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Article *in* Optical Materials Express · December 2016

DOI: 10.1364/OME.7.000016

Yong Li
Southern University of Science and Technology

Dan Luo
Southern University of Science and Technology

Research Article Vol. 7, No. 1 | 1 Jan 2017 | OPTICAL MATERIALS EXPRESS 16
Optical Materials EXPRESS

Full-color reflective display based on narrow bandwidth templated cholesteric liquid crystal film

YONG LI,[1] DAN LUO,[1,*] AND ZENG HUI PENG[2]

[1]*Department of Electrical and Electronic Engineering, Southern University of Science and Technology, Xueyuan Road 1088, Nanshan District, Shenzhen, Guangdong 518055, China*
[2]*State Key Laboratory of Applied Optics, Changchun Institute of Optics, Fine Mechanics and Physics, Chinese Academy of Sciences, Changchun 130033, China*
**luo.d@sustc.edu.cn*

Abstract: We demonstrate narrow bandwidth templated cholesteric liquid crystal films. The reflection bandwidth of these films can be dramatically narrowed with the reduced birefringence of refilled materials. Different materials from liquid crystals with anisotropic refractive index to toluene with isotropic refractive index have been refilled to polymer scaffolds with helical structures which originate from the periodic arrangement of cholesteric liquid crystals. The temperature effect on the linewidth of the films is also studied. A full-color reflective display is experimentally demonstrated based on these flexible reflective films that are refilled with small birefringence liquid crystals. The applications of these flexible films include flexible reflective display, color pixels in digital photographs, printing and colored cladding of variety of objects.

OCIS codes: (160.3710) Liquid crystals; (160.5470) Polymers; (140.3948) Microcavity devices.

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#280681 http://dx.doi.org/10.1364/OME.7.000016
Journal © 2017 Received 14 Nov 2016; revised 29 Nov 2016; accepted 29 Nov 2016; published 2 Dec 2016

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1. Introduction

Cholesteric liquid crystal (CLC) is a self-assembled material with one-dimensional photonic band-gap (PBG), which is firstly categorized by G. Friedel in 1922 [1]. The molecules of CLC are arranged as a helical structure where the director is twisted uniformly in space as a function of position along the helical axis, perpendicular to the director [2]. CLC, due to the excellent electro-optical response and properties of liquid crystal, is sensitive to external stimulus such as mechanical stress [3], optical radiation [4], electric field [5], magnetic field [6] and temperature [7]. The CLC based devices have been reported in a wide range of applications including dynamic Bragg gratings [8], electrically switchable light shutters [9], broad-band polarizers [10], microcavity lasers [11] and reflective display devices [12–15] and so on. For reflective display, the most commonly used method is utilizing electric field to tilt the pitch and dynamic change the reflection of CLCs [16]. Another approach is using interdigitated electrodes as the driven layer to induce reflection notch tuning in polymer stabilized cholesteric liquid crystal [17]. The change of pitch of CLC while bending limits its application in flexible displays due to the requirement of thickness uniformity.

Cholesteric liquid crystal film (CLCF) has attracted a lot of interests in recent years [18–25]. It is usually fabricated based on cholesteric liquid crystals and polymer, which possess the inherent reflection characteristics of CLC as well as the flexibility due to polymer network. Comparing with CLC, the CLCF was characterized by features such as flexible, full-color, single substrate, simplicity and ease of use, which is suitable for flexible reflective displays [26]. In reflective display, highly saturated colors thus narrow band-gap is preferred, which leads to wide color gamut [27, 28]. Therefore, to achieve wider color gamut, a CLCF with narrower reflection band-gap is highly desirable. However, the current report on reflection band-gap of CLCF is around 90 nm, which is far from satisfaction and optimization in practical applications [29]. Recently, the polymer-stabilized blue-phase LCs with band-gap have been reported as three-dimensional photonic crystals [30, 31].

In this letter, we demonstrate templated cholesteric liquid crystal films fabricated by cholesteric liquid crystal and polymer. The reflection band-gap of these films can be dramatically narrowed with the reduced birefringence of refilled materials. The temperature effect on band-gap of films is also studied. A full-color reflective display is experimentally demonstrated based on these flexible films that are refilled with small birefringence liquid crystals. According the experimental result, this display has good electro-optic performances. The applications of these flexible reflective films include flexible reflective display, color pixels in digital photographs, printing and colored cladding of variety of objects.

