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New micro-structure designs of a wide band reflective polarizer with a pitch gradient

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New microstructure designs for fabricating a wide band reflective polarizer (WBRP) from cholesteric liquid crystals are reported. The pitch difference in the WBRP is formed by orienting a single layer consisting of different glassy siloxane cyclic side chain oligomer powders on a heater. The molecular arrangement obtained is frozen by quenching. The experimental results show that various micro-areas exhibiting different reflection wavelengths and pitch gradients are formed in the WBRP. A WBRP exhibiting the reflection properties of the original cholesteric liquid crystals is fabricated by a novel experimental process and the experimental results are in accordance with the microstructure designs of the WBRP.

1. Introduction

Bragg reflection or iridescence is the most characteristic property of cholesteric liquid crystals with a periodic helical molecular structure. The Bragg reflection wavelength of the incident light is related to the cholesteric pitch p and the mean refraction index n by [1, 2]:

$$\lambda = n \times p. \tag{1}$$

Thus the reflection bandwidth $\Delta\lambda$ can be adjusted mainly by the birefringence Δn and the pitch gradient Δp. Because Δn is typically limited to 0.3, the reflection bandwidth $\Delta\lambda$ will be less than 100 nm in the visible spectrum, when it is determined only by Δn. A cholesteric liquid crystal with $\Delta\lambda$ less than 100 nm is insufficient, however, for some applications, such as full colour or black and white displays.

A favourable pitch gradient is effective in obtaining a cholesteric material whose spectral characteristics evolve from a selective to a broad band filter. The pitch gradient of a cholesteric liquid crystal has often been formed by photo-induced molecular diffusion [3, 4]. A UV-intensity gradient over the film thickness, and reactivity difference between the helix-winding chiral monomer and the helix-unwinding nematic one, led to molecular diffusion, with the formation of a pitch gradient. A pitch gradient could also be induced through the diffusion of small molecules, thermal diffusion between two adjacent cholesteric liquid crystal oligomer films with different pitches, or by a temperature gradient [5–8].

In our studies, new microstructures of a wide band reflective polarizer (WBRP) with a pitch gradient have been designed, and another novel experimental route to broaden Δp devised. The WBRP is fabricated without the usual polymerization reaction, photo-induced phase separation, temperature gradient or multilayer structure which are used to form pitch gradients. The mechanisms of broadening are studied in detail and experiments clarifying the broadening phenomenon are described.

2. Experimental

2.1. *Materials*

Glassy siloxane cyclic side-chain oligomers (GSCSOs) with achiral mesogens and chiral mesogens attached via spacers were chosen [9]. The chemical structures of the oligomers are shown in figure 1. The pitch length of a cholesteric structure is tuned by the molar percentage of chiral mesogens in the oligomer molecule. These materials can be easily quenched at room temperature and the cholesteric structural order with its colour properties is then restored within a solid film.

2.2. *Treatment of the substrates for homogeneous alignment of LC molecules*

A 3.0 wt% polyvinyl alcohol (PVA) aqueous solution was stirred until the PVA had been dissolved completely (for about 6 h). The solution was then deposited on the inner surfaces of cleaned substrates by spin coating

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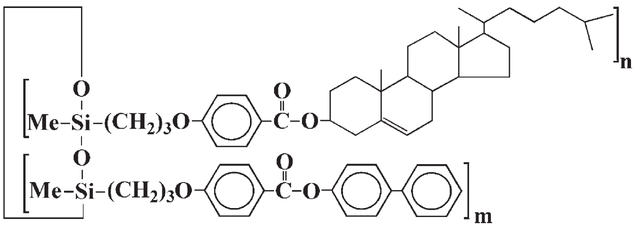

Figure 1. Chemical structures of the GSCSOs.

(4 000 rpm) at room temperature. The deposited film was dried at 353.2 K for about 30 min, and subsequently rubbed with a textile cloth under a pressure of 2.0 g cm^{-2} along one direction.

2.3. *Preparation of the WBRP*

A mixture consisting of different GSCSO powders was coated on a substrate having a PVA alignment layer; a single layer of the mixture was obtained. Another substrate was placed on top, and the mixture, with the upper substrate, was rolled at a constant temperature between the highest glassy transition temperature and the lowest isotropic transition temperature of the glassy oligomers. The received molecular alignment was restored by quenching. A WBRP was then fabricated by the shortened process.

2.4. *Measurements*

The spectral characteristics of the films were obtained by an LCT-5016C LCD Parameters Tester at room temperature. The reflection wavelength λ and reflection bandwidth $\Delta\lambda$ were measured from the spectrum by considering the wavelength for the minimum of transmitted light inside the peak, and the peak bandwidth at half-height, respectively.

