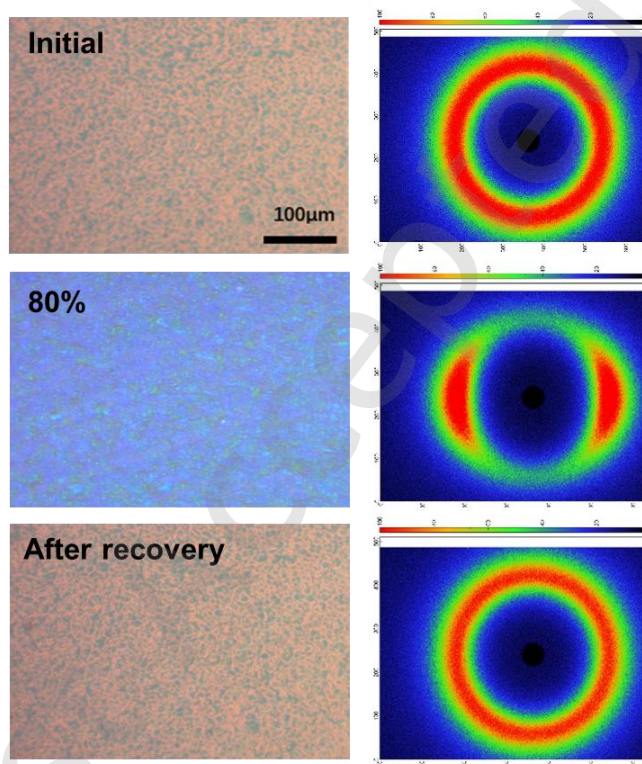


Figure S17 Optical photographs of the initial, 80% strained and recovered RCLCE film, and the corresponding 2D WAXS patterns of polydomain structure and monodomain structure subjected to the stretching.

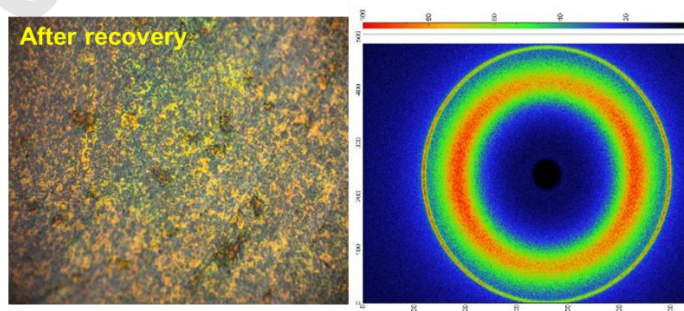


Figure S18 Optical photograph and the corresponding 2D WAXS pattern of RGO/RCLCE film after recovery.

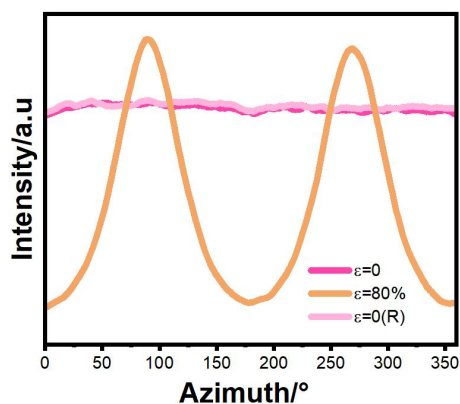


Figure S19 The scattering intensity as a function of the azimuthal angle obtained from the 2D WAXS pattern of the RCLCE film at the initial, 80% strained and recovered state.

