TOPICAL REVIEW — Physical research in liquid crystal

Research progress of cholesteric liquid crystals with broadband reflection characteristics in application of intelligent optical modulation materials*

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Cholesteric liquid crystals (CLCs) have recently sparked an enormous amount of interest in the development of soft matter materials due to their unique ability to self-organize into a helical supra-molecular architecture and their excellent selective reflection of light based on the Bragg relationship. Nowadays, by the virtue of building the self-organized nanostructures with pitch gradient or non-uniform pitch distribution, extensive work has already been performed to obtain CLC films with a broad reflection band. Based on authors' many years' research experience, this critical review systematically summarizes the physical and optical background of the CLCs with broadband reflection characteristics, methods to obtain broadband reflection of CLCs, as well as the application in the field of intelligent optical modulation materials. Combined with the research status and the advantages in the field, the important basic and applied scientific problems in the research direction are also introduced.

Keywords: cholesteric liquid crystals, soft matter materials, broadband reflection characteristics, intelligent optical modulation materials

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1. Introduction

Cholesteric liquid crystals (CLCs), due to their special ability to self-organize into a helical supra-molecular architecture and unique optical and electrical properties, have received increasing attention over the last few decades. Since the first discovery of CLCs in cholesterol benzoate compounds,[1,2] the phase structure of CLCs has been extensively studied and found in substances not connected with cholesterol. In contrast from the nematic liquid crystal (NLC) phase as the characteristic of a purely orientational order of elongated molecules,[3,4] the CLC phase exhibits a spontaneous helical structure with a twist axis perpendicular to the local direction, which comes in whole or in part from the molecular chirality of rod-like molecules. Usually, as shown in Fig. 1, the helical cholesteric structure is considered as a stack of layers with an orientational order of molecules in each plane and a rotation by a constant angle of each plane with respect to its neighbors. The distance along the helical axis that corresponds to a rotation of 360° in the orientation of the rod-like molecules is defined as the pitch (P). Additionally, another important structural parameter to evaluate the CLC phase is the twist sense, which determines the handedness of a helix (left- or right-handed) and strongly depends on the configuration of the chiral group(s) within the molecule. From the macroscopic perspective, the handedness determines the optical reflective feature (see Section 2).

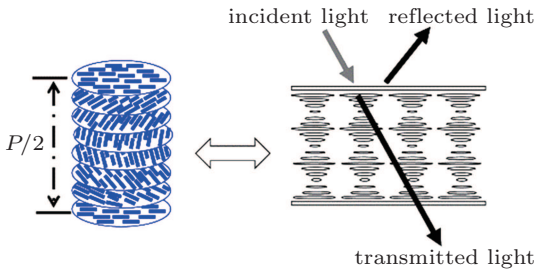


Fig. 1. (color online) The helical structure of the cholesteric liquid crystal phase.

Generally, CLC materials can be obtained by two methods: synthesis of compounds with asymmetric structures by introduction of chiral carbon atoms or cholesteryl groups, and so on.[5–7] The P of the chiral compound is greatly dependent

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on the inherent nature of the chiral group. Additionally, by addition of a small quantity of chiral optically active substances into an NLC, and if the miscibility conditions are satisfied, a CLC mixture can be artificially obtained.[8–12] At this point, the P of the mixture is dependent on the content of the chiral substance, and it can be easily and randomly modulated from a few tens of nanometers to many micrometers. Up to now, scientists have developed hundreds of thousands of kinds commercial CLC mixture formulas for application research.

Due to the unique helical structures of CLCs, they exhibit particular optical properties such as characteristics of selective light reflection,[13,14] optical rotation effect,[15] and circular dichroism,[16] which distinguish them from the common LC materials. Furthermore, CLCs also present special electro-optic effects,[17,18] for example, memory effect, grating effect, as well as the withdrawal effect of spiral. Consequently, CLCs have been extensively studied and widely used in the fields of temperature and pressure sensors,[19] polarizer-free reflective displays,[20] lasing applications,[21] optical data storage media,[22] polarization-independent devices,[23,24] stealth technologies,[25,26] smart-window prototypes,[27] and so on. Reviews[28] have reported the recent advances in the field.

Accordingly, in this critical review, the following key issues are focused on: (i) the basic optical property of CLCs, (ii) the mechanism of the broadband reflection, (iii) the methods to obtain the broadband reflection, (iv) the applications of CLCs in the field of intelligent optical modulation materials. In the conclusion part, the basic and applied scientific problems and the future development directions are elaborated.

2. The optical property of CLCs

As discussed above, due to the unique helical structures, CLCs exhibit particular optical properties such as characteristics of selective light reflection, optical rotation effect, and circular dichroism. In this review, we only focus on the selective light reflection property, which is directly related to the applications of the smart optical modulation materials.

2.1. The selective light reflection of CLCs

Normally, under the irradiation of a white light, CLCs will represent beautiful colors, and the colors will alter with the change of the incident angle, which is the result of selective light reflection of CLCs based on the Bragg relationship. Due to the periodic change of the refractive indices arising from the helical structure, CLCs can selectively reflect the incident light along the screw axis direction. The central wavelength λ and the wavelength range (abbreviation of the bandwidth) $\Delta\lambda$ of the selective reflection are denoted as $\lambda = nP$ and $\Delta\lambda = \Delta nP$, respectively, where $n = (n_o + n_e)/2$ is the average of the ordinary (n_o) and the extraordinary (n_e) refractive indices of the locally uniaxial structure, P is the helical pitch, and $\Delta n = n_e - n_o$ is the birefringence. Within the reflection bandwidth, cholesteric liquid crystals with left-handed helical structure allow right-handed circularly polarized light to go through and reflect left-handed circularly polarized light, and vice versa (as shown in Fig. 2). Beyond the reflection bandwidth, both left-handed and right-handed circularly polarized lights are transmitted.

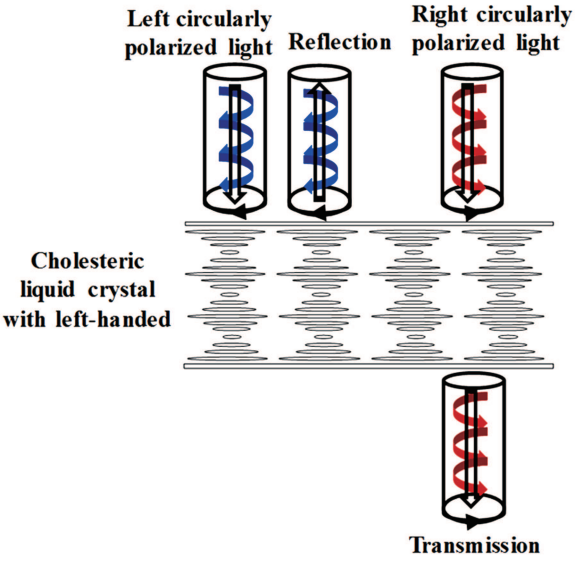


Fig. 2. (color online) Schematic of the selective reflection of circular polarized light of CLCs.

