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Effect of Sectional Polymerization Process on Tunable Twist Structure Liquid Crystal Filters

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Received: 26 March 2019; Accepted: 20 May 2019; Published: 23 May 2019



Abstract: The effect of sectional polymerization process on tunable filters with cholesteric liquid crystal (CLC) and blue phase liquid crystal (BPLC) is demonstrated. The bandwidths of the polymer-stabilized cholesteric liquid crystal (PSCLC) and polymer-stabilized blue phase liquid crystal (PSBPLC) filters can be broadened by the holding treatment without distortion. The reflection bandwidth of the CLC filter can be broadened from 120 nm to 220 nm, and that of the BPLC filter can be broadened from 45 nm to 140 nm. Meanwhile, the intensity of reflection can be retained very well. The central wavelength of polymer-stabilized CLC filter can be thermally tuned from 1614 nm to 1460 nm with a stable wide bandwidth. The tunable C-band CLC filter and BPLC filter show great potential application in multi- and hyper-spectral systems and wide-band color filters.

Keywords: cholesteric liquid crystal; blue phase liquid crystal; filter; C-band

1. Introduction

Optical filters based on liquid crystal (LC) materials have been widely used in many fields, especially in optical communications and spectrometers [1–4]. Various LC materials exhibiting Bragg reflections have been tested, such as cholesteric liquid crystal (CLC) [5-8], ferroelectric liquid crystal [9,10], and blue phase liquid crystal (BPLC) [11–15]. Among them, CLC [16–19] and BPLC [13,20–22] attract great attention due to the self-assembly structures and simple fabrication process. For applications in reflective displays, optical data storage and switchable windows, filters with wide bandwidths are needed to collect mass optical information [23]. Several methods have been used to increase the bandwidth of the CLC and BPLC [23–25]. V. T. Tondiglia et al. realized an increase in bandwidth of CLC by electric field induction [26]. Michel Mitov broadened the CLC bandwidth by stacking cholesteric layers with different pitches [27]. The bandwidth of thick CLC cells composed of high helical twisting power and photoresponsive chiral dopants was broadened by UV light exposure [28]. Akifumi Ogiwara et al. and Yoshihito Hirota et al. broadened the bandwidth of CLC filter by increasing the polymer concentration [29] and by using crosslinker containing modified side-chain polysiloxane polymers [30], respectively. Jia-De Lin realized a largely-gradient-pitched polymer-stabilized blue phase (PSBP) photonic bandgap device based on the reverse diffusion of two injected BP-monomer mixtures with a low and a high chiral concentration [31]. However, some issues still do exist. For instance, the electrically induced and photoinduced bandwidth broadening results in the distortion of the Bragg reflection band because of the pitch distortions under electric field and the formation of pitch gradient under UV irradiation, respectively, and the multi-layer stacking and spatially tuning need a complicated fabrication process [23,27,28].

In this work, the effect of sectional polymerization process on tunable filters with CLC and BPLC is demonstrated. A wavelength and bandwidth tunable C-band CLC filter with a holding treatment during polymerization process is implemented. Compared with the conventional polymerization

process [23,31], the process of holding at a fixed temperature for a fixed time is added in the sectional polymerization process. Prolonging the holding time is effective at improving the homogeneous alignment of the LC material, which can improve the Bragg reflection of the filter [32–34]. Considering this effect, the sectional polymerization process with a holding treatment is used to increase the bandwidth of filter without distortion. The reflection bandwidth of CLC filter can be broadened from 120 nm to 220 nm, and meanwhile the intensity of reflection can be retained very well. The bandwidth of BPLC filter with a holding treatment can be broadened from 45 nm to 140 nm without distortion. The central wavelength of polymer-stabilized CLC filter can be thermally tuned from 1614 nm to 1460 nm with a stable wide bandwidth. The tunable C-band CLC filter and BPLC filter show great potential application in multi- and hyper-spectral systems and wide-band color filters.

