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Thermodynamic parameters studies in *N*-(*p*-*n*-ethyloxybenzylidene)-*p*-*n*-alkoxy anilines

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ABSTRACT

From the computed thermal expansion coefficient from density variation with temperature, on *N*-(*p*-*n*-ethyloxybenzylidene)-*p*-*n*-alkoxy anilines, 2O.Om liquid crystalline (LC) compounds with $m = 3$ –4 and 6–10, different thermodynamic parameters such as Sharma parameter (S_o), Huggins parameter (F), reduced volume ($\sim V$), reduced compressibility ($\sim V^C$), etc., are studied using the volume expansion coefficient (α), which is estimated from density in isotropic and liquid crystalline phases. The parameters like intermolecular free length (L_f), molecular radius (M_r), and Beyer's nonlinearity parameter (B/A) are also computed from density data for the above compounds. Except with $m = 10$, the rest of the compounds exhibit nematic phase where as with $m = 10$, the LC material shows smectic-C in addition to the nematic phase. The results are discussed in the light of these parameters variation with temperature in a particular phase in a LC molecule and with the chain length, m . The thermodynamic parameter S_o exhibits a constant characteristic value in all the compounds like other reported liquid crystalline compounds, liquids, and polymers. Further, the molecular radius, M_r , and molecular free length, L_f , are estimated and the results are compared with the literature data available.

KEYWORDS

Density studies; liquid crystal compounds; molecular radius and free length; thermodynamic parameters

1. Introduction

In order to understand the fundamental aspects and the usage of liquid crystalline compounds (LCs) in technological applications, the study of physical properties such as viscosity, refractive indices, specific heat, etc., will provide more information. In addition to these properties, the density results also will provide the information regarding the nature of phase transitions across different LC phases as well as in between the isotropic to LC phase. Systematic studies and the literature data [1–4] on the nO.Om compounds reveal that these exhibit 13 different phase variants starting from mono variant (N, A, or C) to penta variant (NACIG). Further, it has been observed from the systematic studies on the Schiff's base compounds that the position of the oxygen atom in the molecular moiety plays an important role in deciding the clearing temperature the richness of the nematic phase and the phase variant a compound exhibits. Parameters such as specific volume ($v = 1/\rho$), and molar volume

[V_m = molecular mass/density (ρ)], which are closely related to density are also measured in a number of compounds. In the present manuscript, compounds of *N*-(*p*-ethyloxybenzylidene)-*p*-*n*-alkoxyanilines, (2O.Om) which belong to nO.Om family, the different thermodynamic parameters are estimated.

The thermodynamic parameters such as reduced volume (V^*), Moelwy Hughes parameter (C_1), reduced compressibility (β), available volume (V_a), etc., can be estimated from the computed coefficient of volume expansion (α) from density data. In addition to these, parameters like molecular free length (L_f), molecular radius (M_r), and Beyer's nonlinearity parameter (B/A), which is a measure of the equation of state of a fluid and further defined as the ratio of the coefficients of the quadratic term to the linear term in the Taylor expansion of the equation of state [5–9], can also be evaluated. Extensive studies are carried out on nO.m [10,11], nO.Om [12,] and other homologous series of compounds [13,]. Further, the literature reports these parameters evaluation in liquids [14,15] and polymers also [16]. This manuscript presents the estimation of thermodynamic parameters in *N*-(*p*-*n*-ethyloxy benzylidene)-*p*-*n*-alkyloxy anilines, 2O.Om LC compounds with $m = 3, 4$, and $6-10$ in isotropic and LC phases. Further, for the sake of completeness the authors estimated the molecular free length, L_f , molecular radius, M_r , and Beyer's nonlinearity parameter, B/A [17].

2. Experimental

The present compounds are synthesized following the established procedure reported in the literature [18]. The respective *p*-*n* ethyloxy benzaldehyde and the corresponding alkoxyanilines are taken in equimolar proportions in absolute ethanol and refluxed for 4 hr in the presence of few drops of glacial acetic acid. After refluxing the reactions for 4 hr, the solvent was removed by distillation under reduced pressure. The crude sample was subjected to repeated recrystallization from absolute ethanol in cold to give the pure compound, until the transition temperatures are constant.