2.2. The mechanism of broadband reflection feature of CLCs

According to the above equation $\Delta\lambda = \Delta nP = (n_e - n_o)P$, we can see that the bandwidth $\Delta\lambda$ is dependent on the birefringence Δn and P at normal incidence. The smaller the Δn and P, the smaller the $\Delta\lambda$. For most colorless organic materials, Δn is generally not larger than 0.3, therefore, $\Delta\lambda$ is often less than 100 nm in the visible spectrum.[29] Furthermore, due to the polarization-selectivity rule, when an ordinary light goes through a CLC, the maximal reflected intensity is usually limited to 50%. Figure 3 shows a representative selective light reflection spectrum of CLCs.

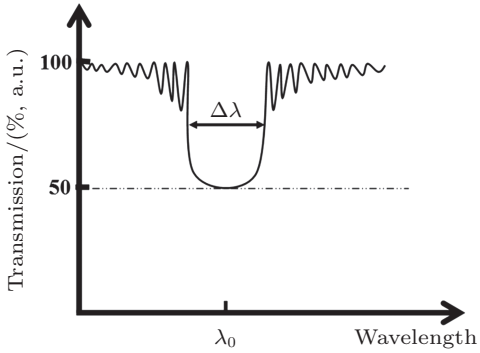


Fig. 3. Representative selective light reflection spectrum of CLCs.

Due to the synthetic complexity, high viscosity, poor chemical and thermal stabilities, as well as the color defect of CLC materials with high Δn,[30–34] it is difficult to modulate $\Delta\lambda$ by simply increasing Δn. Then at the subsequent research, these researchers devoted their work to develop materials with pitch gradient or non-uniform pitch distribution. By the stack of the reflection bandwidth, broadband reflection of CLC materials can be realized. Figure 4 presents the theory of broadening the bandwidth of a CLC.

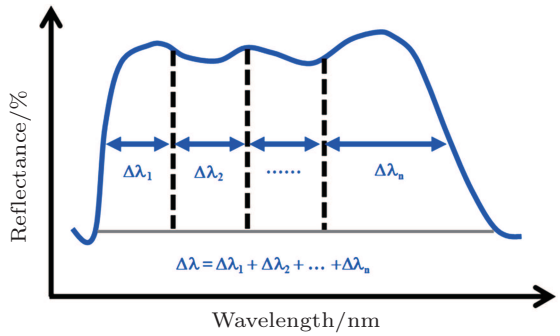


Fig. 4. (color online) Theory of broadening the bandwidth of a CLC.

3. The methods that have been used to obtain the broadband reflection of CLCs

As mentioned above, the selective light reflection bandwidth of CLCs with single pitch is narrow, which is difficult to meet the demand in the applications of color display, light enhancement film, and so on. Consequently, many researchers devoted to broadening the reflection bandwidth by forming pitch gradient distribution or pitch non-uniform distribution in CLC films.

3.1. Thermal diffusion induced broadband reflection

Using a stack of right-handed (RH) CLCs reflecting red, green, and blue, respectively, Li obtained a CLC film which covers the visible region. The results showed that the stacked multi-layer system was not a simple addition of the performance of the single layer, but there was a phenomenon of mutual diffusion between the interface. By utilizing the CLCs and a quarter-wave plate (QWP) in a display backlight, the light enhancement effect for a notebook computer backlight source has been systematically investigated.[35]

As shown in Fig. 5, by stacking three chiral polymer films with different pitch lengths, Wu successfully synthesized a broadband circular polarizer with bandwidth ranging from 400 nm to 736 nm.[36]

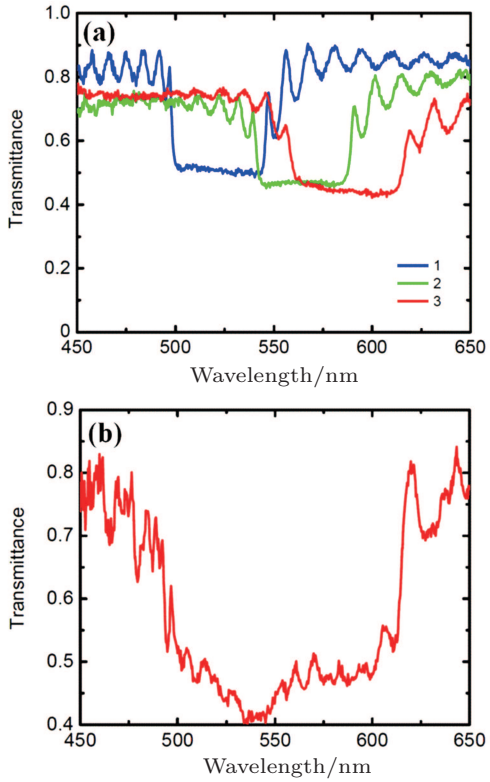


Fig. 5. (color online) Transmission spectra of (a) three individual CLC polymer films and (b) the three stacked CLC polymer film.[36]

By using a simple thermal diffusion process in a glassy CLC polymer, Mitov prepared a film with a wavelength bandwidth greater than 300 nm covering the whole visible region. The evolution of the micro-structure of the film with respect to the processing time and the mechanism of the diffusion was clarified.[29,37–41] The results showed that appropriate heat treatment and process time had important influences on the broadening of the pitch, and the success of the annealing process was mainly due to the disappearance of the initial interface and the establishment of the progressive pitched transitional area (see Fig. 6).

