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Professional paper

INVESTIGATING THERMODYNAMIC AND VISCOSITY PROPERTIES OF SIMPLE LIQUIDS VIA ULTRASONIC AND DENSITY DATA

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Abstract

Thermodynamic properties of liquids are very important in order to understand the complexity of molecular interactions in the system. Correlations between various thermodynamic properties of a liquid are very useful in the study of physicochemical and structural behavior, especially when these properties are difficult to determine experimentally. In order to compute various useful and important thermodynamic properties of liquids from ultrasonic velocity and density data, a number of correlations have been presented. The values of ultrasonic velocity (v) and density (ρ) data for four liquids, methanol, n-hexane, toluene, and acetone were obtained through DSA 5000 M Density Meter. They have been measured at the temperature range from (293.15-333.15) K and are used to compute thermal expansion coefficients (α), isothermal compressibility (β_T), isentropic compressibility (β_s), refractive index (n), internal pressure (P_{int}), and heat capacity ratio (γ). To this end, we incorporate the dipole-dipole molecular interaction, and we investigate the viscosity under temperature variations. The relation between sound velocity and viscosity is also discussed. The calculated values are compared with literature data, and the agreement is found to be good.

Keywords: thermodynamics, sound velocity, density, viscosity, isothermal compressibility, heat capacity ratio

1. Introduction

Thermodynamic properties of pure liquids and liquid mixtures are very important for understanding the physicochemical behavior of such liquids. The study of thermodynamic properties of liquids is of both practical and theoretical interest. These properties can be determined using derived relations based on Flory's statistical theory, the hard sphere equation of state, and hole theory (Flory *et al*., 1964, Abe & Flory, 1965, Lebowitz, Frisch & Helfand, 1961, Carnahan & Starling, 1969, Frisch, 1964, Hoover & Ree, 1964, Pandey & Sanguri, 2001, Pandey & Sanguri, 2008, Pandey, Sanguri & Bhatt, 2003, Pandey, Sanguri & Dwivedi, 2008, Pandey, Sanguri, Dwivedi & Tiwari, 2007). On the other hand, ultrasonic velocity and density data, which can be obtained experimentally relatively easily, were used to study many equilibrium and transport properties (Acosta *et al*., 2001, Ali, Nain, Sharma & Ahmad, 2000, Canosa *et al*., 2001).

On the other hand, some empirical and semi-empirical procedures have been used to determine theoretically sound velocity in such liquids or liquid mixtures (Eyring & John, 1969, Pandey *et al*., 1999; Ali & Nain, 2000). A comparative study of Flory's statistical theory, the hard sphere equation of state, and hole theory, based on theoretically estimated values of certain thermodynamic properties of pure and binary systems, was conducted by Pandey, Chandra Prakash, Sethi, and Sanguri (2012). In particular, ultrasonic velocity and density data have been used to estimate several thermodynamic parameters such are thermal expansivity (α),

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isothermal compressibility (β_T), heat specific ratio (γ), internal pressure (P_{int}), and others for aqueous methyl orange solution (Pandey *et al*., 2019).

On the other hand, viscosity is a well-known property that, in simple terms, measures a fluid's resistance to deformation or flow—essentially indicating how thick or thin the fluid is. It plays a critical role in engineering applications, particularly in chemical engineering, as it directly affects fluid dynamics, as well as heat and mass transfer processes. From a microscopic perspective, viscosity arises from internal friction between molecules in motion under the influence of external forces. This internal friction is primarily governed by molecular interactions and temperature. One of the most widely recognized theoretical approaches to describing viscosity is the Eyring viscosity equation, which is based on the concept of activation energy (Eyring & John, 1969). A natural question that arises is whether there exists a relationship between viscosity and sound velocity in liquids. In the present work, we explore this possibility by utilizing an empirical equation proposed in (Parthasarathy & Bakhshi, 1953).

In Section 2, we present experimental measurements of ultrasonic velocity and density for four liquids. Based on these data, we calculate key thermodynamic quantities and examine their dependence on temperature.

