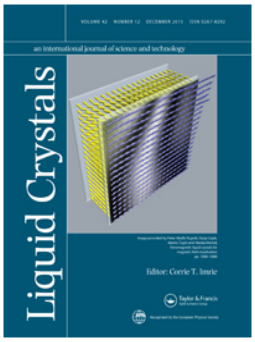

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Role of cell thickness in tailoring the dielectric and electro-optical parameters of ferroelectric liquid crystals

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Here, we report thickness dependence of dielectric and electro-optical parameters in ferroelectric liquid crystals (FLCs) without surface stabilisation. The dependence of dielectric and electro-optical parameters on cell thickness is observed by dielectric spectroscopy and electro-optical measurements. The dielectric permittivity (ε') measured by varying the cell thickness showed increase of ε' with increase of cell thickness which is attributed to the presence of more ions and larger contributions of Goldstone mode in thick cells. The spontaneous polarisation also shows increment with increase of cell gap up to certain thickness range. The rotational viscosity decreases with increase in the cell thickness whereas the response time is more for thicker cells. The decrease in the rotational viscosity is attributed to lowering of elastic deformation with increase in cell thickness and the response time is directly proportional to cell gap. These studies would be utilised to understand the effect of cell thickness on dielectric and electro-optical properties of FLC materials and optimising the material parameters with cell thickness for better and efficient liquid-crystal-based devices.

Keywords: ferroelectric liquid crystal; dielectric permittivity; spontaneous polarisation; rotational viscosity; response time; cell thickness

1. Introduction

The encroachment in dielectric, electro-optical, material and other parameters of liquid crystals (LCs) is in always great demand in order to fabricate better and efficient liquid crystal display devices. The systematic studies on improvement of these parameters become more important in case ferroelectric LCs (FLCs) than their nematic counterpart as much of the research these days in the field of LCs revolves around the FLCs because they are quite promising in terms of high speed, better optical contrast, low threshold voltage, more durability and memory effect.[1–6] The dependence of dielectric, electro-optical, physical, thermal properties of FLCs on various parameters such as cell thickness, material under investigation, phase transition, applied electric field and surface anchoring has been extensively studied by various groups around the world.[7–18] But, the sample cell thickness dependency of dielectric, electro-optical and other material parameters is the subject of considerable interest as most of the practical applications of FLCs such as displays, image control, spatial light modulator or memory devices, which make them popular in today's world, come with constraints of cell thickness.

The cell thickness dependency of dielectric, electro-optical and other material parameters are well reported. Yoshino et al. studied the influence of electric field and cell thickness on dielectric behaviour of FLC at phase transition and confirmed that the phase transition of these compounds is of the second order.[7] Nie and co-workers studied theoretically and validated experimentally that cell gap affects the LC response time where the anchoring energy was also taken into account.[8] Mukherjee et al. found that a pure deformed helix FLC (DHFLC) material (MOPBIC) shows large dielectric constant which increases with increasing cell thickness. The authors attributed the high values of dielectric constant to molecular contributions from different bonds, groups and conformers to the dipole moment and the deformed helix configuration of the molecular structure of material studied.[9] Panarin et al. investigated low-frequency dielectric response of the surface stabilised ferroelectric liquid crystal (SSFLC) cells and found that the dielectric response depends on the type of the smectic layer structure present and the cell thickness.[10] The authors developed an analytical solution to the equation of motion that describes the dynamics of SSFLC cells in the presence of a weak external field for both bookshelf and chevron geometries and observed that experimental values of the dielectric relaxation strength and the relaxation frequency show good agreement with those predicted

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by the theoretical model for the chevron structure. In sufficiently thick FLC cells, the dielectric properties are governed by the helical structure and the relaxation time of the Goldstone mode (GM) depending on helical pitch.[11]The dependence of the relaxation mode/modes on the sample thickness has been investigated.[12] The authors investigated dielectric properties of SSFLC cells both experimentally and theoretically and found that the thickness mode consist of two relaxation processes. Kundu et al. measured thickness-dependent dielectric constant by varying the cell thickness and showed increase of dielectric constant and the ferroelectric transition with increase of cell thickness.[13] The authors described non-linear variation of transition temperature by using an empirical formula of the form $T_x = T_C - \gamma d^{-\beta}$ (where β and γ are sample-dependent parameters). The dependence of switching and memory times of optical switching element utilising FLCs on thickness of cell and material was also studied.[14] The authors observed that switching times, rise time, decay time and memory time, of various electro-optic effects in FLCs are found to be strongly dependent on the molecular structure, cell thickness and temperatures. Some recent studies on role of cell thickness on the dielectric [16] and thermodynamic properties [17,18] of LC materials showing smectic C* phase is reported. Also, the effect of cell thickness and spontaneous polarisation on the demixing phenomenon which is attributed to the thermal hysteresis of antiferroelectric compounds is studied in detail.[18]

