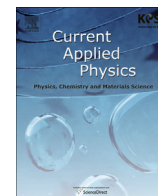




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Anomalous high dielectric strength and low frequency dielectric relaxation of a bent-core liquid crystal with a large kink angle

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ABSTRACT

We investigated the dielectric dispersion property of a bent-core liquid crystal (BLC) with a large kink angle in the frequency range of 1.0 Hz–5.5 MHz in a planar aligned cell. Single dielectric dispersion was observed in the smectic A, nematic, and isotropic phase of the planar aligned sample. The dielectric strengths, relaxation frequencies, distribution parameters, and dc conductivity were measured as a function of temperature. The dielectric strength of the observed relaxation mode was anomalously high (~70), whereas the relaxation frequency was low at ~500 Hz. The relaxation mode observed in the planar aligned cell was attributed to the fluctuations in the polarization direction due to the cooperative motion of the molecules.

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1. Introduction

The bent-core liquid crystal (BLC) has drawn much attention due to its interesting physico-chemical properties [1]. In the 1990s, it was reported that the BLC possesses ferroelectric and an anti-ferroelectric phases [2–6]. The BLC was also found to have biaxial ordering in the nematic (N) phase, which had been regarded as the lost Holy Grail of liquid crystals (LCs) [7–10]. In addition, BLC molecules often form a macroscopic chiral structure from achiral BLC molecules either by polar packing and tilt of the molecules [11,12] or by the structural conformation of the molecules [13–15]. Recently, the BLC attracted scientific interest for its strong flexoelectric properties [16–20]. Thus, the BLC shows a wide variety of phases with different symmetries and new functionalities that are not found in conventional rod-like liquid crystal (RLC) molecules [21–23].

Compared to active studies on the structural and optical studies of BLC molecules, the dielectric property of BLC molecules has been rarely reported [24–30]. Dielectric spectroscopy is an effective method to study the molecular and collective dynamics of molecules [31–33]. Balchandran et al. [24] and Salamon et al. [25]

reported three dielectric relaxation peaks in planar-aligned BLC samples. The relaxation frequency reported by Salamon et al. in the isotropic (I) phase of BLC is in the frequency range of ~3 MHz [25]. Notably, BLC molecules studied in previous literature have a small kink angle θ of approximately 60°. Different kink angles of the BLC often result in different physical properties. For example, the RLC doped with a small amount of BLC with a large θ shows a reduction in the bend elastic constant K_{33} [34,35], while the RLC doped with BLC with a small θ shows a reduction in the splay elastic constant K_{11} [36]. In addition, the BLC with a small θ shows a bistable memory effect via the reorientation of layers similar to the smectic phase of RLC [37].

In this paper, we investigated the dielectric relaxation behavior of the BLC with a large kink angle θ of ~105°. Fig. 1 shows the chemical structure of the BLC molecule, 2,3-naphthalene bis[4-(4-n-dodecyloxybenzoxy)benzoate], used in this study. Hereafter, the BLC would be referred to as B23. The dielectric property of the B23 molecule in the frequency range of 1.0 Hz–5.5 MHz was investigated at the smectic A (SmA), N, and I phase, and the physical origin of the dielectric relaxation mode is discussed in detail.

2. Experimental procedure

The synthesis and X-ray analysis of B23 are described in previous literature [38]. The X-ray diffraction results indicate that the

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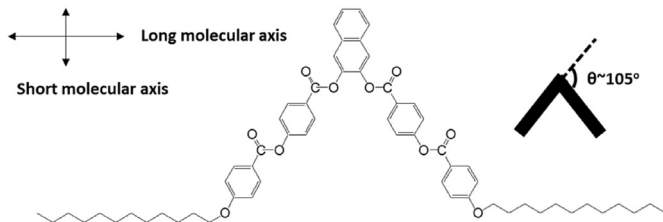


Fig. 1. Chemical structure of the B23 molecule used in this study.

