

Electron Releasing Effect on the Thermodynamic and Transport Properties of Liquid Mixtures at Various Temperatures

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Abstract: Densities (ρ), speeds of sound (u), and viscosities (η) are reported for binary mixtures of *N,N*-diethyl aniline with 1-alkanols (1-hexanol, 1-heptanol and 1-octanol) over the entire composition range of mole fraction at $T = (303.15 - 318.15)$ K and at atmospheric pressure 0.1 MPa. The excess properties such as excess molar volume, excess isentropic compressibility and deviations in viscosity are calculated from the density, speed of sound and viscosity. Excess properties are correlated using the Redlich – Kister equation. The excess partial molar volumes and excess partial molar isentropic compressibilities are calculated for all the binary systems throughout the composition range and also at infinity dilutions. The results are analyzed in terms of rupture of hydrogen bonded chain of the dipolar interaction between solute and 1-alkanols exceeds the intermolecular interaction through dipole-dipole and hydrogen bonding between *N,N*-diethyl aniline and 1-alkanol molecules. The V^E results are analyzed in the light of Prigogine-Flory-Patterson theory. Analysis of each of the three contributions viz. interactional, free volume and P^* to V^E has shown that interactional contribution are positive for all systems, both the free volume effect and P^* contribution are negative for all the mixtures.

Keywords: Density, speed of sound, viscosity, *N,N*-diethyl aniline, 1-alkanols, PFP theory

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I. Introduction

The study of the thermodynamic properties of binary mixtures contributes to an understanding of the behavior of different liquids and their functional groups. This information is very useful in the design of industrial process and in the development of theories for the liquid state and predictive methods. Excess thermodynamic parameters of different mixtures are useful in the study of molecular interactions and arrangements [1]. For engineering applications, liquid mixtures, rather than single component liquid systems are used in processing and product formulations. The thermodynamic parameters derived from the measurement of density, speed of sound and viscosity are useful in understanding the nature and type of intermolecular interactions between the compound molecules.

The variations in densities, speeds of sound and other properties derived using them, pertaining to binary mixtures of alcohols with divergent organic liquids which differ in molecular size, shape, chain length and degree of molecular association than that of normal alcohols, is one of the interesting aspect of research in molecular interactions [2]. In the present work, the liquid components selected for the study of molecular interactions are *N,N*-diethyl aniline and 1-alkanols: 1-hexanol, 1-heptanol and 1-octanol. These solvents are well known organic liquids with a wide range of applications in various fields of chemistry in addition to their use in industries and routine analytical work. *N,N*-diethyl aniline is used as a solvent and also in preparation of dyes and various organic products. Alcohols are interesting as they are biologically and industrially important amphiphilic materials and also versatile solvents used in chemical and technological processes and further, they are inexpensive and easily available at high purity [3].

The aim of the present work is to know the effect of carbon chain length and the influence of polarity of the 1-alkanols in the binary blends of *N,N*-diethyl aniline with 1-alkanols on the excess volume, excess isentropic compressibility and deviation in viscosity values. The present investigation is a continuation of our earlier research [4] on thermodynamic properties of binary liquid mixtures.

A review of the literature has shown that the thermodynamic properties of binary mixtures of *O*-chlorophenol with aniline and *N*-alkylanilines [5], 2-methoxyethanol with diethylamine and triethylamine [6], *m*-cresol with substituted anilines [7] and 2-(2-butoxyethoxy) ethanol with aniline and *N*-alkylanilines [8] have been investigated. There are no information accessible indicating itemized physical properties such as density,

speed of sound and viscosity for the binary mixtures of N, N-diethyl aniline with 1-alkanols. Hence, this research work is undertaken pertaining to these binary blends. The data found in this work may be helpful in various industrial processes such as formulation of PVC cements. In this paper we report densities (ρ), speeds of sound (u) and viscosities (η) of the binary mixtures of N, N-diethyl aniline with 1-alkanols (including pure liquids) at different temperatures over the entire composition range. By using these data, various physicochemical parameters, viz., V^E (excess molar volume), κ_s^E (excess isentropic compressibility), deviation in viscosity ($\Delta\eta$), excess Gibbs energy of activation of viscous flow (G^{*E}), $\bar{V}_{m,1}^{\circ E}$, $\bar{V}_{m,2}^{\circ E}$, $\bar{K}_{s,m,1}^{\circ E}$ and $\bar{K}_{s,m,2}^{\circ E}$ have been calculated.

II. Experimental

2.1 Materials

Chemicals used in the present study were N, N-diethyl aniline, 1-hexanol, 1-heptanol and 1-octanol. All the chemicals were purchased from S.D. Fine Chemicals Ltd. N, N-diethyl aniline was dried over potassium hydroxide pellets for several days [9]. All the chemicals were further purified by distillation and fractional distillation under reduced pressure, and only the middle fractions were collected [9,10]. Before use, the chemicals were stored over 0.4 nm molecular sieves for about 72 h to remove water and degassed. The purity of the liquid samples was checked by gas chromatography. The water contents were determined by Karl-Fischer method. The details of the chemicals and pertaining purification methods were presented in Table 1.

2.2 Apparatus and procedure

All the binary liquid mixtures were prepared by weighing an amount of pure liquids in an electric balance (ER-120A, Afoset) with a precision of ± 0.1 mg by syringing each component into air-tight stopper bottles to minimize evaporation losses. The uncertainty of the mole fraction was $\pm 1 \times 10^{-4}$. The densities of pure liquids and their binary mixtures were measured by using a single-capillary pycnometer (made of Borosil glass) having a bulb capacity of $\sim 10 \text{ cm}^3$. The capillary with graduated marks had uniform bore and could be closed with a well-fitted cap. The marks on the capillary were recalibrated by using triply distilled water. The uncertainty in density measurement was found within $\pm 0.2 \times 10^{-3} \text{ g} \cdot \text{cm}^{-3}$. The speed of sound measurements was performed using a commercially available single crystal ultrasonic interferometer (model F-05) from Mittal Enterprises, New Delhi, India, at 2 MHz frequency at various temperatures. The uncertainty in the measured speed of sound is $\pm 0.5\%$. The temperature of the test liquids during the measurements was maintained within an uncertainty of ± 0.01 K in an electronically controlled thermostatic water bath. The interferometer function was tested by obtaining measurements for pure samples of benzene, toluene, chloroform, chlorobenzene and acetone and the measured speeds of sound of these liquids were in good agreement with those reported in the literature [11]. The viscosities of pure liquids and their mixtures were determined at atmospheric pressure at $T = (303.15 - 318.15)$ K by using an Ubbelohde viscometer, which was calibrated with benzene, carbon tetrachloride, acetonitrile and doubly distilled water [12]. The Ubbelohde viscometer bulb capacity was 15 ml and the capillary tube had a length of about 90 mm with 0.5 mm internal diameter. The viscometer was thoroughly cleaned and perfectly dried, filled with the sample liquid by fitting the viscometer to about 30° from the vertical and its limbs were closed with Teflon caps to avoid the evaporation. The viscometer was kept in a transparent walled bath with a thermal stability of ± 0.1 K for about 20 min to obtain thermal equilibrium. An electronic digital stopwatch with an uncertainty of ± 0.01 s was used for flow time measurements. The experimental uncertainty of viscosity was estimated as $\pm 1.03\%$ and the uncertainty of temperature ± 0.1 K. The purity of all these solvents were compared with the measured densities, speeds of sound and viscosities of the pure liquids with the literature [13-21] and these were enlisted in Table 2.

Experimental data graphically compared [22-33] and in the given supplementary material Figs. from 1S to 9S and also third-order polynomial was used to fit viscosity and speed of sound as a function of concentration, for pure substances and its binary mixture of N,N-diethyl aniline with 1-heptanol, + 1-octanol, were listed in Tables and Figs. 1S, 2S, 10S, 11S and 12S respectively (*Supplementary material*).

