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D. Madhavi Latha[a]; V. G. K. M. Pisipati[a]; P. V. Datta Prasad[b]
[a] Liquid Crystal Research Centre, ECE Department, Koneru Lakshmaiah University, Vaddeswaram, India [b] Department of Physics, The Hindu College, Machilipatnam, India

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# Thermodynamic Parameters of nO.m Liquid-Crystal Compounds—A Dilatometric Study

D. MADHAVI LATHA,[1] V. G. K. M. PISIPATI,[1] AND P. V. DATTA PRASAD[2]

[1]Liquid Crystal Research Centre, ECE Department, Koneru Lakshmaiah University, Vaddeswaram, India
[2]Department of Physics, The Hindu College, Machilipatnam, India



*Different thermodynamic parameters are estimated from volume expansion coefficient, α, for a number of N-(*p-n-*alkoxy benzylidene)*-p-n-*alkyl anilines, nO.m compounds in isotropic phase at* $T_{IN} + 5°C$. *The parameters like intermolecular free length,* $L_f$, *and molecular radius,* $M_r$, *are also computed from density, sound velocity, and refractive index for these nO.m compounds for which the data are available in the literature. The results are discussed in light of these parameters' variations with temperature in a particular phase in a liquid-crystal molecule and in a homologous series. The nature of molecular free length and molecular radius is analyzed and discussed with respect to the different expressions used for the estimation of these parameters and the reasons for their deviations in values from one method to the other.*

**Keywords** Molecular free length and molecular radius; nO.m series; thermodynamic parameters

## Introduction

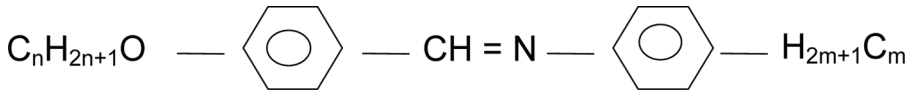
Most of the thermodynamic parameters of a material can be estimated with the combination of density [1,2], ultrasonic velocity [3–5], and birefringence [6] or individually to some extent. The estimation of different thermodynamic parameters is mostly dealt with in pure liquids and binary and ternary mixtures of liquids at ambient and at different temperatures [1,7,8]. However, the data on liquid crystals is meager and is available only on individual systems at different temperatures. In view of this, no definite conclusions could be obtained regarding different parameters and their behavior in a liquid-crystal (LC) molecule and in a particular homologous series.

One of the methods for determining the nature of phase transitions and pretransitional effects across the isotropic to liquid-crystalline state and liquid-crystalline

Address correspondence to V. G. K. M. Pisipati, Liquid Crystal Research Centre, ECE Department, Koneru Lakshmaiah University, Vaddeswaram 522 502, Guntur, Andhra Pradesh, India. E-mail: venkata_pisipati@hotmail.com

state to liquid-crystalline state is the measure of density of the material and its variation with temperature [9–16].

The thermodynamic parameters such as reduced molar volume ($V^{\sim}$), Moclwyn-Hughes parameter ($C_1$), reduced compressibility ($\beta$), fraction free volume ($f$), *etc.*, can be estimated from volume expansion ($\alpha$). In addition to these, parameters like molecular free length ($L_f$) and molecular radius ($M_r$) can be evaluated from the above parameters. This motivated the authors to carry out extensive studies to estimate the above parameters for number of nO.m compounds utilizing the available dilatometric, ultrasonic velocity, and birefringence data from the literature over the past few years. The authors are trying to address the following aspects from these systematic studies: (1) How will these parameters vary in a particular phase of the LC material? (2) How do these parameters behave in the total LC molecule? (3) What will be the behavior of these thermodynamic parameters at and around the phase transformation? (4) What will be their behavior in a particular homologous series? That is, how does the magnitude vary with either $n$ (alkoxy) or $m$ (alkyl) chain number? Are there any parameters that are temperature independent in the material but vary in a homologous series? Are there any parameters that are independent of both temperature and material? The chemical structure of the compounds is given below.




where $n$ and $m$ represent the alkoxy and alkyl chain number.



## Theory and Expressions

### *Thermodynamic Parameters*

The procedure for the estimation of different thermodynamic parameters using the coefficient of thermal expansion ($\alpha$) is described below.

These values of coefficient of volume expansion $(\alpha) = 1/V_m\ (dV_m/dT)$, where $dT = T_2 - T_1$, $dV_m = V_{m2} - V_{m1}$, are taken from the literature [6] for evaluation of the following parameters.

The Moelwyns-Hughes parameter ($C_1$) and reduced molar volume ($V^{\sim}$) are evaluated from the following expressions:

$$C_1 = \left(\frac{13}{3}\right) + \left(\frac{1}{\alpha T}\right) + \left(\frac{4\alpha T}{3}\right) \tag{1}$$

$$V^{\sim} = \left\{\frac{\left(\frac{\alpha T}{3}\right)}{(1+\alpha T)} + 1\right\}^3 \tag{2}$$

The isochoric temperature coefficient of internal pressure ($X$) is given by

$$X = -\frac{2(1+2\alpha T)}{V^{\sim C_1}} \tag{3}$$

where $V_1^{\sim C} = \beta$ is the reduced compressibility.

Sharma parameter [18] $S_0$ is given by the expression

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

The Huggins parameter [1,17] $F$ of a liquid crystal is related to $S_0$ by the equation

$$F = 2\{1 + [S_0/(3 + 4\alpha T)]\} - (3 + 4\alpha T/3) \tag{5}$$

The isothermal microscopic Grunessian parameter ($\tau$) is a measure of volume dependence of the hormonicity of the normal mode frequency ($\nu$) of molecular vibrations of a material and is related to $F$ and $S_0$ as

$$\tau = (2/3)\alpha T + (2 - F + 4\alpha T)/2\alpha T \tag{6}$$

The fraction free volume ($f$) is a measure of disorder due to increase of mobility of molecules in a liquid crystal and can be expressed in terms of isothermal Grunessian parameter ($\tau$) as

$$f = 1/(\tau + 1) = V_a/V \tag{7}$$

where $V_a$ is the available volume. The estimation of $V_a$ is described below.

Thermal parameter $A^*$ is a dimensionless parameter that shows that at low temperatures, a liquid crystal tends to be ordered and thereby makes $A^*$ equal to unity

$$A^* = 1 + (f/\tau) \tag{8}$$

The Grunessian parameter ($\tau_p$) for liquid crystals can be obtained from

$$\tau_p = (2/3)\alpha T + (1/2\alpha T) + 2 \tag{9}$$

The isochoric acoustical parameter, $\Delta$, is given as

$$\Delta = \left(\frac{-\alpha T}{2}\right) \tag{10}$$

### *Intermolecular Free Length ($L_f$)*

The estimation of $L_f$ is described below using the thermodynamic parameters. Using Eqs. (1)–(3), the isothermal ($K'$), isochoric ($K''$), and isobaric ($K$) acoustical parameters are obtained from the following expressions:

$$K' = K + K'' = \frac{1}{2}\left\{3 + \frac{[S^*(1 + \alpha T) + X]}{\alpha T}\right\} \tag{11}$$

$$K'' = 1 + \left(\frac{X}{2\alpha T}\right) \tag{12}$$

and

$$K = \frac{1}{2}\left\{1 + \left[\frac{S^*(1+\alpha T)}{\alpha T}\right]\right\} \tag{13}$$

where

$$S^* = 1 + \left(\frac{4\alpha T}{3}\right) \tag{14}$$

The isothermal acoustical parameter ($K'$) is related to the available volume ($V_a$) of the compound as

$$\frac{V_a}{V_m} = \frac{1}{(K'+1)} = \frac{1}{(K''+K+1)} \tag{15}$$

where $V_m$ is molar volume (molecular weight/density) and the available volume ($V_a$) can be deduced using the thermoacoustical parameter $K'$ as

$$\mathrm{V}_a = \frac{V_m}{K'+1} \tag{16}$$

Then the intermolecular free-length ($L_f$) is given by the relation

$$L_f = \frac{2V_a}{Y} \tag{17}$$

where $V_a$ represents the available volume per mole and $Y$ is surface area per mole given by

$$V_0 = V - V_a \tag{18a}$$

where

$$Y = (36\pi N V_0^2)^{1/3} \tag{18b}$$

The molar volume at 0 K ($V_0$) can be obtained by a different method [19] as

$$V_0 = \frac{V}{V^{\sim}} \tag{19}$$

$N$ is the Avogadro number and $V^{\sim}$ is taken from Eq. (2). Further, $V_0$ can be obtained directly from molar volume data by extrapolating isotropic data to 0 K.

### *Molecular Radius ($M_r$)*

$M_r$ for the LC molecule can be obtained from density, ultrasonic velocity and refractive index results and the relevant expressions are given below.

(i) From density, $\rho$ [6]

$$M_r = \frac{1}{2}3\sqrt{\frac{M\sqrt{2}}{\rho N}} \tag{20}$$

where $M$ is the molecular weight.

(ii) From ultrasonic velocity ($U$)

The following equations from (21) to (24) are employed to estimate the molecular radius using ultrasonic and density data.

