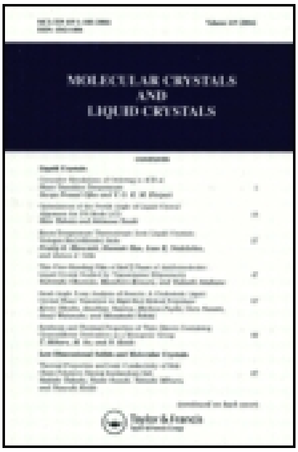

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Dielectric Spectroscopy of Wide Temperature Range Nematic Liquid Crystal Mixtures

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The dielectric studies in two nematic liquid crystal mixtures (E-201, E-47) in the frequency range of 50 Hz–1 MHz and theoretically calculated up to the extended frequency range have been investigated. The real and imaginary parts of dielectric constant at low frequencies exhibit a significant contribution of conductivity. The corrected dielectric parameters show Debye-type dispersion with symmetric distribution of relaxation times. Our results indicate that dielectric increment, empirical-factor and static permittivity vary with temperature. A well known Fuoss – Kirkwood method was used to analyze the dielectric parameters.

Keywords Dielectric permittivity dielectric loss; dielectric relaxation; distribution parameter; nematic liquid crystal

1. Introduction

Liquid crystal (LC) materials have attracted much attention over a number of years because of their unique electro-optic and magneto-optic properties for novel display applications [1–5]. Dielectric spectroscopy provides one of the few techniques for probing the nature of the molecular reorientation and there have been several such studies on nematic liquid crystals. The phenomena associated with molecular orientation and relaxation processes have been discussed by several research groups [6–11]. The E-47 differs from E-201 having cyno-biphenyl and PCH positive additive. The E-47 is identical to E-7, which is a mixture of various compound like cyanobiphenyl (CB), Oxycyanobiphenyl (OCB) and Cyanoterphenyl (CT) derivatives in the following proportion by weight: 5CB (51%), 7CB (21%), 8OCB (16%) and 5CT

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(12%). The preference of choosing E-47 and E-201 is due to the broad nematic range −20 to 61°C and −11 to 88°C, respectively.

The present study of the dielectric behavior in the nematic phase of E-47 compared to the E-201 is expected to provide information about the influence of the components, which will help to understand the dielectric relaxation mechanism and related parameters in liquid crystals [12].

2. Theoretical Background

The director reorientation can be described in terms of real (ε') and imaginary part (ε'') of the complex dielectric permittivity (ε^*) which is given as

$$\varepsilon^* = \varepsilon' - j\varepsilon'' \tag{1}$$

Maier and Meier [7] studied the dielectric relaxation in liquid crystals, both experimentally and theoretically. Generally long-chain molecules show a broader dispersion curve and lower maximum loss that would be expected from the Debye relationship. In such case Cole-Cole suggested that permittivity might follow the empirical equation [13]:

$$\varepsilon^* - n^2 = \frac{\varepsilon_0 - n^2}{1 + (j\omega\tau)^{(1-\alpha)}} \tag{2}$$

Where, α ($0 \leq \alpha \leq 1$) is constant. In some cases the Fuoss and Kirkwood also developed an empirical distribution function in terms of Kirkwood theory of permittivity [15]. Experimental data were analyze using the Fuoss-Kirkwood relation for practical evaluation is

$$\cos\, h^{-1}\left(\frac{\varepsilon''_{\max}}{\varepsilon''}\right) = 2.303\,\beta(\log f_{\max} - \log f) \tag{3}$$

where, numerical factor β is the Fuoss – Kirkwood parameter defines the width of the absorption. The $\beta = 1.00$ corresponds to a simple Debye process, i.e., the dipolar orientation relaxes with a simple rate coefficient.