2. Design Principle

To prepare a polymer-stabilized cholesteric liquid crystal (PSCLC) composite, 0.7 wt % chiral dopant (R5011, HCCH, Nanjing, Jiangsu, China), 5.6 wt % photocurable monomers [2.8 wt % RM257 (HCCH) + 2.8 wt % 12 A (HCCH)], and 0.06 wt % of photo-initiator were mixed with 93.64 wt % nematic liquid crystal host BP006 ($\Delta n = 0.158$, $\Delta \varepsilon = 34.2$ at $\lambda = 633$ nm and T =293 K, HCCH). From 345 K to 298 K, the mixture showed the following phase sequence: Iso, 340 K N*.

The BPLC mixture was composed of 88.9 wt % nematic liquid crystal host HBG ($\Delta n = 0.141$, $\Delta \epsilon = 34.3$ at $\lambda = 589$ nm and T = 298 K, HCCH), 3 wt % chiral dopant (R5011, HCCH), 8 wt % photocurable monomers [4 wt % RM257 (HCCH) + 4 wt % 12 A (HCCH)], and 0.1 wt % of photo-initiator. From 340 K to 330 K, the mixture showed the following phase sequence: Iso 338 K BP, 334.5 K N*.

The precursor mixtures were filled into empty cells with a thickness of 10 µm comprised of two ITO glass substrates with antiparallel polyimide alignment layers at 368 K. The polyimide alignment layers had the effect of enhancing the intensity of reflection because of its surface pinning effect [33–35]. The mixtures were cooled from isotropic phase to chiral nematic phase using a temperature controller (HCS302, Instec Co., Boulder, Colorado, USA). All the Bragg reflection spectra from the cholesteric liquid crystal and blue phase liquid crystal were collected by the measurement system shown in Figure 1. A tungsten bromine lamp was used as the unpolarized light source covering NIR region and visible region, and the reflection spectra were collected by the detector attached to the data acquisition system (DCS300PA, Zolix, Beijing, China). The transmittance was calculated as the ratio of the measured light intensity in measurement system with the LC cell to that with an empty cell.



Figure 1. Schematic diagram of the transmittance measurement system.

3. Experiments

3.1. Bandwidth Tunable PSCLC Filter

The PSCLC filters under different polymerization conditions were obtained and the transmission spectra at 323 K are shown in Figure 2. During the cooling process at a rate of 7 K/min, the mixture showed the planar state of chiral nematic phase at 340 K. In order to make a comparison with the polymerization with a holding treatment, we measured the transmission spectra of polymerization without holding treatment as shown in Figure 2 (dashed lines). These dashed lines were obtained by

cooling the cell from 345 K to the given temperature and curing the cell at the given temperature directly. The central wavelengths of dashed lines were 1592 nm, 1554 nm, 1502 nm and 1446 nm, with cured temperatures of 338 K, 333 K, 328 K, and 323 K, respectively. The results of polymerization with holding time of 15 minutes, 30 minutes and 45 minutes at different holding temperatures are shown in Figure 2 (solid lines). All the polymerization of the cells were implemented at 323 K. The temperature and time symbols in the legend represent the fixed temperature and the fixed time, respectively. For example, the black curve in Figure 2a represents the polymerization process during which the cell was cooled from 345 K to 338 K at a rate of 7 K/min, placed at 338 K for 15 minutes, cooled to 323 K at a rate of 7 K/min, and finally polymerized at 323 K. The bandwidth referred to the full width at half maximum (FWHM) as well as the 3 dB bandwidth [36]. Among the curves with the polymerization process at different holding time, the curves with the holding time of 45 minutes had the widest bandwidth due to the stabilization effect of the longtime holding treatment [32–34]. The curves with a holding treatment at 338 K showed wider bandwidth than those at other temperatures because of the larger temperature difference between the holding temperature and the cured temperature. The bandwidth related to the temperature difference and the holding time, and a large temperature difference and a long holding time contributed to a wide bandwidth. Compared with the filter of polymerization at 323 K without holding treatment, the bandwidth of the filter with a holding treatment for 45 minutes at 338 K and polymerization at 323 K could be broadened to 183% from 120 nm to 220 nm. Meanwhile, the intensity of reflection could be retained because of the orientation effect of the holding treatment, which contributed to the maintenance of the orientational order of the liquid crystal molecules and the formation of uniform planar orientation [32,37]. The results showed that the holding treatment could contribute to the broadening of the bandwidth and the retaining of the intensity of reflection of the PSCLC filter. The location of the central wavelengths after polymerization could be shifted in accordance with need because of the stabilization effect of the longtime holding treatment. The stabilization effect referred to the effect of the holding treatment on the improvement of the homogeneous alignment of the LC material and the strong inertia caused by the longtime holding treatment [32–34]. Compared with other broadening methods, the method using a holding treatment could realize undistorted bandwidth broadening and simplify the fabrication process [26,28,29].

