



Contents lists available at ScienceDirect

Opto-Electronics Review

journal homepage: <http://www.journals.elsevier.com/geo-electronics-review>

Dispersion of fluorescent dye in the nematic liquid crystal: Enhanced photoluminescence and high birefringence

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ARTICLE INFO

Article history:

Received 31 May 2018

Received in revised form

20 September 2018

Accepted 22 October 2018

Available online 23 November 2018

Keywords:

Nematic liquid crystal

Fluorescent dye

Photoluminescence

Birefringence

Dielectric anisotropy

ABSTRACT

In the present investigation optical, electro-optical and dielectric properties have been measured for nematic liquid crystal (NLC) material 1550C which consists of 4'-(trans, trans-4-alkylbicyclohexyl) carbonates and 4'-(4-(trans,trans-4-alkyl)-4-cyanobicyclohexane, dispersed with fluorescent dye (Benzo 2,1,3 Thiadiazole) in two different concentrations. Photoluminescence has been enhanced for a dye dispersed system which is the key finding of this investigation. UV absorbance study has also been performed and found to be increased for composite system. Enhanced birefringence after dispersion of dye into pure NLC is also a prominent result of this investigation. Relative permittivity, threshold voltage and dielectric anisotropy have also been measured and found to be increased. The outcome of the present work may be very useful in the construction of liquid crystal displays (LCDs).

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1. Introduction

Liquid crystals (LCs) are very important for displays, as well as non-display devices. There are several phases of LCs and the most important phase are nematic liquid crystals (NLCs). NLCs have preferred birefringence. NLCs can be easily aligned by the applied electric field and aligned nematics are useful for liquid crystal displays (LCDs). The liquid crystal dispersed with dye is often used for opto-electronic devices and photo induced refractive index gratings [1–3]. This NLC-dye system has also many applications in displays. Display features can be improved by synthesize a new liquid crystal material [4] or by dispersion of dyes [5,6], polymers [7], and nanoparticles [8–10] in LCs. To enhance the optical parameters, dye may play a very much important role as a dopant. Communication tools which require less energy desire fluorescent guest hosts LCDs [11–14]. We can enhance different important parameters of NLC by doping fluorescent dye. So, it is necessary to study the NLC-dye composite system. The dye used in this study is extremely fluorescent [15]. Maximum researches on dye-doped liquid crystals are focused around those properties that affect the performance of practical devices directly [16,17]. Doping of dye into the pure NLC changes the molecular dynamics of pure NLC. Orientation ordering of dye dispersed NLC system will depend on the shape of LC molecules and dye molecules and also

the intermolecular attraction arises from anisotropic short-range and long range forces [18]. Since nematic liquid crystal materials are anisotropic in nature, therefore, if we change the director of NLC materials, then we can change many physical parameters. The relative permittivity perpendicular to the long molecular axis (ϵ_{\perp}) is minimum for a perfectly aligned NLC. If any disturbance produces in alignment, then the increment in the ϵ_{\perp} will occur due to contribution from relative permittivity parallel to the axis ($\epsilon_{||}$). Therefore, dielectric anisotropy ($\Delta\epsilon = \epsilon_{||} - \epsilon_{\perp}$) and birefringence vary throughout the same liquid crystal cell. It has several potential applications [19].

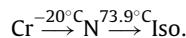
Analysis of dye-nematic composite system for photoluminescence and birefringence property is very noteworthy. Measurement of luminescent property of any display device is very significant. In this study, optical, electro-optical and dielectric parameters of the pure NLC and dye dispersed system have been reported. Generally, nematic mesophase gives photoluminescence (PL) behaviour, therefore, it is important to investigate this property. Therefore, we have measured the photoluminescence property in this study for a pure and dispersed system. UV-visible absorbance study has also been performed here. We have also measured the birefringence in this reported work. The process and method of calculation of birefringence property has been described in detail in the current investigation. We have also measured relative permittivity and dielectric anisotropy in this work. The results of this investigation may be applicable in liquid crystal displays.

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2. Experimental details and methods

2.1. NLC Material

The liquid crystalline material used in this investigation is NLC 1550C, composed of 4'-(trans,trans-4-alkylbicyclohexyl) carbonates and 4'-(4-(trans,trans-4-alkyl)-4-cyanobicyclohexane. The used NLC 1550C in this investigation is given by our research group from the Military University of Technology, Poland. This NLC is a low birefringent material. The inherent properties of NLC 1550C are $\Delta n = 0.059$ (20°C , 589 nm); $n_0 = 1.461$ (20°C , 589 nm) [20–22]. The phase sequence of the NLC 1550C has been given below:



Where 'Cr' represents the crystal phase, 'N' represents the nematic phase and 'Iso' represents the isotropic phase of the NLC 1550C.

2.2. Fluorescent Dye

The dopant used for the present investigation is a fluorescent dye (Benzo 2,1,3 Thiadiazole), which is a disc shape dye, purchased from Sigma Aldrich [23,24]. Here we can see from this Fig. 1 that this disc shape dye consists of six carbon (C) atoms and four hydrogen (H) atoms attached with two nitrogen (N) atoms and one sulphur (S) atom. Dyes are those chemicals which show colour effect and absorb certain wavelength strongly.

