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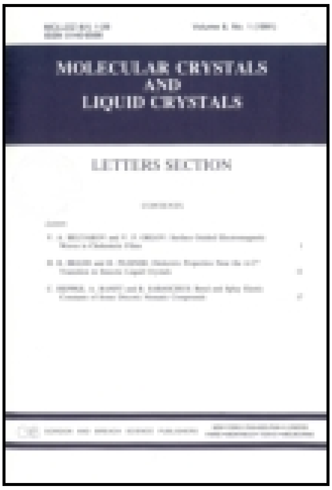

Molecular Crystals and Liquid Crystals
Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gmcl20

Electro-Optical Study of Fluorescent Dye-Doped Ferroelectric Liquid Crystal

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Published online: 08 Apr 2014.

To cite this article: Abhishek Kumar Misra, Pankaj Kr Tripathi, Kamal Kr Pandey & Rajiv Manohar (2014) Electro-Optical Study of Fluorescent Dye-Doped Ferroelectric Liquid Crystal, Molecular Crystals and Liquid Crystals, 591:1, 25-33, DOI: 10.1080/15421406.2013.830353

To link to this article: http://dx.doi.org/10.1080/15421406.2013.830353

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Mol. Cryst. Liq. Cryst., Vol. 591: pp. 25–33, 2014
Copyright © Taylor & Francis Group, LLC
ISSN: 1542-1406 print/1563-5287 online
DOI: 10.1080/15421406.2013.830353

Electro-Optical Study of Fluorescent Dye-Doped Ferroelectric Liquid Crystal

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The present paper reports the electro-optical behavior of a pure and fluorescent dye-doped ferroelectric liquid crystal (FLC) system. Different material parameters, such as spontaneous polarization and optical response time, have been analyzed by electro-optical study. The temperature dependence of these parameters have been discussed for the pure and the fluorescent dye-doped systems. The spontaneous polarization and optical response time have also been found to change strongly by the variation of concentration of the fluorescent dye into the pure FLC. In addition to these parameters, the present paper also reports the temperature dependence of the effective dielectric permittivity for the pure and fluorescent dye-doped FLC systems. The value of spontaneous polarization has improved after doping of fluorescent dye suggesting that fluorescent dye is beneficial for this FLC material.

Keywords Effective dielectric permittivity; fluorescent dye; material parameters; spontaneous polarization

1. Introduction

Ferroelectric liquid crystal (FLC) is an important LC among the all types of LCs which possesses exceptional electro-optical properties with switching time in microsecond [1–4]. In spite of some drawbacks, FLC became a good option for commercial video applications [3, 4]. FLCs have features such as fast response and wide viewing angle that makes them attractive for use in flat panel displays [4–6]. Recently with increasing use of portable displays, such as notebook computer, mobile phone, personal tablets, privacy protection has become an important issue. Therefore, liquid crystal displays with a narrow viewing angle are required [7].

Dye-doped liquid crystals (DDLC) represent an exciting class of functional materials retaining the useful electro-optical properties of LCs and adding further unique power abilities and functionalities [8]. DDLCs provide wide range applications such as privacy windows, reflecting display devices, and dynamic holography. Dispersion of various dyes into FLCs has shown tremendous results due to the unique properties of dyes [9–11]. In

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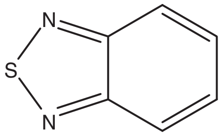

Figure 1. Molecular structure of the fluorescent dye Benzo 2, 1, 3 Thiadiazole used for investigation.

recent time, among the varieties of dyes, fluorescent dyes are currently used for memory-based applications. The dispersion of fluorescent dye in FLCs has shown considerable improvement in existing properties of the pure FLC material [12–14]. In the beginning, dispersion of fluorescent dye into pure FLC was focused on the dielectric properties of the system [14].

In the present work, efforts have been made to study the effect of fluorescent dye on the electro-optical properties of the pure FLC. The effect of fluorescent dye on optical response time is also subject of our current interest. In addition to this, we have also discussed spontaneous polarization and effective dielectric permittivity. The temperature and applied voltage dependence behavior of response time and spontaneous polarization have been reported for pure FLC and fluorescent dye-doped FLC system.