Employing the selective light reflection characteristic of cholesteristic side-chain-liquid-crystalline-polymers (ChSCLCP), and by stacking the EVA/ChSCLCP composite film powders with different reflection wavelengths, Yang successfully developed a novel soft matter composite material for laminated glass (see Fig. 7). The preparation method and the optimized conditions of the composite film were systematically investigated. UV/VIS/IR experimental results illustrated that the obtained wide-band reflective film not only maintained the reflection properties of the original single composite films, but also showed new center reflection wavelengths, which were probably corresponding to the new pitch lengths arising from two adjacent different powders, and the reflection bandwidth was also significantly broadened due to the thermal diffusion between the neighboring composite powders.[42]

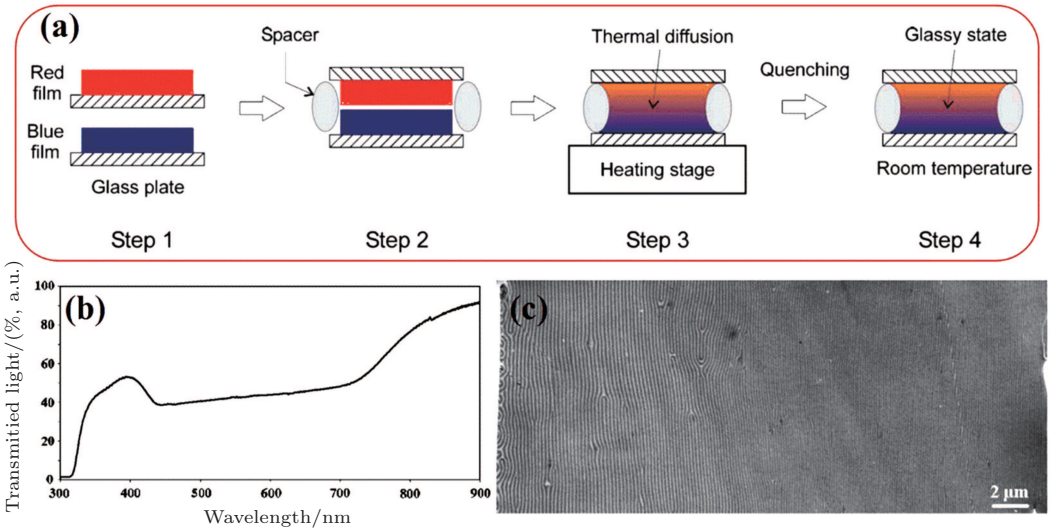


Fig. 6. (color online) (a) Schematic representation of the mechanism of the thermal diffusion induced pitch gradient, (b) the transmission spectrum of the bilayer film after annealing, and (c) the representative TEM micrograph of the bilayer film.[40]

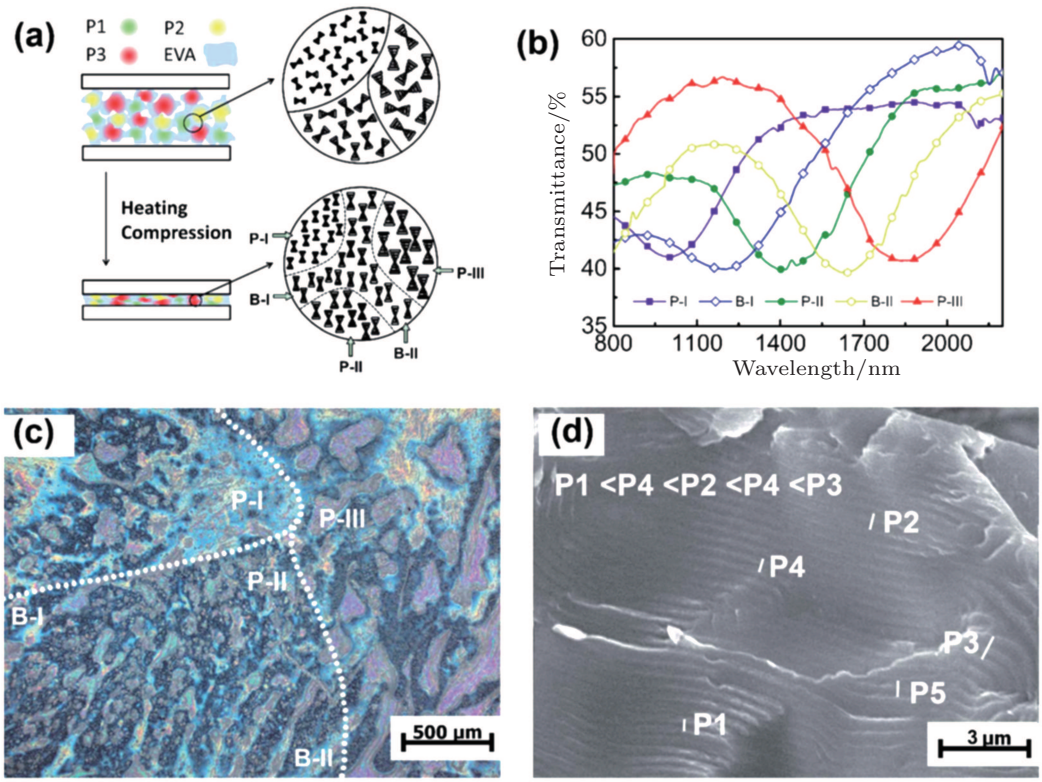


Fig. 7. (color online) (a) Schematic drawing of the mechanism of the thermal diffusion induced pitch gradient, (b) the transmission spectrum of the IR shielding film in different areas, (c) the polarized optical microscope photograph of the film, and (d) the representative SEM micrograph of the cross-section of the film.[42]

3.2. Concentration difference of chiral compound induced pitch with non-uniform distribution

In 1995, Broer first prepared LC films with broadband reflection covering the whole visible region by photopolymerization of chiral LC monomer with bifunctional groups/LC monomers with mono-functional group/ultraviolet absorbance system under UV light.[43] The difference in reactivity between the LC monomers with bifunctional groups and mono-functional group and a gradient in ultraviolet intensity perpendicular to the film arising from the existence of dyes promoted the formation of the pitch gradient. The polymerization rate was fast at the top of the film (lamp-side), resulting in a fast consumption of the most reactive monomer at the same location, the depletion of this monomer started a diffusion process. Therefore, the pitch at the top of the film was relatively smaller, whereas the pitch at the bottom was larger, a pith with non-uniform distribution was formed and broadband reflection was achieved (see Fig. 8).

