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## Liquid Crystals

Publication details, including instructions for authors and subscription information:

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### Optically isotropic liquid crystal mixtures and their application to high-performance liquid crystal devices

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Published online: 31 Mar 2015.

To cite this article: No Hyun Park, Seong Cheol Noh, Prasenjit Nayek, Myong-Hoon Lee, Min Su Kim, Liang-Chy Chien, Joun Hoo Lee, Byeong Koo Kim & Seung Hee Lee (2015) Optically isotropic liquid crystal mixtures and their application to high-performance liquid crystal devices, *Liquid Crystals*, 42:4, 530-536, DOI: [10.1080/02678292.2015.1006698](https://doi.org/10.1080/02678292.2015.1006698)

To link to this article: <http://dx.doi.org/10.1080/02678292.2015.1006698>

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## Optically isotropic liquid crystal mixtures and their application to high-performance liquid crystal devices

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(Received 23 December 2014; accepted 8 January 2015)

Optically isotropic liquid crystal (OILC) mixtures utilising polymerisation-induced phase separation in mixtures of prepolymers and liquid crystals (LCs) have been made and their overall electro-optic performances have been tested by applying an in-plane electric field for flexible display application. Excellent transparent films with a LC to prepolymer weight ratio of 42:58 are achieved and confirmed by SEM images. The average LC droplet size inside the polymer matrix is 215 nm. Consequently, an in-plane driven LC device with a crossed polariser shows a very high contrast ratio over 1000 comparable to the conventional nematic in-plane switching cell, wide viewing angle, a low threshold voltage of 8.2 V, hysteresis free and fast response time. The proposed LC device is free from an alignment layer and the rubbing process and its dark state is not disturbed by mechanical pressure, which can open its application to flexible liquid crystal displays.

**Keywords:** optically isotropic liquid crystals; Kerr effect; in-plane field; flexible LCD

### 1. Introduction

Recently, flexible displays have been accelerating commercialisation of the technology. In order to maintain the top most position in the display market, liquid crystal displays (LCDs) have to adopt new display modes suitable for flexible LCDs in an indispensable way. Liquid crystals (LCs) possess fluidic properties that cause the deformation of LC thickness as well as orientation under a bent display, resulting in a change in electro-optic characteristics of the display. In other words, LCDs have intrinsic difficulty to be flexible devices.

As one of the promising candidates, optically isotropic liquid crystal (OILC) films in which nano-sized LCs are dispersed in a polymer matrix have been studied.[1–4] The films are made using the polymerisation induced phase separation (PIPS) method.[5] Controlling the ratio of the polymer to LC concentration and UV exposure intensity, the droplet size can be reduced to smaller than the wavelength of visible light, and then optically isotropic LCs are formed. Previous works reported on how to achieve the OILC films, but their electro-optic performances in viewpoints of display applications were not revealed in detail.[2,6–8] The in-plane field driven LC device with the OILC films was found to consist of submicron-sized nematic LC domains by the PIPS method [9,10] and encapsulation of LCs via emulsion [11] has advantages of

rubbing free, wide-viewing-angle and fast response time, except high operating voltage.

In this article, we have investigated on how to achieve an OILC film via the PIPS method using a LC with high dielectric anisotropy and high birefringence for reducing an operating voltage and compared its electro-optic characteristics with those of conventional LC devices using nematic LCs only, exploring its application possibility to flexible LCDs.