Schaff's relation [20]

$$r = 3\sqrt{\frac{M}{\rho N}}3\sqrt{\frac{3}{16\pi}\left[1 - \frac{\gamma RT}{Mu^2}\left(\sqrt{1 + \frac{Mu^2}{3\gamma RT}} - 1\right)\right]} \tag{21}$$

Rao's relation [21]

$$r = 3\sqrt{\frac{M}{\rho N}}3\sqrt{\frac{3}{16\pi}\left[1 - \frac{\gamma RT}{Mu^2}\left(\sqrt{1 + \frac{Mu^2}{\gamma RT}} - 1\right)\right]} \tag{22}$$

The above two relations are based on van der Waal's equation of state. The following relations due to Kincaid and Eyring [22,23], Kittel [24], and Glasstone [25] are based on the assumption that the velocity of sound results from the propagation of sound waves inside the molecule and in the free space between them.

Erying's relation

$$r = 3\sqrt{\frac{M}{\rho N}\frac{1}{2}}3\sqrt{\left[1 - \left(1 - \frac{1}{u}\sqrt{\frac{\gamma RT}{M}}\right)^3\right]\sqrt{2}} \tag{23}$$

Kittel's relation

$$r = 3\sqrt{\frac{M}{\rho N}\frac{1}{2}}3\sqrt{\left[\left(1 - \frac{1}{u}\sqrt{\frac{3\gamma RT}{M}}\right)\right]\sqrt{2}} \tag{24}$$

(iii) The refractive index data can be utilized in conjunction with molar volume to compute the molecular radius ($M_r$) using the following relation [26]:

$$M_r = \left[\left(\frac{3}{4\pi N}\right)\frac{(n^2 - 1)}{(n^2 + 2)}V_m\right]^{1/3} \tag{25}$$

## Results and Discussion

The different thermodynamic parameters are estimated for a number of nO.m compounds with varying $n$ and $m$ values using Eqs. (1) to (10). The relevant data are taken from the literature and the corresponding references are depicted in Table 1. The density data, $\rho$, (gm/cm$^3$) and in turn the volume expansion coefficient $\alpha$ ($10^{-4}{}^{\circ}C^{-1}$), are taken from the literature over the past two and half decades. The data are from the senior author Pisipati and his group.

These parameters are evaluated in the isotropic phase of each material at $(T_{IN}+5)^{\circ}C$ (where $T_{IN}$ is the isotropic nematic transition temperature). The general observation is that all the parameters exhibit an odd even variation either with $n$ or $m$ chain number. This is expected along the lines of the isotropic–nematic (IN) transition temperature, which shows an odd–even effect. Further, all these parameters exhibit either a sharp increase or decrease at the transition with the pretranstional phenomena. However, the Sharma parameter, $S_0$, is constant with respect to the temperature and the material with a value $1.11 \pm 0.01$ and this feature persists for all nO.m compounds. This is shown in Fig. 1 for the case of compounds 1O.12, 2O.12, 1O.14, and 2O.14, which exhibit nematic (N), N, N, N, and smectic-A (A) LC phases, respectively. This is in agreement with the reported data on liquids as well on liquid crystals [1,2,4]. The value $(3+4\alpha T)$ increases with the increase of temperature but $S_0$ remains independent of temperature as well as compound and exhibits an independence with $\alpha$ of any material system [1,27,28]. However, $S_0$ shows variation in and around the transition temperature and falls to a minimum value at the transition (Fig. 1).

Another thermal parameter, $A^*$, is also constant like $S_0$ irrespective of temperature in a material with a value around $1.07 \pm 0.02$ (not in a homologous series) but exhibits a minimum at the transition like $S_0$.

To understand the nature of these parameters and their variation in a particular compound and in a homologous series, the percentage of deviations of these parameters from a mean value in a homologous series is estimated. It has been observed that all the parameters exhibit a variation $<10\%$ except for the case of parameters such as the Huggins parameter, $F$; isothermal microscopic Grunessian parameter, $\tau$; and the fraction free volume, $f$. The percentage of deviation observed in these cases varies between $>10\%$ to as high as 30%. This particular variation for the case of $F$ and $\tau$ is illustrated in Fig. 2 for 5O.m compounds.

Further, all the parameters show slight variation in their magnitude in different phases at either higher or lower values depending upon whether the parameter is directly or indirectly proportional to $\alpha T$. Table 2 shows how these parameters vary in different phases for the compounds 1O.12, 2O.12, 1O.14, and 2O.14. However, the variation of any parameter in any liquid-crystalline phase depends on the thermal span of the phase. The parameters exhibit slight variation in a particular phase if it possesses long thermal range. Further, this variation shows a dip/elevation if the molecules are interdigitized [29]. Figure 3 depicts these aspects in the case of 1O.12 and 5O.16 compounds respectively for the case of the parameter, $V_1^{\sim C} = \beta$, the reduced compressibility.

### *Molecular Free Length ($L_f$)*

Equations (17) to (19) are used to evaluate $L_f$ for the nO.m series. The $L_f$ evaluated for all the compounds in two different ways is presented in Table 3 along with other

**Table 1.** Different thermodynamic parameters of nO.m compounds at $T_{IN} + 5°C$

| nO.m | $T$ (°C) | $V\sim$ | $C_1$ | $B$ | $X_1$ | $X$ | $S_0$ | $F$ | $\tau$ | $f$ | $A^*$ | $\tau_P$ | $\Delta$ |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| 4O.4 [31] | 79.4 | 1.241 | 8.179 | 5.861 | −1.578 | −0.538 | 1.119 | 1.153 | 3.658 | 0.215 | 1.059 | 3.923 | 94.87 |
| 4O.5 [31] | 86.1 | 1.235 | 8.277 | 5.744 | −1.560 | −0.543 | 1.119 | 1.170 | 3.669 | 0.214 | 1.058 | 3.972 | 97.55 |
| 4O.6 [31] | 81.0 | 1.237 | 8.242 | 5.785 | −1.566 | −0.542 | 1.119 | 1.164 | 3.665 | 0.214 | 1.058 | 3.954 | 95.86 |
| 4O.7 [31] | 87.8 | 1.228 | 8.390 | 5.619 | −1.541 | −0.549 | 1.120 | 1.188 | 3.681 | 0.214 | 1.058 | 4.028 | 98.96 |
| 4O.8 [31] | 83.2 | 1.248 | 8.074 | 5.999 | −1.598 | −0.533 | 1.118 | 1.134 | 3.647 | 0.215 | 1.059 | 3.871 | 94.91 |
| 4O.9 [31] | 85.2 | 1.252 | 8.024 | 6.071 | −1.609 | −0.530 | 1.118 | 1.124 | 3.641 | 0.215 | 1.059 | 3.845 | 94.93 |
| 4O.10 [32] | 81.9 | 1.301 | 7.471 | 7.161 | −1.760 | −0.492 | 1.111 | 0.985 | 3.589 | 0.218 | 1.061 | 3.569 | 87.24 |
| 4O.12 [33] | 81.7 | 1.297 | 7.513 | 7.050 | −1.746 | −0.495 | 1.112 | 0.998 | 3.592 | 0.218 | 1.061 | 3.590 | 87.82 |
| 4O.14 [34] | 79.5 | 1.282 | 7.661 | 6.706 | −1.699 | −0.507 | 1.114 | 1.041 | 3.605 | 0.217 | 1.060 | 3.664 | 89.31 |
| 4O.16 [34] | 75.0 | 1.237 | 8.240 | 5.786 | −1.567 | −0.541 | 1.119 | 1.164 | 3.665 | 0.214 | 1.058 | 3.954 | 94.22 |
| 5O.1 [35] | 67.4 | 1.230 | 8.369 | 5.642 | −1.545 | −0.548 | 1.120 | 1.185 | 3.679 | 0.214 | 1.058 | 4.018 | 93.20 |
| 5O.2 [36] | 64.0 | 1.216 | 8.627 | 5.390 | −1.506 | −0.559 | 1.120 | 1.222 | 3.708 | 0.212 | 1.057 | 4.147 | 94.13 |
| 5O.5 [37] | 82.8 | 1.260 | 7.920 | 6.229 | −1.632 | −0.524 | 1.117 | 1.103 | 3.631 | 0.216 | 1.059 | 3.793 | 93.22 |
| 5O.6 [37] | 77.9 | 1.247 | 8.091 | 5.977 | −1.595 | −0.534 | 1.118 | 1.137 | 3.648 | 0.215 | 1.059 | 3.879 | 93.65 |
| 5O.7 [37] | 83.1 | 1.260 | 7.917 | 6.233 | −1.632 | −0.524 | 1.117 | 1.102 | 3.630 | 0.216 | 1.059 | 3.792 | 93.27 |
| 5O.8 [38] | 81.0 | 1.227 | 8.420 | 5.587 | −1.536 | −0.550 | 1.120 | 1.192 | 3.685 | 0.213 | 1.058 | 4.043 | 97.34 |
| 5O.9 [39] | 78.8 | 1.254 | 7.992 | 6.117 | −1.616 | −0.528 | 1.118 | 1.118 | 3.638 | 0.216 | 1.059 | 3.830 | 92.92 |
| 5O.10 [40] | 81.2 | 1.225 | 8.452 | 5.555 | −1.531 | −0.551 | 1.120 | 1.197 | 3.688 | 0.213 | 1.058 | 4.059 | 97.64 |
| 5O.12 [41] | 78.9 | 1.344 | 7.154 | 8.282 | −1.901 | −0.459 | 1.102 | 0.858 | 3.567 | 0.219 | 1.061 | 3.410 | 80.77 |
| 5O.14 [41] | 76.5 | 1.239 | 8.208 | 5.825 | −1.573 | −0.540 | 1.119 | 1.158 | 3.661 | 0.215 | 1.059 | 3.937 | 94.35 |
| 5O.16 [41] | 73.9 | 1.254 | 8.000 | 6.106 | −1.614 | −0.529 | 1.118 | 1.119 | 3.639 | 0.216 | 1.059 | 3.833 | 91.70 |
| 6O.2 [36] | 73.4 | 1.221 | 8.524 | 5.484 | −1.520 | −0.554 | 1.120 | 1.208 | 3.696 | 0.213 | 1.058 | 4.095 | 96.03 |
| 6O.4 [42] | 82.3 | 1.241 | 8.190 | 5.848 | −1.576 | −0.539 | 1.119 | 1.155 | 3.659 | 0.215 | 1.059 | 3.928 | 95.75 |
| 6O.5 [43] | 95.0 | 1.321 | 7.309 | 7.658 | −1.824 | −0.476 | 1.107 | 0.927 | 3.577 | 0.219 | 1.061 | 3.488 | 87.66 |
| 6O.6 [43] | 94.0 | 1.288 | 7.603 | 6.833 | −1.716 | −0.502 | 1.113 | 1.025 | 3.600 | 0.217 | 1.060 | 3.635 | 92.19 |
| 6O.7 [43] | 84.9 | 1.274 | 7.746 | 6.534 | −1.675 | −0.513 | 1.115 | 1.063 | 3.613 | 0.217 | 1.060 | 3.706 | 91.76 |
| 6O.8 [44] | 87.2 | 1.295 | 7.528 | 7.013 | −1.741 | −0.496 | 1.112 | 1.003 | 3.594 | 0.218 | 1.061 | 3.597 | 89.40 |

| | | | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| 7O.1 [45] | 77.9 | 1.238 | 8.233 | 5.796 | −1.568 | −0.541 | 1.119 | 1.162 | 3.664 | 0.214 | 1.059 | 3.950 | 94.94 |
| 7O.2 [43] | 74.6 | 1.340 | 7.179 | 8.167 | −1.887 | −0.462 | 1.103 | 0.871 | 3.569 | 0.219 | 1.061 | 3.423 | 80.32 |
| 7O.3 [43] | 85.5 | 1.280 | 7.678 | 6.670 | −1.694 | −0.508 | 1.115 | 1.045 | 3.607 | 0.217 | 1.060 | 3.672 | 91.05 |
| 7O.4 [46] | 81.6 | 1.285 | 7.626 | 6.780 | −1.709 | −0.504 | 1.114 | 1.031 | 3.602 | 0.217 | 1.060 | 3.646 | 89.39 |
| 7O.5 [47] | 88.2 | 1.217 | 8.595 | 5.418 | −1.510 | −0.557 | 1.120 | 1.217 | 3.705 | 0.213 | 1.057 | 4.131 | 100.66 |
| 7O.6 [48] | 85.0 | 1.240 | 8.207 | 5.827 | −1.573 | −0.540 | 1.119 | 1.158 | 3.661 | 0.215 | 1.059 | 3.937 | 96.63 |
| 7O.7 [43] | 88.8 | 1.309 | 7.410 | 7.335 | −1.783 | −0.486 | 1.110 | 0.964 | 3.584 | 0.218 | 1.061 | 3.538 | 87.95 |
| 7O.8 [49] | 88.0 | 1.241 | 8.181 | 5.859 | −1.578 | −0.539 | 1.119 | 1.153 | 3.658 | 0.215 | 1.059 | 3.924 | 97.20 |
| 7O.9 [43] | 89.0 | 1.234 | 8.297 | 5.721 | −1.557 | −0.544 | 1.119 | 1.173 | 3.671 | 0.214 | 1.058 | 3.982 | 98.51 |
| 7O.10 [43] | 87.8 | 1.246 | 8.115 | 5.944 | −1.590 | −0.535 | 1.118 | 1.142 | 3.651 | 0.215 | 1.059 | 3.891 | 96.53 |
| 8O.4 [50] | 85.2 | 1.237 | 8.240 | 5.787 | −1.567 | −0.541 | 1.119 | 1.164 | 3.665 | 0.214 | 1.059 | 3.953 | 96.98 |
| 8O.5 [51] | 89.0 | 1.315 | 7.356 | 7.500 | −1.804 | −0.481 | 1.109 | 0.945 | 3.580 | 0.218 | 1.061 | 3.511 | 87.23 |
| 8O.6 [51] | 91.0 | 1.307 | 7.422 | 7.298 | −1.778 | −0.487 | 1.110 | 0.968 | 3.585 | 0.218 | 1.061 | 3.544 | 88.69 |
| 8O.7 [51] | 91.9 | 1.306 | 7.429 | 7.279 | −1.776 | −0.488 | 1.110 | 0.971 | 3.585 | 0.218 | 1.061 | 3.548 | 89.02 |
| 8O.8 [51] | 91.2 | 1.294 | 7.542 | 6.976 | −1.736 | −0.498 | 1.113 | 1.007 | 3.595 | 0.218 | 1.061 | 3.605 | 90.62 |
| 8O.9 [51] | 92.4 | 1.264 | 7.866 | 6.317 | −1.644 | −0.521 | 1.117 | 1.091 | 3.625 | 0.216 | 1.060 | 3.766 | 95.13 |
| 8O.10 [48] | 91.6 | 1.277 | 7.715 | 6.594 | −1.684 | −0.511 | 1.115 | 1.055 | 3.610 | 0.217 | 1.060 | 3.691 | 93.09 |
| 9O.4 [52] | 90.9 | 1.266 | 7.847 | 6.350 | −1.649 | −0.519 | 1.116 | 1.087 | 3.623 | 0.216 | 1.060 | 3.757 | 94.52 |
| 9O.5 [39] | 97.7 | 1.252 | 8.025 | 6.069 | −1.609 | −0.530 | 1.118 | 1.124 | 3.642 | 0.215 | 1.059 | 3.846 | 98.26 |
| 9O.6 [53] | 95.0 | 1.294 | 7.541 | 6.978 | −1.736 | −0.498 | 1.112 | 1.007 | 3.595 | 0.218 | 1.061 | 3.604 | 91.55 |
| 9O.8 [53] | 95.5 | 1.282 | 7.657 | 6.714 | −1.700 | −0.506 | 1.114 | 1.040 | 3.605 | 0.217 | 1.060 | 3.662 | 93.31 |
| 9O.9 [A] | 96.9 | 1.269 | 7.810 | 6.415 | −1.658 | −0.517 | 1.116 | 1.078 | 3.620 | 0.216 | 1.060 | 3.738 | 95.63 |
| 9O.10 [A] | 93.9 | 1.282 | 7.662 | 6.703 | −1.699 | −0.507 | 1.114 | 1.041 | 3.605 | 0.217 | 1.060 | 3.664 | 92.98 |
| 9O.12 [A] | 95.8 | 1.300 | 7.485 | 7.123 | −1.755 | −0.493 | 1.111 | 0.989 | 3.590 | 0.218 | 1.061 | 3.576 | 90.88 |
| 9O.14 [A] | 91.5 | 1.289 | 7.593 | 6.856 | −1.720 | −0.502 | 1.113 | 1.022 | 3.599 | 0.217 | 1.060 | 3.630 | 91.42 |
| 1O.12 [54] | 64.9 | 1.259 | 7.934 | 6.207 | −1.629 | −0.525 | 1.117 | 1.106 | 3.632 | 0.216 | 1.059 | 3.800 | 88.67 |
| 2O.12 [54] | 81.2 | 1.238 | 8.237 | 5.790 | −1.567 | −0.541 | 1.119 | 1.163 | 3.664 | 0.214 | 1.059 | 3.952 | 95.87 |
| 4O.12 [33] | 81.7 | 1.297 | 7.513 | 7.050 | −1.746 | −0.495 | 1.112 | 0.998 | 3.592 | 0.218 | 1.061 | 3.590 | 87.82 |
| 5O.12 [41] | 78.9 | 1.344 | 7.154 | 8.282 | −1.901 | −0.459 | 1.102 | 0.858 | 3.567 | 0.219 | 1.061 | 3.410 | 80.77 |

(*Continued*)

