

## Phase Transition and Thermodynamic Studies on 6O.CB and 10O.CB Liquid Crystalline Compounds

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### Abstract

In the present exploration, the hexyloxy and decyloxy Cyano biphenyl, liquid crystal compounds are used. Polarizing Optical Microscope is used to record the phase transition temperatures and optical textures. A specially designed Pyknometer is hired for density measurements in the liquid crystalline phase. The Dilatometric data reveals that 6O.CB compound exhibits nematic and 10O.CB compound exhibits only Smectic A phase. The temperature dependence of Thermodynamic parameters were evaluated. The results show that all the thermodynamic parameters exhibit distinctive variations in the neighborhood of phase transition. The outcomes are equated with literature data accessible on number of compounds.

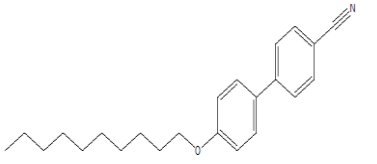
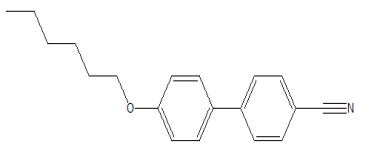
**Keywords:** Liquid crystals, Optical textures, Phase transition, Density jump, Thermal expansion co-efficient and Thermodynamic parameters.

### 1. Introduction

The liquid crystalline compounds are the materials with the properties in between that of an anisotropic crystalline solid and isotropic liquid<sup>1-2</sup>. These materials behave as a solid crystal by showing anisotropy in its mechanical, optical and electrical properties, however they are unable to support the shearing, hence they flow like normal liquids<sup>3</sup>. The transformation from one phase to another phase is known as phase transition associated with abrupt changes and discontinuities. These compounds exhibit polymorphism and the transformation occurs with temperature variation due to the disruption of the molecular

regularity. The type of phase transformation and the occurrence of pre transitional effects in liquid crystalline compounds are better understood with respect to density variations at different temperatures in the liquid crystalline phases.

In the present study, the variation of density at different temperature in liquid crystalline phases is analyzed. The respective phase variants are observed in the neighborhood of liquid crystalline phases and the number of thermodynamic parameters is estimated. These parameters are useful in the study of anharmonicity, internal structure, molecular order, and play a vital role in the equation of state calculations. These parameters also assist in the study of mechanism of ultrasonic absorption in liquids, establishing the coordination with the phonon-phonon interaction absorption<sup>4</sup>. The molecular structures of the compounds used in the present investigations are as shown.

4-Cyano-4'-decyloxybiphenyl 10O.CB	
4-Cyano-4'-hexyloxybiphenyl 6O.CB	



## 2. Experimental

The density of liquid Crystals were measured using dilatometer, which has specially assembled Pycnometer having capillaries of diameter about 250  $\mu\text{m}$  and 6-8 cm length placed on a glass tube which is U shaped. Standardization of the pycnometer is done by measuring molar volume of water. The liquid crystalline material is filled in the pycnometer and is kept in the heating block maintained at temperature around 5°C above the transition temperature, and then the sample is cooled slowly till the level of the sample attains the mark on the capillaries. The extra sample found in the capillary cups is gently taken out with the help of a syringe. A charge coupled device camera will be fixed to the telescope and the sample levels in the capillary are seen on a monitor.

The density of mesogenic material is found to decrease with the rise in temperature, it exhibits a steep increase in the neighborhood of phase transitions. The density jump ( $\Delta\rho/\rho$ ) is estimated as the vertical distance between the density values  $\rho_1$  and  $\rho_2$  which is got by

linear extrapolation from both sides of the transition. The recorded density jump, coefficient of thermal expansion and density slopes around different phase transitions are represented in Table II.

