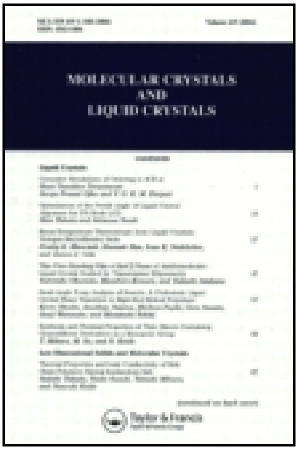

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Measurement of Dielectric Anisotropy of Some Liquid Crystals for Microwave Applications

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Measurement of Dielectric Anisotropy of Some Liquid Crystals for Microwave Applications

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This work presents the measurement of the dielectric anisotropy of selected nematic liquid crystals in the 0.01–6 GHz range, and comparison of the measurements with the birefringence values at optical frequencies. The effect of alignment on the reproducibility of dielectric measurements was also measured. It was found that a good surface alignment is essential to achieve reproducibility. Finally, it was experimentally confirmed that there is a strong correlation between the dielectric anisotropy in the microwave region and the birefringence at visible frequencies.

Keywords: birefringence; dielectric anisotropy; liquid crystals

INTRODUCTION

The growth of the microwave engineering industry, such as telecommunications, remote sensing and global navigation satellite systems etc., would be benefit from the development of materials with a large tunability of their dielectric properties in the microwave region. In comparison to the existing candidate materials such as ferroelectric ceramics, liquid crystals (LCs) have attracted attention in recent years because of low losses at microwave frequencies and comparatively high anisotropy [1–5]. For this reason, research has been focused on the use of liquid crystals in the GHz regime [6–12]. The work reported here includes the assessment of selected nematic liquid crystals in terms of dielectric permittivity modulation in the 0.01–6 GHz range.

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In particular, we discuss the correlation of dielectric properties at GHz frequencies with the refractive indices at optical frequencies. In addition, we examine the effects of the liquid crystal alignment on tunability and hysteresis.

EXPERIMENTAL

The RF measurement system consists of a network analyser (MS4623B, Anritsu) (frequency range 0.01–6 GHz), an RF test feature (BDS2200, Novocontrol), and a computer connected to the network analyser through a GPIB interface. A pair of parallel metal electrodes allows the introduction of either a solid (film-like) or liquid sample to form a capacitive load at the end of a transmission line. The complex dielectric permittivity is calculated by measuring the complex reflection coefficient of both the filled and unfilled capacitor.

The liquid crystals are all commercial products, with the high birefringence values typical of mixtures based on cyanobiphenyls. One mixture with particularly high birefringence, ITC, is a commercial mixture based on isothiocyanates. The electrodes were coated with a polyimide alignment and rubbed to give planar alignment of the LCs. An electric field up to 40 V AC (1 kHz) was applied to the electrodes to switch the LC into a homeotropic geometry. The LC layer thickness was fixed at 28 μm. The parallel ($\varepsilon_{||}$) and perpendicular ($\varepsilon_{\perp}$) components of dielectric permittivity were measured from the amplitude and phase of the reflected signal when a field is applied and then removed, respectively.

RESULTS AND DISCUSSIONS

1. Molecular Relaxation in the GHz Regime

Dielectric spectroscopy is a useful tool to probe the molecular relaxations of polar materials, especially aqueous solutions and liquid crystals. The relaxation process of propan-2-ol around 0.6 GHz was measured in the system because the dielectric spectrum is well known and the results can be used to check the correct performance of our system (Fig. 1). The relaxation time derived from figure 1 is 237 ps, which is in good agreement with published data [13].