2. Experimental Details

One commercially available FLC 16/100, purchased from Clariant Chemicals Co. Ltd., Germany, was used as host material. The phase sequence of the FLC material is Cr- SmC* -SmA - N*- Iso at –20°C, 74°C, 82°C, and 90°C–94°C, respectively. For the pure FLC 16/100, the standard values of spontaneous polarization (Ps), cone angle ($2\theta = 2 \times$ Tilt Angle), and rotational viscosity (γ) at 25°C was –10.5 nC/cm^2, 54.30^0, and 60 mPas, respectively [15, 16]. The fluorescent dye used for doping was Benzo 2,1,3 Thiadiazole, purchased from Sigma Aldrich (India), with the chemical structure as shown in Fig. 1 [13, 14].

Highly conducting (~20 Ω/□ mm) indium tin oxide (ITO)-coated glass plates were used to make sample cells. The desired electrode pattern on the ITO substrates was achieved using a photolithographic technique. The active electrode area was 7 mm × 7 mm in the present case. Uniform thickness (6 μm in our case) of both the cells was maintained by means of Mylar spacer. The planar alignment was obtained on polymer (Nylon 6/6)-coated glass plate by unidirectional rubbing. The empty sample cells were calibrated using analytical reagent (AR) grade carbon tetrachloride (CCl_4) and benzene (C_6H_6). The detailed procedure for preparation of the sample cell has been discussed in our earlier papers [15–18].

The fluorescent dye-doped FLC system was prepared by the mixing of a fluorescent dye in the 1% wt/wt and 2% wt/wt ratio with that of the pure FLC and these samples were denoted as Mixture 1 and Mixture 2 in the whole manuscript. The pure FLC and the fluorescent dye-doped FLC were filled in the isotropic phase by means of capillary action. Then this was cooled gradually upto room temperature with the application of a small a.c. electric field [13–15]. The alignment of the samples was checked by the polarizing microscope under the crossed polarizer-analyzer arrangement [16–18].

The dielectric data was determined by measuring the value of capacitance and the dissipation factor measured with the help of impedance/gain phase analyzer HP-4194A in the frequency range 100 Hz to 10 MHz. In this measurement, there is accuracy of ±0.17% for the measurement of parameters. The temperature control is very essential for the FLC because the different phases exist at different temperatures. The temperature was maintained with the help of Instec Hot Plate HCS 302 with temperature stability of ± 0.01°C. The spontaneous polarization (Ps) was measured by the polarization current reversal peak method by using function generator (Tektronix AFG3021B) and storage type oscilloscope (Tektronix TDS2024C). For the spontaneous polarization measurement, a triangular wave of amplitude of 20 Vpp and frequency 10 Hz was used [8, 9, 11, 19]. The spontaneous polarization was determined by the relation Ps calculated by using the following relation [11, 19]:

$$Ps = \frac{1}{2\mathrm{A}} \int i(t)dt, \tag{1}$$

where $\int i(t)dt$ is the area under the current bump and A is the active area of the sample. For the measurement of response time (τ), a square wave signal of 10 Hz and 20 V_{PP} was used [11, 13, 19]. The response time was determined by using optical switching method [8, 13]. In this method, a square wave of frequency 1 Hz and amplitude 20 V peak to peak is applied to the sample cell. The optical response of molecules as observed by the detector was fed to a storage oscilloscope (HM 407) in electrical form. The output waveform was now used to determine the response time. The response time of FLCs was evaluated using $\tau = \frac{t_{90} - t_{10}}{1.8}$. Here, t_{90} and t_{10} are the time taken by the output waveform to reach 90% and 10% of the maximum of the output waveform [8, 13]. Rotational viscosity was calculated by using spontaneous polarization data and response time as follows [8, 13]:

$$\tau = \frac{\gamma}{P_S E}. \tag{2}$$

Here τ, γ are optical response time and rotational viscosity of the sample, respectively; P_S is spontaneous polarization of the material and E is applied electric field.