2. Experiment

The mixture used in our experiment consisted of nematic liquid crystals (NLC, E7), chiral dopant (R5011, from HCCH), reactive mesogens (RMs), and photo-initiator (Darocur1173 Sigma-Aldrich). The RM material was mixed by RM257, RM82, RM006, RM021, and RM010 (all from Shijiazhuang Sdyano Fine Chemical Co., Ltd), at 30: 15: 20: 20: 15 weight ratio [32, 33]. The center wavelength in reflection spectrum is dominated by equation: $\lambda_0 = n_{av}{*}p$, where λ_0 is the center wavelength in reflection band, n_{av} is the average refractive index of CLC, and p is the pitch length. Both of n_{av} and p are determined by the ratio of initial LC E7 and chiral dopant R5011. Therefore, the reflective color such as red, green, and blue can be produced by adjusting the concentration of chiral dopant R5011 in initial liquid crystals E7.

Four types of refilling materials, NLC LCs-160324 (n_e = 1.88, n_o = 1.50, clearing point T_c = 100°C, Changchun institute of optics, fine mechanics and physics, Chinese academy of sciences), NLC E7 (n_e = 1.74 and n_o = 1.52, clearing point T_c = 61°C, from HCCH), NLC DYX8013 (n_e = 1.56, n_o = 1.48, clearing point T_c = 102°C, Shijiazhuang Sdyano fine chemical Co., Ltd), and toluene, were used to refill into the polymer template. Table 1 lists the different compositions of samples, where A1~A3, B1~B3, and C1~C3 represent samples refilled by NLC LCs-160324, E7, and DYX8013, corresponding to large (Δn = 0.38), middle (Δn = 0.22), and small (Δn = 0.08) birefringence, respectively. D1~D3 represent samples refilled by toluene (isotropic fluid) that is an isotropic material.

Figure 1 demonstrates the fabrication process of cholesteric liquid crystal film. Firstly, the mixture consisted of NLC E7, chiral dopant R5011, and RMs were heated up to 70°C in isotropic phase, and then conducted with a magnetic stirrer at a rotation speed of 1000 rpm and rotation time of 10 minutes (Step I). The mixture was then filled into a LC cell that was assembled by two indium-tin-oxides (ITO) coated glass substrates with anti-parallel rubbing using polyimide (PI) through capillary action. Due to the alignment layer, the CLC would

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form a one-dimensional chiral structure within the cell, where the thickness of LC cell was about 40 µm. Then, the sample was exposed in an ultraviolet (UV) light (UVEC-4II, LOTS) at intensity of 15 mW/cm^2 for 15 minutes. Due to the presence of photo-initiators, the RMs would photo-polymerize through reactive acrylate end groups and form a rigid polymer network with porous (Step II), which retained the director orientation of original CLC molecules [32–35].

Table 1. The composition of samples

	NLC	R5011	RM	Darocur 1173	Refill material	Δn
A 1	69.8 wt%	4.1 wt%	25 wt%	1.1 wt%	LCs-160324	0.38
A 2	70.81 wt%	3.09 wt%	25 wt%	1.1 wt%	LCs-160324	0.38
A 3	71.36 wt%	2.54 wt%	25 wt%	1.1 wt%	LCs-160324	0.38
B 1	69.8 wt%	4.1 wt%	25 wt%	1.1 wt%	E7	0.22
B 2	70.81 wt%	3.09 wt%	25 wt%	1.1 wt%	E7	0.22
B 3	71.36 wt%	2.54 wt%	25 wt%	1.1 wt%	E7	0.22
C 1	70.89 wt%	3.01 wt%	25 wt%	1.1 wt%	DYX8013	0.08
C 2	71.38 wt%	2.52 wt%	25 wt%	1.1 wt%	DYX8013	0.08
C 3	71.88 wt%	2.02 wt%	25 wt%	1.1 wt%	DYX8013	0.08
D 1	70.97 wt%	2.93 wt%	25 wt%	1.1 wt%	Toluene	-
D 2	71.45 wt%	2.45 wt%	25 wt%	1.1 wt%	Toluene	-
D 3	71.94 wt%	1.96 wt%	25 wt%	1.1 wt%	Toluene	-

