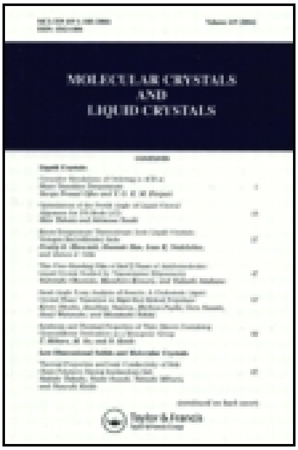

Molecular Crystals and Liquid Crystals

Effect of the Polymer Network Density Formed in Short Pitch Ferroelectric Liquid Crystal on the Electroclinic Effect

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Effect of the Polymer Network Density Formed in Short Pitch Ferroelectric Liquid Crystal on the Electroclinic Effect

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The influence of the polymer network density formed in short pitch ferroelectric liquid crystal on the electroclinic effect has been analyzed. The polymer network is formed from a photo-reactive achiral monomer in a thin planar S_c^* cell. Using an electro-optic response technique, the electric field dependance of the induced tilt angle and the polarization near to the $S_A - S_c^*$ transition has been measured for different polymer concentrations. The results show that at phase transition temperature the electroclinic effect and the electroclinic susceptibility are largely controlled by the polymer network. These results are discussed and applied to a simple phenomenological model developed in [20]. This model confirms our observations for all of the polymer concentration studied. The average distance between two fibers and the interaction energy between the liquid crystal molecules and the polymer network are estimated. Good correlation is found between the experimental results and the proposed model.

Keywords: elasticity, electroclinic effect, PSFLC, structure and morphology

INTRODUCTION

The development of the flexible liquid-crystal displays using polymer stabilized ferroelectric liquid crystals (PSFLC) is currently of high interest [1]. Polymer-stabilized liquid crystals are composite materials in which a low density of the polymer network is dispersed within

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liquid crystal media [2,3]. The polymer network is formed by chemical crosslinking of a small amount (few percent) of photo-reactive monomers dissolved in low molecular weight mesogenic material, through a polymerization reaction photochemically activated by a UV illumination. When the polymerization occurs in an aligned geometry, the resulting polymer network is roughly aligned parallel to the direction initially imposed by the liquid crystal medium in which the network has been formed [4]. Depending on the type of the reactive mesogen, the morphology of the polymer network may correspond to an open structure consisting of anisotropic fibrils [4–6]. The lateral size of fibrils is of the order of a few tenths of a micron [7–10]; their density increases with the initial reactive monomer concentration. The lateral average distances between two successive fiber decreases when the polymer concentration increases [6]. The Freedericksz transition was affected profoundly by the dispersed polymer network [6]. Aligned polymer fibers of sub-μm-diameter stabilize the liquid crystal alignement and create high speed grayscale capability [1]. The polymer fibrils present a large internal boundary and to provide a bulk anchoring mechanism which allows a control of the liquid crystal alignment in the bulk. Much work has been carried out to understand the interaction between the liquid crystal molecules and polymer network [11]. Dierking *et al.* [11] showed that when polymerization occurs at large tilt angles (polymerization with dc bias field), the liquid crystal molecules preserve these angles when approching the phase transition temperature $S_c^* - S_A^*$. These situations cause the increase of the polarization with polymer concentration. However, when the polymerization occurs in the SmA phase, the induced polarization is reduced [11]. It is well known that if the smectic A is formed by chiral molecules, a coupling between the tilt angle and the electric polarization occurs ($P = K\theta$). Thus an external electric field being applied perpendicular to the layer normal causes the director to tilt thereby inducing a polarization. This is the electroclinic effect or soft-mode. This effect is discovered by Garoff and Meyer [12,13]. Generally the field-induced tilt angle and the polarization display linear behavior with the applied field $E(\theta = e_c E)$, e_c is the electroclinic coefficient. The elecroclinic coefficient and the soft mode susceptibility diverge when approching the transition temperature T_c. The early investigations were all carried out on a pure ferroelectric liquid crystal (FLC) [12,13]. It was not until 1992 that this effect was studied for composites: ferroelectric liquid crystal/polymer especialy in polymer dispersed ferroelectric liquid crystal (PDFLC) by Kitzerow *et al.* [14–16]. The e_c found varied between 0.5 and 2.1 10^{-3} (rad μm V^{-1}) for PDFLC cells, while for the pure compound it is between 7.9 and 20.910^{-3} (rad μmV^{-1}) [17].