2.3. Preparation of NLC-Dye composites and LC cell

In the present work, we have dispersed a fluorescent dye in NLC to measure its optical, electro-optical and dielectric nature. Fluorescent dye has been dispersed in pure NLC with two weight percentage: 0.5% (mix 1) and 1% (mix 2). Fluorescent dye has been dispersed in toluene for proper mixing. Then, it is mixed with NLC 1550C. Next, we have heated the sample again to evaporate solvent completely from the sample. We have used planar aligned LC cells having thickness of $8\text{ }\mu\text{m}$ for this investigation. Indium tin oxide (ITO)-coated glass plates have been used to make LC sample cells. To make planar aligned cell, nylon (6/6) has been put onto the surface of two ITO coated glass plates and, then, we have rubbed unidirectionally. The substrates were then placed one over another. Mylar spacer has been placed between two substrates for fixing the cell thickness. The liquid crystal material was filled into the cell by

using capillary method. The details of LC cell preparation have been already reported by our group [25].

2.4. Instruments Used

We have taken the optical textures by using a polarising optical microscope (POM) (progress CT3, Radical). Cary eclipse fluorescence spectrophotometer (Agilent technologies) has been used for photoluminescence (PL) study for a 347 nm excitation wavelength at room temperature. We have used UV-Visible Spectrophotometer (ELICO, SL 210) for UV-visible absorption at room temperature. We have performed the FTIR study in the wavelength range of 400–4000 cm^{-1} by using Fourier Transform Infrared Spectrophotometer (IR affinity-1 Shimadzu). For the measurement of dielectric anisotropy, we have used HP4194A Impedance gain/phase analyser in the frequency range of 100 Hz to 40 MHz. To control the temperature of the cell we have used INSTECH mK 2000 hot plate. The meticulous experimental details have already been reported by our group [26–29].

2.5. Birefringence measurement

The experimental setup of birefringence measurement for pure NLC and dye doped system has been given in Fig. 1. Here we can see from this figure that when a plane polarized light falls onto the liquid crystal material (anisotropic material), its splits into two components due to birefringence and retardation is produced. For birefringence measurement, first of all liquid crystal material has been placed between two crossed polarizer and analyzer. Here we have applied He-Ne laser source ($\lambda = 632.8\text{ nm}$, power = 5 mW) to the liquid crystal cell. Now we have rotated 45° to this LC cell. Here we have managed the temperature by hot plate (mk 2000, Instec Co. USA). For detection of transmitted light, we have used a photo detector (Instec-PD02-P) which is connected to the digital storage oscilloscope (Tektronix TDS-2024C). Then, we have plotted the graph between transmitted intensity and temperature and measure phase difference with the help of following Eq. (1a) and Eq.(1b) [30,31] at different temperature:

$$\Delta\varphi = m\pi + 2\sin^{-1}\sqrt{\frac{I - I_{\min}}{I_{\max} - I_{\min}}} \quad (1a)$$

Here Eq. (1a) is for $m = 0, 2, 4, \dots$ (For even number of peaks).

$$\Delta\varphi = (m + 1)\pi - 2\sin^{-1}\sqrt{\frac{I - I_{\min}}{I_{\max} - I_{\min}}} \quad (1b)$$

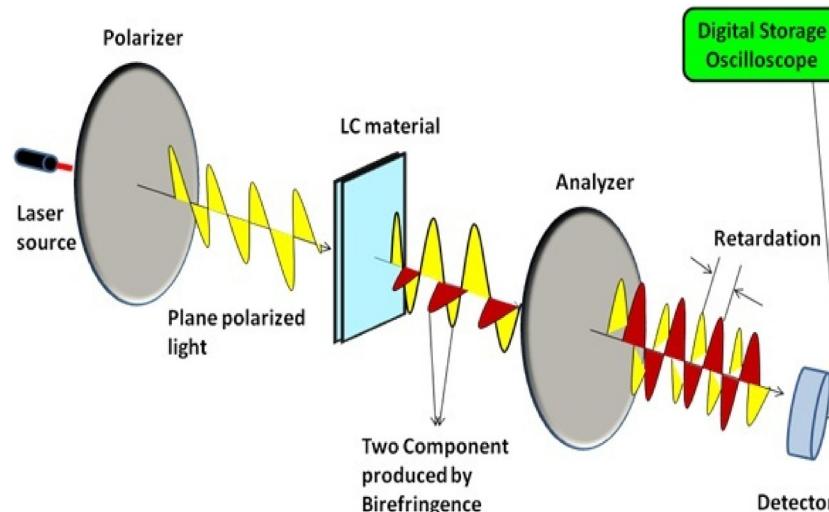


Fig. 1. Experimental set-up of birefringence measurement for pure and dye dispersed liquid crystal system.

