Pixel-isolated liquid-crystal mode by using a patterned anisotropic phase separation for flexible LCDs

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Abstract — A pixel-isolated liquid-crystal (PILC) mode for enhancing the mechanical stability of flexible-display applications is proposed. Because liquid-crystal (LC) molecules in this mode are isolated in each pixel by patterned or phase-separated microstructures, and the two substrates are tightly attached to each other by a solidified polymer layer, the LC alignment is stable against external pressure, and the cell gap of our structure is uniformly preserved against bending deformation of the plastic substrates. The mechanical stability of the PILC structure having plastic substrates was tested for its electro-optic properties.

Keywords — *Liquid crystal, flexible display, phase separation, pixel-isolated liquid-crystal mode, mode stability.*

1 Introduction

In recent years, roll-up displays have drawn considerable attention for next-generation information displays because of their excellent portability, such as light weight, thin packaging, and flexibility. Among several available technologies, it is expected that a liquid-crystal display (LCD) using plastic film substrates is the most promising device because of its superior visibility with low power consumption over other displays, such as organic light-emitting devices or electrophoretic displays. However, there are still major problems in fabricating commercially available plastic LCDs with current technologies based on glass substrates. One of those problems is the instability of LC structures due to the hydrodynamic properties of LCs at bending. The separation of two plastic substrates due to the flexibility is also a problem that needs to be solved. Such problems do not exist in conventional glass-substrate-based LCDs because glass substrates can sustain a stable LC-alignment condition against external bending or pressure.

To solve these problems, several types of polymer walls and/or networks as supporting structures have been proposed and demonstrated.[1–9] These structures were fabricated using an anisotropic phase-separation method from polymer and LC composite systems by applying a patterned electric field[1] or spatially modulated UV intensity.[2,3,8] However, these methods require a high electric field to initiate the anisotropic phase separation or remain residual polymers in unexposed regions that reduce optical properties and increase the operating voltage of the devices.[1–3] Moreover, these methods are not appropriate for a cost-effective roll-to-roll process, which is essential in fabricating large-area plastic LCDs. Thus, an alternate fabrication method should be developed in order for plastic LCDs to be commercialized.

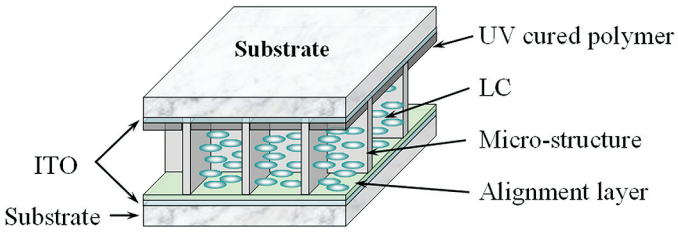

FIGURE 1 — Schematic diagram of a PILC structure made by patterned microstructures.

In this presentation, we propose various fabrication methods for enhancing the mechanical stability in the LC alignment and the adhesion of two substrates for plastic LCDs with a pixel-isolated LC (PILC) mode. The proposed PILC structures for stable and flexible LCDs are examined by measuring the electro-optic (EO) properties under various mechanical stability tests.

2 Fabrication of PILC mode using patterned microstructures

We fabricated a PILC mode using patterned microstructures as shown in Fig. 1. In the proposed structure, the pixel-isolating polymer walls were made by using a stamping method using durable elastomeric poly(dimethylsiloxane) (PDMS),[6,7] which can be applied to a roll-to-roll process for the mass production of large flexible LCDs.[11] The adhesion between the wall structure and the substrate was enhanced by a solidified thin polymer layer using an anisotropic phase-separation process.[4,5]

Figure 2 shows the schematic illustration of procedures for fabricating plastic LC devices by using a micro-transfer molding method. The first step shown in Fig. 2(a) is to produce a master structure with a negative photoresist SU-8 (MicroChem) by using a photolithographic method.

