

# Enhanced contrast ratio and viewing angle of polymer-stabilized liquid crystal via refractive index matching between liquid crystal and polymer network

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**Abstract:** Long standing electro-optic problems of a polymer-dispersed liquid crystal (PDLC) such as low contrast ratio and transmittances decrease in oblique viewing angle have been challenged with a mixture of dual frequency liquid crystal (DFLC) and reactive mesogen (RM). The DFCLC and RM molecules were vertically aligned and then photo-polymerized using a UV light. At scattering state under 50 kHz electric field, DFCLC was switched to planar state, giving greater extraordinary refractive index than the normal PDLC cell. Consequently, the scattering intensity and the contrast ratio were increased compared to the conventional PDLC cell. At transparent state under 1 kHz electric field, the extraordinary refractive index of DFCLC was simultaneously matched with the refractive index of vertically aligned RM so that the light scattering in oblique viewing angles was minimized, giving rise to high transmittance in all viewing angles.

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OCIS codes: (230.2090) Electro-optical devices; (230.3720) Liquid-crystal devices.

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## 1. Introduction

Polymer-dispersed liquid crystal (PDLC) has drawn much attention for various electro-optical devices such as a smart window, a display device, and a grating component [1–10]. Among the various methods to fabricate the PDLC cell [1,2], the polymerization-induced phase separation (PIPS) method has been widely used [3–10]. In the PIPS method, liquid crystal (LC) is mixed with reactive monomers, and then photo-polymerized using a UV light. The polymerized monomers are simultaneously phase-separated from LC due to the decreased miscibility with LC. Consequently, nano- and micro-sized LC domains are remained within the polymer matrix. The operation principle of conventional PDLC is shown in Fig. 1. We assumed bipolar orientation of LC molecules in the droplet. At zero field state [Fig. 1(a)], the bipolar axes of the LC droplets are randomly oriented and the refractive index of LC  $n_{LC}$  became the intermediate value between the extraordinary and ordinary refractive index,  $n_e$  and  $n_o$  respectively. Provided that the refractive index of the polymer  $n_p$  is similar to  $n_o$ , the incident light is scattered due to the mismatching between  $n_{LC}$  and  $n_p$ . Under an electric field [Fig. 1(b)], the bipolar axes are aligned parallel to the electric field and  $n_{LC}$  becomes  $n_o$ . Then, the incident light passes through the cell without scattering, i.e., the cell becomes transparent.

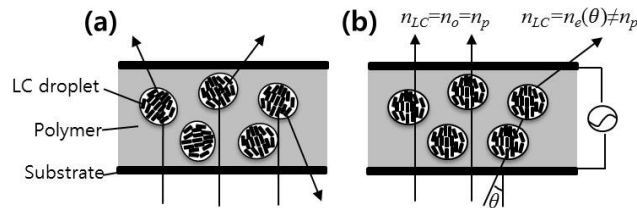


Fig. 1. Schematic illustration of (a) scattering and (b) transparent state of the conventional PDLC cell.

In spite of the simple fabrication process, there have been unsolved problems of PDLC for the optical application. First, the scattering effect [Fig. 1(a)] was imperfect because  $n_{LC}$  was less than  $n_e$  due to the random orientation of the bipolar axes [1]. Second, the viewing angle property of transparent state [Fig. 1(b)] was not good [11,12], i.e., transparent state was changed to scattering state for the oblique view due to the mismatching between  $n_e(\theta)$  and  $n_p$  [Fig. 1(b)], where  $\theta$  is the incident angle of the light and  $n_e(\theta)$  is given by Eq. (1),

$$n_e(\theta) = \left( \frac{\sin^2 \theta}{n_e^2} + \frac{\cos^2 \theta}{n_o^2} \right)^{-\frac{1}{2}} \quad (1)$$

In this paper, we proposed a new method to resolve these problems using a dual frequency liquid crystal (DFLC) whose sign of dielectric anisotropy inverts at the low and high frequency of electric field and a liquid crystalline photo-polymerizable monomer, often called as a reactive mesogen (RM). The mixture of DFLC and RM was injected into an empty cell whose substrates were coated with a vertical alignment polyimide. Then, the vertically aligned DFLC and RM were photo-polymerized using a UV light in presence of a low frequency electric field. The LC and polymerized RM were vertically oriented after the photo-polymerization. Under a high frequency electric field, DFLC was switched to planar state and  $n_e(\theta)$  of DFLC was maximized greater than that of the randomly aligned LC droplets in Fig. 1(a). Consequently, the scattering intensity was increased and the contrast ratio was improved compared to conventional PDLC. Under a low frequency of an electric field, the DFLC molecules maintained the homeotropic orientation and  $n_e(\theta)$  was simultaneously matched with

$n_p(\theta)$  so that the light scattering in oblique viewing angles was minimized, giving rise to high transmittance in all viewing angles.