In Section 3, we analyze the viscosity of these liquids by incorporating dipole–dipole interaction terms and explore a possible empirical relationship between viscosity and sound velocity. In Section 4, we summarize and discuss our main findings.

2. Calculating thermodynamic quantities using the ultrasonic velocity and density data

Extracting numerical values for thermodynamic quantities—such as the thermal expansion coefficient, isothermal compressibility, isentropic compressibility, internal pressure, and heat capacity ratio—is not straightforward from a practical standpoint and cannot be easily achieved using standard tools or measurement instruments. In this section, we demonstrate that these thermodynamic properties can, in fact, be probed and quantitatively estimated using experimental data based on ultrasonic velocity and density.

2.1 Experimental setup: To measure the ultrasonic velocity and density of our samples, we used the DSA 5000 M Density Meter. These measurements were conducted over a temperature range of 293.15 K to 333.15 K. In this study, we examined four simple liquids: methanol, toluene, acetone, and n-hexane. Our objective is to utilize the ultrasonic velocity and density data to calculate various thermodynamic quantities and investigate their relationships with key thermodynamic parameters.

We present our experimental results in Tables 1-4.

Table 1. Experimental data for methanol

Temperature (K)	Refractive Index	Density (kg/m^3)	Speed of Sound (m/s)
293.15	1.32999	791.633	1120.36
298.15	1.32699	786.919	1103.89
303.15	1.32470	782.213	1087.51
313.15	1.32021	772.698	1055.10
323.15	1.31478	763.061	1022.74

Table 2. Experimental data for acetone

Temperature (K)	Refractive Index	Density (kg/m³)	Speed of Sound (m/s)
293.15	1.35982	790.735	1185.95
298.15	1.35701	785.004	1163.52
303.15	1.35346	779.265	1141.20
313.15	1.34754	767.608	1096.56

Table 3. Experimental data for toluene

Temperature (K)	Refractive Index	Density (kg/m³)	Speed of Sound (m/s)
293.15	1.49520	866.778	1327.76
298.15	1.49222	862.125	1306.04
303.15	1.48922	857.462	1284.26
313.15	1.48313	848.093	1241.36
323.15	1.47713	838.656	1199.35

Table 4. Experimental data for n-hexane

Temperature (K)	Refractive Index	Density (kg/m³)	Speed of Sound (m/s)
293.15	1.37664	659.707	1100.87
298.15	1.37297	655.160	1078.31
303.15	1.36936	650.625	1055.90
313.15	1.36340	641.384	1010.70
323.15	1.35730	631.942	967.290

2.2 Thermodynamic properties: Having the experimental data, we shall now continue and focus on the thermodynamic properties. Let us first mention that there is only one thermodynamic property, namely the isentropic compressibility, which can be directly determined using ultrasonic and density data. Based on dimensional analysis between ultrasound velocity *(v)* - density of liquids (ρ) and thermodynamics properties several empirical relations have been obtained (Pandey and Verma, 2001, Pandey, Dey and Upadhaya, 1997, Sanguri, Chhabra, Srivastava and Pandey, 2015, Marcus, 2013, Pandey, Chhabra, Dey, Sanguri and Verma, 2000). First, the thermal expansivity given by the relation

$$\alpha = \frac{75.6\times10^{-3}}{T^{1/9}v^{1/2}\rho^{1/3}}, \tag{1}$$

Thermal compressibility

$$\beta_T = \frac{1.71\times10^{-3}}{T^{4/9}v^{2}\rho^{4/3}}, \tag{2}$$

Internal pressure

$$P_{\text{int}} = \frac{\alpha}{\beta_T} - P_{ext} = 44.2\times T^{4/3}v^{3/2}\rho, \tag{3}$$

where P_{ext} is the external pressure, provide $P_{ext} = 0$, we get the above equation. Finally, we also have the heat-specific ratio

$$\gamma = \frac{C_P}{C_V} = \frac{\beta_T}{\beta_S} = \frac{17.1\times10^{-3}}{T^{4/9}\cdot\rho^{1/3}}. \tag{4}$$

