www.czasopisma.pan.pl PAN www.journals.pan.pl
Opto-Electronics Review 26 (2018) 35–43

Contents lists available at ScienceDirect

Opto-Electronics Review

journal homepage: http://www.journals.elsevier.com/opto-electronics review

# Tunable dielectric and conductivity properties of two 4-n alkoxy benzoic acid

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ARTICLE INFO

*Article history:*
Received 11 July 2017
Accepted 16 December 2017
Available online 30 January 2018

*Keywords:*
Enthalpy
Dielectric anisotropy
Splay elastic constant
Electrical conductivity
Activation energy

ABSTRACT

We have presented dielectric and conductivity studies of two liquid crystal (LC) compounds- p-octyloxybenzoic acid (8OBA) and p-decyloxybenzoic acid (10OBA). Dielectric permittivity study of those compounds gives the evidence of space charge polarization and ionic conductance in the samples. Dielectric permittivity is found to be the highest for 8OBA than 10OBA. Both compounds found to exhibit positive dielectric anisotropy. Splay elastic constant as a function of temperature has also been investigated. Frequency and temperature dependent electrical conductivity of these two LC compounds have been studied in detail. Activation energy has been estimated from both dc and ac conduction process.



## 1. Introduction

Liquid crystal (LC) materials have been extensively investigated due to their numerous applications in device technology, industry, consumer product and medical science [1]. The presence of dual nature, i.e. combination of order and mobility of these magnificent materials makes more functional in display devices. Uses of LC materials in display device depend on its physical properties such as order parameter, dielectric constant, dielectric anisotropy, birefringence, optical transmittance, elastic constant etc. In an attempt to achieve the knowledge about the changes of phase transition temperatures, mesophase textures and enthalpy with the variation of alkyl side chain length between these two compounds p-octyloxybenzoic acid (8OBA) and p-decyloxybenzoic acid (10OBA), we have performed POM and DSC studies for these compounds. A comparative study has been performed on the dielectric and conductivity properties of these two LC compounds. Many researchers already pointed out to the significance of alkoxybenzoic acid compounds in the LC research world, e.g. the hydrogen-bonding gives a strong contribution for generating the novel LC materials due to a dimerization of either symmetrical dimmers or asymmetrical dimers [2–4] which influences the properties of mesogen molecules [5,6]. Q.X. Chen [7] reported that 4-alkoxybenzoic acids have been studied as tyrosinase inhibitors in the food chemistry. Since alkoxybenzoic acid is well known for generating novel LC materials so it is very essential to have the proper idea about its mesophase textures, transition temperatures, dielectric and conductivity behaviour as a function of frequency and temperature of a pure alkoxybenzoic acid compound. Moreover, the importance of dielectric studies is that if these two alkoxybenzoic acid compounds (8OBA, 10OBA) want to be used in a display device, it is fundamental to have the knowledge about the threshold and operating voltages, switching times, operating frequencies, etc. which can be known from the dielectric studies of these compounds [8]. Further, dielectric studies also provide useful information about the molecular structure, intermolecular forces and also molecular dynamics [9]. Rohit *et al.* observed the thermodynamic and dielectric behaviour of Pure DOBA and irradiated DOBA. They found that values of transverse component of dielectric permittivity of the irradiated material in the nematic phase decreases and in the SmC phase increases as compared to those of the pure material [10]. R. Dhar *et al.* [11] measured temperature dependent dielectric permittivity of the binary mixture of 3β −chloro-5-cholestene and DOBA and observed the mixture exhibits positive dielectric anisotropy. E.I. Efremova *et al.* [12] measured dielectric permittivity of the compound 8OBA and reported that the compound exhibits positive dielectric anisotropy in the nematic phase.

## 2. Experimental

The liquid crystal compounds p-octyloxybenzoic acid (8OBA), p-decyloxybenzoic(10OBA) acid have been procured from the Frinton laboratories of USA. The chemical structure and the phase transition temperature quoted from Frinton laboratories of these compounds are shown in Table 1. In order to obtain mesophase textures we have used Optical Polarizing Microscope (POM) (Leica model DM

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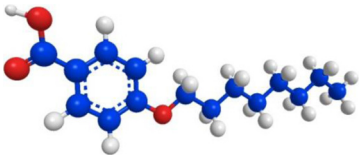
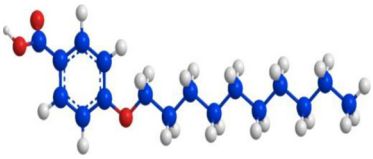
https://doi.org/10.1016/j.opelre.2017.12.002
1230-3402/

**Table 1**
Chemical structures and estimated phase transition temperatures of two LC compounds.

| Compound Name | Chemical structure | Transition temperature(K) |
|---|---|---|
| p-Octyloxybenzoic acid (8OBA) |  | 375($S_c$), 380(N), 420 (I) |
| p-decyloxybenzoic acid (10OBA) |  | 370 ($S_c$), 395(N), 415(I). |