BLC molecule has a trans-conformation, as depicted in Fig. 1. From the layer spacing data of B23, the estimated kink angle θ of B23 was approximately 105° . B23 shows a phase sequence of Crystal (404 K) SmA (485 K) N (492 K) I in the heating cycle, while it exhibits the phase sequence of I (490.5 K) I + N (489 K) N (484.5 K) N + SmA (481 K) SmA (333 K) Crystal in the cooling cycle. The wider SmA phase in the cooling cycle is due to the supercooling effect.

For the dielectric measurements, an indium-tin-oxide (ITO)-deposited glass substrates with a small sheet resistance of $\sim 10 \Omega/\text{cm}^2$ was used. Due to the small sheet resistance of the electrodes, no dielectric loss by the ITO was observed up to a frequency of 5.5 MHz. A commercial planar alignment polyimide (PI) of AL1254 (JSR) was coated on both substrates and rubbed with a cotton cloth. The substrates were assembled in an antiparallel fashion and a $10 \mu\text{m}$ gap was maintained using mylar film.

The dielectric properties of the sample was measured in the frequency range from 1.0 Hz to 5.5 MHz using a LCR meter ZM2376 (NF). The temperature (T) of the sample was controlled by a heating stage TS95 (Linkam) with an accuracy of 0.1°C during the measurements. An electric field of $0.05 \text{ V}/\mu\text{m}$ was applied across the BLC layer to measure the capacitance of the sample. The phase transition temperature was checked by measuring the change of the polarizing optical microscopy (POM) texture and the transmittance (TR) of the sample between crossed polarizers. The dielectric spectra were simultaneously recorded with the POM and TR data with varying T . B23 shows some orientational defects in the heating cycle, while it shows a homogeneous orientation in the cooling cycle. Thus, the dielectric data was taken in the cooling cycle.

3. Results and discussion

Fig. 2 shows the POM textures of the planar-aligned BLC samples at various T in the cooling cycle. The transmission axis of the polarizer is 45° in the rubbing direction of the sample. At 491 K, above the N-I phase transition temperature (T_{NI}), a completely dark state was observed [Fig. 2(a)]. The transmission through the sample started to increase at 490.5 K [Fig. 2(b)] and the sample completely converted into the N phase at 489 K [Fig. 2(d)]. Generally, the N-I phase transition is the first order and the N and I phase domains can coexist. The homogeneous bright texture in Fig. 2(b)–2(c) seems to be related to the surface-induced ordering of the BLC molecules, which is often observed slightly above T_{NI} . The mixed phase (N + SmA) was observed in the temperature range from 484.5 K to 481.5 K, as shown in Fig. 2(e)–2(i). The B23 completely transformed into the SmA phase at 481 K [Fig. 2(j)] and the typical fan-shaped texture of the SmA phase was observed.

Fig. 3 shows the POM textures of the planar-aligned sample in the N and SmA phase when the rubbing direction of the sample was 0° to the transmission axis of the polarizer. The homogeneous dark state in Fig. 3(a) indicates the molecules are uniformly aligned to the rubbing direction in the N phase. In the SmA phase [Fig. 3(b)], the sample shows a dark state except in some places, which is presumably due to the curved smectic layer [39]. The overall

transmission through the cell continues to increase when the temperature falls below 380 K, which could be related to the supercooling effect where the LC material continues to lose its optical anisotropy [Fig. 3(c)].