The analysis of 1-hexanol indicated that the literature ρ data [23] are more scattered as compared to our data. It may arise due to experimental procedure because viscosity ' η ' was well correlated with our experimental values. The AAD values for ' u ' were found to be less than 0.28% (Jerie et al [27]) to our experimental values may be experimental procedure or chemical purity. The 1-heptanol and 1-octanol speed of sound ' u ' values varies with 0.4% and 0.35% (Average Absolute Deviation) with Dzida et al [32] at 303.15K. The deviations may be arising due to experimental procedure.

III. Results And Discussion:

The experimental densities and viscosities for all the binary systems at various compositions were used to calculate the excess thermodynamic functions using following equations:

$$V^E / \text{cm}^3 \cdot \text{mol}^{-1} = [x_1 M_1 + x_2 M_2] / \rho - [x_1 M_1 / \rho_1 + x_2 M_2 / \rho_2] \quad (1)$$

$$\Delta \eta / \text{mPa} \cdot \text{s} = \eta - (x_1 \eta_1 + x_2 \eta_2) \quad (2)$$

$$G^{*E} / \text{J} \cdot \text{mol}^{-1} = RT [\ln \eta V - (x_1 \ln \eta_1 V_1 + x_2 \ln \eta_2 V_2)] \quad (3)$$

where ρ , η and V are density, viscosity and molar volume of the binary mixture. x_1 , M_1 , ρ_1 , η_1 , V_1 and x_2 , M_2 , ρ_2 , η_2 , V_2 are the mole fraction, molar mass, density, viscosity and molar volume of pure components 1 and 2 respectively. R is the gas constant and T is the absolute temperature

The experimental data were used to compute isentropic compressibility (κ_s) by using the following relation:

$$\kappa_s = (u^2 \rho)^{-1} \quad (4)$$

The method used for calculating κ_s^E (Benson - Kiyohara approach) was outlined previously [34].

V^E , κ_s^E and $\Delta \eta$ values were fitted to a Redlich - Kister [35] polynomial equation,

$$Y^E = x_1 x_2 \sum_{i=0}^j A_i (1 - 2x_1)^i \quad (5)$$

where Y^E is the V^E , κ_s^E and $\Delta \eta$. Values of the coefficients A_i were determined by using the method of least - squares. The standard deviations $\sigma(Y^E)$ were calculated by using the formula

$$\sigma(Y^E) = [\sum (Y_{exp}^E - Y_{cal}^E)^2 / (m-n)]^{1/2} \quad (6)$$

where m is the total number of experimental points and n is the number of parameters. The coefficients, A_i and corresponding standard deviation values (σ) were presented in Table 5.

The densities, speeds of sound and viscosities for mixtures of various mole fractions of N, N-diethyl aniline in 1-alkanols were presented in Tables 3. The excess molar volumes, excess isentropic compressibility, viscosity deviation and excess Gibbs energy of activation of viscous flow at various temperatures were presented in Tables 4. Figs. 1, 2, 3 and 4 show values of V^E , κ_s^E , $\Delta \eta$ and G^{*E} as a function of the molar fraction of N, N-diethyl aniline, respectively.

The volume changes arising due to addition of N, N-diethyl aniline to an alkanol results from several opposing effects [36]. The main volume effects accompanying the addition of N, N-diethyl aniline to an alkanol result from changes of free volume, disruption of the alkanol structure, and the so-called condensation effect [37] due to restriction of the rotational motion of the alkanol molecule [38]. A perusal of Table 4 shows that the excess molar volume data for the mixtures N, N-diethyl aniline with 1-alkanols are positive over the entire composition range at all temperatures. In the present mixtures, considerable amounts of positive contributions are expected due to dissociation of intermolecular interactions between like molecules is dominant over the heteroassociation interactions between unlike molecules. Similar results have also been reported for N, N-dimethylformamide with n-alkanols [39] and toluene with n-alkanols mixtures [40].

The V^E values at equimolar concentration of mixtures follow the order:

$$1\text{-octanol} > 1\text{-heptanol} > 1\text{-hexanol}$$

It indicates that clustering decreases with increase of chain length of alkanols owing to less proton-donating tendency of higher alkanols. Therefore, heteroassociation effects decrease in the binary mixtures with an increase of chain length of linear alkanols. Hence above order is justified.

The increase of V^E with increase of temperature suggests the breaking of interactions and aggregates of alkanols and N, N-diethyl aniline at elevated temperature [36]. An examination of data in the Table 4 shows that the excess isentropic compressibility data for all the binary systems are positive over the entire composition range at $T = (303.15 \text{ to } 318.15) \text{ K}$.

The observed values of κ_s^E can be qualitatively explained by considering the factors (i) declustering and breaking of H-bonds in 1-alkanols (ii) repulsive forces due to the electric charge of components, (iii) size and shape of both component molecules along with the effect of the alkyl chain of the 1-alkanols because the hydrophobic character of the 1-alkanol is amplified by an increase in chain length, and consequently, the molecular interactions between 1-alkanols and N, N-diethyl aniline molecules are weakened. It also suggests that the physical dipole-dipole interactions between 1-alkanol monomers and multimers contribute to increase in free-spaces and these results in decrease in sound velocity and it is reflected in the positive deviation in isentropic compressibilities [41]

From the Fig. 2, it is inferred that the positive κ_s^E values increase from 1-octanol to 1-heptanol in their binary blends with N, N-diethyl aniline. This fact signifies decreasing dipole-dipole interaction due to decreasing proton donating ability with increasing chain length of 1-alkanols. It also suggests the rupture of hydrogen bonded chain of the dipolar interactions between N, N-diethyl aniline and 1-alkanol exceed the

intermolecular interaction through dipole-dipole and hydrogen bonding between N,N-diethyl aniline and 1-alkanol molecules.

The positive κ_s^E values for binary systems follow the order:

$$1\text{-octanol} > 1\text{-heptanol} > 1\text{-hexanol}$$

The order indicates that the strength of interactions between component molecules decreases due to decrease in polarizability of 1-alkanol molecules [42, 43] as the carbon chain increases and also decreases in proper dipole alignment of the components.

According to Kauzman and Eyring [44], the viscosity of a mixture strongly depends on the entropy of mixture, which is related to liquid structure and enthalpy and consequently, to the molecular interactions between the components of the mixture. Vogel and Weiss [45] affirm that mixtures with strong interactions between different molecules ($H^E < 0$) and negative deviations from Raoult's law present positive viscosity deviations ($\Delta\eta > 0$), whereas for mixtures with positive deviations of Raoult's law and without strong specific interactions, the viscosity deviations are negative ($\Delta\eta < 0$).

An examination of curves in Figs.3 and 4 suggests that the deviation in viscosity ($\Delta\eta$) and excess Gibbs energy of activation of viscous flow (G^{*E}) for all the binary systems is negative over the entire composition range at different temperatures. The negative values of deviation in viscosity and excess Gibbs free energy of activation of viscous flow for the binary systems investigated suggest that the breaking of the self-associated molecules and weak interactions between unlike molecules, decrease systematically with an increase in chain length of alkanol. With increasing in chain length of alkanol, hydrogen bonding interaction of N,N-diethylaniline with 1-hexanol, 1-heptanol and 1-octanol is weaker due to decreased polarizability as suggested by Mecke [38]. The viscosity deviation values are found to be opposite to the sign of excess molar volumes for all binary mixtures, which is in agreement with the view proposed by Brocos et al [46, 47]

IV. Partial Molar Properties

The interpretation of excess partial molar properties ($\bar{V}_{m,1}^E, \bar{V}_{m,2}^E, \bar{K}_{s,m,1}^E$ and $\bar{K}_{s,m,2}^E$) and excess partial molar properties at infinite dilution ($\bar{V}_{m,1}^\infty, \bar{V}_{m,2}^\infty, \bar{K}_{s,m,1}^\infty$ and $\bar{K}_{s,m,2}^\infty$) of components 2 have previously been described [48].

