# A polarizer-free flexible and reflective electrooptical switch using dye-doped liquid crystal gels

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**Abstract:** A polarizer-free flexible and reflective electro-optical switch using dye-doped liquid crystal (LC) gels is demonstrated. The electro-optical performances of both scattering and absorption based dye-doped LC gels depend on curing temperatures due to domain sizes of polymer networks. Such flexible electro-optical switch is bendable and trim-able because of the vertical polymer networks and gel-like materials. The dye-doped LC gel shows good reflectance ~55%, good contrast ratio~450:1 and fast response~6.4ms at curing temperature 10 °C. The bending curvature is 21 mm. The dye-doped LC gels open a new window for trim-able electronic papers, decorative displays, electrically switchable curtains, and electrically switchable sun control film for the automobiles, homes or commercial buildings

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OCIS codes: (230.3720) Liquid-crystal devices; (160.5470) Polymers.

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#### 1. Introduction

Roll-able, bendable, trim-able, and conformable paper-like flexible displays are useful for electronic paper, electronic tags, and decorative displays.[1] Many liquid crystal (LC) technologies, such as polymer-dispersed liquid crystals (PDLC)[1-6], cholesteric liquid crystals[1, 7-11], and single-substrate LCDs using photoenforced stratification[1, 12-14] or using LC/polymer composites[15-18], and non-liquid crystal technologies, such as electrophoretic imaging [1, 19-21], Gyricon[1, 22], and organic light-emitting diode (OLED)[1, 23-27], have been carried out to achieve transmissive type or reflective type flexible displays. In liquid crystal-based flexible displays, bistability and colors of cholesteric liquid crystals limits the application due to the complexity of driving and color shift at off angle. By combining dye absorption and scattering, contrast ratio of dye-doped PDLC is not good enough.[28-29] The dye solubility with polymer matrix, the order parameter of dye and

dichroic ratio (typically ~10:1) of dye are the factors to limit the contrast ratio of dye-doped PDLC. Recently, we have developed a new polarizer-free LCD using a dye-doped dual-frequency liquid crystal (DFLC) gel on the ITO-only glass substrates [30-31]. Without polarizers, the optical efficiency is high and viewing angle is wide. Although its contrast ratio reaches ~150:1 and response time ~6 ms under frequency modulation, the frequency driving scheme, high driving voltage (~30 V<sub>rms</sub>) and unavoidable dielectric heating effect.[32-33] need to be overcome for TFT-LCDs and flexible displays applications. To avoid the dielectric heating effect of DFLC, negative LC within vertical alignment layer is a good alternative and it is suitable for making a transflective LCD[34]. The gel-like feature of materials, vertically aligned polymer network and low temperature processes drive us to realize a trim-able and bendable polarizer-free flexible display in reflective mode.

In this paper, we demonstrated a reflective, flexible, and trim-able electro-optical switch using dye-doped LC gels which is polarizer-free, fast response, high contrast. The curing temperature has influence on the electro-optical properties of dye-doped LC gels because temperature-dependent domain sizes. The normally white gels exhibit ~55% reflectance, ~450:1 contrast ratio, ~6.4 ms response time, and ~30  $V_{\rm rms}$  at f=1 kHz driving voltage at curing temperature 10 °C. A single pixel flexible reflective display using such dye-doped LC gels is also demonstrated under bending and cutting.

## 2. Sample preparation and operating principle

The dye-doped LC gel we employed is a mixture of negative nematic liquid crystal ZLI-4788 (Merck,  $n_e$ = 1.6567,  $\Delta n$ =0.1647 at  $\lambda$ =589 nm;  $\Delta \epsilon$ = -5.7 at f= 1 kHz) and a diacrylate monomer (bisphenol-A-dimethacrylate) with a dichroic dye S428 (Mitsui, Japan) at 90:5:5 wt% ratios. The structure of the diacrylate monomer is shown as follows:

The monomer chosen is preferred not too rod-like structure; otherwise, scattering decreases. The dye-doped LC mixture was then injected into an empty cell consisting of two glass substrates or flexible substrates whose inner surfaces were coated with a thin conductive layer, indium-tin-oxide (ITO) on glass substrates and indium-zinc-oxide (IZO) on the flexible substrates, and polyimide (PI) layer *without* rubbing treatment. The PI layer provides vertical alignment for the LC directors. The cell gap was 5  $\mu$ m. The filled cell was irradiated by a UV light ( $\lambda$ ~365 nm, I~3 mW/cm²). Both cells were cured at a fixed temperature for 1.5 hr. After photo-polymerization, the formed chainlike polymer networks are along the z direction because the LC directors are aligned perpendicular to the glass substrates during the UV curing process, as shown in Fig. 1(a).