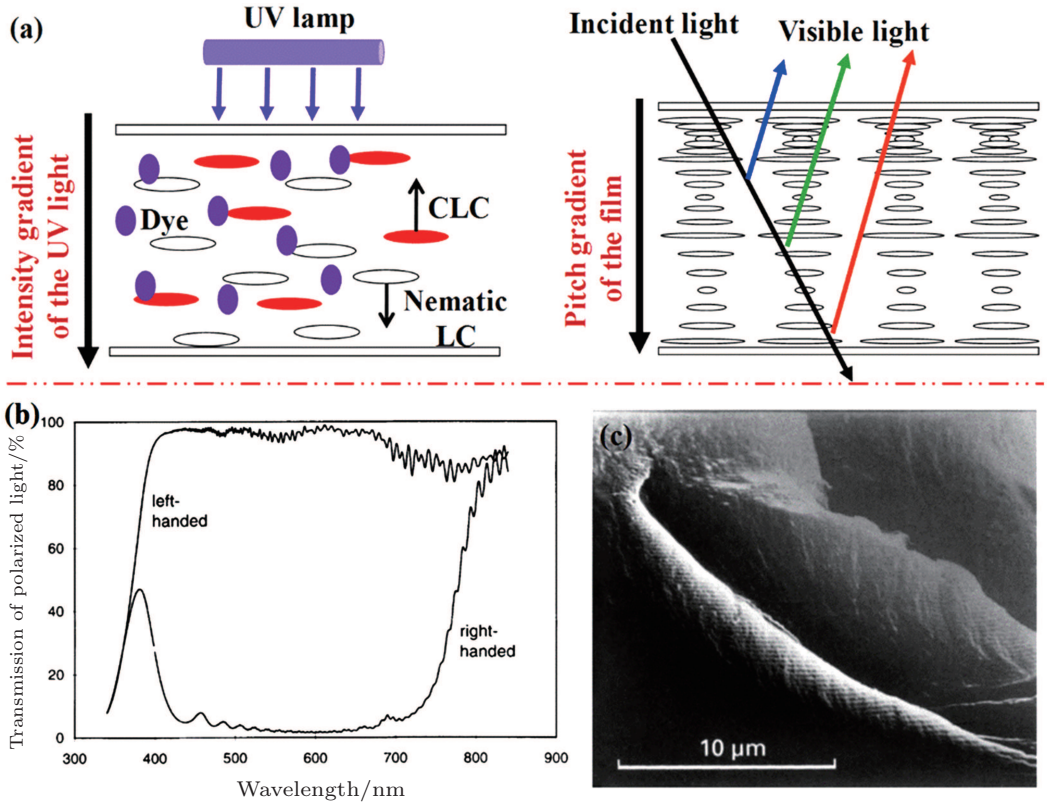


Fig. 8. (color online) (a) Schematic representation of the mechanism of the concentration difference induced pitch gradient, (b) the transmission spectrum of a 15-μm-thick film of the cholesteric pitch-gradient network, and (c) the SEM image of the fracture surface of the network.[43]

By spin-coating and assembling two layers of cholesteric photo-polymerizable monomers/chiral compounds having different pitches at the same temperature, due to the concentration gradient that induced the diffusion of the chiral compound over the film thickness, Sixous obtained a more or less large diffusion between the two layers, which was dependent on various parameters such as the degree of cross-linking of the two layers, the thickness of the layers, the temperature dependence, as well as the time evolution at a given temperature.[44,45] The results showed that CLC polymer films obtained by a system of a left-handed CLC polymer /a right-handed chiral compound can exhibit more wide reflection bandwidth with the existence of a pitch gradient inside the sample, and the contact time has important effects on the reflective bandwidth. As shown in Fig. 9, at shorter time, diffusion occurred at the interface and the reflection bandwidth was broadened, however, after a longer time, the broadening band decreased and a single large peak was observed.

Utilizing the diffusion velocity difference between crystallization phase and liquid crystal phase of photo-polymerizable cholesteric liquid crystal monomer with different pitches, Yang prepared the cholesteric liquid crystal polymer films with non-uniform pitch distribution (see Fig. 10). The effect of the molecular structures of monomers on the microstructure and optical properties of the film was investigated and the method of regulating the bandwidth of the left-handed (or right-handed) circularly polarized light reflected by the film was clarified.[46]

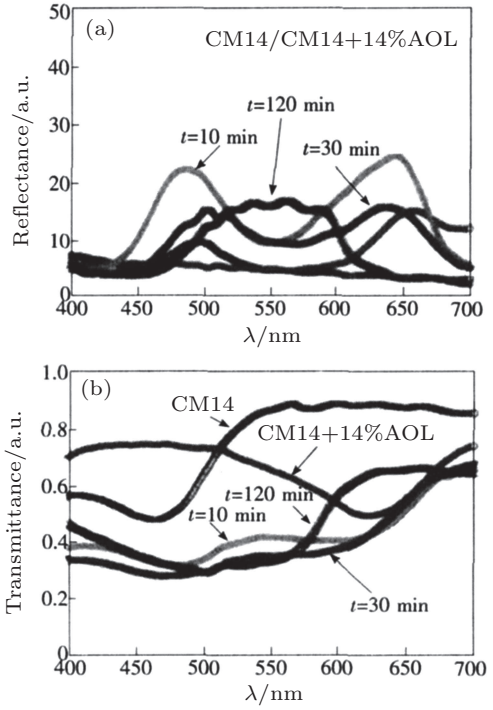


Fig. 9. (a) Reflection and (b) transmission spectra of the CLC polymer film with different contact time.[44]

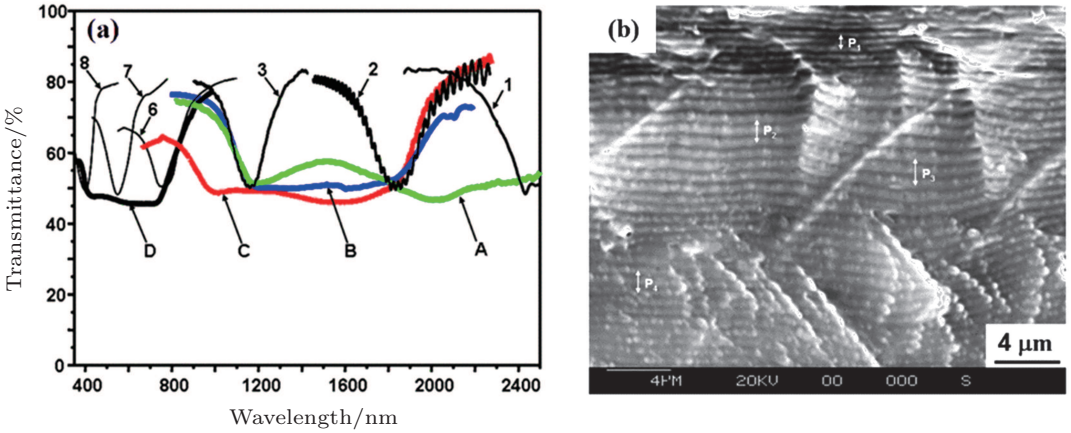


Fig. 10. (color online) (a) Transmission spectrum of the CLC polymer film with non-uniform pitch distribution and (b) SEM image of the fractured surface of the film.[46]