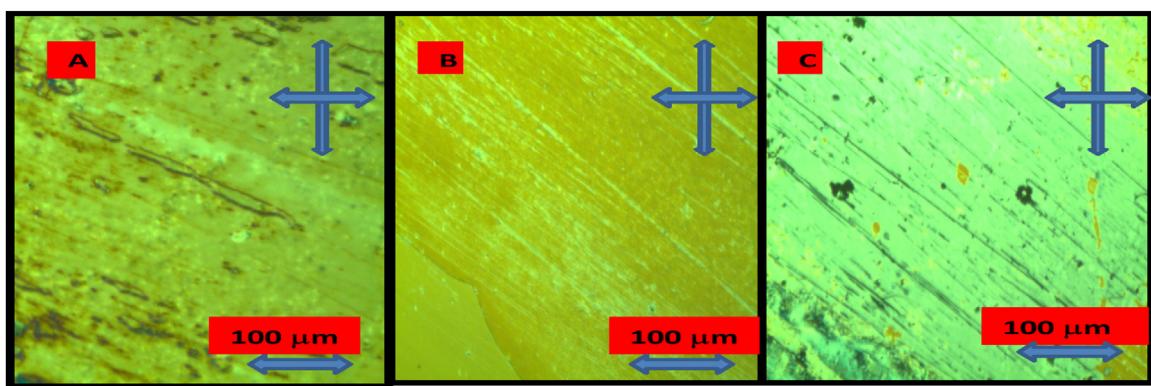


Fig. 2. Polarizing optical microscope aligned textures, (A) pure NLC 1550C, (B) pure NLC 1550C + 0.5% concentration of dye (mix 1), and (C) pure NLC 1550C + 1% concentration of dye (mix 2) at room temperature. The scale bar is the same for all samples.

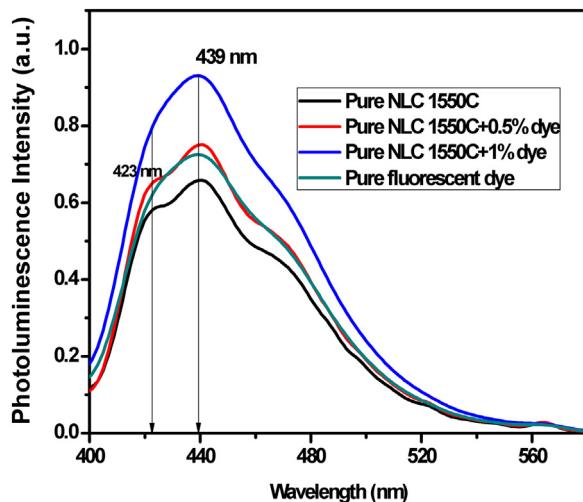


Fig. 3. Photoluminescence (PL) intensity of pure nematic liquid crystal (NLC) and NLC dispersed with fluorescent dye.

Here Eq. (1b) is for $m = 1, 3, 5, \dots$ (For odd number of peaks).

Where m represents the number of maxima peak observed in the graph of intensity and temperature, ' I ' represents the intensity at particular temperature, I_{\max} is the maximum intensity, and I_{\min} represents the minimum intensity. After calculating the phase difference for all temperature we have calculated the birefringence (Δn) using the following formula:

$$\Delta n = \frac{\lambda}{2\pi d} \cdot \Delta\varphi \quad (2)$$

Where, λ represents the wavelength while $\Delta\varphi$ represents the phase difference and d represents the thickness of a liquid crystal cell. Here we can see from Eq. (2) that the birefringence is directly proportional to the wavelength and the phase difference of the light while it is inversely proportional to the thickness of the liquid crystal cell. Therefore, high birefringence will reduce the cell gap.

3. Results and discussion

The polarizing optical microscope (POM) images for pure NLC material and dye dispersed NLC composite systems, under cross polarizer condition, are shown in Fig. 2. The POM image of pure NLC is shown in Fig. 2(A), while POM images of 0.5% dye (mix 1) and 1% dye (mix 2) are shown in Fig. 2(B) and (C), respectively. These POM images show that the homogeneous alignment has been observed for both mixtures. Also, there is no visible aggregation observed in mix 1 (Fig. 2B) but for mix 2 (Fig. 2C) small visible aggregations

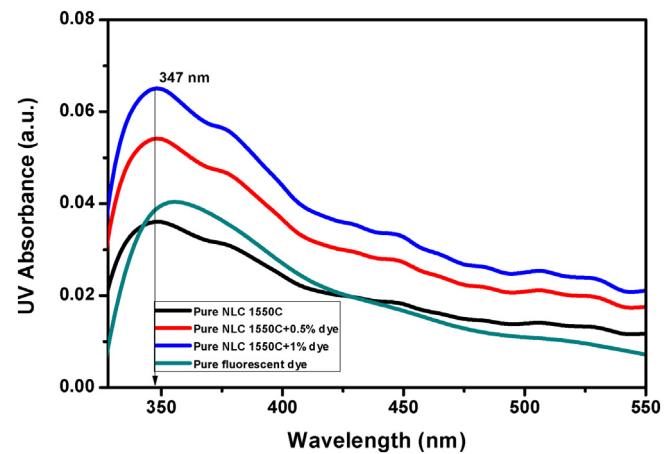


Fig. 4. Wavelength variation of UV absorbance for pure and dye doped NLC mixtures.

have been occurred. POM images also show better alignment for dispersed system as compared to pure NLC. Therefore, we can say that alignment has been improved after dispersion of dye into pure NLC. Here, dye molecules easily adjust into the NLC geometry and improve the alignment of the system.