In general, the negative values of excess partial molar volume component 1 and excess partial molar volume component 2 ($\bar{V}_{m,1}^E, \bar{V}_{m,2}^E, \bar{K}_{s,m,1}^E$ and $\bar{K}_{s,m,2}^E$) values, indicate the presence of significant solute-solvent interactions between unlike molecules, whereas the positive excess partial molar volume component 1 and excess partial molar volume component 2 data indicate presence of solute-solute / solvent-solvent interactions between like molecules in the mixtures [49,50].

A close perusal of Table 6 indicates that the values of $\bar{V}_{m,1}^E$ and $\bar{V}_{m,2}^E$ are positive for all the binary mixtures over the whole composition range. Positive values may be attributed to the breaking up of self-associated structures of the components of the mixtures is dominant over the effect of H-bonding and dipole-dipole interaction between unlike molecules

From the Table 6 it may be inferred that the values of $\bar{K}_{s,m,1}^E$ and $\bar{K}_{s,m,2}^E$ are positive for all the binary mixtures over the whole composition range. The positive values indicate that the rupture of hydrogen bonded chain of the dipolar interaction between solute, N, N-diethyl aniline and 1-alkanol exceed the intermolecular interaction through dipole-dipole and hydrogen bonding between N, N-diethyl aniline and 1-alkanol molecules.

From the Table 7, it can be seen that the values of $\bar{V}_{m,1}^\infty$ and $\bar{V}_{m,2}^\infty$ are positive for all the binary mixtures over the whole composition range. The positive $\bar{V}_{m,1}^\infty$ and $\bar{V}_{m,2}^\infty$ values indicate that the rupture of three-dimensional hydrogen-bonded network of 1-alkanol due to the addition of solute, N,N-diethyl aniline is not compensated by the weak interactions between unlike molecules.

It is seen from the Table 7 that the values of $\bar{K}_{s,m,1}^\infty$ and $\bar{K}_{s,m,2}^\infty$ are positive for all the binary systems at each investigated temperature. The positive values may be due to interruption of dipolar association in the binary mixtures of N, N-diethyl aniline with 1-alkanols or attributed to weak dipole-dipole interactions between unlike molecules in the mixtures [51, 52].

Prigogine-Flory-Patterson Theory:

The Prigogine-Flory-Patterson theory considers excess thermodynamic properties of binary mixtures to be the sum of different contributions [53-55]. The excess molar volume can be expressed as: an interactional term which is proportional to the interaction parameter, χ_{12} , a free volume contribution which arises from the dependence of the reduced volume upon the reduced temperature as a result of the differences between the degrees of thermal expansion of the components and an internal pressures and of the reduced volumes of the components

$$\frac{V^E}{x_1V_1^* + x_2V_2^*} = \frac{(\tilde{V}^{-1/3} - 1)\tilde{V}^{2/3}}{[(4/3)\tilde{V}^{-1/3} - 1]} \Psi_1 \theta_2 \left(\frac{\chi_{12}}{P_1^*} \right) \text{int.contribution} - \frac{(V_1 - V_2)^2 [(14/9)\tilde{V}^{-1/3} - 1] \Psi_1 \Psi_2}{[(4/3)\tilde{V}^{-1/3} - 1]\tilde{V}}$$

$$\text{(fv.contribution)} + \frac{(\tilde{V}_1 - \tilde{V}_2)(P_1^* - P_2^*)}{P_1^* \Psi_2 + P_2^* \Psi_1} \Psi_1 \Psi_2 \text{ (P* contribution)} \quad (7)$$

The reduced volume of pure component \tilde{V}_i was calculated from the isobaric thermal expansivity α_i by equation:

$$\tilde{V}_i = \left(\frac{1 + \left(\frac{4}{3}\right)\alpha_i T}{1 + \alpha_i T} \right) \quad (8)$$

The \tilde{V} of mixture was approximated in equation (8) by

$$\tilde{V} = \Psi_1 \tilde{V}_1 + \Psi_2 \tilde{V}_2 \quad (9)$$

The molecular contact energy fraction of components Ψ_1 is given by

$$\Psi_1 = 1 - \Psi_2 = \frac{\phi_1 P_1^*}{\phi_1 P_1^* + \phi_2 P_2^*} \quad (10)$$

The characteristic volume is $V_i^* = V_i^* / \tilde{V}_i$ and the characteristic pressure is given by

$$P_i^* = \frac{T \tilde{V}_i^2 \alpha_i}{\kappa_{Ti}} \quad (11)$$

where κ_{Ti} is the isothermal compressibility of pure component i .

The hard-core volume fractions of the components 1 and 2 (ϕ_1 and ϕ_2) are defined by

$$\phi_1 = 1 - \phi_2 = \frac{x_1 V_1^*}{x_1 V_1^* + x_2 V_2^*} \quad (12)$$

The κ_T values were calculated from the following expression,

$$\kappa_T = \kappa_S + \frac{T V \alpha^2}{C_p} \quad (13)$$

The χ_{12} parameter required for the calculation of V^E using Flory-Patterson theory was derived by fitting the V^E expression to the experimental equimolar value of V^E for each system investigated. The calculated equimolar values of the three contributions together with the χ_{12} parameter for each system were listed in Table 8. An analysis of each of the three contributions to V^E showed that the interactional contribution was positive for all systems, the free volume contribution was negative for all the mixtures and P* contribution was positive for all the mixtures except 1-octanol which had a negative internal pressure term. It is clear from Table 8 that the dominant role is played by the interactional contribution and this is the main parameter for deciding the sign and magnitude of excess volumes for N,N-diethyl aniline + 1-alkanols.

V. Conclusion

This paper reports experimental data of densities, speeds of sound and viscosities of binary blends of N,N-diethyl aniline with 1-alkanols (1-hexanol, 1-heptanol and 1-octanol) binary mixtures over the entire composition range at $T = (303.15 - 318.15)$ K with 5 K intervals. From the experimental data, various physicochemical parameters, viz., V_m^E , κ_s^E and $\Delta\eta$ of the mixtures, the excess partial molar properties at infinite dilution ($V_{m,1}^{\infty E}$, $V_{m,2}^{\infty E}$, $K_{s,m,1}^{\infty E}$ and $K_{s,m,2}^{\infty E}$) of components were calculated. The results are analyzed in terms of rupture of hydrogen bonded chain of the dipolar interaction between solute and 1-alkanols exceed the intermolecular interaction through dipole-dipole and hydrogen bonding between N,N-diethyl aniline and 1-alkanols molecules. This behavior is characteristic for systems containing an associated component. Moreover, the V^E values have been analyzed with Prigogine-Flory-Patterson theory and shown that the interactional contribution is the main factor for positive values of excess molar quantities.

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Table 1 List of chemicals with details of supplier, CAS number, Purity and water content

Name of the chemical	Source	CAS number	purification method	mass fraction purity	**Water content mass fraction	*Analysis method
N,N-diethyl aniline	S.D. fine Chemicals India	91-66-7	distillation under reduced pressure	0.995	0.00045	GC
1-hexanol	S.D. fine Chemicals India	111-27-3	Fractional distillation	0.985	0.00042	GC
1-heptanol	S.D. Fine Chemicals, India	111-70-6	distillation under reduced pressure	0.995	0.00044	GC
1-octanol	S.D. Fine Chemicals, India	111-87-5	Fractional distillation	0.995	0.00045	GC