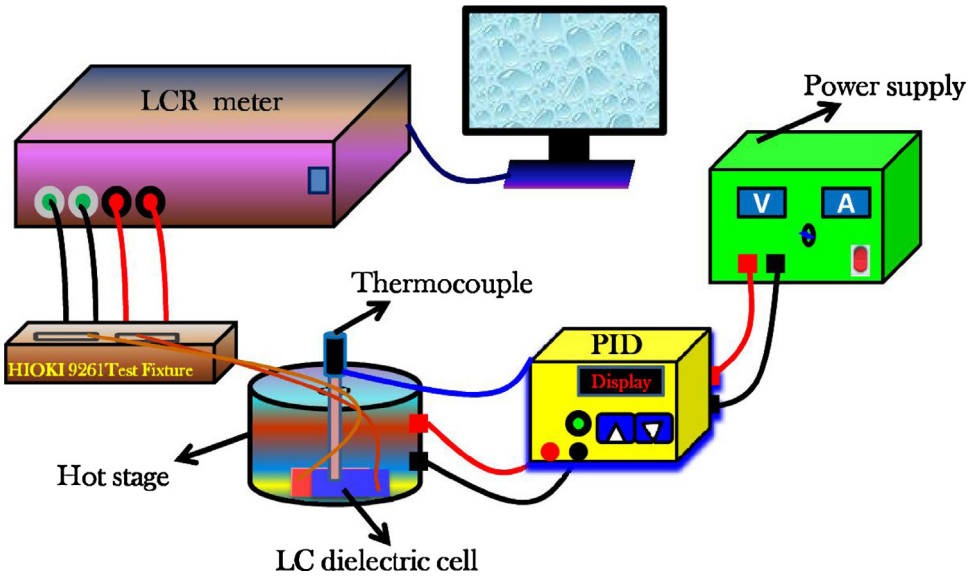



**Fig. 1.** Schematic diagram of experimental set up for dielectric study.

2500 P) having a hot stage (LTS420E). The observation was done under crossed polarization condition at 20× magnification. All the textures of mesophases have been taken during cooling of the sample and the temperature of the sample was controlled by 1 °C/min. The phase transition temperatures of these two compounds have also been measured by Differential Scanning Calorimetry (DSC) [model: Perkin Elmer, DSC-4000]. In a DSC study the temperature of the LC material was controlled at the rate of 5 °C/min. Enthalpy changes have also been carried out during the heating cycle of the sample. The dielectric permittivities of these two compounds have been measured by using a digital LCR bridge (HIOKI 3532-50 LCR HiTESTER). The schematic diagram of experimental set up for dielectric study is shown in Fig. 1. In order to study dielectric permittivities as a function of temperatures, the LC cell was put in a hot stage whose temperature was controlled by 1 °C/min. Before putting the LC cell inside the hot stage, the cell was filled with an LC sample in the isotropic state by capillary action and, then allowed to cool very slowly in the first thermal cycle in order to get best possible alignment. The LC cells used in the dielectric studies were procured by Intec. In,USA. Two types of cell for a homogenous and homeotropic alignment of an LC molecule have been used. These cells are coated with a polyimide (PI) alignment layer. ITO resistance of the empty cell is of 25 ohm. Cut off frequency of the empty cell is of ~112 kHz to 115 kHz. The thickness of the cell is of ~9 μm and dimension of the cell is of 10 × 10 mm.

## 3. Results and discussion

### *3.1. Optical mesophase textures and phase transition temperatures*

The optical mesophase textures of these two thermotropic mesogen 8OBA and 10OBA are shown in Figs. 2 and 3, respectively. Both the compounds are found to exhibit smectic-C ($S_c$) and Nematic (N) meshophase. The compound 8OBA is found to exhibit thread like nematic and schlieren smectic-C texture which is shown in Figs. 2(b) and 2(c), respectively. The isotropic to nematic transition texture and crystalline solid texture for the compound 8OBA are depicted in Figs. 2(a) and 2(d), respectively. The compound 10OBA is also found to display nematic schlieren and schlieren smectic-C as shown in Figs. 3(b) and 3(c) respectively. Whereas, Figs. 3(a) and 3(d) represent the isotropic to nematic transition texture and crystalline solid texture for the compound 10OBA, respectively. In order to get phase transition temperatures more accurately DSC studies have been performed for these two compounds. DSC thermographs for 8OBA and 10OBA have been shown in Figs. 4(a) and 4(b), respectively. Due to the increase of the alkyl chain length at the flexible side chain, the nematic to isotropic transition temperature of the compound 10OBA is reduced. The transition of enthalpies between one state to another state related to the degree of internal order are present in the system. From the comparative studies of DSC thermographs of 8OBA and 10OBA, it