The morphologies of freeze-fractured surfaces of samples perpendicular to the plates were observed by scanning electron microscopy (SEM), after coating with carbon in a sputtering coater.

3. Results and discussion

3.1. *A selective reflective film from the GSCSO*

When a GSCSO is heated to a temperature above its glassy transition, the highly flexible siloxane backbone, coupled with flexible spacers separating the mesogens from the backbone, can supply free room for the motion of the chain segments. The energy of disorientation of the mesogens is therefore low, and the mesogens attached to the cylic siloxane can easily be aligned parallel to the substrates at this time. Due to the 'freezing' of the chain segments of glassy materials below T_g, the molecular arrangement obtained can be stored by quenching [10–13]. A reflective film with a constant reflection wavelength can then be fabricated after the oriented planar texture of the siloxane cyclic side chain liquid crystal oligomer is restored by quenching.

The pitch length of a cholesteric liquid crystal is related to the mole ratio of chiral component (X_c) and helical twisting power (HTP) by:

$$p = 1/(HTP \times X_c) \qquad (2)$$

Thus the selective reflection wavelength of a siloxane cyclic side chain liquid crystal oligomer with oriented planar texture can be adjusted by grafting different concentrations of chiral component into the cyclic siloxane backbone. GSCSOs with different mole ratios of chiral and achiral mesogens grafted to the cyclic siloxane backbone were employed as the starting materials in our studies.

3.2. *Designs of microstructures to broaden* $\Delta\lambda$

Various liquid crystal micro-areas exhibiting different reflection wavelengths and pitch gradients in a single layer are designed to broaden $\Delta\lambda$. To turn the designs into reality, a WBRP is designed to be fabricated from a single layer consisting of different GSCSO powders with different pitch lengths (see the schematic model in the ellipse of figure 2). Different liquid crystal micro-areas with different reflection wavelengths will be formed when the single layer is oriented on a heater; and pitch

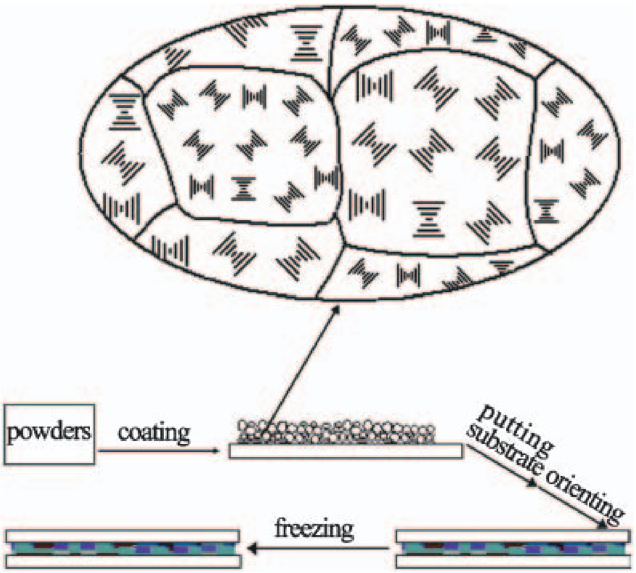

Figure 2. Schematic representation of the sample preparation method.

gradients among the liquid crystal micro-areas will also be formed because of molecular diffusion. A WBRP exhibiting the reflection properties of the different liquid crystal micro-areas can then be fabricated after the molecular arrangement is frozen by quenching. A schematic representation of the preparation of a WBRP with these micro-structures is shown in figure 2.

3.3. *Experiments and mechanism of broadening*

The original powders consisted of GSCSOs whose mole ratios of chiral and achiral mesogens grafted to the cyclic siloxane backbone were 2/1 and 3/2. A mixture of these powders was coated onto a glass substrate and another substrate placed on top. The mixture was oriented at 75°C on a heater and the molecular arrangement obtained was frozen by quenching. A WBRP was then obtained.

The transmission spectrum of the WBRP is shown as curve 3 in figure 3; The transmission spectra of the two original cholesteric liquid crystals are shown as curves 1 and 2. It can be seen that the WBRP can reflect light throughout the reflection wavelengths of the two original liquid crystals. We consider that during rolling between the highest glassy and the lowest isotropic transition temperatures of the GSCSOs, the different powders form different liquid crystal micro-areas.