There have been several reports of the polymer-stabilized liquid crystal (PSLC) modes using the DF LC and RM molecules [13,14]. Huang et al. fabricated a pi-cell DF LC-RM mixture under strong electric field and weak UV light [13]. A fast response and greater contrast of the pi-cell was achieved without a high pretilt surface layer or a refreshing electric field. Brimicombe et al. stabilized the bend orientation of the pi-cell using DF LC-RM mixtures and eliminated the nucleation time of the bend domains [14]. In these previous literatures, the transmittance was controlled by varying the in-plane birefringence using the electric field. On the other hand, we adapted the DF LC-RM mixture to improve the scattering-mode PSLC cell which operates without polarizers. The scattering intensity and viewing angle property of the suggested PSLC cell using the DF LC-RM mixture could be significantly enhanced compared to the conventional PDLC cell by the increased  $n_e$  of LC and the simultaneous matching between  $n_e(\theta)$  and  $n_p$  at all incident angles.

## 2. Experimental procedure

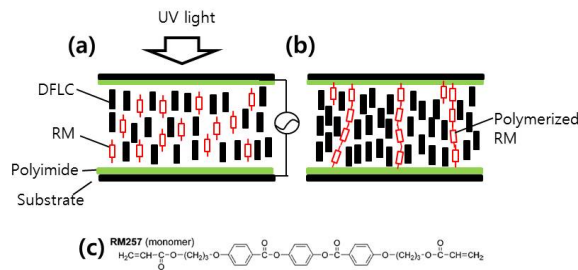


Fig. 2. Fabrication procedure of the proposed PSLC cell using a DF LC-RM mixture. (a) DF LC-RM mixture was injected into an empty cell and then UV-polymerized in presence of 1 kHz 2 V square voltage. (b) DF LC and polymerized RM were vertical aligned after UV exposure. (c) chemical structure of the RM molecule (RM257) used in the paper.

The indium-tin-oxide (ITO)-deposited glass substrate was coated with a vertical alignment polyimide (SE6514, Nissan). The substrate was then baked at 220 °C for 1 h and assembled together. Cell gap was maintained to be 10 μm using bead spacers. Commercially available DF LC (MDA-00-3969, Merck) was mixed with RM (RM257, Merck) at a weight ratio of 97:3 and 95:5. DF LC has a positive and a negative dielectric anisotropy at 1 and 50 kHz of electric field, respectively. The refractive indices of DF LC were  $n_e = 1.7192$  and  $n_o = 1.4978$ , which were similar to those of RM  $n_e = 1.687$  and  $n_o = 1.508$ . The DF LC-RM mixture was injected into the cell and cured using a UV light (365 nm) of 11.5 mW/cm<sup>2</sup> for 90 sec [Fig. 2(a)]. In the course of the photo-polymerization, 1 kHz 2 V bipolar voltage was applied across the cell. Consequently, the DF LC molecules were homeotropic aligned with the intersticed vertical RM polymers [Fig. 2(b)]. RM257 used in this study has a rod-like shape and phase sequence Cr 70 °C N 126 °C Iso. [Fig. 2(c)] To compare the electro-optical properties of the PSLC cell using the DF LC-RM mixture, we also prepared a normal PDLC cell using a commercial LC (CL-002, Q-sys) and non-mesogenic monomer (AT-002, Q-sys). 60 wt% monomer was mixed with LC and injected at room temperature (25°C). No alignment material was coated on the substrate. The cell was then exposed to the UV light of 15 mW/cm<sup>2</sup> for 30 s. To measure transmittance (TR) vs. voltage and  $\theta$ , a commercial optical measurement system LCMS200 (Sesim Photonics Technology) was used. A white light source consecutively passes through the cell, a convergent lens, and a detector, and the intensity was measured as a function of applied voltage and  $\theta$ . The intensity within a diverging angle of  $\pm 10^\circ$  was measured.