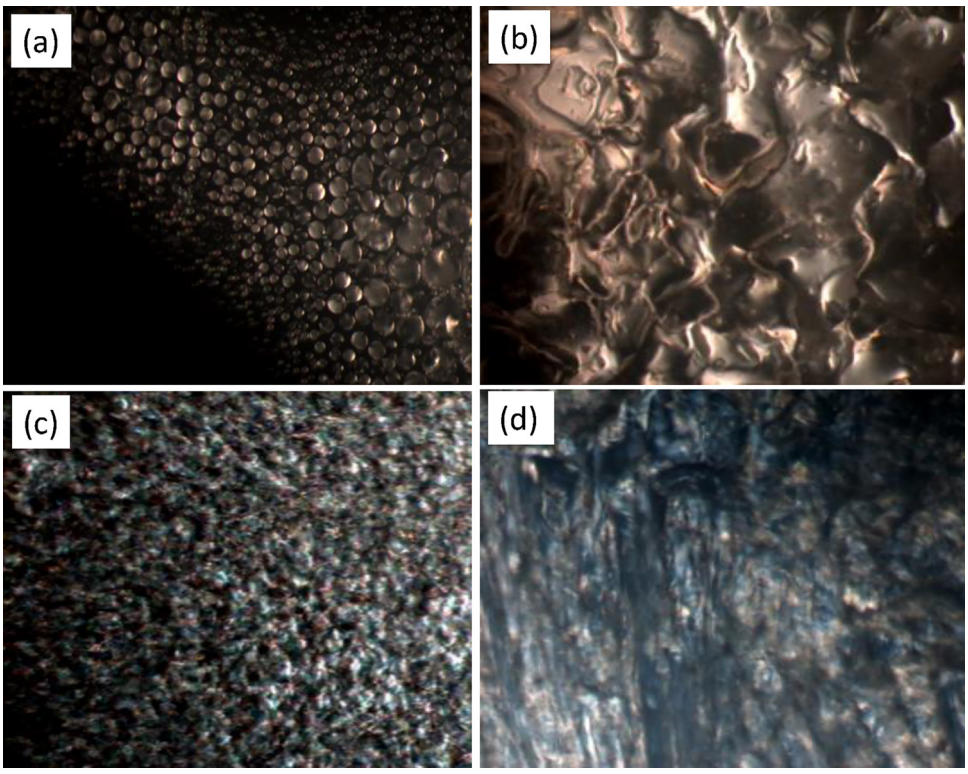



**Fig. 2.** Optical mesophase textures of the compound 8OBA.

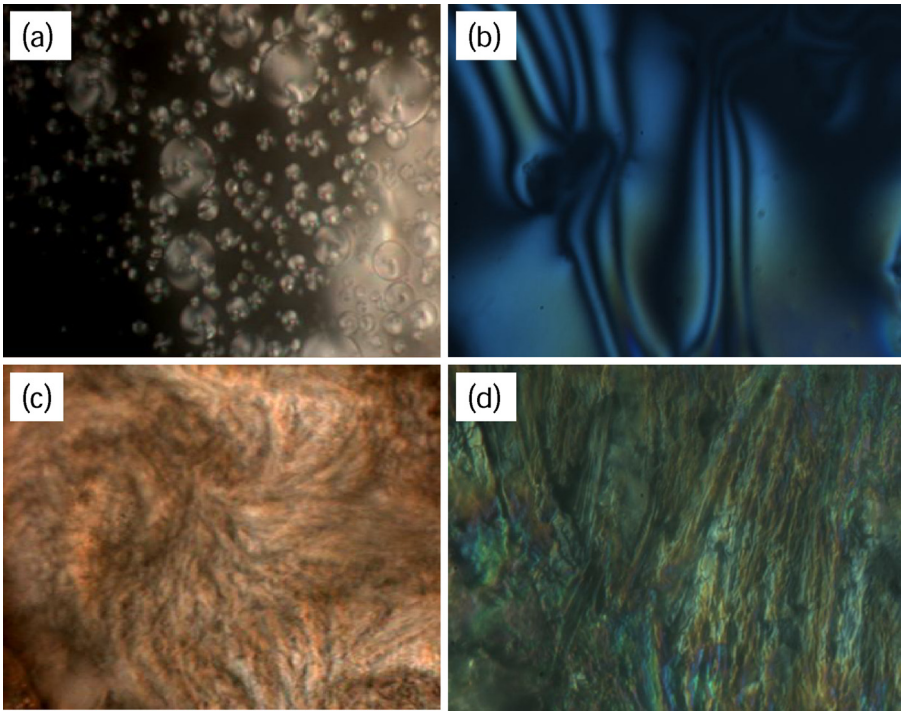



**Fig. 3.** Optical mesophase textures of the compound 10OBA.

**Table 2**
Phase transition temperatures and enthalpy changes (during heating) for the mesogens 8OBA and 10OBA.

| Compounds name | Phase Transition | Temperature (K) | Enthalpy (DH) in kJmol$^{-1}$ |
|---|---|---|---|
| 8OBA | Crystalline solid – Smectic C | 368.84 | 17.6733 |
| | Smectic C – Nematic | 384.13 | 9.3623 |
| | Nematic – Isotropic | 416.65 | 0.5257 |
| 10OBA | Crystalline solid – Smectic C | 370.12 | 5.3451 |
| | Smectic C – Nematic | 395.61 | 0.1736 |
| | Nematic – Isotropic | 409.94 | 0.3062 |

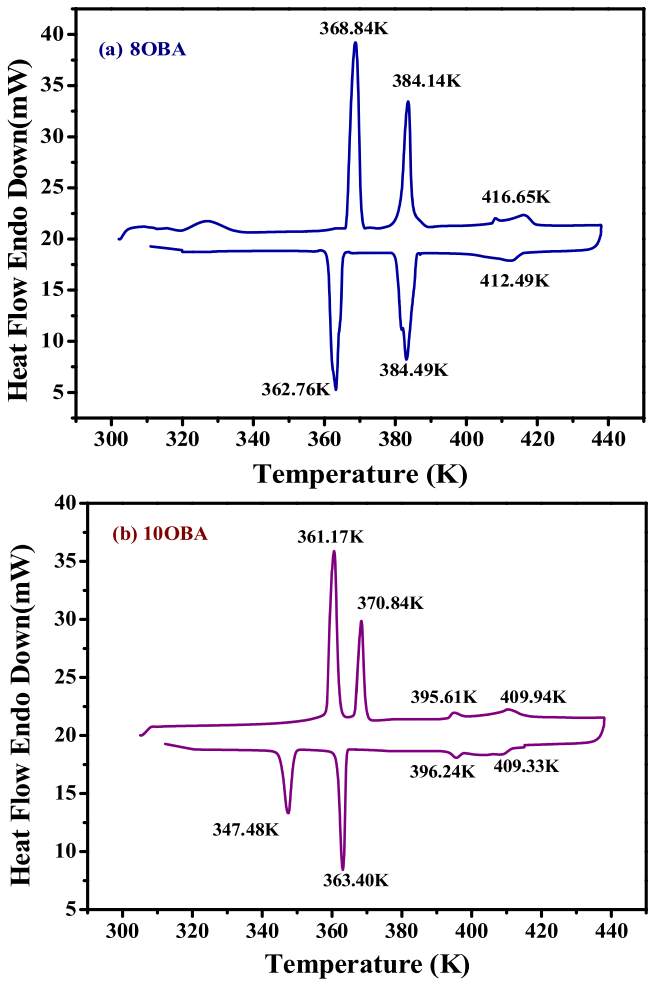



**Fig. 4.** DSC thermographs for the LC materials (a) 8OBA and (b) 10OBA.

has been observed that within the transition between crystalline solid to smectic-C state and smectic-C to nematic state the compound 8OBA is associated with high enthalpies than that of 10OBA. It indicates that the molecules of 8OBA are comparatively more rigid than the molecules of 10OBA. For both the compounds the enthalpy values are larger for solid crystalline to smectic-C than smectic-C to nematic transition. The reason is that, in the case of solid crystalline to smectic-C transition, LC molecules change their positional and orientational order which requires a large amount of energy, whereas during smectic-C to nematic transition molecules maintain the orientational order but a small disruption has been occurred in the positional order. The enthalpy values for different mesophase transitions of these two compounds are shown in Table 2. In case of 10OBA before crystalline solid to smectic- C transition, a transition from crystal to crystal was observed in the DSC thermographs.