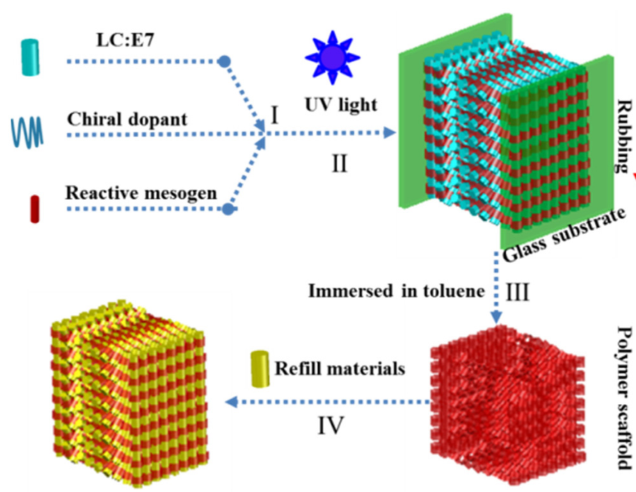

Fig. 1. Fabrication process of cholesteric liquid crystal film. (I) Mixing process, (II) sample exposed in an UV light, (III) sample immersed in toluene for one day to soak out CLC molecules and unpolymerized RM monomers, resulting polymer scaffold, and (IV) sample refilled with material.

In Step III, the polymerized sample was immersed in toluene for one day at room temperature until the original liquid crystal E7 and unpolymerized RM monomers were soaked out completely. After that, only polymer template with micro-cavity structure was left. Then different materials can be refilled into the polymer scaffold to form reflective film (Step IV), where the polymer scaffold and refilled material were represented by red hollow cylinder and yellow cylinder, respectively. The feasibility and effectiveness of the fabrication have been proved by M. E. McConney [34] and M. Mitov [35]. In our experiments, four different materials including liquid crystals LCs-160324, E7, DYX8013, and toluene were refilled into the polymer scaffold.

3. Results and discussion

3.1 Reflectance of cholesteric liquid crystal films

Figures 2(a)-2(c) show the normalized reflectance of CLCF refilled by different LCs, labeled by A1~A3 (LCs-160324), B1~B3 (E7), and C1~C3 (DYX8013). It can be seen that the photonic band-gaps of CLCF refilled by LCs-160324(A1~A3) are centered at 450.1 nm, 530.8 nm and 629.2 nm, and the band-gaps are $\Delta\lambda_{A1}$ = 112.2 nm (from 402.1 nm to 514.3 nm), $\Delta\lambda_{A2}$ = 137.8 nm (from 461.1 nm to 598.9 nm) and $\Delta\lambda_{A3}$ = 165.6 nm (from 562.7 nm to 728.3 nm), respectively. The photonic band-gaps of CLCF refilled by E7 (B1~B3) are centered at 452.7 nm, 539.5 nm and 621.8 nm, and the band-gaps are $\Delta\lambda_{B1}$ = 63.6 nm (from 420.9 nm to 484.5 nm), $\Delta\lambda_{B2}$ = 73.7 nm (from 502.6 nm to 576.3 nm) and $\Delta\lambda_{B3}$ = 90.7 nm (from 576.4 nm to 667.1 nm), respectively. In contrast, for CLCF refilled by DYX8013, the photonic band-gaps of samples C1~C3 are centered at 449.2 nm, 541.9 nm and 640.7 nm, and the band-gaps are as narrow as $\Delta\lambda_{C1}$ = 40.8 nm (from 428.8 nm to 469.6 nm), $\Delta\lambda_{C2}$ = 46.3 nm (from 518.7 nm to 565.0 nm) and $\Delta\lambda_{C3}$ = 63.2 nm (from 609.1 nm to 672.3 nm), respectively. Comparing to samples B1~B3, the band-gaps of samples C1~C3 are largely reduced by ~36%, ~38% and ~30%, respectively. Comparing to samples A1~A3, the band-gaps of samples C1~C3 are dramatically reduced by ~64%, ~66% and ~62%, respectively.

