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Single glass substrate LCDs using a phase separated composite organic film method

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Abstract

We review recent developments in the use of the phase separated composite organic film method and the fabrication of liquid crystal (LC) electro-optical devices using a single glass substrate using this method. The LC layer is confined between a film of solidified polymer layer on one side and the glass substrate on the other. The solidified polymer layer adjacent to and parallel to the LC layer is created by UV induced phase separation of a mixture of LC and prepolymer. Electro-optical properties of these devices demonstrate their high technological potential in light weight and hand-held electronic products.

Keywords: Liquid crystal display; Single glass substrate; Composite system; Phase separation

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1. Introduction

Conventional liquid crystal (LC) electro-optical (EO) devices, such as flat panel displays, are prepared by sandwiching the LC between two glass substrates coated with transparent indium-tin-oxide (ITO) electrode pattern with an overlay of rubbed polymer alignment layer to facilitate alignment of the LC's optical axis in a predetermined configuration. One of primary role of the substrates is to support LC molecular orientation and shield it from external influences such as mechanical bending or compression, which alter the thickness of the LC layer and arrangement of LC molecules in a manner that is detrimental to optical properties and performance of such devices. An electric field is normally applied across the electrodes on opposing sides of the LC layer to change the direction of the optic axis and to alter the optical path length in different regions of the devices. Various LC modes, such as twisted nematic, vertically aligned, in-plane switching modes, and ferro- and antiferro-electric LC mode, have been developed. In these conventional LC modes, the use of two substrates is imperative because of the fluid nature of LCs. There has been a considerable effort in recent years to replace glass by plastic films to make devices that are thin, light weight, and flexible. These are essential requirements for hand-held electronic products such as smart cards, PDA, and mobile phone [1].

In the past 20 years, several techniques of preparing dispersions of microscopic LC droplets in a polymer matrix have been developed [2–4]. These polymer dispersed liquid crystal (PDLC) devices operate in the scattering mode, in which an applied electric field controls the extent of the light scattered caused by a mismatch of refractive indices at the droplet boundary. PDLC structures are the result of isotropic and relatively fast phase separation. In 1999, a method to fabricate phase separated composite films (PSCOFs) of LC and polymer using anisotropic phase separation was developed by Vorflusev and Kumar [5,6]. The rate of phase separation in this method is controlled and deliberately kept low to allow the system to undergo a complete phase separation into regions of nearly pure LC and solid polymer. The PSCOF method has been used to prepare multi-layers and other complex geometrical structures. The operation of such PSCOF devices exploits the birefringence of the LC and relies on changes in the direction of the LC's optic axis in response to an applied electric field as in conventional displays. Since the LC is naturally confined

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between a glass substrates and the phase separated polymer layer, the PSCOF method lends itself to building devices with a single (glass or plastic) substrate [7,8].

Here, we review the PSCOF based method for the fabrication of a single substrate nematic device. The single substrate used in these devices is imprinted with electrodes to permit application of in-plane electric field.

2. Experimental

The method used to prepare PSCOF structures between glass substrates is essentially the same as used for building PDLC and polymer stabilized cholesteric textures (PSCT) devices. One starts with a pair of substrates coated with ITO. One of the substrates is spin-coated with specific alignment layers, such RN-1286 (polyimide), Nylon-6 (N6) or polyvinyl alcohol (PVA), which is unidirectionally rubbed to achieve homogeneous LC alignment and induce anisotropic phase separation during UV exposure. The second substrate is left untreated. They are separated by commonly used glass rod or bead spacers. The materials used in this study are commercially available nematic LC (NLC) E48 from Merck, ferroelectric LC (FLC) Felix 15-100 from Hoechst, and UV curable optical adhesive prepolymers NOA65, 72 and 73 from Norland, and SK 9 from Summers laboratories. Prepolymers are mixtures of monomer, oligomer, and photo-initiator. The prepolymer and the LC are mixed in a ratio ranging from 20:80 to as high 60:40 and introduced in to the cell by capillary action typically at a temperature well above the clearing point of the LC. Phase separation is initiated by exposing the cell to UV light through the substrate without the alignment layer. The source of UV light is a high pressure mercury vapor lamp operated at 200 W of electrical power. The cell is exposed to a collimated beam of UV light for approximately 2–10 min depending on the device to be made and its application. Phase separation results in a solidified film of polymer on the substrate closer to the UV source and an LC film between the polymer film and the second substrate as shown in Fig. 1.