Using the density and velocity data presented in Tables 1–4, we provide numerical values of the thermodynamic quantities at a specific temperature presented in Tables 4–8. From the numerical values obtained, we conclude that the thermal expansion coefficient, isothermal compressibility, and isentropic compressibility all increase with the increase of temperature. Among the studied liquids, n-hexane exhibits the highest values for these properties, followed by methanol and acetone, with the lowest values observed for toluene. An interesting trend is observed for the specific heat ratio and internal pressure, which decrease as the temperature increase.

Table 4. Numerical values of thermodynamic quantities for methanol

Temperature (K)	Thermal expansivity (10^{-4} K^{-1})	Isothermal compressibility (10^{-14} Pa^{-1})	Internal pressure (10^{12} Pa)	Heat capacity ratio	Isentropic coefficient (10^{-9} Pa^{-1})
293.15	1.29881	1.48970	2.55526	0.14803	1.00638
298.15	1.30861	1.53517	2.54090	0.14721	1.04284
303.15	1.31863	1.58272	2.52507	0.14642	1.08096
313.15	1.33937	1.68464	2.48910	0.14491	1.16253
323.15	1.36133	1.79789	2.44620	0.14350	1.25288

Table 5. Numerical values of thermodynamic quantities for acetone

Temperature (K)	Thermal expansivity (10^{-4} K^{-1})	Isothermal compressibility (10^{-14} Pa^{-1})	Internal pressure (10^{12} Pa)	Heat capacity ratio	Isentropic coefficient (10^{-10} Pa^{-1})
283.15	1.26287	1.33149	2.77975	0.14808	8.99159
293.15	1.27568	1.38634	2.74285	0.14733	9.40980
303.15	1.28886	1.44455	2.70413	0.14660	9.85352
313.15	1.31670	1.57346	2.61987	0.14523	10.8342

Table 6. Numerical values of thermodynamic quantities for toluene

Temperature (K)	Thermal expansivity (10^{-4} K^{-1})	Isothermal compressibility (10^{-14} Pa^{-1})	Internal pressure (10^{12} Pa)	Heat capacity ratio	Isentropic coefficient (10^{-10} Pa^{-1})
283.15	1.15755	0.93985	3.60963	0.14362	6.54415
293.15	1.16703	0.97104	3.58240	0.14280	6.80012
303.15	1.17684	1.00410	3.55217	0.14200	7.07097
313.15	1.19707	1.07494	3.48644	0.14048	7.65175
323.15	1.21814	1.15266	3.41427	0.13905	8.28942

Table 8. Numerical values of thermodynamic quantities for n-hexane

Temperature (K)	Thermal expansivity (10^{-4} K^{-1})	Isothermal compressibility (10^{-14} Pa^{-1})	Internal pressure (10^{12} Pa)	Heat capacity ratio	Isentropic coefficient (10^{-9} Pa^{-1})
283.15	1.39235	1.96746	2.07410	0.15730	1.25077
293.15	1.40744	2.05414	2.04236	0.15648	1.31270
303.15	1.42296	2.14627	2.00939	0.15569	1.37855
313.15	1.45613	2.35345	1.93706	0.15419	1.52629
323.15	1.49060	2.58438	1.86340	0.15281	1.69126

Furthermore, to clearly and better illustrate these trends, we also present the interpolation plots of the same thermodynamic quantities as functions of temperature presented in Figures 1–2.