The results indicate that with the decrease of birefringence of refilled material, the corresponding band-gap of CLCF dramatically reduced. The experimental results are consistent with theory expectation, where the band-gap in reflection spectrum of CLCF is proportional to birefringence of refilled materials: $\Delta n * p = (n_e - n_o) * p$, where n_o and n_e are ordinary and extraordinary refractive index of the material refilled. For the same color with similar pitch (p), the smaller birefringence (Δn) of refilled material used, the narrower band-gap obtained.

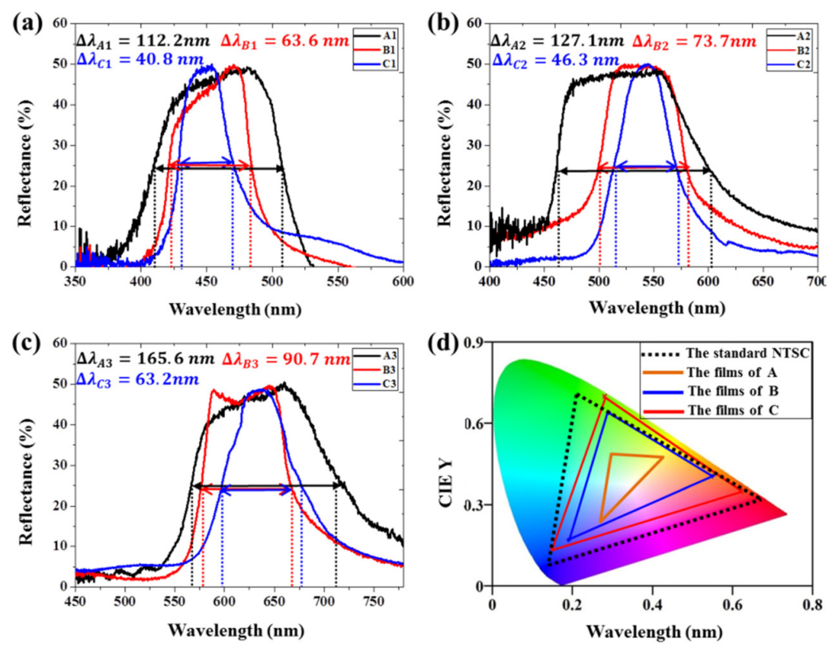

Fig. 2. Reflectance of CLCF at (a) blue, (b) green, and (c) red color. (d) Color gamut of sample refilled with different liquid crystals on the CIE 1931 chromaticity diagram.

Figure 2(d) plots the color gamut of group A~C on the CIE 1931 *xy* chromaticity diagram. In a CIE 1931 color space, the colors saturated (or colors purity) is defined as: $p=\sqrt{(x-x_n)^2+(y-y_n)^2/(x_i-x_n)^2+(y_i-y_n)^2}$, where (x, y) is the chromaticity of the sample point, (x_n, y_n) is the chromaticity of the white point and the (x_i, y_i) is the point on the perimeter whose line segment to the white point contains the chromaticity of the stimulus [36, 37]. The saturation of a color is determined by a combination of light intensity and how much it is distributed across the spectrum of different wavelengths. National television system committee (NTSC) ratio indices are used for color gamut analysis. The standard NTSC is represented by the black-dotted triangle. The color gamut is calculated by equation: $g = S_{\Delta\text{sample}} / S_{\Delta\text{NTSC}}$*100%, where the $S_{\Delta\text{sample}}$ is the triangle area of the sample and the $S_{\Delta\text{NTSC}}$ is the triangle area of the standard NTSC that is 299.7 (unit-less). It is clearly that group C (C1~C3, red triangle, 228.9) has broader color gamut than group B (B1~B3, blue triangle, 138.4) and group A(A1~A3, orange triangle, 63.5), corresponding to color gamut of 21%, 46%, and 76% for film A (LCs-160324), B (E7), and C (DYX8013), respectively. The results indicate that the increase of Δn of refilled material leads to a reduced band-gap and a broader color gamut.