When the temperature is too low or the orienting time too short, the molecules cannot diffuse quickly among the liquid crystal micro-areas, and different pitches are still retained in the various liquid crystal micro-areas. Thus when the planar texture (most axes of the cholesteric helix perpendicular to the substrates) is formed after orienting by rolling, different liquid crystal micro-areas reflect light with different reflection wavelengths, leading to a variety of iridescent colours (see figure 2). A solid WBRP reflecting light throughout the reflection wavelengths of the two original liquid crystals will be obtained after this oriented planar texture is frozen by quenching.

The SEM of a freeze-fractured surface (cross-section perpendicular to the plates) of the fabricated WBRP is shown in figure 4. It is clear that there are different pitches in different micro-areas, as seen, for example, in micro-areas I and II. On the other hand, pitch gradients are also formed among adjacent micro-areas during orientation. As shown in figure 5, transition liquid crystal layers have different directions, which also broadens the reflection bandwidth. The angles between the transition layers III in figure 5 (*a*) and the substrates are about 0°, while the angles between the transition layers III in figure 5 (*b*) and the plates are about 45°. These experimental results comply with the micro-structure designs of the WBRP mentioned above.

3.4. *Orienting temperature dependence of the reflection bandwidth*

Figure 6 represents the orienting temperature dependence of the reflection bandwidth of WBRPs fabricated from the same mixture of glassy oligomer powders. This indicates that the reflection bandwidth decreases as the orienting temperature rises. When the orienting temperature is low (65°C), the mesogens are slow to rearrange because of the strong molecular friction force and steric hindrance of large mesogens, especially the cholesterol mesogens. The planar texture, when the majority of the cholesteric helix axes are perpendicular to the substrates is therefore also slow to form. It can be seen from figure 7 that a uniform oriented layer structure cannot be formed when the mixed powders are rolled at 65°C. As the orienting temperature

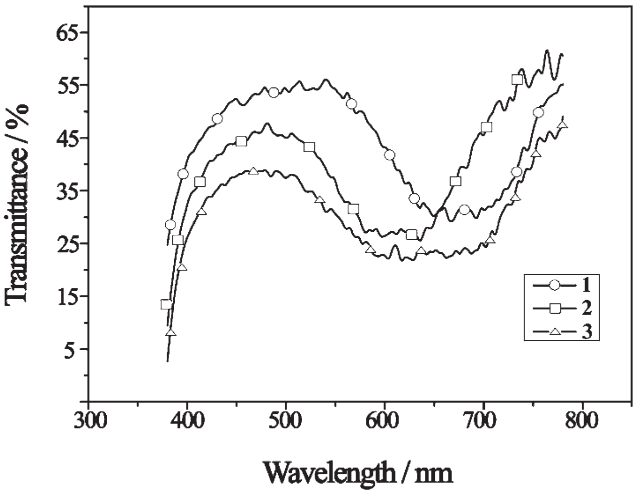

Figure 3. Transmission spectra of the reflection films: 1, one original GSCSO rolled at 75°C; 2, another original GSCSO rolled at 75°C; 3, the WBRP fabricated from a mixture of 1 and 2 rolled at 75°C.

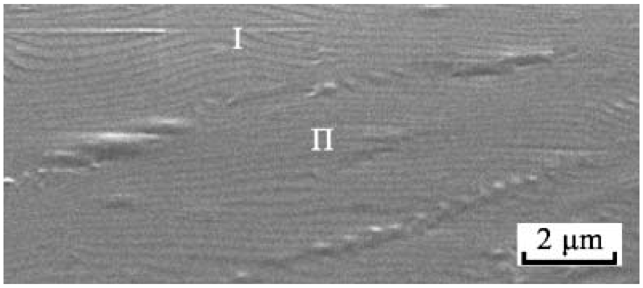

Figure 4. Scanning electron micrograph of the freeze-fractured surface of the WBRP.

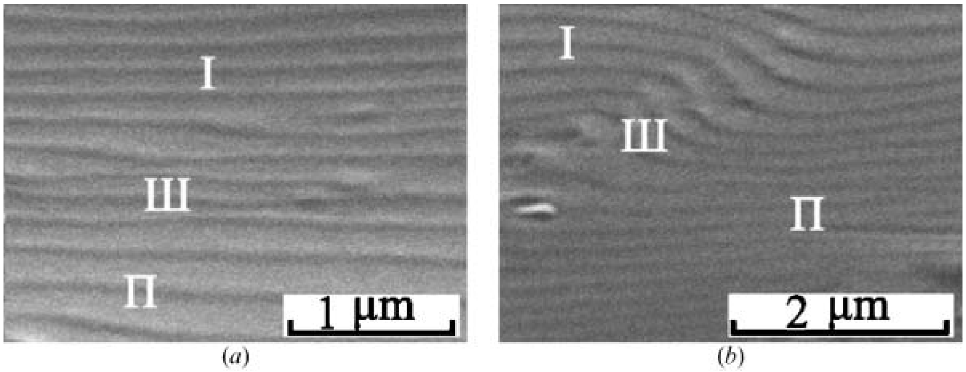

Figure 5. Scanning electron micrographs of freeze-fractured surfaces of the WBRP: (*a*) with angles of about 0° between the transition layers III and the substrates; (*b*) with angles of about 45° between the transition layers III and the plates.