To analyze the measured dielectric data, the dielectric spectra were fitted to the generalized Cole-Cole equation (Eq. (1)) [31,32]:

$$\varepsilon^* = \varepsilon' - j\varepsilon'' = \varepsilon(\infty) + \sum_{i=1}^2 \frac{(\varepsilon)_i}{1 + (j\frac{f}{f_{ri}})^{1-h_i}} + \frac{A_1}{f^n} - j \frac{\sigma_i}{2\pi\varepsilon_0 f^k} - jA_2 f^m \quad (1)$$

where $i = 1, 2, \dots, k$ is an integer for different relaxation processes, $\varepsilon(\infty)$ is the relative permittivity in the high frequency limit, $\Delta\varepsilon_i$, f_{ri} , and h_i are the dielectric strength, relaxation frequency, and distribution parameter ($0 \leq h_i \leq 1$) of the i th mode, respectively. The third term of Eq. (1) represents the contribution of the electrode polarization at low frequencies, where A_1 and n are fitting parameters. $\sigma_i/2\pi\varepsilon_0 f^k$ accounts for the contribution of the ionic conductivity, with σ_i and k as fitting parameters, usually $k = 1$, and $\varepsilon_0 (=8.85 \text{ pF/m})$ is the free space permittivity. The relaxation frequency, dielectric strength, and distribution parameters were estimated by fitting the experimental data using Eq. (1).

Fig. 4 shows the relative permittivity ε'_\perp and the dielectric loss ε''_\perp of the planar-aligned B23. Single dielectric dispersion in the I [Fig. 4(a)], N [Fig. 4(b)], and SmA phases [Fig. 4(c)] was observed. The black and yellow plots in each figure represent the experimental data of ε'_\perp and ε''_\perp , respectively. The red and magenta lines were the fitted curves according to Eq. (1) and the blue and purple lines were the ε'_\perp and ε''_\perp data after subtracting the low frequency correction terms from the experimental data. The ε'_\perp and ε''_\perp peaks decrease with decreasing T due to the increased ordering and more planar orientation of the B23 molecules.

Fig. 5(a) and (b) show the relaxation frequency f_r and the dielectric strength $\Delta\varepsilon_\perp$ of the B23 sample, respectively. The f_r decreases with decreasing T , while $\Delta\varepsilon_\perp$ is weakly dependent on T . The dielectric relaxation frequency f_r of B23 is very small, $\sim 500 \text{ Hz}$, in the I phase compared to the f_r of $\sim 3 \text{ MHz}$ for the BLC with a small kink angle of $\theta \sim 60^\circ$ in the literature [25]. $\Delta\varepsilon_\perp$ in the SmA phases (up to $\sim 380 \text{ K}$) slowly decreases with decreasing T , whereas $\Delta\varepsilon_\perp$ is nearly independent of T in the N and N + SmA phases, except near the phase transition temperature. As shown in Fig. 5(b), a little discontinuity in the dielectric strength values between the I and N phase could be observed, which may be due to the reason of having almost same origin of relaxation mode in both phases and intrinsic characteristic of the BLC of having bent core structure even in the isotropic phase. The origin of relaxation mode in both the phases would be discussed later in this manuscript. The dielectric experiment on BLC carried out by J. K. Vij's group also could not observe any appreciable discontinuity in the dielectric strength values for the relaxation mode of high dielectric strength [30].

The dielectric strength of the B23 molecule is anomalously high, at $\Delta\varepsilon \sim 70$. Below $T = 380 \text{ K}$, $\Delta\varepsilon_\perp$ of the BLC continues to decrease with decreasing T . The temperature dependence of $\Delta\varepsilon_\perp$ below 380 K seems to be due to a supercooling effect, where the LC material keeps on losing its optical and dielectric anisotropy. Fig. 5(c) shows the Arrhenius plot for f_r vs. T^{-1} . A slight discontinuity between the different phases was observed in Fig. 5(a)–5(c). The activation energy of the dielectric relaxation for all phases was estimated by

$$f = f_0 \exp(E_a/K_B T), \quad (2)$$

where f_0 is a constant parameter, $k_B = 1.38 \times 10^{-23} \text{ J/K}$ is the Boltzmann constant, and E_a is the activation energy. The activation energies in the I, N, N + SmA, and SmA phases were 0.13, 0.47, 0.58,

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