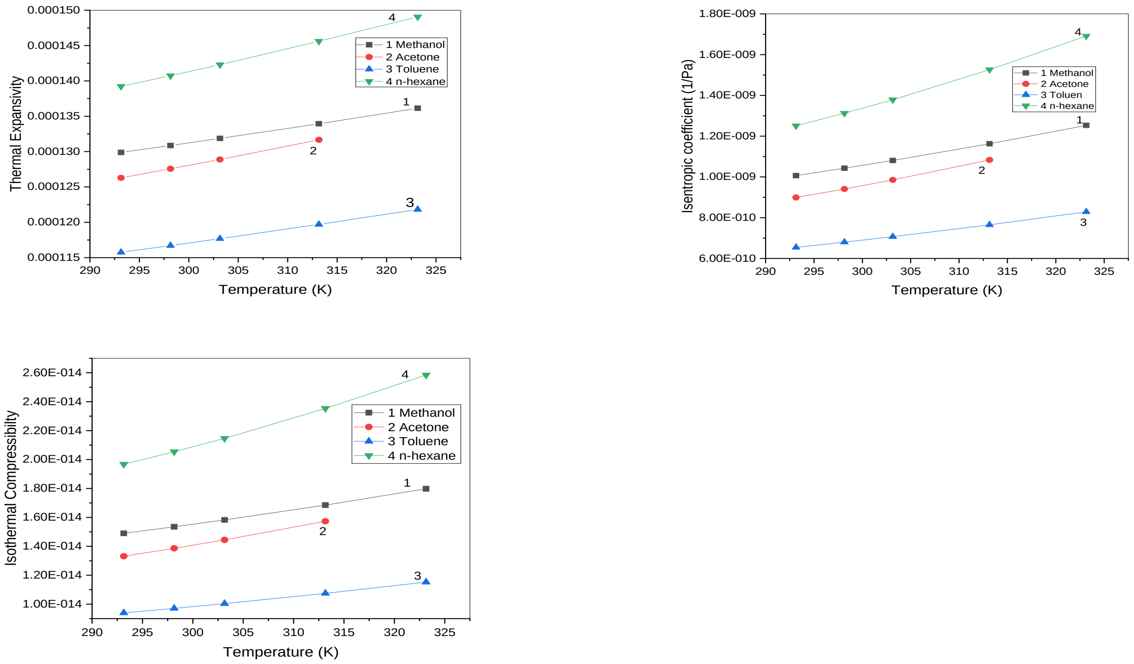

Figure 1 Plots of thermal expansivity, thermal compressibility and isentropic coefficient, as a function of temperature

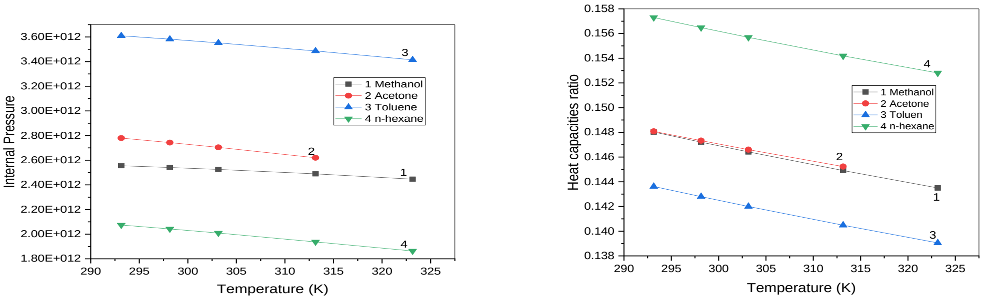

Figure 2 Plots of internal pressure and heat capacity as a function of temperature

3. Viscosity of polar liquids: relating viscosity and sound speed

In this section, we examine the behavior of viscosity under temperature variations for polar molecules. One of the most widely recognized theoretical frameworks for describing viscosity is the Eyring viscosity theory, also known as the Andrade equation. However, in our case—where we study both polar molecules such as methanol and acetone, both with polarity index 5.1, and less polar molecules such as n-hexane (0.1) and toluene (2.4)—we adopt a modified viscosity model that explicitly incorporates dipole–dipole interaction effects. To achieve this, we follow the approach proposed by (Girifalco, 1955), and more recently, extended by (Han, Fang & Chen, 2010). Starting from the Lennard-Jones potential model (Han, Fang & Chen, 2010), which is expressed as:

