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## Flexible Homeotropic Liquid Crystal Displays Using Low-Glass-Transition-Temperature Poly(ethylene terephthalate) Substrates

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This research is aimed at fabricating flexible homeotropic liquid crystal displays (LCDs) using low-glass-transition-temperature poly(ethylene terephthalate) (PET) substrates. The low-temperature technique of nanoparticle-induced vertical alignment (NIVA) was successfully applied to the flexible homeotropic PET-LCD. The PET film generally exhibits birefringence, which greatly interferes with the phase retardation of light propagation in the LCD. This problem can be alleviated by adjusting the optical axis with respect to the axis of the polarizer. The resulting flexible PET-LCD exhibited a low threshold voltage of 2.1 V and a transmittance of 53% at 3.2 V in the flat state.

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Flexible displays have been receiving increasing attention owing to their unique properties such as their light weight, thin packaging, impact resistance, and ease of use compared with conventional glass-based displays.<sup>1)</sup> The development of materials and equipment used to manufacture liquid crystal displays (LCDs) is rather advanced and more competitive than that of other display technologies. Thus, LCD technology is a good candidate for realizing flexible displays with high image quality.<sup>2)</sup>

Flexible displays are composed of flexible substrates (replacing conventional glass substrates), barrier layers, conducting layers, and functional thin-film materials. Flexible substrates made of flexible plastics or other bendable materials represent the fundamental starting element for these displays. In order to meet the cost, performance, reliability, and manufacturing goals of flexible electronics and displays, one strategy is to vary the materials used to manufacture LCDs and adopt manufacturing processes suitable to flexible displays. Poly(ethylene terephthalate) (PET) is a commonly used plastic substrate. Its semicrystalline character exhibits birefringence owing to its biaxial nature,<sup>1)</sup> making it suitable for use in optical compensators, retarders, reflective polarizers, brightness enhancement films, color filters, and scattering films.<sup>3)</sup> However, PET is not suitable as a substrate for polarization-mode LCD applications because the birefringent film changes the polarization state of the LCD owing to inherent phase retardation ( $\sim 70$  nm for a 50- $\mu\text{m}$ -thick film). In addition, conventional PET can only be used in low-temperature processes because of its low glass transition temperature ( $T_g$ ) of 75 °C, resulting in poor thermal stability.

In the manufacturing process of flexible LCDs, the thermal stability of the film is an important concern, particularly when high-temperature processes, such as the formation of polyimide (PI) alignment layers, are involved. If the plastic substrates possess a high  $T_g$ , the precision registration of different layers in the final device can withstand thermal cycling during the fabrication of multi-layer devices. Flexible substrates with better thermal resistances, such as polycarbonate (PC)<sup>4,5)</sup> and poly(ester sulfone) (PES),<sup>6)</sup> are used commercially in flexible displays. However, the costs of these films are relatively high

compared with the PET film. For example, the costs of PET, PC, and PES substrates are 5%, 6–10 times, and 8 times the cost of a glass substrate, respectively.<sup>7)</sup> Taking into account the marketing potential of flexible displays, all the components should be made cost-effective in order to be suitable for daily use. Therefore, it is worthwhile to develop flexible LCDs using low-cost substrates.

Recently, our group introduced a low-temperature processing method called nanoparticle-induced vertical alignment (NIVA),<sup>8–12)</sup> in which mixing a small amount of polyhedral oligomeric silsesquioxane (POSS) nanoparticles in LC induces spontaneous vertical alignment without using polyimide alignment layers for a display cell. In this study, homeotropic alignment of the LCD cell using PET substrates was achieved by the NIVA method and the optical structure of this cell was optimized by the photointensity method. The electrooptical characteristics of the PET-LCD cell were investigated under both flat and bent conditions.