Revised extended version of a paper presented at the 2006 SID Symposium held June 6–9, 2006 in San Francisco, California.
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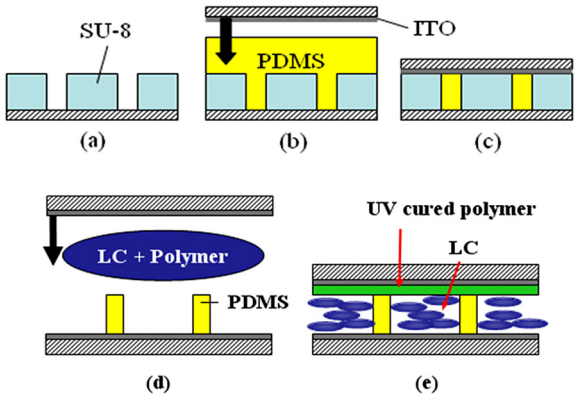

FIGURE 2 — Schematic illustrations of fabrication procedures with stamping method: (a) formation of micropatterned master structure, (b) fabrication of PDMS structure using the master substrate, (c) transferring the PDMS wall structure to a plastic substrate by baking under pressure, (d) forming a LC alignment layer on a plastic substrate for a microstructure, then dropping or injecting polymer/LC composites. The PILC structure is prepared by sandwiching the other plastic substrate. (e) The cross section of a PILC structure after phase separation by UV exposure.

On the master substrate, liquid PDMS is deposited and the excess liquid PDMS is removed by a PDMS block, as shown in Fig. 2(b). The PDMS wall structure produced by the patterned master structure can be effectively transferred to the covered bare indium-tin-oxide (ITO) substrate by heating under pressure, as shown in Fig. 2(c). In our experiment, the heating condition for transferring and solidifying the PDMS structure was 100°C for 10 min. By peeling off the master substrate, the bottom substrate with PDMS wall structures is prepared. Since PDMS provides a very low interfacial free energy and a good chemical stability, the master substrate can be easily detached without the degradation of the microstructure on both substrates.[11] Onto the prepared bottom substrate shown in Fig. 2(d), the homogeneous alignment layer RN1286 (Nissan Chemical Industries) is spin-coated and rubbed to promote a uniform LC alignment. After the mechanical rubbing process, the PDMS walls maintain the initial micropatterned structures attached to the ITO surface. After dropping a LC/prepolymer composite onto the substrate of the microstructure, a LC cell is prepared by laminating a bare ITO substrate on the bottom substrate. However, during these fabrication steps, the cell gap cannot be stably sustained at bending since the two substrates are not strongly attached to each other.

In our structure, such problems are solved by producing a uniformly solidified thin polymer layer on the bare ITO substrate using a complete anisotropic phase separation of the prepolymer/nematic LC (NLC) mixture by UV exposure.[4,5] The materials used are E48 (Merck) for the NLC and UV curable epoxy NOA-72 (Norland) for the prepolymer. A solution of the LC and prepolymer with a weight ratio of 95:5 is deposited on the substrate of the microstructure and covered by a bare ITO substrate, as shown in Fig. 2(d). The UV light is exposed to the bare ITO substrate in the isotropic phase of the NLCs. In our experiment, the source of the UV light is a xenon lamp of λ = 350 nm, operated at an electrical power of 200 W, and the exposure time is 20 min. The solidified polymer layer makes the patterned wall structures strongly attached to the opposite substrate and enhances the mechanical strength of the pixel-isolated LC device, as shown in Fig. 1.

Figure 3(a) shows the SEM images of the master structure of SU-8 with a pixel-area size of 100 × 300 μm^2. The pattern-transferred PDMS structures are shown in Fig. 3(b). By using our microtransfer method, we could successfully and repeatedly fabricate the micropatterned wall structures. Figures 3(c) and (d) show the microscopic textures of our cell under a polarizing microscope in the dark and white states without an applied voltage. Because the phase-separated polymer/LC interface has no preferred azimuthal anchoring direction after phase separation in the isotropic phase of the NLCs, the NLC alignment is promoted only by the single LC alignment layer on the bottom substrate shown in Fig. 1. Therefore, the homogeneously planar structure is obtained as shown in Figs. 3(c) and 3(d). Recently, we have shown that the twisted-nematic structure can be also obtained in the phase-separated LC/polymer system by temperature-controlled phase-separation procedures, where the azimuthal easy axis is induced on the LC/polymer interface by imprinting the LC order on the polymer during phase separation.[10] The low light leakages shown in Fig. 3(c) indicate that there are LC distortions on the side of the wall structures, which might originate from the homeotropic LC anchoring property of the PDMS surface[12] and the geometric effects of the wall structures to LC anchoring. However, the overall transmittance behavior in the pixel area is not affected.