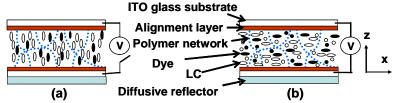


Fig. 1. Schematic operating principle of dye-doped liquid crystal display at (a) voltage-off state and (b) voltage-on state. The alignment layer has no rubbing treatment.

The structure and operation principles of the dye-doped LC gel are schematically depicted in Fig. 1(a) and Fig. 1(b). At V=0, the cell does not scatter light and the absorption is rather weak due to the vertically aligned polymer networks, liquid crystal directors and dye molecules. Therefore, the display has the highest reflectance. When we apply a high voltage at  $f=1\,$  kHz in the dye-doped LC gel, the negative liquid crystals and dye molecules are reoriented in the x-y plane, as Fig. 1(b) depicts. The polymer network scatters light strongly.

Since the alignment layer has no rubbing treatment, the absorption has no preferred direction; therefore, the display appears black because of the strong light scattering and dye absorption. The appearance of color is mainly because of the light absorption of dye. The scattering and reflection assist the multiple absorption due to the path elongation of light propagation.

### 3. Experiment and results

Figure 2 show the morphologies observing under an optical microscope with a single polarizer only. The top region of the two regions in Fig. 2 is the patterned ITO area. The bright part represents the state of V=0. The dark area represents the ITO electrodes with V=30  $V_{rms}$  at f=1 kHz. At the voltage-off state, the cell shows good bright state because of the vertically aligned polymer networks, LC and dye molecules. At 30  $V_{rms}$ , it shows the fine domain textures of the polymer networks, and red color because of dye molecules, as shown in Fig. 2. Our LC cell shows good dark and bright states although the dark state up to now is redish, not truly black.

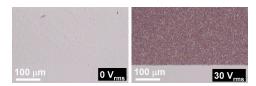


Fig. 2. The microscopic images of dye-doped LC gels at 0 V<sub>rms</sub> and 30 V<sub>rms</sub>.

We adopt the typical reflectance measurement in order to measure the electro-optical properties of dye-doped LC gels. Because the guest-host system we employed appears dark red rather than black, we used a unpolarized green He-Ne laser ( $\lambda$ =543.5 nm, Melles Griot, Model 05-LGR-173) instead of a white light source for characterizing the device performances. A dielectric mirror was placed behind the cell so that the laser beam passed through the cell twice. A large area photodiode detector (New Focus, Model 2031) was placed at ~25 cm (the normal distance for viewing a mobile display) behind the sample which corresponds to ~2° collection angle. A computer controlled LabVIEW data acquisition system was used for driving the sample and recording the light reflectance. In order to prove our dyedoped LC gel is polarization independent, we placed a polarizer between the laser and the LC cell. The reflectance as a function of an angle of the polarizer at different applied voltages is shown in Fig. 3. The variation of reflectance is less than 5% when we rotated the polarizer. It indicates the dye-doped LC gels are indeed polarization independent at all applied voltages. The reflectance at 0  $V_{rms}$  is around 50%.

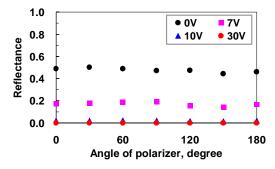


Fig. 3. Reflectance as a function of angle of the polarizer at different applied voltages. The curing temperature was 20  $^{\circ}\text{C}.$ 