Photoluminescence (PL) intensity spectra (in arbitrary unit) of pure NLC, pure fluorescent dye and NLC - dye dispersed composite systems have been given in Fig. 3. PL intensity spectra have been taken at room temperature and we have used a 347 nm excitation wavelength. For the study of photoluminescence, we have prepared two mixtures of NLC-dye composition along with pure NLC. When we have added little amount of dye (mix 1), then prominent change in emission peak emerges. Also, for 1.0 % wt/wt concentration of dyes (mix 2), we get higher emission peak. Photoluminescence has been enhanced for dispersed NLC - dye composite system as compare to pristine NLC for this study. Dispersion of dye increases the scattering area and since incident photons get elastically scattered to excite more NLC molecules and, therefore, PL intensity enhances. Molecules of pure NLC get excited in excited state after dispersion of dye. Constructive combination emissions from nematic and fluorescent dye molecules may also be the reason for enhancement in the PL intensity. As we can also see from Fig. 3 that the deconvoluted spectra (two peaks) are obtained for pure NLC, as well as for NLC - dye dispersed system. There are also some other liquid crystals which show the two distinct peaks which has been studied by the authors of reference [32]. In the present study two distinct peaks for mix 1 along with pure NLC have been also observed. Mix 2 does not show any prominent peak near 423 nm. First emission peak occurred at 423 nm and second emission peak occurred near

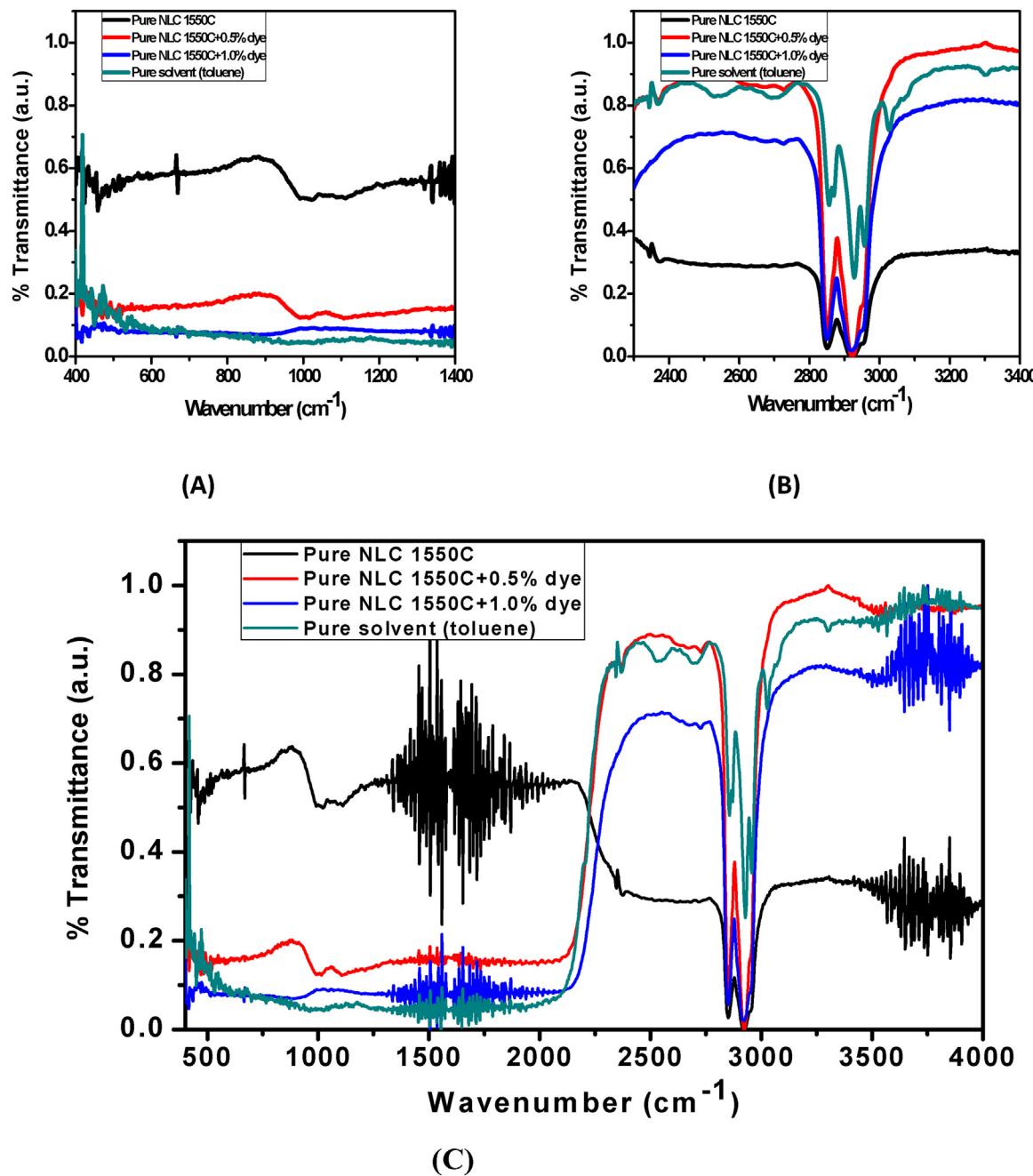


Fig. 5. FTIR spectra of the NLC 1550C and two mixtures (mix 1–0.5% concentration of fluorescent dye and mix 2- 1.0% concentration of fluorescent dye): (A) for lower wavenumber, (B) for higher wavenumber, (C) for full range.