In applications of plastic LC devices, adhesion between two flexible substrates is of great importance. We observed

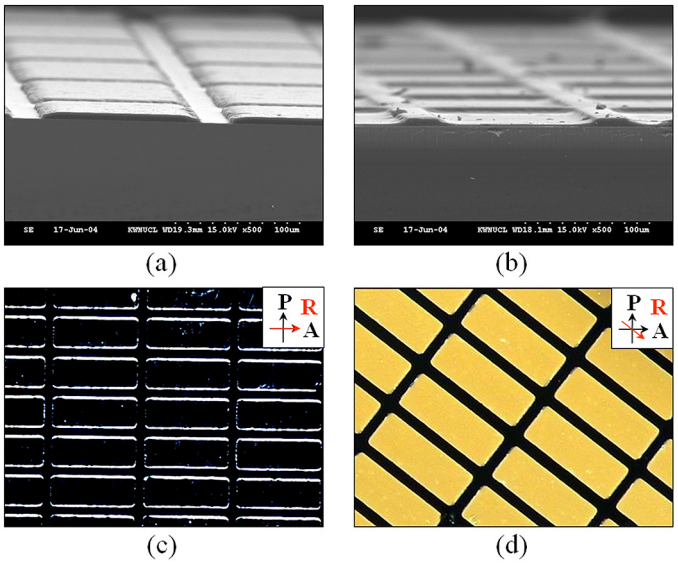

FIGURE 3 — (a) and (b) are SEM images of the micropatterned SU-8 master substrate and the stamped PDMS structure, respectively. (c) and (d) are polarizing microscopic textures of the PILC cell obtained when the rubbing direction of the sample is parallel and 45°-rotated with respect to the polarizer in the absence of an applied voltage, respectively.

the microscopic structure of the PILC cell by using scanning electron microscopy (SEM). Figure 4(a) shows the cross-sectional view of the PILC sample after removing the LC, where the thin polymer layer for tight binding of two substrates can be observed. Figures 4(b) and 4(d) are the SEM images of the top and bottom surfaces, respectively, observed after separating the two substrates, and Figs. 4(c) and 4(e) show their surface profiles. The thickness of the phase-separated binding layer is 200 nm. Using the stamped microstructure and the binding layer, we can provide suitable adhesion between two plastic substrates and good mechanical stability without cell-gap variation under external bending pressure. In our demonstration, PDMS was used for the stamping material. On the contrary, when PDMS is used for the mold structure, any photocurable or thermally curable material can be stamped for production of the wall structure.[11] In this case, the adhesion of the photopolymer between the wall structure and the upper substrate would be more enhanced when the low interfacial energy of the PDMS is considered.

3 Fabrication of PILC mode using anisotropic phase separation

3.1 Microstructures from three-dimensional anisotropic phase separation

We fabricated the pixel-isolating polymer wall structure by photo-polymerization-induced phase separation from LCs and pre-polymer composite material. Using UV intensity variation and polymer wetting properties,[4–6] the LC molecules in our structure can be isolated in pixels where LCs are surrounded by the inter-pixel vertical polymer walls and the horizontal polymer film. The fabrication process is shown in Fig. 5. One of the ITO-coated PES substrates was spin-coated with a homogeneous alignment layer and unidirectionally rubbed. The other substrate was untreated and remained with the bare ITO surface to promote a surface-wetting difference in the vertical direction during the second phase separation. A mixture of nematic LC (LC17) and photocurable pre-polymer (NOA65, Norland) with a weight ratio of 75:25 was filled into the cell by capillary action at isotropic temperature. During the first UV exposure, the UV was illuminated onto the bare ITO-coated PES substrate through the photomask of pattern size of 100 × 300 μm for 90 minutes [Fig. 5 (a)]. The second UV exposure was performed uniformly without the mask for 10 minutes to fully harden the pre-polymers. During these first and second photo-polymerization processes, the anisotropic phase separation occurs in the horizontal and vertical direction, respectively; therefore, vertical polymer walls and planar polymer layers were formed as shown in Fig. 5(b).