*Gas Chromatography

** Karl-Fischer method

Table 2 Densities, viscosity and speeds of sounds data of pure components at different temperatures and 0.1MPa pressure

component	density (ρ / g·cm ⁻³)		speed of sound, (u / m·s ⁻¹)		viscosity (mPa·s)		C _p (J·K ⁻¹ ·mol ⁻¹)
	experimental	literature	experimental	literature	experimental	literature	
N,N-diethyl aniline							
303.15 K	0.9258[4]	0.9259[13] 0.9260[14] 0.9261[15]	1432.8[4]	1434.0[15]	1.711[4]	1.709[13]	291.0[16]
308.15 K	0.9218[4]	0.9219[13]	1415.2[4]		1.537[4]	1.534[13]	
313.15 K	0.9179[4]	0.9180[13] 0.9177[14] 0.9175[15]	1397.2[4]	1396.0[15]	1.400[4]	1.402[13]	296.0[16]
318.15 K	0.9138[4]	0.9139[13]	1378.8[4]		1.276[4]	1.279[13]	
1-hexanol							
303.15 K	0.8122	0.81212[17]	1283.4	1287.90[20]	3.883	3.885[20]	246.52[20]
308.15 K	0.8081	0.80780[19]	1270.2	1271.14[20]	3.355	3.359[20]	251.47[20]
313.15 K	0.8043	0.80451[17]	1256.9	1254.45[20]	2.911	2.914[20]	256.48[20]
318.15 K	0.8002	0.80018[18]	1244.2		2.656	2.655[18]	
1-heptanol							
303.15 K	0.8153	0.81529[21]	1315.5	1313.58[20]	5.065	5.069[20]	275.37[20]
308.15 K	0.8117	0.81174[21]	1298.4	1293.86[20]	4.329	4.333[20]	289.83[20]
313.15 K	0.8081	0.80816[21]	1280.5	1277.15[20]	3.723	3.726[20]	295.35[20]
318.15 K	0.8044	0.80439[18]	1261.5		3.446	3.447[18]	
1-octanol							
303.15 K	0.8183	0.81823[21]	1334.9	1338.02[20]	6.405	6.402[20]	313.60[20]
308.15 K	0.8147	0.81474[21]	1318.7	1314.27[20]	5.422	5.425[20]	320.12[20]
313.15 K	0.8101	0.81009[21]		1297.60[20]	4.625	4.628[20]	325.98[20]
318.15 K	0.8055	0.80555[18]	1300.0 1284.9		4.051	4.051[18]	

The standard uncertainties are $u(\rho) = \pm 0.2 \times 10^{-3}$ g·cm⁻³, $u(u) = 0.5\%$, $u(\eta) = \pm 1.03\%$, $u(T) = 0.01$ K for density and speed of sound, $u(T) = 0.1$ K for viscosity and $u(p) = 1$ kPa

Table 3; Density (ρ), Viscosity (η) and speed of sound (u) of binary liquid mixtures of N,N-diethylaniline with 1-hexanol, 1-heptanol and 1-octanol at T= (303.15 to 318.15) K and 0.1MPa pressure

X ₁ N,N-diethylaniline	density (ρ / g·cm ⁻³)				viscosity (η / mPa·s)				speed of sound (u / m·s ⁻¹)			
	303.15K	308.15K	313.15K	318.15K	303.15K	308.15K	313.15K	318.15K	303.15K	308.15K	313.15K	318.15K
N,N-diethylaniline (1) + 1-hexanol (2)												
0.0000	0.8122	0.8081	0.8043	0.8002	3.883	3.355	2.911	2.656	1283.4	1270.2	1256.9	1244.2
0.1057	0.8245	0.8202	0.8167	0.8124	3.391	2.850	2.403	2.105	1293.7	1278.4	1265.1	1252.4
0.1952	0.8348	0.8306	0.8270	0.8227	3.041	2.524	2.096	1.801	1302.6	1287.2	1273.6	1260.3
0.3004	0.8469	0.8427	0.8391	0.8348	2.695	2.226	1.835	1.569	1314.3	1298.8	1284.7	1270.8
0.4021	0.8585	0.8543	0.8507	0.8464	2.423	1.997	1.652	1.421	1326.8	1311.2	1296.7	1282.1
0.5011	0.8698	0.8656	0.8619	0.8577	2.207	1.814	1.518	1.314	1340.3	1324.6	1309.5	1294.1
0.6117	0.8823	0.8782	0.8744	0.8702	2.014	1.667	1.405	1.218	1356.8	1340.9	1325.3	1308.9
0.7135	0.8938	0.8897	0.8859	0.8817	1.879	1.577	1.332	1.151	1373.7	1357.7	1341.3	1324.0
0.8163	0.9053	0.9012	0.8974	0.8932	1.780	1.525	1.297	1.119	1392.6	1376.4	1359.3	1341.0
0.9045	0.9152	0.9111	0.9073	0.9031	1.725	1.501	1.313	1.148	1410.5	1394.2	1376.7	1357.5
1.0000	0.9258	0.9218	0.9179	0.9138	1.711	1.537	1.400	1.276	1432.8	1415.2	1397.2	1378.8
N,N-diethylaniline (1) + 1-heptanol (2)												
0.0000	0.8153	0.8117	0.8081	0.8044	5.054	4.337	3.735	3.407	1315.5	1298.4	1280.5	1261.5
0.1216	0.8276	0.8240	0.8203	0.8165	4.250	3.658	3.122	2.671	1322.4	1305.9	1286.1	1267.9
0.2426	0.8401	0.8364	0.8327	0.8289	3.593	3.095	2.622	2.129	1330.9	1314.5	1294.8	1276.5
0.3349	0.8498	0.8461	0.8424	0.8385	3.178	2.735	2.308	1.826	1338.6	1322.2	1302.7	1284.2
0.4641	0.8638	0.8600	0.8563	0.8523	2.706	2.323	1.960	1.535	1351.1	1334.7	1315.4	1296.6
0.5612	0.8745	0.8707	0.8669	0.8629	2.425	2.079	1.760	1.398	1361.9	1345.5	1326.3	1307.2
0.6524	0.8848	0.8809	0.8771	0.8731	2.210	1.895	1.617	1.319	1373.2	1356.6	1337.6	1318.4
0.7356	0.8943	0.8904	0.8866	0.8826	2.049	1.763	1.520	1.278	1384.8	1368.4	1349.4	1329.9
0.8144	0.9035	0.8996	0.8957	0.8917	1.923	1.665	1.455	1.257	1397.1	1380.6	1361.8	1342.0
0.9141	0.9154	0.9114	0.9075	0.9034	1.792	1.577	1.405	1.243	1414.5	1398.1	1379.6	1359.6
1.0000	0.9258	0.9218	0.9179	0.9138	5.065	4.329	3.723	3.446	1432.8	1415.2	1397.2	1378.8
N,N-diethylaniline (1) + 1-octanol (2)												
0.0000	0.8183	0.8147	0.8101	0.8055	6.392	5.388	4.585	4.011	1334.7	1318.0	1298.8	1284.3
0.1141	0.8284	0.8248	0.8202	0.8157	5.389	4.420	3.684	3.135	1339.5	1321.5	1301.4	1288.6
0.2548	0.8415	0.8379	0.8334	0.8289	4.382	3.490	2.854	2.356	1346.4	1328.0	1307.3	1295.0
0.3537	0.8512	0.8476	0.8431	0.8386	3.806	2.987	2.428	1.976	1352.5	1334.0	1313.3	1300.5
0.4726	0.8634	0.8597	0.8553	0.8509	3.235	2.519	2.056	1.667	1361.3	1343.3	1322.8	1308.8
0.5725	0.8742	0.8704	0.8661	0.8617	2.842	2.221	1.837	1.503	1370.4	1352.8	1332.7	1317.5
0.6945	0.8879	0.8840	0.8798	0.8755	2.446	1.947	1.653	1.386	1383.9	1366.8	1347.2	1330.5
0.7795	0.8979	0.8940	0.8899	0.8856	2.212	1.800	1.560	1.339	1395.2	1378.2	1359.1	1341.4
0.8584	0.9076	0.9036	0.8996	0.8953	2.016	1.684	1.487	1.305	1407.0	1390.1	1371.3	1353.0
0.9317	0.9168	0.9129	0.9089	0.9047	1.847	1.587	1.421	1.271	1419.5	1402.3	1383.9	1365.2
1.0000	0.9258	0.9218	0.9179	0.9138	6.405	5.422	4.625	4.051	1432.3	1414.8	1396.5	1377.9