The relaxation process for a high birefringence nematic liquid crystal mixture (Mixture ITC) is shown in Figure 2. The relaxation time derived from the figure is close to 1 ns. This spectrum was measured in the planar geometry (no applied field). This suggests that

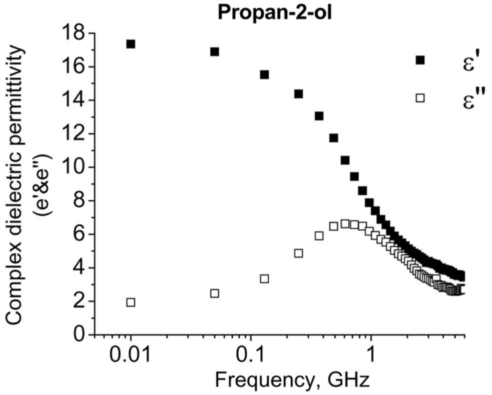

FIGURE 1 Dielectric spectrum of propan-2-ol.

the origin of the relaxation is related to molecular rotation around the long axis. In comparison with other nematic liquid crystals, the relaxation frequency of this material is high. Also shown in Figure 2 is a dielectric loss peak close to 0.1 GHz. However, the losses decrease to low values above 3 GHz. The same material possesses the highest dielectric modulation (22.5% at 3 GHz) amongst the test liquid crystals. However, in order to avoid large dielectric loss at ~1 GHz and the tail up to 3 GHz, for real dielectric modulation applications this liquid crystals mixture can only be used in frequencies above 3 GHz.

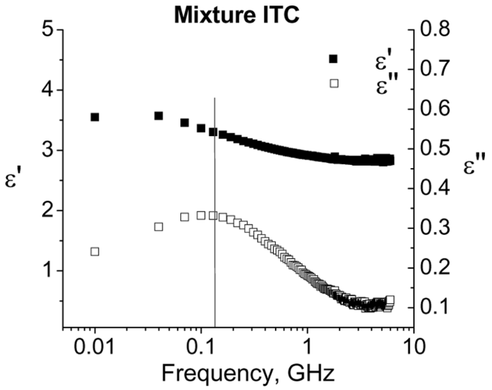

FIGURE 2 Dielectric spectrum of nematic liquid crystal (Mixture ITC) in planar alignment.

2. Dielectric Permittivity Modulations of nematic LCs in GHz

We selected nine nematic liquid crystals for the GHz dielectric measurements, with optical birefringence values ranging from 0.11 to 0.38. The perpendicular ($\varepsilon_{\perp}$) and parallel ($\varepsilon_{||}$) components of dielectric permittivity were measured under zero and finite applied electric fields. The dielectric tunability, or dielectric permittivity modulation, is defined as below.

$$\tau(\%) = \frac{\varepsilon_{||} - \varepsilon_{\perp}}{\varepsilon_{||}} \cdot 100\%$$

We compared our results with the data obtained from the literature. Pentyl cyano-biphenyl (K15) is a well known nematic liquid crystals, which has been measured in the RF frequency range by various methods [1,3,14]. For comparison purposes, we chose data which had been measured in a similar method to ours. Weil *et al.* [1] used a parallel capacitor method to measure the dielectric properties between 0.1 and 1 GHz. The tunability of 5CB at 1 GHz was larger than that measured in the present system (Table 1). In addition, the tan δ loss was 40–50% higher than our data.

We have identified two possible explanations for this disparity. Poor planar alignment may increase the measured value of $\varepsilon_{\perp}$. It is difficult to check the alignment quality visually due to our use of metal electrodes. Moreover, the tuning voltage range in our measurement system is limited to $40\,V_{p-p}$ by the bias tee. Since the cell thickness is about 28 μm, the resulting field strength of 1.43 V/μm may not be high enough to achieve the maximum tunability of the material. The second explanation is that the errors in the Weil experiment may be increased because the measurement was made at the edge of the frequency range, whereas it was in the middle in our case (frequency range: 0.01–6 GHz).