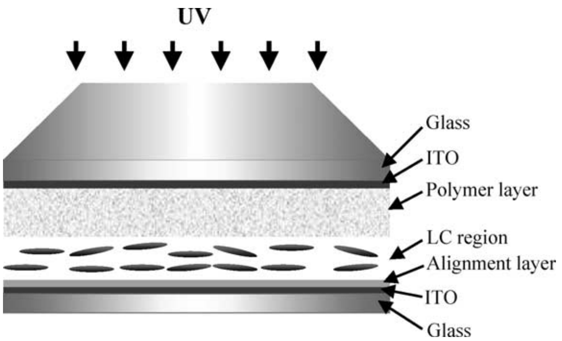

Fig. 1. Schematic representation of the UV exposure process to initiate polymerization and phase separation in mixtures of LC and prepolymer resulting in PSCOF structure.

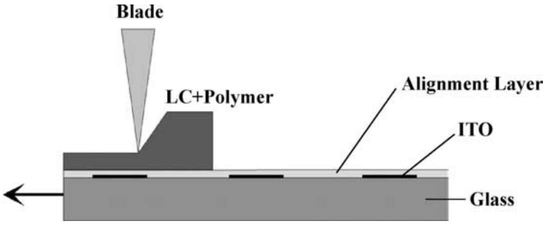

Fig. 2. Schematic illustration of the fabrication process for LCD with a single glass substrate.

In order to fabricate a LC device with a single glass substrate using the PSCOF method, in-plane electrodes are prepared by etching 100 μm wide interdigitated ITO strips on the glass substrate with a separation of 100 μm. The substrate is spin coated with 1 wt% N6 in trichloroethanol. The N6 film is unidirectionally rubbed after drying to achieve homogeneous LC alignment. To obtain an optical uniformity of the LC+prepolymer mixture, we use a two step process. First, the mixture is spread on the substrate using a steel blade, as shown in Fig. 2. The coating direction is kept parallel (or antiparallel) to the rubbing direction to avoid LC misalignment by shear flow. The thickness of the film is about 10 μm. The glass substrate is then spun for 30 s at 1500 rpm to increase the film uniformity. In the last stage, the phase separation is initiated, to obtain the PSCOF structure, by exposing cell directly to a collimated beam of UV light for approximately 60 min to fully cure the polymer.

3. Results and discussion

3.1. PSCOF structure

To determine the internal PSCOF structure, several cells are opened and washed with a solvent to remove the LC an then examined under a scanning electron microscope (SEM). In Fig. 3, we compare the PSCOF sample with a conventional PDLC sample. In a PDLC sample, the LC is trapped in droplets that are dispersed in the polymer medium. However, in PSCOF samples (Fig. 3(a)), phase separation results in a solidified film of polymer and a liquid crystal film between the polymer film and the second substrate. The LC (nematic, FLC, or AFLC) in PSCOF sample acquires a homogeneous alignment due to the influence of the rubbed alignment layer on the adjacent substrate. The LC and polymer films are uniform except in regions where the polymer–LC interface bonds to the opposite substrate on sites such as the one marked with the arrow in Fig. 3(a). The size and the number of these bonding sites are affected by the concentration and chemical nature of the LC compound, the alignment layer, the rate of phase separation, and the cell thickness. The process can be optimized to reduce the average size (to ≤1 μm)