The molecular formula of the compounds is given below.



where $m = 3-4$ and $6-10$.

3. Results and discussion

3.1. Thermodynamic parameter theory and expressions

The procedure for the estimation of different thermodynamic parameters using the coefficient of thermal expansion (α) are given in the references [10,14,19–26] described below.

The values of coefficient of volume expansion (α) = $1/V_m (dV_m/dT)$, where $dT = T_2 - T_1$, $dV_m = V_{m2} - V_{m1}$ are taken from literature [10,14,19–22] for the evaluation the following parameters.

3.1.a. Molecular radius (M_r)

M_r for the LC molecule can be obtained from density and refractive index results and the relevant expressions are given below [27]

(i). From density, ρ

$$M_r = \frac{1}{2} 3 \sqrt{\frac{M \sqrt{2}}{\rho N}} \quad (1)$$

where M is the molecular weight.

(ii). The refractive index data can be utilized in conjunction with molar volume to compute the molecular radius (M_r) using the following relation [27,28].

$$M_r = \left[\left(\frac{3}{4} \pi N \right) \frac{(n^2 - 1)}{(n^2 + 2)} V_m \right]^{1/3} \quad (2)$$

3.1.b. Acoustic nonlinearity parameter, B/A

There are a number of reports in literature describing in detail the theoretical and empirical approach for the estimation of B/A from α and u , respectively, the relevant expressions of the nonlinearity parameter, B/A is given as [29]

$$B/A = 2u\rho[du/dp]_T \quad (28)$$

General formalism for B/A in terms of the acoustical parameters for liquids and polymers has been made using Moelwyns-Hughes parameter (C_1), isobaric acoustic parameter (K), and the isothermal acoustic parameter (K'') (the detailed method of calculation of K and K'' from α is given elsewhere obtained from the thermal expansion coefficient, α , and from the sound velocity, u , respectively. The expressions for B/A from density are given as [30]

$$B/A = C_1 - 1 \quad (3)$$

$$B/A = 2K + 2\gamma K'' \quad (4)$$

$$n = 3C_1 - 12 \quad (5)$$

$$m = 3C_1 - (n + 6) \quad (6)$$

where m and n are the exponents describing the magnitude of attractive and repulsive force, respectively. Several authors [31–35] estimated n and highlighted its importance. The magnitude of m is 6 and is very well reported in literature and in the case of LC materials are also 6.

The measurement of density of the material and its variation with temperature using dilatometric technique is one of the methods to understand the nature of the phase transformations. This particular technique is chosen and the thermal expansion coefficient, α , obtained from density data of the material is used to estimate the thermodynamic parameters such as Moelwyn-Hughes parameter (C_1), reduced volume (V^*), reduced compressibility (β), isochoric temperature coefficient of internal pressure (X), Sharma parameter (S_0), Huggin's parameter (F), Gruneisen parameter (Γ_p), isothermal microscopic Gruneisen parameter (Γ), fraction free volume (f) and acoustical parameters like isothermal (K'), isobaric (K), isochoric (K'') acoustical parameters, isothermal Gruneisan parameter, (Γ_{ith}),

Table 1. The transition temperatures in °C and heats of transition across different phases.

Compound	Phase Variants	Scan Rate	Melting Temp. °C	Solid	Solid	NA/NC	NI
20.03	N	Heating	10 °C/min ΔH J/gm	128.06 182.71		–	–
		Cooling	10 °C/min ΔH J/gm		117.03 387.86		122.82 7.66
		TM			117.00		122.00
20.04	N	Heating	10 °C/min ΔH J/gm	127.00 112.73 38.78			131.606 2.17
		Cooling	10 °C/min ΔH J/gm		99.03 42.00		129.29 2.22
		TM			99.00		129.00
20.06	N	Heating	10 °C/min ΔH J/gm	112.00 104.34 83.82			123.93 2.40
		Cooling	10 °C/min ΔH J/gm		87.53 73.36		120.65 3.08
		TM			88.00		121.00
20.07	N	Heating	10 °C/min ΔH J/gm	104.00 99.36 84.18			118.33 1.65
		Cooling	10 °C/min ΔH J/gm		82.48 59.07		116.11 2.03
		TM			82.00		116.00
20.08	N	Heating	10 °C/min ΔH J/gm	100.00 93.67 70.32			117.75 2.14
		Cooling	10 °C/min ΔH J/gm		77.86 44.40		115.26 2.19
		TM			78.00		115.00
20.09	N	Heating	10 °C/min ΔH J/gm	94.00 97.09 26.58			111.98 1.61
		Cooling	10 °C/min ΔH J/gm		71.26 6.75	74.35 17.52	108.96 2.04
		TM				74.00	108.00
20.010	NC	Heating	10 °C/min ΔH J/gm	97.00 95.50 31.39		100.72 0.53	104.73 1.03
		Cooling	10 °C/min ΔH J/gm			82.41 39.1	100.49 0.69
		TM				82.00	100