**Table I** Phase transition temperature and Optical textures obtained by Polarizing Optical Microscope

Compound	Transition temperature in ( $^{\circ}\text{C}$ )				Optical Textures
	I-N	N-Cr	I-SmA	SmA-Cr	
100.CB	--	--	84.5	59.93	
60.CB	75.9	58.76	--	--	

**Table II**, The Density Jump, Thermal Coefficient and density slope in different phases of liquid crystals.

Compound	Phase Variant	% of $(\Delta\rho/\rho)$	$\alpha_{\text{I-N}} \times 10^{-3}$ ( $^{\circ}\text{C}^{-1}$ )	$\alpha_{\text{N-Cr}} \times 10^{-3}$ ( $^{\circ}\text{C}^{-1}$ )	$\alpha_{\text{I-SmA}} \times 10^{-3}$ ( $^{\circ}\text{C}^{-1}$ )	$\alpha_{\text{SmA-Cr}} \times 10^{-3}$ ( $^{\circ}\text{C}^{-1}$ )	$(\text{d}\rho/\text{d}T)_{\text{I}} \times 10^{-4}$ ( $^{\circ}\text{C}^{-1}$ )	$(\text{d}\rho/\text{d}T)_{\text{N}} \times 10^{-4}$ ( $^{\circ}\text{C}^{-1}$ )	$(\text{d}\rho/\text{d}T)_{\text{SmA}} \times 10^{-4}$ ( $^{\circ}\text{C}^{-1}$ )	$(\text{d}\rho/\text{d}T)_{\text{N-Cr}} \times 10^{-4}$ ( $^{\circ}\text{C}^{-1}$ )	$(\text{d}\rho/\text{d}T)_{\text{SmA-Cr}} \times 10^{-4}$ ( $^{\circ}\text{C}^{-1}$ )
100.CB	I-SmA	0.3056	--	--	3.0	--	3.4	--	3.6	--	--
	SmA-Cr	0.1411	--	--	--	1.4	--	--	--	--	3.8
60.CB	I-N	0.2846	2.8	--	--	--	2.2	--	--	--	--
	N-C	0.1818	--	1.8	--	--	--	2.4	--	2.8	--

### 3. Theory

The theory for calculation of different thermodynamic parameters from the coefficient of thermal expansion ( $\alpha$ ) is given by several authors<sup>5-6</sup>. Thermodynamic parameters play a pivotal role in understanding the internal structure, molecular order and anharmonicity. They play a major role in the equation-of-state calculations, in understanding the mechanism of ultrasonic absorption in liquids and developing the relation with the outcomes of phonon-phonon interaction absorption. In the present study the temperature dependence of density is analyzed, which throws light on the type of phase transformation

and the development of the effects of pre transition. A good number of approaches were put forth to evaluate thermo acoustical parameters of solids, liquids, polymers and liquid crystals. From the density data and coefficient of thermal expansion many thermo dynamical parameters viz., Moelwyn-Hughes parameter ( $C_1$ ), the reduced molar volume ( $\tilde{V}$ ), the isochoric temperature coefficient of internal pressure ( $X$ ), the Sharma parameter ( $S_0$ ), Huggins parameter ( $F$ ), isothermal microscopic Gruneisen parameter ( $\Gamma$ ), Fractional free volume ( $f$ ), Thermal parameter ( $A^*$ ), Gruneisen parameter ( $\Gamma_p$ ), isothermal, isochoric and isobaric Gruneisen parameters ( $\Gamma_{ith}$ ,  $\Gamma_{ich}$ ,  $\Gamma_{iba}$ ), Beyer's nonlinearity parameter ( $B/A$ ) are evaluated for the homologous series of nO.CB liquid crystalline compounds with  $n=6$  and  $10$ .

The distinct expressions of thermodynamic parameters using the thermal expansion coefficient are as below.

**Moelwyn-Hughes parameter ( $C_1$ ):** This parameter serves as an important elemental parameter to determine the thermo acoustical parameters in the molten metals, liquids, liquefied gases, liquid crystals and polymers. The expression for  $C_1$  and reduced molar volume ( $\tilde{V}$ )<sup>7-9</sup> in terms of  $\alpha$  is

$$C_1 = \frac{13}{3} + (\alpha T)^{-1} + \frac{4}{3}(\alpha T)$$

$$(\tilde{V}) = \left[ 1 + \frac{\alpha T}{3(1+\alpha T)} \right]^3$$

The isochoric temperature coefficient of internal pressure ( $X$ )<sup>10</sup>.

