Effect of dc field on texture and electrohydrodynamic instability in cyanobiphenyl mixture

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Effect of dc electric field on a homeotropically oriented liquid crystal mixture of positive dielectric anisotropy, has been studied. Detailed studies on electrohydrodynamic (EHD) instability were carried out on liquid crystal cells of four different thicknesses. Microscopic studies of its textures at different voltage were also carried out. Existence of charge injection, charge diffusion and convective flow of liquid in the liquid crystal cells was observed. Effects of anisotropy in viscosity, dielectric permittivity and other physical parameters on electric current through the sample cells were discussed.

1 Introduction

Electrohydrodynamic (EHD) instability in nematic liquid crystals was studied extensively by many workers. Williams\textsuperscript{1-3} and his co-workers studied effects of electric field on nematic liquid crystals having negative dielectric anisotropy and observed the occurrence of EHD instability for the first time. Nakagawa and co-workers\textsuperscript{4} and Rout and Choudhary\textsuperscript{5,6} studied EHD instability of pure cyanobiphenyls with positive dielectric anisotropy. In homeotropic orientation of these liquid crystals in the nematic phase they observed EHD instability as roll-like domains. This type of instability was found similar to the Felici-instability \cite{Felici} in isotropic liquids in a dc field\textsuperscript{7}, this was analogous to the thermally induced Rayleigh-Benard instability\textsuperscript{8,9}, and hence named as Felici-Benard instability\textsuperscript{5,6} \cite{Felici-Benard}. Charge injection from the electrodes to the liquid crystals, diffusion of charges and convective flow of liquid accompanied by changes were considered to be the main reasons for FBI. The principles of FBI were used to explain EHD instability of homeotropically oriented cyanobiphenyls\textsuperscript{5,6} and their binary mixtures\textsuperscript{9}. Since the liquid crystals possess anisotropy in dielectric, conductive and mechanical properties the influences of these anisotropy are required to be considered.

The device parameters and physical properties of pure materials can be enhanced or changed drastically by mixing the two or more compounds in different ratios or concentrations. From the preliminary physical data on mixtures of two or more cyanobiphenyls\textsuperscript{10} it is clear that some of these mixtures have very interesting properties and may be very useful for devices.

In view of the importance of mixed liquid crystals we have taken the mixture $S_{2c}$ to study which is a eutectic mixture of two cyanobiphenyls and has a small nematic range. With this short nematic range, the values of different physical parameters may be changed with a considerable higher rate with rise in temperature and the effect of those changes on textures and EHD instability in a dc electric field can be observed easily. The usefulness of the material on device applications can also be studied.

2 Experimental Details

Commercially available mixture of two cyanobiphenyls, $S_{2c}$, was kindly gifted by M/s BDH, UK, for the present study. Liquid crystal cells of four different thicknesses were prepared with $SnO_2$ coated glass plates between which the sample was sandwiched. Mylar spacers of four thicknesses (17, 23, 50 and 75$\mu m$) were used to determine the thickness of the sample. Homeotropic alignment of the molecules of the liquid crystals was obtained by treating the glass plates with CTAB (Cetyl trimethyl ammonium bromide). The temperature was controlled and measured with 0.1$^\circ C$ interval with a Mettler central processor (FP-80) and a hot stage
Fig. 1 — Microphotographs of domain patterns in sample SX2 (magnification 150 x) for the 23 µm thick cell with applied voltage 1(a) 3 V, 1(b) 3.5 V, 1(c) 4 V, 1(d) 5 V, 1(e) 7 V at temperature 47.2°C. For the 17 µm thick cell with applied voltage 1(f) 4.8 V at 46.8°C. For the 50 µm thick cell with voltage 1(g) 4 V, 1(h) 4.8 V, 1(i) 5.5 V at 46.8°C. For the 75 µm thick cell with applied voltage 1(j) 4 V, 1(k) 5 V, 1(l) 6 V, 1(m) 8 V and 1(n) 11 V.
3 Results and Discussion

3.1 Microscopic studies

The variation in texture of $S_{2C}$ provides the following sequence of phase transition:

$$S_{2C} \rightarrow 44.6^\circ C \rightarrow N \rightarrow 47.5^\circ C \rightarrow I$$

where $S_{2C}$ = smectic phase, $N$ = nematic phase and $I$ = isotropic phase. The phase transition temperatures observed were found slightly different from those of the reported values.$^{10}$