Aminoethyl-aminopropylisobutyl-POSS (AM-0275) was purchased from Sigma-Aldrich. The display medium of the POSS-doped LC mixture consisted of negative dielectric anisotropic liquid crystal (MLC-6882,  $\Delta\epsilon = -3.1$ ,  $K_{33} = 12.8$  pN, Merck) and POSS at a 95/5 w/w. The medium was dissolved in a small amount of acetone with the application of ultrasound for 1 h, followed by heating at 50 °C for 1 h to evaporate the acetone solvent. The good dispersion of POSS in the LC was confirmed by the absence of any visible clusters in the mixture. The substrates in the LCD cells consisted of two PET films (with optical axes parallel to each other) coated with transparent electrodes of indium-tin-oxide (ITO). The thickness of the PET film was 125  $\mu\text{m}$  (Toray NX-01). The cell gap was maintained by bead spacers with a diameter of 8.5  $\mu\text{m}$ . The display medium was introduced into the empty cell by capillary action in the isotropic state to avoid nonuniform injection. The filled cell was then slowly cooled to room temperature.

The experimental setup for measuring the electrooptical properties of the sample cells is shown in Fig. 1. The He-Ne laser beam ( $\lambda = 633$  nm) was used as the incident light and the voltage of 1 kHz square waves was applied. The computer-controlled LabVIEW data acquisition system was used to drive the applied voltage, and light transmittance data of the sample were recorded. The electrooptical property of the sample cell under different bending curvature

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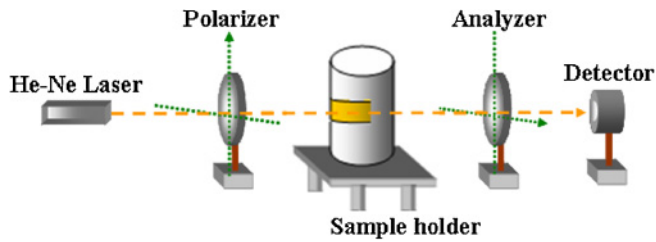


Fig. 1. (Color online) Schematic representation of electrooptical measurement system for flexible LC cell.

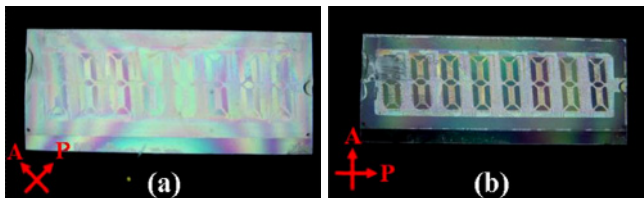


Fig. 2. (Color online) Off-state PET-LCD cell with characteristic angle between the optical axis of PET and the axis of polarizer (P) or analyzer (A) of (a) 45° and (b) 0°.

conditions was measured by attaching the sample onto a cylindrical holder with various curvatures ( $D = 5, 8, 10,$  and  $15$  cm) followed by insertion between the crossed polarizers.

The PET substrate exhibits significant birefringence that greatly interferes with the performance of the LC cell. The dependence of polarization of incident light on the electrooptical properties of a LC cell implied that film substrates with birefringence may change the polarization state. Amorphous film substrates, such as PC or PES, do not exhibit birefringence and are more suitable for flexible LCDs. However, all the components should be made cost-effective in order to be suitable for daily use, and the high cost of PC or PES will limit the marketing potential of flexible LCDs made of these materials.

In order to use birefringence substrates in polarization-mode LCDs, we used the photointensity method to find the optical axis of PET substrates. We rotated a PET-LCD between a crossed polarizer and analyzer to determine the characteristic angle between the optical axis of PET and either the polarizer or analyzer by recording the minimum (0°) and maximum (45°) light intensities detected using a photodiode. Figure 2 shows the pictures of a PET-LCD with the characteristic angles of (a) 45° and (b) 0°. At 45°, the PET substrate produced great retardation that significantly affected the uniformity of transmittance over the display area. Adjusting the characteristic angle to 0° suppressed this defect and provided visually acceptable uniformity. Accordingly, the characteristic angle was fixed at 0° in our subsequent experiments. As shown in Fig. 2, the NIVA is only observed in the patterned-electrode regions. The POSS nanoparticles were absorbed on the ITO surfaces but not on the bare PET surfaces because of the formation of hydrogen bonding between the amino groups in the side chain of POSS and the oxygen groups of ITO.

