

Article

Fabrication of Multilayer Broadband Reflective Cholesteric Liquid Crystal Films via Poly(vinyl Alcohol) Interlayers and Their Infrared Shielding Properties

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Abstract

Cholesteric liquid crystals (CLCs) possess the unique ability to selectively reflect incident circularly polarized light, exhibiting tremendous potential in diverse optical applications. In this study, a trilayer composite architecture of polymer-stabilized cholesteric liquid crystals (PSCLCs) was successfully fabricated. Introducing poly(vinyl alcohol) (PVA) as intervening barrier layers enabled the formation of independent and mutually non-interfering broadband reflection bands within each respective layer. Initially, a single-layer system was evaluated to identify the effects of component concentrations and polymerization conditions on the reflection bandwidth. Under optimal conditions, a maximum reflection bandwidth of 890 nm was achieved. Building upon these parameters, the effective concatenation of two independent reflection bands was accomplished by precisely regulating the concentration of the chiral dopant R5011 in the adjacent layers. Subsequently, the trilayer PSCLC film was constructed, ultimately broadening the total reflection bandwidth to 1650 nm. Characterization via polarized optical microscopy (POM) confirmed that the liquid crystal molecules consistently maintained a well-defined planar texture throughout the fabrication process of the multilayer films. Additionally, the film shows good infrared shielding performance. Its ability to regulate ambient light makes it highly promising as an optical filter and thermal management component in LC smart windows and emerging displays.

Keywords: cholesteric liquid crystals; broadband reflection; poly(vinyl alcohol); multilayer films; infrared shielding



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1. Introduction

Near-infrared (NIR) radiation makes up approximately half of the solar spectrum and is the primary driver of indoor temperature rise. Meanwhile, the absorption of solar radiation by materials can cause severe thermal accumulation, which significantly degrades the performance, color fidelity, and lifespan of display devices [1–3]. Due to their unique helical structures, cholesteric liquid crystals (CLCs) exhibit distinctive optical properties such as circular dichroism and selective reflection, holding broad application prospects

in fields including color patterning [4–6], anti-counterfeiting [7–11], smart windows [12], and infrared reflectors [13–16]. Among these, CLC-based IR reflectors have attracted considerable research interest for their efficiency in shielding solar IR light.

The selective reflection of CLCs originates from their unique helical structure, which can be described by Bragg's law [17–20]. Under normal incidence conditions, CLCs reflect circularly polarized incident light with the same handedness as their helical twist. The reflection wavelength λ is governed by the equation $\lambda = nP \sin\varphi$, where P , n , and φ represent the helical pitch, the average refractive index of the material, and the angle of the incident light relative to the liquid crystal surface, respectively [21–24]. The average refractive index is defined as $n = \sqrt{\frac{n_o^2 + n_e^2}{2}}$, where n_o and n_e are the ordinary and extraordinary refractive indices of the local uniaxial structure, respectively [25]. The helical pitch is expressed as $P = 1/(HTP \times c)$, where HTP and c denote the helical twisting power and the concentration of the chiral dopant, respectively, with HTP being primarily dependent on the intrinsic nature of the chiral compound [26–28]. Polymer-stabilized cholesteric liquid crystals (PSCLCs) are formulated by incorporating a low concentration of polymerizable monomers into CLC materials, followed by in situ polymerization. The most prominent feature of PSCLCs is their capacity to anchor the CLC pitch through elastic interactions between the polymer structural network and the LC molecules, thereby providing a fundamental basis for the establishment of a CLC pitch gradient [29,30].

Conventional CLC films possessing a uniform pitch typically exhibit a narrow reflection bandwidth, which severely limits their practical applications in fields requiring broadband reflection. To overcome this limitation, constructing a gradient or non-uniform pitch distribution has emerged as an effective strategy to achieve a broader reflection bandwidth [31,32]. Broer et al. [33] pioneered a molecular diffusion approach; doping a UV-absorbing dye into the LC system forms a UV intensity gradient under UV irradiation, which consequently causes differential consumption rates of mono- and di-functional monomers along the direction of the UV intensity gradient. In regions on the near-UV side, the di-functional chiral monomers are consumed more rapidly, driving the diffusion of unreacted monomers toward the near-UV side and thereby inducing the formation of a pitch gradient distribution.

Serving as an ideal separator for multilayer LC systems, the PVA film not only exhibits excellent optical transmittance—ensuring minimal adverse effects on the overall optical performance of the composite film—but also, owing to its intrinsic hydrophilicity, is immiscible with lipophilic substances such as LCs. Consequently, it effectively suppresses the mutual diffusion of components between adjacent layers [34,35].

In our previous studies, highly reflective bilayer PSCLC films exceeding the 50% theoretical reflection limit were successfully fabricated by combining dual-handed (left- and right-handed) chiral systems via a PVA barrier layer [36].

It is also worth noting that, besides pure LC films, hybrid smart windows have recently gained significant attention. For instance, by combining a tunable scattering LC layer with thermochromic VO₂ or nanoporous particles, researchers achieved excellent dynamic control over both visible and IR radiation [37–39].

Distinct from the pursuit of exceeding the 50% reflection limit or utilizing active hybrid materials, the present work specifically focuses on maximizing the passive reflection bandwidth within a purely single-handed chiral system.

In this work, multilayer broadband reflective PSCLC films were constructed through the further incorporation of PVA intermediate layers. The optimal composition and polymerization conditions for the single-layer PSCLC film were systematically investigated, alongside an examination of the effect of the chiral dopant (R5011) concentration on both the central wavelength and the reflection bandwidth of the single-layer film. Building

upon these parameters, bilayer films were subsequently fabricated. By utilizing PVA to effectively avert interlayer molecular diffusion while concurrently enhancing interlayer adhesion, and by precisely regulating the R5011 content in the upper and lower layers, the effective concatenation of two independent reflection bands was successfully achieved, thereby broadening the reflection bandwidth of the film. Based on the bilayer system, a trilayer composite film was successfully developed, further extending the overall reflection bandwidth to 1650 nm. Polarized optical microscopy (POM) characterization indicated that the multilayer film maintained a well-defined planar texture throughout the entire fabrication process. Furthermore, the excellent infrared shielding performance of these constructed films highlights their significant value in ambient light management. This paves a solid foundation for their application as optical filters and thermal regulatory components in smart windows and advanced displays.

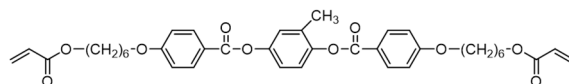
2. Materials and Methods

2.1. Materials

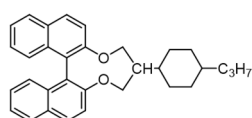
The nematic liquid crystal host SLC-1717 ($n_e = 1.720$, $n_o = 1.519$, $\Delta n = 0.201$ measured at 589 nm) was acquired from Shijiazhuang Chengzhi Yonghua Display Material Co., Ltd. (Shijiazhuang, China). The chiral dopant R5011 and the UV-polymerizable monomer C6M were purchased from Shanghai Boer Chemical Reagent Co., Ltd. (Shanghai, China) and Beijing Kexin Jingyuan Technology and Trade Co., Ltd. (Beijing, China), respectively. The photo-initiator IRG651 and the UV absorber UV-327 were supplied by Aladdin Co., Ltd. (Shanghai, China) and Annaiji Chemical Reagent Co., Ltd. (Shanghai, China). Poly(vinyl alcohol) (PVA-1799, 98–99% hydrolyzed), utilized for the intermediate barrier layers, was obtained from Shanghai Macklin Biochemical Co., Ltd. (Shanghai, China). The chemical structures of these compounds are illustrated in Figure 1.