isobaric Gruneisan parameter, (Γ_{iba}), isochoric Gruneisan parameter, (Γ_{ich}), reduced Bulk modulus, ($B^{\sim}$), isothermal Anderson-Gruneisan parameter, (δ) and the magnitude of n and m the attractive and the repulsive forces. In addition to these, the parameters like molecular free length (L_f), molecular radius (M_r), and nonlinearity parameter, (B/A), are also be evaluated. Recently, extensive studies are reported regarding the estimation of the thermodynamic parameters for a number of nO.m LC compounds utilizing the available dilatometric, ultrasonic velocity, and birefringence data [10].

The dimensionless thermodynamic parameter C_1 , defined as the pressure coefficient of bulk modulus as introduced by Moelwyn-Hughes [36], serves as severe test for the equation of state for liquids and solids and liquid crystals [10–12]. The calculated values of the Moelwyn-Hughes parameter C_1 for the materials are presented in Table 2 in both isotropic and LC phases. It is already known that C_1 offers the simplest scale for the determination of molecular ordering, structure, interactions, and anharmonicity in some materials [37]. Further, C_1 signifies the nonlinear variation with temperature. It is quite interesting to note that $(B/A) + 1 = C_1$. The ratio of $C_1/ (B/A)$ is approximately equal to unity which is true in our case as well as in other LC materials also [10–12].

The isobaric Gruneisen parameter Γ_{iba} is found to be less than the isothermal Gruneisen parameter Γ_{ith} . This trend is similar in the case of these materials except in the vicinity of phase transition. The fractional free volume (V_a/V) determines the disorder, which is due

Table 2. Variation of anharmonic parameters in different LC phases of all the compounds.

Comp.	Phase variant	C_1	$V^{\sim}$	$V^{\sim}C_1$	X	X'	S_0	F	F	A^*	Δ
20.O3	Iso,	8.16	1.24	5.892	− 0.537	− 1.583	1.119	1.149	0.215	1.059	107.71
	N	7.77	1.27	6.489	− 0.514	− 1.669	1.116	1.068	0.217	1.060	100.559
20.O4	Iso,	8.03	1.25	6.058	− 0.531	− 1.607	1.118	1.126	0.216	1.059	107.965
	N	7.85	1.26	6.340	− 0.520	− 1.648	1.116	1.088	0.216	1.060	103.182
20.O6	Iso,	7.94	1.26	6.202	− 0.525	− 1.628	1.117	1.106	0.216	1.059	104.739
	N	7.80	1.27	6.440	− 0.516	− 1.662	1.116	1.075	0.217	1.060	100.388
20.O7	Iso,	7.87	1.26	6.315	− 0.521	− 1.644	1.117	1.091	0.216	1.060	102.590
	N	7.68	1.28	6.673	− 0.508	− 1.694	1.115	1.045	0.217	1.060	97.511
20.O8	Iso,	7.925	1.26	6.221	− 0.524	− 1.631	1.117	1.104	0.216	1.060	103.024
	N	7.65	1.28	6.732	− 0.506	− 1.703	1.114	1.037	0.217	1.060	96.867
20.O9	Iso,	7.80	1.27	6.430	− 0.516	− 1.661	1.116	1.076	0.217	1.060	99.684
	N	7.27	1.33	7.805	− 0.472	− 1.843	1.106	0.910	0.219	1.061	88.769
20.O10	Iso,	7.41	1.31	7.339	− 0.486	− 1.783	1.110	0.964	0.218	1.061	91.863
	N	6.75	1.44	11.917	− 0.383	− 2.282	1.066	0.528	0.219	1.061	71.047
	SmC	6.66	1.51	15.226	− 0.336	− 2.561	1.030	0.296	0.217	1.060	61.225

Table 3. Variation of thermoacoustic parameters and molecular free length in different LC phases of all the compounds.