The textures of $S_{2C}$ at different voltages for different cells are shown in Fig I [Figs 1(a)-1(e) are for 23 $\mu$m thin cell, Fig. 1(f) is for 17 $\mu$m thin cell, Figs 1(g)-1(i) are for 50 $\mu$m thick cell and Figs 1(j)-1(n) are for 75 $\mu$m thick cell]. Before application of electric field to the cells, the liquid crystal film appeared dark under the microscope with the polarizer and analyzer in crossed position. The darkness was due to homeotropic alignment of the molecules. In some cells, a few bright spots were observed indicating the improper alignment of molecules in those areas. When the voltage was applied above 2 V, the whole area of the cell under the microscope became dark. It indicated the completion of homeotropic alignment of the molecules all-over the cell.

In the liquid crystals of positive dielectric anisotropy (i.e. $\Delta\varepsilon > 0$), Freudenkzsz transition from planer to homeotropic alignment is possible if an electric field is applied along the cell thickness. The threshold voltage for this transition depends on the splay elastic constant and dielectric anisotropy$^{11}$ $V_{th} = \frac{C}{K_{11}}(\Delta\varepsilon)^{1/2}$, where $V_{th}$ = threshold voltage for above-mentioned Freudenkzsz transition, $C$ = constant, $K_{11}$ = splay elastic constant]. Our observation shows that $S_{2C}$ mixture contains positive dielectric anisotropy. It was seen that at temperature close to $T_{SN}$ ($T_{SN} = $ Smectic to nematic transition temperature of the mixture $S_{2C}$) high voltage was required to align the molecules completely. It showed that elastic constants of the nematic liquid crystal decrease with the increase of temperature. It was also observed that keeping the applied voltage above $V_{th}$, if the material is cooled down, the homeotropic alignment remains unchanged after N-$S_{2C}$ phase transition and in the smectic phase.

From the above observations, it can be suggested that the material can be used in optical switching devices with a small operating dc voltage [i.e. within 2-2.5 V], for a small temperature range, 45 - 46.5°C. Since the material can retain the homeotropic alignment in the smectic phase so it can be used in optical memory devices at room temperature range [smectic range of $S_{2C}$ is from 0°C to 44.6°C].

Above the threshold voltage $V_{th}$, the voltage required to start EHD instability, the cellular domain patterns started appearing. $V_{th}$ was found to be 2.9 V for 23 $\mu$m cell and 3.6 V for 75 $\mu$m cell at 47°C. At lower temperature, $V_{th}$ was found slightly higher, 3.2 V for 23 $\mu$m cell, at temperature 46°C (Fig. 2). From the relation$^{7}$, $V_{th} = k\eta\mu/\varepsilon_0$, $\eta$ = viscosity, $\mu$ = drift mobility of the charges in the liquid crystal, $k$ = constant and $\varepsilon$ = permittivity we can see that $\eta$ decreases with increasing temperature$^{12}$. The molecules were kept aligned homeotropically, hence $\varepsilon$ can be taken as constant. From the current-voltage characteristics, it was found that the current did not change much with temperature at $V_{th}$ in the nematic phase and hence $\mu$ is not so much influenced by temperature in the short nematic range of $S_{2C}$. It may be concluded that $V_{th}$ decreases with decreasing $\eta$. At a temperature, close to nematic to isotropic transition temperature ($T_{NI}$), it was found that $V_{th}$ was slightly higher again (3.2 V at 47.4°C for 23 $\mu$m cell). The order parameter decreases significantly at this temperature$^{13}$ and the thermal vibrations of the directors was high.

![Fig 2 - Threshold voltage - temperature characteristics for 23 $\mu$m cell](image-url)
about their mean directions therefore higher electric field was necessary to orient the molecules along its direction. To make the optical patterns visible, a voltage was required to be applied which was higher than the threshold voltage, $V_{th}$, necessary to start EHD instability in the liquid crystal at that respective area. Careful analysis did not show any definite relation between cell thickness and $V_{th}$. However for 23 μm cell $V_{th}$ was low. With increase of voltage above $V_{th}$, the roll-like domain pattern appeared with shorter roll-length, an increase in roll-length with applied voltage was observed. But with a further increase in voltage, contraction in roll-length was observed tending towards a round shape patterns [Figs 1(a)-1(d), 1(g)-(i) and 1(j)] with the appearance of central crosses at and above 5 V. At 6 V the domain patterns started distorting and above 6.5 V fast turbulent flow was observed. Above 7.5 V it became completely irregular and looked like a boiling liquid [Figs 1(e), 1(f) and 1(k)-(n)].