(a) **Nematic liquid crystal: SLC-1717** Mixture of LCs with positive dielectric anisotropy

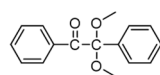
(b) **UV-polymerizable free radical monomer: C6M**



(c) **Right-handed chiral monomer: R5011**



(d) **Free radical photo-initiator: IRG651**



(e) **UV absorption dyes: UV-327**

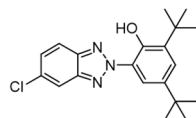


Figure 1. Chemical structures of the experimental materials used.

2.2. Measurements

Optical transmittance data across the 600 to 2400 nm spectral region were collected at room temperature relying on a UV/VIS/NIR spectrophotometer (JASCO V570, JASCO Corporation, Tokyo, Japan). Baseline normalization was established using a blank LC cell as the 100% reference. The reflection bandwidth, denoted as $\Delta\lambda$, corresponds to the width measured at half the depth of the transmission peak. The alignment and internal

morphology of the resultant CLC layers were captured via an Olympus BX51 polarized optical microscope (POM, Olympus Corporation, Tokyo, Japan). Additionally, an NTC temperature sensor (10 K/3435) was integrated into the simulated model to monitor internal temperature fluctuations dynamically.

2.3. Preparation of Samples and Cells

Fabrication of Single-Layer Broadband Reflective Films: Single-layer LC mixtures were formulated according to the compositions of samples A, B, C, and D listed in Table 1, followed by the addition of a small amount of dichloromethane (DCM) as a solvent. The mixtures were subjected to ultrasonic agitation to ensure the complete dissolution of all components. Subsequently, the samples were placed in an oven at 60 °C for over 6 h to guarantee the thorough evaporation of the DCM solvent; this entire procedure was strictly conducted in the dark. The resulting LC mixtures were infiltrated into 20 µm thick LC cells via capillary action and maintained at 60 °C for 10 min to promote the formation of a well-defined planar alignment texture. Finally, the samples were exposed to 365 nm UV irradiation for 20 min to complete the monomer polymerization, thereby yielding the single-layer broadband reflective PSCLC films.

Table 1. Ratios and polymerization conditions of samples.

Sample No.	SLC-1717/C6M/R5011/ IRG651/UV-327 (wt%)	UV Intensity (mW/cm ²)
A1	93.40/5.00/1.00/0.30/0.30	1.1
A2	88.40/10.00/1.00/0.30/0.30	1.1
A3	83.40/15.00/1.00/0.30/0.30	1.1
A4	78.40/20.00/1.00/0.30/0.30	1.1
A5	73.40/25.00/1.00/0.30/0.30	1.1
B1	83.60/15.00/1.00/0.30/0.10	1.1
B2	83.40/15.00/1.00/0.30/0.30	1.1
B3	83.20/15.00/1.00/0.30/0.50	1.1
B4	83.00/15.00/1.00/0.30/0.70	1.1
B5	82.80/15.00/1.00/0.30/0.90	1.1
C1	83.40/15.00/1.00/0.30/0.30	0.1
C2	83.40/15.00/1.00/0.30/0.30	0.6
C3	83.40/15.00/1.00/0.30/0.30	1.1
C4	83.40/15.00/1.00/0.30/0.30	1.6
C5	83.40/15.00/1.00/0.30/0.30	2.1
D1	83.70/15.00/0.70/0.30/0.30	1.1
D2	83.40/15.00/1.00/0.30/0.30	1.1
D3	83.10/15.00/1.30/0.30/0.30	1.1
D4	82.90/15.00/1.50/0.30/0.30	1.1
D5	82.00/15.00/2.40/0.30/0.30	1.1

Fabrication of Bilayer Broadband Reflective Films: Following the polymerization of the upper LC film, the glass substrate on the far-UV side was carefully removed. A 0.5 wt% aqueous PVA solution was uniformly deposited onto the exposed LC surface, facilitated by surface tension. Following a slow heating and drying process to completely evaporate the solvent, a dense PVA intermediate layer with a thickness of approximately 3.5 µm was formed. Upon obtaining the single-sided glass substrate bearing the PSCLC-PVA composite layer, it was assembled with another bare ITO-coated glass substrate equipped with a 20 µm thick PET spacer to construct a new LC cell. The LC mixture of group D was infiltrated into the newly formed cell via capillary action, and the cell was identically incubated in an oven at 60 °C for 10 min to establish a stable planar texture. The sample

was then exposed to UV irradiation (365 nm, 1.1 mW/cm²) for 20 min to complete the polymerization, successfully constructing the bilayer broadband reflective film.

Fabrication of Trilayer Broadband Reflective Films: LC mixtures D1 and D2 were individually infiltrated into two distinct 20 µm thick LC cells and subjected to UV polymerization, yielding two independent broadband reflective films. The glass substrates on the far-UV side of both samples were peeled off. A 0.5 wt% PVA aqueous solution was uniformly deposited onto the exposed surface of each LC layer, facilitated by surface tension. Following a slow heating and drying process, two single-sided glass substrates, each coated with a PSCLC-PVA composite layer, were obtained. Next, a 20 µm thick PET spacer was inserted between these two prepared glass substrates to construct a novel 60 µm thick LC cell. Finally, the LC mixture D4 was infiltrated into the gap between the two PVA layers via capillary action. The assembly was then exposed to UV irradiation (365 nm, 1.1 mW/cm²) for 20 min to complete polymerization. Thus, a trilayer broadband reflective film featuring dual PVA intermediate layers was successfully fabricated. The overarching fabrication procedure for this trilayer architecture is illustrated in Figure 2.

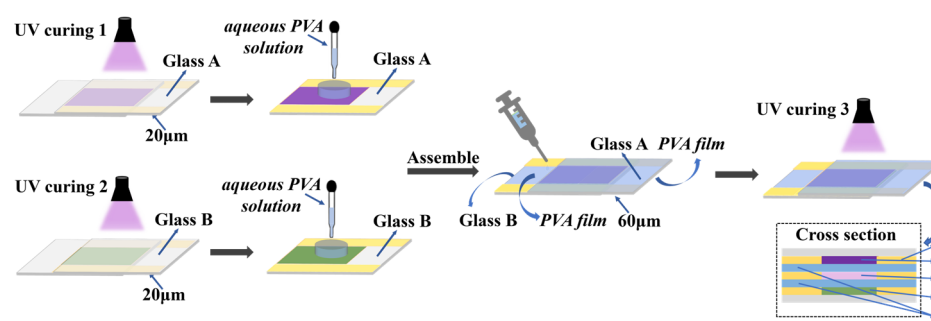


Figure 2. Preparation process of trilayer PSCLC films with broadband reflection. Note that both UV curing step 1 and UV curing step 2 are performed on independent single-layer films prior to the assembly of the 60 µm thick cell.

