

NOVEL POLYMER WIDE VIEWING ANGLE COMPENSATION FILMS FOR LIQUID CRYSTAL DISPLAYS (LCDs)*

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Abstract We report on generating uniaxial negative birefringent compensation films, made of specifically designed polyimides. These polymers were synthesized via a polycondensation of dianhydride [such as 2,2'-bis(3,4-dicarboxyphenyl) hexafluoropropane dianhydride] and 2,2'-bis(trifluoromethyl)-4,4'-diaminobiphenyl. The uniaxial negative birefringent ($n_x = n_y > n_z$) polyimide substrates are achieved using a solution-casting method in conventional solvents, which exhibit the desirable optical phase retardation [$(n_x - n_z) \times d$] values from 50 to 400 nm varying with the film thickness. In these polyimide films, the long chain rigid molecules adopt intrinsic planar orientation. In detail, the majority of phenylene-imide rings and phenylenes preferentially adopt nearly planar conformations parallel to the film substrate. In addition, these films also possess high transparency (or transmittance) and little color shift. The unique color dispersion curve indicates that this type of materials is very suitable for the applications in LCDs due to an excellent mimic for the retardation color dispersion curve with respect to LC molecules. Significantly low in-plane retardation (< 1 nm) allows this new technology based film to achieve sufficiently high contrast ratio while highly negative retardation dramatically suppresses the gray scale inversion to improve the viewing angle performance in a variety of new mode LCDs.

Keywords Compensation films, Liquid crystal displays, Polyimides

INTRODUCTION

Flat panel active matrix thin-film-transistor (TFT) liquid crystal displays (LCDs) have been widely accepted for applications in notebook personal computers and desktop computers, wireless phones/images, and digital TVs with light weight and low power consumption since their discovery by Schadt and Helfrich^[1]. Optical polymer films play a critical role in improving the contrast ratio, gray scale, viewing angle and brightness in displays. New optical compensation films were used to improve the angular dependence of contrast and gray scale inversion, in which the optical retardation of the discotic molecular construction in hybrid molecular orientation mimicked that of the liquid crystals (LCs)^[2]. In addition, other wide viewing LCD monitor technologies have been developed including in-plane switching (IPS) mode, multi-domain vertically aligned (MVA) mode, and optically compensated bent (OCB) mode^[3–6]. However, the LC response time in IPS, which does not have negative optical compensation films, still needs to be improved. In the other two modes, high quality uniaxial negative optical compensation films are desired to improve the optical performance of the black state and suppress the light leakage. Besides, a multi-domain photoalignment surface in each pixel upon exposure to polarized UV can also be achieved in displays, which will provide the displays with improved wide viewing angles and less color-shift^[5, 6]. A great challenging question is whether we can develop a new uniaxial negative birefringent film with low in-plane retardation and high negative out-of-plane retardation characteristics. One of the most effective ideas is to

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adjust the structure and chemical properties of aromatic polyimides.

Uniaxial negative optical birefringent polymer compensation films, so called C-plates, have been widely used in normally white twisted nematic (TN) LCD with improved wide viewing angle and high contrast ratio^[7, 8]. As compared to the optical ellipse of the positive uniaxial birefringence ($n_x = n_y < n_z$, $\Delta n = n_z - n_x = n_z - n_y > 0$) in uniaxial stretched films like a polycarbonate film (A-plate) (Fig. 1a). The optical refractive indices along the n_x , n_y and n_z follow the relationships of $n_x = n_y > n_z$, $\Delta n = n_x - n_z = n_y - n_z < 0$ in uniaxial negative compensation films as shown in Fig. 1b. The advantage of this polymer film technology focuses on solution casting, high negative out-of-plane birefringence and low in-plane as well as high temperature stability, which can dramatically suppress film stripe defects and non-uniformity in biaxially stretching processing as well as processing difficulties. The uniaxial negative optical phase retardation in films will be used to compensate that of the positive LCs in normally white TN displays and limit the off-axis light leakage over the entire visible range from the LC displays in the address state^[9].