### *3.2. Dielectric permittivities as a function of frequency and temperature*

The dielectric studies of LC materials have acquired great practical attention since the realization of technical applications of the electro-optical effects [13]. Moreover, information about the mobile charge carriers and the dielectric relaxation can be achieved from low frequency dielectric study [14]. Fig. 5a and c show the behaviour of transverse component of dielectric permittivity ($\varepsilon_\perp$) with frequency of the compounds 8OBA and 10OBA, respectively. From the low frequency studies of transverse component of dielectric permittivity ($\varepsilon_\perp$), it has been revealed that values of $\varepsilon_\perp$ are high at low frequency which may indicates the presence of space charge polarization effect [15]. Further, dielectric loss factor (tanδ) *vs.* frequency curves for both the compounds) [Figs. 5(b) and 5(d)] are found to exhibit relaxation peak at high tanδ value (tanδ > 1) region. This is in turn that at low frequency $\varepsilon_\perp$ may also be effected by the ionic conductance. Some authors [16] also observed the presence of ionic conductance at the low frequency part. In dynamically scattering nematic and smectic display devices ionic conductance has a great importance. But the movement of ions gives a leakage current and should be minimized to achieve the best performance of the display [16]. It has also found that the value of $\varepsilon_\perp$ is decreasing gradually with increasing frequency revealing Maxwell-Wagner interfacial polarization which shows close agreement with Koop's phenomenological theory [17]. Whereas lower values $\varepsilon_\perp$ at the high frequency signifies the lowest electrical loss characteristics. At low frequency region, values of $\varepsilon_\perp$ are highly effected by temperature and found to increase with increase in temperature. The peak of tanδ *vs.* frequency curves for both the compounds are found to shift with increase in temperature. This is the evidence of the presence of temperature dependent relaxation process in both compounds. In an attempt to understand the behaviour of transverse and longitudinal component of dielectric permittivity with temperature, we have been measured the dielectric permitivities ($\varepsilon_\perp$ and $\varepsilon_{||}$) as a function of temperature at frequency 100 kHz for both the compounds. We have chosen the frequency 100 kHz because it has been expected that the values of dielectric data at100 kHz are not influenced by low-frequency ionic effects, as well as high-frequency effects due to the ITO resistance [18]. Moreover, studies of temperature dependent dielectric permittivity give an idea about the dipole behaviour of the LC material. The temperature dependence of dielectric permitivities (at 100 kHz) for the compounds 8OBA and 10OBA are depicted in Figs. 6(a) and 6(c), respectively. The perpendicular component of dielectric permittivity ($\varepsilon_\perp$) has been measured from the homogeneously aligned LC samples, whereas the parallel component of dielectric permittivity ($\varepsilon_{||}$) obtained from homeotropically aligned sample. It has been observed that both the component of dielectric permittivity ($\varepsilon_{||}$ and $\varepsilon_\perp$) for the compound 8OBA increase with increase in temperature at the smectic phase but they also found to decrease with increasing temperature at the nematic phase. In case of 10OBA the values of $\varepsilon_{||}$ and $\varepsilon_\perp$ are found to increase with increase in temperatures at the nematic phase but near nematic to isotropic transition temperature both the components start to decrease with increase in temperature. The reason is that some molecules of 4-(n-alkyloxy) benzoic acid (nOBA) compounds consists dimerized form of the molecules by hydrogen bonding and they may consider as a mixture of monomers and dimers. With the variation of temperatures one of these two molecular forms dominates and leads to changes of dipole alignment angle which may vary the values of dielectric permittivity [10]. To use these compounds in any display devices it is very important to have an idea about their dielectric anisotropy ranges. It was found that both the compounds 8OBA and 10OBA exhibit positive dielectric anisotropy ($\Delta\varepsilon = (\varepsilon_{||} - \varepsilon_\perp) \geq 0$) as shown in Figs. 6(b) and 6(d), respectively. For both the compounds the values of $\Delta\varepsilon$ are also found to decrease with increaseing temperature. Liquid crystals with positive dielectric anisotropy have several applications. For instance, in twisted nematic display positive dielectric anisotropy is required. Further a positive dielectric anisotropy is essential so that electrical reorientation can be accomplished [19].

### *3.3. Temperature dependent splay elastic constant*

Elastic constants in liquid crystals are very remarkable and have an immense practical significance. In order to manipulate the direction of the director in display devices, the concept of elas-

