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Polyimide films as negative birefringent compensators for normally white twisted nematic liquid crystal displays

Fuming Li, Frank W. Harris and Stephen Z. D. Cheng*
Maurice Morton Institute and Department of Polymer Science, The University of Akron, Akron, OH 44325-3909, USA
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Novel and reproducible uniaxial negative birefringent compensators for twisted nematic liquid crystal displays which are operated in the normally white mode have been developed for improving viewing angle characteristics and contrast ratio. The compensator design is based on the in-plane orientation of solution-cast aromatic semi-rigid polyimide films which possess a high degree of transmission and a birefringence dispersion similar to the liquid crystals over the visible spectral region. The films have been assembled into liquid crystal displays, and the isocontrast map of the displays using this compensator has shown that the viewing angle has been extended 10° or more on each side along the horizontal direction compared to that of the uncompensated displays. Copyright © 1996 Elsevier Science Ltd.

(Keywords: polyimide film; compensator; liquid crystal display)

Introduction

Thin-film-transistor active matrix twisted nematic (TN) liquid crystal displays (LCDs) can offer simultaneously the advantages of high information content, colour capability, grey scale and fast response. They are widely used in televisions, lap-top computers and information-related equipment. The TN-LCDs operated in the normally black (NB) display mode are not practical due to the manufacturing tolerance, low contrast ratio and poor colour-reproduction associated with this mode. Most of the TN-LCDs are operated in the normally white (NW) mode, that is the TN-LCD is light-transmissive under the undriven state (field-off state) and dark in the driven state (field-on state). One of the major drawbacks of the NW-TN-LCDs is asymmetric viewing angle characteristics along the horizontal and vertical directions, as well as narrow viewing angles[1]. This is a critical issue for applications such as avionics displays, where cross-cockpit viewing is required. Loss of the contrast ratio at high viewing angles for NW-TN-LCDs is the result of light leakage in the dark state. From a molecular point of view, the liquid crystals are aligned in a twisted nematic orientation induced by surface alignment layers in the undriven state, but in the driven state they are in a quasi-homeotropic state having a uniaxial positive birefringence (UPB). Without consideration of the pretilt angle in active matrix NW-TN-LCDs, they are in perfect homeotropic state under enough driving voltage. Therefore, under the driven state, a light beam travelling normal to the cell is parallel to the long optical axis of the liquid crystals and does not see any birefringence. The cell shows the black state. However, an off-axis light beam, with an incident angle θ with respect to the normal direction of the LCD surface, undergoes a retardation due to the contribution from both ordinary (n_o) and extraordinary (n_e) refractive indices of the liquid crystal. Consequently, light leakages occur. The light leakages in the black state cause a low contrast ratio in the oblique direction. In order to eliminate the light transmission in the oblique viewing direction, an optical compensator (OC) can be inserted between the cross-polarizers. The optical compensator has negative birefringence while the homeotropically aligned liquid crystal has positive birefringence, as shown in *Figure 1*. Therefore, the retardation of the LC (R_{LC}) is compensated by the opposite negative retardation of the OC (R_{OC}). Theoretically, without consideration of the pretilt angle and if the driving voltage is high enough to drive the LC into the perfect homeotropic state, the total retardation becomes $R = R_{LC} + R_{OC} = 0$. Therefore, the off-axis of the light transmission to the normal direction (leakage) under the driven state ideally approaches zero[2].

Several techniques have been reported to generate such uniaxial negative birefringence (UNB) compensators. One of the known examples is the use of ultra-super-twisted (UST) liquid crystal cells, which have an opposite symmetry and were used to compensate this driven (homeotropic) state and improve the viewing angle[3]. The cost and non-uniformity of the large area UST-LC compensators limit the application of this technique. Certain types of films having negative birefringence were recognized and used to compensate this homeotropic state[4–8]. One approach uses biaxially stretched polymer films. However, it is very difficult to produce polymer films which are uniformly stretched in different directions parallel to the film plane using mechanical force fields. A gradient of birefringence along the direction perpendicular to the film developed during cooling is also a major problem. This approach was recently mentioned by Kuo *et al.*[6] without detailed information. A UNB compensator has also been fabricated using vacuum deposition of alternating multiple thin layers (each 0.02 nm thick) or silicon dioxide and