3. Results and Discussion

3.1. Experimental Mechanism of the Multilayer Broadband Reflection

Figure 3 schematically illustrates the fabrication mechanism underlying the bilayer and trilayer broadband reflective PSCLC films. The crux of this architecture lies in the incorporation of the PVA intermediate layer. Upon the slow evaporation of the aqueous PVA solution, a dense physical barrier layer is formed. This layer not only effectively prevents interlayer molecular diffusion but also circumvents the issues of refractive index mismatch and weak interlayer adhesion typically associated with conventionally stacked LC films, thereby mitigating interfacial scattering and optical losses. Initially, during the formation of the upper film, the helical pitch and the spatial distribution of all molecular components within the composite system remain uniform prior to polymerization. Upon UV irradiation, owing to the absorption by the UV dye, an attenuation gradient of UV intensity is generated along the thickness direction of the LC cell. On the side closer to the UV source, the high-intensity UV radiation triggers the rapid polymerization and consequent depletion of the polymerizable monomers. This localized reduction in monomer concentration drives the diffusion of unreacted monomers from the far-UV side toward the near-UV side. Conversely, the non-polymerizable chiral dopant R5011 undergoes a counter-migration toward the far-UV side. This migration process gradually establishes a concentration gradient of the chiral compound within the film; specifically, a low chiral dopant concentration at the near-UV side corresponds to a longer helical pitch, whereas a high chiral dopant concentration at the far-UV side induces a shorter helical pitch. Meanwhile, this non-uniform pitch distribution is anchored by the polymer network,

realizing the broadband reflection characteristic of the single-layer PSCLC film. A right-handed LC mixture, identical to that of the first layer, is infiltrated into the lower layer. Benefiting from the physical barrier effect of the PVA intermediate layer, the lower system undergoes an analogous process of UV intensity attenuation and chiral migration during the secondary polymerization, yielding a second pitch gradient distribution. Because both the upper and lower layers possess chiral molecules with identical handedness, the synergistic effect of the two independent right-handed reflection bands enables their effective concatenation. This significantly broadens the overall reflection range, ultimately realizing the broadband reflection of the bilayer film.

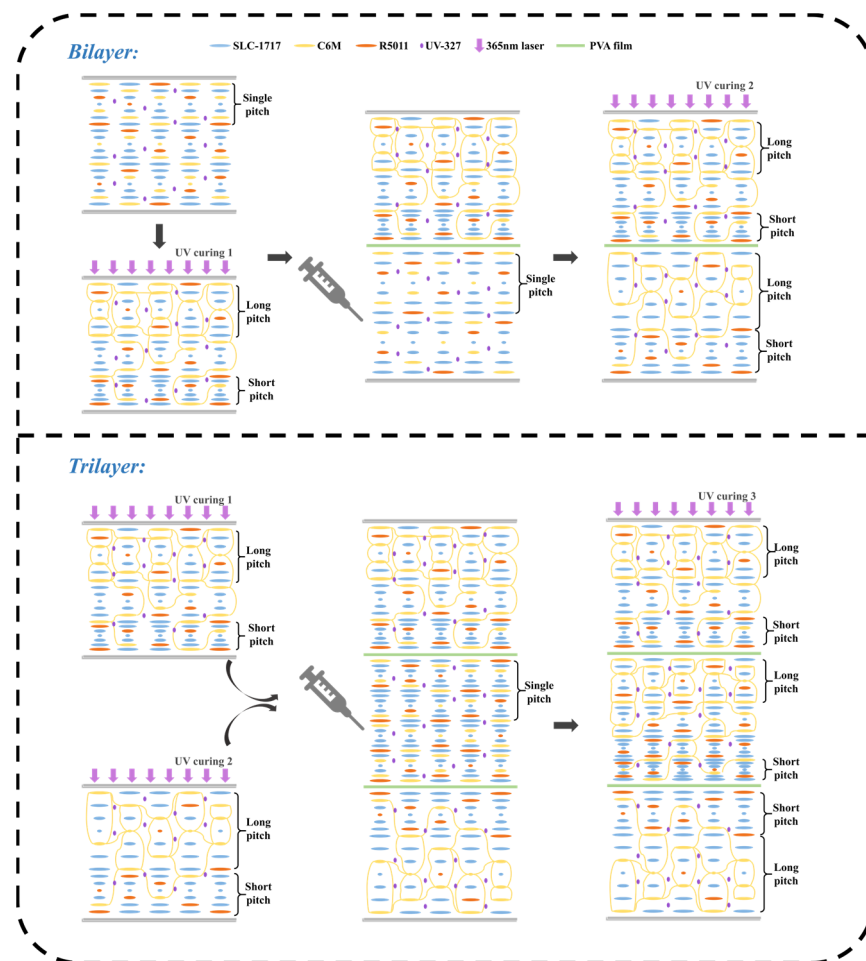


Figure 3. Preparation mechanism of bilayer and trilayer PSCLC films with broadband reflection.

By the same rationale, the fabrication of the trilayer broadband reflective film is founded upon the aforementioned mechanism of concatenating multiple independent reflection bands. Within the trilayer composite system segregated by dual PVA barrier films, all three LC layers (D1, D4, and D2) independently undergo monomer polymerization and chiral diffusion along the thickness direction of the cell. This engenders distinct, non-uniform pitch distributions within each layer, which are subsequently anchored by their respective polymer networks. Through precise regulation of the chiral dopant content across these three systems, their respective reflection bands are spectrally staggered and seamlessly connected. Concurrently, as the PVA films effectively preserve the structural integrity of each layer and enhance interlayer adhesion, the three independent pitch gradients function as a cohesive entity along the thickness direction. Upon illumination, incident light of varying wavelength bands is selectively reflected by the LC layers possessing the

corresponding helical pitches, culminating in the successful fabrication of a composite PSCLC film exhibiting ultra-broadband reflection characteristics.

3.2. Effect of C6M Concentration on the Broadband Reflection of Single-Layer PSCLC Films

Although various non-optimized conditions were tested in our preliminary trials, the remaining parameters in each group were kept constant at their optimal values to clearly isolate the effect of a single variable. For the samples in Group A, the mass fractions of the polymerizable monomer C6M were formulated at 5 wt%, 10 wt%, 15 wt%, 20 wt%, and 25 wt%, respectively. The samples were subjected to UV irradiation (365 nm, 1.1 mW/cm²) for 20 min to induce polymerization; their corresponding Vis-NIR transmission spectra and reflection bandwidths are illustrated in Figure 4. Within the single-layer PSCLC film system, as the concentration of the polymerizable monomer C6M increased, the reflection bandwidth of the film exhibited an initial increase followed by a subsequent decrease. At a concentration of 15 wt%, the reflection bandwidth $\Delta\lambda$ reached a maximum value of 890 nm. This phenomenon originates from the competitive interaction between the polymerization rate of the monomers and the molecular diffusion rate within the film.