The standard uncertainties are $(x_1) = 1 \times 10^{-4}$, $u(\rho) = \pm 0.54 \times 10^{-3}$ g·cm⁻³, $u(u) = 0.3\%$, $u(\eta) = 1.03\%$, $u(T) = 0.01$ K for density and speed of sound, $u(T) = 0.1$ K for viscosity and $u(p) = 1$ kPa

Table 4 Excess molar volume (V^E), excess isentropic compressibility (κ_c^E), deviation in viscosity ($\Delta\eta$) and excess Gibbs free energy of activation of viscous flow ($G^{\ddagger E}$) of

Liquid mixtures of N,N-diethylaniline with 1-hexanol, 1-heptanol and 1-octanol at T= (303.15 to 318.15) K

N,N-diethylaniline (1) + 1-hexanol (2)												
x ₁	V ^E /cm ³ ·mol ⁻¹			Δη/mPa·s				κ _c ^E /TPa ⁻¹				
	303.15 K	308.15 K	313.15 K	303.15 K	308.15 K	313.15 K	318.15 K	303.15 K	308.15 K	313.15 K	318.15 K	
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.1057	0.0216	0.0247	0.0279	0.0307	-0.262	-0.312	-0.348	-0.405	2.226	2.549	2.877	3.207
0.1952	0.0384	0.0429	0.0483	0.0530	-0.418	-0.476	-0.520	-0.585	3.689	4.020	4.462	4.877
0.3004	0.0546	0.0583	0.0631	0.0675	-0.535	-0.583	-0.623	-0.672	4.876	5.310	5.764	6.168
0.4021	0.0647	0.0684	0.0721	0.0762	-0.587	-0.627	-0.651	-0.680	5.560	5.979	6.453	6.887
0.5011	0.0677	0.0725	0.0759	0.0796	-0.588	-0.630	-0.635	-0.650	5.792	6.224	6.749	7.198
0.6117	0.0643	0.0685	0.0725	0.0768	-0.541	-0.576	-0.582	-0.594	5.631	6.125	6.608	7.160
0.7135	0.0546	0.0591	0.0634	0.0681	-0.454	-0.480	-0.501	-0.521	4.926	5.369	5.990	6.535
0.8163	0.0385	0.0433	0.0479	0.0521	-0.330	-0.346	-0.380	-0.410	3.750	4.203	4.780	5.264
0.9045	0.0216	0.0249	0.0268	0.0312	-0.193	-0.210	-0.231	-0.260	2.264	2.597	2.942	3.363
1.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
N,N-diethylaniline (1) + 1-heptanol (2)												
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.1216	0.0289	0.0316	0.0353	0.0390	-0.396	-0.438	-0.471	-0.511	2.741	3.103	3.479	3.789
0.2426	0.0505	0.0546	0.0586	0.0637	-0.647	-0.681	-0.709	-0.740	4.586	5.008	5.450	5.833
0.3349	0.0626	0.0664	0.0704	0.0754	-0.752	-0.762	-0.762	-0.762	5.442	5.841	6.256	6.739
0.4641	0.0702	0.0748	0.0781	0.0824	-0.790	-0.790	-0.790	-0.790	6.029	6.491	6.983	7.445
0.5612	0.0704	0.0753	0.0779	0.0821	-0.745	-0.745	-0.745	-0.745	6.040	6.527	7.014	7.582
0.6524	0.0651	0.0701	0.0737	0.0778	-0.655	-0.609	-0.587	-0.569	5.790	6.407	6.856	7.343
0.7356	0.0566	0.0604	0.0647	0.0701	-0.536	-0.508	-0.488	-0.469	5.124	5.630	6.191	6.746
0.8144	0.0429	0.0489	0.0517	0.0560	-0.398	-0.384	-0.369	-0.378	4.060	4.644	5.110	5.635
0.9141	0.0238	0.0271	0.0281	0.0320	-0.195	-0.193	-0.184	-0.175	2.347	2.733	3.100	3.466
1.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
N,N-diethylaniline (1) + 1-octanol (2)												
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.1141	0.0311	0.0336	0.0370	0.0410	-0.467	-0.525	-0.533	-0.559	2.112	2.413	2.731	2.608
0.2548	0.0563	0.0609	0.0650	0.0685	-0.814	-0.907	-0.908	-0.946	4.465	5.337	6.175	5.505
0.3537	0.0678	0.0719	0.0755	0.0796	-0.925	-1.025	-1.014	-1.051	5.729	7.500	9.682	7.070
0.4726	0.0738	0.0784	0.0817	0.0857	-0.937	-1.031	-1.002	-1.030	6.610	7.940	9.913	8.183
0.5725	0.0733	0.0774	0.0807	0.0849	-0.861	-0.941	-0.898	-0.916	6.707	7.540	9.152	8.329
0.6945	0.0653	0.0698	0.0741	0.0782	-0.685	-0.739	-0.688	-0.693	5.949	6.205	7.279	7.429
0.7795	0.0554	0.0595	0.0639	0.0677	-0.519	-0.556	-0.506	-0.505	4.846	4.809	5.506	6.076
0.8584	0.0396	0.0443	0.0473	0.0514	-0.344	-0.365	-0.324	-0.319	3.410	3.241	3.621	4.302
0.9317	0.0228	0.0261	0.0269	0.0286	-0.169	-0.177	-0.154	-0.149	1.771	1.618	1.762	2.238
1.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000

Table 5 Coefficients of Redlich–Kister equation and standard deviation (σ) values for liquid mixtures of N, N-diethyl aniline with 1-alkanols at T=(303.15 - 318.15) K

Binary mixtures		Functions	A ₀	A ₁	A ₂	σ
303.15 K						
N,N-diethylaniline+ 1-hexanol	V ^E /cm ³ ·mol ⁻¹		0.272	0.013	-0.051	0.001
	Δη/mPa·s		-2.351	0.362	-0.237	0.003
	κ _c ^E /TPa ⁻¹		23.23	1.382	2.565	0.026
N,N-diethylaniline+ 1-heptanol	V ^E /cm ³ ·mol ⁻¹		0.283	0.015	0.002	0.001
	Δη/mPa·s		-3.118	0.774	-0.435	0.001
	κ _c ^E /TPa ⁻¹		24.32	1.857	5.201	0.054
N,N-diethylaniline+ 1-octanol	V ^E /cm ³ ·mol ⁻¹		0.295	0.020	0.051	0.001
	Δη/mPa·s		-3.694	1.227	-0.495	0.001
	κ _c ^E /TPa ⁻¹		26.08	1.758	6.656	0.023
308.15 K						
N,N-diethylaniline+ 1-hexanol	V ^E /cm ³ ·mol ⁻¹		0.288	0.015	-0.020	0.001
	Δη/mPa·s		-2.489	0.561	-0.563	0.006
	κ _c ^E /TPa ⁻¹		24.90	1.734	5.416	0.047
N,N-diethylaniline+ 1-heptanol	V ^E /cm ³ ·mol ⁻¹		0.300	0.024	0.029	0.001
	Δη/mPa·s		-3.162	0.978	-0.787	0.001
	κ _c ^E /TPa ⁻¹		26.14	2.636	8.807	0.069
N,N-diethylaniline+ 1-octanol	V ^E /cm ³ ·mol ⁻¹		0.311	0.028	0.083	0.001
	Δη/mPa·s		-3.742	1.523	-1.282	0.001
	κ _c ^E /TPa ⁻¹		27.51	2.011	10.18	0.052
313.15 K						
N,N-diethylaniline+ 1-hexanol	V ^E /cm ³ ·mol ⁻¹		0.305	0.009	0.003	0.001
	Δη/mPa·s		-2.543	0.649	-1.005	0.009
	κ _c ^E /TPa ⁻¹		26.94	2.249	8.256	0.024
N,N-diethylaniline+ 1-heptanol	V ^E /cm ³ ·mol ⁻¹		0.315	0.015	0.045	0.001
	Δη/mPa·s		-3.212	1.214	-1.125	0.001
	κ _c ^E /TPa ⁻¹		27.89	3.034	12.35	0.075
N,N-diethylaniline+ 1-octanol	V ^E /cm ³ ·mol ⁻¹		0.327	0.022	0.098	0.001
	Δη/mPa·s		-3.916	1.813	-1.506	0.001
	κ _c ^E /TPa ⁻¹		29.44	2.173	11.99	0.049
318.15 K						
N,N-diethylaniline+ 1-hexanol	V ^E /cm ³ ·mol ⁻¹		0.319	0.016	0.042	0.001
	Δη/mPa·s		-2.603	0.835	-1.649	0.001
	κ _c ^E /TPa ⁻¹		28.70	2.929	11.82	0.034
N,N-diethylaniline+ 1-heptanol	V ^E /cm ³ ·mol ⁻¹		0.332	0.017	0.085	0.001
	Δη/mPa·s		-3.355	1.390	-1.541	0.001
	κ _c ^E /TPa ⁻¹		29.79	3.849	15.18	0.081
N,N-diethylaniline+ 1-octanol	V ^E /cm ³ ·mol ⁻¹		0.343	0.018	0.126	0.001
	Δη/mPa·s		-4.018	2.018	-1.519	0.001
	κ _c ^E /TPa ⁻¹		31.12	4.120	15.39	0.063