Com.	Phase Variant	Temp. $^{\circ}\text{C}$	n	$B^{\sim}$	Γ_{ith}	E	F	Γ_{ich}	Γ_{iba}	δ	θ
20.O3	Iso.	128	12.455	0.170	3.576	0.507	− 0.583	1.871	1.705	3.409	4.742
	N	118	11.297	0.154	3.383	0.508	− 0.669	1.760	1.623	3.246	4.520
20.O4	Iso.	134	12.087	0.165	3.514	0.508	− 0.607	1.836	1.678	3.357	4.672
	N	124	11.547	0.158	3.425	0.508	− 0.648	1.784	1.640	3.280	4.569
20.O6	Iso.	126	11.800	0.161	3.467	0.508	− 0.628	1.809	1.658	3.316	3.618
	N	116	11.377	0.155	3.396	0.508	− 0.662	1.768	1.628	3.256	4.536
20.O7	Iso.	121	11.592	0.158	3.432	0.508	− 0.644	1.789	1.643	3.287	4.578
	N	111	11.019	0.150	3.336	0.509	− 0.694	1.733	1.604	3.207	4.465
20.O8	Iso.	120	11.765	0.161	3.461	0.508	− 0.631	1.806	1.655	3.311	4.611
	N	110	10.935	0.149	3.323	0.509	− 0.703	1.724	1.598	3.196	4.449
20.O9	Iso.	113	11.393	0.156	3.399	0.508	− 0.661	1.770	1.629	3.258	4.539
	N	103	9.795	0.128	3.133	0.511	− 0.843	1.606	1.527	3.053	4.212
20.O10	Iso.	105	10.215	0.136	3.203	0.510	− 0.783	1.651	1.552	3.103	4.302
	N	98	8.234	0.084	2.872	0.517	− 1.282	1.403	1.469	2.939	3.806
	SmC	91	7.956	0.066	2.826	0.522	− 1.561	1.334	1.492	2.983	3.669

Table 4. Variation of V_a and B/A in 20.Om compounds in isotropic and nematic phase.

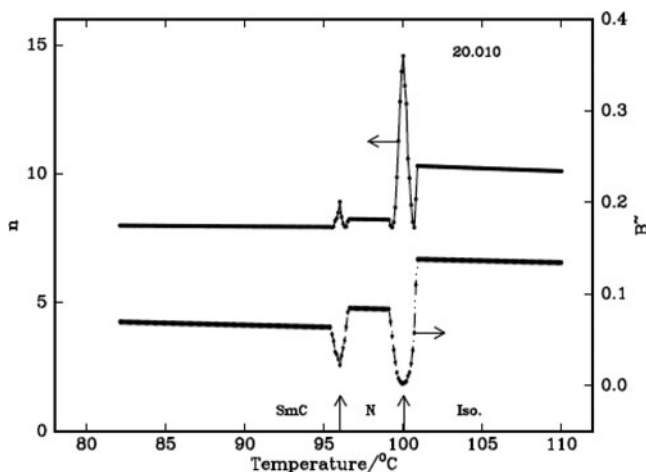
Comp.	Phase	Temp.	V_a	$B/A(C_1)$	$B/A(k)$
20.O3	Iso.	128	49.758	7.058	9.705
	N	118	51.441	6.658	9.179
20.O4	Iso.	134	53.082	6.931	9.539
	N	124	53.568	6.744	9.294
20.O6	Iso.	126	59.284	6.832	9.409
	N	116	59.496	6.685	9.216
20.O7	Iso.	121	62.372	6.760	9.314
	N	111	58.921	6.732	9.277
20.O8	Iso.	120	66.142	6.636	9.151
	N	110	69.770	6.086	8.419
20.O9	Iso.	113	69.010	6.691	9.223
	N	103	72.821	6.128	8.476
20.O10	Iso.	105	75.117	6.278	8.676
	N	98	81.821	5.534	7.662
	SmC	91	82.329	5.395	7.457

Table 5. Variation of M_r , L_{f1} , and L_{f2} in 2O.Om compounds in isotropic and nematic phase.