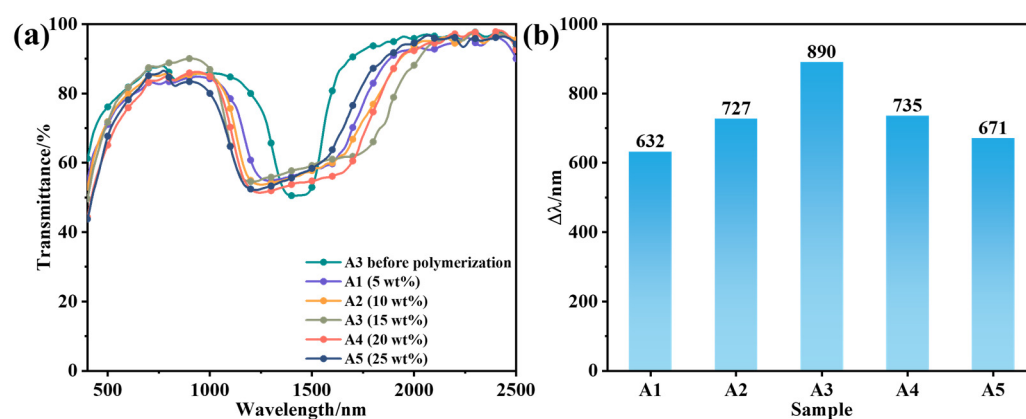


Figure 4. Group A samples: trends of (a) Vis-NIR transmission spectra; (b) $\Delta\lambda$ with the content of polymerizable monomer C6M.

At relatively low C6M concentrations, an increase in C6M accelerates the photopolymerization rate of the monomers, leading to the gradual formation of a denser polymer network upon curing. During this stage, the diffusion of molecules within the system is not yet significantly restricted by the density of the polymer network. Driven by the local concentration gradients induced by the attenuation of UV intensity, both C6M and R5011 can undergo sufficient diffusion. At the same time, the gradually densifying polymer network can promptly and effectively anchor the helical pitch, averting the ineffective molecular anchoring that would result from an overly sparse network. However, upon further increasing the C6M content, the reflection bandwidth of the film paradoxically begins to decline. This is attributed to the excessively high monomer content, which triggers an overly rapid polymerization rate and the swift formation of a highly dense polymer network. Such a rigid network structure not only hinders the unreacted C6M from further diffusing toward the UV source, but also indirectly suppresses the migration of the chiral dopant R5011. Because the molecules within the system are deprived of sufficient time to diffuse and establish an adequate concentration difference, the ultimately formed pitch gradient narrows, which consequently diminishes the reflection bandwidth of the film. Therefore, the optimal single-layer broadband reflection performance is achieved when the C6M concentration is optimized at 15 wt%.

3.3. Effect of UV-327 Concentration on the Broadband Reflection of Single-Layer PSCLC Films

Utilizing the aforementioned polymerization conditions, the samples of Group B were fabricated, with the mass fractions of UV-327 formulated at 0.1 wt%, 0.3 wt%, 0.5 wt%, 0.7 wt%, and 0.9 wt%, respectively. Figure 5 presents the Vis-NIR transmission spectra of the Group B samples and their $\Delta\lambda$. As illustrated, with an increase in the UV-327 content, the reflection bandwidth of the single-layer PSCLC films exhibited an initial increase followed by a subsequent decrease. The maximum $\Delta\lambda$ was achieved at a UV-327 concentration of 0.3 wt%. This phenomenon primarily originates from the UV intensity gradient established by UV-327 along the thickness direction of the LC cell.

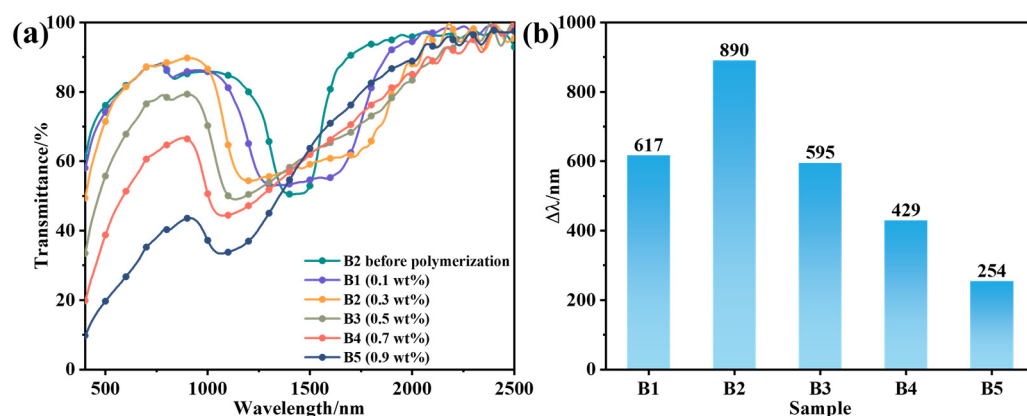


Figure 5. Group B samples: trends of (a) Vis-NIR transmission spectra; (b) $\Delta\lambda$ with the content of UV-absorbing dye UV-327.

At relatively low UV-327 concentrations, the optical intensity gradient within the LC system is inconspicuous, and the polymerization rate is comparatively slow. Consequently, the polymer network fails to promptly anchor the diffusing molecules, resulting in the formation of a narrow pitch gradient. Upon an appropriate increase in UV-327 concentration, its UV absorption induces a suitable attenuation of UV intensity within the single-layer film. Owing to the relatively higher UV intensity on the near-UV side, the polymerization rate of the monomer C6M is faster than that on the far-UV side. This differential in polymerization rates drives the unreacted C6M to diffuse toward the near-UV side, whilst the chiral dopant R5011 concomitantly migrates toward the far-UV side. An optimal amount of UV-327 not only induces the formation of a more favorable concentration gradient of the chiral compound but also ensures that the polymer network can promptly anchor the non-uniform pitch distribution. This facilitates the formation of a continuous pitch gradient, thereby progressively broadening the reflection bandwidth.

However, as the UV-327 concentration is further augmented, the reflection bandwidth of the films begins to decrease. This is attributed to the fact that an excessive amount of UV-327 causes an overly drastic attenuation of UV intensity, establishing an excessively steep optical intensity gradient. Under these conditions, the polymerization rate of C6M on the near-UV side is exceedingly rapid, leading to the formation of a dense polymer network prior to adequate molecular diffusion. Consequently, the chiral dopant R5011 is deprived of sufficient diffusion time, resulting in a diminished pitch gradient. Furthermore, as depicted in Figure 6, while samples B1 and B2 exhibited a well-defined planar texture following polymerization, samples B3–B5 underwent phase separation induced by the extreme disparity in polymerization rates between the near-UV and far-UV sides. This phase separation disrupted the planar texture of the LC system, inducing optical scattering within the film and ultimately narrowing the reflection bandwidth. It is noteworthy that among all the variables investigated in the single-layer systems, only the excessive concentration

of the UV-absorbing dye induced such macroscopic phase separation. Variations in other parameters (e.g., C6M concentration and UV intensity) primarily altered the reflection bandwidth while consistently maintaining a well-defined planar texture. Therefore, the optimal UV-327 concentration for the single-layer broadband reflective PSCLC film is determined to be 0.3 wt%.

