Nematic liquid crystal/dimethylsulphoxide mixture based tuning condenser

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Abstract
Here, we demonstrate experimentally the existence of temporally variable dielectric behavior of a nematic liquid crystal (NLC), the most widely used liquid crystal (LC) phase among all LCs materials, by mixing dimethylsulphoxide (DMSO) into former. The intermolecular interactions and nanosegregation of the molecular LC structures have influenced their self-assembly by mixing DMSO into it. We mainly examined the dielectric parameters such as dielectric permittivity, dielectric loss factor, and absorption/energy dissipation and observed that the NLC/DMSO mixture shows a nearly dielectric-conductor (D-C) transition as time passes. The presence of DMSO in this analysis was confirmed by Fourier transform infrared spectroscopy while time-dependent dielectric studies were carried out using dielectric spectroscopic techniques. The promising idea of showing D-C transition is truly proven that may open the possibilities for real-time variable and supercapacitors.

Introduction
In a world controlled by symmetry and invariance postulates, liquid crystal (LC) materials, since their discovery, have been proved as a backbone in the modern digital world [1–3]. The LCs, nowadays, are the tempting materials used in the electronic-industry as the electro-optical component in different devices. Despite their enormous success in display devices, there are several applications of LC in another arena like biology (as the combination of order and mobility is a basic requirement for structure formation in a living system) biomedicines, analytical chemistry and nanophysics [4–7]. The impact of LC on modern technology such as a biosensor, memory devices, LC-based lasers, holography has been profound [8, 9]. Besides these modern familiar applications, there are other devices with improved properties of LC by doping nanoparticles or mixing with other inorganic/organic solvents as LCs are favorable material for the effective transfer of ions and electrons [6, 10–13].

On the other side, dimethylsulphoxide (DMSO), which is an organosulfur compound and produced as a by-product of wood pulping, has been applied as an organic solvent in chemical transformations and is utilized widely in industrial processes as well [14]. It is a polar aprotic solvent and effectively miscible in organic as well as inorganic compounds. Albeit, several observations have been done in which DMSO was used as a solvent to improve the solubility of nanoparticles in LCs but more recently, there has been an explosion of interest as in the present studies DMSO alone could change the dielectric properties at the macroscopic level.

The macroscopic properties such as dielectric permittivity, loss factor, absorption etc evolving a powerful tool to study the performance of LC-based advanced devices. Various dielectric observations of LC in wide frequency range have been investigated as such regions provide the dynamics of dipolar motion. Enormously, dipole moment provides information about the molecular structure of LC materials. Henceforth, various experiments have been performed for dielectric measurements of molecules having a strong dipole moment along with some weak dipole moment. Additionally, the organic molecules have very high polarizability which helps to improve the dielectric properties of LC when combined with them.
Recently, nematic liquid crystal (NLC) has received much attention and deep interest due to its electro-optical properties in different applications. The NLC is the most commonly used LCs of LC family due to its long-range orientational order and short-range positional order of molecules and can be easily aligned by applying low electric and magnetic fields. The hydrodynamics of NLC has been theoretically explained in a simplified manner \[15\]. The interactions between different integrants with LCs are common for various intriguing phenomena of a kind that faster electro-optic response, memory effect and different transition from one phase to another.

Getting motivated by these experimental results, the notion would be interesting to find the temporally variable dielectric behavior of LC in addition with DMSO. So firstly, in this article, we present our vision for the effective changes in dielectric parameters of NLC by mixing DMSO into it. The concentration of DMSO into NLC was optimized by observing the behavior of dielectric parameters with frequency at different concentrations of the former. After optimizing the appropriate concentration of DMSO, the temporal variation of dielectric behavior of the mixture has been investigated for several months continuously to elucidate the inter-surface interaction between the molecules that is the ultimate finding of this reported research work.