439 nm on wavelength scale for mix 1. PL intensities of both convoluted peaks for dye dispersed system have been enhanced as compared to pure NLC. No prominent shifting in wavelength scale observed in this PL study for both mixtures but dispersion of dye only affects the intensity of photoluminescence. If we excite NLC molecules dispersed with fluorescent dye, the electronic transition from the HOMO (highest occupied molecular orbital) to the LUMO (lowest unoccupied molecular orbital) will happen for both materials. So this phenomenon also confirms the enhancement of photoluminescence. Also since the dopant material used in this study is a dye of having fluorescent in nature which is a highly scattering material and, hence, excited photons gets multiplied by this highly light scattering dopant. Also since dyes mostly emit in the visible spectrum and, hence, it couples with the emission of

pure NLC and, therefore, enhanced photoluminescence intensity observed in this case.

UV absorbance in arbitrary unit has been shown in Fig. 4 for pure NLC, pure fluorescent dye and NLC-dye dispersed system. From Fig. 4 we can see that the absorbance has been increased for fluorescent dye dispersed NLC in comparison to pure NLC. This increment in the absorbance value follows the concentration of dye. If we are increasing the concentration of dye, UV absorbance is increasing according to that. Mix 1 (pure NLC 1550C + 0.5% dye) and mix 2 (pure NLC 1550C+ 1% dye) both are showing higher value of absorbance in comparison to pure NLC. We have found absorbance peak at 347 nm wavelength for pure NLC. Any prominent shift in absorbance wavelength for dispersed system has not been observed in this study.

Fourier transformed infrared spectroscopy (FTIR) measurement has been performed in the present study at room temperature. Figure 5(A) shows FTIR study at lower wave number while Figure 5(B) shows FTIR study at higher wave number. Figure 5(C) shows the FTIR study for full range. Lower wave number peak is occurring due to the intrinsic bands which are frequently observed in thermotropic liquid crystals. We can see from the Fig. 5(A) that the normalized % Transmittance (in arbitrary unit) has been reduced for the dye dispersed system as compared to pure NLC in the lower wavenumber region. In Fig. 5(B), the band at 2355 cm^{-1} has occurred for pure NLC and mix 1. This may be due to the vibration of terminal functional group or may be due to the binding of dye molecules with the terminal carbon atom. We can see that for mix 2 (pure NLC 1550C + 1% dye) this band disappears due to a high concentration of dye. Presence of fluorescent dye also influences the vibrational bands near $2800\text{--}3000\text{ cm}^{-1}$ for pure NLC and dye dispersed system. This band is due to the CH/CH₂/CH₃ stretching vibrations. Some small peaks also occur near at 3400 cm^{-1} which may be due to the moisture (-OH). Therefore, appearance of all these peaks suggests that the dye highly affects the NLC geometry.

Liquid crystal used in this investigation is NLC, which is a birefringent material. Therefore, we have also performed the birefringence measurement for pure NLC and dye dispersed system. Figure 6(A) shows the variation of birefringence (Δn) with change in temperature for pure NLC and dye dispersed composite system. This figure shows that the birefringence has been enhanced for dispersed system. When laser light falls onto the NLC material, then phase difference has been produced from which we have calculated the birefringence. Figure 6(B) shows the variation in output intensity with respect to temperature for pure and dye composite system. When we are increasing the temperature, intensity varies from minima to maxima because molecular orientation changes. Therefore, we are observing change in phase difference. Dispersion of dye into pure NLC improves the orientational order for mix 1 (pure NLC 1550C + 0.5% dye), as well as for mix 2 (pure NLC 1550C + 1% dye). Therefore, both the composites of NLC dispersed with dyes show improvement of orientation order and, hence, the birefringence as we can see in Fig. 6(A). This improved orientation order can also be seen in the POM textures and in the model given in Fig. 6(C). Figure 6(C) shows that the orientational order improved when the disc shape dye dispersed into the NLC geometry. The dopant used in this study is a disc shape fluorescent dye. The effect of disc shape dye molecule has also been reported by some researchers in recent years [33,34]. When dye molecules dispersed into the matrix of pure NLC, it affects the molecular orientation. On the other hand, angular momentum conservation concept of interaction of nematic with dye excites dye molecules after interaction with light [35,36]. This may also be the reason for increment in the value of birefringence. Figure 6(A) also shows that near isotropic point both the composites showing almost similar values of birefringence as compared to pure NLC material. Since intermolecular forces between the components of the NLC 1550C (alkylbicyclohexyl carbonates and cyanobicyclohexane) and dye molecules possess weak van der waal character only and which is broken out easily at higher temperature point (near isotropic), therefore towards the isotropic point birefringence does not change much more and also falls to zero. In the inset of Fig. 6(A) concentration of dye along with birefringence has been plotted for 30°C temperature. For pure NLC, birefringence comes to 0.060 and for mix 1 and mix 2, birefringence values are found to be of 0.062 and 0.065. Many photonic and opto-electronic devices require materials having high birefringence value. Therefore, this study about the enhanced birefringence may be very useful in photonic and opto-electronic industries.