Figure 5(c) shows the cross-sectional SEM image of our PILC cell. The spatially distributed polymer walls formed by the first UV exposure act as supporting structures from external point pressure and bending distortion and maintain the cell gap of the plastic LC cell. The residual

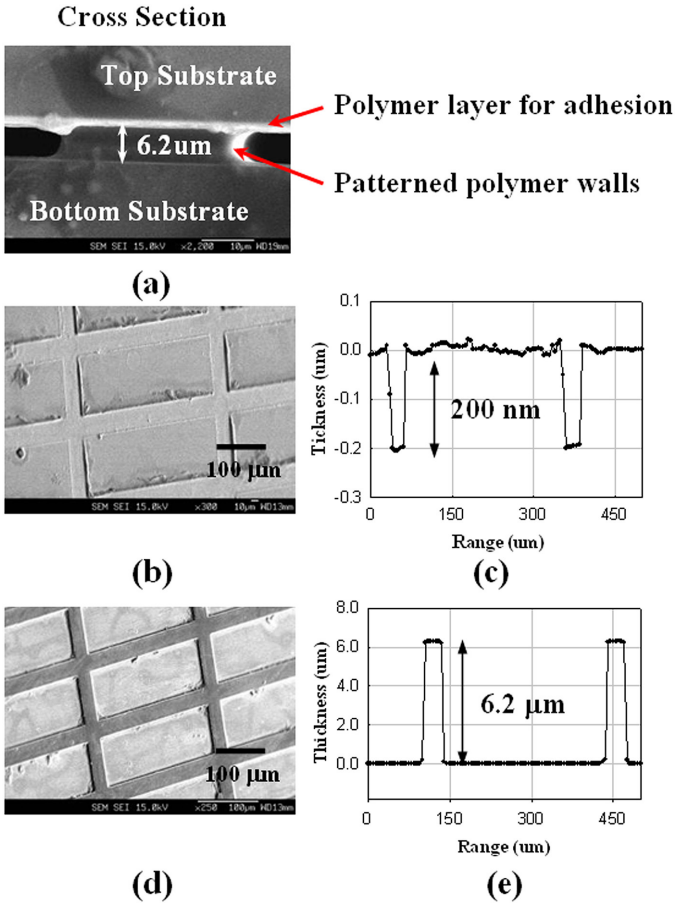

FIGURE 4 — (a) Cross-sectional SEM image. (b) and (d) are SEM images of the top and bottom surfaces, respectively, observed after separating two substrates, and (c) and (e) are their surface profiles.

pre-polymers are completely expelled from the bulk LC layer onto the bare ITO-coated PES substrate, forming a thin polymer layer during a second UV exposure. Due to this second UV exposure, our PILC mode demonstrates good EO properties and enhanced mechanical stability with good adhesion between the plastic substrates and the polymer walls. Figure 5(d) shows the polarizing optical microscopic image after UV irradiation. In this structure, the LCs are well-aligned by the single alignment layer along the rubbing direction without any residual polymer within each pixel.

3.2 Polymer-wall formation using patterned alignment layer

Three-dimensional polymer structures for the PILC mode can be made only by surface-induced phase separation without patterned UV exposure. This method uses the phase separation between the LC and prepolymers induced by the difference in the wetting property on a patterned surface. Figure 6 shows the schematic diagram for fabricating a PILC mode using phase separation, depending on the surface wetting property. To spatially modify the surface wetting condition to the LC/polymer mixtures, a homeotropic