The mechanism of the holding treatment is illustrated in Figure 3, showing the CLC pitch variation in the sectional polymerization process. During the holding process, the CLC material had a stable and homogenous pitch distribution. When the temperature was cooled rapidly, the pitch at the bottom region close to the temperature controller was shortened because of the change of helical twisting power [18], while the pitch at the top region still had the former pitch because of the long distance from the temperature controller. After UV irradiation, the CLC textures were stabilized by the polymer chain and the pitch distribution was maintained [29]. Therefore, the bandwidth of the filter with a holding treatment was broadened.

The bandwidth of the filter with a holding treatment was related to the central wavelength of the Bragg reflection at the holding temperature and that at the cured temperature. The ideal bandwidth of the filter with a holding treatment was expected to cover both the reflection band of polymerization without holding treatment at the holding temperature and at the cured temperature. However, if the temperature difference between the holding temperature and the cured temperature was small, the reflection band at the holding temperature and that at the cured temperature overlapped partially, resulting in a narrower bandwidth than the sum of the two bandwidths without holding treatment. If the temperature difference was large, the temperature of the material at the top would decrease during the cooling process and the effect of the holding treatment would be weakened. Therefore, an appropriate temperature difference could be found to obtain a maximum bandwidth. The bandwidth of the filter with a holding treatment was also related to the holding time, which could improve the homogeneous alignment of the LC material and improve the Bragg reflection of the filter at the holding temperature if prolonged [32–34]. The polymer network could be weall maintained

because of the strong inertia caused by the longtime holding treatment. Therefore, a long holding time could contribute to the broadening of the bandwidth.



Figure 2. Transmission spectra of polymer-stabilized cholesteric liquid crystal (PSCLC) filters under different polymerization conditions. Results of polymerization at 323 K with holding treatment at different temperatures for (**a**) 15 minutes, (**b**) 30 minutes, and (**c**) 45 minutes.



Figure 3. The mechanism illustration of the holding treatment. The cholesteric liquid crystal (CLC) pitch distribution in cell (**a**) at the holding temperature, and (**b**) at the cured temperature in the sectional polymerization process.

The transmission spectra of the PSCLC filter with standing treatment for 30 minutes at 338 K and polymerization at different temperatures ranging from 333 K to 293 K at a step of 10 K are shown in Figure 4. The spectra were measured at the polymerization temperature. Among these curves, the bandwidth of the filter with polymerization at 323 K reached the maximum value because of the appropriate temperature difference between the holding temperature and the cured temperature. For comparison, the transmission spectrum of polymerization at 338 K without treatment is also shown in Figure 4 (dashed line). Compared with the bandwidth at 338 K without holding treatment, the bandwidth with a holding treatment polymerized at 323 K broadened from 138 nm to 189 nm. Similar to the conclusion from Figure 2, the results in Figure 4 also indicated that the holding treatment could contribute to the broadening of the bandwidth and the retaining of the intensity of reflection of the PSCLC filter.



Figure 4. Transmission spectra of the PSCLC filter with a holding treatment for 30 minutes at 338 K and polymerization at different temperatures.