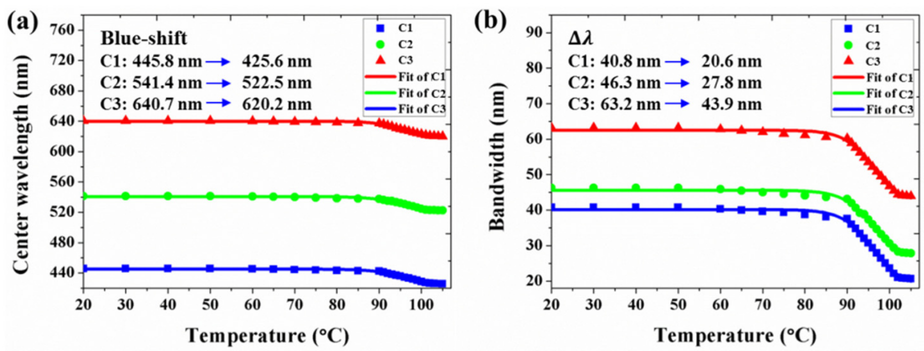

Fig. 3. (a) Temperature effect on: (a) center wavelength and (b) bandgap in reflection spectrum of samples C1~C3.

The temperature effects on center wavelength as well as band-gap in reflection spectrum of samples C1~C3 are demonstrated in Fig. 3(a)-3(b), respectively. A spectrometer with Y-type fiber (AvaSpec-ULS2048, Avantes) was used to capture the measured spectra. In Fig. 3(a), when the temperature increased from 20 °C to 88 °C, the center wavelengths of C1~C3 were all almost unchanged. When the temperature further increased above 88 °C to 107 °C, blue-shifts of the center wavelength were clearly observed for all three samples. The center wavelengths were shifted from 445.8 nm to 425.6 nm (blue-shift of ~20.2 nm), from 541.4 nm to 522.5 nm (blue-shift of ~18.9 nm), and from 640.7 nm to 620.2 nm (blue-shift of ~20.5 nm) for C1, C1, and C3, respectively, as shown in Fig. 3(a). Above 102 °C, the liquid crystal DYX8013 was in isotropic phase, there was no significant change on center wavelength with the increase of temperature. A plot of the band-gap as a function of temperature is shown in Fig. 3(b). When the temperature was below 88 °C, the band-gap had no significant change with the increase of temperature. On heating from 88 °C to 102°C, the band-gap Δλ narrowed from 40.8 nm to 20.6 nm (narrowing of ~20.2 nm), from 46.3 nm to 27.8 nm (narrowing of ~18.5 nm), and from 63.2 nm to 43.9 nm (narrowing of ~19.3 nm) for C1, C1, and C3, respectively. Over 102 °C, where the LCs were in isotropic phase, there were no change of band-gaps for C1~C3, respectively. The results obtained here indicated that the band-gap became narrowed when the refilled liquid crystal was heated up to the isotropic phase. It leads to a conclusion that besides original chiral dopant concentration and birefringence of refilled

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material, the temperature also has significant influence on the reflective color (center wavelength) as well as the band-gap of CLCF.

In addition, the band-gap of CLCF can be further narrowed if isotropic material was refilled. Herein, an isotropic liquid, toluene (n = 1.490, 633 nm, 25 °C), was refilled into the CLCF. Figures 4(a)-(c) show the normalized reflectance of CLCF refilled with toluene, labelled by samples D1~D3 at room temperature. The band-gaps of D1~D3 were 22.4 nm (from 448.3 nm to 470.7 nm), 30.6 nm (from 550.6 nm to 581.2 nm), and 36.1 nm (from 611.8 nm~647.9 nm), corresponding to blue, green and red color, respectively. The images of D1~D3 are shown in the inset Figure. CLCFs with narrower band-gap were obtained by refilling toluene, which is narrower than that of DYX8013 (Δn = 0.08) at room temperature. In our experiment, the refractive index of empty polymer film and refilled toluene at different wavelengths were measured by ellipsometer (iHR320, HORIBA), shown in Fig. 4(d). The fit curves were also demonstrated. Here, a refractive index difference of ~0.06 was presented between polymer film and toluene. The further studies on refractive index difference of polymer film and refilled material effect on resultant center wavelength and band-gap will be reported in our future work.