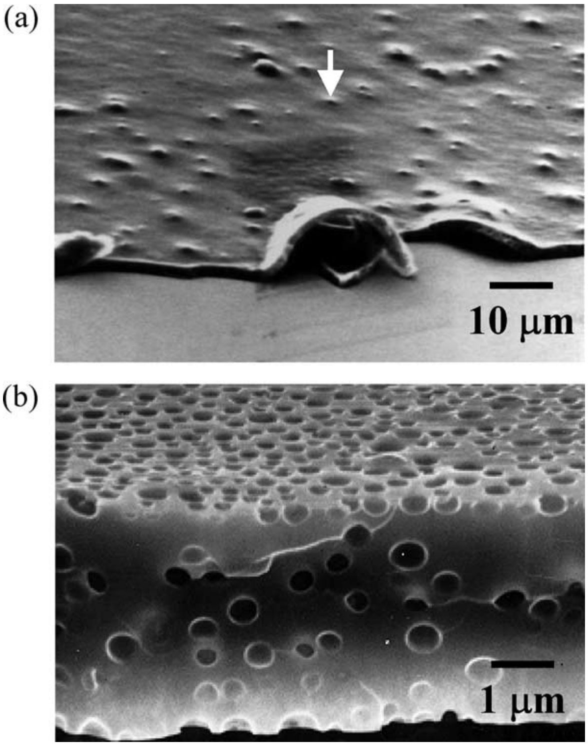

Fig. 3. Scanning electron micrographs of the polymer film deposited on one of the substrates, taken after washing away the LC in (a) PSCOF and (b) PDLC samples.

and control the number of these binding sites resulting in an almost perfectly uniform LC film.

The mechanism responsible for the formation of PSCOF is anisotropic polymerization [9]. A complete theory of the anisotropic phase separation by UV exposure would describe the evolution of the PILC structure in terms of spatial and temporal distribution of LC, prepolymer, polymer, and all the intermediate oligomers. In a previous report [10], a simple model was developed to describe the anisotropic phase separation due to the UV intensity gradient in the direction of sample thickness (*z*-axis) using mean-field kinetic theory. According to the model, the LC molecules are expelled from the polymerized volume causing them to move closer to the second substrate, towards the region of lower UV intensity. Droplet formation is inhibited because of relatively fast diffusion of the relatively small LC molecules.

The chemical nature of the alignment layer also influences the rate of phase separation and the quality of the polymer/LC films. The presence of an alignment layer, that LC molecules like to wet, enhances the uniformity of polymer layer even at slightly higher rate of phase separation. Fig. 4 shows contact angles of different prepolymers on N6 surface as a function of time after dropping and, on right hand side, alignment textures of E48 after phase separation. The variations in the measured contact angles lie within experimental errors. The contact angles of NOA 72 and 73 decrease and stabilize at $35 \pm 5°$ within 10 s. The contact angle of SK 9, however, stabilizes at $\sim 10°$. It means that the SK 9 has higher affinity for N6 than NOA 72 or 73. The LCs used in this study are found to the wet N6 alignment layer and dewet bare ITO surface. The interactions between the alignment layers and LC on one hand and prepolymer SK 9 on the other, compete inhibiting complete phase separation and resulting in polymer network structures upon UV exposure. However, when NOA 72 and 73 are used, the LC molecules near the alignment layer respond to its anchoring potential and align parallel to the rubbing direction. These oriented LC molecules determine the microscopic structure of the polymer–LC interface