$$\varphi(r) = 4\varepsilon[(\frac{\sigma}{r})^{12} - (\frac{\sigma}{r})^{6}], \tag{5}$$

where r denotes the effective distance between a pair of molecules, ε is a characteristic energy, actually the maximum energy of attraction between a pair of molecules. Finally, σ is a characteristic diameter of the molecule, often called the collision diameter. For polar molecules, due to the dipole-dipole interaction, we have to consider the electrical dipole potential. By taking the average dipole-dipole intermolecular potential the total potential reads (Han, Fang & Chen, 2010)

$$\varphi(r) = 4\varepsilon[(\frac{\sigma}{r})^{12} - (\frac{\sigma}{r})^{6}] - (\frac{1}{(4\pi\varepsilon_0)^2}\cdot\frac{2}{3kT})\cdot\frac{p_1^2 p_2^2}{r^6}$$

(6)

where p_1 and p_2 are the dipole moments of the molecules. For pure liquids, we can write in general $p_1 = p_2 = p$. Without going into details, one can now show that the viscosity of polar molecules is given by (Han, Fang & Chen, 2010)

$$\eta = \frac{N_A h}{V}\cdot\exp(\frac{5.14\varepsilon}{kT} - \frac{f}{2})\cdot\exp(\frac{1}{(4\pi\varepsilon_0)^2}\cdot\frac{5.635p^4}{3k^2T^2\sigma^6}). \tag{7}$$

Let us now define the three parameters:

$$\eta_0 = \frac{N_A h}{V}\exp(-\frac{f}{2}),\ A = \frac{5.14\varepsilon}{k},\quad C = \frac{1}{(4\pi\varepsilon_0)^2}\cdot\frac{5.635p^4}{3k^2\sigma^6}, \tag{8}$$

where f is the number of degrees of freedom. Finally, we obtain a simple law of viscosity (Girifalco, 1955):

$$\eta = \eta_0 \exp(\frac{A}{T} + \frac{C}{T^2}). \tag{9}$$

As a special case, by setting C=0, we obtain the Andrade equation, $\eta = \eta_0 \exp(A/T)$. To fit the above viscosity model, we shall use the data for our liquids presented in (Kaye & Laby, 2025), for methanol and acetone, (Santos, Nieto de Castro, Dymond, Dalaouti, Assael & Nagashima, 2006) for toluene, and the reference (Michailidou, Assael, Huber & Perkins, 2013), for n-hexane. The fitting of viscosity with a temperature dependence is given in Figs. 3-4. We also present the numerical values from the fitting in Table 9.

Table 9. Numerical values for parameters using the viscosity law of Andrade and Girifalco.

Substance	Model	R^2	η_0	A	C
Methanol	Andrade	0.99974	0.0083±0.0002	1246.5792 ± 8.3228	-
Methanol	Girifalco	0.99987	0.0050 ± 0.0011	1526.8171 ± 129.9530	-39268.9706 ±18628.4393
Acetone	Andrade	0.99793	0.0169 ± 0.0013	864.8360 ± 22.7692	-

Acetone	Girifalco	0.99955	0.0045 ± 0.0022	1680.1449 ± 300.0052	-125422.4494 ±47044.3468
Toluene	Andrade	0.99535	5.5261 ± 0.8217	1339.4997 ± 30.6558	-
Toluene	Girifalco	0.99883	46.1981 ± 12.6240	301.1397 ± 133.4234	123407.0806 ±16037.2519
n-Hexane	Andrade	0.99933	15.7963 ± 0.4391	872.5760 ± 7.6329	-
n-Hexane	Girifalco	0.99997	6.6483 ± 0.4526	1371.5124 ± 39.0122	-71171.4768 ± 5667.3103

Based on the fitting results, we find that the Girifalco model provides a better fit compared to the Andrade equation. The main reason for this improved fit is that our samples contain polar molecules, which influence the activation energy through dipole-dipole interactions. Specifically, methanol and acetone have strong polarity, whereas n-hexane and toluene are less polar. In the case of less polar molecules, polarity can arise from induction, i.e., temporary electric fields caused by the presence of nearby charged molecules, even without direct contact. In other words, the Girifalco equation may partially account for polarity due to induction, as observed with n-hexane. In the final part of this paper, we will use the numerical values for viscosity at a specific temperature as obtained from the Girifalco model fit, in order to study a possible relation with the velocity of sound in liquids.