Then we removed the polarizer. That means the incident light was unpolarized green laser

beam. Figure 4 (a) plots the voltage-dependant reflectance of the dye-doped LC gels at various curing temperatures. The reflectance is normalized to that of a pure LC cell with the same cell gaps. The reflectance decreases gradually as  $V > V_{th}$  because of the increase of the scattering and the absorption. As curing temperature decreases, the increases (~40% to ~55%) of maximum reflectance at 0 V<sub>rms</sub> result from the better vertical alignment of LC directors, dye molecules and polymer networks at low curing temperature. The contrast ratio (CR) is defined as a reflectance ratio of 0  $V_{rms}$  to 30  $V_{rms}$ . The CR is ~450: 1 at 10 °C, 250: 1 at 20 °C, 200: 1 at 30 °C, and 300: 1 at 40 °C. The contrast ratio decreases as T< 30 °C and then increases as T>30°C. That is because the increase of a curing temperature results in larger polydomains; therefore, the contrast ratio and threshold voltage decrease. Moreover, the decay time increases, as shown in Fig. 4(b). When the temperature is higher than 30°C, we found the cell has dynamic scattering, a fluctuation of liquid crystal directors in polymer domains, to help rebooting the contrast ratio in spite of the larger domain size. The reason why the larger domain has the dynamic scattering is still unclear. Besides the curing temperature, the UV curing intensities, monomer concentrations and dye concentrations also affect the performance of the dye-doped LC gels. Based on our experimental results (results are not shown here), UV curing intensity does not affect the electro-optical properties of dye-doped LC gels dramatically. However, weak UV curing intensity needs the longer curing time in order to complete the polymerization. Higher monomer concentrations, higher driving voltages. The polymer network of dye-doped LC gels is not stable as the monomer concentration is too low. To lower the driving voltage, a high birefringence and high absolute value of dielectric anisotropy ( $\Delta \varepsilon$ ) of a negative LC and slightly lower polymer concentration could be considered.

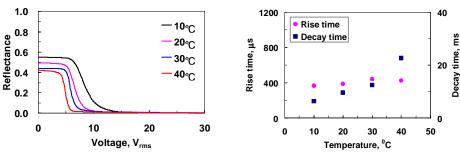


Fig. 4. (a) Voltage-dependent reflectance at various curing temperature. (b) Curing temperature-dependent response time

Response time is also an important issue for guest-host displays. The response time of the dye-doped LC gels was measured using 30  $V_{rms}$  squared pulses with time duration 500ms at f=1 kHz. The curing temperature-dependent rise times and decay times are shown in Fig. 4(b). The rise times are about 0.4 ms and the decay time decreases with decreasing curing temperatures. A typical response time of a guest-host display is around 50 ms. The response time of our dye-doped LC gel ( $\sim$ 6.4ms) is  $\sim$ 7x faster because polymer network helps LC directors to relax back. The rise time is  $\sim$ 0.4 ms and decay time is  $\sim$ 6 ms at 10  $^{\circ}$ C.

To validate the red color of dye-doped LC gels comes from the interaction between monomer and some components of dye, not the change of chemical structures of dye by UV light, we measured the transmission spectrum of dye-doped LC gels at 0  $V_{rms}$  before and after UV curing by using a spectrometer (Ocean Optics USB2000), as shown in Fig. 5. The black dye (S-428) we used consists of several molecular components. Without monomer, dye in LC shows black color. However, the color changes to red when we mixed dye, LC and the monomer before UV illumination. After illuminating UV light, the spectrum is similar, but transmission decreases slightly due to degrade of alignment of LC and dye molecules as monomer reacts to form polymer networks. Therefore, we can attribute the red color to the interaction between monomer and dye components. Besides the dye absorption, the scattering

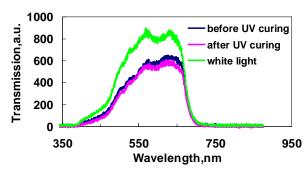


Fig. 5 Transmission spectra of dye-doped LC gels before (blue line) and after UV curing (pink line) at voltage-off state. The curing temperature was 20°C. Green line indicates the transmission spectra of white light source.

To prove principles, we also fabricated a single pixel polarizer-free reflective LCD using the dye-doped LC gels in glass substrates. To avoid specular reflection, we laminated a diffusive reflector, a white paper, on the backside of the bottom glass substrate. The ambient white light was used to illuminate the samples. Figure 6 (a) shows the photos of the displays using a 5  $\mu m$  dye-doped LC gel at different viewing angle. The voltage is applied in the middle squared region which is also ITO patterned region. Since no polarizer is required, the optical efficiency is high and the viewing angle is not limited by a polarizer.