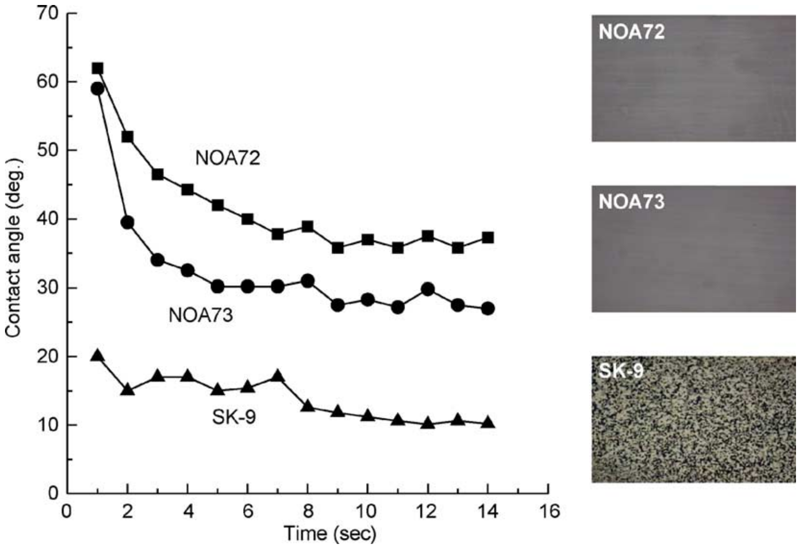

Fig. 4. Evolution of contact angle as function of time for different prepolymers on Nylon 6 after dropping and alignment textures with E48 after phase separation.

which becomes imprinted with the morphology of the rubbed alignment layer. The interface itself acquires aligning properties due to its physical structure. This goes to show that the presence of appropriate alignment layer can be used to promote anisotropic phase separation in the direction of sample thickness.

Measurements on PSCOF cells prepared with different concentrations of the LC show that the thickness of the LC layer depends directly on the concentration of LC in the LC–prepolymer mixture and that only a small amount (2–5%) of the LC is trapped in the polymer film. The isotropic to nematic transition temperature, of the LC after phase separation, deviates from that of the pure LC by less than 1 K. This small change is most likely the consequence of an increase in impurity concentration resulting from the expulsion of impurities, originally contained in the prepolymer, in to the LC volume.

The resultant structure depends on a number of parameters including concentration, the temperature of phase separation, spatial gradient in the rate of polymerization, diffusion coefficients of the LC, prepolymer, and wetting properties of various components. A generalized representation of the structures/morphologies that can result, is shown in Fig. 5. At low polymer concentrations, PSCT or cellular [11] structures are obtained. At higher polymer contents the result depends on the phase separation rate. For fast phase separation, PDLC/PDFLC are obtained. As the polymerization is slowed down, the droplets become larger and are formed closer to the substrate away from the UV source. At the slowest polymerization rates, the diffusion rates of LC and prepolymer determine whether columnar or film structure is obtained.

3.2. Advantages of the PSCOF structure

The PSCOF technology, because of its simple fabrication process and unique and a variety of possible internal structure, provides a number of advantages as discussed below.

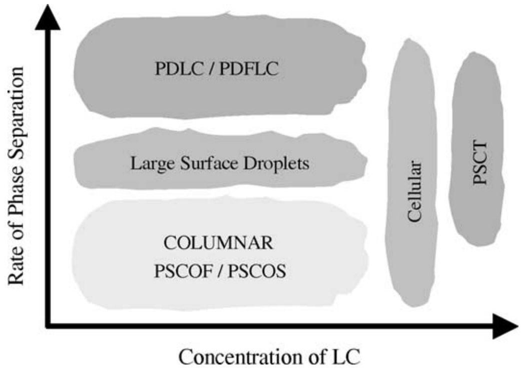

Fig. 5. Phase diagram for polymer+LC mixtures, qualitatively showing how the results of phase separation depend on the concentration and the rate of phase separation.

3.2.1. Mechanical ruggedness and flexible devices

The polymer–LC interface in PSCOF binds to the substrate on the other side of the LC at a large number of randomly distributed but microscopic points thus providing it rigidity and strength and thereby decreasing the sensitivity to external mechanical deformations without compromising performance. Mechanical deformations are known to render the surface stabilized FLC devices impractical to use and have been one of the major obstacles to their commercialization [12]. Under a large local pressure, PSCOF cells exhibit temporary changes in thickness. Their thickness reverts back to the original value within 5–15 s after the pressure is released. Because of the internal structure, where the two substrates are bonded to each other by polymer as discussed above, PSCOF method lends itself to the fabrication of cells using plastic substrates that are very flexible. Measurements on such a flexible 5 cm long cells, deformed in the middle by 1 cm to form an arc, reveal no internal changes in the director configuration showing that PSCOF devices are ideally suited for flexible displays [13]. Fig. 6 shows a comparison of the characteristics of the plastic PSCOF cell, without and with 40% bending, with a glass cell. The change observed in transmission levels upon bending is due to the fact that the cells' local optic axis was now on a curved surface. The transmission levels return to original levels when the pressure is released. We have