Komitov *et al.* [18] have studied the electroclinic effect in PDFLC films containing the liquid crystal which exhibits a high electroclinic coefficient, $e_c = 0.9 - 2.810^{-2}$ (rad μm V^{-1}) [19], and have found $e_c \sim 8.710^{-3}$ (rad μmV^{-1}) for PDFLC films. Recently, we showed that the electroclinic effect in polymer stabilized ferroelectric liquid crystal (PSFLC) is largely controlled by the polymer network [20]. A value of $e_c = 0.26$ (rad μm V^{-1}) was found for pure FLC which exhibits a high polarization (l00 nCcm^{-2}), however, it is about 0.18 (μmV^{-1} rad) with a polymer concentration of 5%. These results are interpreted as a consequence of the strong anchoring of the liquid crystals molecules on the polymer fibers surfaces due to the twist morphology of the polymer fibers [21,20]. In this article, we try to complete the work reported in [20]. For polymer concentration densities varied from 0 to 7% considerable changes on the electroclinic effect is observed. The proposed model [20] confirms all our experimentals results of the induced tilt and polarization as function of the polymer concentration. The average distance between two fibers and the coupling interaction between polymer network and liquid crystal molecules were evaluated for all our studies concentration. This results is in accordance with our estimated values obtained by atomic force microscopic (AFM).

EXPERIMENTS

The liquid crystal compound used in these studies is the ROLIC 8823 (Rolic research Ltd.) which exhibits the following phase sequence: Cr -27°C $S_c{}^*$ 63.5°C $S_A{}^*$ 65°C I. In the $S_c{}^*$, at low temperature, the helical pitch is about 0.3 μm and the spontanuous polarization (P_s) is close to 100 nC/cm^2. The PSFLC mixture was prepared by mixing the Diacrylate monomer with weight concentrations between 2 and 7% (the molecular structure of the monomer is shows in Fig. 1). The FLC compound and the diacrylate were dissolved in the isotropic phase to make a homogeneous mixture. A 2 μm thick EHC-cell (EHC Inc., Japan) with polyimide aligning layers was filled with the mixture in its isotropic phase. In order to obtain a good alignment in the $S_c{}^*$ phase, the cell was slowly cooled-down (0.1°C/minute) from the isotropic phase under an applied electric field (5 V/μm) into the $S_c{}^*$

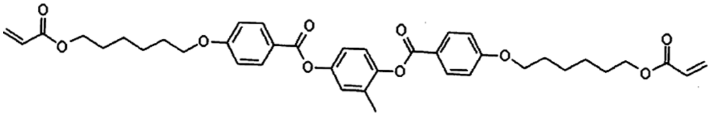


FIGURE 1 The molecular structure of the monomer.