### 3. Operation principle

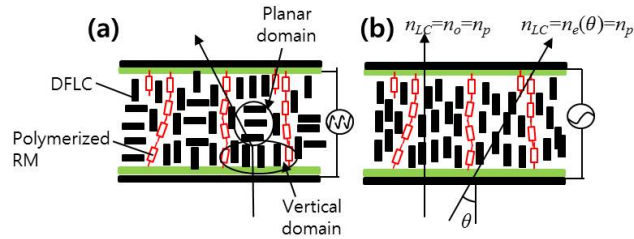


Fig. 3. Schematic illustration of proposed PDLC using RM and DF-LC at (a) scattering and (b) transparent state.

Figure 3 shows the operation principle of the proposed PSLC cell using the DF-LC-RM mixture. We refer the proposed PSLC as DF-PSLC hereinafter. Under a 50 kHz bipolar voltage [Fig. 3(a)], DF-LC has a negative dielectric anisotropy and the bulk molecules switch to planar state. Meanwhile, DF-LC molecules adjacent to the polymerized RM are strongly bound to the polymers and not switched. Consequently, micro-sized LC domains with different refractive indices are formed in the cell and the incident light is strongly scattered. Because the DF-LC molecules can be switched to planar state by the electric field,  $n_e(\theta)$  of DF-PSLC at scattering state [Fig. 3(a)] can be greater than that of conventional PDLC where the bipolar axes of the droplets are randomly oriented [Fig. 1(a)]. The increased scattering intensity of DF-PSLC can give greater contrast ratio. On the other hand, under a 1 kHz bipolar voltage [Fig. 3(b)], the DF-LC molecules are homeotropic aligned and the cell becomes transparent. We should note that the light sees the same refractive indices of DF-LC and polymerized RM, i.e.,  $n_e(\theta) = n_p(\theta)$  at all incident angle in the transparent state [Fig. 3(b)], resulting in a good viewing angle property without a scattering or haze. On the other hand, the conventional PDLC cell using non-mesogenic monomers [Fig. 1(b)] cannot avoid a light scattering for the oblique incidence light due to the mismatching between  $n_e(\theta)$  and  $n_p$ .

### 4. Results and discussion

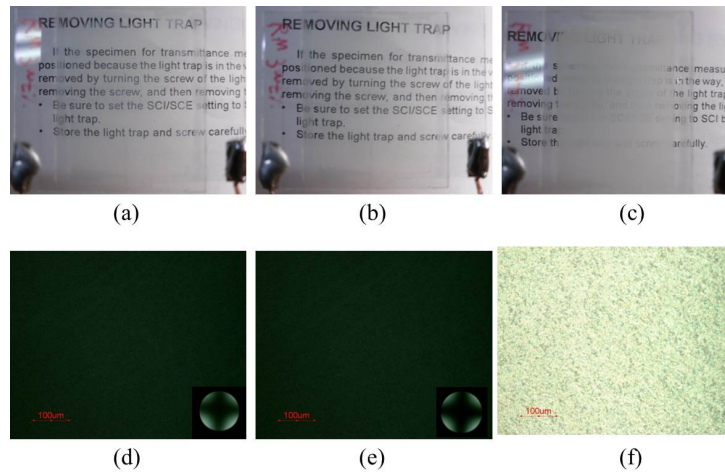


Fig. 4. 3.0 wt% RM mixed DF-PSLC cell images at (a) 0 V and (b) 1 kHz 40 V, and (c) 50 kHz 40 V state. POM image of the 3.0 wt% RM mixed PSLC cell at (d) 0 V, (e) 1 kHz 40 V, and (f) 50 kHz 40 V state. The inset images in (d) and (e) are the conoscopy image of the cell.

Figures 4(a) and 4(b) shows the 3.0 wt% RM mixed DF-PSLC cell images at 0 V and 1 kHz 40 V applied state, respectively. The picture was obtained under an ambient light. Both in

Figs. 4(a) and 4(b), the letters under the DF-PSLC cell were clearly seen confirming a good transparent state. Thus, the DF-LC molecules were uniformly homeotropic aligned both at 0 V and 1 kHz 40 V applied state. Upon applying 50 kHz 40 V [Fig. 4(c)], the PSLC cell was switched to scattering state and the background letters were not seen. As described above, the bulk DF-LC molecules were switched to planar state by the high frequency bipolar voltage, while the DF-LC molecules adjacent to the polymers and the substrate surfaces were homeotropic aligned. Under this situation, the incident light is strongly scattered by the mismatching of the refractive indices between the planar and homeotropic domains [Fig. 3(a)]. Figures 4(d)–4(f) represent the polarizing optical microscopy (POM) images of the DF-PSLC cell between crossed polarizers at 0 V, 1 kHz 40 V, and 50 kHz 40 V state, respectively. Both at 0 V and 1 kHz 40 V applied state, the cell showed uniform dark texture without a defect. In addition, the conoscopy images in the inset of Figs. 4(a) and 4(e) showed vertical orientation of the optic axis of the DF-PSLC cell. Thus, the homeotropic orientation of the DF-LC and RM molecules in transparent state [Fig. 3(b)] was confirmed. On the other hand, the POM texture showed bright and dark micro-sized domains at 50 kHz 40 V applied state [Fig. 4(f)]. The bright domains are the phase-separated DF-LC droplets dispersed in the polymer network and the dark domains are the polymerized RM and DF-LC molecules as described in Fig. 3(a).