3. Results and Discussion

Figure 2 shows the relation between temperature and spontaneous polarization for pure FLC and fluorescent dye-doped FLC with different doping concentration at frequency 10 Hz and at $V_{\mathrm{pp}} = 20$ volt. The horizontal axis is the temperature for different doping concentration of dye. It is clear that that spontaneous polarization decreases rapidly with reaching minimum as the temperature reaches to 70°C and then sample goes into SmA phase. Figure 2 shows the same trend, i.e., the magnitude of spontaneous polarization is continuously decreasing and it vanishes at SmC* -SmA phase transition point or near this phase transition point [9, 11, 13, 20–22] for all the mixtures, but there is a drastic difference in Mixture 1 and Mixture 2. The value of spontaneous polarization for Mixture 1 has increased to almost double in comparison to the pure FLC, while the value of spontaneous polarization has decreased for the Mixture 2 as compared to Mixture 1 with increase in concentration of fluorescent dye. The value of spontaneous polarization for Mixture 2 is also high as compared to pure FLC. The reason for this increment is that whenever fluorescent dye is introduced in pure FLC, it tries to fit in the geometry of FLC system. The dipole moment of fluorescent dye and FLC molecules may be in the same direction or in the opposite direction; whenever dipole moments of fluorescent dye and FLC molecules are in the opposite direction the net spontaneous polarization decreases, but if the directions are the same then spontaneous polarization increases (as in our case). The increment in the value of spontaneous polarization after doping of fluorescent dye into FLC

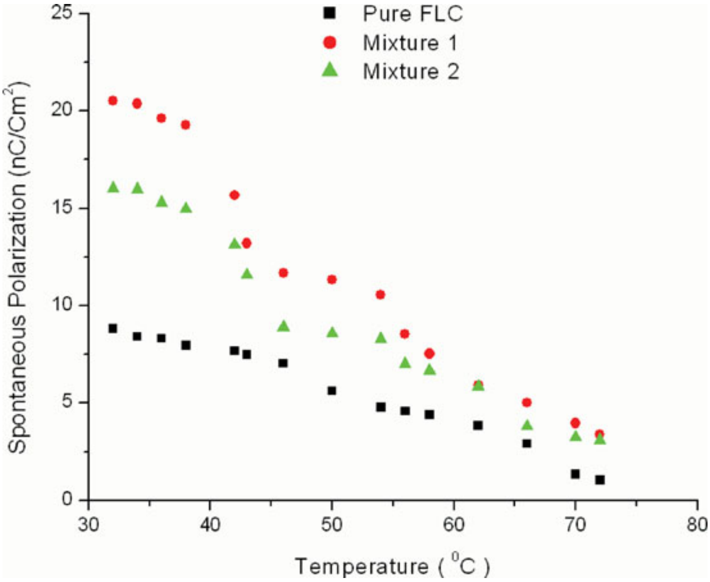

Figure 2. Temperature variation of spontaneous polarization for the pure FLC, Mixture 1, and Mixture 2.

shows improvement in the optical response; therefore, this fluorescent dye is very useful for improving the response time of FLC material. The alignment was checked by placing sample cell between two-crossed polarizers of polarizing microscope CENSICO 7626. We have performed dielectric measurement to calculated conductivity σ(dc) term for pure FLC and dye-doped FLCs. The value of σ(dc) term has been obtained by fitting the dielectric data into Cole–Cole equation. We have found that conductivity increases for dye-doped FLCs in comparison to pure FLC. The value for all the samples has been given in Table 1.

In order to understand the sensitivity of the applied voltage [23, 24] to the doping concentration of the fluorescent dye, we observe the optical response time. In general, the response time of the FLC is much faster when using higher values in the electric field. The electric field applies an external torque to each molecule but when the field is removed or reduced, interactions between LC molecules provide the dominant restoring forces. These interactive forces are much weaker than the torque caused by an external electrical field, which leads to a slower relaxation time. The relaxation time of the molecules dominates the overall response time in FLC. Not all molecules feel weak forces equally

Table 1. Variation of rise, fall and response with concentration of dye at constant temperature of 35°C

Sample	Ionic conductivity σ (dc) in Ω^{-1} cm^{-1}	Rise time (in μs)	Fall time (in μs)	Response time (in μs)
Pure	$6.13 \times 10^{-9} + 0.043$	52	268	320
Mixture 1	$7.02 \times 10^{-9} + 0.051$	58	269	327
Mixture 2	$8.11 \times 10^{-9} + 0.049$	97	325	422

when the external electrical field is removed. Molecules close to the bonding surfaces feel the strongest restoring force and have the fastest temporal response. They also require the highest voltages to switch.

The value of rise time, fall time, and response time at constant temperature for pure FLC, Mixture 1 and Mixture 2 has been presented in Table 1 at constant temperature of 35°C, frequency 10 Hz, and Vpp (peak to peak voltage) = 20. The value of total response time (i.e., the sum of rise and fall time) is 320 μs, 327 μs, and 422 μs for pure sample, Mixture 1 and Mixture 2, respectively.