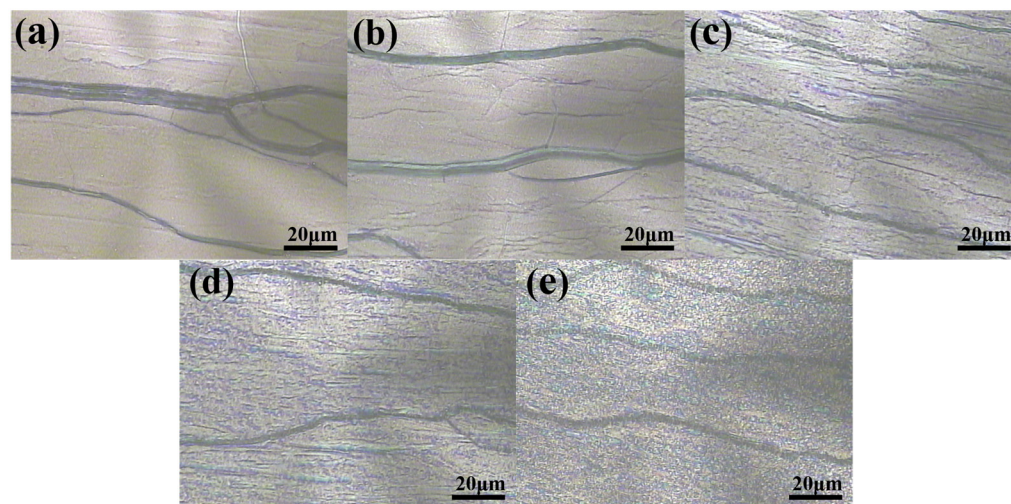


Figure 6. POM images of Group B samples with different UV-327 contents after polymerization: (a) B1 (0.1 wt%); (b) B2 (0.3 wt%); (c) B3 (0.5 wt%); (d) B4 (0.7 wt%); (e) B5 (0.9 wt%).

3.4. Effect of Polymerization Light Intensity on the Broadband Reflection of Single-Layer PSCLC Films

For the single-layer films of the Group C samples, the composition of each sample was kept identical. The samples were subjected to 365 nm UV irradiation for 20 min at diverse intensities of 0.1 mW/cm², 0.6 mW/cm², 1.1 mW/cm², 1.6 mW/cm², and 2.1 mW/cm², respectively, to induce polymerization. Figure 7 presents the Vis-NIR transmission spectra of the Group C samples and their $\Delta\lambda$. As illustrated, with an increase in UV intensity, the reflection bandwidth of the single-layer PSCLC films exhibited an initial increase followed by a subsequent decrease, reaching a maximum value at a UV intensity of 1.1 mW/cm². Modulating the UV intensity dictates the polymerization rate of the monomer C6M within the system, thereby indirectly influencing the diffusion rate of the small molecules.

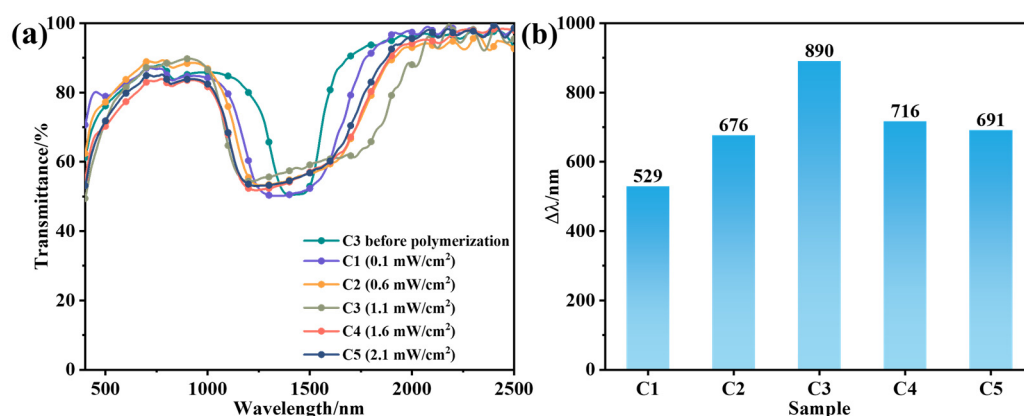


Figure 7. Group C samples: trends of (a) Vis-NIR transmission spectra; (b) $\Delta\lambda$ with the polymerization light intensity.

Owing to the presence of UV-327, a UV intensity gradient is established along the thickness direction of the film. When the UV intensity is relatively low but progressively

increased, the optical intensity on the near-UV side increases correspondingly. This accelerates the polymerization rate of C6M on the near-UV side, resulting in a more rapid depletion of the monomers. The accelerated polymerization generates a more substantial monomer concentration gradient, which in turn drives the unreacted C6M to diffuse more rapidly toward the near-UV side, whilst simultaneously propelling the chiral dopant R5011 to migrate toward the far-UV side. The gradually enhanced light intensity not only facilitates the formation of a more pronounced concentration gradient of the chiral dopant but also induces a moderately accelerated polymerization reaction, ensuring that the polymer network can promptly and effectively anchor the pitch distribution. Consequently, a broader pitch gradient is established within the film, thereby progressively widening the reflection bandwidth.

However, when the UV intensity becomes excessively high, the reflection bandwidth of the film begins to decline. This is attributed to the fact that an excessive UV intensity induces an overly rapid photopolymerization of C6M, where the reaction rate significantly surpasses the molecular diffusion rate. Consequently, a dense polymer network is rapidly formed within the film. This rigid network hinders the diffusion of both the unreacted monomers and the chiral dopant R5011, thereby impeding the development of a broader pitch gradient and ultimately narrowing the reflection bandwidth.

During multi-layer fabrication, the incident UV light inevitably experiences partial attenuation. However, since the UV-absorbing dye is kept at a relatively low concentration (0.3 wt%), the extent of light attenuation is limited. Consequently, the transmitted UV light reaching the underlying layers remains well above the effective threshold required to initiate photopolymerization. As firmly demonstrated by the final multi-layer systems, maintaining a constant incident UV intensity at the single-layer optimized value (1.1 mW/cm^2) is sufficient to successfully broaden the reflection bands in all subsequent layers. Consequently, to avoid cumbersome experimental procedures, further investigation into the specific optimization of irradiation conditions for each individual layer was not pursued.

