

# Optical Tuning of Polymer Stabilized Liquid Crystals Refractive Index

## G. Nabil Hassanein<sup>1,2\*</sup>

<sup>1</sup>Department of Physics, Taibah University, Saudi Arabia <sup>2</sup>Department of Physics and mathematical engineering, Higher institute of engineering and Technology ,Egypt

## Abstract

The effect of alignment layer on the response and transmission characteristics of polymer stabilized liquid crystal (PSLC) thin film driven by a modulated electrical signal is studied. In aligned PSLC thin film, the dispersed liquid crystal droplets in the polymer matrix are aligned along the rubbing direction of the alignment layer. Aligned PSLC thin film shows faster response to the applied electric field compared to non aligned PSLC. Total response time of aligned and non-aligned PSLC film is 12.4 and 27.2 ms, respectively under driving field strength of 2.03 V/µm. Thus alignment layer greatly improved the response time and reduced the driving, threshold, and saturation voltages. The study shows how possible to improve and tune the electro-optic properties of PSLC to be applied in planar photonic devices.

**Keywords:** Polymer stabilized liquid crystal; Alignment; Refractive index; Optical switching; Photonics

## Introduction

Planner light wave circuit (PLC) devices have numerous advantages in terms of their use in optical fiber communication systems. They offer compactness and ruggedness, as well as excellent controllability, compatibility, high reliability, and mass productivity. PLC technology potentially provides low-voltage and low-power electrical tuning of devices by using thermo-optic or electro-optic (EO) effects [1]. Nevertheless, the thermo-optic method deals with disadvantages of power consumption, heat dissipation, and potential geometric deformation of waveguide structure under high heater temperature. These problems can be resolved by using the EO effect as an alternative.

As opposed to inorganic EO materials, polymers are very attractive for optical devices due to advantages of large optical nonlinear coefficient, fast response times, low dielectric constants, simple fabrication processes, and easy fabrication of multi-layer structure. EO polymers have also been investigated because of their large EO coefficient, high speed, and low driving voltage. However, the stability of EO polymer remains an issue that needs to be tackled [2].

Polymer stabilized liquid crystal (PSLC) can be an alternative EO material due to its wide applicability. PSLC is promising in the scientific and industrial research and development fields [3]. Unlike the conventional liquid crystal (LC) [4-6], PSLC films are mechanically flexible, easy to prepare, and can modulate light without using a polarizer while providing high optical efficiency. In comparison with pure LC devices, PSLC-based devices have lower LC material content, and hence provide economic advantage compared to the more expensive pure LC. Solid PSLC thin film is also appropriate for patterning through typical photolithography or other techniques. Thus is a promising material for various PLC devices; PSLC received attention in the field of optical communication [7].

PSLC thin films consist of micrometer-sized LC droplets embedded in optically transparent polymeric matrix and is formed by phase separation of LC components from a homogeneous mixture with either a pre-polymer or a polymer.

The operation principle of PSLC films is based on the electric field controlled optical transmission from LC micro-droplets. Light scattering depends on the shape and size of LC droplets, as well as their compatibility with the polymer matrix, refractive indices of both phases, and birefringence.

In the on-state, when an electric field is applied to a PSLC thin film, the refractive indices of the aligned LCs inside the droplets and polymer matrix are matched. This causes a change in optical scattering from the film that change from opaque to transparent state. In the off-state, surface anchoring causes non-uniform director field within the droplets. As a result, there is a mismatch between the effective refractive index of the LC and the refractive index of the polymer. Thus PSLC thin film appears milky white in opaque state due to high light scattering [8-10].

Although PSLC has advantages compared with LC, but on the other hand the large amount of interface between the LC and polymer causes anchoring force which strongly affects the reorientation of the LC director responding to the applied voltage and generally requires higher driving voltage [10].

It is advantageous to apply a PSLC device with fast response time, low threshold and saturation voltage, and on-state high transmission. Therefore, much attention is required to improve these properties. The response time of a PSLC film mainly depends on droplet size, shape, LC rotational viscosity, and applied electric field [10-12]. LC droplets orientation significantly influences PSLC thin film properties. Ordered LC droplets improve EO characteristics of PSLC thin film. The surface anchoring of the alignment layer of polymer network during curing process also affects the EO properties of the PSLC devices [13].