Comp.	Phase	Temp.	Mr 10—8cm	Lf1 10—8cm	Lf2 10—8cm
20.03	Iso,	128	4.405	0.825	0.849
	N	118	4.389	0.842	0.824
20.04	Iso,	134	4.479	0.850	0.841
	N	124	4.461	0.755	0.742
20.06	Iso,	126	4.630	0.879	0.866
	N	116	4.609	0.883	0.866
20.07	Iso,	121	4.696	0.896	0.880
	N	111	4.674	0.903	0.880
20.08	Iso,	120	4.766	0.906	0.892
	N	110	4.743	0.918	0.894
20.09	Iso,	113	4.844	0.928	0.910
	N	103	4.819	0.960	0.915
20.010	Iso,	105	4.900	0.965	0.928
	N	98	4.877	1.028	0.926
	SmC	91	4.852	1.043	0.913

to increased mobility of the molecules in a system. It is found that the fractional free volume behaves like that of the thermal expansion coefficient. For most of the materials under present investigation, the fractional free volume V_a/V_0 values are around 0.23, which are of the same order as observed for saturated hydrocarbons and other liquids and liquid crystals [38].

The results are compared with those obtained for similar nature of compounds, viz., nO.Om compounds also. This has been carried out to find out the variation of different parameters with respect to the chain length. The data pertinent to the transition temperatures in °C and heats of transition across different phases and above parameters for all the compounds are presented in Tables 1–5. The variation of the Sharma parameter S_0 , isothermal microscopic Gruneisen parameter Γ , reduced bulk modulus B^* and n in 2O.O10, acoustic nonlinearity parameter B/A , in 2O.O10 are shown in Figs. 1 and 2 and molecular radius, M_r , with temperature in different LC phases for all the compounds are shown in Fig. 3. Further, the variation of molecular radius, M_r , estimated from density with chain number is shown in Fig. 4. The increment of molecular radius per methylene unit is found to be 0.072 Å from density. The variation of available volume, V_a , with chain length is found to be less in isotropic phase and increases in LC phase and also it is observed that the increment for methylene unit is 3.45 and

**Figure 1.** Variation of magnitude of repulsive exponent, (n) and reduced Bulk modulus, (B^*) with temperature in 2O.O10 compound.

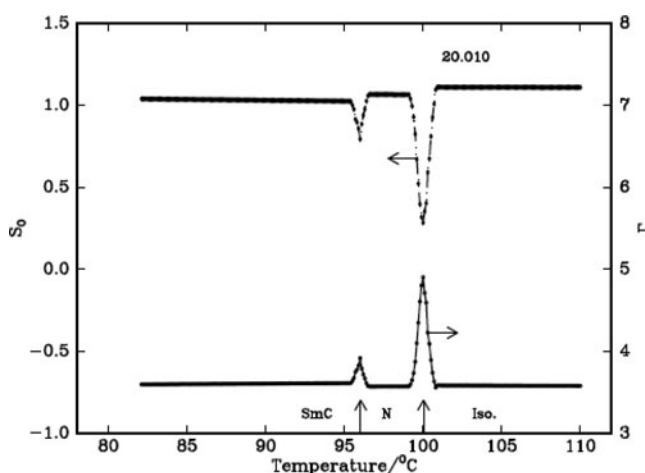


Figure 2. Variation of Sharma parameter (S_o) with temperature in 2O.O10 compound.

4.16 in isotropic and nematic phases in 2O.Om series, respectively. The variation of nonlinearity parameter and fractional free volume V_a/V with temperature for 2O.O10 compound are shown in Figs. 5 and 6 and the variation of ratio S_o/n with temperature for 2O.O10 compound is shown Fig. 7.

The salient features observed from the Tables 2–5 and the Figs. 1–7 are.