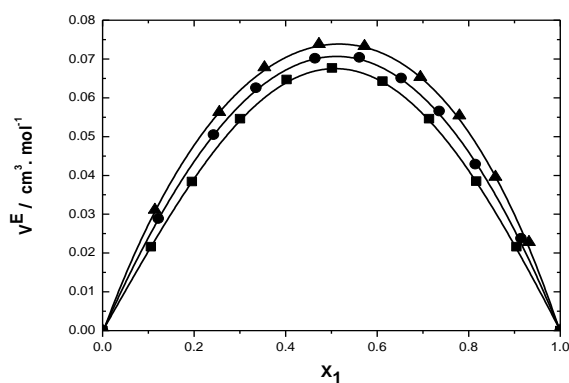


Figure 1 Variation of excess molar volume (V^E) with mole fraction (x_1) of N, N-diethyl aniline in the binary liquid mixtures of N,N-diethyl aniline with 1-hexanol (■), 1-heptanol (●) and 1-octanol (▲) at 303.15 K

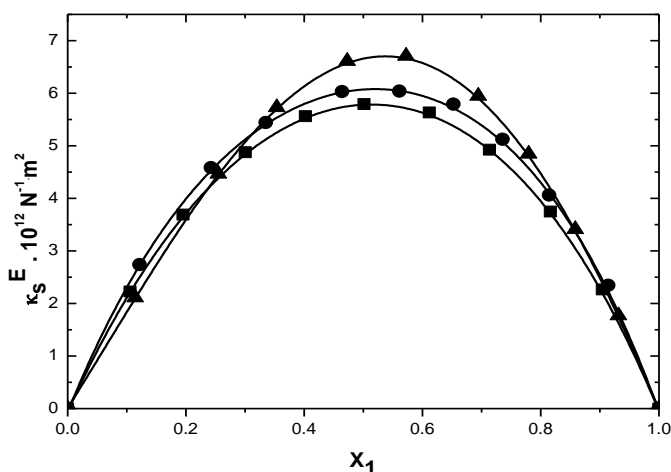


Figure 2 Excess isentropic compressibility (κ_s^E) with mole fraction (x_1) of N, N-diethyl aniline in the binary liquid mixtures of N,N-diethyl aniline with 1-hexanol (■), 1-heptanol (●) and 1-octanol (▲) at 303.15 K

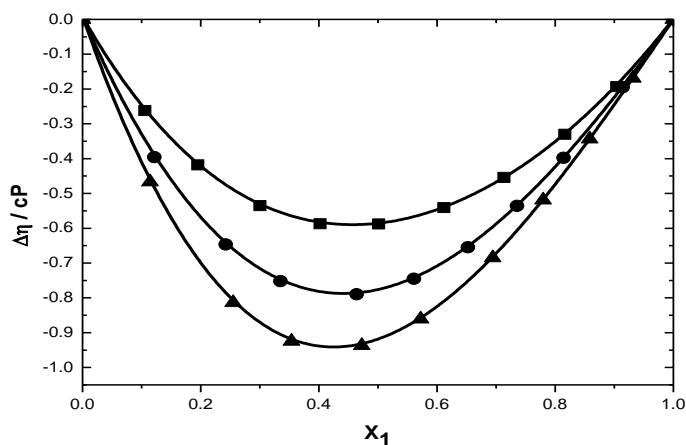


Figure 3 Variation of deviation viscosity ($\Delta\eta$) with mole fraction (x_1) of N, N-diethyl aniline in the binary liquid mixtures of N,N-diethyl aniline with 1-hexanol (■), 1-heptanol (●) and 1-octanol (▲) at 303.15 K

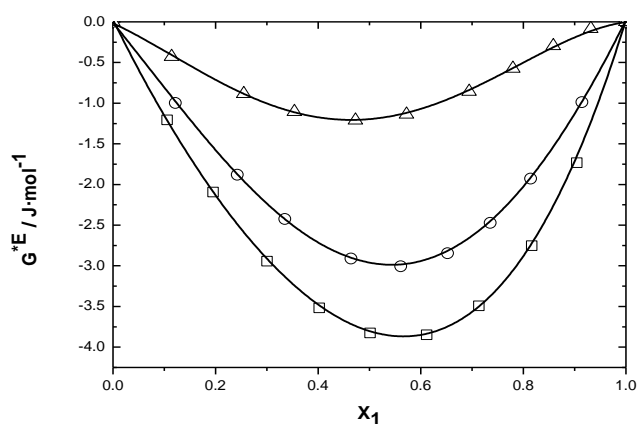


Figure 4 Excess Gibbs energy of activation of viscous flow (G^{*E}) with mole fraction (x_1) of N, N-diethyl aniline in the binary liquid mixtures of N, N-diethyl aniline with 1-hexanol (∇), 1-heptanol (\square) and 1-octanol (Δ) at 303.15 K

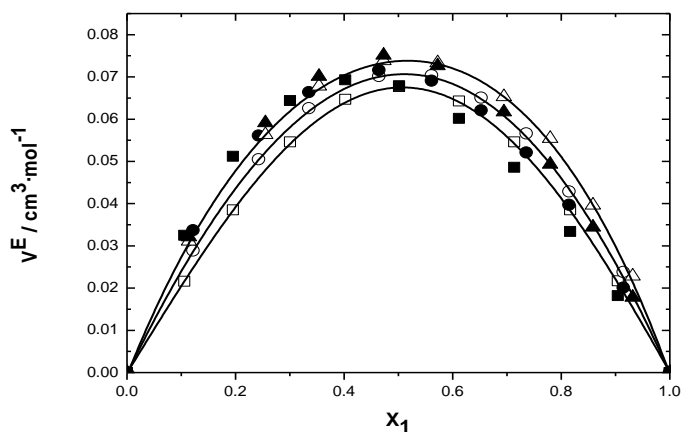


Figure 5 Excess molar volumes of the binary mixtures against mole fraction x_1 at $T = 303.15$ K for N,N-diethyl aniline + 1-hexanol (∇), N,N-diethyl aniline + 1-heptanol (\square), N, N-diethyl aniline + 1-octanol (Δ) and (---) calculated with PFP theory using Parameters