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* To whom correspondence should be addressed

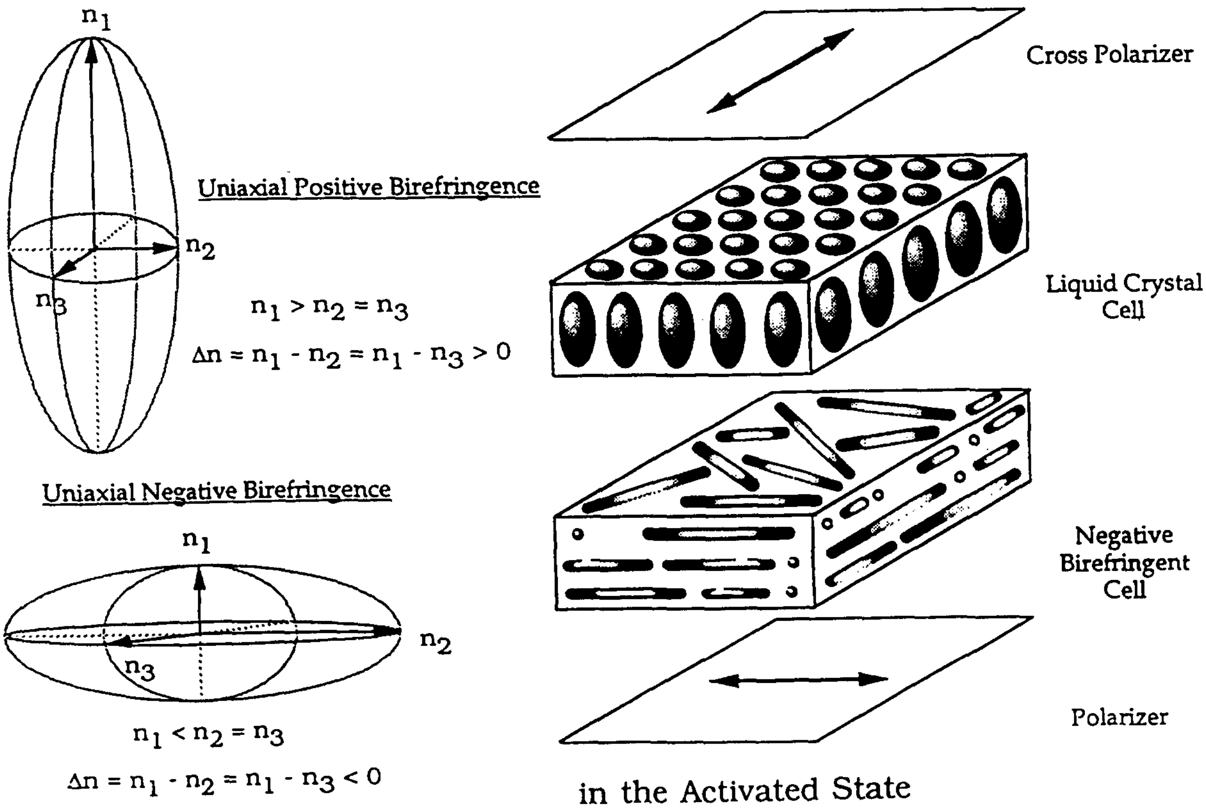

Figure 1 Optical principle of the NW-TN-LCD with a UNB compensator

titanium dioxide[7,8]. Although the film has good optical UNB properties, the epitaxial processing is complicated. High cost and poor environmental stability of this film are also restrictive. A very recent method proposed is based on pressing polymers above their softening points between two substrates. Birefringence is developed, due to the internal mechanical constraints, during cooling[5]. Obviously, it is hard to obtain enough birefringence and large uniform film dimensions with this method. The birefringence is also process dependent. Novel and simple methods are thus needed to develop compensating films with reproducible UNB.