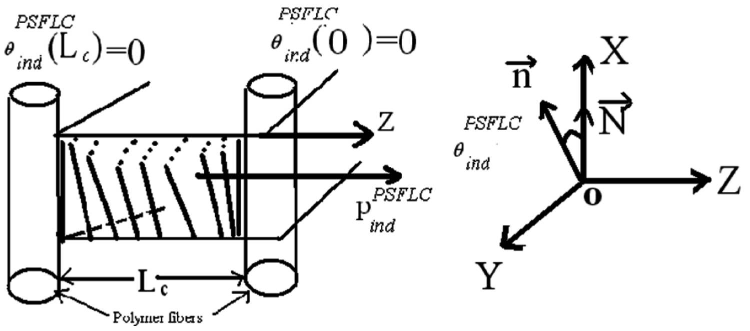
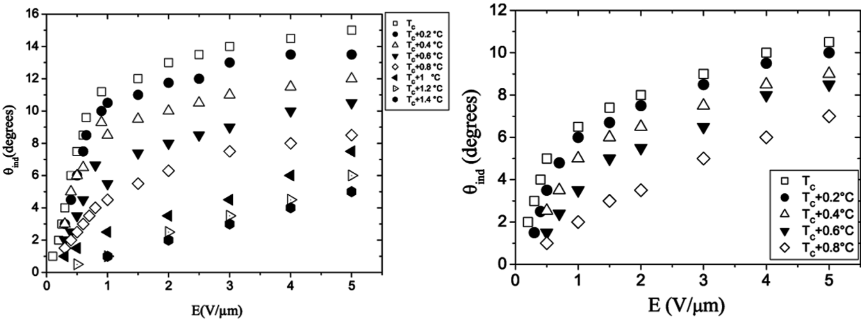


FIGURE 2 Tilt angle induced by electric field (E//Z) in the S_A^* phase; distortion due of the anchorage at the polymer boundaries (a), reference system (OXYZ) Cartesian coordinate to describe the orientation of the liquid crystal director $\vec{n}$ in the presence of the polymer network director $\vec{N}$ (on the right).

phase. The sample cells were then exposed to ultraviolet light (wavelength $\lambda = 365\,\text{nm}$) at 25°C with an intensity of $5\,\text{mW/cm}^2$ for 30 minutes without any applied electric field. During these studies, the cell was placed on a hot stage (Linkam TMS 93) for temperature control. The texture observations of the cells were carried out by means of a polarized optical microscope (POM)(LEICA DMRXP). The estimated values of the distance between fiber was carried out by AFM. The transmitted light intensity through the sample was detected by a PDA55 photodiode (Thorlabs Inc. USA) by means of an optical fiber connected to the optical microscope. The driving electric field was supplied from an HP33120A function generator. The spontaneous polarization was determined from the well-known reverse current method with triangular wave voltage at 50 Hz frequency [22].

ELECTRO-OPTICS RESULTS AND DISCUSSION

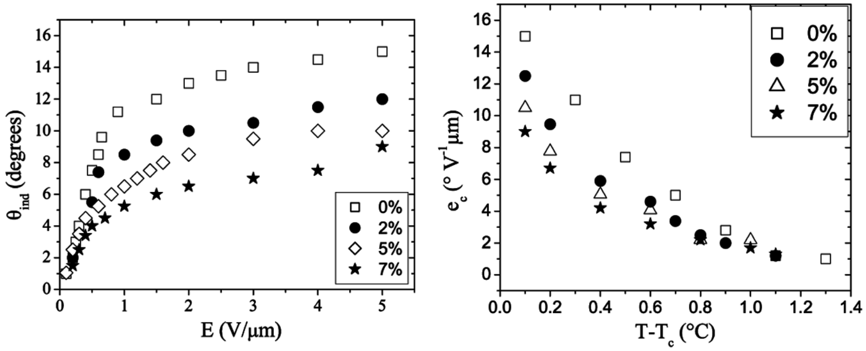
The coupling between tilt angle and the applied electric field are illustrated in Fig. 3 for bulk FLC and the sample with a 7% polymer concentration. As shown in all figures, this behavior is the same as that reported by J. S. Patel [23,24]. Near the $S_c^* - S_A$ transition temperature $T_c{}^*$ the field induced molecular tilt first linearly increases with E for small electric fields, then deviates significantly from linear dependance at moderate E fields. For high fields, θ becomes less elecric field dependance. As temperature increases, the deviation from linear behavior becomes less pronounced. The most important difference in the behavior of the field induced tilt between the FLC stabilized by the surface and that stabilized by anisotropic polymer network is



(a) (b)

FIGURE 3 Electric field dependance induced tilt angle at the bulk FLC (a) and at 7% polymer concentration (b) at different temperatures.