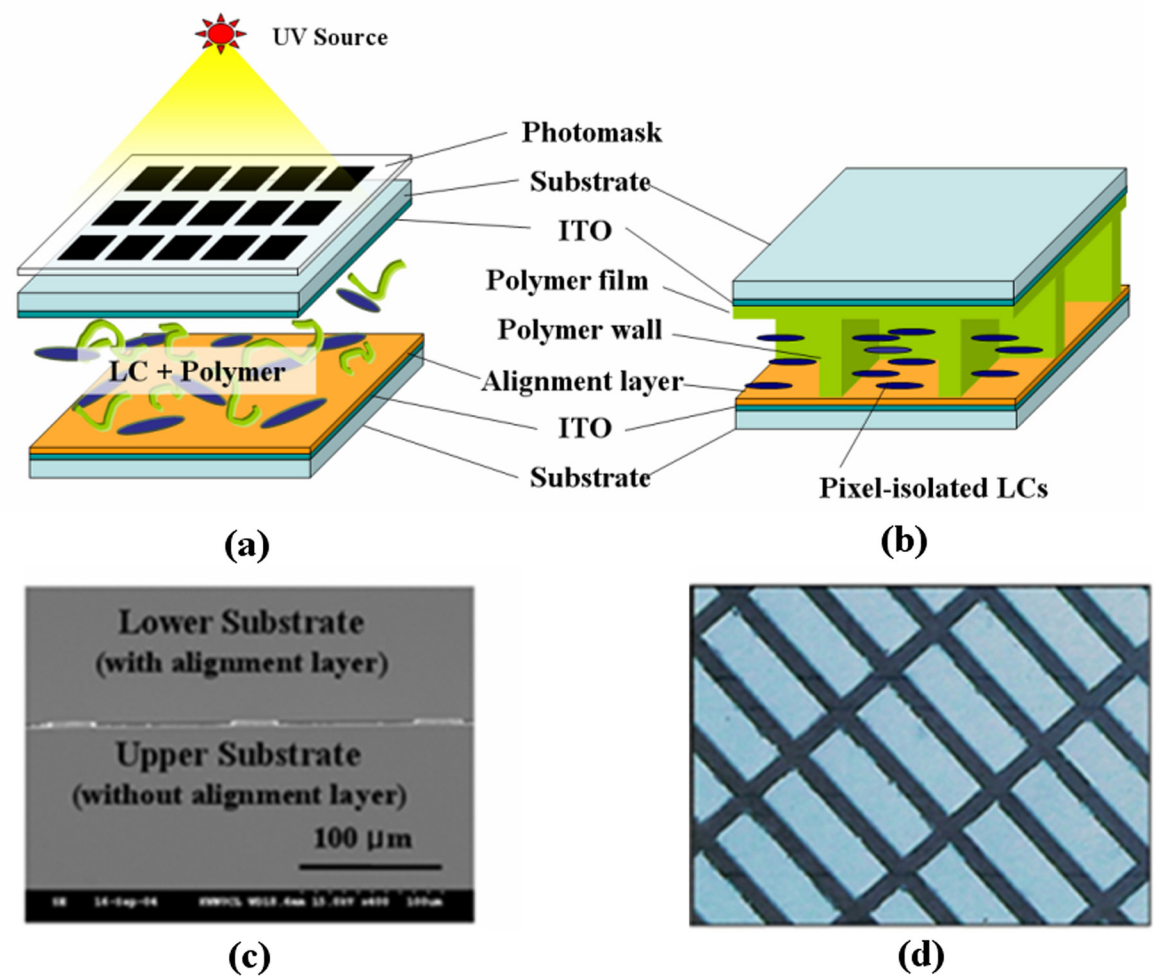

FIGURE 5 — (a) Schematic diagram of PILC mode made by photo-polymerization-induced anisotropic phase separation, (b) pixel structure after UV exposure, (c) the cross-sectional SEM image of PILC sample, and (d) the polarizing optical microscopic texture of the PILC cell.

alignment layer (JALS684, JSR Co.) was patterned on one of the bare ITO substrates by using a micro-contact printing method. On the other substrate, JALS684 was uniformly coated. The mixture of negative NLC (MLC-6610, Merck) and photocurable prepolymer (NOA65, Norland) was introduced into the cell by capillary action at an isotropic temperature. After filling the mixture, the cell was slowly cooled down to room temperature. Because the wettability of the LC is higher than that of the prepolymer on the patterned alignment layer, phase separation between the LC and the prepolymer are spontaneously promoted and the LC molecules are aggregated on the patterned alignment layers.