**Table 1.** Continued

| nO.m | $T$ (°C) | $V\sim$ | $C_1$ | $B$ | $X_1$ | $X$ | $S_0$ | $F$ | $\tau$ | $f$ | $A^*$ | $\tau_P$ | $\Delta$ |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| 1O.O12 [A] | 90.1 | 1.326 | 7.271 | 7.797 | −1.842 | −0.472 | 1.106 | 0.911 | 3.574 | 0.219 | 1.061 | 3.469 | 85.77 |
| 1O.O1 [A] | 79.0 | 1.170 | 9.805 | 4.648 | −1.383 | −0.595 | 1.121 | 1.340 | 3.850 | 0.206 | 1.054 | 4.736 | 104.76 |
| 1O.O6 [A] | 89.2 | 1.321 | 7.312 | 7.647 | −1.823 | −0.477 | 1.107 | 0.928 | 3.577 | 0.218 | 1.061 | 3.490 | 86.34 |
| 1O.O8 [A] | 94.8 | 1.312 | 7.379 | 7.429 | −1.795 | −0.483 | 1.109 | 0.953 | 3.582 | 0.218 | 1.061 | 3.523 | 88.87 |
| 1O.O9 [A] | 96.1 | 1.339 | 7.182 | 8.157 | −1.886 | −0.462 | 1.103 | 0.872 | 3.569 | 0.219 | 1.061 | 3.424 | 85.34 |
| 1O.O10 [A] | 93.9 | 1.245 | 8.118 | 5.940 | −1.590 | −0.535 | 1.118 | 1.142 | 3.651 | 0.215 | 1.059 | 3.892 | 98.19 |
| 1O.O12 [A] | 90.1 | 1.326 | 7.271 | 7.797 | −1.842 | −0.472 | 1.106 | 0.911 | 3.574 | 0.219 | 1.061 | 3.469 | 85.77 |
| 1O.O16 [A] | 90.8 | 1.299 | 7.491 | 7.107 | −1.753 | −0.493 | 1.112 | 0.991 | 3.590 | 0.218 | 1.061 | 3.579 | 89.74 |
| 1O.14 [54] | 65.3 | 1.206 | 8.824 | 5.226 | −1.480 | −0.566 | 1.121 | 1.247 | 3.731 | 0.211 | 1.057 | 4.245 | 95.77 |
| 2O.14 [54] | 75.1 | 1.272 | 7.771 | 6.485 | −1.668 | −0.515 | 1.116 | 1.069 | 3.616 | 0.217 | 1.060 | 3.719 | 89.55 |
| 4O.14 [34] | 79.5 | 1.282 | 7.661 | 6.706 | −1.699 | −0.507 | 1.114 | 1.041 | 3.605 | 0.217 | 1.060 | 3.664 | 89.31 |
| 5O.14 [41] | 76.5 | 1.239 | 8.208 | 5.825 | −1.573 | −0.540 | 1.119 | 1.158 | 3.661 | 0.215 | 1.059 | 3.937 | 94.35 |
| 7O.14 [55] | 82.2 | 1.285 | 7.625 | 6.784 | −1.710 | −0.504 | 1.114 | 1.031 | 3.602 | 0.217 | 1.060 | 3.646 | 89.52 |
| 8O.14 [55] | 87.7 | 1.323 | 7.297 | 7.700 | −1.830 | −0.475 | 1.107 | 0.922 | 3.576 | 0.219 | 1.061 | 3.482 | 85.70 |
| 9O.14 [55] | 88.3 | 1.268 | 7.816 | 6.404 | −1.657 | −0.517 | 1.116 | 1.080 | 3.620 | 0.216 | 1.060 | 3.741 | 93.48 |
| 1O.O14 [56] | 88.2 | 1.292 | 7.563 | 6.926 | −1.729 | −0.499 | 1.113 | 1.013 | 3.597 | 0.218 | 1.060 | 3.615 | 90.17 |
| 11O.14 [55] | 93.6 | 1.257 | 7.955 | 6.174 | −1.624 | −0.526 | 1.117 | 1.110 | 3.634 | 0.216 | 1.059 | 3.811 | 96.43 |
| 12O.14 [55] | 92.5 | 1.266 | 7.839 | 6.364 | −1.651 | −0.519 | 1.116 | 1.085 | 3.622 | 0.216 | 1.060 | 3.753 | 94.83 |
| 13O.14 [55] | 92.9 | 1.280 | 7.681 | 6.662 | −1.693 | −0.508 | 1.115 | 1.046 | 3.607 | 0.217 | 1.060 | 3.674 | 92.98 |
| 14O.14 [55] | 93.5 | 1.365 | 7.032 | 8.936 | −1.977 | −0.443 | 1.096 | 0.791 | 3.563 | 0.219 | 1.062 | 3.349 | 81.09 |
| 15O.14 [55] | 93.6 | 1.276 | 7.721 | 6.582 | −1.682 | −0.511 | 1.115 | 1.056 | 3.611 | 0.217 | 1.060 | 3.694 | 93.68 |
| 16O.14 [55] | 92.9 | 1.282 | 7.656 | 6.715 | −1.700 | −0.506 | 1.114 | 1.040 | 3.605 | 0.217 | 1.060 | 3.661 | 92.65 |
| 18O.14 [55] | 93.8 | 1.282 | 7.030 | 5.748 | −1.978 | −0.688 | 1.705 | 1.036 | 3.311 | 0.232 | 1.070 | 3.349 | 126.22 |
| 12O.12 [57] | 93.0 | 1.285 | 7.624 | 6.785 | −1.710 | −0.504 | 1.114 | 1.031 | 3.602 | 0.217 | 1.060 | 3.645 | 92.24 |
| 12O.14 [57] | 92.5 | 1.266 | 7.839 | 6.364 | −1.651 | −0.519 | 1.116 | 1.085 | 3.622 | 0.216 | 1.060 | 3.753 | 94.83 |
| 12O.16 [57] | 90.6 | 1.292 | 7.564 | 6.924 | −1.729 | −0.499 | 1.113 | 1.014 | 3.597 | 0.218 | 1.060 | 3.615 | 90.78 |

[A]. Pisipati, V. G. K. M., & Prabhu, C. R. (unpublished work).

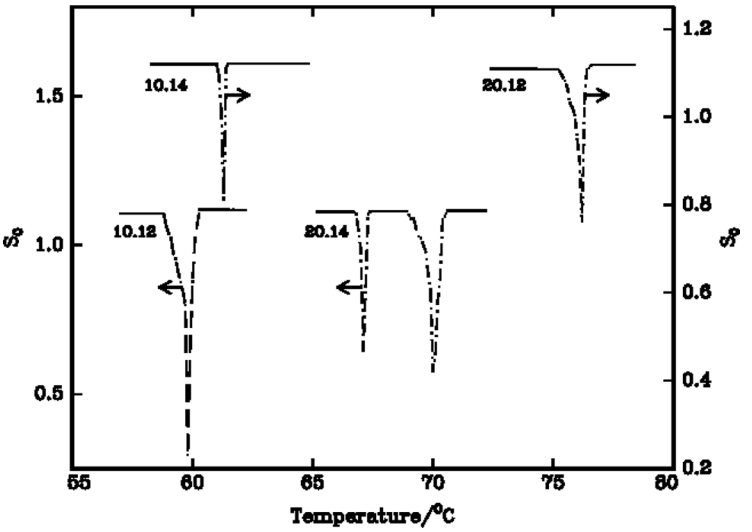



**Figure 1.** Temperature variation of $S_0$ in 1O.12, 2O.12, 1O.14, and 2O.14 compounds.



thermodynamic parameters, viz. the isothermal ($K'$), isochoric ($K''$) and isobaric ($K$) acoustical parameters, $S^*$, and the available volume. The data reveal that the $L_f$ estimated from different $V_0$ has almost identical values for all the compounds. The $L_f$ obtained from Eq. (19) is presented in Table 4 for ready reference for all the nO.m compounds.

In the case of 5O.m compounds the $L_f$ estimated from $V_0$ obtained from extrapolation technique also exhibits values close to those obtained from other two methods. Table 5 illustrates values obtained for $V_0$ from different methods along with the estimated $L_f$ values using $V_0$ in the case of 5O.m series. The $V_0$ values, which increase with the $m$ value like molar volume, show some kind of irregularity, which

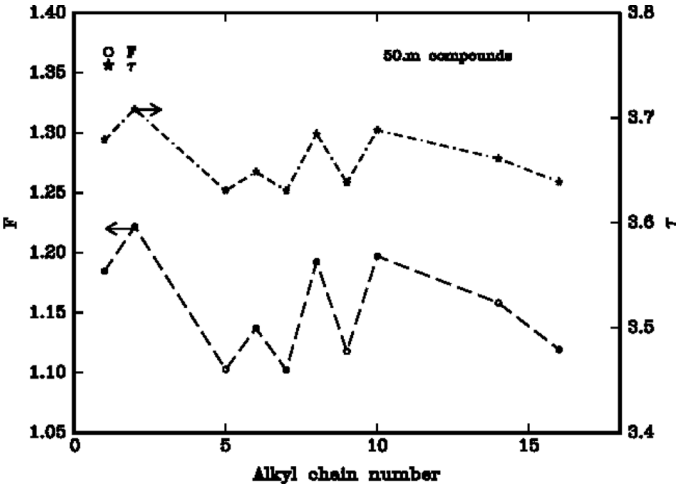



**Figure 2.** Variation $F$ and $\tau$ in 5O.m compounds.

**Table 2.** Thermodynamic parameters in different LC phases of 1O.12, 2O.12, 1O.14, and 2O.14

| Thermodynamic parameter | Phase | 1O.12 | 2O.12 | 1O.14 | 2O.14 |
|---|---|---|---|---|---|
| $C_1$ | Isotropic | 7.94 | 8.12 | 8.84 | 7.79 |
| | Nematic | 7.23 | 7.33 | 8.35 | 7.57 |
| | Smectic-A | | | | 7.46 |
| $V^{\sim}$ | Isotropic | 1.26 | 1.24 | 1.20 | 1.27 |
| | Nematic | 1.33 | 1.32 | 1.23 | 1.29 |
| | Smectic-A | | | | 1.30 |
| $V^{\sim I}1$ | Isotropic | 6.00 | 5.9 | 5.2 | 6.27 |
| | Nematic | 7.82 | 7.6 | 5.7 | 6.96 |
| | Smectic-A | | | | 6.96 |
| I | Isotropic | −0.52 | −0.54 | −0.56 | −0.52 |
| | Nematic | −0.47 | −0.48 | −0.54 | −0.50 |
| | Smectic-A | | | | −0.46 |
| $X^{'}$ | Isotropic | −1.61 | −1.59 | −1.47 | −1.65 |
| | Nematic | −1.84 | −1.82 | −1.55 | −1.73 |
| | Smectic-A | | | | −1.77 |
| $F$ | Isotropic | 1.11 | 1.15 | 1.25 | 1.08 |
| | Nematic | 0.91 | 0.94 | 1.18 | 1.02 |
| | Smectic-A | | | | 0.98 |
| $\tau$ | Isotropic | 3.63 | 3.65 | 3.73 | 3.62 |
| | Nematic | 3.57 | 3.58 | 3.67 | 3.59 |
| | Smectic-A | | | | 3.58 |
| $F$ | Isotropic | 0.22 | 0.22 | 0.21 | 0.22 |
| | Nematic | 0.22 | 0.22 | 0.21 | 0.22 |
| | Smectic-A | | | | 0.22 |
| $\Delta$ | Isotropic | 89.4 | 94.3 | 95.6 | 89.4 |
| | Nematic | 77.4 | 82.5 | 90.6 | 85.7 |
| | Smectic-A | | | | 82.9 |
| $\tau_P$ | Isotropic | 3.80 | 3.89 | 4.25 | 3.72 |
| | Nematic | 3.40 | 3.50 | 4.00 | 3.61 |
| | Smectic-A | | | | 3.56 |



may be attributed to the difference in $dM_V/dT$ data from compound to compound. However, it has been observed that the $L_f$ values exhibit no such trend. This may be attributed to large value of $Y$ (18b).