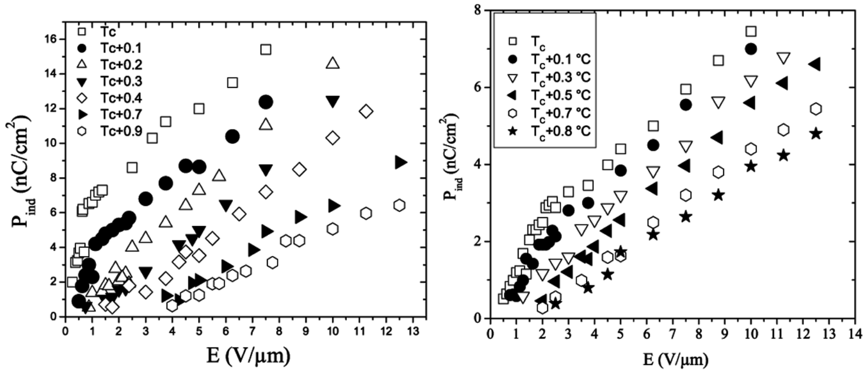
the magnitude of the variation of θ *versus* E. To illustrate the effect of the polymer network on the electroclinic effect we plot the variation of the induced tilt angle near the phase transition temperature T_C. Figure 4(a) shows the result of the induced tilt angle near the phase transition temperature *versus* electric field for several polymer concentrations. As seen in this figure, a sharp decrease of the induced tilt is observed when the polymer concentration increases. At $T_c + 0.1$°C, and at $E = 1\ V/\mu m$, the induced tilt θ are about 15°, 12.4°, 10.5°, and 9° for 0, 2,5 and 7% polymer concentrations, respectively. This result indicates that the polymer network imposes an elastic torque that counteracts the reorientation of the molecular director when an electric field is applied. It is proposed that increases in polymer concentration produces increases the elastic torque. The effect of the polymer concentration on the electroclinic effect is in accordance with the theoretical studies which are reported by Patel *et al.* [23,24], these authors have demonstrated that the induced tilt angle is reduced by increasing elastic constant K [23,24]. To quantitatively characterize the electroclinic behavior we shall focus our attention on the linear parts of θ(E) plots at relatively weak fields. From the gradient of the Fig. 4(a) we obtain the values of the electroclinic coefficients e_c. These values are presented in Fig. 4(b) *versus* temperature and for each polymer concentration. As shown in this figure, the effect of the polymer network on the electroclinic effect is mostly observed near the phase transition temperature T_c. At $T_c + 0.1$°C, we obtain the values of $e_c = 15$, 12.75, 11, and 9 (degrees $\mu m\ V^{-1}$) for 0, 2, 5, and 7% polymer concentrations, respectively. The effect of the polymer network density on the electroclinic effect was confirmed by electric measurements of the field induced polarization P in the smectic A phase. The electric



(a) (b)

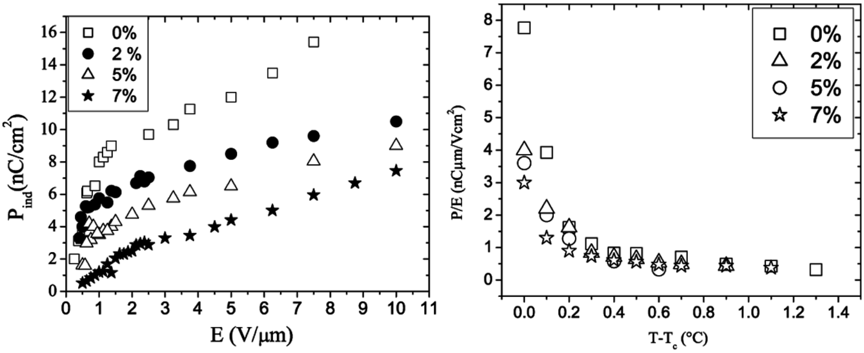
FIGURE 4 Electric field dependance of the induced tilt angle near the phase transition temperature at several polymer concentration (a). Temperature dependance of the electroclinic coefficients (b) at different polymer concentration.