3.3. Polymerization induced molecular re-distribution

Based on CLC materials containing one polymeric LC component and other non-reactive LCs, Li obtained various single layer reflective polarizers over a bandwidth from 400 nm to 1000 nm with a non-linear pitch gradient by polymerization induced molecular re-distribution. In contrast from the work of Broer, a CLC blend containing only one polymerizable LC compound and no UV dye to promote the band broadening, due to the nonlinear pitch distribution created via the photo polymerization and molecular re-distribution, the UV polymerization of the cross-linkable CLC compound induced a molecular segregation of the non-cross-linkable nematic LC compound, the segregated nematic diffused along the UV field to form a density gradient at sites where the low molecular weight nematic is rich, the CLC pitch is longer, and vice versa (as shown in Fig. 11). Finally, a nonlinear pitch distribution was generated and this technology was employed to fabricated broadband polarizers in the visible as well as in infrared.[47–49]

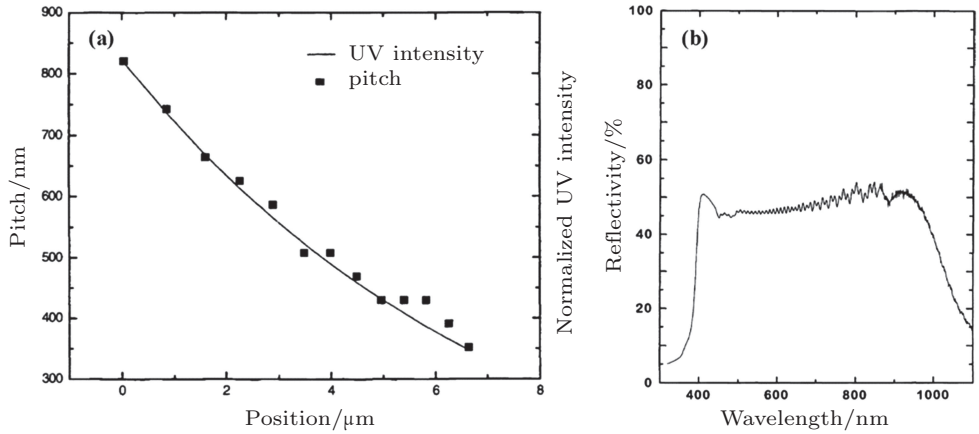


Fig. 11. (a) The CLC pitch of a film and the UV intensity variation as a function of position across the film thickness of an ultra-broadband CLC polarizer. (b) The reflection spectrum of an optimized CLC film.[48]

3.4. Photo-isomerization of chiral dopants induced pitch gradient

Using the difference of the helical twisting power (HTP) caused by photo-isomerization of chiral dopants under the irradiation of ultraviolet light, van de Witte prepared a monolayer CLC film with pitch gradient distribution that can reflect the whole visible region.[50] Figure 12 shows the chemical structures of the photo-isomerizable CLC and the circularly polarized transmission spectra of the film.

By doping azobenzene chiral compounds into cholesteric liquid crystal, Yang synthesized a series of films with large pitch gradient. The films were prepared due to the difference of helical twist power caused by the photo-induced cis-trans isomerization (see Fig. 13). These films can shield infrared light with wavelengths of 1000–2400 nm.[51]

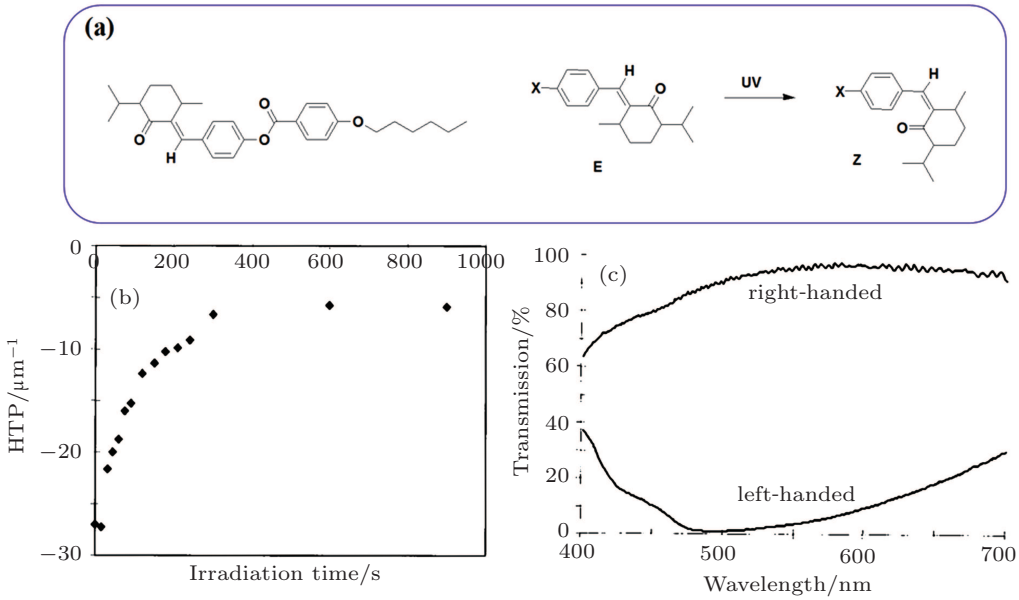


Fig. 12. (color online) (a) Chemical structure of the chiral compound and the light isomerization process of the chiral group, (b) the helical twisting power of the chiral compound as a function of irradiation time, and (c) the circularly polarized transmission spectrum of the resulting film.[50]