In this article, we report thickness dependence of dielectric and material constants in non-SSFLC cells. The experimental results have been analysed by dielectric spectroscopy and electro-optical measurements. Thickness-dependent dielectric permittivity (ε') showed increase of ε' with increase of cell thickness. The spontaneous polarisation also shows increment with increase of cell thickness. The rotational viscosity decreases with increase in the cell thickness whereas response time increases for thicker cells. The underlying studies would be useful for understanding the effect of cell thickness on dielectric and electro-optical properties of FLC materials and optimising the material parameters for better and efficient LC-based devices.

2. Experimental

2.1. *Liquid crystal sample cell fabrication*

The LC sample cells have been fabricated by using highly conducting (30 ohms/square) and optically transparent sputtered indium tin oxide glass substrates. The desired (square) electrode patterns having the area 4.5 mm × 4.5 mm were obtained by using positive photolithographic (using positive photoresist) technique. The homogeneous alignment of LC sample cells has been achieved by using conventional rubbed polyimide technique. The thickness of the cells was maintained by using Mylar spacers of varying thicknesses.

2.2. *Detail of FLC materials used*

Two commercially available FLC materials, namely KCFLC 10S and ZLI 3654, were used in the present studies. The FLC mixture, KCFLC 10S, was purchased from Kingston Chemicals Limited, UK. In KCFLC 10S, the compound dialkyl ortho-difluoroterphenyls was used as host and the addition of 10% of chiral dopant, cyanohydrin, to host mixture generated a FLC mixture with a SmC* phase. The chemical structure of these two major components of KCFLC 10S is already reported.[19] Two basic compounds of FLC mixture ZLI 3654 are also available in the some report.[20]

The phase sequence of the FLC mixtures used is as follows:

$$cryt \xleftrightarrow{<5^{\circ}C} SmC* \xleftrightarrow{64.5^{\circ}C} SmA* \xleftrightarrow{99.5^{\circ}C} N \xleftrightarrow{112^{\circ}C} iso_{(\mathrm{KCFLC\ 10S})}$$

$$cryt \xleftrightarrow{<-30^{\circ}C} SmC* \xleftrightarrow{62^{\circ}C} SmA* \xleftrightarrow{76^{\circ}C} N* \xleftrightarrow{86^{\circ}C} iso_{(\mathrm{ZLI\ 3654})}$$

where *cryst* is the crystalline phase, *SmC** is chiral smectic C phase, *SmA** is the chiral smectic A phase, *N* is the nematic phase, *N** is chiral nematic and *iso* is the isotropic phase. The other parameters specified by the manufacturers of both FLC mixtures have been listed in Table 1. In the present study, in order to obtain the non-SSFLC configuration the thicknesses of cells are varied from ~4 μm to ~20 μm, since the helical pitch of FLC materials used, KCFLC 10S and ZLI 3654, are 2.5 and 3 μm respectively.

2.3. *Apparatus and measurements*

The ε' of FLC materials filled cells has been measured using an impedance analyser 6540A (Wayne Kerr, UK) in a frequency range of 20 Hz–1 MHz. The

Table 1. Material parameters of FLC mixtures, KCFLC 10S and ZLI 3654.

Parameter/property	KCFLC 10S	ZLI 3654
Spontaneous polarisation	31 nC/cm^2	29 nC/cm^2
Rotational viscosity	~285 mPa.s	~380 mPa.s
Optical tilt angle	23°	25°
Helical pitch	~2.5 μm	~3 μm
Optical anisotropy (birefringence)	0.18	0.13

optical micrographs of the sample cells were taken with the help of polarising optical microscope (Ax-40, Carl Zeiss, Germany) fitted with charge-coupled device camera. The material parameters such as spontaneous polarisation, rotational viscosity and response time were measured by automatic LC tester (ALCT, Instec, USA).