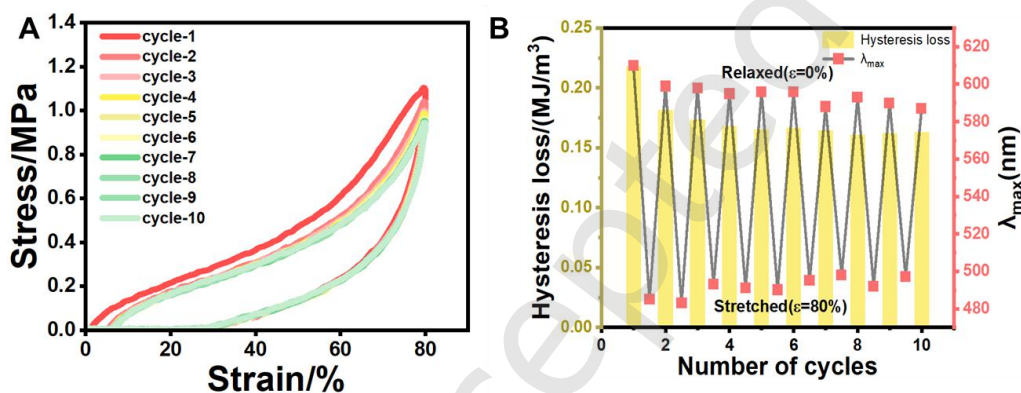


Figure S20 (A) Hysteresis curves of RGO/RCLCE film obtained from ten times load and unload tests at room temperature up to strain of 0.8. (B) Changes of reflectance wavelength and hysteresis loss of RGO/RCLCE during 10 times stretching-recovery cycles under a length strain of 0.00 to 0.8.

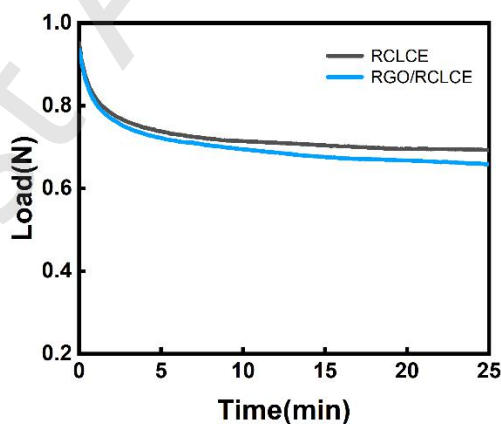


Figure S21 The stress relaxation performance of RCLCE and RGO/RCLCE with $\varepsilon = 80\%$.

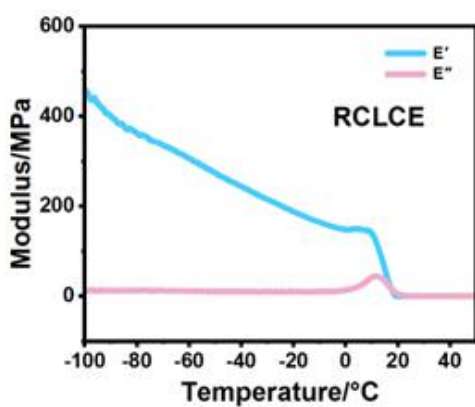


Figure S22 Storage modulus (E') and loss modulus (E'') of RGO/RCLCE film versus temperature.

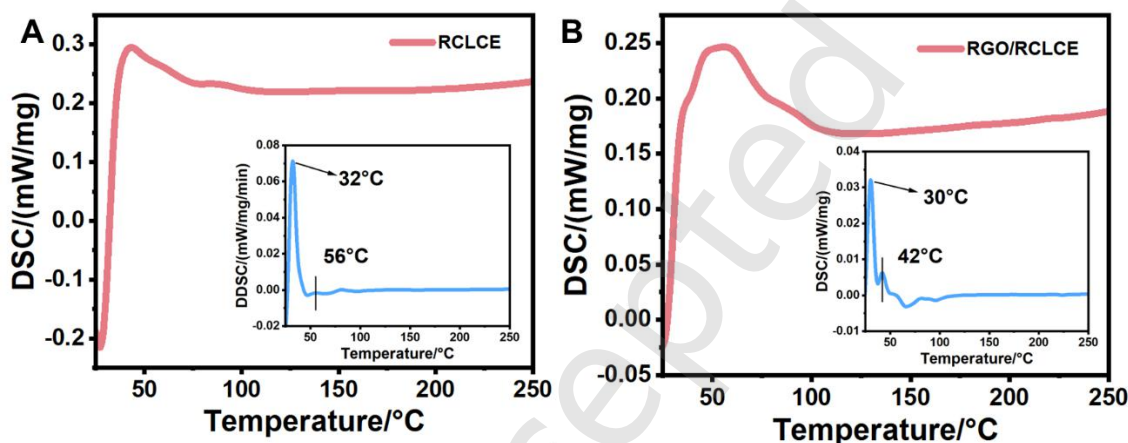


Figure S23 DSC curves of (A) RCLCE and (B) RGO/RCLCE films.