1. All the parameters exhibit a constant value in a particular phase except in the vicinity of phase transition where they exhibit singularity.
2. The parameters reduced volume, $\tilde{V}$, reduced compressibility, $\tilde{V}^{cl}$, and the isochoric temperature coefficient of internal pressure, (X) and their variation with temperature is similar to that of thermal expansion coefficient α , as excepted in all compounds as these parameters are proportional to α .
3. The parameters C_1 , X , F , Γ , Δ , and Γp and their variation with temperature show decrease with the decrease of temperature, i.e., exhibiting low values in LC phases

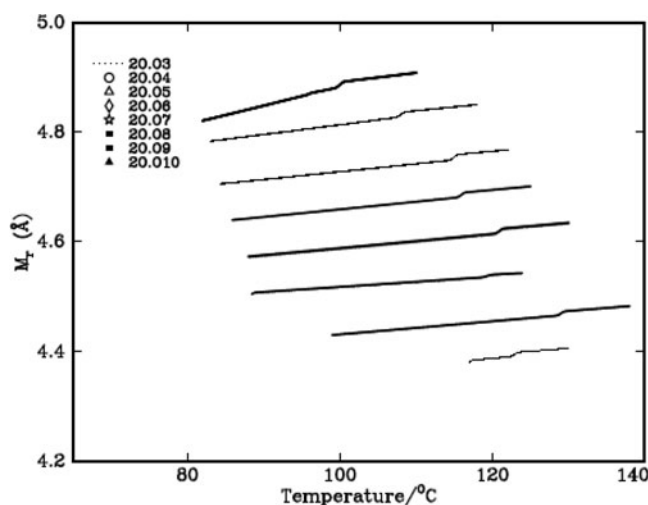


Figure 3. Variation of molecular radius (M_r) with temperature in 2O.Om compounds.

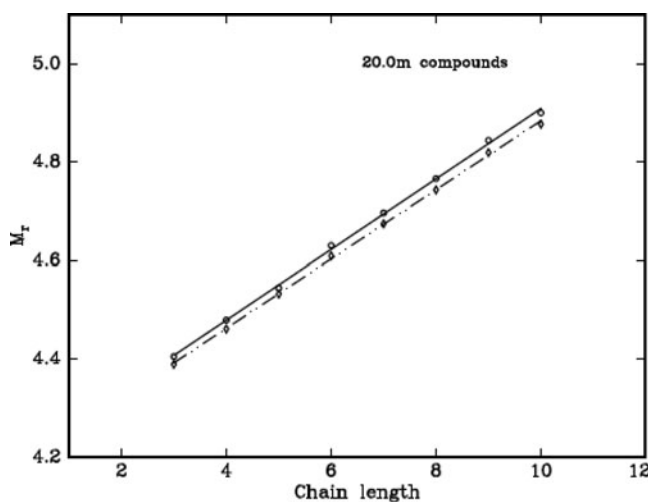


Figure 4. Variation of molecular radius (M_r) with chain length in 20.0m compounds.

compared to the isotropic phase which shows the molecular ordering increases as the temperature decreases in liquid crystal phases.

4. The Sharma parameter, S_0 is constant around 1.13 ± 0.05 irrespective of the compound including the liquid crystals. However, the value of $(3+4\alpha T)$, which is used in the determination of Sharma parameter, increases with the increase of temperature but S_0 tends to be independent of the value of α for any material. However, at and in the immediate vicinity of the LC phase transformations the value of S_0 abruptly falls to a minimum as the parameter S_0 is indirectly proportional to α .
5. The relations viz., $S_0 = 5.5 * f$ and $A^* = 5.25 * f$ hold good in these compounds also as in the case of other LC compounds.
6. The isothermal, isochoric, isobaric Gruneisen parameters also show the consistent values in any particular phase but decreases slightly in LC phases from isotropic phase.