$$X = - \frac{2(1 + 2\alpha T)}{\tilde{V} C_1}$$

The Sharma parameter ( $S_0$ ) is estimated by the following equation<sup>11-12</sup>.

$$S_0 = \frac{-X}{2} (3 + 4\alpha T)$$

**Huggins parameter (F):** The Sharma parameter and Huggins parameter (F) are interrelated for liquid crystals the Huggins parameter is given by

$$F = 2 \left[ 1 + \left( \frac{S_0}{3 + 4\alpha T} \right) \right] - \left( \frac{3 + 4\alpha T}{3} \right)$$

**The isothermal microscopic Gruneisen parameter ( $\Gamma$ ):** Measures the dependence of the volume of harmonicity with respect to normal mode frequency  $\nu$ . This parameter depends on Huggins parameter (F) and Thermal expansion coefficient  $\alpha$

$$\Gamma = \left( \frac{2}{3} \right) \alpha T + \left( \frac{2 + F + 4\alpha T}{2\alpha T} \right)$$

**The Fractional free volume (f):** In liquid crystallinity as the mobility of molecules increases, the fractional free volume measures the disorderness, which is expressed as

$$f = \frac{1}{(\Gamma + 1)}$$

**Thermal parameter ( $A^*$ ):** The thermal parameter signifies the tendency of the liquid crystal to attain the ordered structure at low temperature and will be equal to unity

$$A^* = 1 + \frac{f}{\Gamma}$$

**Gruneisen parameter ( $\Gamma_p$ ):** The Gruneisen parameter ( $\Gamma_p$ ) for mesogenic compounds is

$$\Gamma_p = \left( \frac{2}{3} \right) \alpha T + \left( \frac{1}{2\alpha T} \right) + 2$$

**Isothermal, isochoric and isobaric Gruneisen parameters ( $\Gamma_{ith}$ ,  $\Gamma_{ich}$ ,  $\Gamma_{iba}$ ):** are similar to the respective acoustical parameters.

$$\Gamma_{ich} = \Gamma_{ith} + \Gamma_{iba}$$

**The isochoric Gruneisen parameters  $\Gamma_{ich}$**  could be evaluated using the following equation

$$\Gamma_{ich} = -\frac{E - F}{F}$$

Where,

$$E = -[2 + (\alpha T)^{-1}][2\alpha(\bar{V})^{C_1-1}] \text{ and } F = 2\alpha$$

The isothermal Anderson-Gruneisen parameter  $\delta$  is very much required in the analysis of variation of bulk modulus with respect to temperature in solids.

$$\delta = 2\Gamma_{iba}$$

The Anderson-Gruneisen parameter is different from Moelwyn-Hughes parameter with the introduction of the parameter  $\theta$

$$\theta = 2(\Gamma_{it'h} - \Gamma_{iba}) + 1$$

The literature gives different methods to evaluate  $B/A$ <sup>13</sup>. The expression for  $B/A$  in terms of Moelwyn-Hughes parameter  $C_1$  which is determined from coefficient of thermal expansion<sup>14</sup>.

$$B/A = (C_1 - 1)$$

## 4. Results and Discussion

### Isotropic to SmecticA transition

The compound 10O.CB exhibits isotropic to smecticA transition. This transition is associated by the concurrent setting of orientational ordering which is long range and the periodic translational ordering arising from the layered arrangement of the molecules from totally disordered isotropic liquid. This transformation portrays the density jump of 0.3056% and coefficient of thermal expansion of  $3.0 \times 10^{-3} \text{ }^\circ\text{C}^{-1}$ . These values are in agreement with PB16A<sup>15</sup> compound but lower when compared to nO.m and PB18A<sup>16-19</sup> compounds. This is due to the nucleation. (Growth of translucent SmecticA phase is seen visually at the base of the pycnometer bulb). The isotropic liquid which is transparent

found to flow over it with distinct boundary underscoring the co-existence of both the phases.