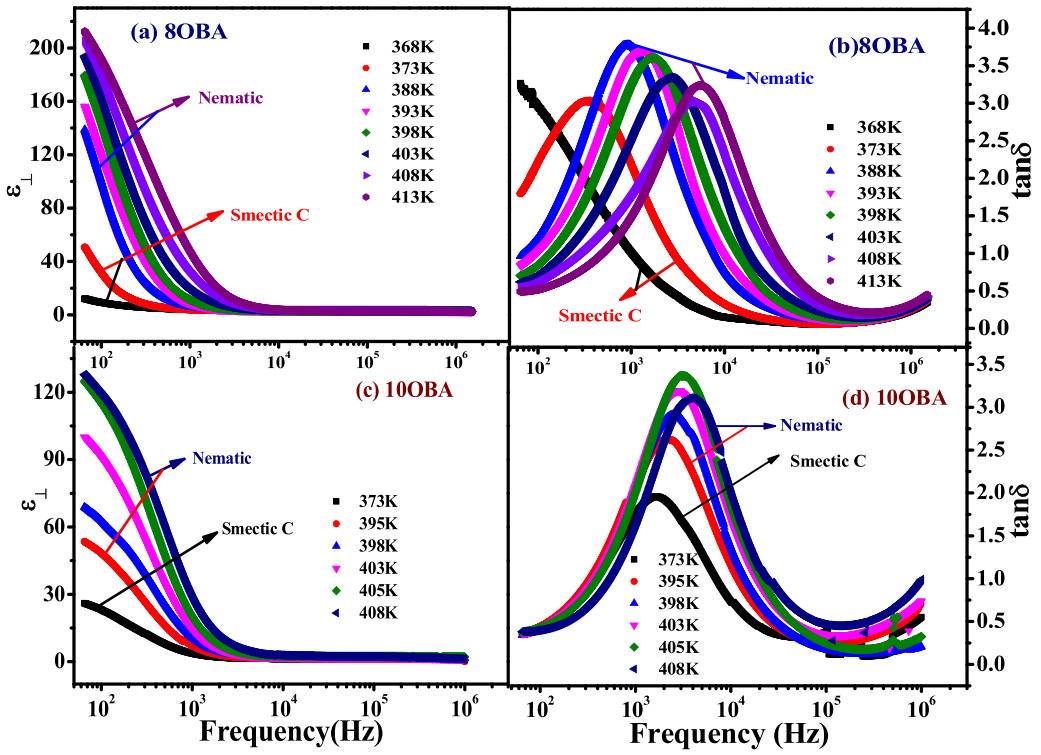



**Fig. 5.** (a) and (c) represents frequency dependent dielectric permittivities ($\varepsilon_\perp$) at different temperature obtain from homogenously aligned cell for the compound 8OBA and 10OBA respectively. (b) and (d) represents dielectric loss factor (tanδ) for the compound 8OBA and 10OBA, respectively.

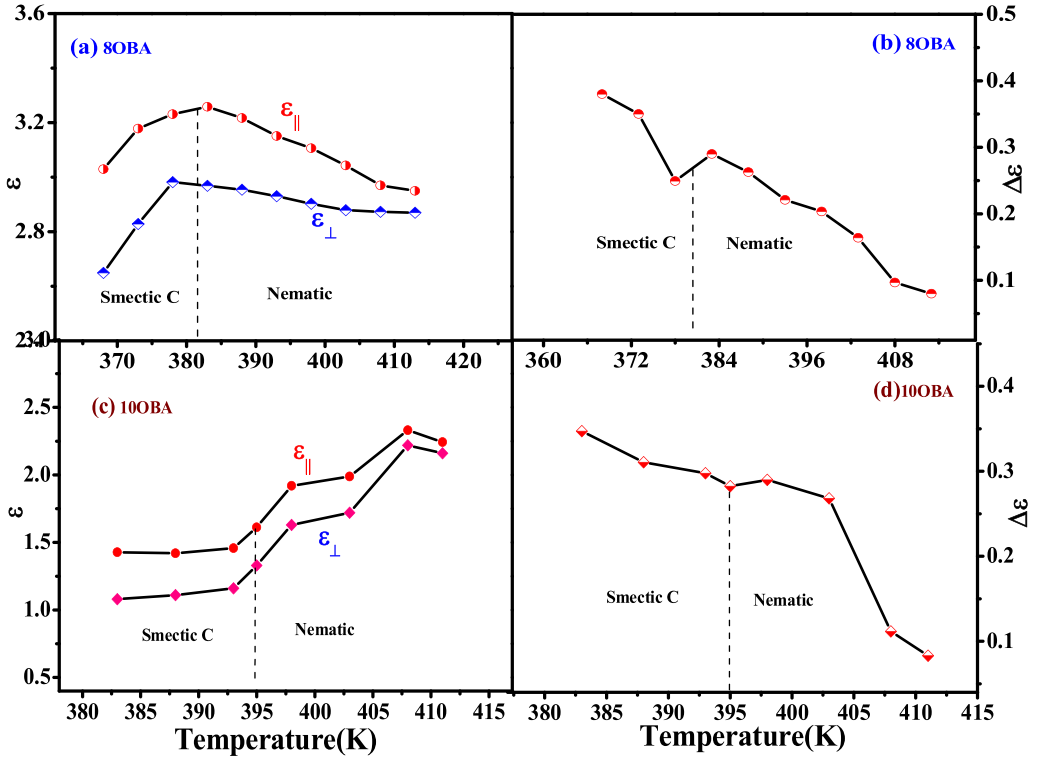



**Fig. 6.** Behaviour of dielectric permittivities ($\varepsilon_\parallel$ and $\varepsilon_\perp$) with temperature of the LC compounds (a) 8OBA (c) 10OBA and dielectric anisotropy($\Delta\varepsilon$) as a function of temperature (b) 8OBA and (d) 10OBA.

tic constants in applied LC medium is extremely important [20]. Usually three basic types of elastic constants: (1) splay ($K_{11}$) (2) twist ($K_{22}$) and (3) bend ($K_{33}$) are used to describe the deformation occurred in liquid crystals [21]. In case of splay of the material, the bending of molecules occurs perpendicular to the director. For the compounds 8OBA and 10OBA, we have attempted to examine only the behaviour of splay elastic constant ($K_{11}$) with temperature. In order to switch, specially the nematic display devices the param-