### 2. Switching principle of OILC films with the Kerr effect

The OILC film is optically isotropic, so that the device appears to be black under a crossed polariser in the absence of an electric field. With an applied in-plane field, a birefringence  $\Delta n$  is induced along the field direction, so that the transmittance starts to appear. In the OILC film, the induced birefringence  $\Delta n_{\text{ind}}$  follows the expression [12]:

$$\Delta n_{\text{ind}} = \lambda K E^2 \quad (1)$$

where  $\lambda$  is the wavelength of an incident light,  $K$  is the Kerr constant of the film and  $E$  is the applied electric field. Therefore, in order for a device to be operated in a low voltage,  $K$  of the film should be as large as possible. On the other hand, the threshold field  $E_{\text{th}}$  in

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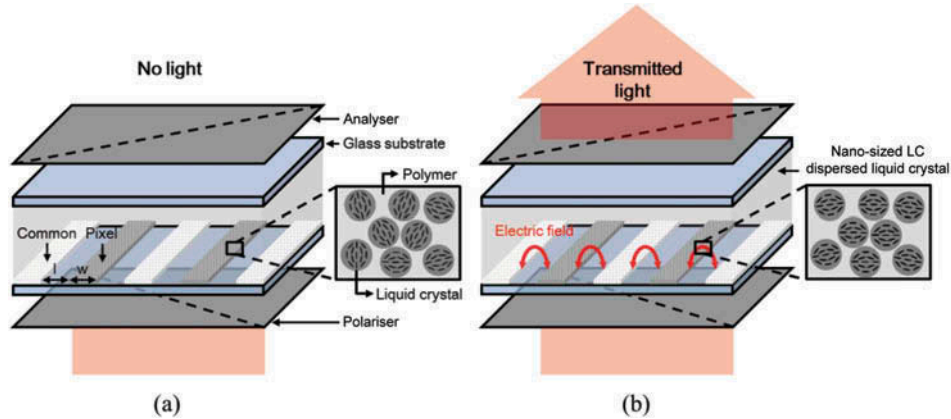


Figure 1. (colour online) Schematic diagram of the cell structure and orientation of LC molecules in the (a) voltage-off state (optically isotropic state), and (b) voltage-on state (optically anisotropic state).

an in-plane switching device [13] follows the expression in which the cell gap ( $d$ ) is replaced by the diameter ( $R$ ) of LC droplets in the OILC film:

$$E_{\text{th}} = \frac{\pi}{R} \left( \frac{K_{\text{LC}}}{\varepsilon_0 \Delta \varepsilon} \right)^{\frac{1}{2}} \text{ or } \left( \frac{\Delta n_{\text{ind}}}{\lambda K} \right)^{\frac{1}{2}} \quad (2)$$

where  $K_{\text{LC}}$  and  $\Delta \varepsilon$  are the elastic constant and dielectric anisotropy of a LC, respectively. By substituting  $E_{\text{th}}$  into  $E$  in Equation (1), we can obtain a proportional expression related to  $K$  with physical properties of a LC and the droplet size of a LC in a polymer matrix as follows:

$$K \propto \Delta n_{\text{ind}} \Delta \varepsilon \frac{\varepsilon_0 R^2}{K_{\text{LC}} \lambda \pi^2} \quad (3)$$

As clearly indicated above, a LC with high birefringence and dielectric anisotropy with low elastic constant and large size of a LC droplet must be used to maximise  $K$ . However, when  $\Delta n$  of a LC is larger than 0.3 at 550 nm and the size of the LC droplet reaches 400 nm close to the visible wavelength, the OILC film causes scattering of the visible light, which will deteriorate the dark state and following the contrast ratio of the device because an average scattering cross-section coming from refractive index mismatch between LC droplets and the polymer matrix is related to the following Equation (4):

$$\sigma_{\text{avg}} \propto k^4 R^6 \quad (4)$$

where  $k = 2\pi n_p / \lambda$ . [14,15] Therefore,  $R$  should be as smaller as possible to minimise any scattering, which depolarises an incident linearly polarised light. Consequently, there are clear limitations controlling

birefringence and LC droplet size, so that a LC with high dielectric anisotropy must be utilised to lower an operating voltage. In this work, two different LCs with different  $\Delta \varepsilon$  are evaluated to improve electro-optic properties of the device with OILC films and their suitability to flexible displays is compared with those of conventional nematic LC devices.