Figure 3 shows photographs of flat and bent price-tags consisting of PET-LCD in off and on states. The POSS nanoparticles accumulated on the ITO surfaces induced the LC molecules to align vertically between the ITO layers in the cell and resulted with the dark to bright transition of flat

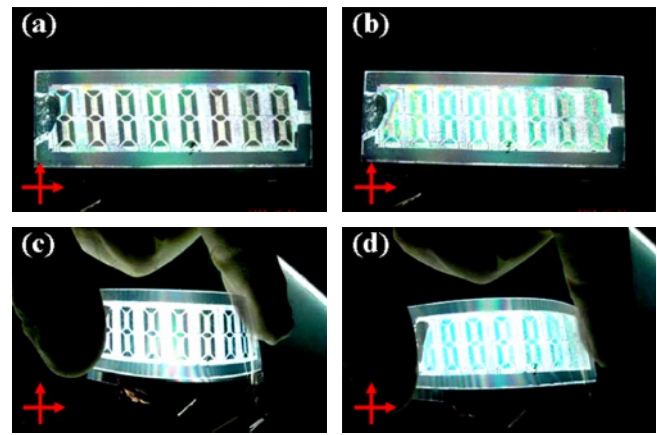


Fig. 3. (Color online) Photographs of PET-LCD under (a) flat condition and off state, (b) flat condition and on state, (c) bent condition and off state, and (d) bent condition and on state.

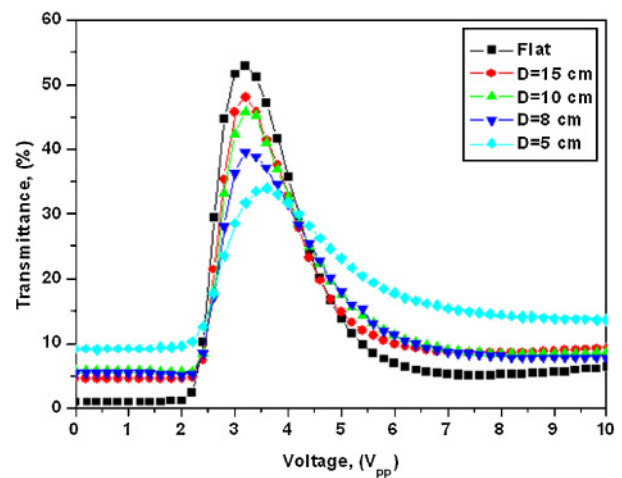


Fig. 4. (Color online) Measured optical transmittance of PET-LCD cell at various bending curvatures vs applied voltage.

PET-LCD while the external voltage was being applied. The bent price-tag exhibited good flexibility, but suffered from subtle light leakage caused by the strain-induced birefringence of PET.

A plot of the voltage-dependent optical transmittance of PET-LCD under different bending curvatures is shown in Fig. 4. When the cell was flat, the dark-state transmittance was only 1%, and the transmittance began to increase at its threshold voltage ( $V_{th}$ ) of 2.1 V, and reached its maximum value of 53% at 3.2 V. The measured contrast ratio of a flat cell was 49 : 1. When the cell was bent, the value of  $V_{th}$  was almost unchanged from 2.1 V, and the maximum values of optical transmittance of the cell were 48, 45, 39, and 33% as the bending curvature changed to  $D = 15, 10, 8,$  and  $5$  cm, respectively. The dark-state transmittance of the bent cell was 5 to 9% and the contrast ratios were 11 : 1, 8 : 1, 7 : 1, and 4 : 1 as the cell was bent to  $D = 15, 10, 8,$  and  $5$  cm, respectively. Our homeotropic PET-LCD results showed that the contrast ratios were 49 : 1 and 4 : 1 for the flat and bent states ( $D = 5$  cm), respectively, and the transmission of the bent state was 40% lower than that of the flat state. The reasons for this deviation may be the cell gap variation and slight strain-induced birefringence of PET substrates for the bent homeotropic PET-LCD.