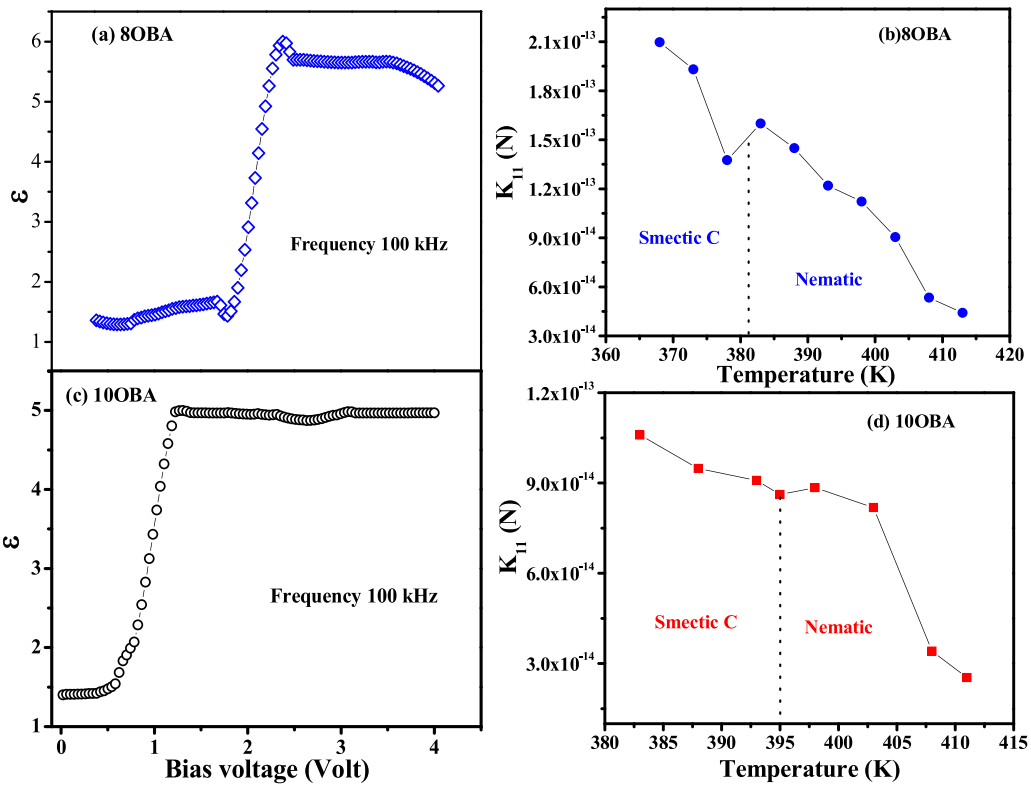



**Fig. 7.** Dielectric permittivity as a function of bias voltage at 100 kHz (a) for 8OBA and (c) for 10OBA and also variation of Splay elastic constant ($K_{11}$) with temperature (b) for 8OBA and (d) for 10OBA.

eter $K_{11}$ plays a vital role and it strongly depends on the external field. The values of $K_{11}$ can be evaluated by using the following equation derived from Freedericks transition [22]:

$$V_{th} = \pi\sqrt{\frac{K_{11}}{\varepsilon_0 \Delta\varepsilon}} \tag{1}$$

where, $\varepsilon_0$ is the permittivity of free space ($\varepsilon_0 = 8.85 \times 10^{-12}\ Fm^{-1}$). $V_{th}$ is the threshold voltage and it can be determined from the behaviour of dielectric permittivity with bias voltage across the cell [23] as shown in Figs. 7(a) and 7(c), respectively. The values of $V_{th}$ for the compounds 8OBA and 10OBA are 0.784 V and 0.583 V, respectively. Figs. 7(b) and 7(d) represent the behaviour of splay elastic constant ($K_{11}$) with temperature for the compounds 8OBA and 10OBA respectively. It has been found that parameter $K_{11}$ exhibits similar behaviour as that of $\Delta\varepsilon$. Equation (1) also indicates that $K_{11}$ is directly proportional to the $\Delta\varepsilon$ of the materials which is also reflected in our experimental results.

### 3.4. Conductivity Study

The acquaintance of electrical conductivity is essential to demonstrate the switching behaviour of liquid crystal films under the action of electric and magnetic fields. To get the awareness about the electrical conductivity of these two compounds we also performed frequency and temperature dependent conductivity studies. Figs. 8(a) and 8(b) show the frequency dependent AC conductivity ($\sigma_{ac}$) for the compounds 8OBA and 10OBA, respectively. The values $\sigma_{ac}$ have been calculated by using the following relation:

$$\sigma_{ac} = \omega\varepsilon\varepsilon_o \tan\delta \tag{2}$$

where, $\omega$ is the angular frequency, $\varepsilon$ is the dielectric permittivity and $\tan\delta$ is the dielectric loss factor.

It has been found that at the low frequency region, the effects of frequency on $\sigma_{ac}$ are very little (almost frequency independent). This may occurred due to the random diffusion of ionic charge carriers via activated hopping [24]. But at this region the effect of temperature on $\sigma_{ac}$ is more prominent and found to increase with increase in temperatures. The reason is that the increasing temperatures cause to increase the mobility of charge carriers and hence leads to increase the conductivity. This behaviour of the conductivity does not affect the contrast ratio of display significantly, but it may increase the current consumption and also the cut-off frequency. In case of 8OBA, $\sigma_{ac}$ is found to merge for all temperatures at the high frequency region. According to the Jonscher, the origin of the frequency dependence of conductivity lies in the relaxation phenomenon working due to mobile charge carriers. So, the frequency dependent conductivity curves are also analyzed by fitting by Jonscher power law [25–27].

$$\sigma(\omega) = \sigma_{dc} + A\omega^n \tag{3}$$

Where, $\sigma(\omega)$ indicates the total conductivity, $\sigma_{dc}$ is the dc conductivity at a particular temperature, 'A' is the temperature dependent pre-exponential factor and 'n' is the frequency exponent in the range of $0 < n < 1$. Estimated values of A and n from $\sigma_{ac}$ *vs.* frequency curves at different temperatures for the compound 8OBA and 10OBA are given in Table 3.