A single pixel polarizer-free reflective and flexible LCD using dye-doped LC gels is shown in Fig. 6 (b). The flexible substrates are provided by EOL/ITRI (Electronics& Optoelectronics Research Laboratories, Industrial Technology Research Institute, Taiwan) [35-36]. IZO was over coated on the top of flexible substrates made by polycarbonate with thickness 120 µm. The cross shaped microstructures made by photo-spacers, resins, were developed on the flexible substrates by photolithography process in order to maintain the cell gap under bending. The width of photo-spacers is 10 µm and the pitch of photo-spacers is 430 μm. We also laminated a white paper as a diffusive reflector on the backside of the bottom flexible substrate. The ambient white light was used to illuminate the sample. The vertically aligned polymer networks can further help to maintain the cell gap under bending. However, the CR is degraded because of two reasons. First, the white light source is used to observe the dye-doped LC gels rather than the laser light. Secondly, the microstructures of photo-spacers somehow hinder the polymer network, LC directors and dye molecules to align vertically. Figure 7 is the transmission as a function of radius of curvature under bending at 0 and 30 V<sub>rms</sub>. The measurement method is two-point bending technique[1]. The transmission of dyedoped LC gels is almost the same as the radius of curvature larger than 21 mm. Figure 8 (a) and (b) are the bending performances of dye-doped LC gels at 0 V<sub>rms</sub> in transmissive mode and at 30 V<sub>rms</sub> in reflective mode. The dye-doped LC gel is trim-able as well because our material is gel-like, as shown in fig. 8(c). The flexible display performance remains the same after cutting by a scissor. In fig. 8(d), we demonstrated a single pixel polarizer-free reflective and flexible LCD using the dye-doped dual frequency LC gels under bending when we applied voltages at different driving frequencies. Without sealing dye-doped dual frequency LC gels by glue, the performance remains the same after scissoring a corner of the display. Since no polarizer in needed, the residual birefringence of polycarbonate does not affect the performance of our flexible display.

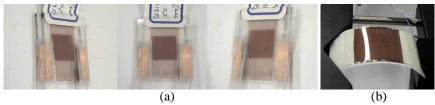


Fig. 6. (a)A single pixel polarizer-free reflective LCD using the dye-doped LC gels in glass substrates at curing temperature 10 °C. (497 KB) (b) A single pixel polarizer-free reflective and flexible LCD using dye-doped LC gels at curing temperature 20 °C. (728 KB) White papers were used as diffusive reflectors.

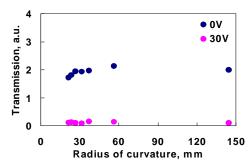


Fig. 7. The transmission as a function of bending radius curvature of a single pixel dye-doped LC gels at at 0 and 30  $V_{rms}$ . The curing temperatures was 20  $^{\circ}$ C.

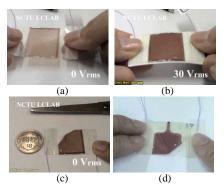


Fig. 8. A single pixel of a polarizer-free reflective and flexible LCD using dye-doped LC gels under bending at (a) 0  $V_{rms}$  in transmissive mode (794KB ) and (b) 30  $V_{rms}$  in reflective mode (216KB), and (c) by a scissor cutting(1.35MB) (d) A single pixel of a polarizer-free reflective and flexible LCD using dye-doped dual frequency LC gels under bending. (569 KB) The curing temperatures were 20  $^{\circ}\text{C}$  in (a) ,(b) and (c). The curing temperature was 10  $^{\circ}\text{C}$  in (d). White papers were used as diffusive reflectors in (b) ,(c) and (d)