The values of the density slopes in isotropic to smecticA transition is  $3.6 \times 10^{-4} \text{ }^\circ\text{C}^{-1}$  and smecticA to crystal transition is  $3.8 \times 10^{-4} \text{ }^\circ\text{C}^{-1}$ . The higher values of slope from smecticA to crystal transition than isotropic to smecticA suggest the further packing of the molecules with both positional and translational order.

### Isotropic to Nematic transition

The isotropic to nematic transition is observed in 6O.CB compound, this transition is associated by density jump of 0.2846% and thermal expansion coefficient of  $2.8 \times 10^{-3} \text{ }^\circ\text{C}^{-1}$ . The value of density jumps are on par with 7O.1 and 6O.O4 compounds. These values are smaller compared to 7O.O3, 7O.O5, 7O.O7, 7O.O9 compounds in literature<sup>20-22</sup>. The smaller values are common. The distinct density jump and coefficient of thermal expansion maxima suggests that the isotropic to nematic transition is of first order. The density slopes in isotropic phase  $2.2 \times 10^{-3} \text{ }^\circ\text{C}^{-1}$ , in nematic it is  $2.4 \times 10^{-3} \text{ }^\circ\text{C}^{-1}$  and in crystalline phase the value is  $2.8 \times 10^{-3} \text{ }^\circ\text{C}^{-1}$ . The higher values of density slope in nematic and crystalline phases suggest that the molecular packing are accompanied by the growth of orientational order a long range order from the totally disordered molecular arrangement of isotropic phase.

**Table III** Thermodynamic parameters of 10O.CB

T	A	C <sub>1</sub>	$\tilde{V}$	X	S <sub>o</sub>	F
330	3.80E-04	12.47126	1.115617	-0.63921	1.119131	1.472011
331	3.80E-04	12.44768	1.11594	-0.63895	1.119156	1.471242
332	1.40E-03	7.099647	1.352059	-0.45339	1.101564	0.83366
333	3.60E-04	12.83112	1.110912	-0.64305	1.118754	1.48321
334	3.60E-04	12.80662	1.11122	-0.6428	1.118779	1.482479
354	3.60E-04	12.34633	1.117347	-0.6378	1.119264	1.467881
355	3.60E-04	12.3247	1.117652	-0.63755	1.119288	1.467153
356	3.00E-03	6.68677	1.610446	-0.25918	0.942384	-0.16482
357	3.40E-04	12.73002	1.112194	-0.642	1.118859	1.480164
358	3.40E-04	12.70746	1.112484	-0.64177	1.118882	1.479474

Table III (Continued)...

T	$\Gamma$	f	A*	$\Gamma_p$	$\delta$	B/A
330	15.92735	0.059076	1.003709	6.070841	5.436169	11.47126
331	15.88271	0.059232	1.003729	6.059048	5.42487	11.44768
332	5.358124	0.157279	1.029353	3.385598	2.99561	6.099647
333	16.60782	0.056793	1.00342	6.250758	5.611417	11.83112
334	16.56153	0.056943	1.003438	6.23851	5.599655	11.80662
354	15.6909	0.059913	1.003818	6.008375	5.378808	11.34633
355	15.64996	0.06006	1.003838	5.997563	5.368441	11.3247
356	3.571167	0.218762	1.061258	3.180165	3.18446	5.68677
357	16.41674	0.057416	1.003497	6.200215	5.565569	11.73002
358	16.37409	0.057557	1.003515	6.188935	5.554726	11.70746

Table III (Continued)...

T	$\Delta$	$\Gamma_{ith}$	$\Gamma_{ich}$	$\Gamma_{iba}$	$\theta$
330	105.4699	5.735632	3.017547	2.718085	7.035094
331	105.7459	5.723838	3.011403	2.712435	7.022807
332	75.26327	3.049823	1.552018	1.497805	4.104037
333	107.0679	5.915558	3.109849	2.805709	7.219698
334	107.3474	5.90331	3.103482	2.799827	7.206965
354	112.8908	5.673163	2.983759	2.689404	6.967517
355	113.1656	5.66235	2.978129	2.684221	6.956259
356	46.13433	2.843385	1.251155	1.59223	3.502309
357	114.5977	5.865012	3.082228	2.782784	7.164456
358	114.8763	5.853732	3.076369	2.777363	7.152738