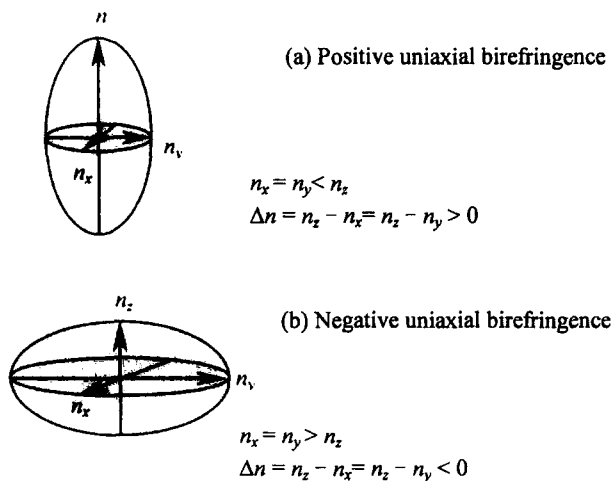


Fig. 1 Linear optical refractive indices for optical ellipses with (a) positive uniaxial birefringence and (b) negative uniaxial birefringence

The operation principle of liquid crystal displays with a normally white mode is illustrated in Fig. 2^[10, 11]. In the off-state, the incident light passes through the first polarizer and is twisted 90° along the LC molecular direction, in which the LC molecules are aligned along the rubbing direction of the alignment surface on ITO glass substrates in a 90° twist configuration. When the polarized light passes through the second crossed polarizer, a bright (white) state is formed. After applying the voltage to the LC cell, the LC molecules will be oriented near a homeotropic configuration along the external electric field. The polarized light will be blocked by the second polarizer to form a black state. In addition, the tilt molecular orientation on the surface will cause the light to deviate from the homeotropic arrangement, which may further induce the color shift (like a blue shift) in the black state of LCDs.

The optical phase retardation from the positive LC molecules is related to the LC cell gap (d), the LC birefringence (Δn), the off-axis incident angle (θ) and the light wavelength (λ) as well as the consideration of the surface tilt of the LC molecules, as shown in Fig. 3. The related optical phase retardation (δ) in this configuration is calculated from the equation of $\delta = 2\pi\Delta n d/\lambda$, in which $\Delta n = (n_e - n_o)\sin^2\theta$, and the off-angle light leakage in the addressed state further leads to low viewing, low contrast ratio and gray scale inversion as well as color shift in LCDs. Due to the small amount of tilt LC molecules on the surface, the depolarization of surface LC molecules should be considered in some LCD modes.

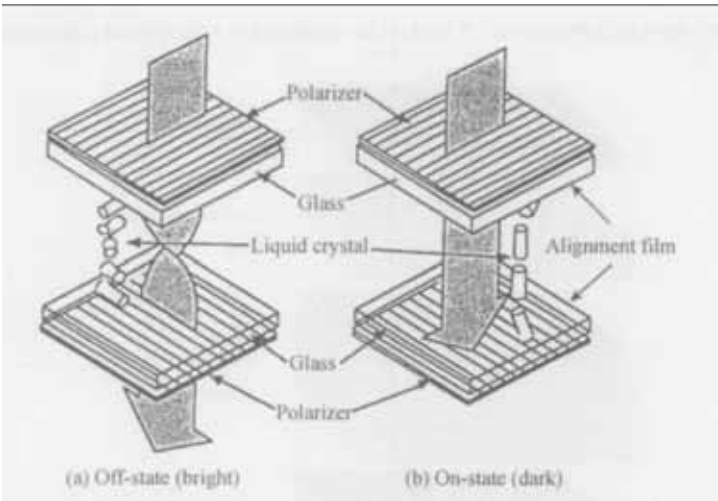


Fig. 2 Schematic drawing of LC operation principles in a normally white mode, without the external field (a): off-state, and with the external field (b): on state