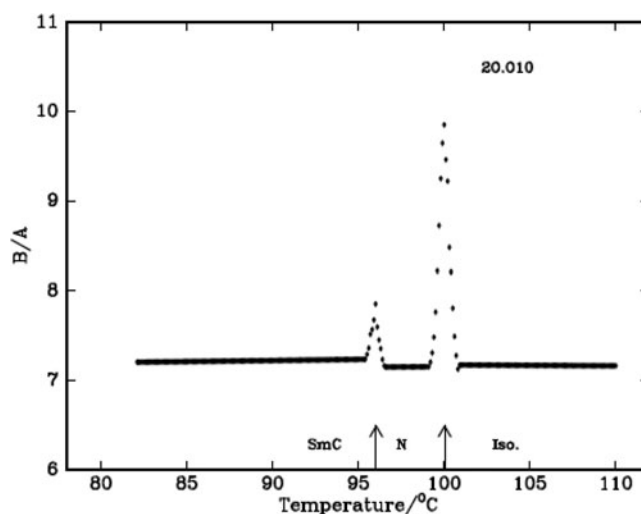


Figure 5. Variation of nonlinearity parameter (B/A) with temperature in 20.010 compound.

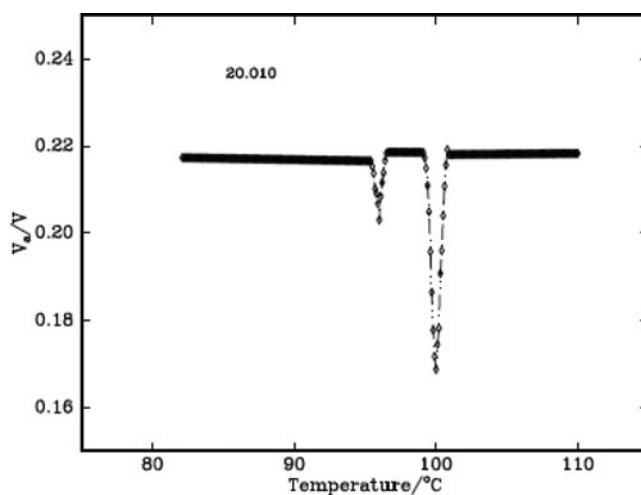


Figure 6. Variation of fractional free volume V_a/V with temperature in 2O.010 compound.

7. The available volume, V_a , is observed to be less in isotropic phase and increases in LC phase and also it is observed that the increment for methylene unit is 3.45 and 4.16 in isotropic and nematic phases in 2O.Om series, respectively.
8. The molecular free length (L_f) are calculated from available volume V_a and molar volume (V_o). However, V_o can be evaluated using different expressions. The V_o is estimated in two ways and the values at different temperatures in LC phases reveal that V_o values show agreement with one another. It has been reported earlier from the systematic studies on the benzoic acids as well as in a number of nO.m compounds found that the slope value should be around 8×10^{-4} to obtain reasonable agreement. The molecular free length values observed in all the six compounds are of comparable magnitude and the isotropic values are slightly lower compared to LC phases.
9. Further, the increment of molecular radius for methylene unit is found to be $0.064 \text{ \AA}$ from density.

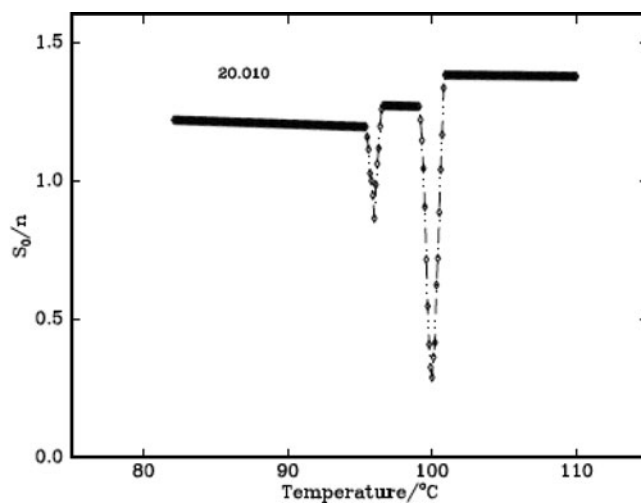


Figure 7. Variation of ratio S_0/n with temperature in 2O.010 compound.

10. The data reveals that the B/A value almost remains constant in a particular phase except at the vicinity of the phase transition where it shows a peak and the peak magnitude depends on α , and further, the magnitude of B/A is slightly smaller in LC phases compared to that in isotropic phase.
11. Generally, this parameter, B/A lies in the range 5.5–9.5, with extreme variations of 2 and 13. These extreme values are reported in some cases [10,30,39].

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