Turbulent flow of liquid broke the regularity, hence the local periodicity of the space charge and the electric field within the cell were disturbed accordingly. As a result, molecular orientations suffered spatial and temporal fluctuations.

### 3.2 Electrical Measurements

**(i) Current-voltage characteristics** — Figs 3(a)-3(d) show the current (I)-voltage (V) characteristics at different temperature ($T$) for all the four cells. The current in the thin cells (17 μm and 23 μm cells) was found higher than that of thick cells (cells with thickness 50 and 75 μm) for the same applied voltage. When the voltage was below 2 V, a very small current was found to flow. It was due to ohmic conduction of current through the liquid crystal. Above 2 V, the current increases with increasing voltage. At this stage charge injection took place and diffusion current started to flow, resulting in a higher current. Above $V_{th}$, the force exerted on space charge was sufficient to start convection flow in the liquid crystal, and hence the cellular domains appeared and the charge transfer with liquid flow was induced. As the voltage increases above $V_{th}$, the current due to charge transfer with convective flow of liquid becomes dominant and then the total current increases very fast with

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**Fig. 3(a)** — Current-voltage characteristics at different temperatures for S2C for cell thickness 17 μm

**Fig. 3(b)** — Current-voltage characteristics at different temperatures for S2C for cell thickness 23 μm
rise in voltage. When the liquid crystal was in isotropic phase, the same type of current-voltage characteristics was observed.

In the smectic phase currents started increasing at 2.5 V, and above 3.5 V, the slopes of $I-V$ curves became small and constant. For thick cells it appeared almost zero. In microscope, no flow of liquid was observed up to 10 V in this phase. Combining it with electrical observations, one can easily conclude that above 2.5 V diffusion current appeared, in addition to the ohmic part but no electric current was found due to fluid convection. As a result, in smectic phase, slope of the current-voltage curves gradually tends to get a constant value.

(ii) Existence of charge diffusion — Figs 4(a)-4(d) show the $I(t)-t$ transient characteristics of the sample, where $I(t)$ is the current at the time, $t$, after switching off the supply voltage. The traces recorded by memory-scope were reproduced as shown in Figs 4(a-d). The sample $S_{2c}$ showed a slow relaxation.

Nakagawa,$^4$ Rout and Choudhary$^5,6$ also observed this type of slow relaxation in pure cyanobiphenyls. We found same type of relaxation in another mixture$^9$.

When $2V (< V_{th})$ was applied, charge injection from the electrode started and spatial difference in charge distribution appeared. As a result, charge started to diffuse into the sample. After a considerable time ($>4$ min), the power supply was switched off, then relaxation of diffused charge started. If the diffusion process takes place the current $I(t)$ would follow an empirical relation for large values of $t$: 

$$I(t) \propto (Dt)^{1/2}$$

where $D$ is diffusion constant.$^5,6$ Figs 4(a-d) were drawn between $I(t)$ and $t^{1/2}$ to examine the applicability and validity of the said relation. It was observed that the relation holds good for $t \geq 1.5$ sec for the thick cells and $t \geq 1$ sec for thin cells. For $t < 1.5$ sec for the thick cells and $t < 1$ sec for thin cells, the above relation does not hold good. Initially, dielectric relaxation process may be active where the flow of current decreases as.
Fig. 4 — The transient current for S$_2$C at different temperatures for cell thickness (a) 50 μm, (b) 75 μm, (c) 17 μm, and (d) 23 μm.

Fig. 5 — (a) Plots of $I(t)$ against $t^{1/2}$ for S$_2$C for cell thickness 17 μm. (b) Plots of $I(t)$ against $t^{1/2}$ for S$_2$C for cell thickness 23 μm.
\[ I(t) = \exp\left(-t / \tau\right) \]  

(2)

\( \tau \) is the dielectric relaxation time. But \( \tau \) is very small i.e., relaxation is very fast. Just after switching off the power supply, dielectric relaxation process might dominate the diffusion process and then deviation from the diffusion relation was observed. The relation given in Eq. (1), an approximate relation taken for large \( t \), may not be suitable below the above limit (i.e., for \( t < 1 \) sec) which may be another reason for such deviation. In the smectic phase, diffusion of charges was small and temperature dependent. In the nematic phase, the diffusion was faster compared to that of the smectic phase. Rise in temperature causes a slight increase in diffusion of charge in the nematic and isotropic phases. Smectic to nematic transition shows an increase in diffusion rate but no significant change was observed in the case of nematic to isotropic transition. In the smectic phase, deviation from the above relation was high in the thick cells. For thin cells, relation for relaxation of diffused charges holds good for \( t \geq 1 \). The higher electric fields help charges to diffuse into the liquid with higher amount. On the removal of the electric field, the contribution of charge diffusion to the current \( I(t) \) was high, and hence it started to dominate over dielectric relaxation within 1 sec. In the smectic phase, charge diffusion was small and a large deviation from the above diffusion relation was found. The electrostatic field between the smectic layers and the slow molecular velocity may be the reasons of such a low diffusion in the smectic phase, particularly in the thick cells.