TABLE 1 Data of K15 in Comparison to the Data in the Literature [1]

5CB (1 GHz)	$\varepsilon_{||}$	$\varepsilon_{\perp}$	$\Delta\varepsilon$	tan $\delta_{||}$	tan $\delta_{\perp}$	Tunability (%)
Cambridge	3.18	3.07	0.11	0.055	0.095	3.5
Ref. [1]	3.27	3.13	0.14	0.084	0.151	4.3

3. The Effect of Alignment on the Dielectric Permittivity Modulation of Nematic Liquid Crystals

In the voltage controlled tunability measurements, we measured the effect of the surface alignment on measurement reproducibility. We used cells both with and without surface alignment. We found that in test cells without alignment the value for $\varepsilon_{\perp}$ could not be reproduced after the external field was removed and reapplied. The value was larger than the initial value, indicating an incomplete return to planar geometry. In the test cells with a rubbed polyimide layer, the value of $\varepsilon_{\perp}$ in the planar state (0 V) returned to its initial value when the voltage was returned to zero. The aligned test cell could be switched many times without changing the measured values of $\varepsilon_{||}$ and $\varepsilon_{\perp}$. The dielectric permittivity of E7 is plotted against applied voltage in Figure 3. It increases when the applied voltage exceeds about 1 V (the Freedericksz transition threshold voltage) and saturates at 40 V, where the molecules align with the field direction.

4. Correlation between Dielectric Anisotropy in the Microwave Regime and the Optical Birefringence

The real and imaginary parts of the dielectric permittivity which were measured at 3 GHz and the refractive index values at 589 nm given by the manufacturer are listed in Table 2. The dielectric anisotropy varies between 0.25 and 0.83, with low associated losses (typically $\tan\delta_{\perp} \sim 0.05$ and $\tan\delta_{||} \sim \tan\delta_{\perp}/2$). The dielectric anisotropy $\Delta\varepsilon$ at 3 GHz is plotted versus birefringence Δn at 589 nm for two sets of data in Figure 4. One set of data is taken from Table 2. The other set is from the work by Lim *et al*. at 30 GHz [14].

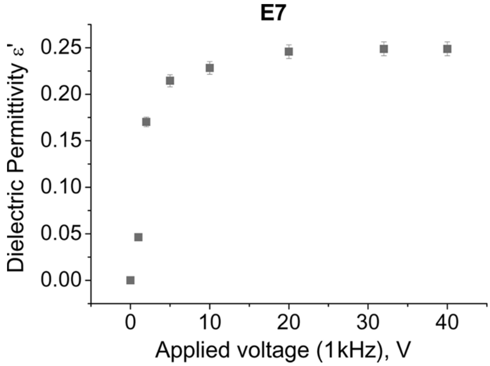

FIGURE 3 Dielectric permittivity *vs*. applied voltage for E7.

TABLE 2 Dielectric permittvity (3 GHz) and refractive index (589 nm) of the tested nematic liquid crystals

	3 GHz						589 nm		
Nematic LCs	$\varepsilon_\perp$	$\varepsilon_{\|\|}$	$\Delta\varepsilon$	tan $\delta_\perp$	tan $\delta_{\|\|}$	τ (%)	n_e	n_o	Δn
ZLI2222/000	2.77	2.91	0.14	0.04	0.025	4.6	1.6134	1.5010	0.1124
K15	3.07	3.18	0.11	0.095	0.055	3.4	1.71	1.53	0.18
E7	2.87	3.11	0.24	0.05	0.02	7.7	1.7462	1.5216	0.2246
BL093	2.89	3.13	0.24	0.06	0.02	7.7	1.7697	1.5241	0.2456
BL048	2.95	3.23	0.28	0.04	0.02	8.7	1.7904	1.5277	0.2627
MDA1602	2.83	3.11	0.28	0.06	0.04	9.0	1.7779	1.5113	0.2666
BL111	2.89	3.21	0.32	0.06	0.03	10.0	1.8025	1.5321	0.2704
BL037	2.96	3.32	0.36	0.06	0.027	10.8	1.808	1.526	0.2820
Mixture ITC	2.79	3.6	0.81	0.05	0.035	22.5	1.915	1.535	0.38

For both the data measured at 3 GHz and the data measured by Lim *et al.* at 30 GHz, the magnitude of dielectric anisotropy $\Delta\varepsilon$ correlates with the optical birefringence. Our measurements show a high correlation coefficient of 0.88 whereas the data measured by Lim *et al.* shows a correlation coefficient of 0.8. The normalised data, tunability (τ%, 3 GHz) versus $\Delta n/n_e$, is plotted in Figure 5. The correlation coefficient here is 0.87.