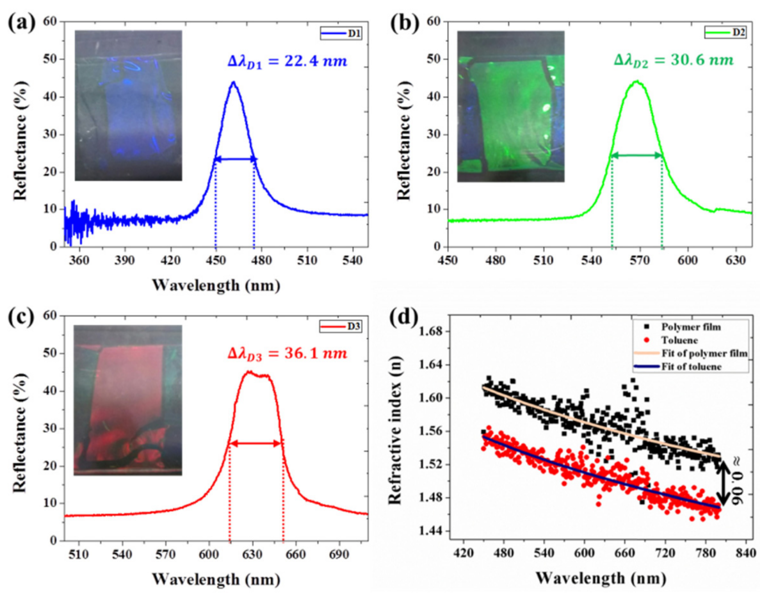

Fig. 4. Reflection spectrum of CLCF samples at (a) blue color, D1; (b) green color, D2; and (c) red color, D3. (d) The refractive index of the empty polymer film and toluene at different wavelengths. The solid curves represent the fit data.

3.2 Full-color reflective displays based on narrow bandwidth CLCF

Figure 5(a) demonstrates a schematic diagram of full-color reflective display fabricated based on CLCF with narrow band-gap (using samples C1-C3). This reflective device consists of dark layer, reflective layer, driven layer and two parallel polarizers. In the driven layer, a 90° twisted nematic (TN) LC cell (the thickness of the cell was about 8 μm) was placed between two parallel polarizers. The director of LC near to the top substrate was aligned parallel to the transmission axis of the top polarizer, while the director of LC near to the bottom polarizer was aligned perpendicular to the transmission axis of the bottom polarizer. The reflective layer was deposited on the bottom external side of the bottom polarizer. A dark layer was then adhered to the reflective layer to absorb light. In voltage-off state, the incident light firstly passed through the top polarizer and then the TN cell. A linearly polarized light, created by the top polarizer, entered the TN LC cell and had its polarization rotated by 90° as

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it arrived at the bottom polarizer. Since the direction of polarization of light was perpendicular to transmission axis of the bottom polarizer, thus without an applied filed, the light cannot pass the bottom polarizer. The cell appeared dark. When an external alternating current (AC) voltage was applied, the LC molecules of TN cell were switched from the 90° twisted state to the homeotropic texture state, leading to a black-to-transparent transition. The polarization in the light entering the cell was unaffected and therefore the light can pass the bottom polarizer. Thus the device was bright and the color reflected from the reflective layer can be observed by observers.

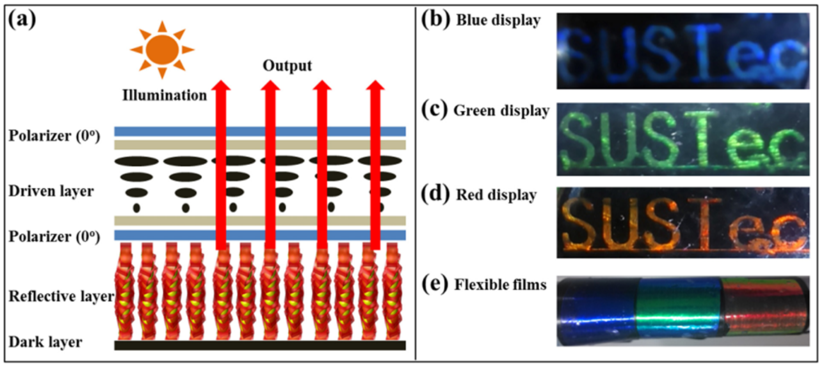

Fig. 5. (a) Schematic diagram of reflective display consisting of a dark layer, reflective layer, driven layer and parallel polarizers. 2-inch device of reflective display operated at the (b) blue color (c) green color and (d) red color. The external electric field was applied in the "SUSTec" parts. (e) The red, green and blue films were deposited on a flexible substrate; this device can be used as reflective layer for flexible display.