Alignment layer; A rubbed polyimide alignment layer is simply used to align LC molecules in the LC thin film along the rubbing direction [6,14]. Various alignments techniques were previously investigated by researchers to control the EO properties of PSLC films [15-17]. In previous studies, some complex PSLC mixtures have been created by mixing different types of LC materials with different monomers, cross linker, and photo-initiating materials [14,16,18]. The curing time of

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<sup>\*</sup>Corresponding author: Hassanein GN, Department of Physics, Taibah University, Saudi Arabia, Tel: +966 14 861 8888; E-mail: ghassanein@taibahu.edu.sa

some PSLC films is quite extensive ~40 minutes for pre-polymerization then 25 minutes for full curing and some PSLC films are cured at high temperature [15,19]. In such studies, most PSLC films were relatively thicker. But for most planar photonic devices, lower film thickness is required. The materials composition of PSLC mixture should also be simple to avoid any degradation, especially in the case of polymerbased photonic devices.

Alignment layer is also used in this study in order to improve the EO properties of PSLC-based devices. The effect of alignment layer on the response and transmission characteristics of the PSLC thin film is investigated.

## Experimental

A simple PSLC mixture is prepared by mixing only one commercial pre-polymer (NOA65) with one commercial LC material (E7) processed at room temperature. The thickness of the prepared PSLC thin film in our study is 5  $\mu$ m, which can be considered as a thinner film compared with PSLC films from other studies [6,14]. A modulated driving signal is applied, rather than normal square signal [16,20], to enhance the response characteristics and to lower the driving voltage. In as much as the PSLC thin film can substantially improve response and transmission characteristics, it can therefore be used for a wide variety of tunable planar photonic devices.

#### Materials

We used nematic LC E7 (Merck) which is an eutectic mixture of cyanobiphenyls and terphenyls capable of giving a nematic phase of high chemical and photochemical stability at room temperature. The nematic-isotropic phase transition (clearing point) of the nematic LC E7 is ~60°C. E7 has positive dielectric anisotropy,  $\Delta\epsilon$ =13.8 ( $\Delta\epsilon$ = $\epsilon_{\parallel}$ - $\epsilon_{\perp}$ ), where the parallel and perpendicular components of dielectric constant is  $\epsilon_{\parallel}$ =19 and  $\epsilon_{\perp}$ =5.2 at 20°C and 1 kHz frequency. Ordinary and extraordinary refractive indices of E7 are n<sub>o</sub>=1.5185 and n<sub>e</sub>=1.737 ( $\lambda$ =633 nm at 20°C) [21]. The polymeric material used was a photopolymerizable pre-polymer (Norland Optical Adhesive, NOA65). The refractive index of the UV-cured NOA65 polymer was n<sub>p</sub>=1.524 ( $\lambda$ =633 nm at 20°C) with a dielectric constant of  $\epsilon_p$ =4. Both NOA65 polymer and LC material have very high resistivity (~10<sup>12</sup> \Omega.cm) [12].

## **PSLC** mixture preparation

E7 liquid crystal material is mixed with NOA 65 using a hot plate magnetic stirrer and heated above the isotropic clearing temperature in order to achieve good mixing. In the mixing processes, we used higher LC content (70 wt %) relative to the polymer content (30 wt %) in order to reduce the value of the driving voltage, since as the LC content in the PSLC mixture increase the driving voltage decrease [10].

## **Cell preparation**

We used two empty cells with upper and lower Indium Tin Oxide (ITO) glass substrates as transparent electrodes. Both cells were with a cell gap spacing of 5  $\mu$ m. One cell contained anti-parallel rubbed polyimide (PI) alignment layers while the other cell was without any alignment layers. The two cells were filled with PSLC mixture at 65°C by capillary action, and then cured under UV intensity of 33 mW/cm<sup>2</sup> at 20°C for 15 minutes. LC droplet formations within the polymer matrix were carried out using the polymer-induced phase separation process [10]. During UV curing, formation of distinct nematic LC droplets was observed early in the phase separation process of the E7/ NOA65 mixture.

### LC Droplets morphology investigation

To study the LC droplet morphology in the two PSLC films, the two fabricated PSLC cells were investigated using a polarizing microscope. The droplet shape and size within the two PSLC films were observed between the crossed polarizers of the polarizing microscope. Their images were captured for comparison.

#### EO measurements

The EO response of the two PSLC thin films in both cells was measured without the using polarizers as PSLC materials are polarization independent. A schematic of the experimental setup is shown in Figure 1. The driving signal frequency is 1 KHz and modulated by low frequency square signal of 5 Hz frequency. Typically, 1 KHz frequency is used for driving LC-based devices in order to ensure stable EO performance and to avoid conductivity effects [6,22]. The 5 Hz modulating signal frequency is also used to provide sufficient pulse duration time for the saturation of the output transmitted optical signal. The undershooting driving method (amplitude-modulated signal) was applied [6] to enhance PSLC switching properties, as well as to achieve a relatively fast and stable response with low driving voltage. The applied driving voltage was from 0–20 volts (peak-to-peak voltage).