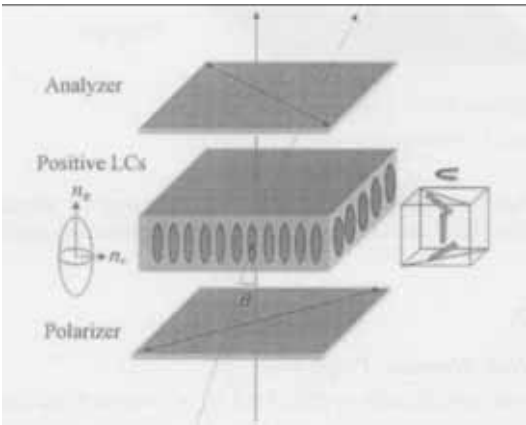


Fig. 3 Schematic representation of the optical phase retardation along the off-angle direction after applying the external electric field

Basically, we can introduce the negative optical ellipse to compensate the positive LC ellipse so that we can achieve the net zero optical phase retardation (shift) of $R = R_{LC} + R_{OC} \approx 0$, as illustrated in Fig. 4. Uniaxial negative birefringent compensation films are extremely important to improve the viewing angle and gray scale while the high contrast ratio remains. The optical phase retardation can be reduced when a negative optical compensation film is integrated. The overall optical properties will be optimized using a computer simulation. This new film technology allows us to achieve a much better optical performance in normally white TFT displays than that in uncompensated TFT LCDs. As we know, an improved efficiency is also dependent on a reduced number of processing steps in manufacturing, which requires the development of new optical polymers and new constructive ideas in the film fabrication. Therefore, it is necessary for us to further explore negative optical birefringent films along with related molecular mechanisms. This will improve our understanding of the molecular design and assembly of negative birefringent films as well as other optical properties.

In this work, we focus on a newly designed polyimide as negative birefringent optical film substrates. The insertion of bulky groups into aromatic backbones generates an organo-soluble aromatic polyimide, along with desired optical properties. Using combined characterization methods, we report our experimental results on the new polyimide material possessing intrinsic negative birefringence in the bulk with an in-plane orientation on the

substrates, and provide a new understanding of molecular orientation and optical characteristics.

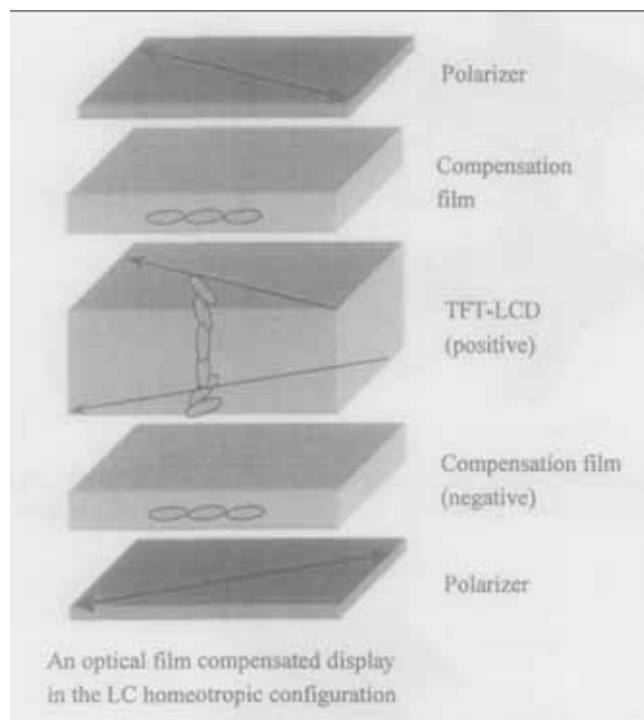


Fig. 4 Schematic representation of the insertion of negative optical compensation films into LC displays between the polarizers and the LC cell