We have found that the values of 'n' is greater than one for both the compounds which do not obey the Jonscher's prediction. A model has already been developed by some authors pointing that there is no physical argument to restrict the value of n below 1 and they suggest that there does not exist a 'universal fractional power law' [25,28]. Fig. 9 depicts $\ln\sigma_{ac}$ *vs.* $\frac{10^3}{T}$ curves for these two compounds and these curves were also investigated by using Arrhenius relation [29]:

$$\sigma_{ac} = \sigma_0 \exp\left(-w_A/k_\beta T\right) \tag{4}$$

where, $\sigma_0$ is the pre-exponential factor, $w_A$ is the activation energy and $k_\beta$ is the Boltzmann constant.

**Table 3**
Estimated values of A and n for the compounds 8OBA and 10OBA.

| Compound Name | Temperature (K) | A | n |
|---|---|---|---|
| 8OBA | 373 | 4.489E-16 (±2.034E-17) | 1.827 (±3.03E-3) |
| | 378 | 2.722E-16 (±1.131E-17) | 1.858 (±2.70E-3) |
| | 393 | 2.481E-16 (±3.167E-18) | 1.868 (±8.53E-4) |
| | 403 | 2.209E-16 (±3.065E-18) | 1.877 (±9.27E-4) |
| | 413 | 1.701E-16 (±1.901E-18) | 1.887 (±7.46E-4) |
| 10OBA | 388 | 4.589E-14 (±2.452E-15) | 1.475 (±3.30E-3) |
| | 398 | 4.017E-15 (±1.514E-15) | 1.658 (±2.62E-2) |
| | 403 | 2.794E-14 (±9.520E-15) | 1.515 (±2.83E-2) |
| | 408 | 9.406E-16 (±1.29E-16) | 1.707 (±9.50E-3) |

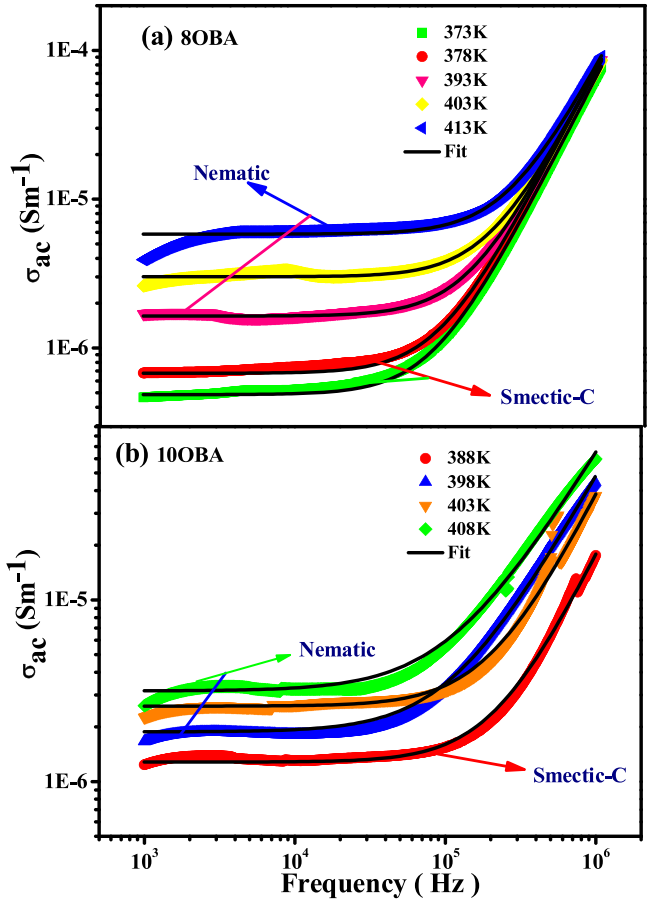



**Fig. 8.** Frequency dependent ac conductivity ($\sigma_{ac}$) at different temperature (a) for the compound 8OBA and (b) for the compound 10OBA.

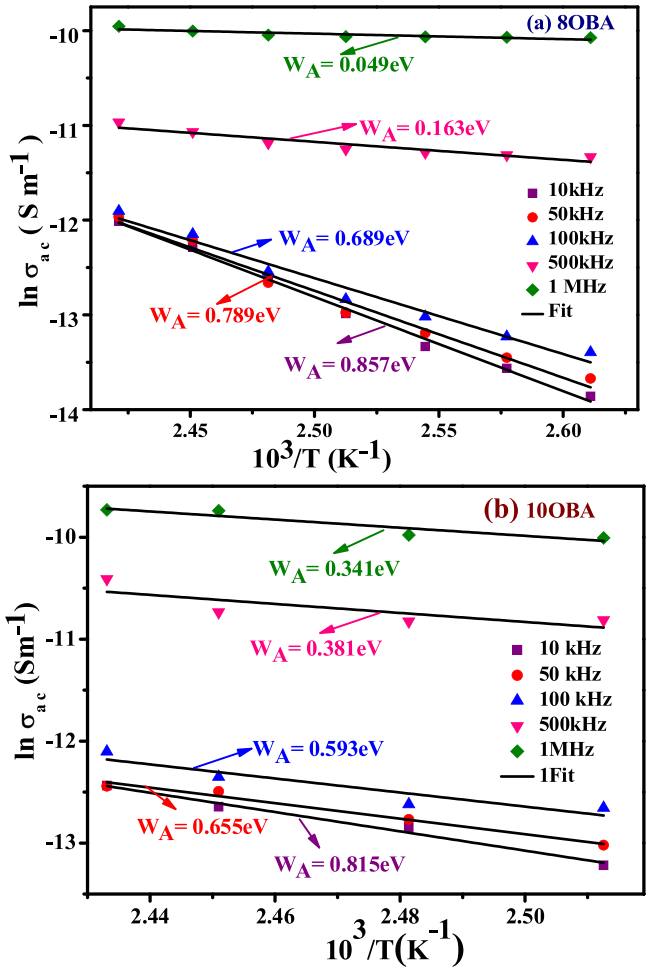



**Fig. 9.** $\ln(\sigma_{ac})$ *vs.* $10^3/T$ with varying frequency of (a) 8OBA and (b) 10OBA LC compounds. The continuous solid line shows the fitted curve.