Variation of relative electric permittivity with change in temperature has been shown in Fig. 7. This graph has been plotted at 1

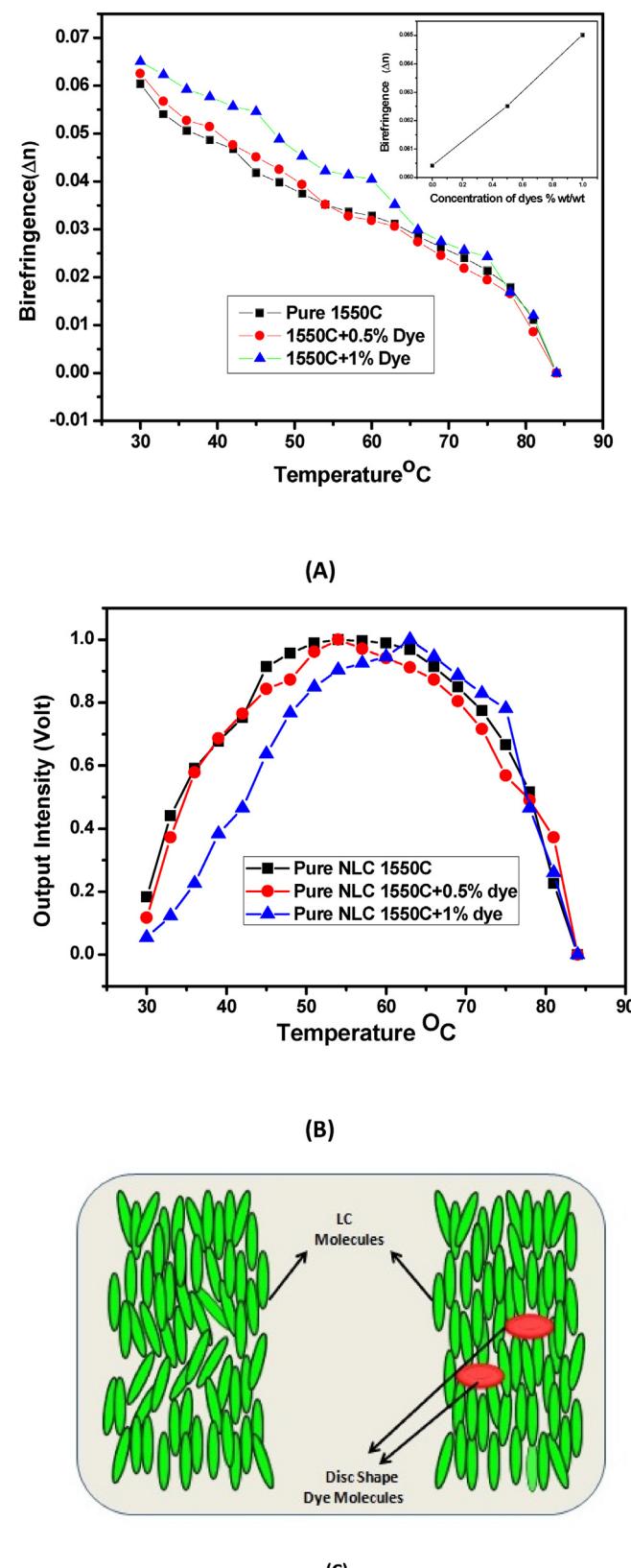


Fig. 6. (A) Variation in birefringence (Δn) for pure NLC 1550C and dye dispersed NLC system with change in temperature (inset of this Fig. shows variation of birefringence with respect to concentration of dye at 30°C temperature); (B) variation of normalized output intensity with respect to temperature for pure NLC 1550C and dye dispersed system; (C) change of orientation of nematic liquid crystal molecule with the addition of dye in a pure liquid crystalline system.

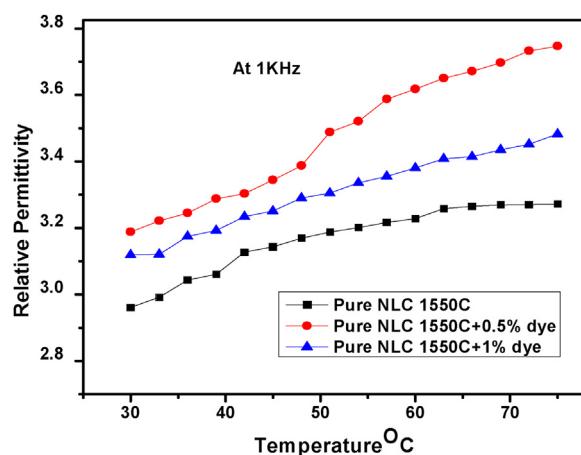
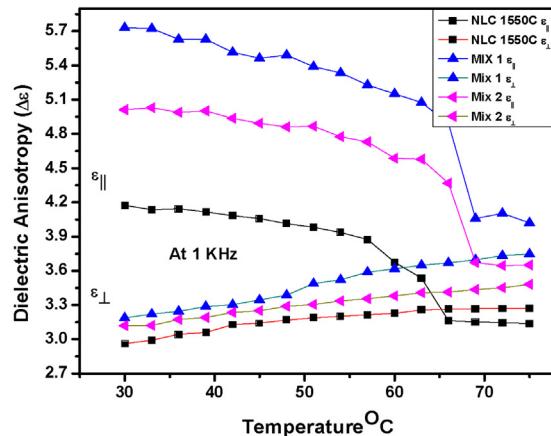


Fig. 7. Variation of relative permittivity with respect to temperature for pure NLC 1550C and dispersed system at 1 KHz frequency.