RESULTS AND DISCUSSION

Highly Transparent and Colorless Aromatic Polyimide

This polymer (6FDA-PFMB) was specifically synthesized by a one-step imidization via a polycondensation of 2,2'-bis(3,4-dicarboxyphenyl) hexafluoropropane dianhydride (6FDA) and 2,2'-bis(trifluoromethyl)-4,4'-diaminobiphenyl (PFMB)^[12], as seen in Fig. 5. An equal molar ratio of the dianhydride and diamine was used in the reaction. The corresponding polymerization was conducted in a three-neck flask refluxing in a solution of *m*-cresol under a dry N₂ atmosphere with a tertiary amine catalyst (isoquinonline) at elevated temperatures for 24 h. The imidization process was detected in Fourier transmission (FT)-IR and proton nuclear magnetic resonance spectroscopy. The intrinsic viscosity of this polymer was 0.76 dL/g measured in chloroform at 30°C. The glass transition temperature was 320°C with the thermal expansion coefficient of 27 ppm/K as measured using TA Instruments TMA 2940 at a heating rate of 5 K/min under a nitrogen atmosphere. Optical films with thickness of 1–20 μm were obtained by casting solutions at a concentration of 5 wt%–15 wt% in cyclopentanone after filtration through a PTFE filter of 0.45 μm, which were used recording for in-plane and out-of-plane birefringence and transmission spectra. A highly transparent and colorless transmission spectrum is demonstrated between 400 nm and 800 nm in Fig. 5, without the obvious observation of any yellowing shift at 420 nm in conventional aromatic polyimides. The transmission level is about 90% in the photopic region while there is about 10% Fresnel loss from a 10 μm optical film.

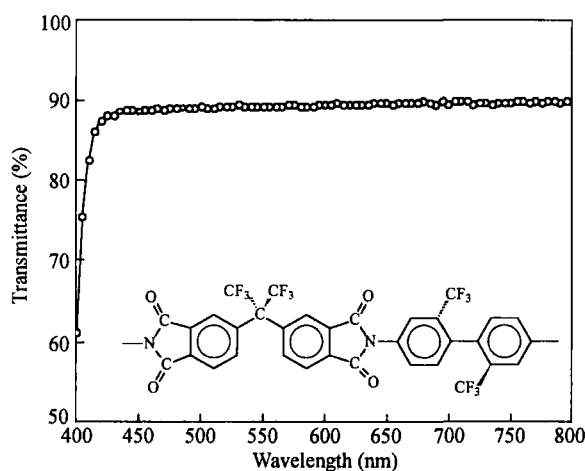


Fig. 5 UV/Vis transmission spectrum of 6FDA-PFMB as a function of the wavelength from 400 nm to 800 nm at a thickness of 10 μm , along with the chemical structure

High Uniaxial Negative Birefringent Optical Film Substrates

Refractive index and birefringence measurements for solution-cast films were conducted using a Metricon 2010 optical prism coupler, coupled with a He:Ne laser (at 633 nm). The negative birefringence for the polyimide films is measured with different thicknesses. The relationship between the in-plane ($n_x = n_y$) and out-of-plane (n_z) refractive indices and film thickness is illustrated in Fig. 6. The refractive indices were detected in both of the $n(\text{TE})$ and $n(\text{TM})$ modes, which represented molecular orientation along the in-plane ($n_x = n_y$) and out-of-plane (n_z) directions, respectively. It is evident that the polyimide long-chain molecules possess intrinsic uniaxial negative birefringence, because the refractive index along the in-plane direction is always greater than that along the out-of-plane direction. This indicates that the phenylenes and conjugated imide rings preferentially orient parallel to the substrate surface after solution casting and solvent evaporation from the glass hard substrates. As the polyimide film thickness increases, the in-plane refractive index, $n(\text{TE})$, gradually decreases. The out-of-plane refractive index, $n(\text{TM})$, on the other hand, gradually increases. The in-plane refractive index shows a certain dependence on the film thickness. As the polyimide film thickness increases, the negative birefringence is reduced. The reason for this behavior is presumably due to a decrease in the in-plane molecular orientation with increasing film thickness. This behavior is in agreement with our previous observations and the behavior of rod-like molecules^[13–15]. Anisotropic aromatic main-chain polyimide 6FDA-PFMB has a negative birefringence of 0.042 in a 6 μm film^[7].