The $L_f$ value shows practically no variation in a particular phase in a compound. However, it increases with the decrease of temperature, exhibiting a peak as in the case of $\alpha$ with pretransitional effects. The high value in liquid-crystalline phases compared to that in isotropic phase is along expected lines as the molecular order increases in LC phases. These features are illustrated in Table 6 and Fig. 4 for the case of compounds 1O.12, 2O.12, 1O.14, and 2O.14. Because the $L_f$ values obtained from these methods do not vary much from one another (the differences between these values is about $<3\%$) and because the variation with temperature is similar,

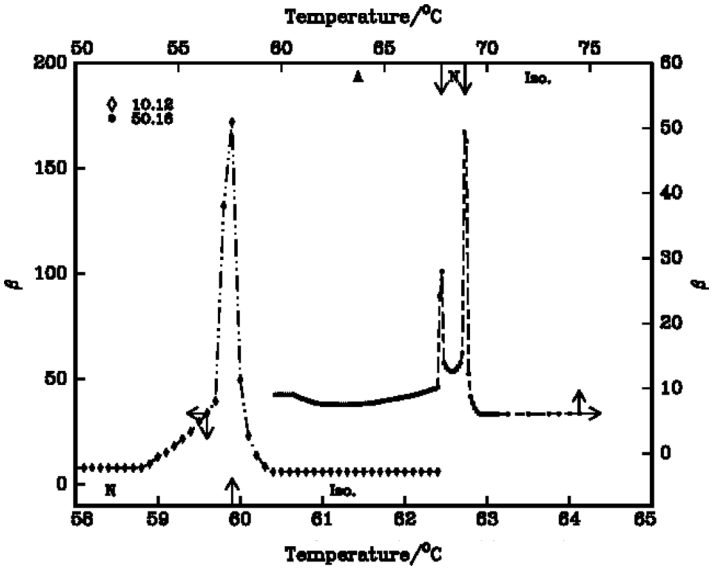



**Figure 3.** Temperature variation of $V_1^{\sim C} = \beta$ in 1O.12 and 5O.16 compounds.



the $L_f$ values estimated from Eq. (19) are presented in Table 6 and Fig. 4 as a representative case.

Tables 3 and 4, which present data for all nO.m compounds, reveal that $L_f$ exhibits an even–odd effect like other thermodynamic parameters and this is depicted in Fig. 5 for the case of 5O.m and nO.14 series. From Fig. 5, it is inferred that the 5O.m series exhibits an odd–even effect, and nO.14 homologues show an even–odd effect similar to that obtained for other thermodynamic parameters. Further, how $L_f$ varies in isotropic, nematic, and smectic-A phases in 5O.m series is shown in Fig. 6. Like the case of isotropic, the $L_f$ exhibits an odd–even effect in nematic and smectic-A phases also. The $L_f$ value is slightly higher in nematic phase compared to isotropic phase. However, in smectic-A phase the $L_f$ values are slightly higher compared to nematic, whereas they are equal in the case of 5O.12, and less in smectic-A than nematic phase for 5O.14, and 5O.16 compounds. The lower values in the case of the last two compounds and the identical value in the case of 5O.12 can be attributed to the interdigitation of the molecules in the compounds in smectic-A phase. Pisipati and Rananavare reported [29] from their X-ray investigations on these compounds that the interdigitation starts from the 5O.12 compound and completes in 5O.16. The same tendency is exhibited by these compounds in $L_f$ value also.

### *Molecular Radius, $M_r$*

Molar radius, $M_r$, is calculated from density data using the relation given in Eq. (25). Further, molar radius of a molecule can be obtained from ultrasonic in conjunction with density data using the different expressions given in Eqs. (21) to (24). Molecular radius is estimated for all compounds using the density results. However, for 5O.m compounds the same is obtained using sound velocity available in the literature.

**Table 3.** Molecular free length, $L_f$ (Å), and other thermodynamic parameters of nO.m compounds at $T_{IN} + 5°C$

| nO.m | $T$ (°C) | $S^*$ | $k''$ | $k$ | $k'$ | $V_a$ | $V_0$ | $V_0$ | $L_f$ | $L_f$ |
|---|---|---|---|---|---|---|---|---|---|---|
| 4O.4 | 79.4 | 1.385 | 0.068 | 3.590 | 3.658 | 69.44 | 254.01 | 260.56 | 0.848 | 0.834 |
| 4O.5 | 86.1 | 1.373 | 0.030 | 3.638 | 3.669 | 73.26 | 268.78 | 276.92 | 0.862 | 0.845 |
| 4O.6 | 81.0 | 1.378 | 0.044 | 3.621 | 3.665 | 76.19 | 279.22 | 287.24 | 0.874 | 0.857 |
| 4O.7 | 87.8 | 1.361 | −0.014 | 3.695 | 3.681 | 79.60 | 293.02 | 303.32 | 0.884 | 0.864 |
| 4O.8 | 83.2 | 1.399 | 0.110 | 3.537 | 3.647 | 83.36 | 303.98 | 310.26 | 0.903 | 0.891 |
| 4O.9 | 85.2 | 1.406 | 0.130 | 3.512 | 3.641 | 86.92 | 316.49 | 322.20 | 0.917 | 0.906 |
| 4O.10 | 81.9 | 1.507 | 0.353 | 3.236 | 3.589 | 91.86 | 329.68 | 323.90 | 0.943 | 0.954 |
| 4O.12 | 81.7 | 1.497 | 0.336 | 3.256 | 3.592 | 98.19 | 352.71 | 347.68 | 0.963 | 0.973 |
| 4O.14 | 79.5 | 1.466 | 0.275 | 3.330 | 3.605 | 99.23 | 357.74 | 356.45 | 0.964 | 0.967 |
| 4O.16 | 75.0 | 1.378 | 0.044 | 3.620 | 3.665 | 105.01 | 384.82 | 395.84 | 0.972 | 0.954 |
| 5O.1 | 67.4 | 1.363 | −0.005 | 3.684 | 3.679 | 61.08 | 224.72 | 232.42 | 0.809 | 0.791 |
| 5O.2 | 64.0 | 1.337 | −0.105 | 3.813 | 3.708 | 64.18 | 238.01 | 248.59 | 0.819 | 0.795 |
| 5O.5 | 82.8 | 1.421 | 0.171 | 3.460 | 3.631 | 76.34 | 277.16 | 280.59 | 0.880 | 0.872 |
| 5O.6 | 77.9 | 1.397 | 0.103 | 3.545 | 3.648 | 79.41 | 289.72 | 295.94 | 0.888 | 0.876 |
| 5O.7 | 83.1 | 1.422 | 0.172 | 3.459 | 3.630 | 83.68 | 303.79 | 307.52 | 0.907 | 0.900 |
| 5O.8 | 81.0 | 1.358 | −0.025 | 3.710 | 3.685 | 86.14 | 317.38 | 328.95 | 0.907 | 0.885 |
| 5O.9 | 78.8 | 1.410 | 0.142 | 3.496 | 3.638 | 90.55 | 329.42 | 334.82 | 0.930 | 0.92 |
| 5O.10 | 81.2 | 1.354 | −0.038 | 3.726 | 3.688 | 92.79 | 342.21 | 355.13 | 0.929 | 0.906 |
| 5O.12 | 78.9 | 1.601 | 0.490 | 3.077 | 3.567 | 101.93 | 363.61 | 346.43 | 0.980 | 1.010 |
| 5O.14 | 76.5 | 1.382 | 0.057 | 3.604 | 3.661 | 109.08 | 399.35 | 410.20 | 0.985 | 0.968 |
| 5O.16 | 73.9 | 1.409 | 0.139 | 3.500 | 3.639 | 117.28 | 426.76 | 433.92 | 1.013 | 1.000 |
| 6O.2 | 73.4 | 1.347 | −0.066 | 3.762 | 3.696 | 68.12 | 251.82 | 262.04 | 0.837 | 0.815 |
| 6O.4 | 82.3 | 1.384 | 0.064 | 3.595 | 3.659 | 76.28 | 279.13 | 286.47 | 0.875 | 0.860 |
| 6O.5 | 95.0 | 1.550 | 0.422 | 3.155 | 3.577 | 81.43 | 291.26 | 282.09 | 0.908 | 0.927 |
| 6O.6 | 94.0 | 1.478 | 0.299 | 3.301 | 3.600 | 84.71 | 304.96 | 302.63 | 0.916 | 0.920 |
| 6O.7 | 84.9 | 1.450 | 0.240 | 3.373 | 3.613 | 87.10 | 314.71 | 315.34 | 0.922 | 0.921 |
| 6O.8 | 87.2 | 1.494 | 0.330 | 3.264 | 3.594 | 91.17 | 327.61 | 323.31 | 0.795 | 0.948 |
| 7O.1 | 77.9 | 1.379 | 0.048 | 3.616 | 3.664 | 68.58 | 251.26 | 258.37 | 0.844 | 0.828 |
| 7O.2 | 74.6 | 1.591 | 0.479 | 3.090 | 3.569 | 73.84 | 263.52 | 251.80 | 0.880 | 0.907 |
| 7O.3 | 85.5 | 1.463 | 0.268 | 3.339 | 3.607 | 77.30 | 278.80 | 278.12 | 0.887 | 0.889 |
| 7O.4 | 81.6 | 1.473 | 0.289 | 3.313 | 3.602 | 80.92 | 291.50 | 289.76 | 0.902 | 0.905 |
| 7O.5 | 88.2 | 1.340 | −0.093 | 3.797 | 3.705 | 83.26 | 308.45 | 321.79 | 0.893 | 0.868 |
| 7O.6 | 85.0 | 1.382 | 0.058 | 3.603 | 3.661 | 86.62 | 317.11 | 325.70 | 0.912 | 0.896 |
| 7O.7 | 88.8 | 1.522 | 0.379 | 3.205 | 3.584 | 91.79 | 328.98 | 321.56 | 0.943 | 0.958 |
| 7O.8 | 88.0 | 1.385 | 0.068 | 3.591 | 3.658 | 93.82 | 343.19 | 352.08 | 0.937 | 0.922 |
| 7O.9 | 89.0 | 1.371 | 0.022 | 3.648 | 3.671 | 96.97 | 355.97 | 367.06 | 0.946 | 0.926 |
| 7O.10 | 87.8 | 1.394 | 0.094 | 3.558 | 3.651 | 100.96 | 368.61 | 376.97 | 0.962 | 0.948 |
| 8O.4 | 85.2 | 1.378 | 0.045 | 3.620 | 3.665 | 83.59 | 306.33 | 315.10 | 0.901 | 0.884 |
| 8O.5 | 89.6 | 1.536 | 0.402 | 3.178 | 3.580 | 88.53 | 316.94 | 308.33 | 0.933 | 0.950 |
| 8O.6 | 91.0 | 1.519 | 0.374 | 3.211 | 3.585 | 92.11 | 330.21 | 323.10 | 0.944 | 0.958 |
| 8O.7 | 91.9 | 1.517 | 0.371 | 3.214 | 3.585 | 95.73 | 343.22 | 336.02 | 0.956 | 0.970 |
| 8O.8 | 91.2 | 1.490 | 0.324 | 3.271 | 3.595 | 98.52 | 354.17 | 349.90 | 0.964 | 0.972 |
| 8O.9 | 92.4 | 1.430 | 0.192 | 3.433 | 3.625 | 102.15 | 370.32 | 373.78 | 0.970 | 0.964 |