Figure 1 schematically depicts the operation principle of the LC device with OILC mixtures. [16] In an electric field off state, LC molecules within nano-size (less than 300 nm) of LC droplets in a polymer matrix are randomly oriented, so that the cell exhibits an optically isotropic state in the visible light and it appears to be dark under a crossed polariser, as shown in Figure 1 (a). When an in-plane field is applied, LC molecules reorient along the field direction due to the Kerr effect, so that an optical birefringence is induced, resulting in a bright state, as shown in Figure 1(b).

### 3. Experimental results and discussion

In order to prepare the OILC film, a LC with relatively high dielectric anisotropy ( $\Delta \varepsilon = 42.6$ ), MLC-2053 from Merck Advanced Technology in Korea, was used and also the eutectic nematic mixture E7 with  $\Delta \varepsilon = 14.4$  was tested for a comparison.  $\Delta n$  of E7 and MLC-2053 was 0.217 and 0.235 at 589 nm, respectively. The ultraviolet (UV) curable optical adhesive Norland optical adhesive 65 (NOA65, USA) was used as a prepolymer. A photoinitiator, Irgacure-651 (Merck) with 1 wt%, was added to complete the polymerisation of the mixture with a proper LC droplet size. Using capillary force, the prepared mixture was injected into an in-plane field driven cell at a clearing temperature. UV light with an intensity of 150 mW/cm<sup>2</sup> at 365 nm was irradiated for 5 s at room temperature. The in-plane field cell has an

electrode width of 4  $\mu\text{m}$  and a distance of 4  $\mu\text{m}$  between signal and common electrodes. The cell gap was 10  $\mu\text{m}$ . After UV irradiation, the film was transparent and its electro-optic properties of the cells were investigated between crossed polarisers using a custom-made set up consisting of a photo-detector, oscilloscope (Agilent 33521A), laser source (633 nm), amplifier (FLC A400) and a function generator (Tektronix DPO 2024B).

We have prepared several mixtures using both LC and NOA65 with different weight percent ratios and their dark state has been observed as can be seen in polarising optical microscopic (POM, Nikon eclipse E600 POL) images in Figure 2. At first, the relative value of light leakage intensity in six mixtures with different LC wt% ratios was evaluated and it was measured to be 9, 29, and 63 when MLC-2053 was 40, 45, and 55 wt%, respectively, while it was 8, 13, and 31 when E7 was 40, 45, and 55 wt%, respectively. In addition, one more mixture using 42 wt% of MLC-2053 was prepared and its light leakage was 12. In all mixtures, the higher the prepolymer ratio, the better transparent OILC film with less scattering was obtained. In other words, the scattering increased with increasing LC amount, resulting in a light leakage in a dark state, in which the increased scattering was expected to originate from the increased LC amount, which made LC droplets larger, according

to Equation (4). Therefore, higher content of LC in a prepolymer is preferred to minimise the operating voltage, but at the same time the film should be as much transparent as possible not to depolarise an incident linearly polarised light to achieve a perfect dark state. In terms of these viewpoints, the mixture of the LC and prepolymer with a weight percent ratio of 42:58 (MLC-2053) and 45:55 (E7) was an optimal condition in our studies. Under the optimal condition, we confirmed a clear switching from a dark to a bright state with bias voltage for MLC-2053, in which a uniform bright state over a whole area was obtained, indicating that the birefringence was induced uniformly in the OILC film by the Kerr effect as shown in Figure 2(h).

In order to judge the dark quality of the film, cells with vertical alignment (VA) and homogenous alignment (HA) with interdigitated electrodes were made and their dark levels are compared in the POM as shown in Figure 3. The relative value of light intensity of a dark state in VA, HA, and OILC cells was 3, 12, and 12, respectively, indicating that the dark level is quite much comparable to that of the HA cell although the VA and HA cells used a LC with much smaller birefringence ( $\Delta n < 1$ ).