3.5. Effect of R5011 Concentration on the Broadband Reflection of Single-Layer PSCLC Films

Based on the aforementioned experiments, the mass fractions of R5011 in the Group D samples were formulated at 0.7 wt%, 1.0 wt%, 1.3 wt%, 1.5 wt%, and 2.4 wt%, respectively. The concentrations of C6M and UV-327 were maintained constant at 15 wt% and 0.3 wt%, respectively. The samples were subsequently subjected to UV irradiation (1.1 mW/cm^2) for 20 min to induce polymerization. Figure 8 illustrates the Vis-NIR transmission spectra of samples D1–D5, alongside their corresponding central wavelengths and reflection bandwidths. As depicted in Figure 8a, the reflection bandwidths of samples D1–D5 post-polymerization were invariably broader than those prior to polymerization. The emergence of this phenomenon is attributed to the presence of the UV dye, which establishes a UV intensity gradient along the thickness direction of the film, causing the polymerization rate of the monomer C6M on the near-UV side to outpace that on the far-UV side. This differential in polymerization rates generates a monomer concentration gradient, which in turn drives the unreacted C6M on the far-UV side to diffuse toward the near-UV side. Conversely, the chiral dopant R5011 undergoes a counter-migration toward the far-UV side. This process establishes a concentration gradient of the chiral compound, effectively broadening the reflection bandwidth. As illustrated in Figure 8b, with the increment of the R5011 content, the central wavelength of the LC films progressively blue-shifted, and the corresponding reflection bandwidth gradually decreased. The blue-shift in the central wavelength is primarily dictated by the inverse proportional relationship between the LC helical pitch and the chiral dopant concentration. Because the identical chiral dopant

R5011 was employed throughout this system, its HTP remained constant. As the initial mass fraction of R5011 in the system escalates, the helical pitch of the LC contracts accordingly, thereby causing a progressive blue-shift in the corresponding central wavelength. Concurrently, a higher initial concentration of R5011 within the system dictates a smaller relative concentration differential that can be established during the molecular migration process. This engenders a narrower distribution range of the helical pitch within the LC system, ultimately leading to a gradual reduction in the selective reflection bandwidth of the samples.

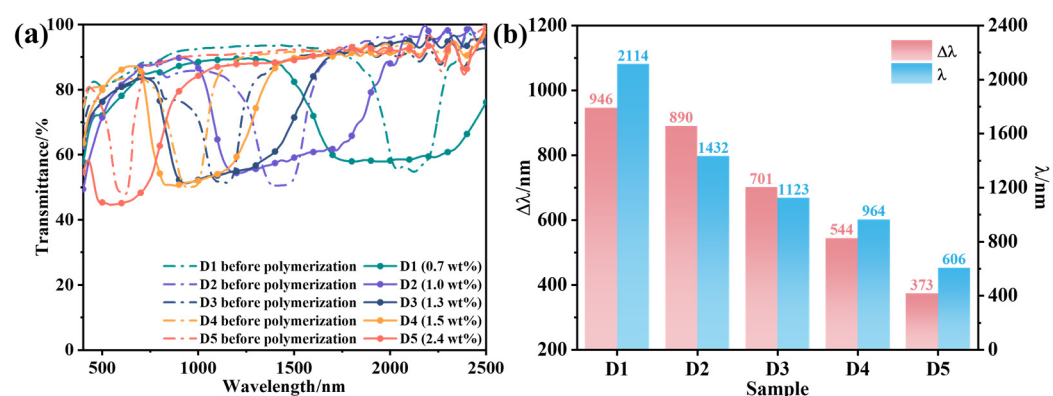


Figure 8. Group D samples: trends of (a) Vis-NIR transmission spectra; (b) $\Delta\lambda$ and λ with the content of chiral dopant R5011.

3.6. Effect of R5011 Concentration on the Broadband Reflection of Bilayer PSCLC Films

Through the aforementioned experiments, the optimal composition and polymerization conditions for the single-layer PSCLC film were established. To further broaden the reflection bandwidth of the films, bilayer broadband reflective films incorporating a PVA intermediate layer were fabricated in this section. In the experimental design, sample D2 was designated as the upper layer of the bilayer architecture. Excluding the D2-PVA-D2 combination, which would result in completely overlapping bandwidths, samples D1, D3, D4, and D5—featuring progressively increasing chiral dopant concentrations—were respectively selected as the lower-layer systems and subjected to a secondary UV polymerization. Figure 9 illustrates the Vis-NIR transmission spectra of the bilayer PSCLC films with varying lower-layer compositions. Because the composition of the upper D2 layer was fixed, the spectral position of its corresponding reflection band remained essentially invariant. The variations in the overall reflection spectra of the bilayer films primarily stemmed from the shift in the central wavelength, induced by differences in the R5011 concentration within the lower layer. The spectral curves indicate that as the R5011 concentration in the lower LC system increased, the helical pitch within the lower film decreased. This reduction led to a progressive blue-shift in its central reflection wavelength, along with a narrowing of its intrinsic reflection bandwidth. As depicted in Figure 9, regarding the D2-PVA-D1 sample, the reflection band of the lower film is situated within the longer wavelength region. Owing to the effective physical barrier of the PVA intermediate layer, the bilayer film exhibited two distinct reflection peaks prior to polymerization. After the secondary polymerization, the long-wavelength reflection band of the lower layer and the short-wavelength reflection band of the upper D2 layer were successfully concatenated. The synergistic effect of these two broadband reflection bands significantly broadened the overall reflection bandwidth of the bilayer film, effectively extending its reflection range into the IR region. The selective reflection bandwidth reached a maximum value of 1489 nm.