field polarization dependance is presented in Figure 5. As shown in this figure, P *versus* E behavior present a similar behavior compared to the θ *versus E* behavior. Figure 6(a) shows the electric field polarization dependance at phase transition temperataure for several polymer concentrations. As shows in this figure, a sharp decrease of the induced polarization occurs when the polymer concentration increases. An opposite behavior was observed by Dierking *et al.* [11], who showed that when the polymerization occurs in the SmC*, the



(a) (b)

FIGURE 5 Electric field dependance induced polarization at the bulk FLC (a) and at 7% of the polymer concentration (b) for different temperatures.



(a) (b)

FIGURE 6 Electric field dependance of the induced polarization near the phase transition temperature at different polymer concentration (a). Temperature dependance of the electroclinic susceptibility (b) at different polymer concentration.

induced polarization near the phase transition temperature increases with increasing polymer concentration. However, when the polymer network was formed in the S_A phase, the polarization is reduced as function of the polymer concentration. To illustrate the effect of the polymer network on the field induced polarization, we plot in Fig. 6(b) the electroclinic susceptibility; P/E, corresponding to the linear regime (at low field) for all the polymer concentrations studied. As shown in this figure, the contribution on P/E arising from the electroclinic susceptibility for all system is clearly observed near the phase transition temperature [12,13]. As shown in this figure, the electroclinic susceptibility decreases from 8 to 3.5 (nc μm V^{-1} cm^{-2}) when the polymer concentration increases from 0 to 7%, respectively. This behavior confims the behavior of the electroclinic coefficients with polymer concentration (Fig. 4). Figure 6(b) shows clearly the effect of the polymer network density on the electroclinic response especialy near the phase transition temperature. This figure indicates that the polymer network stabilizes the smectic A order. This stabilization is due to the elastic torques which is imposed by the presence of the polymer network. This elastic torques is not uniform between two succesives groups of the polymer fiber in the direction of the applied electric field. The strength of the elastic torques increases with increasing polymer concentration. Due to the strong elastic torques, the reorientation of the molecular director when an electric field is applied is not homogenous across the cell in the Z direction (Z//E). An opposite behavior was observed by Dierking *et al.* [11]. These authors showed that the elastic torques are reduced by the presence of the polymer network which in turn causes an increase of the induced polarization [11]. We can remember here that, although the range of the SmA phase is very narrow, the effect of the polymer network is clearly observed and affects the electroclinic effect. This influence can be explained in the following way. In the smectic A phase, the molecules are aligned in a direction parallel to the normal layer. The stability of this structure is governed by an elastic constant which keeps the long molecular axis parallel to the smectic normal layer. However, due to the thermal energy, the director will be fluctuating, giving locally an instantaneous tilt angle between the director and the smectic normal layer. If the smectic A phase is cooled down and allowed to approach the smectic A–smectic C transition temperature T_c, the elastic constant controlling the tilt fluctuation gets soft. Thus the fluctuation amplitude increases drastically, and its susceptibility diverges at T_c. The softening of this elastic constant means that the phase will lose its stability gradually until it becomes unstable at T_c. When a weak electric field is applied in a direction perpendicular to the director, it can easily perturb the tilt fluctuation depending on how near the system is to T_c. Now, in the presence of the polymer network, due to the strong anchoring of the liquid crystal molecules on the polymer fibers, the elastic constant of the PSFLC cells becomes much higher in comparison with the FLC bulk. This high value of the elastic stress causes a lower fluctuation amplitude in comparison with the fluctuation amplitude in the FLC bulk. As a conclusion, near the phase transition temperature T_c, the electroclinic effect is governed by the elastic stress produced by the presence of the polymer network.