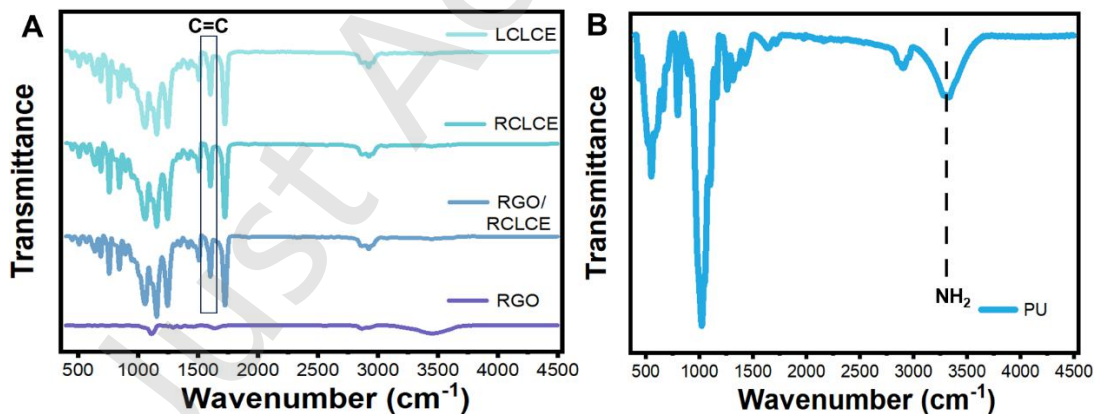


Figure S24 (A) FTIR spectra of RGO, CLCE and RGO/RCLCE films. (B) FTIR spectrum of adhesive layer.

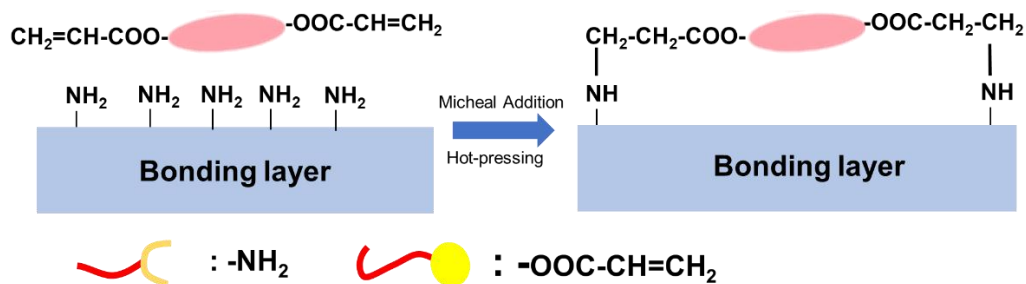


Figure S25 The interfacial chemical reaction of CLCE and bonding layer.

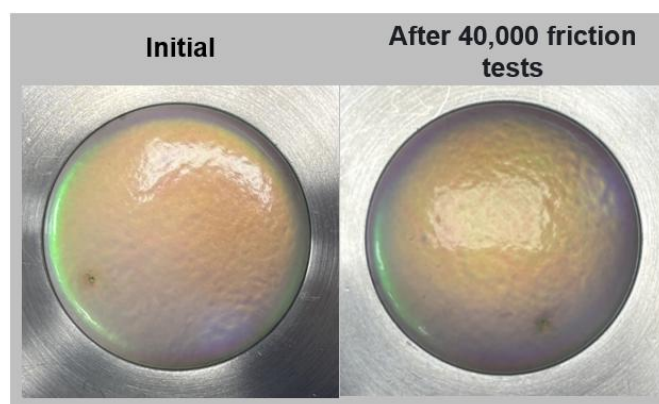


Figure S26 The photographs of the composite textile at the initial status and after 40000 friction tests.