Results and discussion

Aromatic polyimides exhibit excellent electrical and mechanical properties, along with high thermal, light and dimensional stability. This unique combination of properties has lead to many industrial applications for polyimide films. For many years it has been recognized that segmented rigid-rod polyimide thin films, with a thickness from 1 to 30 μm, exhibit an anisotropic structure in the directions parallel and perpendicular to the film surface, which is called 'in-plane orientation'[9–12]. This anisotropic structure leads to differences in the thermal, mechanical, dielectric and optical properties parallel and perpendicular to the film surface[13]. This 'in-plane orientation' produces a UNB character in the film. However, aromatic polyimides are traditionally difficult to process because they are insoluble in conventional solvents and do not flow below their decomposition temperatures. In the past, they were usually processed in the form of their poly(amic acid) precursors which were solution-cast into a film and then either thermally or chemically imidized. The imidization history often affects the ultimate morphology and structure of the films[14,15], which are generally difficult to control, and also affects their optical properties. The strong optical absorption in the low wavelength side of the visible spectral region, crystallinity, and the dark brown colour of the traditional polyimide films also preclude their use as optical retarders. Recently, we designed and synthesized a family of organo-soluble, segmented rigid-rod polyimides in our laboratory. These polyimides were synthesized in refluxing *m*-cresol at high temperatures through a one-step process in which the intermediate poly(amic acid)s were not isolated[16]. The resulting polyimides can be dissolved in common organic solvents and directly processed to form films having the characteristics of low optical insertion loss and reproducible UNB suitable for use as retarders. The improved contrast performance in NW-TN-LCDs using these films as a compensator has also been demonstrated[17].

Figure 2 shows the aromatic polyimides produced by the polymerization of 2,2′-bis(trifluoromethyl)-4,4′-diaminobiphenyl (PFMB) and different dianhydrides[16]. The incorporation of substituents at the 2- and 2′-positions in this diamine leads to changes of electronic state, interruption of the bond conjugation, non-coplanar conformations and decreases in the chain packing, without sacrificing the rod-like linear conformation. Dianhydrides with different linkage groups critically affect refractive indices (1.52–1.65) and UNB (0.04–0.10). *Table 1* gives the in-plane (n_e) and the out-of-plane (n_o) refractive indices and birefringence ($\Delta n = n_e - n_o$) determined at 589 nm wavelength for several PFMB-based polyimides and different dianhydrides. Differences in the linkage and conjugation of the dianhydrides also bring differences in crystallinity, solubility and optical absorption in the visible region. The relationship

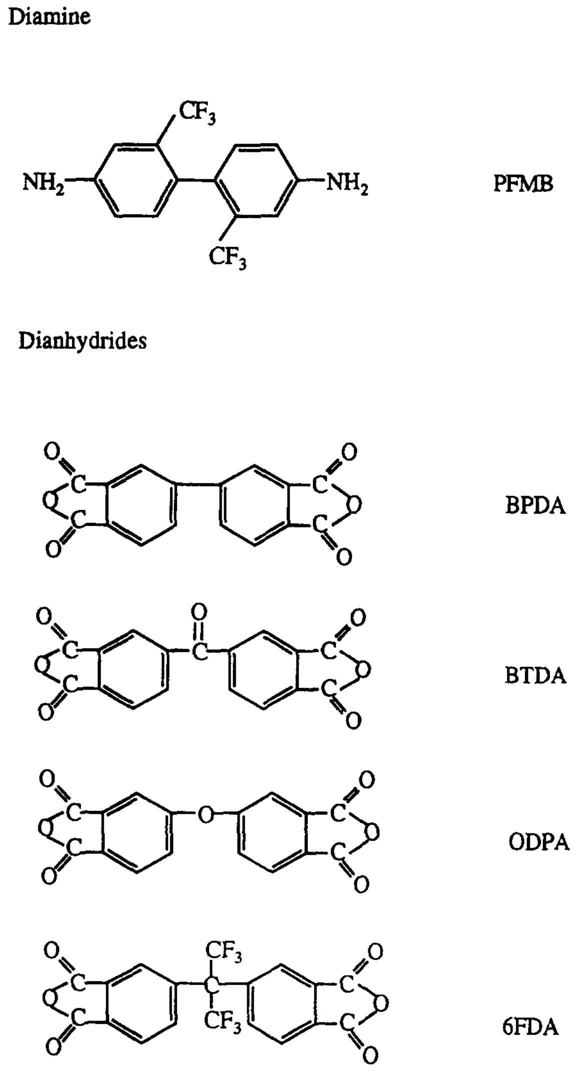

Figure 2 Aromatic polyimides based on different dianhydrides with PFMB

between the structures and properties can be determined and used to design the required polyimides as UNB for optical applications[18].