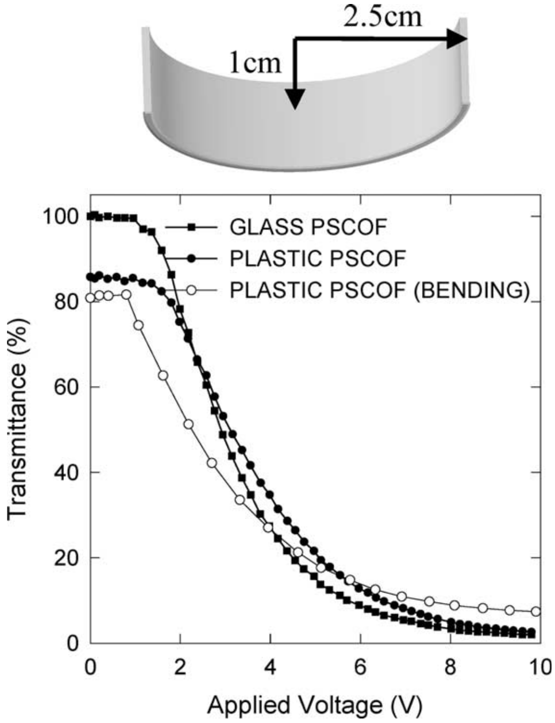

Fig. 6. Transmittance curves for PSCOF cells with glass substrate, plastic substrate, and plastic substrate under bending. Square wave of 1 kHz is applied to obtain these characteristic curves.

made similar cells with nematic and (quite remarkably) ferroelectric LCs, all able to withstand the mechanical deformation.

3.2.2. Modification of the electro-optical behavior

Additional major advantage of the PSCOF emerges from the modification of electro-optical behavior of ferroelectric and antiferroelectric LCs [5]. For example, the surface stabilized FLC cells exhibit bistability and strongly field dependent switching times. In a PSCOF device containing the liquid crystal Felix 15-100, switching at low fields is two orders of magnitude faster than in SSFLC (Fig. 7(a)). Also, the switching angle, which is the difference in molecular orientations in the ‘on’ and ‘off’ states, is found to continuously depend on the applied field (Fig. 7(b)) providing these devices with in intrinsic grey scale.

3.2.3. Active photonic devices

PSCOF also has been employed to fabricate active photonic devices, such as microlens array and one- and two-dimensional diffraction gratings, by affecting anisotropic phase separation in two and three dimensions with the help of photo-mask [14]. The presence of photo-mask produces additional intensity gradients in the plane perpendicular to z-axis (i.e. xy plane). Since the spatially periodic polymerization causes a spatially periodic chemical potential of the LC, a diffusive flow of LC from the region of high chemical potential to low chemical potential occurs until equilibrium is restored. Therefore, the phase separation occurs not only in the direction perpendicular to substrate but also in directions parallel to x and y directions. For example, the use of a surface relief array of hemispheres as a photo-mask results in microles array of (nematic or ferroelectric) LCs. Fig. 8 shows that schematic diagram of a nematic microlens, its focusing of a beam of light, and its microscopic view. The focal length varies with the applied voltage as well as the LC concentration, and the sample thickness.