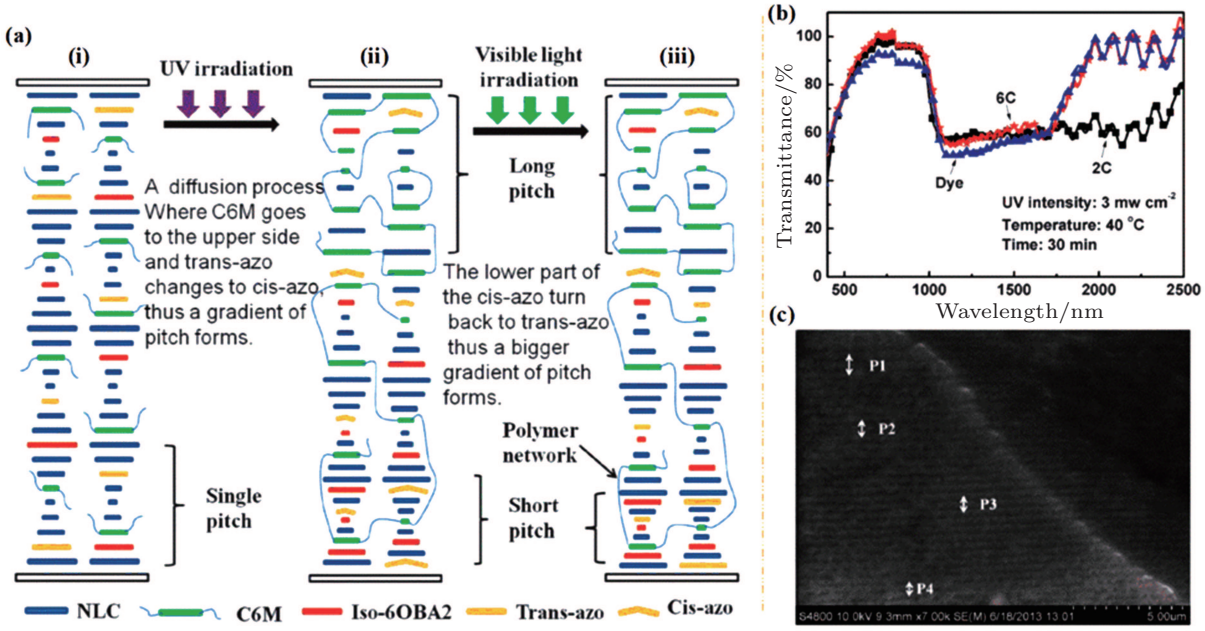


Fig. 13. (color online) (a) Schematic presentation of the possible mechanism of the chiral dopant induced pitch gradient, (b) the transmission spectra of the films, and (c) the SEM image of the fracture surface of the film.[51]

3.5. Thermally controllable broadband reflection

In 2003, Yang *et al.* synthesized a chiral compound of which the HTP increases with increasing temperature (the chemical structure was illustrated in Fig. 14(a)).[52] By UV irradiation polymerization of the photo-polymerized LC monomer, a (polymer network/nematic liquid crystal/chiral dopant) composite film exhibiting a cholesteric phase at room temperature was developed. Due to the thermal sensitivity of the HTP of the chiral dopant and the effect of the polymer network on the molecular rearrangement of the LC, the bandwidth of the selective reflection spectrum of the composite film became wider and narrower reversibly with increasing and decreasing temperature, respectively. Thus the pitch gradient distribution of the CLC molecules can be realized through temperature changes. Figures 14(b) and 14(c) show a schematic of the mechanism of the thermally controllable broadband reflection of the cell and the transmission spectra of the samples. After that, novel chiral compounds containing binaphthalene structure,[53,54] two chiral center structure,[55] as well as benzoic acid isosorbide ester structure,[56] the helical twisted power of which decreased or increased with the variation of temperature, were synthesized by Yang. And single layer CLC films with non-uniform pitch distribution by utilizing the chiral compounds were demonstrated.

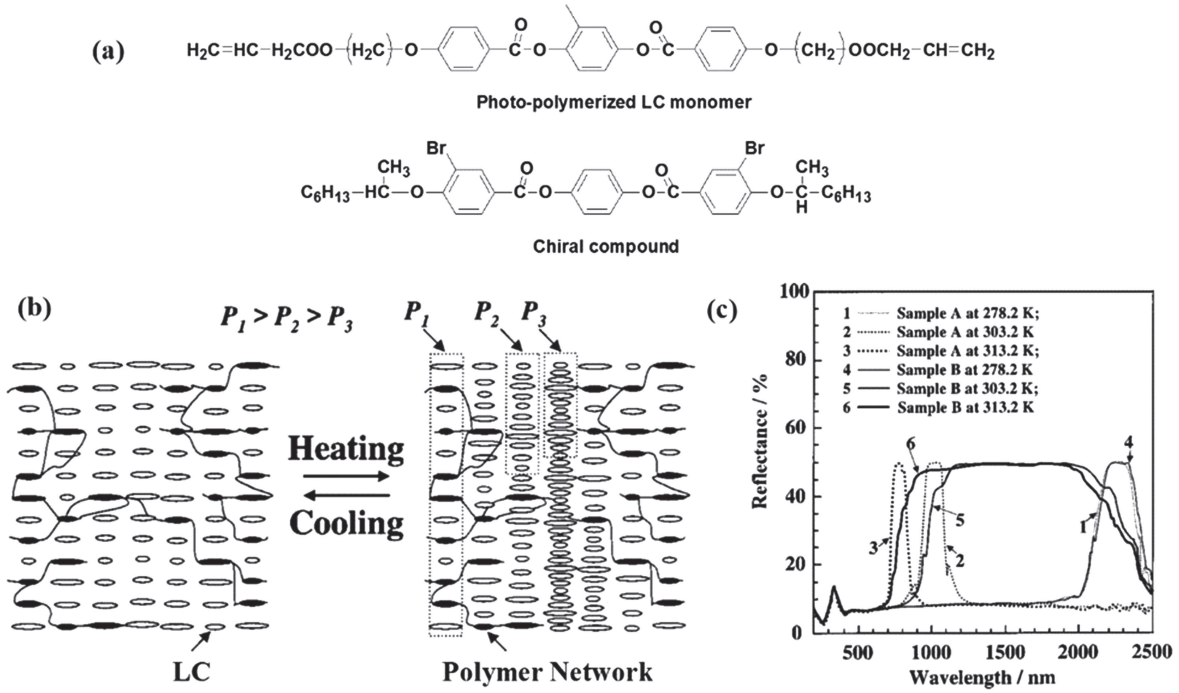


Fig. 14. (a) The chemical structure of the photo-polymerized LC monomer and chiral compound, (b) schematic of the mechanism of the thermally controllable broadband reflection of the cell, and (c) the transmission spectra of the samples.[52]