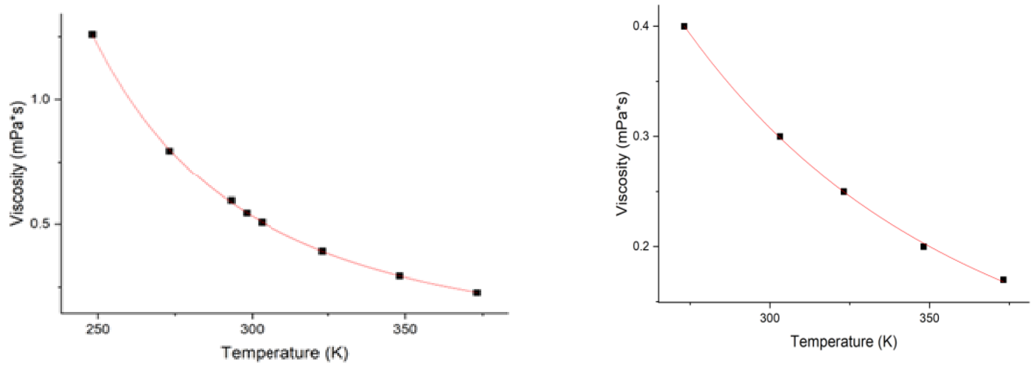

Figure 3. Fitting the viscosity law for methanol (left panel) acetone (right panel)

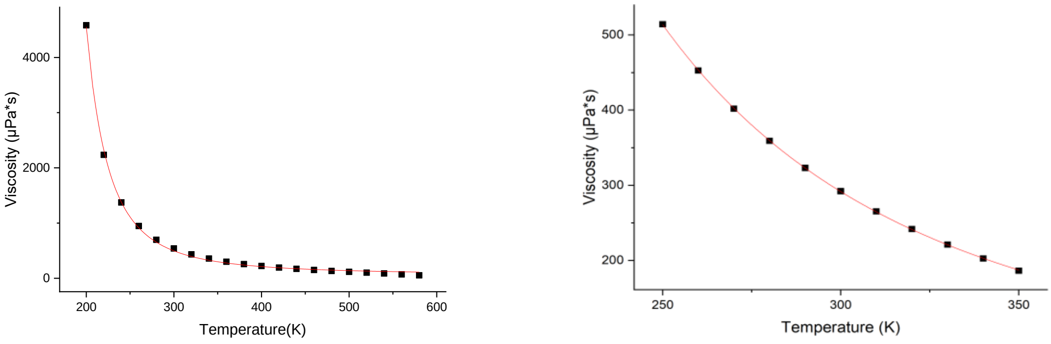

Figure 4. Fitting the viscosity law for toluene (left panel) n hexane (right panel)

Specifically, we aim now to explore a relation between the velocity of sound and viscosity using our results and by following an empirical law proposed in (Parthasarathy & Bakhshi, 1953) where the following equation was proposed

$$v^{1/3} / \rho = A_0 + B / \eta^{1/2} . \tag{10}$$

This relation involves two constants, A_0 and B. The constant A_0 in the last equation has been proposed to be 13.56 for all groups of liquids. In contrast, the constant B varies across different liquid groups, as indicated by distinct diverging straight lines. In our study, we applied this viscosity-based empirical law and calculated the values of B for the simple liquids at various temperatures, as presented in Tables 10–13.