The high correlation coefficients indicate that, over a wide range of liquid crystals, the dielectric anisotropy in the microwave regime is

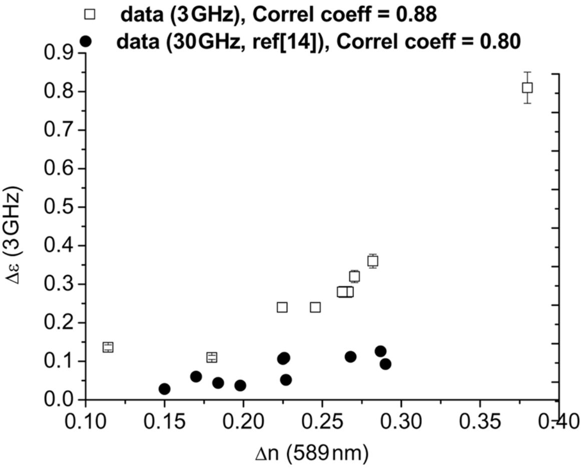

FIGURE 4 Dielectric anisotropy $\Delta\varepsilon$ (in microwave regime) vs birefringence Δn (589 nm).

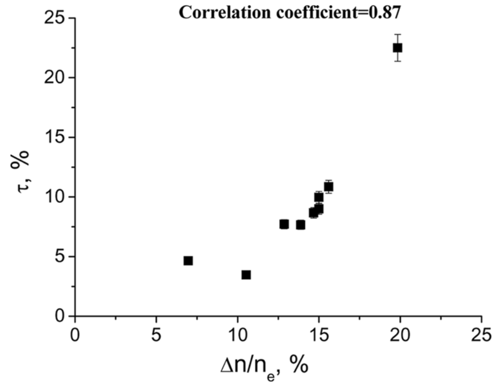

FIGURE 5 Tunability T versus $\Delta n/n_e$ measured for a range of liquid crystals.

correlated with the birefringence in the visible. This finding is supported by other research work by Nose *et al*. [3].

The correlation is useful in addition to its scientific interest. We can select a liquid crystal for $\Delta\varepsilon$ in MMW regime, according to its birefringence value at optical frequencies.

We also evaluate the data in terms of the correlation between the components of the anisotropy, $\varepsilon_{||}$ or $\varepsilon_{\perp}$ at 3 GHz, with n_e^2 or $n_o^2(\lambda = 589\,\text{nm})$, as shown in Figures 6 and 7.

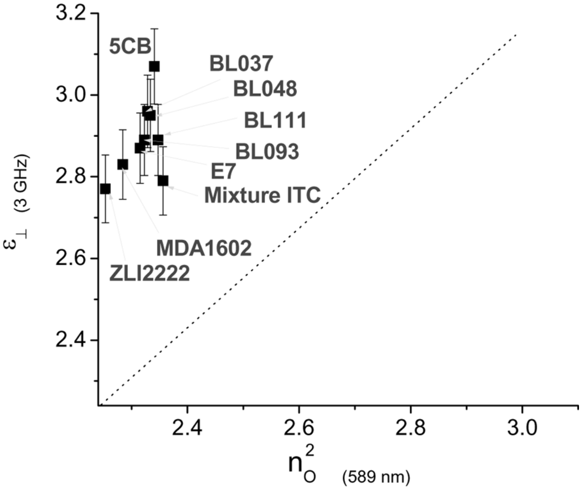

FIGURE 6 $\varepsilon_{\perp}$ (3 GHz) vs n_o^2 (589 nm) for measured liquid crystals.