It can be inferred that for Mixture 1, i.e., low doping concentration of fluorescent dye having highest spontaneous polarization under the same electric field will have smaller response time. On the other hand while the spontaneous polarization of Mixture 2 is lower than Mixture 1 the response time value is much increased. The reason is that behavior of response time is governed by spontaneous polarization and rotational viscosity. The value of spontaneous polarization is increased for fluorescent dye-doped sample as compared to the pure FLC. The value of spontaneous polarization for Mixture 1 is higher as compared to pure FLC and Mixture 2. This means that response time should be lowest for Mixture 1 due to largest value of spontaneous polarization but value of response time is higher for Mixture 1 in comparison to the pure FLC. This is due to the fact that rotational viscosity is much increased for Mixture 1 as compared to pure FLC (discussed in later part of the article) because rotational viscosity is dominant over spontaneous polarization value. The interplay of these two leads an increase in the response time for fluorescent dye-doped systems. Taking this into consideration the rise and fall time performances of fluorescent dye doping concentration, we can conclude that Mixture 1 is optimal.

The response time (τ) of the pure and the doped system is plotted on the temperature scale in Fig. 3 at frequency 10 Hz and 20 V. The response time of the pure and the doped system decrease with increasing temperature [13, 14, 23]. The response time for the doped

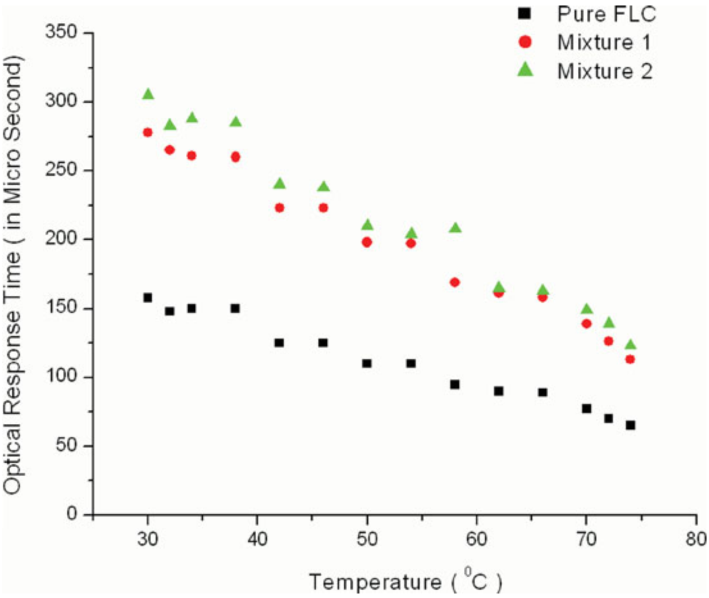

Figure 3. Temperature variation of optical response time for the pure FLC, Mixture 1, and Mixture 2.

system is higher than that of the pure FLC because of the presence of fluorescent dye particles. The presence of fluorescent dye particles creates additional constraints in the molecular motion of the FLC molecules under the influence of applied field. Due to these constraints, the surface anchoring energy of the doped FLC system reduces. The two types of surface anchoring energies, which define the anchoring effects in liquid crystals system, include the polarization and dispersion contribution. The dispersion anchoring energy is defined as energy per unit area for a 90° rotation in the substrate plane, whereas polar anchoring energy is defined as energy per unit area required to rotate the near surface director by 90° out of plane. The anchoring energy coefficient is inversely proportional to the optical response time. Therefore, FLC molecules do not respond quickly with the applied field same as in case of the pure FLC molecules and a higher response time for the fluorescent dye-doped FLC composite system is required.

The temperature dependence of the effective dielectric permittivity at constant frequency 1000 Hz for the pure FLC, Mixture 1 and Mixture 2 has been shown in Fig. 4. Here term effective is used for the dielectric permittivity for which we have removed contribution of sheet resistance, ITO coatings, and lead inductance of the cell [13, 14, 25]. Therefore, the data require correction in the Cole–Cole equation [25–27]. The Cole–Cole equation is given as

$$\varepsilon * = \varepsilon'(\infty) + \frac{\delta\varepsilon'}{1 + (i2\pi f\tau)^{1-\alpha}}, \tag{3}$$

where $\delta\varepsilon'$ is the dielectric strength and $\varepsilon'(\infty)$ is the high-frequency limit of the dielectric permittivity, f is frequency, α is the distribution parameter, and τ is the relaxation time. After adding correction terms due to the ionic conductivity and electrode polarization problem in the low-frequency region while ITO sheet resistance and lead inductance problem in the high frequency region data, the real and imaginary parts of the above equation become as