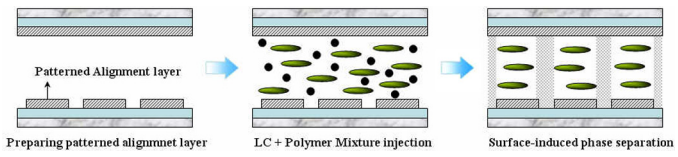

FIGURE 6 — The schematic diagram of a PILC mode where phase separation is induced by surface-wetting properties.

Figure 7(a) shows the polarizing optical microscopic images of the phase-separated cell before UV exposure. Because the LC and the prepolymer were completely phase-separated, there were no polymer-dispersed LC droplets either on the patterned alignmnet layers or the ITO surfaces. Figure 7(b) shows the texture in the presence of an applied voltage before UV exposure. Within the intra-pixel regions on the patterned alignment layer, an axially symmetric LC reorientation was observed, which originated from the sym-

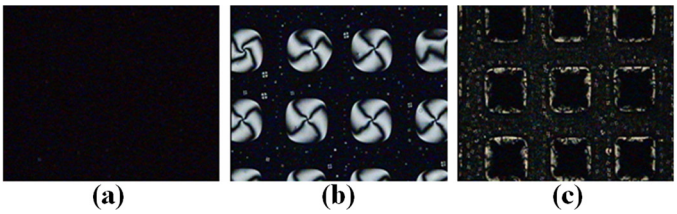

FIGURE 7 — Polarizing optical microscopic images of PILC cell where the phase separation is induced by surface wetting properties of LC and polymer on the patterned alignment layer. Before UV exposure, (a) and (b) are obtained in the absence and presence of an applied voltage, respectively. (c) is obtained in the absence of an applied voltage after UV exposure.

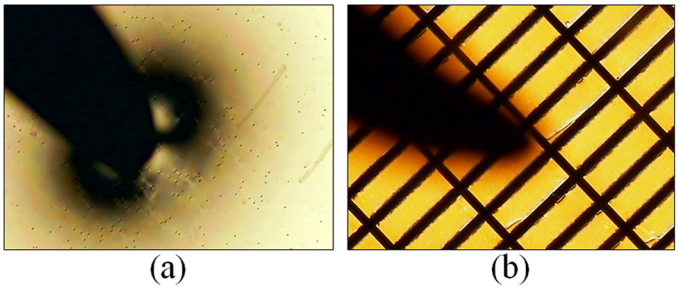

FIGURE 8 — Alignment textures of (a) a normal sample and (b) a PILC sample fabricated using the plastic substrates. The polarizing microscopic textures were taken in the presence of an external point pressure with a sharp tip.

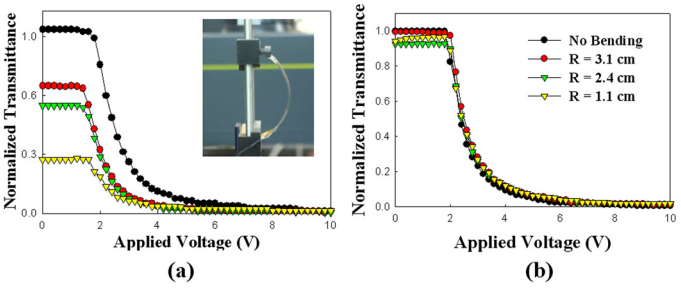

FIGURE 9 — EO properties of (a) a normal and (b) a PILC sample depending on the degree of bending.

metric boundary condition produced by the uncured prepolymer in the inter-pixel regions. In this method, only a one-step UV exposure is required for producing the PILC mode. To harden the prepolymers and to fix the phase-separated pattern, we performed UV irradiation without photomasking. Figure 7(c) shows the resultant texture in the absence of an applied voltage, where the LC molecules are isolated polymer microwall structures.