3. Results and discussion

The variation of ε' with cell thickness has been studied in FLC materials, KCFLC 10S and ZLI 3654. Figure 1 shows the behaviour of ε' of FLC materials KCFLC 10S and ZLI 3654 as a function of frequency for cells with different thickness. It is clearly reflected from Figure 1(a) that ε' of FLC material KCFLC 10S is highly thickness dependent and ε' increases gradually with increase of cell gap. We confirmed this behaviour of ε' with cell thickness in another FLC material ZLI 3654. The behaviour ε' of ZLI 3654 as a function of frequency for cells having different thicknesses has been shown in Figure 1(b). It can be clearly seen from Figure 1(b) that ZLI 3654 shows thickness-dependent dielectric constant measured by varying the cell thickness. In this way, both FLC materials showed increase of dielectric constant with increase of cell thickness. The reason behind the increase of ε' with increase of cell thickness in FLC materials is twofold. First, the increase in ε' with increase in cell thickness can be understood by the fact that it is directly related to the net charge stored within a LC cell which acts as a capacitor. As the cell thickness increases, the presence of ions in FLC material can contribute largely to ε' in the low-frequency regime resulting in the higher values of ε'.[21] Second, the contribution of GM varies with cell thickness. In thick cells, the large value ε' in the SmC* phase represents the contribution of GM indicating the presence of helical structure. Unlikely in thin cells, ε' approaches to a much lower value as the contribution of GM is suppressed for the thinner sample.[9]

Like ε', spontaneous polarisation (P_S) of FLC materials shows variation with cell thickness. Figure 2 shows the behaviour of P_S for FLC materials KCFLC 10S and ZLI 3654 as a function of applied voltage for cells having different thickness. It is clearly reflected from figures that thickness-dependent P_S measured by varying the cell thickness showed first increase with increase in cell thickness and then decrease in thicker cells. The increase of P_S with cell thickness can be explained by the taking the variation of dielectric strength ($\Delta\varepsilon$) with P_S and tilt angle into account [22–24]

$$\Delta\varepsilon = \frac{P_S^2}{2K_3 q^2 \theta^2} \tag{1}$$

where K_3 is the twist elastic constant and q is the wave vector of the helix. As can be seen from Equation (1), P_S is directly proportional to $\Delta\varepsilon$ (which is proportional ε'). So, increase in P_S is due to increment in the magnitude of ε' which also indicates that the dipole moment of the molecules plays important role in the enhancement of the dielectric constant. The similar behaviour of P_S with cell thickness in DHFLCs has also been reported.[9,25] The decrease in P_S in case of thicker cells is due to poor alignment of cells. Due to poor alignment of thicker cells, the dipoles are randomly oriented and P_S decreases. The dark and bright states of the KCFLC 10S-filled cells, which are treated for homogeneous surface alignment with increasing cell thickness, are shown in Figure 3. It is clearly seen from Figure 3 that the alignment of cells becomes poorer with increase of cell gap, which in turn reduces the P_S for thicker cells.

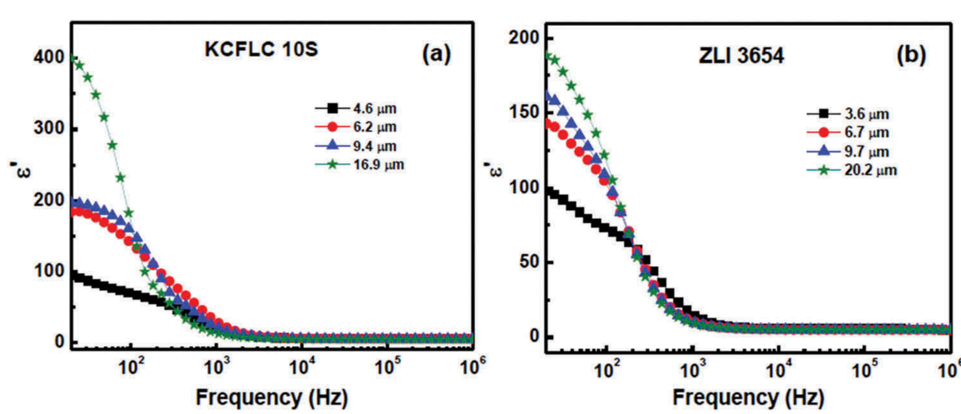

Figure 1. (Colour online) Behaviour of dielectric permittivity (ε') with frequency at room temperature for cells of different thicknesses filled with FLC material (a) KCFLC 10S and (b) ZLI 3654.