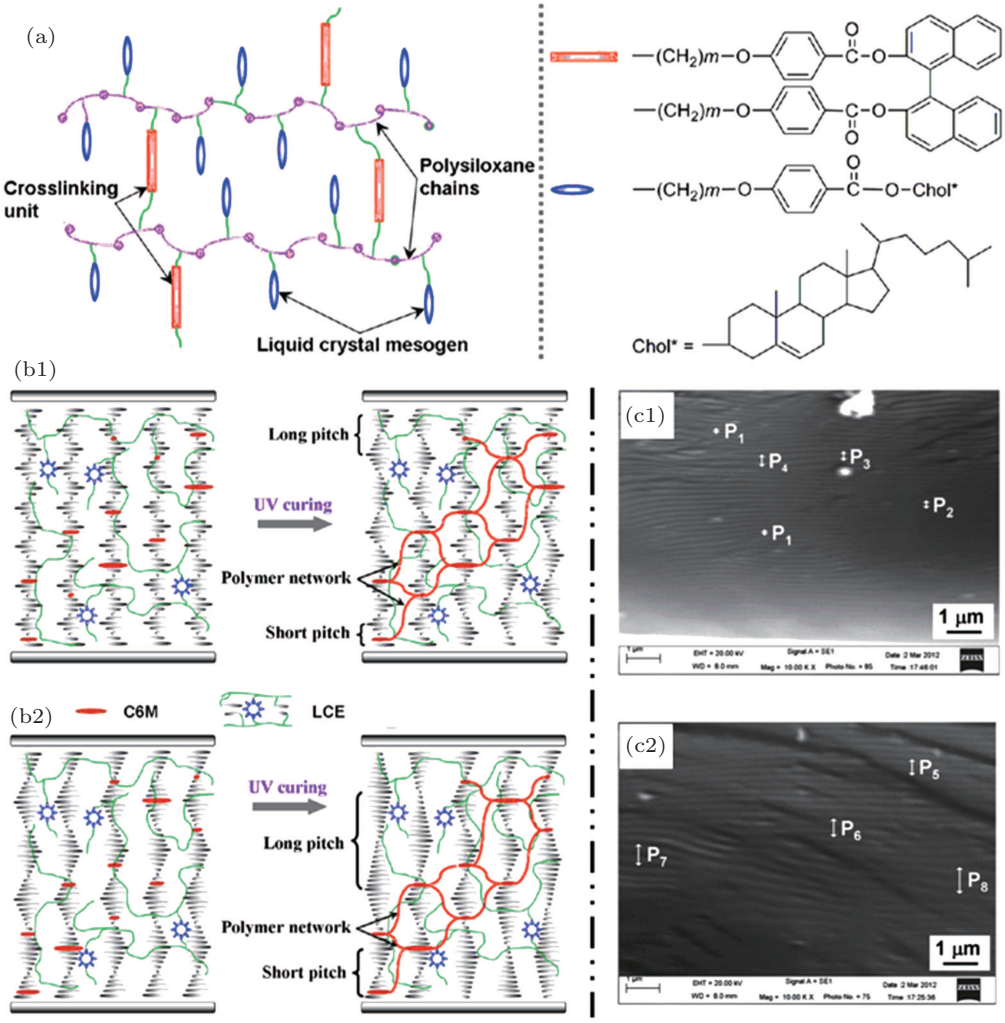


Fig. 15. (color online) (a) Schematic representation of the elastomer, possible mechanism of thermally controllable broadband reflection of the ChLCE with shorter spacer (b1) and longer spacer (b2), the corresponding SEM photographs of the fractured surfaces of the ChLCE with shorter spacer (c1) and longer spacer (c2).[57]

Based on the temperature sensitive characteristics of the HTP of the chiral compound, Yang designed and synthesized a series of side-chain polysiloxane cholesteric LC elastomer (ChLCE) with binaphthalene derivatives as crosslinkings and cholesterol derivatives as mesogenic groups.[57–59] Interestingly, the HTP of all the elastomers exhibited temperature dependence. By UV photo-polymerziation of the ChLCE/photo-polymerizable LC monomer/photo-initiator composite at appropriate temperature, both visible and near-infrared broadband reflective films with pitch non-distribution were obtained. Figures 15(b1) and 15(b2) present the possible mechanism of thermally controllable broadband reflection of the ChLCE with different phase structures. For LCE with shorter flexible spacer exhibiting cholesteric phase structure, the composite mixture was filled into a homogeneously treated cell at high temperature and a uniform short pitch was obtained before polymerization, then the short pitch was fixed in some local regions surrounded by the polymer network after UV curing. When cooling to a low temperature, the pitch far from the polymer network area became longer due to the thermally sensitivity of the HTP of the ChLCE, which led to the non-uniform distribution of the pitch. For LCE with longer flexible spacer exhibiting smectic A-cholesteric (SmA-Ch) phase structure, after polymerization of the composite at a temperature slightly higher than the phase transition of SmA-Ch, the short pitch of the cholesteric composite can be stabilized by the network. Then slow cooling to low temperature, due to the tendency of phase transition from cholesteric phase to SmA phase, for the ChLCE distant from the region of the polymer network, the pitch was obviously increased, which also led to the non-uniform distribution of the pitch in the film. The broadband reflection mechanism was confirmed by SEM investigations (see Figs. 15(c1) and 15(c2)).

3.6. Electronically controllable broadband reflection

Reports have shown that the electrically controllable method can make the LC molecules reorientation, which leads to the pitch changes and further controls the reflection wavelength.[60–62] Based on the above idea, Yang prepared a CLC/chiral ionic liquid (CIL) composite and filled them into a planar treated cell. When an electric field was applied to the cell, the anions and the cations of CIL moved towards the anode and the cathode of the power supply, respectively, thus forming a density gradient of the chiral groups, which resulted in wideband reflection. By adjusting the intensity of the electric field, the reflection bandwidth can be controlled accurately and reversibly. Moreover, the electric field-induced states can be memorized after the applied electric field is turned off. The mechanism of the electrically controllable reflection was successfully demonstrated by a combination of SEM and transmission spectrum investigation (as shown in Fig. 16).[63,64]

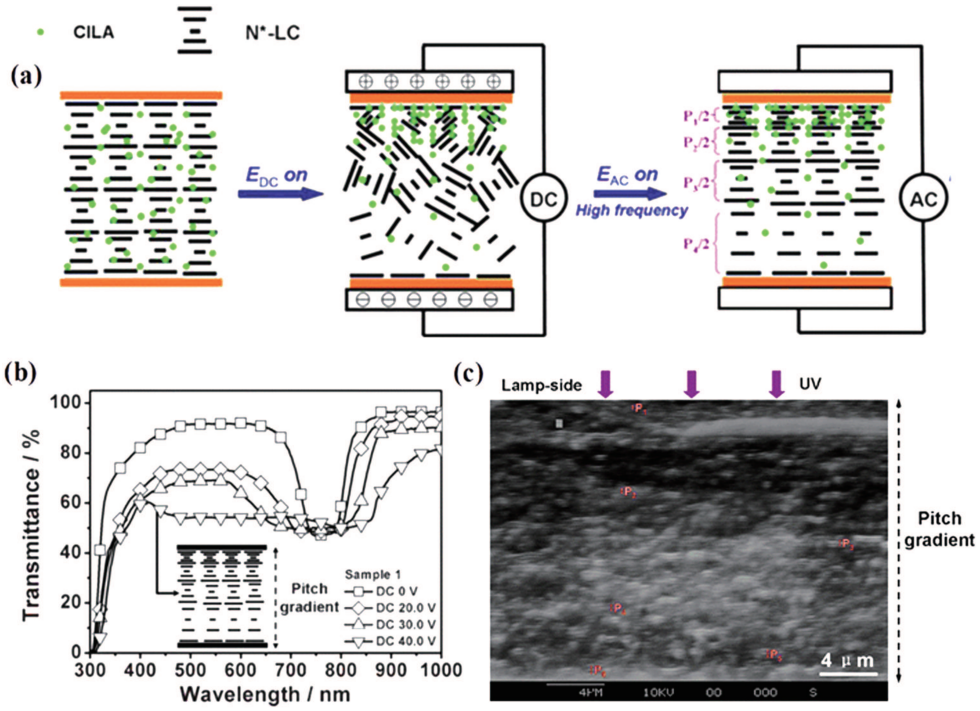


Fig. 16. (color online) (a) Schematic representation of the mechanism of electrically controllable broadband reflection of the cell, (b) the transmission spectra of the films, and (c) the SEM image of the fracture surface of the film.[63]