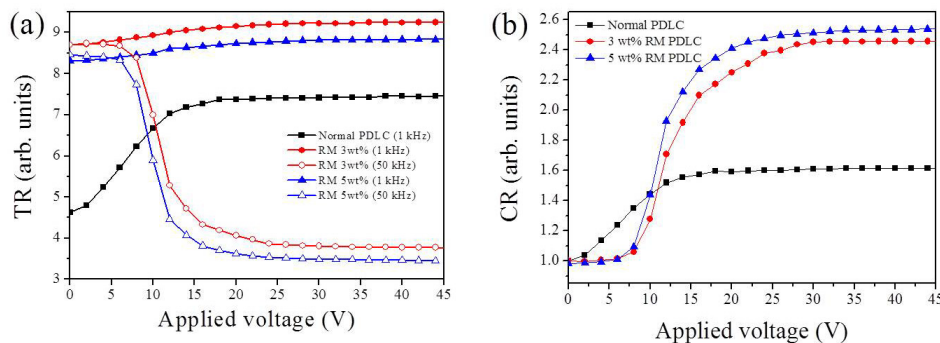


Fig. 5. (a) TR and (b) CR of the normal PDLC, 3.0 wt% RM-, and 5.0 wt% RM-mixed DF-PSLC cells vs. applied voltage. CR of normal PDLC was defined as normalized TR(V) by TR at 0 V state, whereas CR of DF-PSLC was defined as TR(V) under 1 kHz applied voltage normalized with TR(V) under 50 kHz voltage.

Figure 5(a) shows TR of the normal PDLC and the DF-PSLC cells vs. applied voltage. The light was normally incident to the cells. TR of the 3.0 wt% and the 5.0 wt% RM-mixed DF-PSLC cell at 0 V was greater than TR of the normal PDLC cell under 40V. Under 1 kHz applied voltage, TR of the DF-PSLC cells was slightly increased with greater voltage and this is presumably due to the increased order parameter of the DF-LC molecules by the applied voltage. Thus, the DF-PSLC cell became more transparent coinciding with Figs. 4(a) and 4(b). TR of the 3 wt% RM-mixed DF-PSLC cell under 1 kHz 40 V was about 30% greater than that of the normal PDLC cell under the same voltage. This means the DF-PSLC cell is more transparent than the normal PDLC due to the uniform homeotropic orientation. Under 50 kHz applied voltage, TR of the DF-PSLC cells was decreased and TR at 40 V applied state was about 20% smaller than TR of the normal PDLC cell at 0 V state. Thus, scattering intensity of DF-PSLC was greater than that of normal PDLC. This is certainly due to the more planar orientation of the DF-LC molecules switched by the electric field [Fig. 3(b)] and increased  $n_e(\theta)$  compared to the random oriented LC molecules in the normal PDLC cell [Fig. 1(a)].

Figure 5(b) represents the contrast ratio (CR) of the normal PDLC and the DF-PSLC cells vs. applied voltage. CR of the normal PDLC cell was defined as normalized TR(V) by TR at 0 V state, whereas CR of DF-PSLC was defined as TR(V) under 1 kHz applied voltage normalized by TR(V) under 50 kHz voltage. CR of the DF-LC cell was about 50% greater than



that of the normal PDLC cell at 40 V. From these results, greater TR and CR values of the proposed DF-PSLC cell were quantitatively confirmed.