Next, the dark states of VA and OILC cells were compared again not only at normal but also oblique directions, as shown in Figure 5. In the VA cell, the

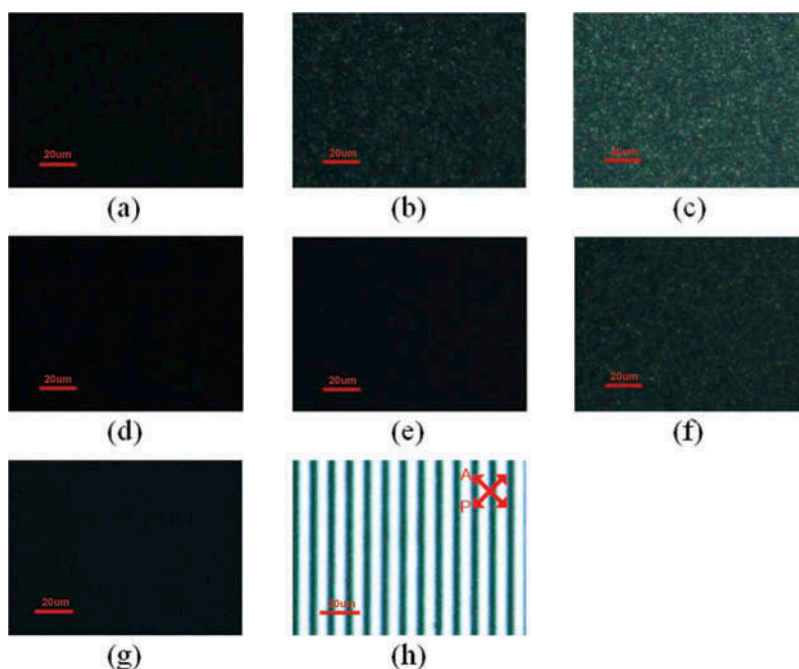


Figure 2. (colour online) POM images of the mixtures with different weight percents of the LC in the prepolymer: MLC-2053 with (a) 40, (b) 45, and (c) 55, and E7 with (d) 40, (e) 45, and (f) 55. POM images of (g) and (h) indicate a dark and a bright state in the mixture with MLC-2053 wt% of 42.

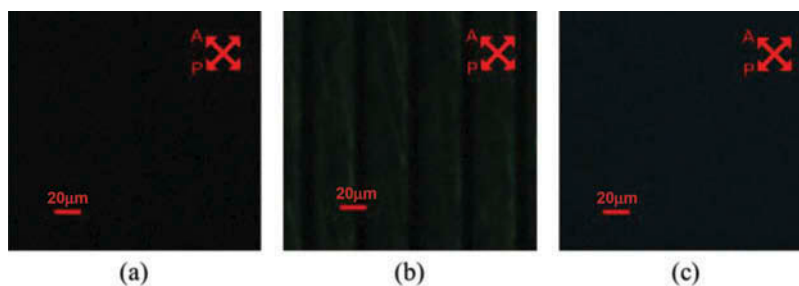


Figure 3. (colour online) The dark state images of (a) VA cell, (b) IPS cell and (c) OILC cell, at a normal direction.

optic axis of LC is vertically aligned, so that a conoscopic image shows a LC texture with crossed dark lines, proving vertical alignment of LC whereas the OILC shows just a dark state because there is no optic axis, as shown in Figure 4(a) and 4(b), respectively. When both cells are viewed at an off normal axis especially at an azimuthal angle of  $45^\circ$  with respect to the crossed-polariser axes, a strong light leakage does occur in the VA cell while a relatively good dark state is kept in the OILC cell, as shown in Figure 4(c) and 4(d), respectively. In general, the VA cell requires an optical compensation film to suppress the light leakage at the off normal direction [17]; however, the OILC cell does not require any compensation film because all viewing directions can yield a good dark state, which is a strong advantage of low cost and flexible displays because either bending or curving of a LCD will cause a change in retardation of the optical compensation films, resulting in distortion of the displayed image. Next, we applied mechanical pressure to both cells using a pen, testing so-called touch mura. In the VA cell, fluidic nematic LCs were vertically aligned, so that the external pressure distorted the orientation of LCs, generating a strong light leakage, whereas the OILC cell exhibited no distortion of a dark state at all at the same external pressure because the LC in each droplet is surrounded by a polymer matrix, and also the LC in droplets might be less ordered than that in the VA cell in a dark state, as shown in Figure 4(e) and 4(f).