(*Continued*)

**Table 3.** Continued

| nO.m | $T$ (°C) | $S^*$ | $k''$ | $k$ | $k'$ | $V_a$ | $V_0$ | $V_0$ | $L_f$ | $L_f$ |
|---|---|---|---|---|---|---|---|---|---|---|
| 8O.10 | 91.6 | 1.456 | 0.253 | 3.357 | 3.610 | 105.70 | 381.61 | 381.61 | 0.984 | 0.984 |
| 9O.4 | 90.9 | 1.433 | 0.200 | 3.423 | 3.623 | 87.35 | 316.49 | 319.08 | 0.921 | 0.916 |
| 9O.5 | 97.7 | 1.406 | 0.129 | 3.512 | 3.642 | 90.72 | 330.37 | 336.35 | 0.930 | 0.919 |
| 9O.6 | 95.0 | 1.491 | 0.324 | 3.271 | 3.595 | 95.11 | 341.89 | 337.75 | 0.953 | 0.961 |
| 9O.8 | 95.5 | 1.467 | 0.277 | 3.328 | 3.605 | 101.63 | 366.37 | 364.96 | 0.972 | 0.975 |
| 9O.9 | 96.9 | 1.439 | 0.215 | 3.405 | 3.620 | 104.66 | 378.84 | 381.10 | 0.979 | 0.975 |
| 9O.10 | 93.9 | 1.466 | 0.274 | 3.331 | 3.605 | 108.35 | 390.65 | 389.28 | 0.993 | 0.995 |
| 9O.12 | 95.8 | 1.504 | 0.348 | 3.242 | 3.590 | 115.69 | 415.31 | 408.48 | 1.018 | 1.030 |
| 9O.14 | 91.5 | 1.480 | 0.303 | 3.296 | 3.599 | 122.20 | 439.81 | 436.14 | 1.035 | 1.040 |
| 1O.O1 | 79.0 | 1.256 | −0.553 | 4.403 | 3.850 | 76.13 | 293.12 | 315.70 | 0.845 | 0.842 |
| 1O.O6 | 89.2 | 1.549 | 0.421 | 3.156 | 3.577 | 99.01 | 354.13 | 343.09 | 0.969 | 0.989 |
| 1O.O8 | 94.8 | 1.530 | 0.392 | 3.189 | 3.582 | 110.46 | 395.62 | 385.64 | 1.004 | 1.021 |
| 1O.O9 | 96.1 | 1.591 | 0.478 | 3.091 | 3.569 | 114.36 | 408.15 | 390.10 | 1.018 | 1.049 |
| 1O.O10 | 93.9 | 1.393 | 0.092 | 3.559 | 3.651 | 115.60 | 422.09 | 431.73 | 1.006 | 0.991 |
| 1O.O12 | 90.1 | 1.561 | 0.439 | 3.135 | 3.574 | 124.24 | 444.03 | 428.43 | 1.046 | 1.107 |
| 1O.O16 | 90.8 | 1.502 | 0.345 | 3.246 | 3.590 | 128.27 | 460.55 | 453.21 | 1.053 | 1.106 |
| 1O.12 | 64.9 | 1.419 | 0.165 | 3.467 | 3.632 | 86.86 | 315.48 | 319.64 | 0.918 | 0.910 |
| 2O.12 | 81.2 | 1.378 | 0.046 | 3.619 | 3.664 | 88.26 | 323.41 | 332.62 | 0.918 | 0.900 |
| 4O.12 | 81.7 | 1.497 | 0.336 | 3.256 | 3.592 | 98.19 | 352.71 | 347.68 | 0.963 | 0.972 |
| 5O.12 | 78.9 | 1.601 | 0.490 | 3.077 | 3.567 | 101.93 | 363.61 | 346.43 | 0.980 | 1.012 |
| 1O.O12 | 90.1 | 1.561 | 0.439 | 3.135 | 3.574 | 124.24 | 444.03 | 428.43 | 1.046 | 1.070 |
| 1O.14 | 65.3 | 1.320 | −0.181 | 3.912 | 3.731 | 92.08 | 343.58 | 361.20 | 0.919 | 0.889 |
| 2O.14 | 75.1 | 1.446 | 0.230 | 3.386 | 3.616 | 96.46 | 348.79 | 350.05 | 0.954 | 0.951 |
| 4O.14 | 79.5 | 1.466 | 0.275 | 3.330 | 3.605 | 99.23 | 357.74 | 356.45 | 0.964 | 0.966 |
| 5O.14 | 76.5 | 1.382 | 0.057 | 3.604 | 3.661 | 109.08 | 399.35 | 410.20 | 0.985 | 0.967 |
| 7O.14 | 82.2 | 1.473 | 0.429 | 3.312 | 3.741 | 111.89 | 418.62 | 401.06 | 0.942 | 1.000 |
| 8O.14 | 87.7 | 1.553 | 0.221 | 3.149 | 3.369 | 124.95 | 420.99 | 430.50 | 1.011 | 1.073 |
| 9O.14 | 88.3 | 1.438 | 0.317 | 3.408 | 3.725 | 119.15 | 443.87 | 435.91 | 0.930 | 1.015 |
| 1O.O14 | 88.2 | 1.486 | 0.160 | 3.282 | 3.441 | 131.17 | 451.39 | 463.41 | 1.092 | 1.072 |
| 11O.14 | 93.6 | 1.416 | 0.204 | 3.477 | 3.682 | 127.83 | 470.62 | 472.60 | 1.003 | 1.032 |
| 12O.14 | 92.5 | 1.434 | 0.268 | 3.419 | 3.687 | 127.25 | 469.19 | 465.96 | 1.032 | 1.037 |
| 13O.14 | 92.9 | 1.462 | 0.553 | 3.341 | 3.894 | 127.64 | 497.05 | 457.51 | 0.976 | 1.053 |
| 14O.14 | 93.5 | 1.651 | 0.272 | 3.016 | 3.288 | 149.89 | 492.87 | 503.57 | 1.119 | 1.159 |
| 15O.14 | 93.6 | 1.455 | 0.277 | 3.361 | 3.638 | 140.98 | 512.84 | 509.83 | 1.078 | 1.082 |
| 16O.14 | 92.9 | 1.467 | 0.553 | 3.328 | 3.881 | 137.00 | 531.73 | 489.68 | 1.022 | 1.080 |
| 18O.14 | 93.8 | 1.652 | 0.548 | 3.015 | 3.563 | 153.48 | 546.85 | 512.83 | 1.124 | 1.173 |
| 12O.12 | 93.0 | 1.473 | 0.290 | 3.312 | 3.602 | 126.58 | 455.94 | 453.15 | 1.047 | 1.050 |
| 12O.14 | 92.5 | 1.434 | 0.268 | 3.419 | 3.687 | 127.25 | 469.19 | 465.96 | 1.032 | 1.037 |
| 12O.16 | 90.6 | 1.486 | 0.315 | 3.282 | 3.597 | 135.96 | 489.02 | 483.91 | 1.073 | 1.080 |