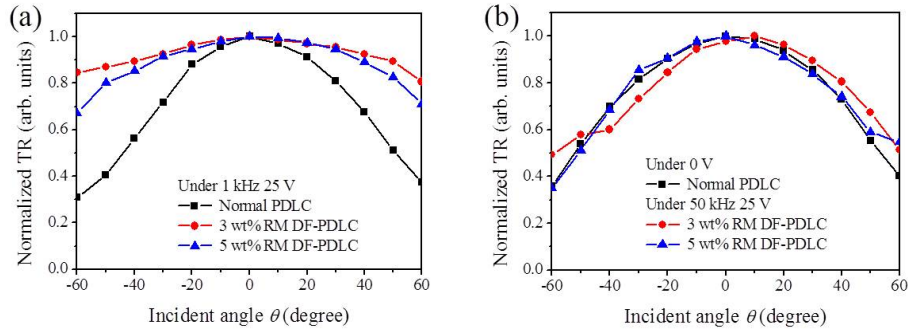


Fig. 6. TR of normal PDLC and DF-PSLC cells vs. incident angle  $\theta$  at (a) transparent and (b) scattering state. TR at transparent state was obtained by applying 1 kHz 25 V voltage. TR of the normal PDLC and DF-PSLC cells at scattering state was measured under 0 V and 50 kHz 25 V, respectively.

Let us turn to the viewing angle property of the proposed DF-PSLC cell. Figures 6(a) and 6(b) shows normalized TR of the normal PDLC and DF-PSLC cells at transparent and scattering state, respectively, vs. incident angle  $\theta$ . TR at transparent state was obtained by applying 1 kHz 25 V voltage, while TR at scattering state was measured under 0 V and 50 kHz 25 V for the normal and DF-PSLC cells, respectively. Normalized TR of the normal PDLC at  $\theta = \pm 60^\circ$  was about 0.3, while that of the 3.0 and 5.0 wt% RM-mixed DF-PSLC cell was about 0.8 and 0.7, respectively. Thus, it was clearly confirmed that the proposed DF-PSLC cells showed less change of TR to the oblique incident light than the normal PDLC cell. Certainly the better viewing angle dependence of the DF-PSLC cell is due to the simultaneous matching of the refractive indices between DF-LC and RM which were vertically aligned [Fig. 3(b)]. The poor viewing angle property of the normal PDLC is due to mismatching of refractive index between LC and polymer matrix [Fig. 1(b)]. In the scattering state [Fig. 6(b)], the DF-PSLC cell showed nearly similar viewing angle property. Under 50 kHz voltage, both vertical and planar domains exist in the DF-PSLC cell [Fig. 3(a)]. This configuration is optically equivalent to the random oriented bipolar droplets in the normal PDLC cell [Fig. 1(a)], thus giving the same angular dependence of TR at scattering state.

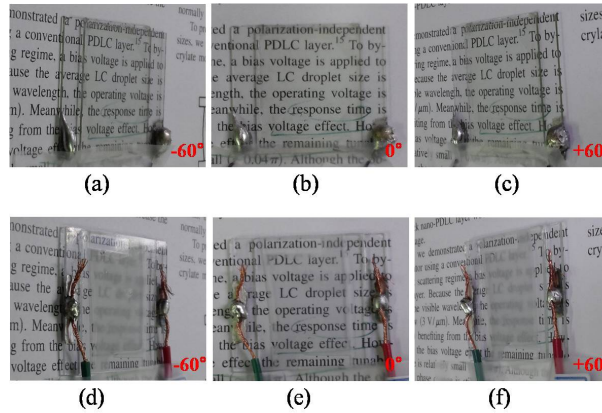


Fig. 7. (a)–(c) 3 wt% DF-PSLC and (d)–(f) normal PDLC cells under 1 kHz 25 V bipolar voltage. Incident angle  $\theta = -60^\circ$  in (a) and (d),  $\theta = 0^\circ$  in (b) and (e),  $\theta = +60^\circ$  in (c) and (f).

Figure 7 shows the demonstration image of the DF-PSLC [Figs. 7(a)–7(c)] and normal PDLC cells [Figs. 7(d)–7(f)] at transparent state. 1 kHz 25 V bipolar voltage was applied

across the cell. It was clearly observed that the DF-PSLC cell was always transparent both for normal [Fig. 7(b)] and oblique view [Figs. 7(a) and 7(c)]. On the other hand, the normal PDLC cell became hazy for oblique view [Figs. 7(d) and 7(f)] due to the mismatching of the refractive index between LC and polymer matrix.

## 5. Conclusion

We proposed a new type of PDLC using a mixture of DF-LC and RM. The DF-LC and RM molecules were homeotropic aligned and TR of the DF-PSLC cell at transparent state was greater than the normal PDLC cell due to the uniform vertical orientation. Scattering effect of the DF-PSLC cell was also increased by switching DF-LC molecules using high frequency electric field, thus giving greater CR. In addition, the proposed DF-PSLC showed good viewing angle property due to the simultaneous refractive index matching between DF-LC and RM at oblique incident angle.

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