increases, chain segments can move more flexibly because of their high energy and low viscosity. The mesogens in different liquid crystal micro-areas can then be oriented easily by rolling, and planar cholesteric texture can be formed in the different liquid crystal micro-areas. As shown in figure 7, oriented layer structure was formed when the mixture was oriented at 75°C, and different pitches were formed in the different liquid crystal micro-areas.

On the other hand, with further increase in temperature, more free space for the motion of chain segments will be induced because of the higher energy and lower viscosity. The rate of molecular diffusion then increases, reducing the pitch difference. As shown in figure 7, when the mixture was rolled at 90°C, the resulting orientation was more uniform and the pitch difference almost disappeared; an almost uniform pitch was formed. It can also be seen from figure 6 that the reflection bandwidth decreases rapidly when the mixture is rolled at 90°C.

4. Conclusions

In this study, a WBRP was fabricated from a single layer consisting of different GSCSO powders. Various pitches in the different liquid crystal micro-areas, and pitch gradients among adjacent micro-areas, were formed after the mixture was oriented on a heater. The WBRP obtained reflected light throughout the reflection wavelengths of the original cholesteric liquid crystals. The reflection bandwidth of the WBRP decreased with increasing orienting temperature.

Due to surface anchoring of the various interfaces among the liquid crystal micro-areas, the molecular orientation also changed; transition liquid crystal layers

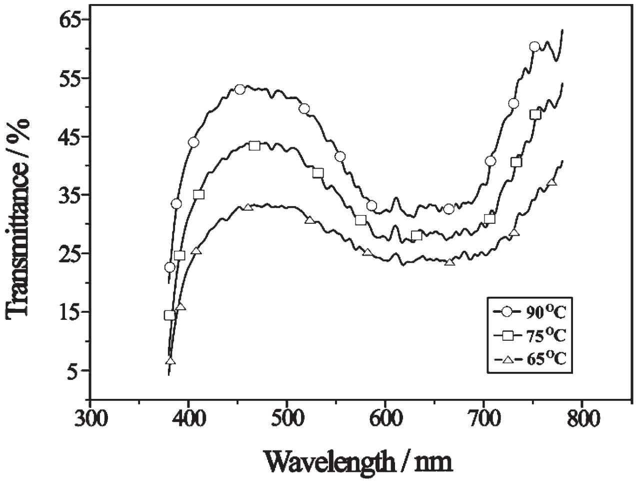

Figure 6. Transmission spectra of reflection films oriented at different temperatures: 1, 65°C; 2, 75°C; 3, 90°C.

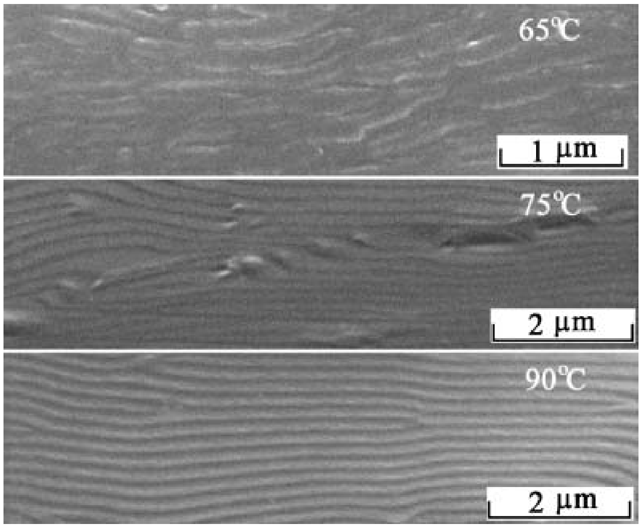

Figure 7. Scanning electron micrographs of freeze-fractured surfaces of WBRPs rolled at different temperatures.

even bent near the interfaces, which could be reduced theoretically, by a higher orienting temperature. However high temperatures also decreased the bandwidth because of faster molecular diffusion. In order to delay molecular diffusion among the liquid crystal micro-areas, the GSCSO powders will be coated with a cladding polymer having a higher glassy temperature; experimental work will be reported later.

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