Once we confirmed voltage-off and -on switching and a dark state of the OILC cell, voltage-dependent transmittance (V-T) was measured by applying a square wave with 1 kHz. As presented in Figure 5, V-T curves for the cell with MLC-2053 were much shifted to the left compared to that with E7, showing that both threshold ( $V_{th}$ ) and an operating voltage ( $V_{op}$ ) at which the maximum transmittance was much lower with MLC-2053 than with E7.  $V_{th}$  and the voltage of 42 wt% MLC-2053 at which the transmittance is 10% and 90% with respect to the maximum is 8.2 and 46 V, respectively, while those of E7 are 12 and 55 V, respectively.

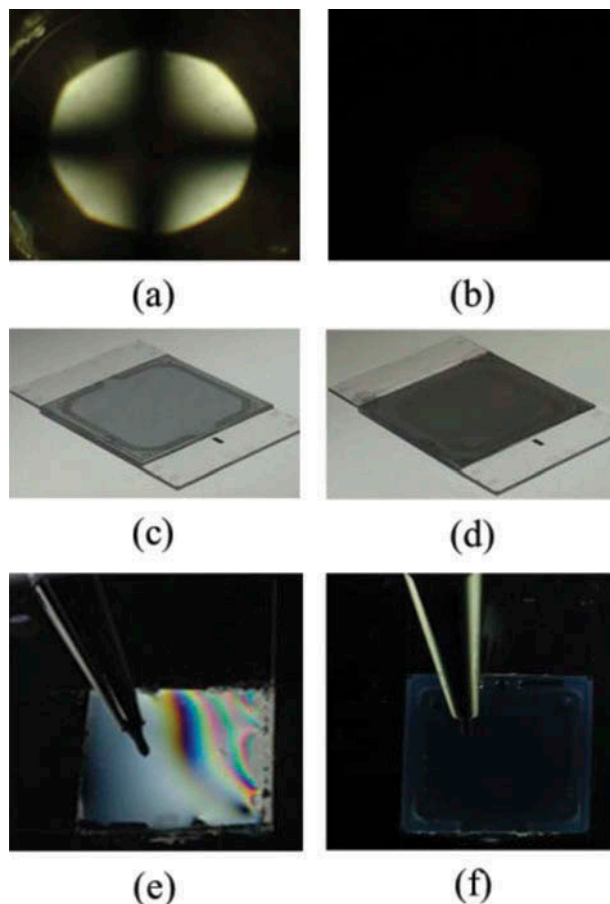


Figure 4. (colour online) Conoscopic images of (a) VA cell and (b) OILC cell, and comparison of light leakage in an oblique viewing direction at an azimuthal polar angle of  $45^\circ$  with respect to polariser axes: (c) VA cell and (d) OILC cell. Comparison of touch-mura images: (e) VA cell and (f) OILC cell.