3.3. Single glass substrate LCDs

LCDs with a single glass substrate can be fabricated with the PSCOF method as described in the Section 2. As shown schematically in Fig. 9, the LC layer is confined between the glass substrate and the solidified polymer layer which replaces the second glass substrate in conventional cells. The LC acquires a homogeneous alignment under the influence of the rubbed alignment layer on the substrate. Thickness of the LC layer mainly depends on the concentration and the thickness of the film of the mixture. The LC and polymer films obtained are uniform. It is important to note that such PSCOF devices can also be made using flexible plastic substrates.

To determine the internal structure of these devices, side-view of one of the cells was obtained under a scanning electron microscope (SEM). A 3 μm thick solidified film of polymer, Fig. 10, is formed on top of the LC. Since the prepolymer and the LC were mixed in 1:1 ratio, the thickness of LC layer for this device is estimated to be ∼3 μm.

Fig. 11 shows the microscopic and macroscopic textures under polarizing microscope with/without an applied voltages. With zero applied voltage, the uniform dark state is achieved due to good LC alignment (Fig. 11(a)). The small number of defects visible as faint spots in the photograph are due to nonuniform mixing of the LC and prepolymer (Fig. 11(b)). Above certain field strength (0.2 V/μm), the LC molecules start to reorient and align along the electric field due to their positive dielectric anisotropy. At higher field strengths (>1.5 V/μm), one can obtain the white state in which the LC molecules have rotated by 45° with respect to the rubbing direction (Fig. 11(c)). These results demonstrate that single glass substrate devices are uniform and possess gray scale capability. Fig. 11(d) shows a 1.5 cm×2 cm cell between crossed polarizers with an applied field of 1.5 V/μm. Except for a small area enclosed by the circle, the whole sample is in the uniform white state. The darker area is caused by non-uniformity of the LC and prepolymer coat.

Fig. 12 compares the field dependence of optical transmission of a one substrate device (PSCOF_ONE) with

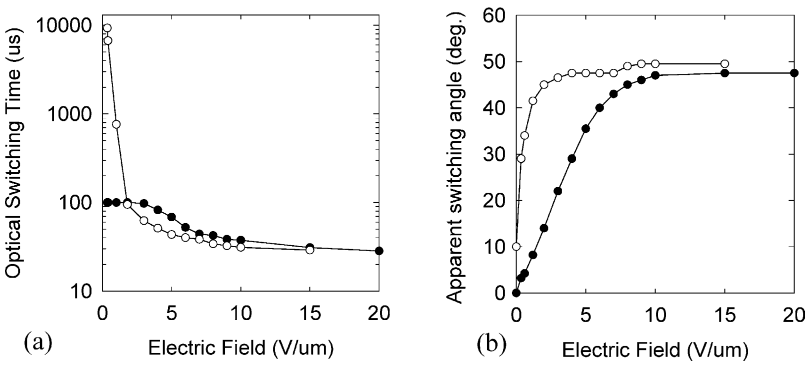

Fig. 7. Comparison of electro-optical properties of surface stabilized FLC (○) and PSCOF (●) at room temperature. (a) Optical switching times and (b) apparent switching angle versus the amplitude of the square-wave electric field measured with 40 wt% of the LC Felix 15-100 for PSCOF.