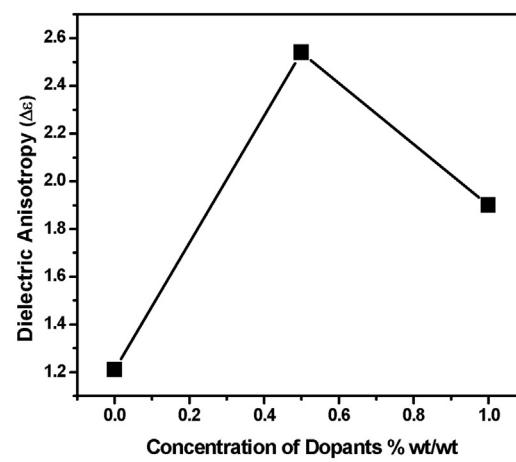
KHz frequency. In the present study, relative electric permittivity increases for mix 1 (pure NLC 1550C + 0.5% dye) while it decreases for mix 2 (pure NLC 1550C + 1% dye) but still higher than that of pure one. When we have dispersed the dye into pure NLC, three interactions take place:

- 1) Interaction between NLC – NLC molecules
- 2) Interaction between Dye – Dye molecules
- 3) Interaction between NLC – Dye molecules.

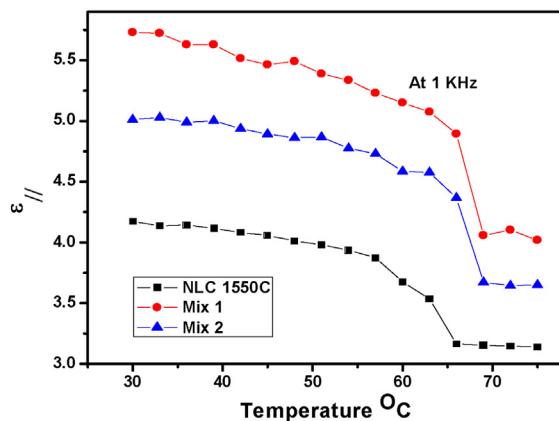
The interactions between NLC and dye molecules are dipolar, electrostatic and van der Waals in which dipolar interaction is dominating. Here permittivity is increasing because interaction of NLC molecules with dye molecules supports the dipole moment. Dipole moment of the dye molecule is in the same direction as the dipole moment of NLC molecule. Also, the enhancement in the value of permittivity occurs because charge storage capacity of dye dispersed NLC system increases. Since we know that, relative permittivity, $\epsilon_r = C/C_0$, where C is the capacitance of the cell filled



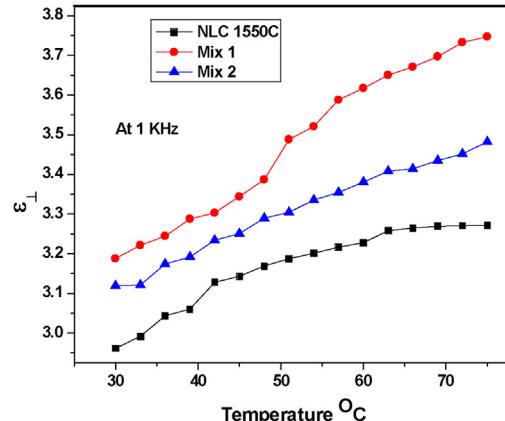
(A)



(B)



(C)



(D)

Fig. 8. (A) Shows the variation of dielectric anisotropy with respect to temperature for pure and dye dispersed system at 1 KHz frequency' (B) ariation of dielectric anisotropy with respect to concentration of dye at 1 KHz frequency; (C) parallel component of dielectric permittivity ($\epsilon_{||}$) of pure and dye dispersed sample as a function of temperature at 1 KHz frequency; (D) perpendicular component of dielectric permittivity (ϵ_{\perp}) of pure and dye doped sample as a function of temperature at 1 KHz frequency.

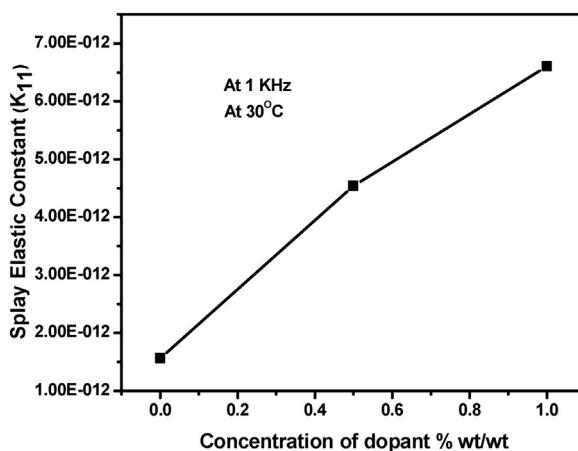


Fig. 9. Variation of splay elastic constant (K_{11}) with respect to different concentration of dye in pure NLC at 1 kHz frequency and 30°C temperature.

with liquid crystalline material and C_0 is the capacitance of the air. Therefore, the permittivity is enhancing due to enhancement in the capacity. On the other hand, when concentration of dye increases, the number of domains within the system decreases. Therefore, the distance between the domains increases and the dipole-dipole interactions are reduced. Therefore, relative electric permittivity decreases for mix 2 as compared to mix 1, but still it is higher than that of pure NLC.