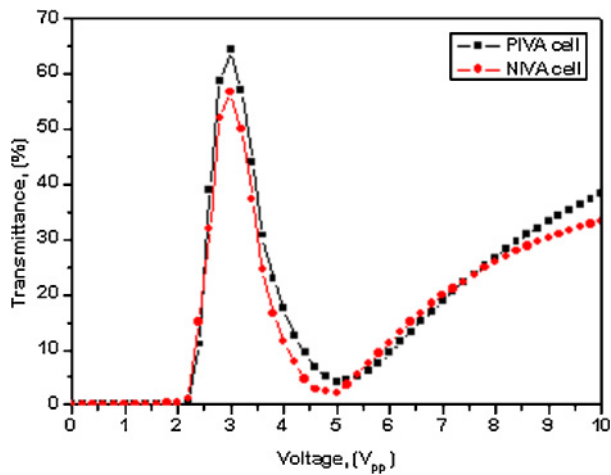


Fig. 5. (Color online) Measured optical transmittance vs applied voltage of PIVA and NIVA on glass cells.

Moreover, nanoparticle aggregation may increase the dark-state transmittance as a result of light scattering. The voltage-dependent optical transmittances of the liquid crystal cells aligned vertically using conventional vertical alignment layers of PI (PIVA) and the NIVA method are shown in Fig. 5. In order to study the effects of light scattering due to the aggregation of nanoparticles, both cells were fabricated by comparable manufacturing processes using the same glass substrates to eliminate the noises caused by the manufacturing process and substrate material of PET. The results show that the dark level of the PIVA was only 0.103%, compared with 0.133% of the NIVA cell. The maximum values of optical transmittance of the PIVA and NIVA cells were 64 and 57%, respectively. Therefore, the contrast ratios of the PIVA and NIVA cells were 621 : 1 and 428 : 1, respectively. The value of  $V_{th}$  was approximately 2.1 V, and the saturated voltage was approximately 3.1 V for both cells. On the basis of the above results, it is likely that nanoparticle aggregation may be one of the factors that induce light scattering and decrease the contrast ratio of the NIVA cell.

The birefringence of the PET substrate cannot be ignored as the bending curvature of the film increases. Hence, the performance of our NIVA PET-LCD was degraded at a larger magnitude of transmittance during the bending state. The birefringence variations of a bare PET film between flat and bent states were analyzed by calculating the phase retardation ( $\Delta nd$ ) using<sup>13)</sup>

$$\frac{I_{\perp}}{I_{\parallel}} = \tan^2 \frac{\pi \Delta nd}{\lambda}, \quad (1)$$

where  $I_{\perp}$  and  $I_{\parallel}$  are the intensities of the transmitting light with the film placed between crossed and parallel polarizers, respectively, and  $\lambda$  is the beam wavelength. The measured  $\Delta nd$  of the phase retardation in our instrument was limited to  $D = 10$  cm and showed that  $\Delta nd$  decreased from 312 to 289 nm (characteristic angle of  $45^{\circ}$ ) when the PET film changed from flat to bent with  $D = 10$  cm. These results suggest that the decrease in the phase retardation of PET substrates during bending resulted in the decrease in the transmittance of the LC cell.

The cell gap deviation during bending for a flexible LCD must be overcome. Various methods of producing microstructures in a cell gap have been proposed to maintain the cell gap during bending, such as the photoinduced phase separation method,<sup>14–17</sup> photolithography,<sup>18</sup> and imprinting techniques.<sup>2,19,20</sup> For example, a flexible LCD film using an amorphous PES substrate fabricated by adopting the phase-separated composite film method (PSCOF) to maintain the cell gap has been reported.<sup>6)</sup> The contrast ratios of the PSCOF LCD in the flat and bent states ( $D = 4$  cm) changed slightly from 72 : 1 to 60 : 1.

A homeotropic PET-LCD was successfully fabricated in this work. In this display, NIVA technology was used to realize an alignment-layer-free vertically aligned LCD. Our results indicate that the biaxial nature of PET substrates interferes with the optical transmittance and contrast ratio of the display cell. This problem can be alleviated by carefully adjusting the characteristic angle. The PET-LCD exhibited a low  $V_{th}$  of 2.1 V and an acceptable contrast ratio of 49 : 1 with a transmittance of 53% at 3.2 V in the flat state. Bending the PET substrate did change its phase retardation, and resulted in a decreased transmittance and contrast ratio of the cell. These results may be explained by considering the cell gap deviation and slight strain-induced birefringence of PET substrates for the bent homeotropic PET-LCD. Although the PET-LCD discussed in this study has problematic issues of a poor viewing angle and low contrast ratio, it is hoped that it can serve as a basis for future study in cost-efficient flexible LCDs.

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