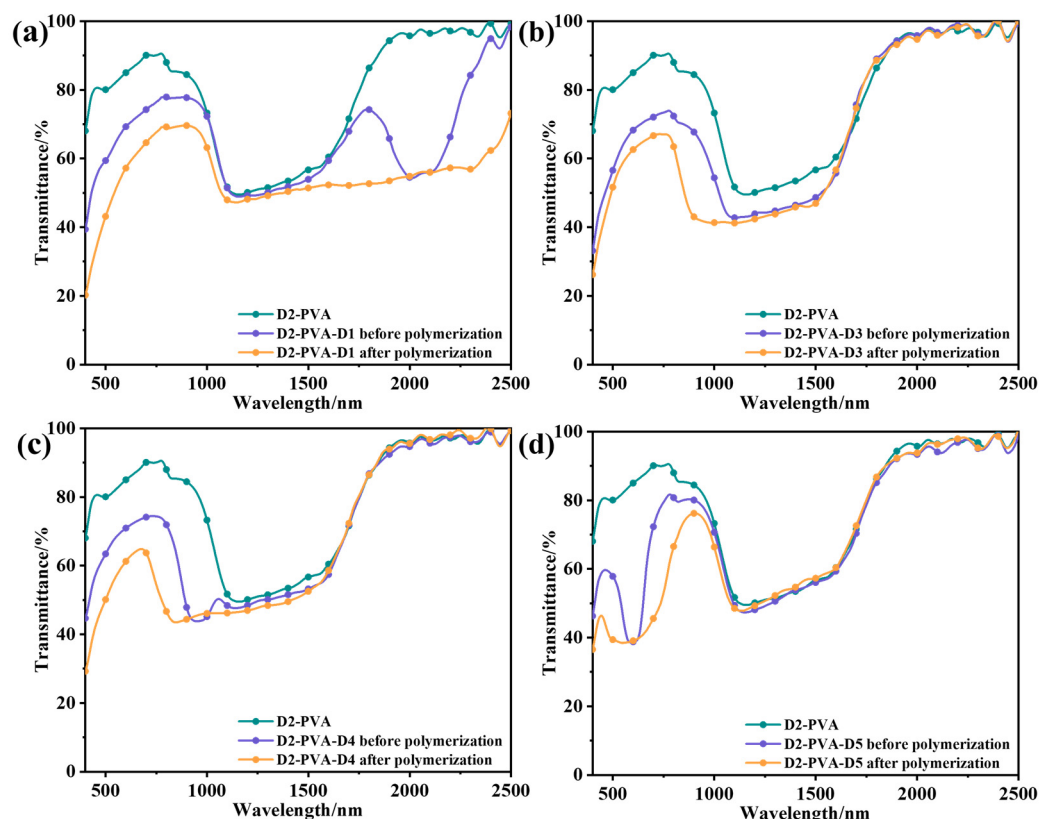


Figure 9. Vis-NIR transmission spectra of bilayer PSCLC films: (a) D2-PVA-D1; (b) D2-PVA-D3; (c) D2-PVA-D4; (d) D2-PVA-D5.

As the R5011 concentration in the lower layer was further elevated from D1 to D3 and D4, the central wavelength of the lower film underwent a pronounced blue-shift, accompanied by a narrowing of its intrinsic bandwidth. This resulted in a spectral overlap with the upper D2 layer. The results indicated that the reflection bandwidths of both D3 and D4 partially overlapped with that of D2; however, because the reflection band of D4 was situated in a shorter wavelength range, its spectral overlapping region with D2 was comparatively smaller. Therefore, the D2-PVA-D4 bilayer film exhibited a broader overall reflection bandwidth than the D2-PVA-D3 configuration.

When the chiral dopant concentration in the lower layer was further increased to that of D5, the central reflection wavelength of the single-layer D5 sample was located at 606 nm. Under these conditions, the reflection bands of D5 and D2 could not be concatenated, manifesting as two independent reflection peaks both prior to and following polymerization. As a result, the film failed to achieve a continuous broadband reflection. In summary, by rationally adjusting the chiral dopant concentration within the upper and lower CLC systems, the spectral positions of the reflection bands for each layer can be precisely modulated, achieving the spectral concatenation of multiple reflection bands. In this study, the optimal concatenation of the upper and lower reflection bands was realized when D2 and D1 were selected as the upper and lower layers, respectively, yielding a maximum reflection bandwidth of 1489 nm for the bilayer film.

3.7. Performance Characterization and Applications of Optimal Bilayer and Trilayer Broadband Reflective PSCLC Films

Through the aforementioned experiments, the optimal composition and polymerization conditions for the single-layer broadband reflective PSCLC films have been established. Given that the fundamental compositions—with the exception of the variations in the chiral dopant (R5011) content—and the polymerization conditions of the bilayer and tri-

layer systems are identical to those of the single-layer system, and considering that the concentrations of C6M and UV-327, alongside the UV polymerization parameters, have been thoroughly investigated and optimized in preceding sections, these variables were not redundantly explored for the multilayer systems in this section.

As depicted in Figure 10a, following ultraviolet (UV)-induced polymerization, the reflection bandwidth of the D2-PVA-D1 bilayer PSCLC film was significantly broadened compared to those of the polymerized upper and lower single-layer films. To further broaden the reflection bandwidth, a trilayer composite film was fabricated utilizing the D2, D4, and D1 systems. Specifically, following the polymerization of the single-layer D2 and D1 films and the subsequent coating of PVA films onto their surfaces, the unpolymerized D4 CLC mixture was infiltrated into the gap between the two PVA layers. Transmission spectra revealed that upon the infiltration of D4, but prior to its polymerization, an isolated narrow-band reflection peak was present in the spectrum. This corresponds to the intrinsic narrow-band reflection characteristic of the initial D4 LC mixture. After the third UV-induced polymerization step, a pitch gradient was established within the D4 layer along the thickness direction of the LC cell. The three independent pitch gradients thus merged into a cohesive entity along the thickness direction. The overall reflection bandwidth of the multilayer LC film was further extended to 1650 nm. As illustrated in Figure 11, throughout the entire fabrication process of the bilayer film—namely, prior to and following the single-layer UV polymerization, after the deposition of the PVA film, and before and after the secondary polymerization—the LC films consistently exhibited a well-defined planar texture. This demonstrates that the incorporation of the PVA film did not disrupt the intrinsic molecular alignment of the LC films. It should be emphasized that all presented POM images are highly representative of the macroscopic morphology under their respective conditions. They were captured randomly across the samples rather than being selectively chosen for specific local features, thereby confirming the excellent large-area uniformity of the planar alignment.

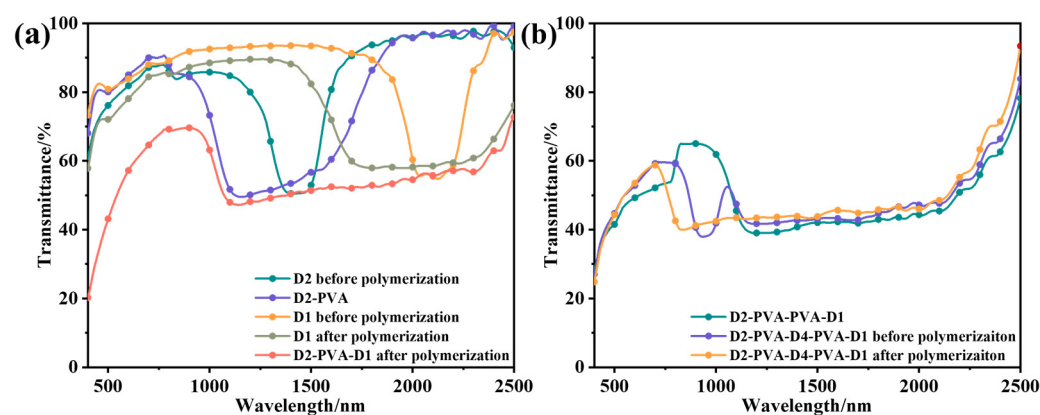


Figure 10. Vis-NIR transmission spectra of the optimal multilayer PSCLC films: (a) bilayer structure; (b) trilayer structure.