To use the NLCs in the LCD devices, we have to know about the dielectric anisotropy ($\Delta\epsilon$) of the system. Therefore, we have also measured the dielectric anisotropy here. Fig. 8(A) gives the change in $\Delta\epsilon$ with respect to temperature. Dielectric anisotropy increases after the dispersion of dye into pure NLC. Figure 8(B) shows the change in $\Delta\epsilon$ with change in concentration of dopant at 30°C temperature. The value of $\Delta\epsilon$ for pure NLC is of 1.21 whereas for mix 1 and mix 2, it is of 2.54 and 1.90. NLC 1550C is a positive anisotropic material (for positive dielectric anisotropic LC, $\epsilon_{||} > \epsilon$). When dye dispersed into the NLC dipolar contribution increases, hence $\Delta\epsilon$ also increases. Figures 8(C) and (D) show the parallel component and perpendicular component of permittivity for pure NLC and dye-NLC system. As we can see from these figures that after doping of dye, the parallel and perpendicular component of permittivity both increase but increment fraction in the parallel component is high as compared to the perpendicular component. Better alignment of NLC shows the lower value of ϵ_{\perp} ($\epsilon_{||} > \epsilon$). Dielectric anisotropy is the difference of $\epsilon_{||}$ and ϵ . Therefore, by the combined effect of parallel and perpendicular components dielectric anisotropy has been enhanced. Also, alignment has been improved for composite system which can also be confirmed by the POM images in Fig. 2. These results can also be understood by interactions of dye molecules and NLC molecules. As we can also see in the c) that irregularities of pure NLC molecules reduce after dispersion of dye and due to which orientational order increases. Presence of disc shape dyes in pure matrix support or enhance the pure LC molecular orientation around the dye molecule. Therefore, we are getting increased dielectric anisotropy for dispersed system. For mix 2, small decrement in $\epsilon_{||}$, as well as in ϵ_{\perp} has been observed, therefore for mix 2 $\Delta\epsilon$ decreases than mix 1.

Splay elastic constant (K_{11}) has also been measured in this study for pure NLC and dye dispersed system using dielectric anisotropy and threshold voltage values. Threshold voltage has been explained in the later part of this article. Figure 9 shows the change in K_{11} with change in concentration of dye. We can see from the given figure that the splay elastic constant has been increased for dye dispersed

Table 1

Change in threshold voltage with change in concentration of dye of pure NLC and dye dispersed system at 30°C temperature.

Pure 1550C	1.20 V
1550C + 0.5% fluorescent dye wt/wt (mix 1)	1.44 V
1550C + 1% fluorescent dye wt/wt (mix 2)	2.10 V

system. Formula between the K_{11} , threshold voltage (V_{th}) and $\Delta\epsilon$ is given below:

$$K_{11} = (V_{th})^2 \epsilon_0 \Delta\epsilon / \pi^2 \quad (3)$$

Here ϵ_0 represents the permittivity of free space. Here we can see from Eq. (3) that K_{11} depends upon the $\Delta\epsilon$. Since in this investigation, $\Delta\epsilon$ is increased for dye dispersed system. therefore increased $\Delta\epsilon$ gives rise to increase in the value of K_{11} . Here when we are dispersing the dye into the nematic liquid crystal, then two interactions take place which are given as follows:

- 1 Interaction between dye and NLC molecules
- 2 Interaction between dye-dye molecules.

The combined effect of these two interactions is responsible for increment in the K_{11} . Also since K_{11} directly depends upon the threshold voltage and since threshold voltage is increasing in our present study, as we can see in Table 1, so this also confirms the increasing nature of splay elastic constant.

For the measurement of driving voltage of liquid crystal material, we have also measured the threshold voltage (V_{th}). Table 1 shows the change in V_{th} with change in concentration of dye at 30°C temperature. Here V_{th} increases as we increase the concentration of dye. It has been found that the value of threshold voltage continuously increases on increasing the concentration of dye in comparison to pure NLC 1550C. V_{th} for pure NLC has been found to be of 1.20 V. But when we dispersed the fluorescent dye into the pure NLC, the values of V_{th} have been increased. For 0.5% of dye (mix 1) the value of threshold voltage has been found to be of 1.44 V whereas for 1.0% of dye (mix 2) V_{th} comes to 2.10 V, respectively. Clearly we can observe that the values of V_{th} for dispersed system are much higher as compared to pure NLC. Some ions get adsorbed at the layer of alignment due to increasing ionic concentration. Due to this, effect of applied field gets diminished and, therefore V_{th} increases.

4. Conclusions

In this investigation, we have analysed some very useful optical, electro-optical and dielectric properties. Enhanced photoluminescence has been observed after dispersion of fluorescent dye into the NLC 1550C. We have also measured the UV-visible absorbance and found that it has been increased for dispersed system. We have also performed the FTIR study and we have observed IR band at 2355 cm⁻¹ and 2800–3000 cm⁻¹. These IR bands show that dye molecules alter the terminal functional group and CH/CH₂/CH₃ stretching vibrations of LC matrix. Increment in the birefringence is also a one of the main finding of present work. Relative permittivity increases after the dispersion of dye. Dielectric anisotropy has also been increased for the dye dispersed system. In this study, both; photoluminescence and birefringence parameters have been enhanced, therefore this NLC-dye composite system is very important for display point of view. The results of current investigation may also be very useful in liquid crystal displays (LCDs).

Conflict of Interest

Authors have no conflict of interest.

Acknowledgements

The authors are thankful to Department of Science and Technology for the grant of Indo-Polish Project and UPCST for grant of project. Authors are also thankful to APJ ABDUL KALAM CENTRE FOR INNOVATION for experimental facilities.

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