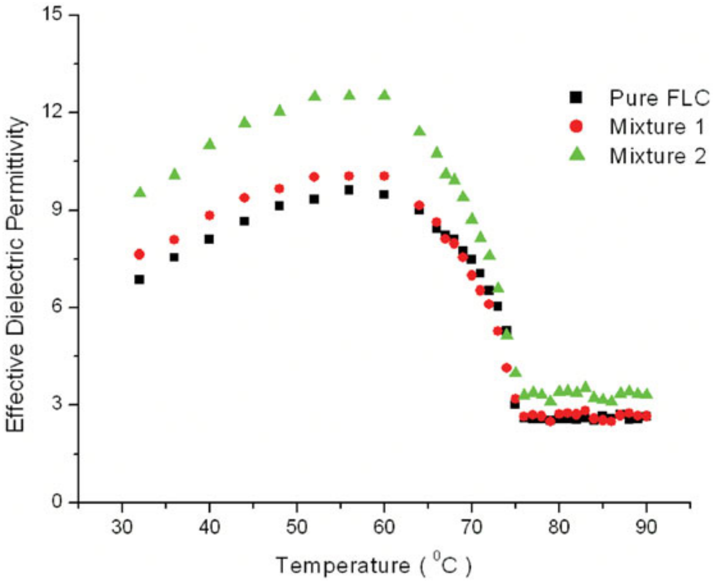

Figure 4. Temperature variation of effective dielectric permittivity for pure FLC, Mixture 1, and Mixture 2.

follows:

$$\varepsilon' = \varepsilon'(dc)f^{-n} + \varepsilon'(\infty) + \frac{\delta\varepsilon'[1 + (2\pi f\tau)^{(1-\alpha)}\sin(\alpha\pi/2)]}{1 + (2\pi f\tau)^{2(1-\alpha)} + 2(2\pi f\tau)^{(1-\alpha)}\sin(\alpha\pi/2)} \tag{4}$$

and

$$\varepsilon = \frac{\sigma(\mathrm{dc})}{\varepsilon_0 2\pi f^k} + \frac{\delta\varepsilon'(2\pi f\tau)^{(1-\alpha)}\cos(\alpha\pi/2)}{1 + (2\pi f\tau)^{2(1-\alpha)} + 2(2\pi f\tau)^{(1-\alpha)}\sin(\alpha\pi/2))} + Af^m. \tag{5}$$

Here, $\sigma(dc)$ is the dc ionic conductance, ε_o is free space permittivity, and f is the frequency while n, m, and k are the fitting parameters. The terms $\varepsilon^1(dc)f{-}n$ and $\sigma(dc)/\varepsilon_0 2\pi f^k$ are added in the Eqs. (4) and (5) for additional contribution due to the electrode polarization and ionic conductance in the low-frequency effect [10, 13]. The $Af\,m$ term is added in Eq. (5) in the high-frequency effect due to the contribution of ITO sheet resistance and lead inductance of sample cell. By the least square fitting of experimental data we have removed these contributions and we got effective dielectric permittivity.

The effective dielectric permittivity of the pure FLC increases slightly with increase in temperature [13, 16, 25]. The maximum value of dielectric permittivity for the pure FLC has been noticed at 60°C. Any further increase in temperature causes a small decrease in the dielectric permittivity. A sharp decrement in the value of dielectric permittivity has been observed at the vicinity of SmC*-SmA phase transition temperature. This sharp decrease in the dielectric permittivity is due to the change in geometry of the phase. This situation arises when fluorescent dye molecules are added in the matrix of pure FLC, they try to fit into the molecular geometry of pure FLC. The presence of fluorescent dye molecules in FLC matrix perturbs the intermolecular interaction field of intrinsic molecular geometry and offers some additional constraints to the molecular dynamics of pure FLC. These perturbations take place in the system and they can affect the molecular properties of the system. We can see from the figure that there was a little difference in the permittivity for the different liquid crystal phase of pure FLC and Mixture 1 while for Mixture 2 there was large difference (approx 1.4 times) in the value of permittivity than others. This is due to this fact that when fluorescent dye is doped into the pure FLC then there is a tendency of dye molecules to fit in the geometry of pure FLC. The dipole moment of dye molecules is in the same direction supporting the dipole moment of pure FLC molecules enhancing the value of effective dielectric permittivity. Therefore, one can say that doping of fluorescent dye will effectively enhance the dielectric permittivity of pure FLC with its large dipole moment. The doping of fluorescent dye had also enhanced its sensitivity to applied electric field.