In order to analyze the thermal stability of the PSCLC filter with a holding treatment, the transmission spectra of the filter with a holding treatment for 30 minutes at 338 K and polymerization at 313 K at different temperatures from 348 K to 303 K at a step of 5 K were measured. As shown in Figure 5, the central wavelength blue-shifted from 1614 nm to 1460 nm with the decrease in temperature with an almost stable bandwidth and intensity of reflection because of the polymerization with the polymer network keeping the orientational order of the liquid crystal molecules, which improved the homogeneous alignment of the LC material [29,32].

In order to analyze the influence of the cell thickness on the polymerization with a holding treatment, we measured the transmission spectra of PSCLC filters with different cell gaps of 8 μ m, 10 μ m, and 20 μ m at 323 K. As shown in Figure 6, the PSCLC filters with different gaps polymerized at the same temperature without holding treatment had the similar bandwidths. The bandwidths of thicker cells with a holding treatment were slightly wider than those of thinner cells with a holding treatment because of the larger pitch gradient formed by the thicker cell gap. The results showed that the bandwidth of 20 μ m cell could be broadened from 120 nm to 232 nm by holding treatment.



Figure 5. Transmission spectra of the PSCLC filter with a holding treatment in thermal modulation.



Figure 6. Transmission spectra of PSCLC filters with different cell gaps of (**a**) 8 μ m, (**b**) 10 μ m, and (**c**) 20 μ m. The cells with a holding treatment were held at different temperatures for 45 min and polymerized at 323 K.

3.2. Bandwidth Tunable PSBPLC Filter

The effect of the sectional polymerization process on the characteristics of polymer-stabilized blue phase liquid crystal (PSBPLC) filter was also investigated. In Figure 7, the dashed lines are the transmission spectra of the PSBPLC filter cured at 337.5 K and 336 K at a cooling rate of 1 K/min, and the bandwidth of the single Bragg reflection band at 336 K was 45 nm. During the polymerization process with a holding treatment, the cooling rate was 1 K/min. When the cell was cooled from 340 K to 337.5 K, then placed at 337.5 K for 5 minutes, and finally polymerized at 336 K, two reflection bands could be obtained due to the stabilization effect of the holding treatment. The temperature gradient of blue phase liquid crystal was so large that the LC orientations at different holding temperatures could be stabilized within a short cooling time because of inertia. Therefore, two reflection bands could be obtained. When the cell was cooled from 340 K to 337.5 K, placed at 337.5 K for 5 minutes, then cooled to 337 K and placed at 337 K for 3 minutes, and finally polymerized at 336 K, three reflection bands could be obtained. The total bandwidth of the filter with a two-stage holding treatment could reach up to 140 nm. The reflection bands could be easily distinguished because of the narrow bandwidth and the large temperature gradient of PSBPLC filter. The difference in temperature gradients, CLC of 9.7 nm/K and BPLC of 80 nm/K, contributed to the difference between the transmission spectra of the PSBPLC filter with a holding treatment and those of the PSCLC filter.



Figure 7. Transmission spectra of the polymer-stabilized blue phase liquid crystal (PSBPLC) filter with sectional polymerization process.

4. Conclusions

The effect of sectional polymerization process on tunable filters with CLC and BPLC is demonstrated. The bandwidths of the PSCLC and PSBPLC filters can be broadened by the holding treatment without distortion. The reflection bandwidth of the CLC filter can be broadened from 120 nm to 220 nm, and that of the BPLC filter can be broadened from 45 nm to 140 nm. Meanwhile, the intensity of reflection can be retained very well. The central wavelength of polymer-stabilized CLC filter can be thermally tuned from 1614 nm to 1460 nm with a stable wide bandwidth. Three reflection bands can be obtained in PSBPLC filter with a holding treatment. The tunable C-band CLC filter and BPLC filter show great potential application in multi- and hyper-spectral systems and wide-band color filters.

Author Contributions: Writing-original draft, C.S; Writing-review & editing, J.L.

Funding: This research was funded by National Natural Science Foundation of China, grant numbers 61727808 and 61775135.

Conflicts of Interest: The authors declare no conflict of interest.

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