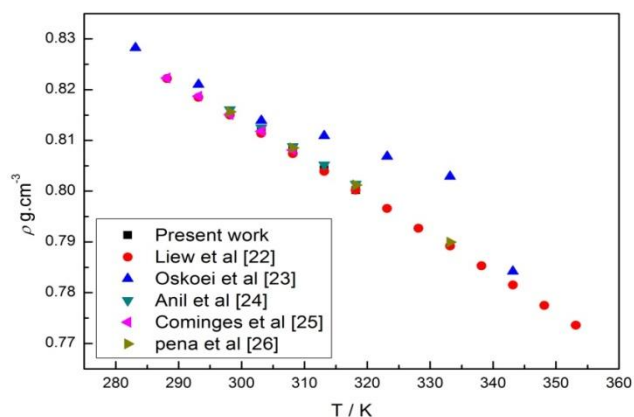


Figure 1 density of 1-hexanol

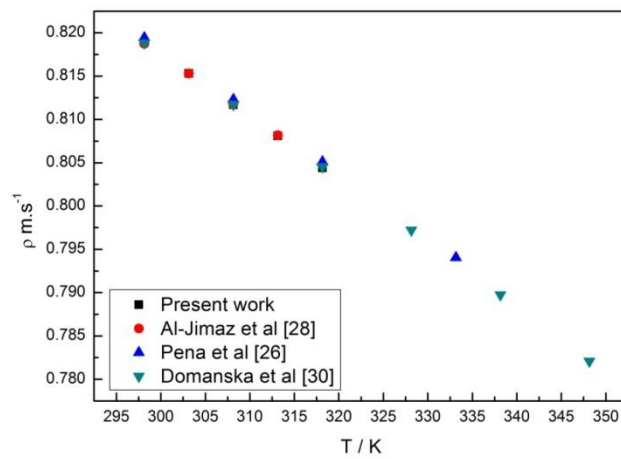


Figure 2 density of 1-heptanol

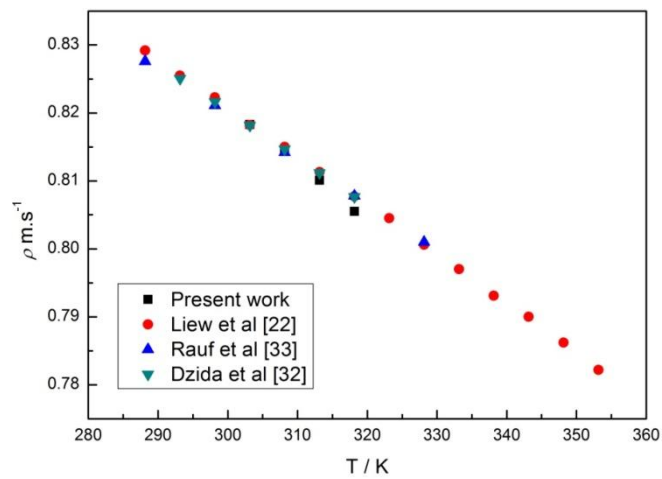


Figure 3 density of 1-octanol

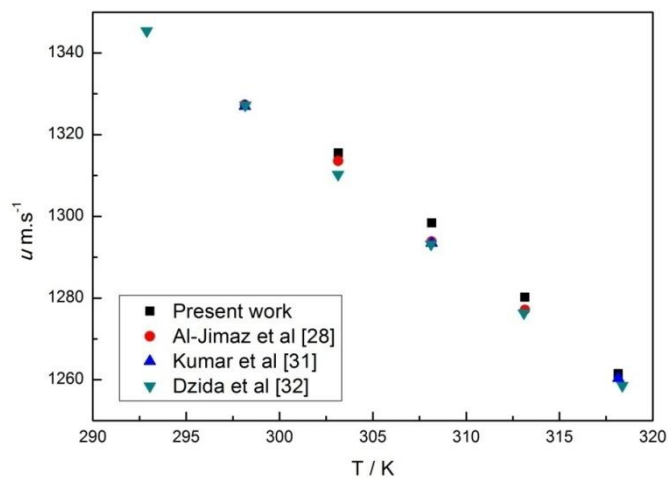


Figure 4 speed of sound of 1-hexanol

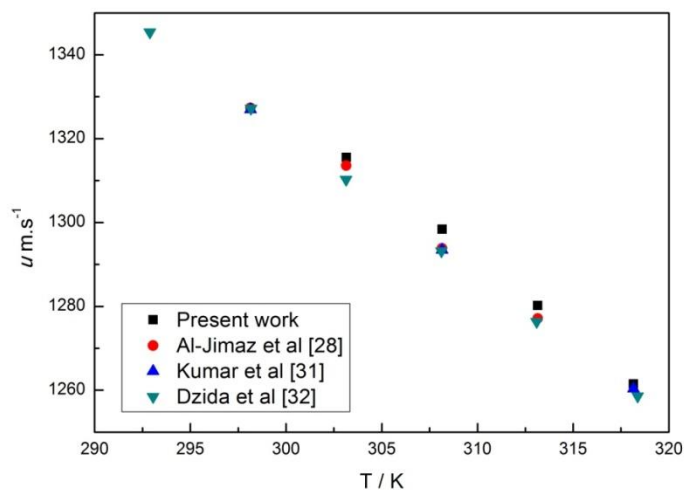


Figure 5s Speed of sound of 1-heptanol

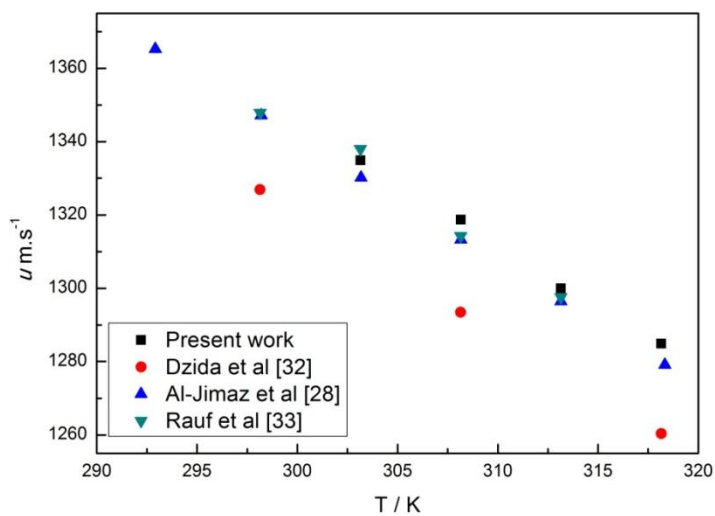


Figure 6s Speed of sound of 1-octanol

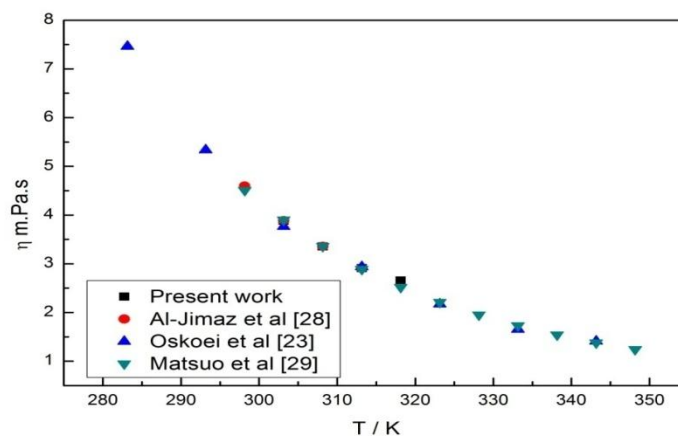


Figure 7s Viscosity of 1-hexanol

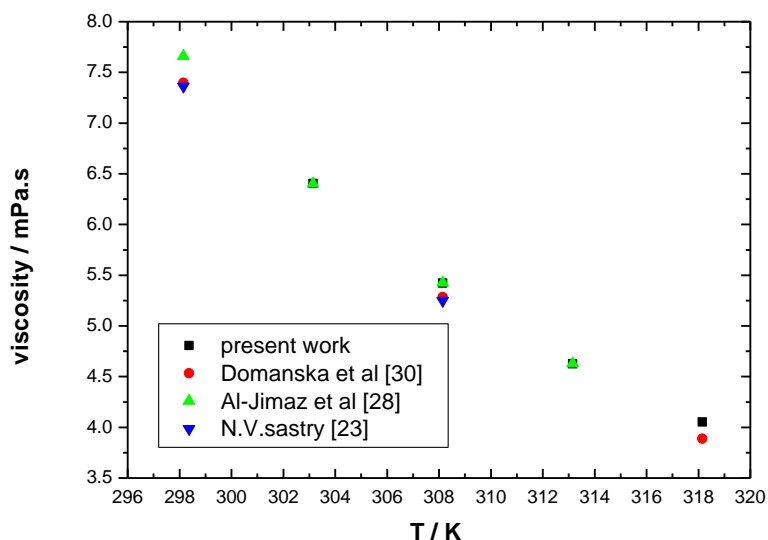


Figure 8s Viscosity of 1-heptanol

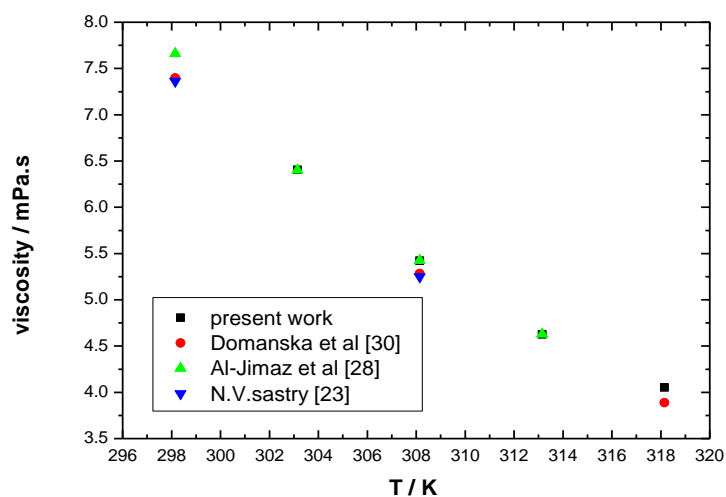


Figure 9s Viscosity of 1-octanol

Table 1S: Coefficients of the polynomial for the correlation of the viscosity as a function of concentration at different temperatures for the N,N-diethylaniline (1) +1-heptanol (2), N,N-diethylaniline (1) +1-octanol (2) binary system

T / (K)	b_0 / (mPa.s)	b_1 / (mPa.s)	b_2 / (mPa.s)	b_3 / (mPa.s)	σ
N,N-diethylaniline (1) +1-heptanol(2)					
303.15	5.054	-7.250	5.439	-1.545	0.013
308.15	4.337	-6.079	4.163	-0.894	0.016
313.15	3.735	-5.528	4.089	-0.910	0.023
318.15	3.407	-6.925	7.510	-2.760	0.046
N,N-diethylaniline (1) +1-octanol(2)					
303.15	6.392	-9.593	7.304	-2.406	0.015