*Molecular Radius from ρ.* The molecular radius for a number of nO.m compounds for different homologues is estimated from Eq. (20). Table 7 illustrates these values. It has been observed that the molecular radius increases with the increase of either $n$ or $m$ number as expected and as reported in a number of liquids [30]. Further, the

**Table 4.** Molecular free length, $L_f$ (Å), in nO.m compounds at $T_{IN} + 5°C$ from volume expansion coefficient from Eq. (19)

| n/m | 1 | 2 | 3 | 4 | 5 | 6 | 7 | 8 | 9 | 10 | 12 | 14 | 16 |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| 1 | | | | | | | | | | | 0.910 | 0.889 | |
| 2 | | | | | | | | | | | 0.900 | 0.951 | |
| 3 | | | | | | | | | | | 0.972 | 0.966 | |
| 4 | | | | 0.834 | 0.845 | 0.857 | 0.864 | 0.891 | 0.906 | 0.954 | 0.973 | 0.967 | 0.954 |
| 5 | 0.791 | 0.795 | | | 0.872 | 0.876 | 0.900 | 0.885 | 0.920 | 0.906 | 1.010 | 0.968 | 1.000 |
| 6 | | 0.815 | | 0.860 | 0.927 | 0.920 | 0.921 | 0.948 | | | | | |
| 7 | 0.828 | 0.907 | 0.889 | 0.905 | 0.868 | 0.896 | 0.958 | 0.922 | 0.926 | 0.948 | | 1.000 | |
| 8 | | | | 0.884 | 0.950 | 0.958 | 0.970 | 0.972 | 0.964 | 0.984 | | 1.073 | |
| 9 | | | | 0.916 | 0.919 | 0.961 | 0.975 | 0.975 | 0.995 | 1.030 | 1.040 | 1.015 | |
| 10 | 0.842 | | | | | 0.989 | | 1.021 | 1.049 | 0.991 | 1.107 | 1.072 | 1.106 |
| 11 | | | | | | | | | | | | 1.032 | |
| 12 | | | | | | | | | | | 1.050 | 1.037 | 1.080 |
| 13 | | | | | | | | | | | | 1.053 | |
| 14 | | | | | | | | | | | | 1.159 | |
| 15 | | | | | | | | | | | | 1.082 | |
| 16 | | | | | | | | | | | | 1.080 | |
| 18 | | | | | | | | | | | | 1.173 | |

**Table 5.** Molar volume at 0 K ($V_0$) and molecular free length ($L_f$) in Å in isotropic phase of 5O.m compounds

| | | From Eq. (19) | | From Eq. (18) | | From extrapolation technique | |
|---|---|---|---|---|---|---|---|
| 5O.m | $T$ (°C) | $V_0^1$ | $L_f$ (Å) | $V_0^2$ | $L_f$ (Å) | $V_0^3$ | $L_f$ (Å) |
| 5O.5 | 82.8 | 280.59 | 0.872 | 277.16 | 0.880 | 322.96 | 0.794 |
| 5O.6 | 77.9 | 295.94 | 0.876 | 289.72 | 0.888 | 337.95 | 0.801 |
| 5O.7 | 83.1 | 307.52 | 0.900 | 303.79 | 0.907 | 351.66 | 0.822 |
| 5O.8 | 81 | 328.95 | 0.885 | 317.38 | 0.907 | 370.15 | 0.818 |
| 5O.9 | 78.8 | 334.82 | 0.920 | 329.42 | 0.930 | 387.37 | 0.834 |
| 5O.10 | 81.2 | 355.13 | 0.906 | 342.21 | 0.929 | 404.62 | 0.830 |
| 5O.12 | 78.9 | 346.43 | 1.010 | 363.61 | 0.980 | 424.11 | 0.884 |
| 5O.14 | 76.5 | 410.20 | 0.968 | 399.35 | 0.985 | 477.18 | 0.874 |
| 5O.16 | 73.9 | 433.92 | 1.000 | 426.76 | 1.013 | 508.02 | 0.902 |

**Table 6.** Magnitude of $L_f$ (Å) in different phases in 1O.12, 2O.12, 1O.14, and 2O.14 compounds

| | Molecular free length $L_f$ (Å) | | |
|---|---|---|---|
| Compound | Isotropic | Nematic | Smectic-A |
| 1O.12 | 0.910 | 0.976 | |
| 2O.12 | 0.902 | 0.973 | |
| 1O.14 | 0.911 | 0.934 | |
| 2O.14 | 0.951 | 0.989 | 0.997 |



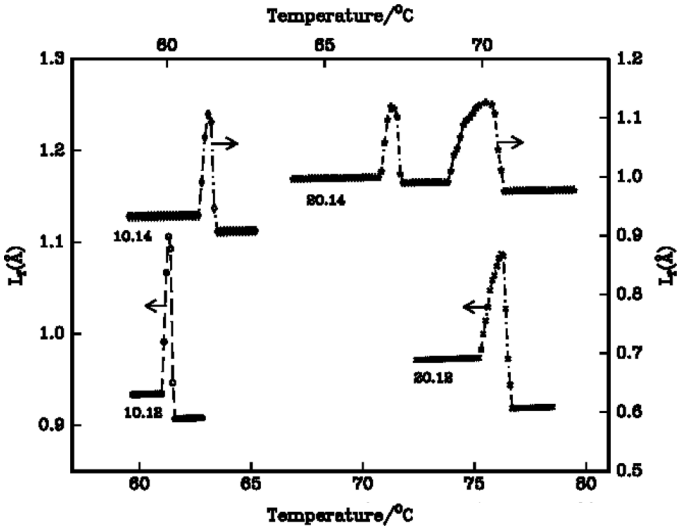



**Figure 4.** Variation of $L_f$ (Å) with temperature in 1O.12, 2O.12, 1O.14, and 2O.14 compounds.

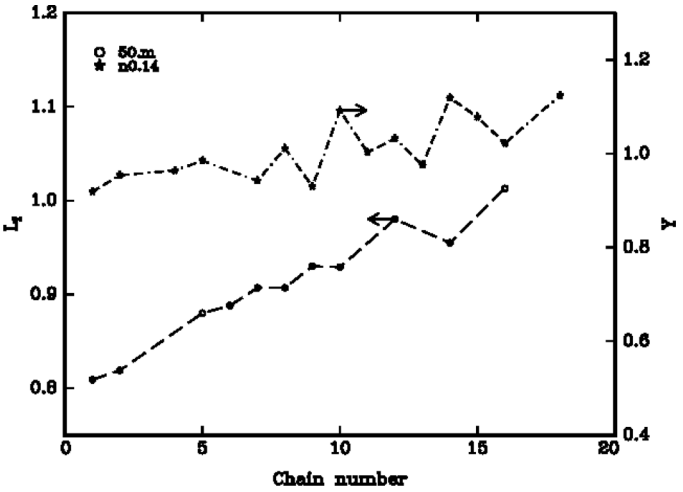



**Figure 5.** Variation of $L_f$ (Å) in 5O.m and nO.14 compounds.



interesting aspect is that the core radius for a particular homologue depends only on the alkoxy chain number rather than constant for the whole nO.m series. This is clearly depicted in Fig. 7 and Table 7. A similar type of observation is made regarding the increment of molecular radius for methylene ($CH_2$) unit also (Table 8).

*Molecular Radius from Sound Velocity.* Molecular radius can also be estimated from sound velocity in conjunction with density from Eqs. (21) to (24). Different authors proposed expressions based on some assumptions for the estimation. Because the sound velocity data for 5O.m compounds are available in literature, only for that series are the $M_r$ values evaluated using above equations. The values are given in Table 9.