As clearly understood from Equation (2), an operating or threshold field is proportional to  $\frac{1}{\sqrt{\Delta\epsilon}}$  in a device with nematic LC only and  $\frac{1}{\sqrt{K}}$  for the OILC film, and thus the drop in the threshold voltage in MLC-2053 is clearly associated with the large value of  $\Delta\epsilon$  and also large  $K$ . On the other hand,  $K$  is linearly proportional to the product of  $\Delta\epsilon$  and  $\Delta n$ ,

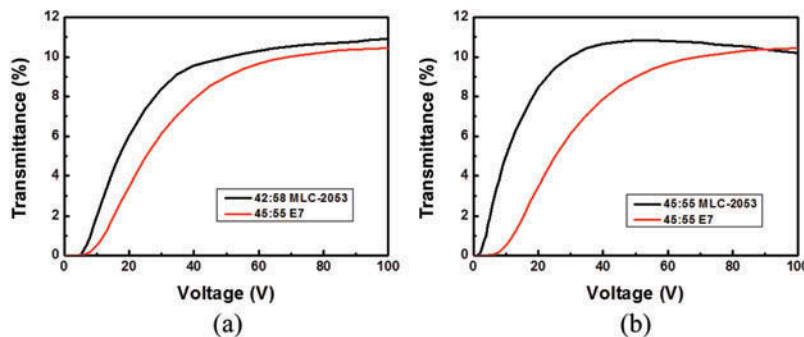


Figure 5. (colour online) Voltage-dependent transmittance curves of the OILC cells using MLC-2053 and E7. Here, two different mixtures of MLC-2053 with (a) 42 wt% and (b) 45 wt% are compared. When wt% of MLC-2053 is 42, the contrast ratio is 1880:1 and when it is 45, the contrast ratio drops to below 1000:1 due to light leakage associated with light scattering.

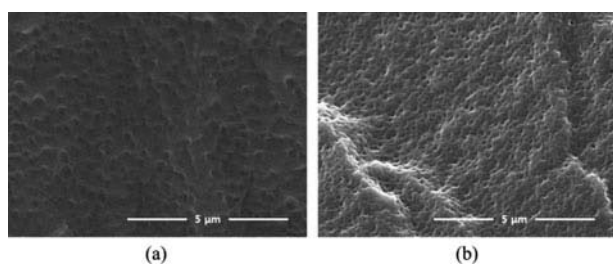


Figure 6. SEM image of holes associated with LC droplets in the polymer matrix: (a) MLC-2053 and (b) E7.

[18] so that  $K$  of MLC-2053 should be 3.2 times larger than that with E7. From the relationship between the in-plane field induced  $\Delta n$  and  $\lambda E^2$ ,  $K$  was evaluated through fittings considering only a low field region.[19] The calculated  $K$  was  $2.66 \times 10^{-9}$  and  $7.53 \times 10^{-10} \text{ mV}^{-2}$  for MLC-2053 and E7, respectively, indicating that  $K$  of MLC-2053 is about 3.5 times larger than that of E7, which is a slightly higher value than expected. In addition,  $K$  is proportional to  $R^2$ , so that we need to get information on the LC droplet size in a polymer matrix. Figure 6 shows scanning electron microscope (SEM) (Hitachi S-4700) images of the polymer matrix after the LC is washed in the composite using a solvent. The average diameter of the droplets in MLC-2053 and E7 is 215 and 194 nm, respectively. Therefore, taking into account  $R$ ,  $K$  of MLC-2053 should be about 3.9 times of that of E7, which is in good agreement with the measured Kerr constant. Since  $V_{\text{th}}$  is proportional to  $\frac{1}{\sqrt{K}}$ , it should show about 6.1 V in MLC-2053, but the measured  $V_{\text{th}}$  is in slightly disagreement with the measured value of 8.1 V.