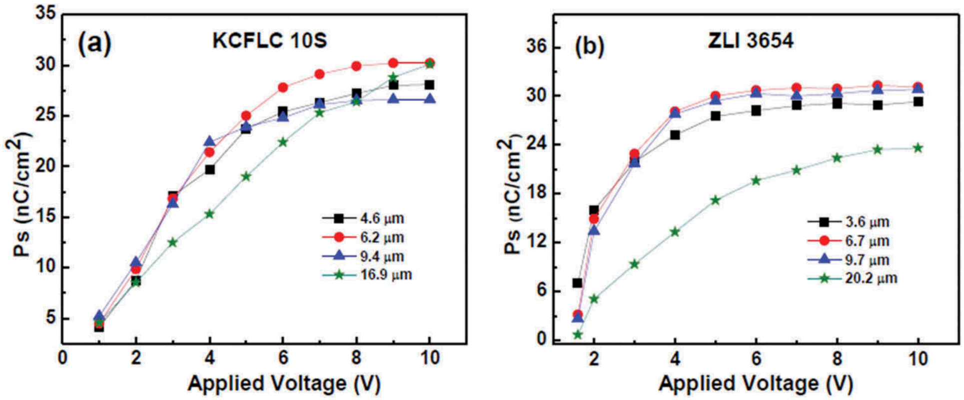

Figure 2. Behaviour of spontaneous polarisation with applied voltage at room temperature for cells of different thicknesses filled with FLC material (a) KCFLC 10S and (b) ZLI 3654.

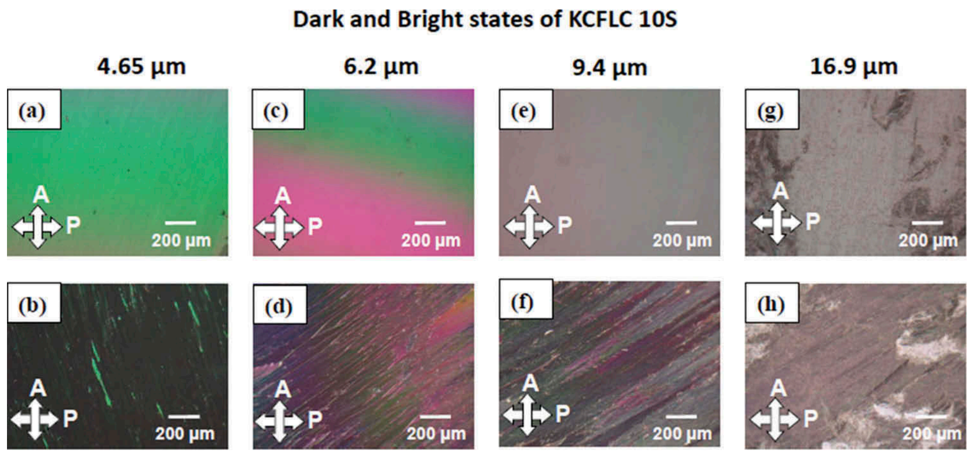

Figure 3. Optical micrographs of the FLC material KCFLC 10S with bright and dark states showing the alignment of filled cell having the thickness of (a, b) 4.65 μm, (c, d) 6.20 μm, (e, f) 9.40 μm and (g, h) 16.90 μm. Scale bar: 200 μm .

The behaviour of rotational viscosity (γ) with cell thickness has been observed as γ is a parameter which determines the ease of response of mesogens to electrical stimulus and also could be responsible for memory effect in certain types of FLCs. Figure 4 shows the variation of γ with applied voltage for cells of varying thickness for FLC materials KCFLC 10S and ZLI 3654. As reflected from Figure 4(a), the thickness-dependent γ measured by varying the cell thickness showed decrease in the former with increase in cell thickness in case of KCFLC 10S mixture. Another FLC material ZLI 3654 also exhibits the similar behaviour which is shown in Figure 4(b). The decrease in γ with increase in cell thickness can be explained on the basis of decrement of elastic deformation in thicker cells. The elastic deformation is directly linked to viscosity of the material.[26] Higher the cell thicknesses, the lower the compactness of the layer structure and hence lower the viscosity.

The behaviour of response time (τ) with cell thickness has also been studied as the response time, among many intrinsically physical parameters of LC material, is the most important parameter for addressing the fast LC display devices. It has been studied theoretically and validated experimentally that the anchoring energy and cell gap affect the LC response time.[8] The relation of response time with the anchoring energy and cell thickness for strong and weak anchoring respectively can be expressed as follows:[8]

$$\tau \approx \frac{\gamma}{K\pi^2}\left(d^2 + \frac{4dK}{W}\right) \tag{2}$$

$$\tau \approx \frac{4\gamma d}{W\pi^2} \tag{3}$$

where τ is the response time, γ is the rotational viscosity, K is bend elastic constant, d is cell gap and W is the anchoring energy strength coefficient respectively.