4 Electro-optic properties of a plastic LCD with the PILC mode

We tested the mechanical stability of the PILC cell against external pressure and bending distortion. Figure 8 shows the polarizing optical microscopic textures of the normal sample without a polymer structure and the PILC sample, prepared with plastic substrates in the presence of an external point pressure by a sharp tip. Between the flexible substrates, the alignment texture of the normal sample was

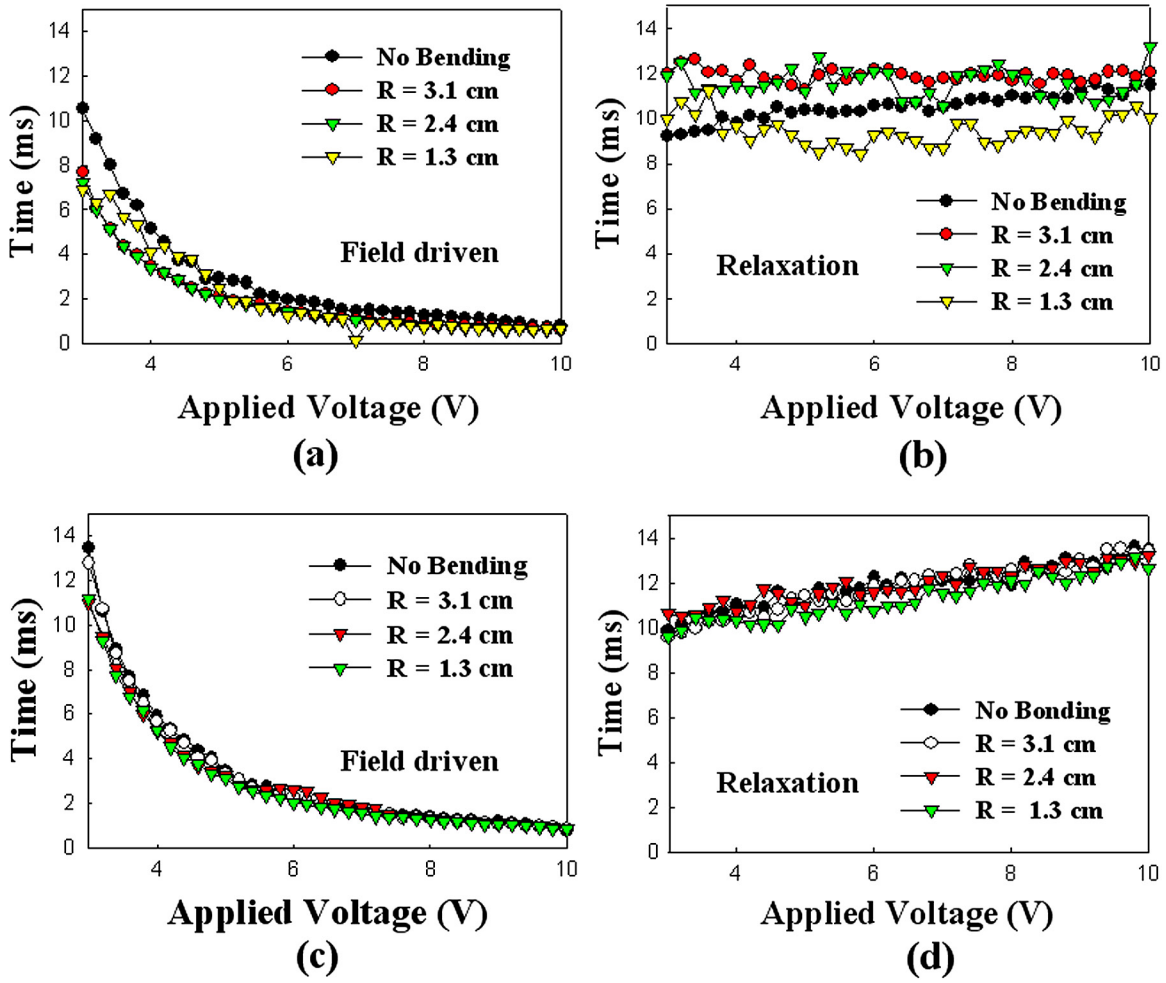

FIGURE 10 — Dynamic properties of field-driven and field-off relaxation times for a normal sample [(a) and (b)] and a PILC sample [(c) and (d)] depending on the degree of bending.

FIGURE 11 — A 3-in. plastic LCD sample in the PILC mode.

severely degraded by the distortion of the LC alignment due to cell-gap variation in a relatively large area as shown in Fig. 8(a). However, that of the PILC sample showed no appreciable changes because the distortion of the LC alignment was restricted and the cell gap was sustained by the polymer-wall structure, where the polymer-wall structures are shown as dark lines in Fig. 8(b).