Table 10. Numerical value for B constant for methanol

Temperature (K)	Density (kg/m³)	Speed of Sound (m/s)	Viscosity (Pa·s)	B Constant
293.15	791.633	1120.36	5.8622×10^{-4}	-0.32800
298.15	786.919	1103.89	5.4547×10^{-4}	-0.31639
303.15	782.213	1087.51	5.0859×10^{-4}	-0.30551
313.15	772.698	1055.10	4.4482×10^{-4}	-0.28571
323.15	763.061	1022.74	3.9197×10^{-4}	-0.2682

Table 11. Numerical value for B constant for acetone

Temperature (K)	Density (kg/m³)	Speed of Sound (m/s)	Viscosity (Pa·s)	B Constant
293.15	790.735	1185.95	3.2838×10^{-4}	-0.24548
298.15	785.004	1163.52	3.1312×10^{-4}	-0.23971
303.15	779.265	1141.20	2.9881×10^{-4}	-0.23417
313.15	767.608	1096.56	2.7276×10^{-4}	-0.22373

Table 12. Numerical value for B constant for toluene

Temperature (K)	Density (kg/m³)	Speed of Sound (m/s)	Viscosity (Pa·s)	B Constant
293.15	866.778	1327.76	5.42515×10^{-4}	-0.31554
298.15	862.125	1306.04	5.08378×10^{-4}	-0.30545
303.15	857.462	1284.26	4.77781×10^{-4}	-0.29612
313.15	848.093	1241.36	4.25403×10^{-4}	-0.27942
323.15	838.656	1199.35	3.82449×10^{-4}	-0.26494

Table 13. Numerical value for B constant for n-hexane

Temperature (K)	Density (kg/m³)	Speed of Sound (m/s)	Viscosity (Pa·s)	B Constant
293.15	659.707	1100.87	3.12537×10^{-4}	-0.23945
298.15	655.160	1078.31	2.97023×10^{-4}	-0.23343
303.15	650.625	1055.90	2.82628×10^{-4}	-0.22770
313.15	641.384	1010.70	2.56808×10^{-4}	-0.21705
323.15	631.942	967.290	2.34398×10^{-4}	-0.20736

From these numerical estimations, we observe that, in general, as the temperature increases, the density, speed of sound, and viscosity all decrease. Interestingly, the B constant is negative for all four liquids, and its value generally increases (becomes less negative) with rising temperature.

4. Conclusions

Measuring the thermodynamic properties of liquids is of particular interest to understand the complexity of molecular interactions in the system. Furthermore, such quantities are difficult to determine experimentally. Ultrasonic velocity and density data, on the other hand, offers a possibility to probe these important thermodynamic properties of liquids. In the present work we experimentally measured the values of ultrasonic velocity (v) and density (ρ) data for four liquids: methanol, n-hexane, toluene and acetone which were obtained through DSA 5000 M Density Meter, measured at the temperature range from (293.15-333.15) K. Our experimental results for velocity of sound and density are in accordance with previous results reported in the literature (see for example, Riddick, J.A. and Bunger, W.B, 1970). We then computed the thermal expansion coefficients (α), isothermal compressibility (β_T), isentropic compressibility (β_S), refractive index (n), internal pressure (P_{int}), and heat capacity ratio (γ), as well as the interpolation plot of these quantities under temperature variation. It is found that the thermal expansion coefficient, isothermal compressibility, and isentropic compressibility all increase with the increase of temperature. Interestingly, we find that n-hexane exhibits the highest values for these properties, followed by methanol and acetone, with the lowest values observed for toluene. It is also observed that the specific heat ratio and internal pressure decrease as temperature increases.

To this end, we investigated the viscosity under temperature variation. In particular, we fitted the law of viscosity by incorporating the dipole-dipole interactions. We further showed that the Girifalco model provides a better fit compared to the Andrade equation. This is related to the fact that our samples contain polar molecules, which, of course, influence the activation energy through dipole-dipole interactions. Namely, methanol and acetone have strong polarity, whereas n-hexane and toluene are less polar. Using the numerical fitting law, we further explored the relation between sound velocity and viscosity for the used liquids. We evaluated numerically the constant B, which is found to be negative and generally increases with the increase of temperature.

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