The behavior of the electroclinic response *versus* polymer concentration is in accordance with our previous study [20]. In [21] we showed that the polymer network formed in short pitch FLC presents a twisted morphology. This structure of the polymer fiber is transfered onto network from the helical pitch of the pure FLC during the polymerization process. Due to the strong anchoring of the liquid crystal molecules on the polymer fiber, this twisted morphology causes an elastic force when the director field distorted upon application of the electric field. This elastic force increases with increasing polymer concentration. We therefore assume that the rotation of the director is inhomogeneous across the cell (X direction) and is homogeneous in the Y and Z direction. We neglect then any splay deformation of the director, and we only consider the elastic energy arising from a twist deformation (Fig. 2). This result can be interpreted by the rigid model of the network introduced by Li *et al.* [25]. In [20] we have adopted the model structure of the Li *et al.* In this model, we were considered that the polymer network formed is rigid. The smectic layers are separated by an average distance between two succesive cylinders (Fig. 2).

The equilibrium values of P and θ are found by minimizing the free energy f_t with respect to P and θ, respectively [20]. The ratio $P_s/\theta \approx \chi C$ is found almost constant for all studied concentrations. Averaging the $\theta(z)$ [20] and $P(z)$ values over the $0 \le z \le L_c$ domain gives the expression of the mean induced tilt and polarization:

$$\left\langle \theta_{ind}^{PSFLC} \right\rangle = \frac{\epsilon_0 \chi C E}{\alpha(T - T_c')} \left[1 - \frac{\tan\mathrm{h}(L_c/2a)}{(L_c/2a)} \right] \tag{1}$$

$$\left\langle P_{ind}^{PSFLC} \right\rangle = \epsilon_0 \chi C E + \frac{\epsilon_0 \chi^2 C^2 E}{\alpha(T - T_c')} \left[1 - \frac{\tan\mathrm{h}(L_c/2_a)}{(L_c/2_a)} \right] \tag{2}$$

where

$$a = \sqrt{\frac{K_2}{\alpha(T - T_c')}}. \tag{3}$$

T_C' is the phase transition temperature of the PSFLC films and can be written as $T_c' = T_c + \frac{W_p}{\alpha}$ [20].

Equations 1 and 2 show that, in the S_A^* phase, the main parameter that governs the electroclinic effect is the distance between two successive groups of the polymer fibers separated by the average distance L_c (Fig. 2). According to Eqs. (1) and (2), an increase in polymer concentrations lead in the reduction of the electroclinic effect.

The electroclinic coefficient in the case of the cell stabilized by the polymer network is given by [20]:

$$e_C^{PSFLC} = \frac{\epsilon_0 \chi C}{\alpha(T - T_c')} [1 - H]. \tag{4}$$

The most interesting result is the role played by the polymer network for stabilized of the smectic A order. At $T_c + 0.1$°C, the electroclinic coefficient values are 15, 12.75, 11, and 9 (°μmV) for 0, 2, 3.5, 5, and 7% polymer concentrations, respectively (Fig. 4(b)). We used the values of $\alpha \approx 8.8 \times 10^{+3}\mathrm{N/m^2K}$ and $a = 0.13\,\mu\,\mathrm{m}$ which were evaluated at $T - T_c = 0.1$°C [20], with a reasonable value of the twist elastic constant, typically, $K_2 \approx 10^{-11}\mathrm{N}$. Using this value for the elastic constant K_2, and the values of the parameters α and a and data of Fig. 4(b), Eq. (4) was graphically resolved to evaluate L_c. We obtain a mean interfiber distance $L_c \approx 1250\,\mathrm{nm}, 1100\,\mathrm{nm}, 960\,\mathrm{nm},$ and $780\,\mathrm{nm}$ for 2, 3.5, 5, and 7% polymer concentrations, respectively (Fig. 9(a)). These calculated values are in accordance with those measured by means of