**Table IV** Thermodynamic parameters of 6O.CB

T	A	$C_1$	$\tilde{V}$	X	$S_o$	F
329	2.80E-04	15.30793	1.086744	-0.66289	1.11646	1.54006
330	2.80E-04	15.2754	1.086992	-0.66268	1.116486	1.539482
331	1.80E-03	6.80083	1.421747	-0.40038	1.077662	0.60598
332	2.40E-04	16.98618	1.07563	-0.67207	1.115212	1.565833
333	2.40E-04	16.94881	1.075846	-0.67189	1.115237	1.565334



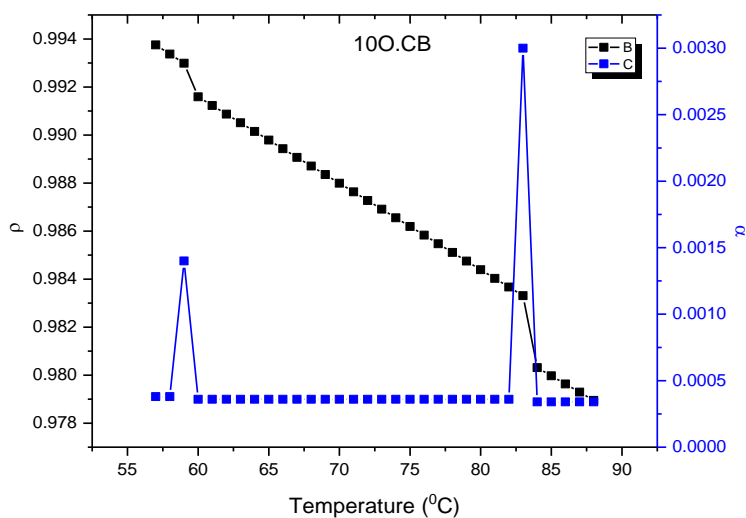
346	2.40E-04	16.48283	1.078649	-0.66957	1.115563	1.558853
347	2.40E-04	16.44845	1.078865	-0.6694	1.115587	1.558355
348	2.80E-03	6.652225	1.579155	-0.2823	0.973592	-0.0169
349	2.20E-04	17.45634	1.073013	-0.67424	1.1149	1.571869
350	2.20E-04	17.41942	1.073212	-0.67408	1.114924	1.571411

Table IV (Continued)...

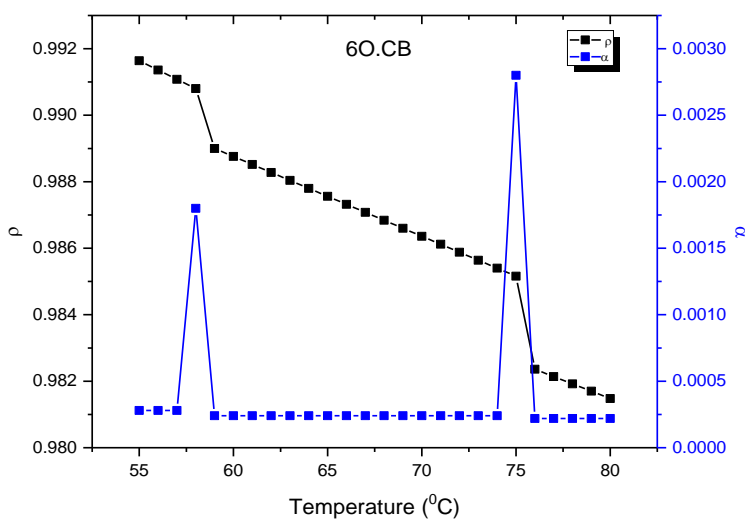
T	$\Gamma$	f	A*	$\Gamma_p$	$\delta$	B/A
329	21.2758076	0.044892	1.00211	7.489116	6.82063	14.30793
330	21.21464113	0.045015	1.002122	7.472855	6.80487	14.2754
331	4.584158711	0.179078	1.039065	3.236408	2.932916	5.80083
332	24.42908009	0.039325	1.00161	8.32822	7.64518	15.98618
333	24.35892314	0.039434	1.001619	8.309536	7.627007	15.94881
346	23.48390648	0.040843	1.001739	8.076555	7.400408	15.48283
347	23.41932283	0.040951	1.001749	8.059362	7.383688	15.44845
348	3.667200575	0.214261	1.058426	3.162736	3.106316	5.652225
349	25.31159953	0.038006	1.001502	8.563299	7.879749	16.45634
350	25.24231385	0.038106	1.00151	8.54484	7.861765	16.41942