When the electric field was applied, the LC molecules within the LC droplets started to align parallel to the field (i.e., positive dielectric anisotropy effect). As the applied electric field increased beyond the threshold value, the LC molecules aligned along the field direction. In this case, the LC refractive index (n<sub>2</sub>) matched with the refractive index (n) of the polymer material, which caused the probing laser beam to pass through the transparent PSLC film without inducing any scattering. The output transmitted optical signal was detected by the detector; it was then displayed on the oscilloscope. The rise time (switch-on time) and fall time (switch-off time) of both PSLC cells were measured as functions of the driving field strength, and were directly measured from the output traces of the oscilloscope. Total response time corresponds to the sum of rise and fall times. Under the transmission-voltage characteristics study, threshold field and saturation field strengths were then determined. Using the saturation field strength value, the effective refractive index for the two PSLC films were calculated and plotted as functions of the driving field strength.

## **Results and Discussion**

## LC droplets morphology

Figure 2a shows the microscopic image of the PSLC thin film





Figure 2: PSLC film as seen by 20X objective between crossed polarizers under a polarizing microscope for (a) with the alignment layer and (b) without the alignment layer.

with the alignment layer (i.e., between the crossed polarizer under the polarizing microscope). All dispersed LC droplets were almost spherical or ellipsoidal in shape. A uniform LC texture was viewed. Alternating bright and dark states were also observed upon rotating the PSLC cell, suggesting that the LC molecules inside the droplets are homogeneously aligned along the rubbing direction of the rubbed PI alignment layer. Droplets with parallel LC molecules anchoring condition that are aligned tangentially to the droplet sidewalls are called bipolar droplets [23].

Figure 2b shows the microscopic image of PSLC thin film without the alignment layer (i.e., between the crossed polarizer under the polarizing microscope). Most of the dispersed LC droplets were almost spherical or ellipsoidal, but some were shaped irregularly. Upon rotating the PSLC cell under the polarizing microscope, no other changes were observed in the color of the LC droplets. The observed extinction configuration of the droplets indicates that droplets have different morphologies: radial, bipolar, equatorial, and hybrid-aligned irregularly shaped. In the radial and equatorial configuration, LC molecules were anchored with their long axes perpendicular to the droplet walls [23]; in the bipolar configuration, the LC molecules were tangentially aligned to the droplet walls. In the case of radial droplets, one point defect was observed at the center of the droplet; in the bipolar droplet, two point defects were observed at the droplet poles. This variation in droplet configurations was due to different anchoring conditions of the droplets. Inside the irregularly shaped droplets, LC molecules were randomly aligned such that they were able to minimize their elastic free energy [21].

#### The response time of PSLC film

**Rise time:** Figure 3 shows the rise time of both PSLC films, with and without the alignment layer, as functions of the electric field strength (E). Rise time is defined as the time for transmitted light intensity to increase from 10% to 90% of the total transmitted intensity. As applied electric field strength increased, the rise time decreased; this is due to the rise time being inversely proportional to the square of the applied field strength (E) [10,21]. At low field strength, significant difference in rise time between the PSLC film with and without the alignment layer was observed. Rise times were 20 and 26 ms for with and without the alignment layer, respectively, at the field strength of 0.625 V/ $\mu$ m. However, at higher field strength, the rise time of both PSLC films were almost the same (1.2 ms at 2.03 V/ $\mu$ m). One reason is that at high field strength, almost all LC molecules inside the LC droplets are perfectly aligned in the direction of the applied field, regardless of the rubbing direction in the alignment layer.

**Fall time:** Figure 4 shows the fall time of both PSLC thin films, with and without the alignment layer, as functions of the applied electric field strength (E). Fall time is defined as the time taken for the transmitted light to fall from 90% to 10% of the total transmitted intensity. The fall time of PSLC film with the alignment layer was lower than that without the alignment layer. This was due to the increased anchoring strength in the PSLC film with the alignment layer, which was with the presence of the rubbed PI alignment layer.

As applied electric field strength increased, the fall time of both PSLC film increased. Theoretically, fall time is independent on applied field strength (E), directly proportional to LC material viscosity, and inversely proportional to the LC material elastic constant [10,21]. However, the increase in fall time in this study was due to the interaction between the anchoring forces of LC molecules at the LC droplet inner walls and the visco-elastic forces of the LC material. The competition between the applied field strength and the elastic and viscous torques of the LC material governed the response characteristics [24].