4. The application of CLCs with broadband reflection property

According to the principle of selective optical reflection characteristic of CLCs, we can conclude that once the pitch or pitch gradient of cholesteric liquid crystals can be regulated arbitrarily, the reflection wavelength and bandwidth could be regulated, which endows the materials with extensive applications.

4.1. Brightness enhancement films for liquid crystal display

As we all know, the LC itself does not shine. It needs a backlight source system to provide the light, and the brightness of the LC display (LCD) significantly affects the quality of the image. Due to the high proportion of power consumption by the backlight source system (for example, if the power consumption of LCD for a notebook computer is about 6.5 W, among which the power consumption of the backlight system is about 5.0 W, and the driving power consumption of the LCD is about 1.5 W), it is not a wise choice to increase the brightness of the backlight source itself.

Research has found that brightness enhancement films can increase the utilization of the backlight in LCD from about 1% to more than 40%. According to the calculation of 3M Corp, this will save 33,000,000 barrels of crude oil or 23,000,000 tons of coal if brightness enhancement films are utilized in LCD screens of 21 inches or larger in the five years from 2006 to 2010. But at present, the technology of brightness enhancement films is monopolized by 3M Corp (USA) and Merck Corp (Germany).

As mentioned previously, the selective reflection bandwidth of circularly polarized light in cholesteric liquid crystal is $\Delta\lambda = \Delta nP$. If $\Delta\lambda$ covers the wavelength range of visible light, the CLCs can be prepared into brightness enhancement films. Using CLCs materials that can reflect the whole visible region in prototype flat-panel LCDs, Broer reported that by inserting a stack of the pitch-gradient cholesteric layer and the quarter-wave foil between the backlight system and the dichroic polarizer, the light yield can be improved by 40% without any optimization of the backlight system.[43]

By utilizing a cholesteric liquid crystal film and a quarter-wave plate combination in a display backlight to both polarize and enhance the backlight output, Li demonstrated a 45% increase in forward propagating light intensity from a notebook computer backlight outfitted.[35] Based on the mechanism of concentration difference induced pitch with non-uniform distribution, by mixing the particles with different pitches of the cholesteric phase together with the crystalline phase and making the LC monomer molecules cross-linked by photopolymerization in the planarly oriented cholesteric phase, Yang prepared a CLC gel film with accurately controllable reflection bandwidth between 400nm and 750 nm of the circularly polarized light.[46] A mono-layer of brightness enhancement film with light utilization of more than 70%, low cost, and simple technology has been prepared by using this method and it is used in large area production.

4.2. Efficient infrared shielding films

It has been reported that infrared radiation of wavelengths 800–2000 nm accounts for more than 90% of the whole solar infrared radiation energy.[65,66] In the modern office building, glass windows and doors bear a thermal load of 30%–60% in summer, which is the main cause of indoor overheating. Efficient handling of solar radiation can significantly reduce energy consumption by reducing the demand for air-conditioning. It has been predicted that if smart window technology is available on a nationwide basis to modulate the heating and cooling control extremes in different climates, a total of $20 billion energy saving is estimated for each year in the national budget. An estimated 5% market penetration of the construction glass sector for the new smart window technology would create a billion-dollar market.[48]

Li has demonstrated a smart window for energy saving by stacking IR polarizers with both left- and right-handedness. Such a prototype IR smart window has an overall reflectivity above 80% in the near IR spectral region from 700 nm to 1700 nm, which spans the majority of the solar IR radiation power.[67,68]

As mentioned above, by stacking EVA/ChSCLCP composite film powders of different reflection wavelengths, Yang successfully developed a new type of IR shielding film with IR spectral region from 800 nm to 2000 nm. Model buildings equipped with and without the film were set up and an energy conservation efficiency of up to 40.4% was obtained, which proves the film's practical applications in energy-saving smart windows.[42] Additionally, by using chiral compounds, of which the helical twisting power increases with increasing temperature based on side-chain polysiloxane LC elastomers system[57–59] and others, Yang also prepared IR shielding films with pitch gradient distribution for smart windows.

4.3. Laser protection films for military and medical application

At present, lasers have been widely used in applications for medical treatment, welding and cutting, as well as binding weapons, and so on. Especially in the field of binding weapons (mainly in the near infrared waveband), lasers can cause instant blinding to police officers, military and civil aviation pilots, and other persons. The laser protection products that are currently available in the market are mainly absorption and reflection types. Their common drawback is that the protection bandwidth is narrow, and they only shield a specific wavelength. For military requirements, laser protection films are expected to be wide enough, especially between 750 nm and 10000 nm. The wider the bandwidth of the infrared light shields, the better the medical and military laser protective film will be. If the shielding bandwidth of the film is wide enough, then there is no need to stack films with different shielding bands.

Due to the unique selective optical reflection characteristics, CLCs stand out as one of the most promising candi-

dates for laser protection products whose protection wavelength can be super wide. By controlling the migration direction of polymerizable monomers during the polymerization process, liquid crystal films with the coexistence of the cholesteric phase and smecitic-like short-range ordering structure were prepared. These films can shield incident light between wavelengths of 580–14000 nm.[69]

5. Conclusion and outlook

In summary, CLCs with broadband reflection characteristics have very far-reaching performance, implications, and extensive market demand in both basic scientific research and industrialization, especially for use as efficient infrared shielding films and laser protection films. As one of the most important soft matters, the research of CLCs is a long-lasting, fascinating, and fashionable topic. Based on our research experience in this field, our recommendations for specific future research directions are as follows: (i) research should focus on the novel cholesteric architectures based on the fundamental relationships between the chirality of the structure and the molecular chirality, (ii) attention should be paid to the design and synthesis of CLC materials with desired functions and films with super wide reflection bandwidth, and (iii) research should look for challenging applications, solve the urgent problems of fabrication technology, and reduce the cost of mass production.

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