AFM experiments. The tapping mode AFM image of a $40 \times 40\,\mu m^2$ region of the polymer network, and the corresponding height profile are presented in Figs. 7 and 8 for 2 and 7% polymer concentrations respectively. Figure 7 (lower part) shows an example of the height profile for sample polymer concentration of 2%. We can distinguish from the height profile two successives groups of fibers in the z direction. The average distance between them was evaluated to about 1300 nm, which is in accordance with the calculated value. For all polymer concentration studies, the estimated values of L_c by AFM are presented in Fig. 9(a). This figure shows that an increase of the polymer

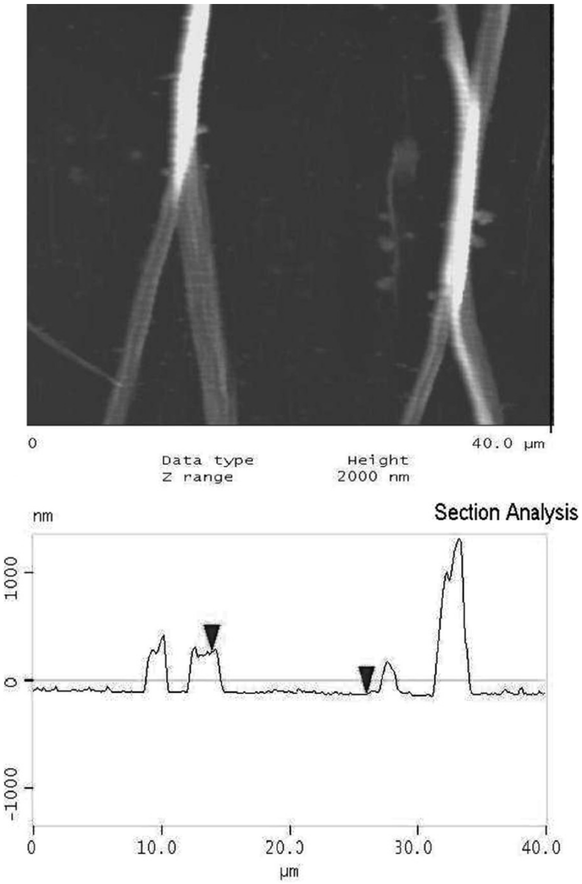


FIGURE 7 Tapping mode AFM images of polymer network strucure of $40 \times 40\,\mu m^2$ (upper) and the height profile of the network structure (lower part) of the 2% polymer concentration.