## 5. Discussion

In order to further understand the mechanism of dye-doped LC gels, the reflectance (R) as function of voltage-induced tilt angle ( $\theta$ ) can be expressed as

$$R(\theta) = a \cdot e^{-\sigma(\theta) \cdot N \cdot 2d} \cdot e^{-\alpha(\theta) \cdot c \cdot 2d}$$
 (1)

where d is cell gap ( $\sim$ 5 µm), N is number density of domains of polymer networks, c is the dye concentration ( $\sim$ 5 wt %) ,and  $\mathcal{A}$  is an LC alignment factor resulting from polymer networks.  $\alpha(\theta)$  and  $\sigma(\theta)$  are absorption coefficient and scattering cross section respectively. At V=0, LC directors are perpendicular to the substrates ( $\theta \sim 0$  and  $a \sim 1$ ); therefore, the scattering

of the dye-doped LC gels is weaker (i.e.  $e^{-\sigma(\theta)\cdot N\cdot 2d}$  ~1) and the absorption rely on  $\alpha_{\perp}$  which stands for the absorption coefficient when the polarization of an incident light is perpendicular to the principal molecular axis of the dye molecules. Hence, we can calculate  $\alpha_{\perp} \sim 1.386~\mu m^{-1}$  from the experiment results (R=0.55, a=1 at curing temperature 10 °C ) and  $\alpha_{//} \sim 13.86~\mu m^{-1}$  since the dichroic ratio of dye molecules is ~10:1. When V> V<sub>th</sub>, the absorption coefficient  $\alpha(\theta)$  is:

$$\alpha(\theta) = \frac{\alpha_e(\theta) + \alpha_\perp}{2} \tag{2}$$

where

$$\alpha_e(\theta) = \frac{\alpha_{\perp} \cdot \alpha_{\parallel}}{\sqrt{{\alpha_{\parallel}}^2 \cdot \cos^2(\theta) + {\alpha_{\perp}}^2 \cdot \sin^2(\theta)}}$$
(3)

When the applied electric field increases, the tilt angle of LC directors increases; therefore,  $\alpha(\theta)$  increases as well. Based on anomalous diffraction approach [28, 37],  $\sigma(\theta)$  can be expressed as

$$\sigma(\theta) \approx \left[\frac{r}{\lambda}(n(\theta) - n_p)\right]^2$$
 (4)

where r is the domain size of dye-doped LC gels,  $\lambda$  is wavelength of light,  $n_p$  is refractive index of polymers. Usually  $n_p \sim n_o$ . The refractive index  $n(\theta)$  of LC directors with title angle  $\theta$  is:

$$n(\theta) = \frac{n_e \cdot n_o}{\sqrt{n_e^2 \cdot \cos^2(\theta) + n_o^2 \cdot \sin^2(\theta)}}$$
 (6)

At V=0,  $\sigma(\theta) \approx 0$  because  $n(\theta) = n_0 = n_p$ . When V>>V<sub>th</sub>(threshold voltage),  $n(\theta) = n_e$ ,  $\sigma(\theta) \approx [r \cdot \Delta n/\lambda]^2$ . Therefore, the scattering increases with an applied voltage. By combing scattering and absorption, reflectance decreases as voltage-induced tilt angle increases. The scattering increases as the domain size of polymer network decreases as well because  $(\sigma(\theta) \cdot N \cdot 2d)$  is proportional to 1/r. Larger domain size results in not only poor scattering state and also poor vertical alignment at voltage-off state. Hence, the performance of dyedoped LC gels depends on the domain size of polymer networks which is curing temperature dependent.

#### 6. Conclusion

We have demonstrated a high-contrast and polarizer-free reflective and flexible electro-optical switch using dye-doped LC gels. The increase of the curing temperature resulting in larger domain sizes affects EO properties of dye-doped LC gels. Since no polarizer is needed, the viewing angle is wide and the brightness is high. The contrast ratio is 450:1 at 30  $V_{rms}$  and the response time is around 6.4 ms at curing temperature 10°C under laser beam measurement. The maximal reflectance is about 55%. The curing temperature is low ( $<40^{\circ}$ C) which is more favorable for the flexible display applications. In addition, it is bendable and trim-able. That is because the polymer networks vertical to the substrate and the micro-structures of photospacer help to maintain the cell gap during the bending. The gel-like materials assist stabilizing the flexible display under cutting. However, the issues we have to overcome are the high driving voltage and red color of dye-doped LC gels. Improving LC materials with larger value of dielectric anisotropy and dye could lower driving voltage and adjust the color. Our dye-doped LC gels provide a stable LC mode and open a new window in paper-like flexible displays. The potential application is electronic papers, electronic tags, electrically switchable curtains, and electrically switchable sun control film for the automobiles, homes or commercial buildings.

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