Figure 9 shows the EO properties of a normal plastic LC cell and the plastic PILC cell in the presence of an external bending pressure controlled by a pair of linear stages, where R represents the curvature of bending radius. As the bending stress on the normal plastic LC cell increases, the LC molecules are severely distorted and the cell gap is decreased, which resulted in the decrease of the transmittance as shown in Fig. 9(a). However, the PILC cell shows almost the same transmittance properties irrespective of the degree of the bending pressure over the entire operating-voltage range. Notice that the transmittances shown in Fig. 9(a) are degraded by about 70% when the bending distortion was increased to R = 1.1 cm.

Figure 10 shows the dynamic properties of field-driven and field-off relaxation times for the normal sample and the PILC sample, depending on the degree of bending, respectively. The dynamic properties of the normal sample showed much variation due to the cell-gap variation under bending distortion, whereas the PILC mode showed stable dynamic properties.

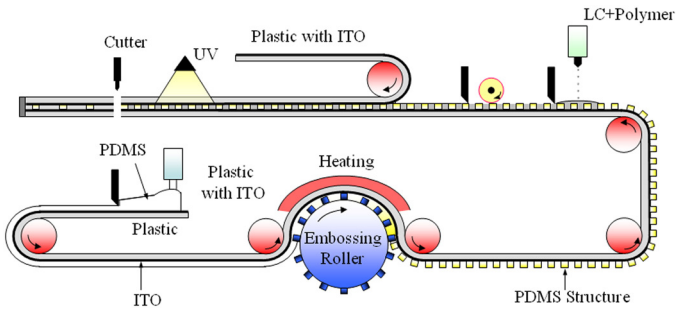

FIGURE 12 — Schematic diagram of continuous roll processing for the fabrication of plastic LCDs using the methods presented here.

Figure 11 shows a photograph of a 3-in. prototype sample of the plastic LCD in the proposed PILC mode. The resolution was 124 × 76 pixels with a pixel size of 100 × 300 μm^2, where the polymer walls were formed within each pixel with a 30 μm width. A full-color flexible LCD was achieved by laminating patterned color filter sheets on the top substrate. The response time (field driven + relaxation time) was about 20 msec. The contrast ratio was about 130:1 even at bending distortion.

5 Conclusion

Mechanical stability is a key issue for flexible LCDs with plastic substrates. To obtain mechanical stability, stabilization of the LC mode and adhesion of two plastic substrates are essential. In this demonstration, we presented stability-enhanced plastic LCDs using the PILC mode, where LC molecules are isolated in the pixels by the horizontal polymer layer and the vertical polymer walls. The polymer walls act as a supporting structure from mechanical pressure and maintain the cell-gap uniformly against bending distorion. Moreover, the polymer layer acts as an adhesive layer for strong adhesion of the two plastic substrates. Therefore, the plastic LCDs with the PILC mode show good EO properties against external pressure and bending.

We presented various fabrication methods of the PILC structure using anisotropic phase separation from a LC and polymer mixture. Among these methods, the fabrication of a patterned wall structure using the stamping method is the most compatible with continuous roll-to-roll processing. The schematic diagram in Fig. 12 shows the concept of the process flow. First, prepolymer for the micro wall structures is deposited onto an ITO-coated plastic film. After the thickness of the prepolymer layer is uniform, the micro wall structures are formed by using a stamping process with an embossing roller. On the micro wall structures, an isotropic LC/prepolymer mixture is deposited. Following the thickness control and the planarization of the mixture, the other plastic susbtrate with ITO is laminated by using roll pressing. Two plastic substrates are strongly attached to each other by the phase-separated thin polymer layer after LiV irradiation. Finally, plastic LCDs are simply prepared by cutting to a desired size. Our sequential process is suiable for cost-effective mass production of flexible LCDs.

Acknowledgments

This research was supported in part by Samsung Co., Ltd., and a grant (F0004052) from the Information Display R&D center, one of the 21st century Frontier R&D programs funded by the Ministry of Commerce, Industry, and Energy of Korean government.

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