We have estimated $w_A$ of these two compounds at the nematic phase from the Arrhenius fit of temperature dependent ac conductivity data for the frequency of 10 kHz, 50 kHz, 100 kHz, 500 kHz, and 1 MHz for frequency of 10 kHz, 50 kHz, 100 kHz, 500 kHz, and 1 MHz. The estimated values of $w_A$ are of 0.857 eV (10 kHz) to 0.049 eV (1 MHz) for 8OBA. Similarly, the estimated values of $w_A$ are of 0.8154 eV (10 kHz) to 0.342 eV (1 MHz) for 10OBA. So it is clear that $w_A$ decreases with increase in frequency for both compounds. As the frequency increases, the smaller time window is available to respond the fast changing electric field as a result the hopping-transportation mechanism becomes restricted at the nearest neighbouring defects' sites. For this reason the values of $w_A$ decreases with increase in frequency. By extrapolating, low frequency part of ac conductivity curve to Y-axis for each temperature, we have estimated dc conductivity ($\sigma_{dc}$) for both compounds. In the nematic phase the values of $\sigma_{dc}$ of these two compounds are found to the order of $10^{-7}$–$10^{-6}$ S-m$^{-1}$. From Fig. 10, it has been observed that $\sigma_{dc}$increases with increase in temperature which indicates that the negative temperature co-efficient of the resistance of these two compounds is under investigation. In order to investigate further the dc conduction process, we have fitted $\ln\sigma_{dc}$ *vs.* $\frac{10^3}{T}$ curves using the Arrhenius relation of conductivity.

$$\sigma_{dc} = \sigma_0 \exp\left(\frac{-W_A}{K_B T}\right) \tag{5}$$

where, $\sigma_0$ is the pre-exponential factor. The values of activation energy obtained from Arrhenius fitted of $\ln\sigma_{dc}$ *vs.* $\frac{10^3}{T}$ curves are of

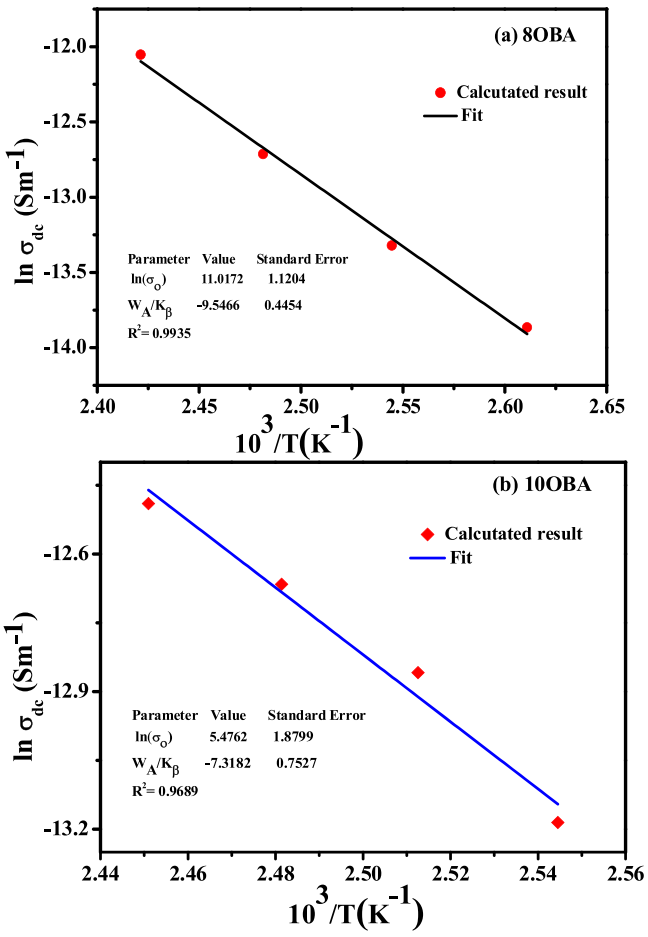



**Fig. 10.** $\ln(\sigma_{dc})$ *vs.* $10^3/T$ (a) 8OBA and (b) 10OBA LC compounds. The continuous solid line shows the fitted curve.

0.779 eV and 0.630 eV for the compound 8OBA and 10OBA, respectively.

## 4. Conclusions

Effect of alkyl chain length on the phase transition temperatures is found to exhibit between these two compounds 8OBA and 10OBA. Due to the increase of the alkyl chain length at the flexible side chain, the nematic to isotropic transition temperature of the compound 10OBA is reduced. At low frequency region, effect of space charge polarization and ionic conductance has been found to exhibit for both the compounds. Presence of ionic conductance in LC compounds has a great impact for using in the dynamically scattering nematic and smectic display devices. From the low frequency studies of dielectric permittivity of these two compounds it has been observed that 8OBA exhibits significantly higher values of dielectric permittivity than 10OBA at the high temperature nematic phase. Both compounds are also found to exhibit positive dielectric anisotropy and the variation of alkyl chain length does not effect much on this property between these two compounds. The threshold voltage, splay elastic constants, activation energy from ac and dc conduction process have been found to effect very slightly by the alkyl side chain length. Dielectric anisotropy and splay elastic constant behaves similarly with temperatures. The activation energy obtained from ac conduction process is found to decrease with increase in frequency. From the temperature dependent dc conductivity studies it was visualized that the values of dc conductivity increases linearly with increase in temperature.

## Acknowledgements

One of the authors, Mrs. Sangita Patari is thankful to Dr.S. K. Mandal and Dr. C. Bera, Assistant professor of NIT Agartala, Department of Physics for their fruitful suggestions and comments.

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