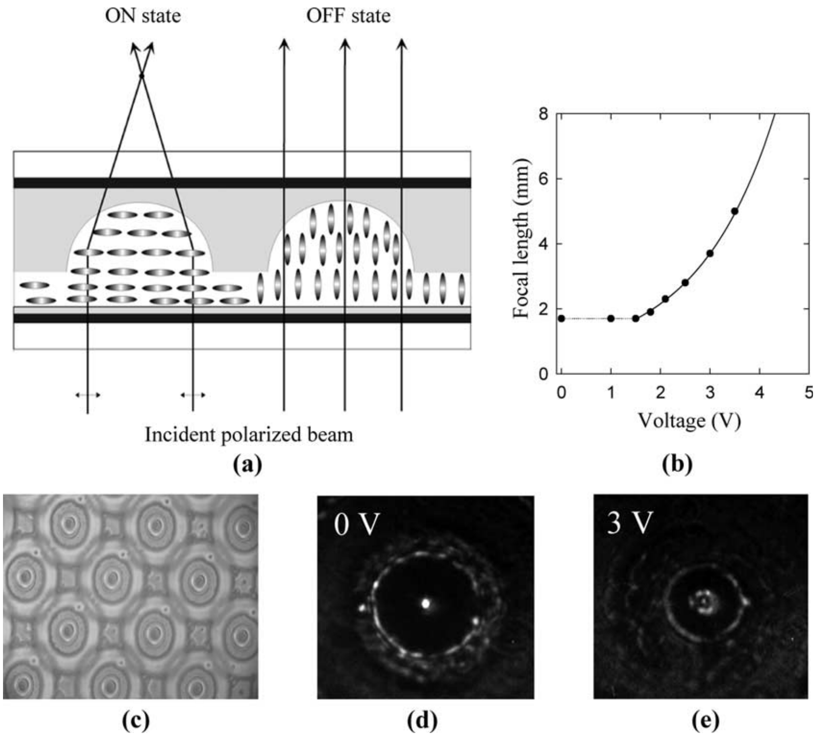

Fig. 8. Nematic microlens array: (a) schematic diagram of microlens, (b) dependence of the microlens' focal length on voltage, (c) microscopic textures of a microlens array under a polarizing microscope with no voltage applied. Concentric rings of different colors signify changing optical thicknesses, and areas of uniform shading outside the lenses indicate cell uniformity. (d) A focussed laser beam through the lens as imageed at 1.7 mm (the focal point) with no voltage applied, (e) defocussed beam with 3 V imaged at a distance of 1.7 mm.

a conventional two substrate (LC_TWO) and a two substrate PSCOF (PSCOF_TWO) cell. All devices were operated in the in-plane switching mode. The two-substrate cells show almost the same behavior. Their transmittance begins to increase at a field of about 0.2 V/μm, and reach its maximum value at 0.8 V/μm. In contrast, transmission through the PSCOF_ONE cell reaches saturation at 1.5 V/μm. It is possible to reduce the driving voltage by optimizing the concentration, dielectric anisotropy of LC, overall cell gap, and the electrode pattern. The maximum contrast of the one-glass sample is about 200:1 which is comparable to normal IPS sample. Fig. 13 shows the switching behavior of the sample. The field driven and relaxation times are 7.8 and 20 ms at 1.5 V/mm, respectively. The cell exhibits good switching characteristics at all gray levels.

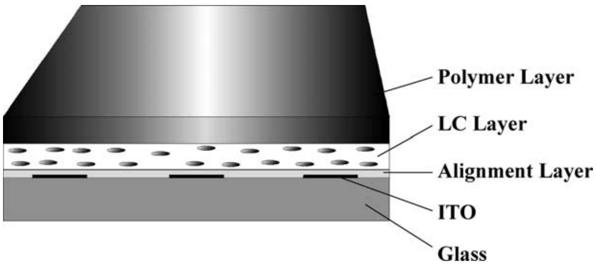

Fig. 9. Schematic diagram of fabricated LC device with a single glass substrate.

4. Concluding remarks

We have reviewed the recently developed phase separated composite organic film method and fabrication of liquid crystal based electro-optical device with a single glass substrate using the method. The PSCOF technology has advantages in the ease of fabrication, mechanical ruggedness and flexibility, and high resistivity. Moreover the active photonic devices, such as microlens array and diffraction gratings, can be easily fabricated using suitable photo-mask

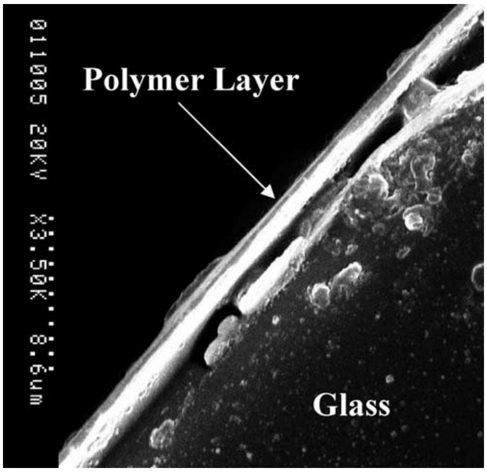

Fig. 10. SEM image showing the glass covered with the phase separated polymer overlayer.