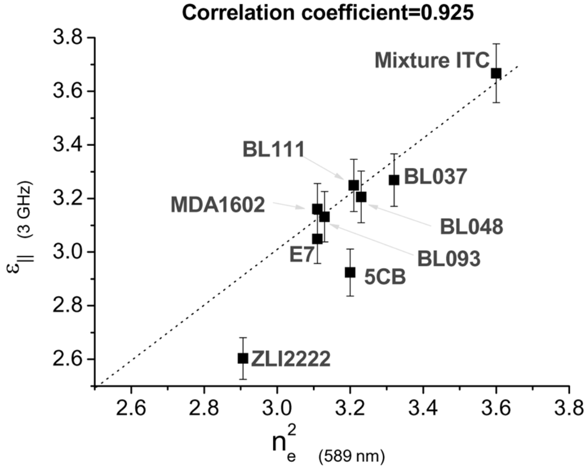

FIGURE 7 $\varepsilon_{\parallel\ (3\mathrm{GHz})}$ vs $n^2_{e\ (589\mathrm{nm})}$ for tested liquid crystals.

An interesting feature found in Figures 6 and 7 is the different correlation coefficients exhibited by the parallel and perpendicular components. The correlation coefficient for the parallel components was 0.925, whereas, for the perpendicular components, a lower correlation

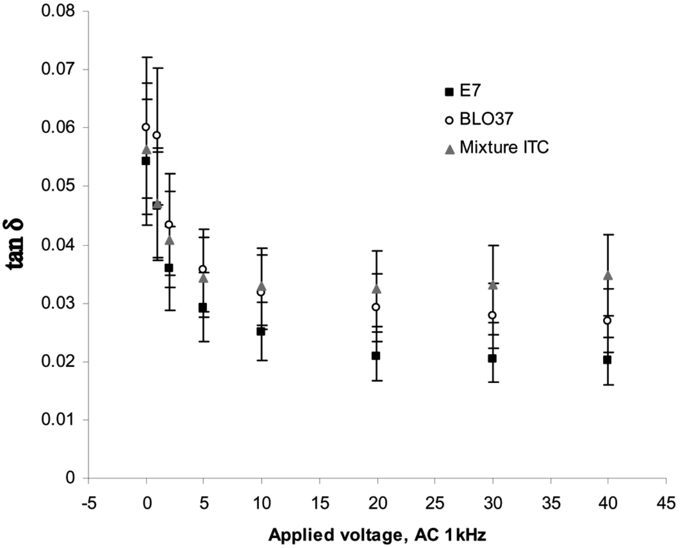

FIGURE 8 Loss tan δ versus applied voltage for three liquid crystals.

coefficient 0.486 was obtained. To understand the physical origin of this behaviour is currently the subject of further work.

A further experimental finding is that the measured dielectric loss $\tan\delta_{||}$ was almost half of $\tan\delta_{\perp}$ (see Table 2). Figure 8 shows, for three liquid crystals, how the loss $\tan\delta_{\perp}$ ($V = 0$) decreases towards $\tan\delta_{||}$ ($V = 40\,V$), as the directors are re-oriented to approach the homeotropic state by increasing the voltage. Similar behaviour was also found by other researchers [15]. This may be attributed to different molecular relaxations in the planar and homeotropic states [16]. In the initial planar state (no electric field), the dielectric dispersion at high MHz frequencies is caused by the molecular rotation around long axis with a characteristic relaxation frequency $f_{\perp}$. In the homeotropic state (applied external field), the dielectric absorption in the MHz regime is dominated by tumbling about the molecular short axis with a relaxation frequency ($f_{||}$). Due to the different rotational viscosities for the two relaxation processes, generally $f_{\perp} > f_{||}$ [16]. Therefore, the tail of the loss curve at 3 GHz is lower for the $f_{||}$ absorption than for the $f_{\perp}$ absorption. Consequently, we expect that $\tan\delta_{\perp} > \tan\delta_{||}$. This is also a subject of further investigation.

CONCLUSIONS

In summary, we have measured dielectric properties of nematic liquid crystals with a range of birefringence values in the region from 0.01 GHz up to 6 GHz. At 3 GHz, the maximum tunability was found to be 22.5% for a high birefringence liquid crystal (Mixture ITC). The materials studied all have low dielectric loss. We also find that good surface alignment is essential to achieve a good reproducibility of the dielectric modulation. Finally, we experimentally confirmed that there is a strong correlation between the dielectric anisotropy in the microwave region and the birefringence at visible frequencies.

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