To evaluate the practical application potential of the multilayer PSCLC broadband reflective films in smart windows and energy-efficient architecture, an infrared (IR) shielding model was constructed to simulate temperature variations within an enclosed space under solar irradiation. In the simulated test, the model house was constructed from wood with internal dimensions of 9.5 cm × 6 cm × 7 cm. The window size was set to 2 cm × 3 cm. An infrared lamp was utilized to simulate the thermal radiation of sunlight, and its irradiation distance and power were maintained strictly constant throughout all comparative experiments. A blank LC cell (composed of ITO-coated glass substrates) and single-layer (D2), bilayer (D2-PVA-D1), and trilayer (D2-PVA-D4-PVA-D1) PSCLC films

were attached to the model window as IR reflectors. The dynamic variation in the internal temperature under continuous simulated solar irradiation over a duration of 0 to 35 min was recorded and is summarized in Figure 12. The experimental results demonstrate that following 30 min of continuous irradiation, substantial heat accumulated within the model equipped with the blank ITO LC cell, resulting in a significant temperature elevation from an initial 19.9 °C to 28.1 °C; after 30 min, the temperature remained stable. In contrast, the models equipped with the PSCLC films exhibited a mitigated temperature ascent. Furthermore, as the number of film layers increased and the reflection bandwidth broadened, the final steady-state temperature progressively decreased. Notably, the trilayer PSCLC film demonstrated the optimal IR shielding performance, yielding a temperature reduction of 4.1 °C compared to the blank control group. These findings indicate that the broadband reflective films constructed via the incorporation of PVA intermediate layers can effectively obstruct IR thermal energy from solar radiation, exhibiting substantial application potential in mitigating indoor heat accumulation and facilitating energy conservation and emission reduction.

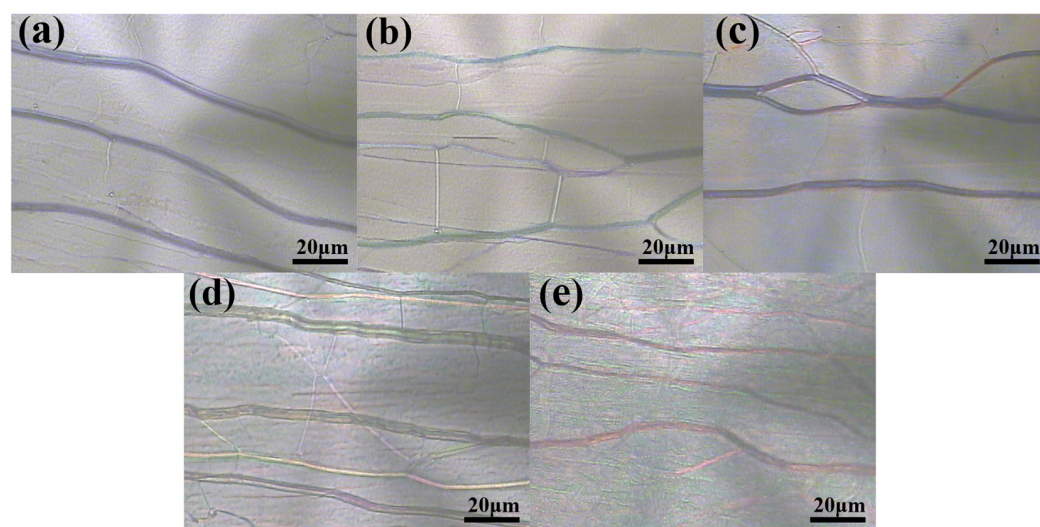


Figure 11. POM images at different fabrication stages of the bilayer PSCLC film: (a) single layer before polymerization; (b) single layer after polymerization; (c) after PVA coating; (d) bilayer before polymerization; (e) bilayer after polymerization.

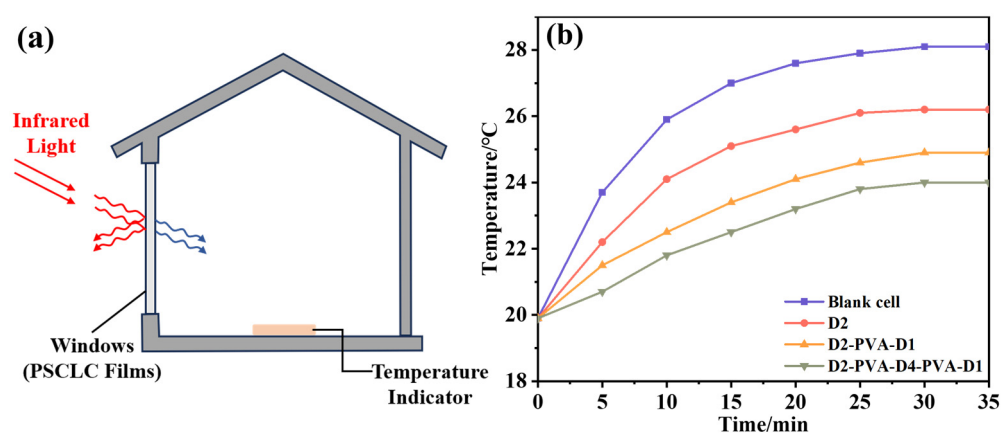


Figure 12. Infrared shielding test of the multilayer PSCLC broadband reflective films: (a) schematic diagram of the infrared shielding test model (The red arrows represent the incident and reflected infrared light, while the blue arrows represent the transmitted infrared light.); (b) internal temperature variation curves over time for models with different films (the blank cell is composed of ITO-coated glass substrates).

4. Conclusions

In this study, multilayer broadband reflective PSCLC films were successfully constructed utilizing PVA barrier layers. This approach effectively resolves issues typically associated with conventionally stacked LC films, such as refractive index mismatch, interlayer molecular diffusion, and weak interlayer adhesion, thereby mitigating interfacial scattering and optical losses. Under the synergistic effect of UV irradiation and the UV-absorbing dye, the optical intensity attenuation generated along the thickness direction of the single-layer PSCLC film induces a disparity in polymerization rates. This differential drives molecular diffusion within the system, yielding a continuous pitch gradient. Owing to the physical barrier effect of the PVA layer, the upper and lower LC layers can form their respective independent reflection bands during the secondary polymerization. By precisely regulating the content of the chiral dopant R5011 in each layer, the reflection bands of the respective layers are spectrally concatenated, ultimately realizing the bilayer broadband reflection effect. Building upon this foundation, a trilayer PSCLC architecture was constructed via the further incorporation of intermediate layers, which further broadened the reflection bandwidth, ultimately achieving an ultra-broad reflection band of 1650 nm. The fabricated bilayer broadband reflective PSCLC films were characterized via polarized optical microscopy (POM). Observations revealed that throughout every fabrication stage of the multilayer films—including prior to and following the single-layer polymerization, after the deposition of the PVA film, and before and after the secondary polymerization—the LC film system consistently maintained a well-defined planar texture. This demonstrates that the incorporation of the PVA intermediate layer did not disrupt the intrinsic molecular alignment of the LC films. Finally, the infrared shielding performance of the optimal sample was evaluated. Under simulated solar irradiation, compared to the blank ITO cell, the temperature ascent in the model equipped with the PSCLC film was significantly mitigated. This confirms the great promise of the film for energy-saving applications and optical display devices. It should be noted that the current films function as passive infrared reflectors. While they effectively reduce indoor heat accumulation, they currently lack dynamic temperature-responsive adaptability (e.g., transmitting IR in winter while reflecting in summer). Integrating these ultra-broadband films with environmentally responsive materials for intelligent thermal management will be the focus of our future research.

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Abbreviations

The following abbreviations are used in this manuscript:

PET	polyethylene terephthalate
POM	polarized optical microscopy
CLCs	cholesteric liquid crystals
PSCLCs	polymer-stabilized cholesteric liquid crystals
PVA	poly(vinyl alcohol)
IR	infrared
UV	ultraviolet

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