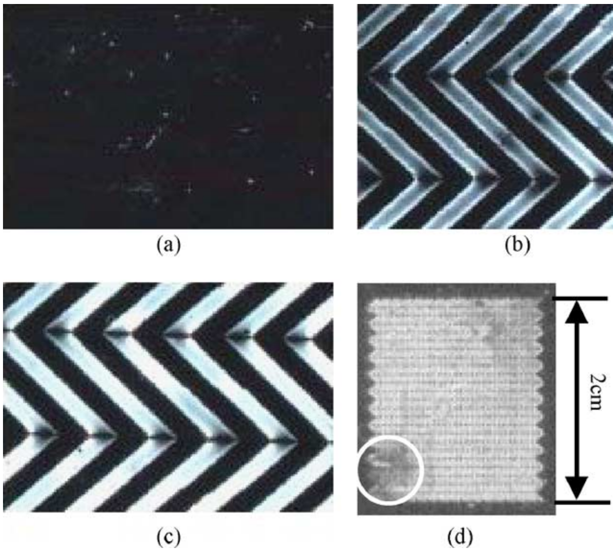
(a) (b)
(c) (d)

Fig. 11. Microscopic textures of one substrate device with zig-zag electrodes under polarizing microscope with an applied field of: (a) 0, (b) 0.7, and (c) 1.5 V/μm. (d) Photograph of 1.5 cm × 2 cm sample between crossed polarizers with 1.5 V/μm showing uniformity if the grey state. The area enclosed in the circle appears darker due to defects.

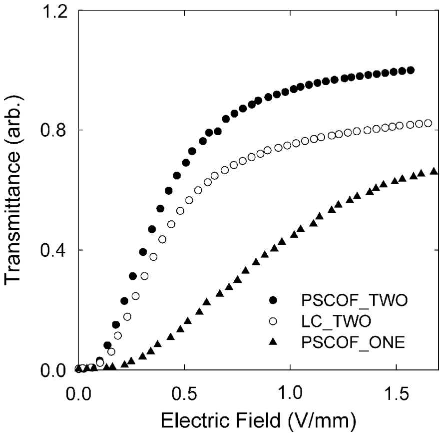

Fig. 12. Transmission vs. applied field for the one substrate (PSCOF_ONE), two-substrate (PSCOF_TWO), and normal IPS cell (LC_TWO).

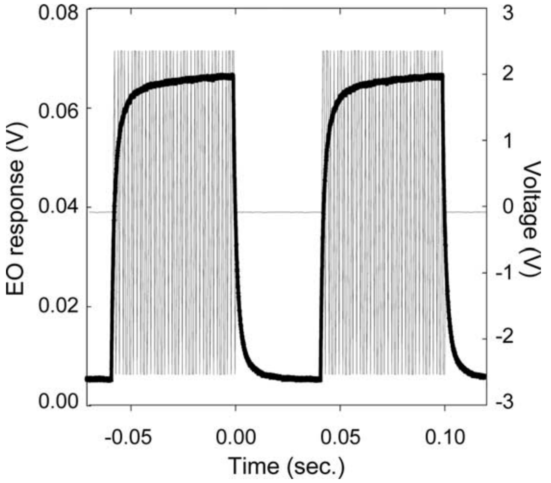

Fig. 13. Optical transmission and the switching waveform applied to a sample with a single glass substrate.

with PSCOF method. And one can also fabricate the LCDs with a single glass substrate. In these devices, the solidified polymer overlayer acts as the substrate. The electro-optical performance of these displays is comparable to the normal two glass substrates displays. The method demonstrated here opens the introduction of a new class of devices in to the market.

Acknowledgements

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