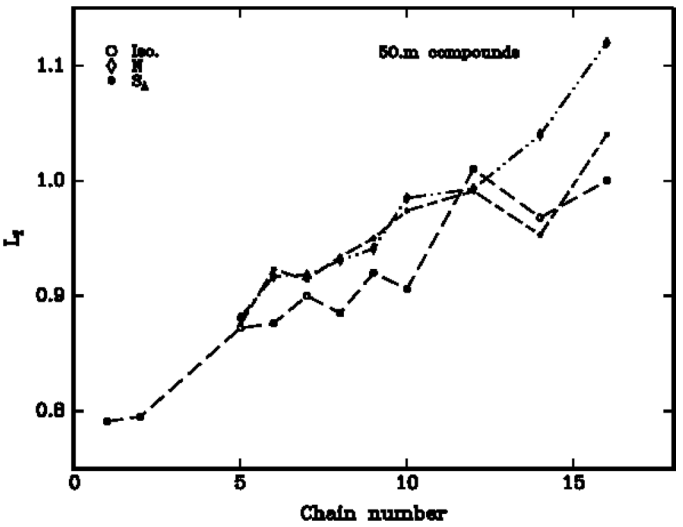



**Figure 6.** Variation of $L_f$ (Å) in different phases of 5O.m compounds.

**Table 7.** Molecular radius (Å) of nO.m compounds from density

| nO.m | $M_r$ (Å) | nO.m | $M_r$ (Å) | nO.m | $M_r$ (Å) | nO.m | $M_r$ (Å) |
|---|---|---|---|---|---|---|---|
| 4O.4 | 4.561 | 6O.2 | 4.545 | 9O.4 | 4.911 | 12O.12 | 5.549 |
| 4O.5 | 4.647 | 6O.4 | 4.707 | 9O.5 | 4.980 | 12O.14 | 5.593 |
| 4O.6 | 4.707 | 6O.5 | 4.782 | 9O.6 | 5.042 | 12O.16 | 5.681 |
| 4O.7 | 4.781 | 6O.6 | 4.853 | 9O.8 | 5.159 | | |
| 4O.8 | 4.844 | 6O.7 | 4.903 | 9O.9 | 5.215 | 1O.14 | 5.037 |
| 4O.9 | 4.910 | 6O.8 | 4.971 | 9O.10 | 5.270 | 2O.14 | 5.074 |
| 4O.10 | 4.982 | | | 9O.12 | 5.381 | 4O.14 | 5.118 |
| 4O.12 | 5.095 | 7O.1 | 4.544 | 9O.14 | 5.484 | 5O.14 | 5.303 |
| 4O.14 | 5.118 | 7O.2 | 4.626 | | | 7O.14 | 5.379 |
| 4O.16 | 5.238 | 7O.3 | 4.710 | 10O.1 | 4.767 | 8O.14 | 5.431 |
| | | 7O.4 | 4.781 | 10O.6 | 5.104 | 9O.14 | 5.487 |
| 5O.1 | 4.377 | 7O.5 | 4.862 | 10O.8 | 5.295 | 10O.14 | 5.550 |
| 5O.2 | 4.459 | 7O.6 | 4.911 | 10O.9 | 5.352 | 11O.14 | 5.600 |
| 5O.5 | 4.698 | 7O.7 | 4.979 | 10O.10 | 5.403 | 12O.14 | 5.593 |
| 5O.6 | 4.766 | 7O.8 | 5.042 | 10O.12 | 5.504 | 13O.14 | 5.680 |
| 5O.7 | 4.844 | 7O.9 | 5.103 | 10O.14 | 5.550 | 14O.14 | 5.735 |
| 5O.8 | 4.910 | 7O.10 | 5.165 | 10O.16 | 5.523 | 15O.14 | 5.767 |
| 5O.9 | 4.976 | | | | | 16O.14 | 5.811 |
| 5O.10 | 5.035 | 8O.5 | 4.918 | 1O.12 | 4.905 | 18O.14 | 5.901 |
| 5O.12 | 5.141 | 8O.6 | 4.985 | 2O.12 | 4.943 | | |
| 5O.14 | 5.303 | 8O.7 | 5.050 | 4O.12 | 5.095 | | |
| 5O.16 | 5.424 | 8O.8 | 5.102 | 5O.12 | 5.141 | | |
| | | 8O.9 | 5.175 | 10O.12 | 5.504 | | |
| | | 8O.10 | 5.229 | 12O.12 | 5.549 | | |



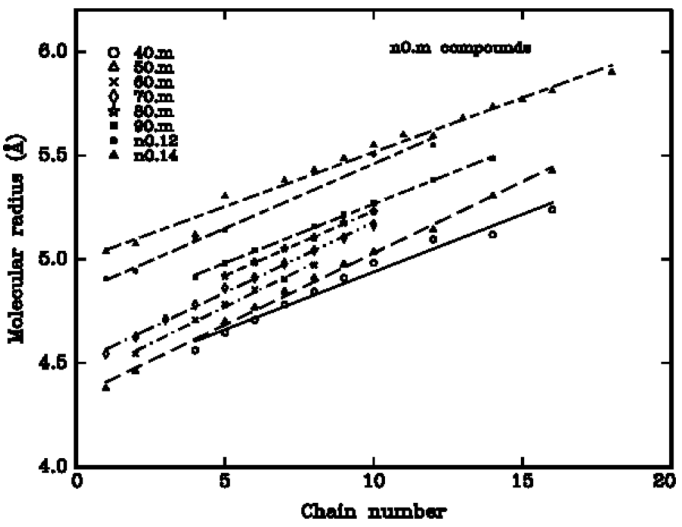



**Figure 7.** Variation of molecular radius, $M_r$ (Å), with either *m* or *n* in different homologues of nO.m compounds.

**Table 8.** Core radius and increment per methylene unit in nO.m series

| nO.m | Increment per $CH_2$ in Å | Core radius in Å |
|---|---|---|
| 4O.m | 0.056 | 4.372 |
| 5O.m | 0.069 | 4.346 |
| 6O.m | 0.705 | 4.421 |
| 7O.m | 0.068 | 4.501 |
| 8O.m | 0.062 | 4.614 |
| 9O.m | 0.057 | 4.705 |
| nO.12 | 0.062 | 4.846 |
| nO.14 | 0.052 | 4.990 |

The $M_r$ values estimated from Schaff [20] and Rao [21] are identical and hence, only the values due to Schaff for 5O.m series are given in Table 9. This is expected because those two expressions are very nearly equal to one another except the term $\gamma$ in the denominator of Eq. (22) due to Rao. The table reveals that the values obtained through Schaff's and Rao's equations are lower compared to those obtained by other two equations due to Kittel as well as from density data. The estimated values due to Erying are very low and hence they are not considered. The values due to Kittel are slightly higher compared to the values due to Kittel and from density are not only in good agreement with one another but also identical. Figure 8 illustrates the variation of $M_r$ with alkyl chain length in 5O.m series estimated from ultrasonic velocity $U$ (due to Schaff); refractive index, $n$; and density, $\rho$.



*Molecular Radius from Refractive Index.* The molecular radius can also be estimated from the refractive index using Eq. (25). The values are estimated for 5O.m series and the data are presented in Table 9 and Fig. 8. The data show that the values calculated using the refractive index are of comparable magnitude with those obtained from sound velocity due to Schaff and Rao. Pandy *et al.* [6] also

**Table 9.** Molecular radius of 5O.m compounds from sound velocity, density, and refractive index

| Method | Schaff's | Rao's | Kittel's | Density | Refractive index |
|---|---|---|---|---|---|
| 5O.1 [35] | 3.048 | The molecular | The molecular | 4.377 | 3.385 |
| 5O.2 [36] | 3.105 | radius is same | radius is same | 4.459 | 3.435 |
| 5O.5 [37] | 3.272 | as in the case | as in the case | 4.699 | 3.570 |
| 5O.6 [37] | 3.320 | due to Schaff | due to density | 4.767 | 3.617 |
| 5O.7 [37] | 3.374 | | | 4.845 | 3.669 |
| 5O.8 [38] | 3.420 | | | 4.911 | 3.715 |
| 5O.9 [39] | 3.465 | | | 4.976 | 3.750 |
| 5O.10 [40] | 3.506 | | | 5.035 | 3.771 |
| 5O.12 [A] | 3.587 | | | 5.141 | 3.884 |

[A]. Pisipati, V. G. K. M. (unpublished work).

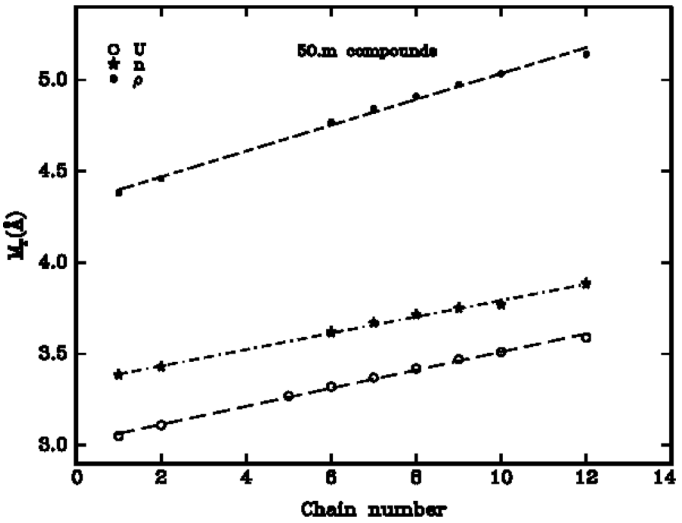



**Figure 8.** Variation of molecular radius, $M_r$ (Å), in 5O.m series estimated from different methods.



reported small values of molecular radius obtained through refractive index in pure and binary mixtures compared to that obtained from density. However, one thing can be pointed out: that these expressions need refinement to get concurrent results.

The salient features from this analysis are

1. All the thermodynamic parameters except the Sharma parameter, $S_0$, exhibit either an odd–even or even–odd effect with respect to $m$ or $n$ number, respectively, in a homologous series. $S_0$ is independent of both temperature and material.
2. Generally all thermodynamic parameters are constant in a particular phase except at the phase transformation, where they exhibit a peak either in the positive or negative direction depending on whether that particular parameter is directly or indirectly proportional to $\alpha T$, respectively. Following the same trend, the magnitude of these parameters is slightly higher or lower in the liquid-crystalline phases, viz. nematic and smectic-A.
3. The molecular free length, $L_f$, also exhibits an odd–even or even–odd effect in a homologous series depending on $n$ or $m$ number and exhibits slightly higher values in nematic and smectic-A phases compared to isotropic phase,
4. The molecular radius, $M_r$, increases with the increase of either $n$ or $m$ in homologous series, but the core radius as well as increment of molecular radius per methylene unit exhibit deviations from homologous series to series instead of constant.
5. The expressions used for the estimation of molecular radius from density, sound velocity, and refractive index need refinement to get concurrent results from these three experimental results.

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