The response time of the OILC cells has been measured considering 80% transmittance change. In devices with nematic LC only, a rise time mainly depends on a rotational viscosity ( $\gamma_1$ ) of a LC and

applied voltage, and a decay time for the conventional nematic LC device is given by [20]:

$$\tau_{\text{off}} = \gamma_1 d^2 / \pi^2 K$$

In the OILC film,  $d$  can be replaced by  $R$  again. The measured rise times by applying the voltage at which the maximum transmittance occurs are 870 and 820  $\mu\text{s}$ , and the decay times are 4.5 ms and 940  $\mu\text{s}$  for MLC-2053 and E7, respectively. The decay time of MLC-2053 is about 4.8 times slower than that of E7, unfortunately. In the OILC film,  $R$  of MLC-2053 is about 1.1 times larger than that of E7 and the viscosity of MLC-2053 is about 3.5 larger than that of E7, so that the decay time of MLC-2053 should be about 4.2 times slower than E7, which is slightly in disagreement with experimental measured data, which might be associated with the difference in elastic constants between two LCs and minor error in measuring the droplet size. In the OILC cell, a LC with high dielectric anisotropy for lowering driving voltage but at the same low rotational viscosity for a short decay time needs to be developed.

One of the most important requirements for the OILC mixtures such as polymer stabilised blue phase (PSBP) and OILC to be applied to thin-film-transistor LCDs is that the device should be free of hysteresis. In a PSBP-LCD, an applied voltage causes electrostriction such that a cubic structure of a double-twisted cylinder is deformed although the disclination is assumed to be fixed by polymer networks, exhibiting a large hysteresis with 3% to 6%.[21–23] Figure 7 compared hysteresis for two OILC cells with MLC-2053 and E7 by applying a voltage to the maximum transmittance and decreasing the applied voltage to zero. We have defined the hysteresis as the voltage difference between increasing and decreasing voltages at 50% of the maximum

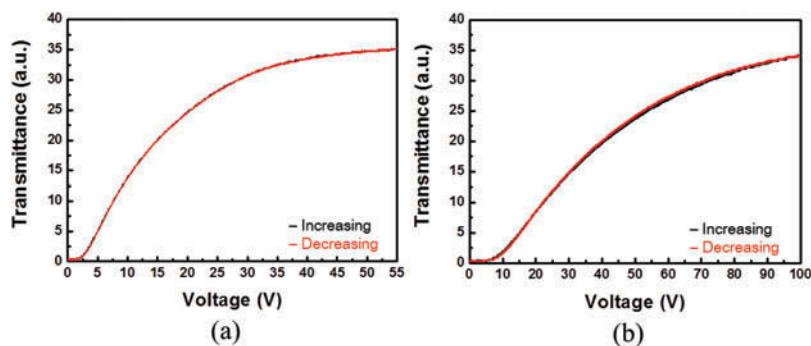


Figure 7. (colour online) Comparison of hysteresis of an optically isotropic LC composite: (a) MLC-2053 and (b) E7.

transmittance. The measured hysteresis was 0.4 and 0.5 V for OILC cells with MLC-2053 and E7, respectively, which was only 0.4% for MLC-2053 and 0.5% for E7 when calculated using the same method given in references.[24] The results support that the hysteresis of OILC cells is much lower than that of PSBP cells, because the field-induced LC orientation in the OILC cells is just associated with the transition from random ordering to ordering of LC along the field direction in LC droplets embedded in a polymer matrix.

#### 4. Conclusion

We proposed optically isotropic LC films comprising nano-sized LC droplets in a polymer matrix. By utilising a LC with high dielectric anisotropy, the proposed in-plane field driven device utilising the film has great advantages such as possessing a non-rubbing process without the requirement of an alignment layer, wide-viewing angle, being hysteresis free, touch mura free, possessing fast response time, low operating voltage with a threshold of 8.2 V and a contrast ratio comparable to conventional IPS cells. In particular, the alignment of the LC does not change under external forces, so that the proposed device has high potential to be applicable to flexible LCDs. Also, further optimisation and controlling droplet size of the composites with more number of high-performance LCs in the composite would improve electro-optic properties of OILC to yield sub-millisecond response time with proper operating voltage.

#### Funding

This study was supported by the Basic Science Research Program of National Research Foundation (NRF) of Korea [grant number 2013R1A1A2005630]; the NRF of

Korea grant funded by the Korean Government (MSIP) [grant number 2014R1A4A1008140]; LG displays.

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