One of the polyimides, 6FDA-PFMB has been used to make retarders in NW-TN-LCDs. 6FDA-PFMB was found to dissolve in common organic solvents such as acetone, tetrahydrofuran, methyl ethyl ketone, PGMEA, chloroform and 2-pentanone. Films with different thicknesses, and therefore different retardations (retardation is equal to birefringence multiplied by the thickness of the film) can be obtained by spin-casting or by the dip-emersion method. The film thickness is uniform to within 1.5% over areas 25 cm square. After solvent evaporation at 100°C in a vacuum oven, 6FDA-PFMB films are highly transparent (around 88%) in the 0.4–1.5 μm spectral region, as shown in *Figure 3*. The Fresnel loss is about 6% at each surface or around 12% for both surfaces. The birefringence of a 10 μm thick 6FDA-PFMB film cast from 2-pentanone was determined in the visible wavelength region between 500 and 800 nm (*Figure 4*). An equilateral prism coupling technique was used to locate radiation modes in films cast on fused silicon wafers. The incident angles at which these modes were observed were used to calculate the refractive indices in both transverse electric (TE) and transverse magnetic (TM) mode polarizations within the visible wavelength region (optical waveguides possess two independent sets of optical propagation modes which can be excited by rotating the polarization state of the incoming laser beam by 90°). The symbols in *Figure 5* are experimental data and the solid lines represent the refractive index dispersion theory[19]. The dependence of birefringence on wavelength of the 6FDA-PFMB film compared with one of the LC mixtures developed for display application (Merck ZLI-1840) is shown in *Figure 6*. Although the Δn of 6FDA-PFMB film is about three times smaller, the Δn dispersion of this film is similar to that of LC mixtures. A good birefringence dispersion match between the retarder and the LC mixture over the entire visible range leads to a null phase retardation for all three primary colours employed in displays. As a result, a high contrast ratio can be achieved[20,21].

Table 1 Ordinary and extraordinary refractive indices and birefringence of several aromatic polyimides based on PFMB diamine

Polyimide	n_o	n_e	Δn
BPDA-PFMB	1.540	1.634	0.094
BTDA-PFMB	1.576	1.639	0.063
ODPA-PFMB	1.587	1.639	0.052
6FDA-PFMB	1.522	1.562	0.040

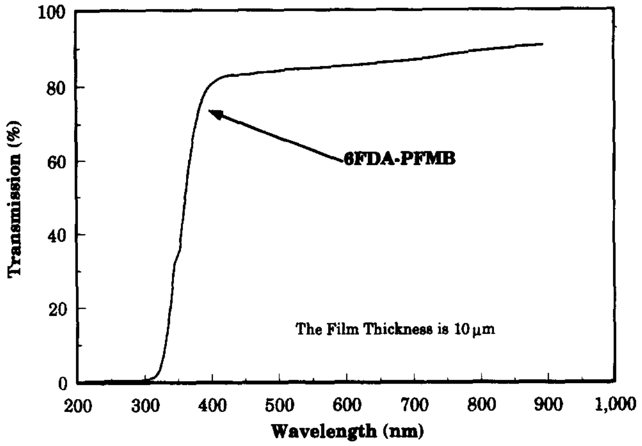

Figure 3 U.v.–visible transmission spectrum of 6FDA-PFMB

To demonstrate the potential use of this UNB film as a phase retardation compensator, we performed experiments in the NW-TN-LCD cell with a cell gap of 5.5 μm and a liquid crystal UPB with Δn equal to 0.15 at 589 nm. Uncompensated and compensated cells show different isocontrast maps (*Figure 6*). The region of high contrast has been expanded by 10° or more in the horizontal direction relative to the uncompensated cell in both the left and right viewing hemispheres. Note that the vertical viewing region has been decreased by addition of the compensator. The remaining viewing range is still well within the requirements for displays. Optimization of the compensation value of the film will bring better results.