Then, three 2-inch full-color displays devices with logo of "SUSTec" were demonstrated. Here, the logo was on the bottom substrate of the TN cell. When an external AC voltage (1 kHz, V_{pp} = 12 V) was applied, the driven layer with logo was transparent, resulting a clearly observation of the logo of "SUSTec" in blue, green and red colors (shown in Figs. 5(b)-5(d)) Fig. 5(e) demonstrates the image of flexible displays based on CLCF with reflection color of red, green and blue on flexible substrate (black PVC tape). The application of this flexible reflective film includes flexible reflective display, E-paper, color pixels in digital photographs, printing and colored cladding of variety of objects.

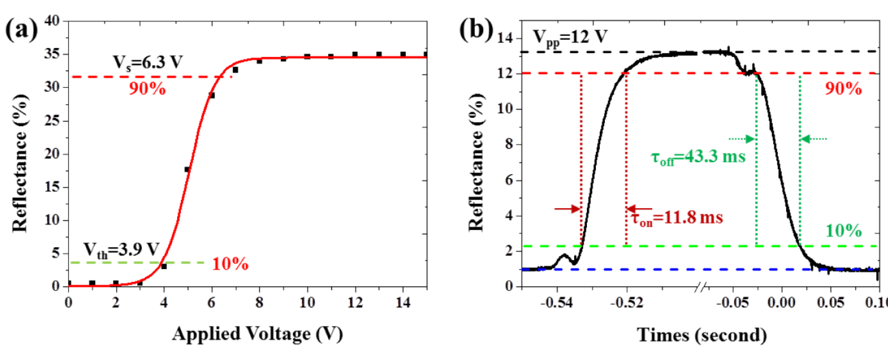

Fig. 6. (a) Variations in reflectance of the display under cross-polarizers as a function of the applied AC voltage. (b) Dynamic reflectance (response) of the display applied with an AC voltage of 12 V, where the τ_{on} and τ_{off} were 11.8 ms and 43.3 ms, respectively.

The electro-optic (EO) performances of reflective display are shown in Fig. 6. Figure 6(a) plots the variations in reflectance verse applied voltage, where the threshold (V_{th} = 6.3 V) and saturation (V_s = 6.3 V) voltages are defined as the reflectance increases to 10% of the

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minimum reflectance and reduces to 90% of the maximum reflectance, respectively. Figure 6(b) shows the response times of display with an AC voltage, where the τ_{on} and τ_{off} represent the time required to change the display reflectance from 90% to 10% and from 10% to 90% of its maximum, respectively. With applied AC voltage of V_{pp} = 12 V, τ_{on} = 11.8 ms and τ_{off} = 43.3 ms are measured.

4. Conclusion

In summary, we demonstrate templated cholesteric liquid crystal films based on cholesteric liquid crystal and polymer. Different materials from liquid crystals with anisotropic refractive index to toluene with isotropic refractive index have been refilled to polymer scaffolds with helical structure which origins from periodic arrangement of cholesteric liquid crystals. For blue, green, and red colors, with the decrease of birefringence from Δn = 0.38 to 0.08, the bandgap of film dramatically reduces from 112.2/127.1/165.6 nm to 40.8/46.3/63.2 nm, respectively. The temperature effect on bandgap of films is also studied. The bandgap can be further narrowed to 22.4/30.6/36.1 nm by refilling toluene with isotropic refractive index (Δn = 0). A full-color reflective display with direct electrode patterns of a logo "SUSTec" is experimentally demonstrated based on these flexible films that are refilled with small birefringence liquid crystals. Further studies could include an array of RGB films with high reflectivity as a full-color reflective layer. The applications of these flexible reflective films include flexible reflective display, color pixels in digital photographs, printing and colored cladding of variety of objects.

Funding

Natural National Science Foundation of China (NSFC) (61405088, 61378075); Shenzhen Science and Technology Innovation Council (JCYJ20150601155130435, JCYJ20160226192528793, JCYJ20150930160634263, and KQTD2015071710313656).