3. Experimental

The frequency dependence of the complex dielectric permittivity has been studied at different temperatures in two nematic liquid crystal mixtures (E-201 and E-47). Planar aligned cells each of thickness 10 μm made of indium tin oxide (ITO) coated glass substrates have been used for dielectric measurements. These ITO coated glass substrates were spin coat (~1000 rpm) with a solution of nylon 6/6 and m-cresol and then uni-directionally rubbed to obtain the perfect planar alignment. The nematic liquid crystals were filled in empty sample cell by capillary action at the isotropic temperature of NLCs and then placed into hot stage (Model THMS600) attached with temperature controller (Model Linkam TP94). Alignment was confirmed under cross polarizer at 10x magnification with the help of polarizing microscope (model Olympus BX-51P) interfaced with charge couple device camera. The dielectric measurements were carried out using a programmable and automatic RCL meter (FLUKE PM 6306) in the frequency range from 50 Hz to 1 MHz. The cell was calibrated using air and benzene as a standard references [15,16].

4. Results and Discussion

In the present samples, measured dielectric data below 1 kHz are affected due to ionic conductance and electrode polarization effect, whereas data above 100 kHz are affected due to the combined effect of lead inductances and electrode surface resistance [17]. In order to remove low and high frequency effects, the dielectric spectra were fitted with the generalized Cole-Cole equation using MATLAB software:

$$\varepsilon^* = \varepsilon' - j\varepsilon'' = \varepsilon'(\infty) + \sum_i \frac{(\delta\varepsilon)}{1+(j\omega\tau)^{(1-\alpha)}} + \frac{A_1}{\omega^n} - \frac{j\sigma_{(50Hz)}}{\varepsilon_0\omega} - jA\omega^m \tag{4}$$

Where, $\varepsilon'(\infty)$ is the relative permittivity in the high-frequency limit, $\delta\varepsilon$, τ and α are the dielectric strength, relaxation time and distribution parameter of i^{th} mode, respectively. The third and fourth terms in Equation (4) represent the contribution of the electrode capacitance and ionic conductance at low frequencies. The imaginary term $A\omega^m$ in Equation (4) takes into account partially the effect of ITO coating. A_1, n, A and m are constants. σ is the ionic conductance and ε_0 is the free space permittivity. In order to carry out the dielectric analysis, plots of $\varepsilon'_\perp$ and $\varepsilon''_\perp$ versus frequency for the mesogens are shown in Figures 1 and 2 at 30°C. It is evident that

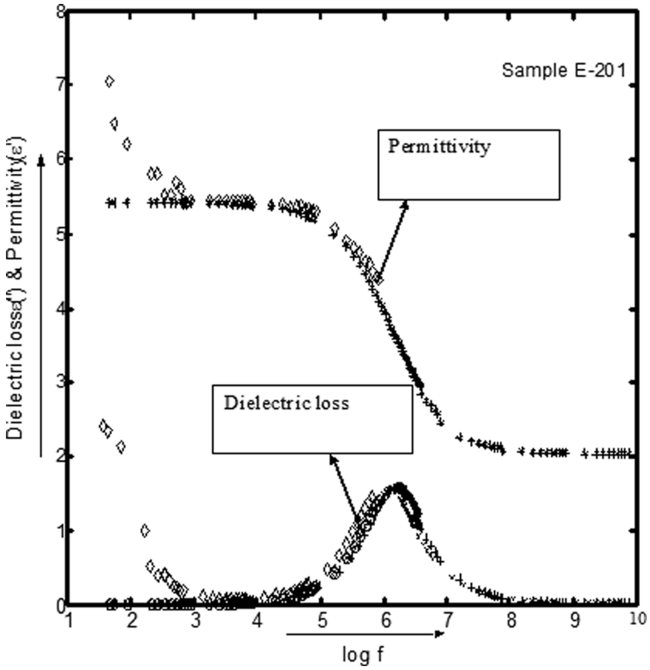

Figure 1. Frequency dependence of measured permittivity (ε') –◇ and loss (ε'') – ◇ for E-201 at 30°C. After subtracting correction terms theoretically calculated values of ε'–* and ε''–* are also shown.