The response time behavior for fluorescent dye has been checked by plotting the temperature dependence of rotational viscosity for all the samples and has been shown in Fig. 5. The temperature dependence of electro-optical rotational viscosity is similar as reported in the earlier papers for other FLCs [11, 13], but the magnitude of electro-optical rotational viscosity for fluorescent dye-doped FLC increase as compared to the pure FLC. It is clear that rotational viscosity increases with doping of fluorescent dye and decreases with increasing temperature near the SmC*–SmA phase transition. For a pure FLC at the SmC*–SmA phase transition, the rotational viscosity starts to exhibit a minimum value due to the cone angle, which is approximately zero at this point. Actually, when fluorescent dye molecules are added in the matrix of a pure FLC, they try to fit into the molecular geometry or molecular structure of the pure FLC. Therefore, rotational viscosity for Mixture 1 is increased as compared to pure FLC and Mixture 2. For Mixture 2, rotational viscosity is

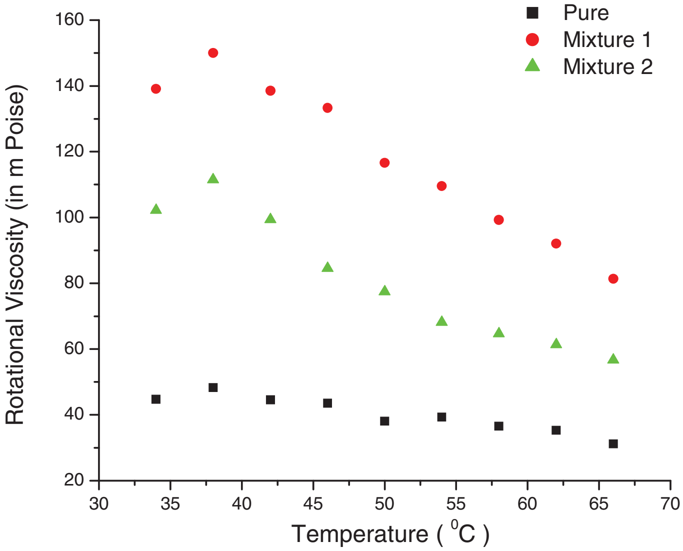

Figure 5. Temperature variation of rotational viscosity for pure FLC, Mixture 1, and Mixture 2.

increased in comparison to pure FLC but its value is decreased in comparison to Mixture 1. This is due to the fact that when dye molecules cannot fit into molecular geometry of the pure FLC, then the dye molecules will obstruct the motion of FLC molecules; hence, the value of rotational viscosity decreases for Mixture 2 in comparison to Mixture 1. This is due to the reason that the Goldstone mode of relaxation in FLCs is due to the phase fluctuation, therefore Zth component of rotational viscosity is the only active component of γ. Hence, the resultant of rotational viscosity of Goldstone mode strongly depends upon the values of tilt angle, which is decreasing considerably for higher concentration of dye in pure FLC due to the fact that the rotational viscosity decreases for Mixture 2.

4. Conclusion

Comparative study of pure FLC and fluorescent dye-doped system is described in the present paper. The fact that the magnitude of spontaneous polarization increases for low concentration of the dye is an indication of achieving good molecular alignment, which is also clear from the behavior of effective dielectric permittivity. The increment in spontaneous polarization with applied voltage variation suggests that doping of fluorescent dye had significantly improved the sensitivity of FLC to an applied electric field. The response times for Mixture 1 and Mixture 2 have also been improved in comparison to the pure FLC. We have achieved fast optical response time for fluorescent dye-doped FLC because low optical response time is considered as a drawback for fluorescent dye-doped system to use in display of high quality moving pictures. By considering the rise and fall time and spontaneous polarization performances of fluorescent dye doping concentration we can conclude that Mixture 1 is optimal.

Acknowledgments

Dr. Abhishek Kumar Misra is thankful to UGC, New Delhi, for the IIIrd year grant of Dr. D. S. Kothari Post-Doctoral Fellowship No. F.4.2/2006 (BSR)-13-234/2008(BSR). The authors are also thankful to DST, New Delhi, for providing project SR/FTP/PS-091/2012

entitled “Ferroelectric Liquid Crystals versus Fluorescent Dye Doped Ferroelectric Liquid Crystals.”

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