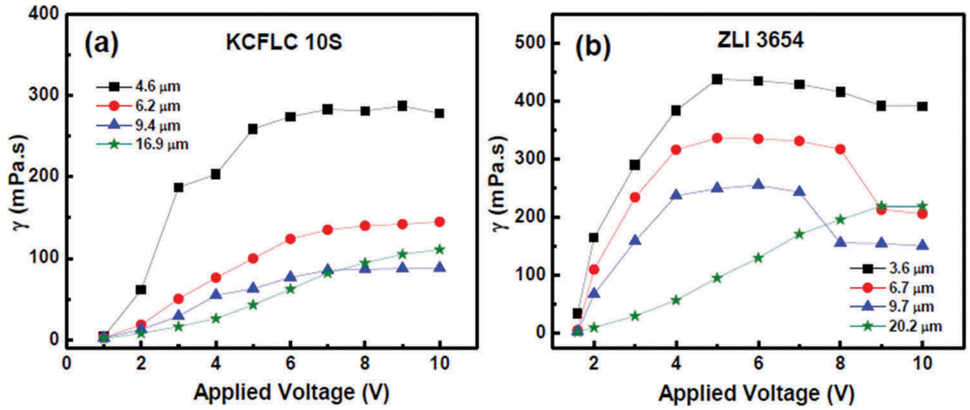

Figure 4. (Colour online) Behaviour of rotational viscosity with applied voltage at room temperature for cells of different thicknesses filled with FLC material (a) KCFLC 10S and (b) ZLI 3654.

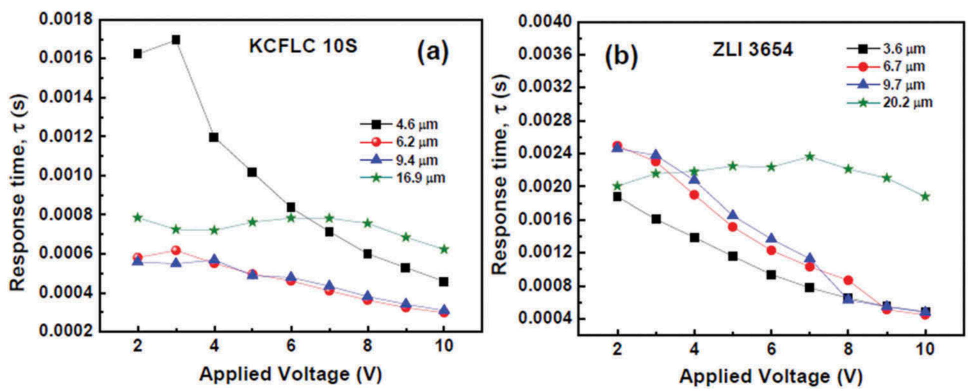

Figure 5. (Colour online) Variation of response time with applied voltage at room temperature for cells of different thicknesses filled with FLC material (a) KCFLC 10S and (b) ZLI 3654.

From Equations (2) and (3), it can be seen that τ is directly proportional to the cell gap in both cases. If one keeps anchoring energy constant, then τ should increase with increase in the cell thickness. Figure 5 shows the variation of τ with applied voltage for cells of varying thickness for FLC materials KCFLC 10S and ZLI 3654. It is seen from Figure 5(a) that τ measured by varying the cell thickness showed an initial decrease in τ for comparatively thinner cells due to the rapid decrement of γ (as seen in Figure 4(a)) for those cells. For thicker cells of KCFLC 10S material, the value of τ increased. The increase in τ with increment of cell thickness as explained in Equations (2) and (3) is observed in another FLC material ZLI 3654 (which is shown in Figure 5(b)). Actually the cells of varying thickness used in the studies were prepared by keeping almost the similar surface treatment for all cells. Therefore, τ is thickness dependent and increases with increase in the cell thickness.

It can be stated from observations made in the present study that cell thickness plays an important role to optimise the dielectric and material constants. If one uses thick cells then high dielectric permittivity and better spontaneous polarisation can be obtained but at the same time the alignment in such cells will be poor and the same will not be worthy for faster display devices and memory applications too. Keeping these things in mind, the LC cells having thickness ~4–7 μm may be preferred for the fabrication of LC/FLC-based display and non-display devices.

4. Conclusions

The thickness dependence of dielectric and electro-optical parameters in FLCs has been studied. Thickness-dependent ε' measured by varying the cell thickness showed increase of ε' with increase of cell thickness. The spontaneous polarisation also shows an increment with increase of cell thickness. The rotational viscosity decreases with increase in the cell thickness whereas the response time is more for thicker cells. The studies carried out would be utilised to understand the effect of cell thickness on dielectric and electro-optical properties of FLC material and optimising the material parameters by taking the cell gap into account for better and efficient LC display devices.

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Disclosure statement

No potential conflict of interest was reported by the authors.

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