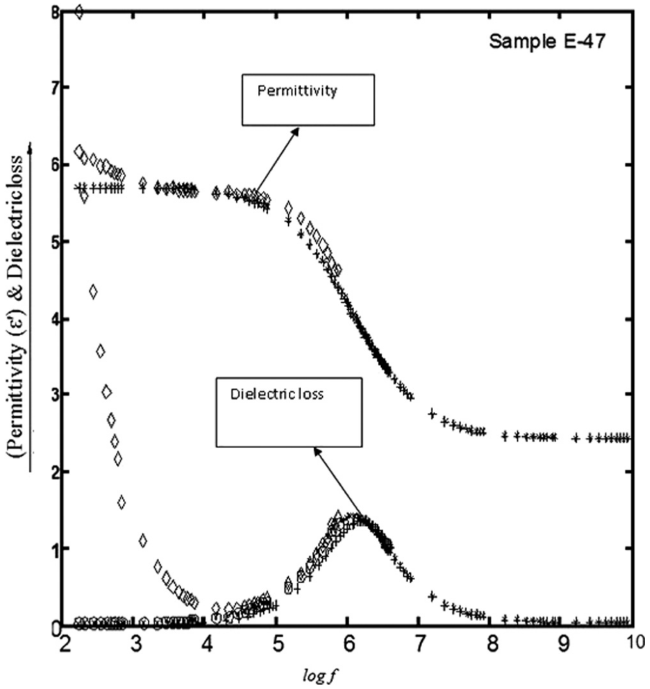

Figure 2. Frequency dependence of measured permittivity (ε′) – ◇ and loss (ε″) – ◇ for E-47 at 30°C. Theoretically calculated values of ε′– * and ε″– * are also shown.

there is a heavy dominance of ionic conductance in the low frequency region. Using Equation (4), the low frequency conductance was subtracted and the corrected data was analyzed theoretically up to the extended frequency. The corrected experimental data points and the theoretically drawn curve for $\varepsilon'_\perp$ and $\varepsilon''_\perp$ are shown in Figures 1 and 2. The values of static dielectric permittivity for both mixtures are of the same order as observed for other nematic mixtures exhibiting positive dielectric anisotropy [18–22]. Our results show that in higher region no significant change in permittivity was observed.

For the demonstration of dielectric properties, Fuoss-Kirkwood method has been used to find the relaxation times and empirical factor [22]. A typical plots between $\cosh^{-1}$ ($\varepsilon''_{\max}/\varepsilon''$) and $\log_{10}$f without bias field at 30°C is shown in Figure 3.

The calculated value of β are found 0.83 and 0.82 for E-201 and E- 47, respectively at 30°C. These values are of the same order as observed for other nematic mixtures [14, 23–27].

The interpretation of perpendicular alignment dispersion in these mixtures is complicated by the possibility of internal rotation of the terminal components. Here two alternatives are possible; firstly when the molecule behaves as a rigid entity and the secondly when the terminal constituent rotation makes a significant contribution. Moreover the values of β for both the nematogens with different constituents are low and of the same order, which suggest that, the dielectric relaxation in these mixtures is a co-operative phenomenon involving the components. At the same time, without bias field, the lower value of β indicates a far less well-defined molecular order, giving a much wider range of individual dipole reorientation rates.

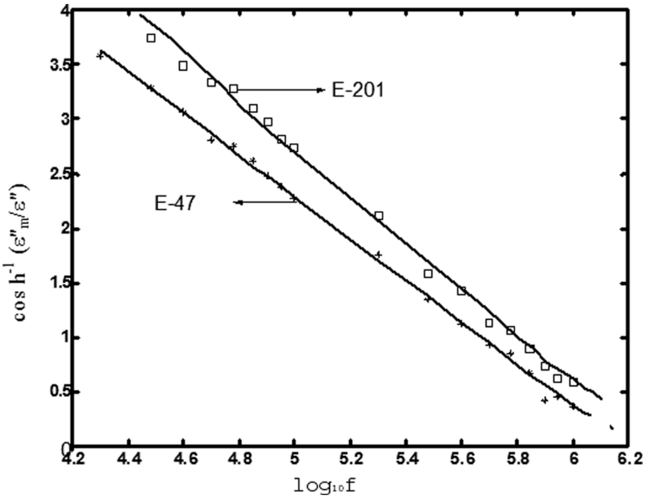

Figure 3. Plot of $\cosh^{-1}(\varepsilon''{}_m/\varepsilon'')$ versus $\log_{10}$f for relaxation frequency of both liquid crystals, (*) and (□) represents experimental data and solid line represent theoretical plot.