Table IV (Continued)...

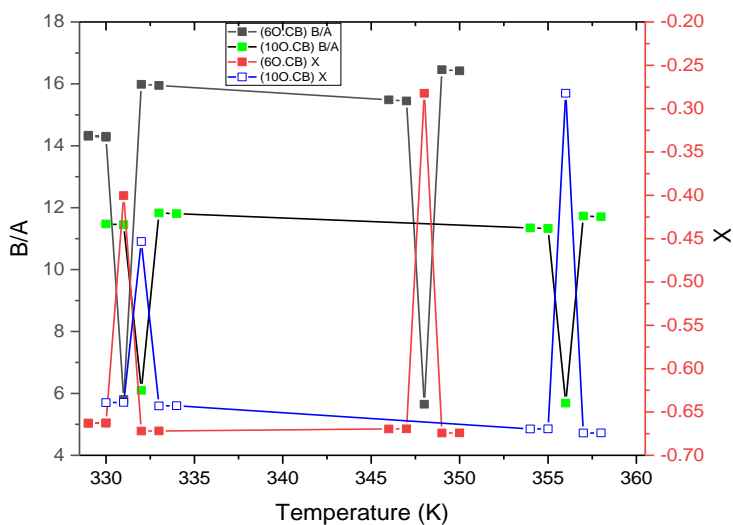
T	$\Delta$	$\Gamma_{ith}$	$\Gamma_{ich}$	$\Gamma_{iba}$	$\theta$
329	109.0448	7.153963	3.743648	3.410315	8.487296
330	109.3425	7.137701	3.735266	3.402435	8.470532
331	66.26286	2.900415	1.433957	1.466458	3.867913
332	111.5642	7.993088	4.170498	3.82259	9.340995
333	111.8704	7.974403	4.160899	3.813504	9.321799
346	115.8362	7.741416	4.041212	3.700204	9.082424
347	116.1401	7.724224	4.03238	3.691844	9.064759
348	49.11997	2.826112	1.272954	1.553158	3.545909
349	117.6553	8.228171	4.288297	3.939875	9.576593
350	117.9636	8.209711	4.278829	3.930883	9.557658



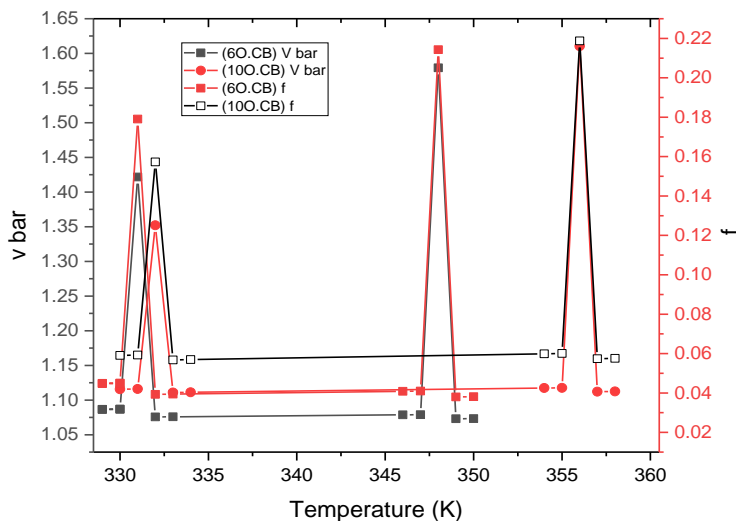
**Fig. 1** Temperature variation of density and coefficient of thermal expansion in 10O.CB