The effect of polyimide film thickness on the uniaxial negative optical retardation values is also evaluated, as shown in Fig. 7. The optical negative retardation values increase from *ca.* 50 nm to 400 nm as the film thickness increases from 1 μm to 10 μm . The optical retardation values are calculated on the basis of the product of $\Delta n = n_x - n_z$ (birefringence) and d (thickness), which are measured independently. Therefore, the optical phase retardation values are adjustable to a desired value by varying the film thickness. Note that this material exhibits the typical characteristic features of uniaxial optical negative retardation film, with the in-plane retardation of less than 1 nm. The optical retardation of the typical C-plate is controlled between 50 nm and 400 nm for applications in flat panel LC displays. In the conventional C-plate, in-plane refractive index n_x is equal to n_y , and it is larger than that of the out-of-plane refractive index n_z . The color dispersion curve of the negative out-of-plane retardation between 400 nm and 800 nm is illustrated in Fig. 8. A monotonic decay color dispersion curve is desired as a function of the wavelength to meet the requirement of primary color match between LC molecules and optical compensation films employed in LCDs. As a result, the high contrast ratio and wide viewing angle compensation film is achieved in displays, along with significant improvement of the gray scale and color shift^[9, 16].

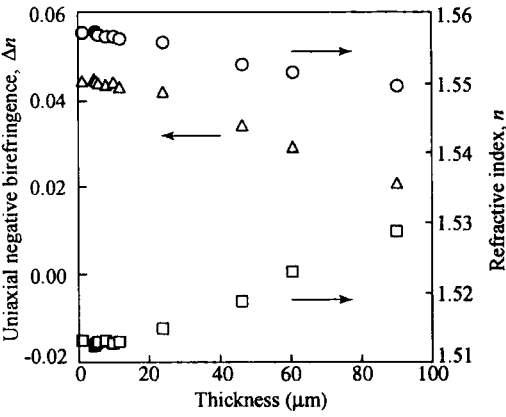


Fig. 6 Refractive indices (in-plane $n_x = n_y$, out-of-plane n_z) and negative birefringence (Δn) of 6FDA-PFMB optical films as a function of the film thickness

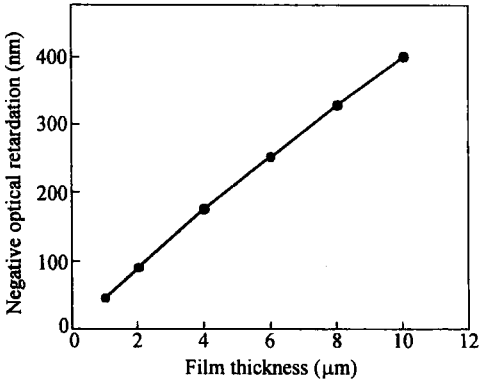


Fig. 7 Negative optical retardation of 6FDA-PFM optical films as a function of the film thickness ranging from 1 μm to 10 μm

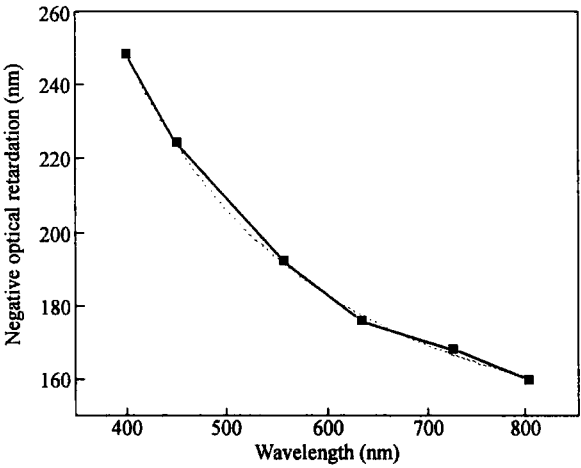


Fig. 8 Negative out-of-plane optical retardation of a 6 μm thick 6FDA-PFMB optical film as a function of the wavelength from 400 nm to 800 nm