The large positive anisotropy of these mesogens suggests that there are large dipole moments along the long molecular [28]. Results on the alkyl-cyno-biphenyl series of liquid crystals indicates that there is significant anti-parallel ordering of the molecular dipoles to which dipole-dipole interactions must contribute. Madhusudana and Chandrasekhar [29,30] suggested that inclusion of short range dipole-dipole interaction raised the order parameter and predicted a small experimental change in the mean permittivity at the nematic to isotropic transitions. For a quantitative analysis the results presented in Figure 4 shows the temperature dependence on dielectric increment $(\delta\varepsilon)^{-1}$.

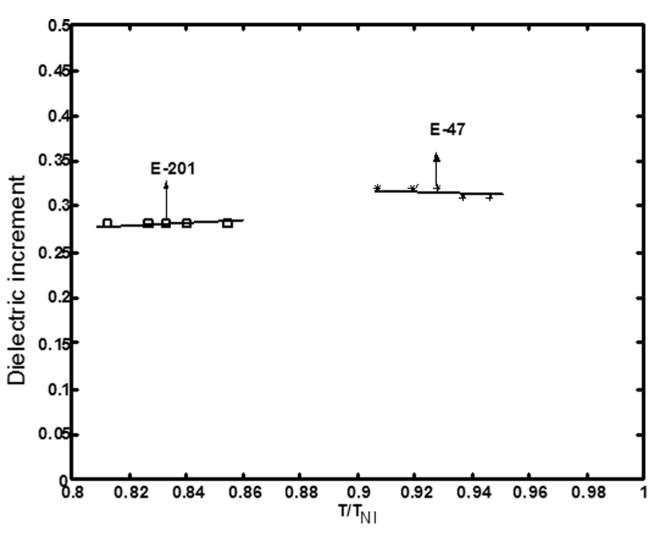

Figure 4. Dielectric increment $(\delta\varepsilon)^{-1}$ as a function of temperature.

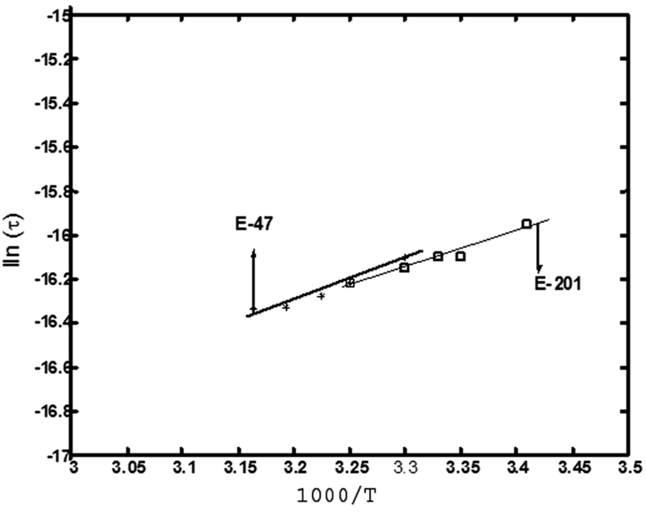

Figure 5. Arrhenius plot for both nematic mixtures studied.

The activation energy was evaluated from the plot of lnτ versus $10^3/T$. In the nematic liquid crystals potential barrier due to the molecular forces hindering the reorientation of the molecules in the perpendicular direction and activation energy due to viscosity effects. Arrhenius plot as shown in Figure 5 yields the total activation energy and samples shows single activation energy.

5. Conclusion

The dielectric studies have been carried out in the nematic liquid crystal mixtures experimentally (50 Hz–1 MHz) and theoretically in the extended frequency range. The effect of combined conductance on the real and imaginary part of the dielectric permittivity has been studied. Our results indicate a Debye type mechanism and empirical factor β gives the width of absorption of the samples. The empirical factor was found as 0.82 and 0.83 for E-201 and E-47 mixtures, respectively at 30°C. The distinctly different value of β for the perpendicular component in the nematic phase is interpreted as a distribution of relaxation times. These factors correspond to better understanding of the molecular order along the director.

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