(iii) Current - Temperature characteristics — Figs 6(a) - (d) show the \( I - T \) characteristics at different applied voltage. A sharp increase in electrical conductivity occurred within the temperature range 44-45°C for thin cells (23 and 17 \( \mu \)m), and for thick cells (50 and 75 \( \mu \)m), it was within 44.8-45.5°C. But in the nematic and isotropic phases, rate of increase of conductivity with increasing temperature was low. The sharp change in current was due to SmA-N phase transition. This transition was associated with drastic change in viscosity, elasticity and an easier fluid flow. As a result, convective part of the current becomes dominant. Charge injection from the electrodes plays a major role in EHD instability. Richardson thermionic emission's law was found useful to explain the current-temperature characteristics in the nematic phase of pure cyanobiphenyls. However, in our previous work the Richardson equation was not so useful. Though charge injection was a major cause for EHD instability, current depends mainly on the flow of liquid during the instability. A rise in temperature induces a decrease in order parameter and elastic properties. Hence at higher
Fig. 6 — (a) $I$-$t$ characteristics at different voltages for $S_{2C}$ for cell thickness 17 µm
(b) $I$-$t$ characteristics at different voltages for $S_{2C}$ for cell thickness 23 µm
(c) $I$-$t$ characteristics at different voltages for $S_{2C}$ for cell thickness 50 µm
(d) $I$-$t$ characteristics at different voltages for $S_{2C}$ for cell thickness 75 µm
temperature molecular alignment can be deformed easily and this change plays a major role in fluid flow. This was the reason for increase in total conductivity of the fluid with rise in temperature. For thin cells, an increase in current was observed above 44°C [Figs 6(a and b)] which was lower than that of thick cells. During microscopic observations it was also observed that $T_{SN}$ decreases slightly on the application of the electric field. It is in contradiction with the theoretical relation

$$\Delta T = T_0 \Delta \varepsilon E^2 / 2pq$$

where $\Delta T$ is the change in phase transition temperature, $\Delta \varepsilon$ the permittivity difference in two phases, $E$, electric field, $p$, density of the liquid crystal, $q$, heat of phase transition and $T_0$ is the phase transition temperature. In the thin cells, for the same applied voltage, electric field $E (=V/d$ where $d =$ thickness of the cell) was high. The high electric field induces a higher charge diffusion in the liquid crystal and hence space charge creates a perturbation in the electrostatic field within smectic layers which may be the reason of slight decrease in $T_{SN}$.

4 Conclusion

The current flow in the liquid crystal mixture $S_2$ consists of three terms: (a) ohmic conduction (b) diffusion resulting from the gradient of the charge carrier concentration and (c) convection part which is due to charge transfer with convective flow of liquid. Convective flow starts above a threshold voltage and hence EHD instability appears. Viscosity plays a major role in fluid flow consequently in the electrical conduction through the sample cell.

The liquid crystal mixture $S_2$ shows a planer to homeotropic transition in the nematic phase. Therefore the mixture has positive dielectric anisotropy. It is also found that the threshold voltage, for Freedericksz transition, is slightly temperature dependent in the nematic phase and its value is very high in the smectic phase. As the threshold voltage depends on the splay elastic constant, hence splay elastic constant of the liquid crystal decreases with increasing temperature and in the smectic A phase its value is higher enough than that in the nematic phase.

The threshold voltage of the material for Freedericksz transition is small and it can be used in optical switching devices with a small operating dc voltage in the temperature range 45–46.5°C. It consists of a very small nematic range and it would be a suitable material for infrared to visible image converters. A sharp increase in conductivity occurred at a small temperature range [within 45.5–46°C], the sample $S_2$ can be used as a temperature sensor and in other important applications. The mixture has a small nematic range and a broad smectic range. It consists of positive dielectric anisotropy, and it may be used in surface controlled bistable memory devices, both in nematic and smectic phase.

References

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