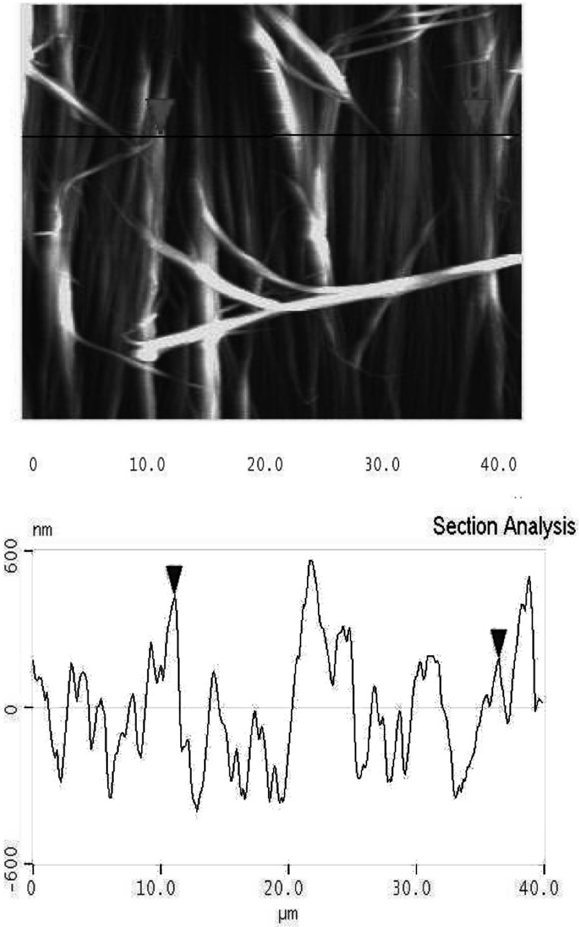
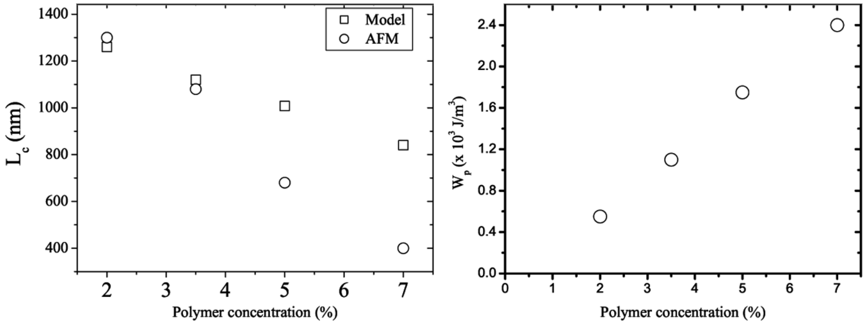


FIGURE 8 Tapping mode AFM images of polymer network strucure of $40 \times 40\,\mu\text{m}^2$ (upper) and the height profile of the network structure (lower) of the 7% polymer concentration.

concentration leads in the reduction of the L_c. This figure also shows that the values of L_c obtained by AFM are in a accordance with that evaluated by theoretical model. According to expression $\Delta T_c = T_c - T'_c = \frac{W_p}{\alpha}$, the coupling coefficient, W_p, characterizing the interaction energy between the FLC and the polymer network is determined, the smectic C^*-smectic A^* phase transition temperature is shifted by introducing the polymer network and especially in the direction of the $S_c{}^*$ phase. This shift is due to the strong interaction between



(a) (b)

FIGURE 9 The average distance between fibers L_c (a) and the coupling interaction W_p in the SmA phase (b) as a function of polymer concentration.

polymer network and liquid crystal molecules. Because of the confining geometry, we expect that the smectic A–smectic C* phase transition temperature T_c in the confining geometry is lower than the corresponding phase transition temperature in the bulk. An opposite behavior has been observed by Dierking *et al.* [11] who show that when the polymer network formed in the S_{C^*} phase, the phase transition temperature is shifted as function of polymer concentration in the direction of the S_{A^*} phase. However, when the polymer network formed in the $S_C{}^*$ phase, the phase transition temperature is shifted in the direction of the S_{A^*} phase. When the polymer concentrations increase from 0 to 7%, ΔT_c was found to vary from 0.2°C to 0.6°C, respectively. W_p linearly increases with polymer concentration (Fig. 9(b)). The values of the W_p which are found here are higher comparable with the values (10–80 J/m^3) reported by Dierking *et al.* [11] in some other PSFLC systems, and is within an order of magnitude than those reported by Furue *et al.* [26] and Li *et al.* [25].

CONCLUSION

In this work, we report the influence of the polymer concentration on the electro-clinic effect of the short pitch ferroelectric liquid crystals stabilized by the polymer network. Our results clearly show that the electroclinic switching is observed at all polymer concentrations studied. Our measurements reveal that an increase in the polymer concentration leads to a decrease in the electroclinic effect. To describe the reduction of the electroclinic effect, we have adopted a one-dimensional model developed in [20]. This model takes into account both the coupling interaction between the polymer network and the liquid crystal, and the elastic energy due to the distortion of the liquid crystal medium in the presence of an applied electric field. The average distance between fibers and the coupling energy between the liquid crystal molecules and the polymer are evaluated for different polymer concentrations.

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