For instance, a conventional LC display without an optical compensation film shows the narrow viewing angle, gray scale inversion and low contrast ration at the 70°-off angle, as demonstrated in Fig. 9(a). After the integration of our negative optical compensation films into the display as proposed in Fig. 4, it is evident that the viewing angle has been improved dramatically, along with high contrast ratio and little color shift, as shown in Fig. 9(b). If the LC parameters such as the cell gap, Δn , the color dispersion *etc.* in displays are more clear, the wide viewing angle optical performance can be further promoted.

Packing Models of Negative Birefringent Optical Film Substrates

On the basis of optical performance in uniaxial negative birefringent optical films, the packing model of this intrinsic negative birefringent film substrate is illustrated in Fig. 4, in which phenylene-imide rings and phenylenes preferentially lie in an anisotropic planar array. SERS signals from the original film surface further confirm that the phenylene-imide rings and phenylenes adopt a nearly planar orientation on the surface of solution-cast films^[17]. The anisotropic in-plane orientation in imide-phenylene conjugated structure and phenylenes discussed here is quite similar to that in other aromatic polyimide thin films, in which imide-phenylene rings orient parallel to the surface substrate, exhibiting uniaxial negative birefringent compensation film features^[7, 8, 13–15].

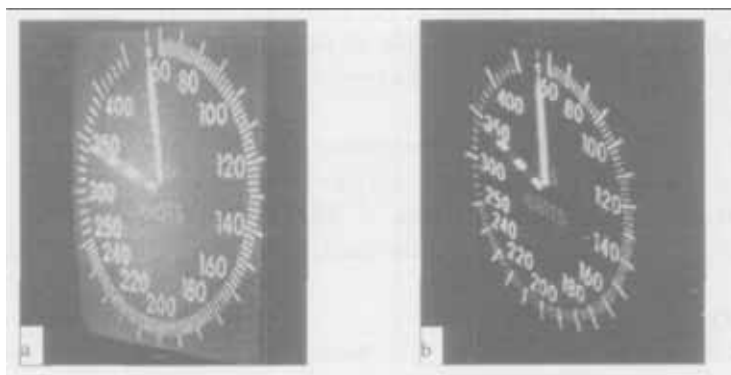


Fig. 9 (a) A conventional LC display without any optical compensation film at the viewing angle of 70° off-axis angle, exhibiting an obvious low contrast ratio and low viewing as well as color shift and (b) an negative optical film compensated LC display along the 70° off-axis angle with improved wide viewing angle and high contrast ratio and little color shift.

Oblique Optical Retardation Films

In order to further improve the viewing angle performance, the oblique optical retardation film is designed to mimic the tilt profile of LC molecules on the alignment surface. Rod-like and discotic LC molecules are engineered into a hybrid configuration on polymer alignment surfaces or inorganic SiO_x substrates through an oblique vapor deposition [2, 18–21]. The tilt configuration will be fixed via UV exposure without the obvious change in LC order parameters while the thermal stability and mechanical properties are also very important in the design of this complicated construction.

CONCLUSIONS

The aromatic polyimide 6FDA-PFMB has been successfully designed and synthesized for next generation uniaxial negative birefringent optical films. The uniaxial negative birefringent films ranging from 1 to 10 μm possess optical phase retardation values from 50 nm to 400 nm, which are quite suitable for applications in LC displays. Long chain aromatic polyimide molecules preferentially lie down in the film substrate, leading to negative birefringent optical performance in the free-standing optical films. Nearly planar molecular conformations in the aromatic backbone in the polyimide substrate have been confirmed. The high pretilt polyimide alignment surface will lead to the development of next generation oblique negative retardation optical films through “soft-epitaxy” growth with an additional discotic or rod-like LC birefringent layer. This new and effective technology can significantly improve the viewing angle and contrast ratio in LC displays.

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