The increasing rate of fall time of PSLC film with the alignment layer was slower than that without the alignment layer. At low field strength (<1 V/µm), LC molecules started to respond to the driving voltage to be aligned along the field direction. Inasmuch as only the elastic relaxation was involved, the difference between the fall time values of both PSLC films was small. At low field strength, the fall time of PSLC film with and without the alignment layer was 5.6 and 11 ms, respectively, at 0.625 V/µm. As field strength increased (>1 V/  $\mu$ m), the difference between the fall time of PSLC film with and without the alignment layer also increased. At high fields, LC molecules were almost perfectly aligned along the field direction. LC molecules with the alignment layer could easily move back to their original position during switch off. However, LC molecules without the alignment layer took longer time to revert to their original position. The increase in fall time was due to the slow change of orientation of the nematic director inside the LC droplets at the droplet boundary surface. At the droplet boundary surface with the polymer (i.e., at the interface), the LC molecules were oriented by the electric field in different ways by those inside the droplet [24-26]. At higher field strength, the fall times of both PSLC films were 11.2 and 26 ms respectively, at 2.03 V/ $\mu$ m.

Total response time: Figure 5 shows the total response time of the two PSLC films with and without the alignment layer with applied field strength. Total response time was calculated by adding both rise and fall times. Total response time decreased as field strength increased. For the PSLC film with the alignment layer, total response time was lower than the PSLC film without the alignment layer. Total response time for the PSLC film with and without the alignment layer was 12.4 and 27.2 ms, respectively, at field strength of 2.03 V/ $\mu$ m, indicating that the effect of adding an alignment layer reduces the response time to almost 50%. In the PSLC with the alignment layer, the aligned LC droplets provided an efficient control of the optical properties of the PSLC film and faster EO response.

## The Transmission - voltage characteristics

Figure 6 shows the transmission-voltage characteristics of both PSLC thin films with and without the alignment layer. Transmission (T);  $T = \frac{I}{I_o}$ , where I is the transmitted light intensity and I<sub>o</sub> is the initial light intensity. Transmission was nearly constant at field strength





lower compared with the threshold value. Then, it sharply increased as field strength increased, and reached saturation value and maximum transmission. Threshold field ( $E_{th}$ ) and saturation field strengths ( $E_{sat}$ ) are defined as the field strength at which transmission reaches 10% and 90% of the maximum transmission, respectively. Generally, the pre-switching oscillations at the low field strength are due to the imperfect optical interference effects to the transmittance of the PSLC film [21]. In this study, the threshold and saturation field strength of the PSLC film with the alignment layer were 0.266 and 0.922 V/µm, respectively. In comparison, the threshold and saturation field strength for the PSLC film without the alignment layer were 0.308 and 1.26 V/µm, respectively.

#### **Refractive index tuning**

When an external electric field is applied to LC materials, the polar LC molecules inside the LC droplets tend to align along or perpendicular to the field direction depending on the sign of the dielectric anisotropy. As a result, the refractive index of the PSLC film is changed. Thus, it is possible to tune the refractive index of PSLC films to attain tunable PSLC-based planar photonic devices. The effective refractive index  $n_{\rm eff}$  (V) of PSLC films can be calculated from the equation [27].

$$n_{eff}(V) = n_o + \left(\frac{n_e - n_o}{3}\right) \exp\left[-\left(\frac{V}{V_{sat}}\right)^2 \cdot \ln(10)\right]$$

where  $n_o$  is the LC ordinary refractive index ( $n_o=1.5185$ ),  $n_e$  is the LC extraordinary refractive index ( $n_e=1.737$ ) [21], and  $V_{sat}$  is the saturation voltage. Variable  $V_{sat}$  is the value of the saturation field strength multiplied by PSLC film thickness (5 µm).

Figure 7 shows the variation of the effective refractive index  $n_{eff}$  as a function of electric field strength (E). The effective refractive index of the PSLC thin film with the alignment layer decreased sharply at lower field strength compared with the PSLC thin film without the alignment layer. The effective refractive index of the PSLC thin film with the alignment layer was tunable at lower field strength compared with the PSLC film without the alignment layer. For example, for the two PSLC films with and without the alignment layer, the effective refractive index of 0.92



and 1.26 V/ $\mu$ m, respectively. Thus, the PSLC film with the alignment layer is appropriate for optical tuning, offering functionality in planar photonic devices [28].

#### Conclusion

We studied the influences of adding an alignment layer on morphologies of LC droplets and EO properties of PSLC films. We found that LC droplets uniformly exhibit the rubbing direction of the alignment layer. Switching the properties of PSLC film with the alignment layer evidently improved (i.e., threshold voltage, saturation driving voltage, and response time declined by about 14%, 27%, and 50% respectively). The tuning characteristics could also be achieved at the lower field strength region after adding the alignment layer. This improvement could be attributed to the addition of the alignment layer. In overall, an alignment layer can extensively assist in the production of PSLC-based tunable planar photonic devices.

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