308.15	5.388	-9.414	8.461	-2.936	0.038
313.15	4.585	-8.905	9.174	-3.500	0.045
318.15	4.011	-8.760	9.882	-3.903	0.045
303.15	6.392	-9.593	7.304	-2.406	0.015

Parameters of the equation $u = b_3x_1^3 + b_2x_1^2 + b_1x_1 + b_0$

Table 2S: Coefficients of the polynomial for the correlation of the speed of sound as a function of concentration at different temperatures for the N,N-diethylaniline (1) +1-octanol (2) binary system

$T / (K)$	$b_0 / (m \cdot s^{-1})$	$b_1 / (m \cdot s^{-1})$	$b_2 / (m \cdot s^{-1})$	$b_3 / (m \cdot s^{-1})$	σ
303.15	1334.73	38.91	17.13	41.55	0.471
308.15	1318.02	24.70	50.51	21.56	0.642
313.15	1298.85	13.90	72.33	11.47	1.072
318.15	1284.31	35.19	14.71	43.68	0.741

Parameters of the equation $u = b_3 x_1^3 + b_2 x_1^2 + b_1 x_1 + b_0$

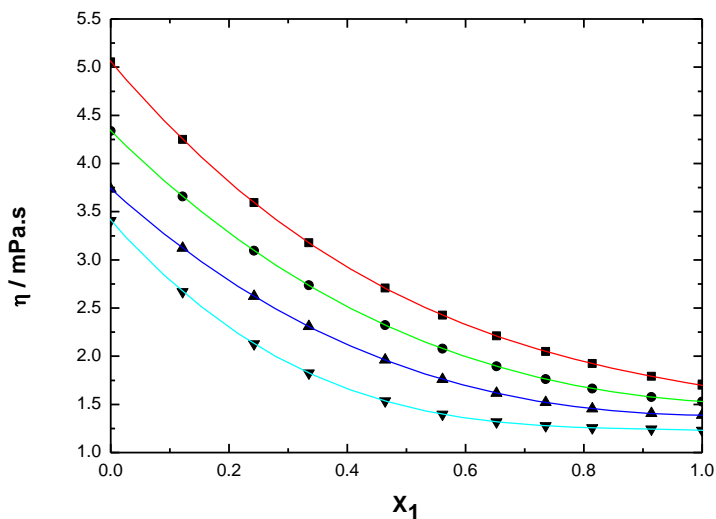


Figure10s: viscosity of binary mixture of N,N-diethylaniline+1-heptanol following the third-order polynomial equation

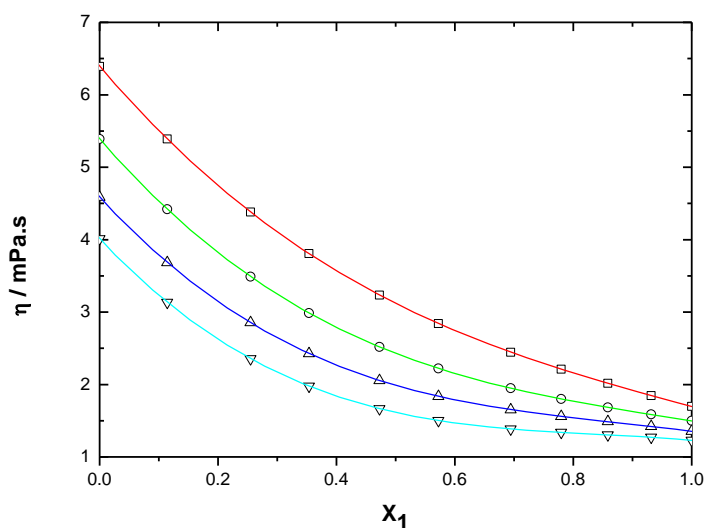


Figure11s: viscosity of binary mixture of N,N-diethylaniline+1-octanol following the third-order polynomial equation

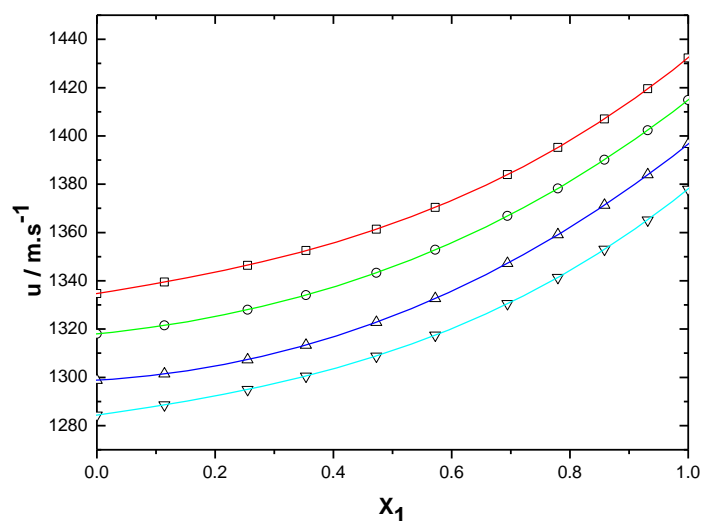


Figure12s: speed of sound of binary mixture of N,N-diethylaniline+1-octanol following the third-order polynomial equation

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