**Experimental**

For the experiments, we used homogeneously aligned LC sample cells (Instec, Inc., USA), coated with indium tin oxide film as an electrode, having an effective area \(10 \times 10\) mm\(^2\) and thickness 5 μm. In the present investigations, room temperature NLC material namely (5 CB, 98%) with chemical formula \(C_{18}H_{19}N\) and integrant (DMSO, 99%) with chemical formula \((CH_3)_2SO\) were used. Both were procured from Sigma-Aldrich, USA and used without further purification. The phase sequence of the NLC material used (as given by the manufacturer) is as follows:

\[
\text{cryst} \quad 23^\circ C \leftrightarrow \text{nematic} \quad 35^\circ C \leftrightarrow \text{iso}
\]

Where cryst represents crystalline and iso represents the isotropic phase.

The chemical structures of 5 CB and DMSO have been shown in figure 1:

The dielectric measurements were carried out with an LCR meter (E4980A, Keysight, USA) and the Fourier transform infrared (FTIR) spectra of pure NLC and doped NLC/DMSO mixture were recorded using FTIR spectrometer (Tensor 37, Bruker, Germany). Mixtures were prepared by weighing, mixing and stirring (through ultrasonication) the appropriate amount of the components at room temperature. The compositions were 1 part DMSO and 4 part 5CB (w/w), 1 part DMSO and 2 part 5CB (w/w), 1 part DMSO and 1 part 5CB (w/w), 2 part DMSO and 1 part 5CB (w/w) etc and called them as 1:4, 1:2, 1:1, 2:1 etc respectively. A small amount of mixtures were placed at the openings of the sample cells of thickness 5 μm.

**Results and discussion**

First, we ensured the presence of DMSO in NLC which was carried out through FTIR spectrometer in the wavenumber range \(500–4000\) cm\(^{-1}\). The observed IR spectra illustrate various functional groups present in these systems. As reflected from figure 2(b) that the strong absorption near 2227 cm\(^{-1}\) is distinguished due to the
C≡N group in 5CB ensures the characteristic of 5CB in the mixture. Similarly, the existence of DMSO is detected through the absorption peak of S=O near 1024 cm⁻¹ in IR spectra. It can also be noticed in figure 2(b) that in 5CB/DMSO mixture, C–H bonds in the alkyl chain of 5CB and symmetric and asymmetric C–H stretching of DMSO are significantly remarked in the range (2800–3100 cm⁻¹).

In order to bring out the strong impact of the guest DMSO molecules in 5 CB, we have observed the dielectric properties of pristine NLC (5CB) such as dielectric permittivity (ε'), dielectric loss (ε″) and dielectric loss factor (tan δ). The complex dielectric permittivity is given by the formula:

\[ \varepsilon^* = \varepsilon' - i\varepsilon'' \]  

(1)

Where \( \varepsilon' = \varepsilon / \varepsilon_0 \) is the real part of dielectric permittivity and \( \varepsilon'' \) gives the absorption or dielectric loss.

Figure 3 shows the variation of these properties with frequencies in the range of 20 Hz to 1 MHz at different concentrations. As reflected in figure 3(a) the value of \( \varepsilon' \) for pristine 5CB is \(~107\) @ 20 Hz. As we started increasing the concentration of DMSO, the permittivity increased but at a particular concentration (1/1 wt%) of DMSO in 5 CB, its value became maximum (i.e. 148). There was no further improvement by increasing the
concentration of DMSO. The dielectric loss is given by the imaginary part of the complex permittivity which has been shown in Figure 3(b). It is clear from the graph that we observed two peaks of absorption one was due to the presence of 5 CB and the other was of DMSO. But when the concentration of DMSO was large enough, one peak has suppressed. The dielectric loss factor, given by equation (2) is shown in Figure 3(c)

$$\tan \delta = \varepsilon'' / \varepsilon'$$

Figure 3(c) shows the dielectric loss factor which is basically the ratio of energy dissipation and energy stored in dielectric materials. It describes the parameters relating to the internal friction for movement of coupling chargers (dipoles), free charges (mainly ions) in dielectric systems and also power loss (such as heat) inside the material.

Further, we have extended this ideology to observe the temporal dielectric behavior of pristine NLC (5 CB) and DMSO / NLC mixture. For this, we have taken the cell of optimized concentration (1/1 wt%) and observed the same for around 4 months continuously. It has been observed that the mixing of DMSO in 5CB showed some special behavior with time. During the temporal observation, it was noticed that firstly the dielectric permittivity of DMSO /NLC mixture started to increase and then it began to decrease towards very low value (nearly zero).