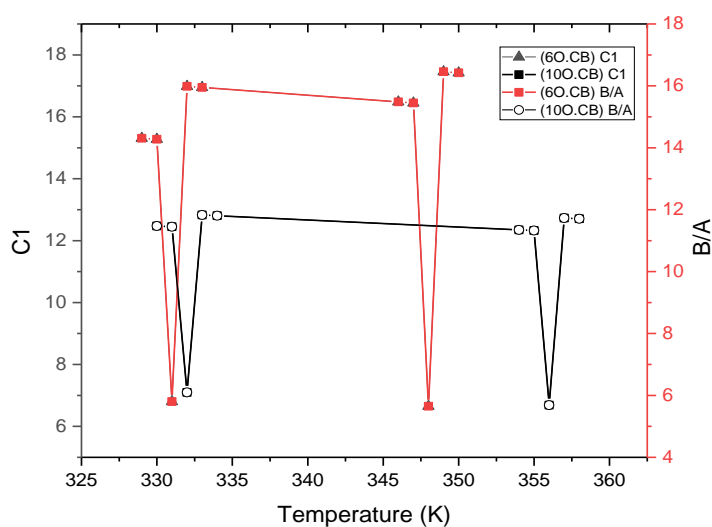
**Fig. 2** Temperature variation of density and coefficient of thermal expansion in 6O.CB



**Fig. 3** Variation of B/A and X with temperature in 6O.CB & 10O.CB



**Fig. 4** The variation of  $\tilde{v}$  and f with temperature in 6O.CB & 10O.CB



**Fig. 5** Variation of  $C_1$  and  $B/A$  with temperature in 6O.CB & 10O.CB

## 5. Salient Features

- The optical textures are recorded from polarizing optical microscope. From the observed textural features, it can be understood that the compound 6O.CB exhibit nematic transition whereas 10O.CB exhibit smecticA transition.
- The percentage of density jump from I-N transition in 6O.CB is 0.2846 and that in 10O.CB is 0.3056 at I-SmA. The higher value of jump in 10O.CB could be due to high thermal range and high transition temperature in this compound.
- The density slope increases from  $2.2 \times 10^{-4}$  to  $2.4 \times 10^{-4}$  and  $2.8 \times 10^{-4}$  for I-N-Cr in compound 6O.CB, in case of 10O.CB the density slopes are  $3.4 \times 10^{-4}$ ,  $3.6 \times 10^{-4}$  and  $3.8 \times 10^{-4}$  for I-SmA-Cr transitions.
- The density slope values increases from I-N, I-SmA, N-Cr and SmA-Cr, these higher values indicate that the further packing of molecules with both positional and translational order while it is passing from Isotropic to further LC phases.
- According to density jump, the coefficient of thermal expansion maxima values suggest that I-N and I-SmA transitions are of first order.
- Using density and thermal expansion coefficient data, the number of thermodynamic parameters are estimated and represented in Table III and IV.
- The thermal expansion coefficient maxima shows anomalous behavior shooting to the high value consequently, the parameters  $C_1$ ,  $S_0$ ,  $F$ ,  $\Gamma$ ,  $\Gamma_p$ ,  $\Delta$  etc. decreases during phase

transitions and the parameters  $\tilde{V}$ ,  $f$ ,  $A^*$  and  $X$  increases during phase variation. These variations are clearly represented in tables 3 & 4 and in Figs. 3-8.

- The B/A parameter possess almost constant value in a specific phase excluding the neighborhood of phase transition, it exhibits a low value and is dependent on thermal expansion coefficient.

## 6. Conclusions

From our investigations we have observed that 6O.CB compound exhibit only nematic and that of 10O.CB shows SmecticA transition. These transitions are of first order. By density and thermal expansion data various thermodynamic parameters are evaluated and noticed that there is abrupt increase or decrease of these parameters during phase transformation. Compared to 6O.CB and 10.CB compounds the transition temperatures of 6O.CB and 10O.CB compounds are higher. This can attributed to the existence of electro negative atom present in the molecular moiety of 6O.CB and 10O.CB samples.

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