In conclusion, we have developed a new and reproducible method to obtain UNB compensators which can be used in NW-TN-LCDs for improving the viewing angles and contrast ratio. This method has been adopted by LCD industries. Polyimide molecular weights and molecular weight distributions, solvent, solution concentrations, evaporation rate, annealing and film thicknesses also are expected to affect the ultimate

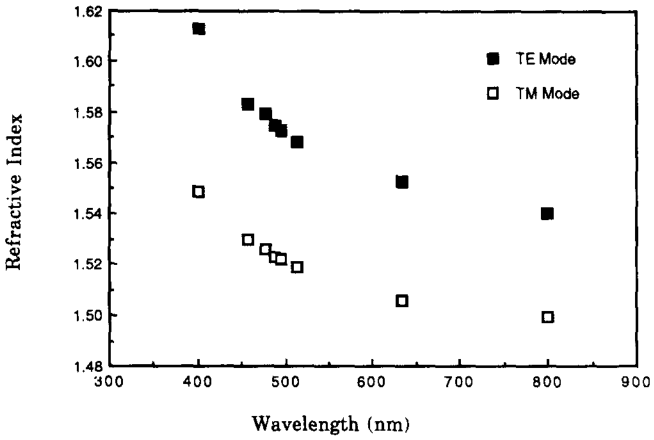

Figure 4 Refractive indices n_0 and n_e of 6FDA-PFMB as a function of wavelength

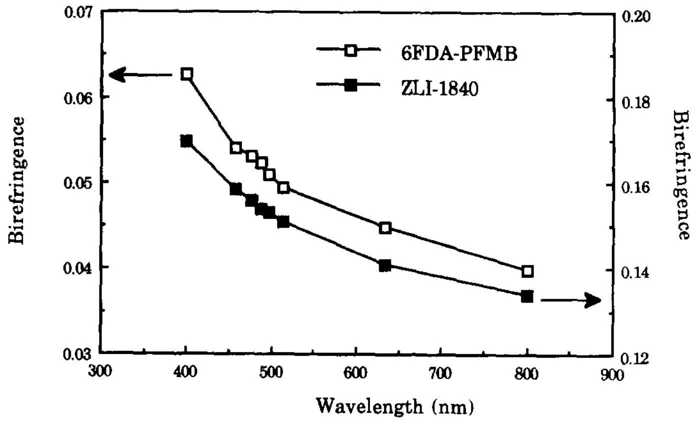

Figure 5 Birefringence dispersion of 6FDA-PFMB and LC (ZLI-1840, Merck) as a function of wavelength

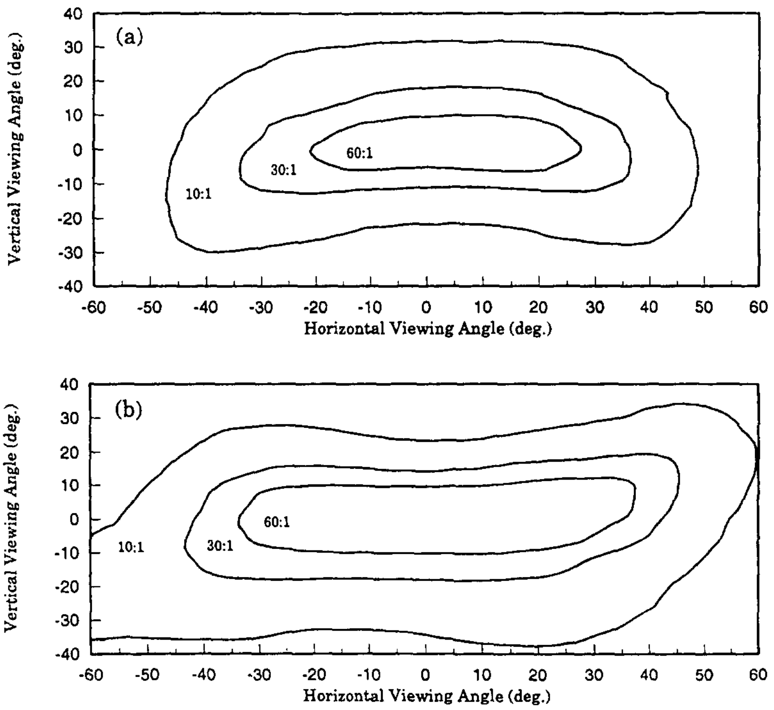

Figure 6 Isocontrast maps of the NW-TN-LCDs without (a) and with (b) a 6FDA-PFMB UNB compensator

UNB optical properties in these phase retardation compensators. These effects will be discussed in the near future.

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