The temporally variable dielectric behavior of pristine NLC (5 CB) and DMSO/NLC mixture at room temperature has been shown in Figure 4. It is clear from figure 4, that in case of pure 5 CB there is a slight change observed in dielectric permittivity due to the injection of impurities from glue, polyimide layers etc [16]. When DMSO was incorporated in 5CB, the increase in dielectric permittivity over time is attributed due to the increased number of ionic impurities. The dielectric characteristics are primarily dictated by space charge polarization in low-frequency region and ionic density given by the following equation [17]

$$\rho(z, t) = \rho_0(z) + \rho_1(z)e^{\omega t}$$

Where \(\rho_0\) represents the steady state density of ions and \(\rho_1(z)\) represents the amplitude of the oscillation of the ion density at the frequency equal to the frequency of the external applied electric field which is further given by following equation

$$\rho_1(z) = \frac{\mu E}{\kappa D}\rho_0\left(\frac{e^{\kappa z}}{1 + e^{\kappa d}} - \frac{e^{-\kappa z}}{1 + e^{-\kappa d}}\right)$$

Where \(\kappa = \sqrt{\frac{\mu E}{\kappa D}}\) and \(\mu / D = q / K_B T\), where \(K_B\) is Boltzmann Constant.

Further it is clear from equation (4) that \(\rho_1(z)\) is directly proportional to the electric field \(E\). And due to these injected ions, space charge polarization is the key factor to the effective polarization in lowfrequency region. Additionally more assumptions have been reported by Sawada et al and the dielectric permittivity is concluded by equation (5).

$$\varepsilon'(\omega) = -\left[\frac{4\pi nq^2D}{\omega K_B TA}\right]\left[\frac{1 + 2e^{\Delta} \sin(A) - e^{2\Delta}}{1 + 2e^{\Delta} \cos(A) + e^{2\Delta}}\right]$$

Where \(A\) is the effective electrode area of the sample cell. This equation clearly explains the dependency of dielectric constant on various parameters through space charge polarization.
When DMSO has incorporated in the alignment-layer sample cell filled with 5CB, the interaction of polyimide layer with the former began to start. More impurity ions are produced due to annihilation of polyimide layer which serves as a protective screen between the material used (pure and doped) and ITO. Now more ions from the ITO layer moving towards the region of the doped sample and the concentration of ions began to collect in the region. With these impurity ions, firstly, the increase in dielectric permittivity over time is attributed according to equation (5). It has been increased continuously until a fixed value (~400) for around 3 months, but when the concentration of the ions was large enough, it stopped following the mentioned equation (5) and began to show some strange behaviour. This is reasonable that the higher impurity ions in doped sample cell allow the creation of a platform for the hopping mechanism. The agglomeration of ions takes place that helps the charge carriers (conducting ions) to reach from one plate to another through hopping mechanisms. Now the transport of these conducting ions is possible via this platform results in the enhancement of conductivity and the dielectric permittivity gradually decreases to a minimum value. This variation in dielectric permittivity over the time illustrates the property of tunable condenser. To check this effect properly, the experiment was also performed for other NLC namely ZLI-1565 and the same results have been observed. These observed effects are key elements in a point to point display or non-display systems and can be used for designing a variety of variable capacitors. These findings offer a combination of advantages in terms of both performance and manufacturing.

**Conclusion**

We systematically investigated the effect of DMSO on the dielectric parameters of 5 CB in the wide frequency range. It has been observed that the dielectric permittivity and loss factor increases with increasing the concentration of DMSO in 5 CB by w/w %. The improvement could observe at a particular concentration of DMSO and when we started further increment in DMSO, the dielectric characteristics remained constant. We have also observed the temporal effect of 5CB/DMSO mixture which illustrates the property of tunable condenser and method developed in the study works as a powerful tool to fabricate various devices in which evaluation of ions contained in LC materials due to controlled molecular interactions.

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**Data availability statement**

‘The data that support the findings of this study are included within the article’.

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