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Research Paper

"Density, Sound Velocity and Viscosity of binary liquid mixtures of 1,3-Dioxolane + alkanols at T = 298.15K and 3 M.Hz."

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Abstract: The excess molar volume (V_m^E) , excess viscosity (η^E) , acoustic impedance (Z^E) and excess adiabatic *compressibility (have been investigated from sound velocity (u), density (ρ) and viscosity (η) measurements of six binary liquid mixtures of 1, 3-dioxolane with pentanol. Hexanol, heptanol, octanol, nonanol* and decanol over the entire range of mole fraction at 298.15K. The excess adiabatic compressibility(β_{ad}^E) have *been calculated from sound velocity measurements of these binary mixtures at 298.15K. The deviation has been fitted by a Redlich-Kister polynomial equation and the results are discussed in terms of molecular interactions. The excess properties are found to be eighter negative or positive depending on the molecular interactions and* nature of the liquid mixtures. Excess properties provide important information in understanding the solute*solvent interaction in a solution.*

KeyWords: 1,3-dioxolane, mole fraction, excess viscosity, adiabatic compressibility, density, sound velocity, molecular interaction.

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I. INTRODUCTION

The sound velocity (u), density (ρ) and viscosity (η) of binary liquid mixtures are used experimentally to understand molecular interaction between the components of the mixtures and find applications in several industries and cosmetics [1-3]. The variation of sound velocity and other ultrasonic parameters of binary liquid mixtures have been studied by many researchers and thy have shed height upon structural changes associated with liquid mixtures of weakly and strongly components [4-10]. The study of molecular association in binary liquid mixtures having alcohol as one of the components is of particular interest, since alcohols are strongly selfassociated liquids having a three-dimensional network of hydrogen bonds and can be associated [11-12].

The thermodynamic and transport properties of a binary liquid mixture such as sound velocity (u), density (ρ) and viscosity (η) are important from practical and theoretical points of view to understand liquid theory. Their properties are extremely useful for the design of process equipment in chemical industries. The data of the properties associated with the liquids and liquid mixtures likesound velocity (u), density (ρ) and viscosity (η) find extensive application in solution theory and molecular dynamics. Such results are necessary for interpretation of data obtained from thermochemical, electrochemical, biochemical and kinetic studies.

The present paper is a part of our ongoing research program in the measurement of thermodynamic and transport properties of binary liquid mixtures. The liquids were chosen in the present investigation on the basis of their industrial impotence.1,3-dioxolane (cyclic diether) have played a major role in the pharmaceutical chemistry. Therefore, the applications of these compounds attract us to study their behavior in alcohols. Alcohols are used as hydraulic fluids in pharmaceutical and cosmetics, in medications for animals, in manufacturing of perfumes, paint removers, flavors and dyestuffs, as defrosting and as an antiseptic agent. The experimental results have been used to discuss the nature of interaction between unlike molecules in terms of hydrogen bonding, dipole-dipole interactions and dispersive forces. It is well known that ethers interact with alcohols by dipole-dipole interaction, formation of new hydrogen bonds or hetro-associations and dispersion forces.

In the present paper several parameters such as adiabatic compressibility (β_{ad}) , molar volume (V_m) and acoustic impedance (Z) of a binary system 1,3-dioxolane with pentanol, hexanol, heptanol, octanol, nonanol and decanol have been reported using the experimental values of sound velocity (u), density (ρ) and viscosity (η) of the binary liquid mixtures at temperature 298.15K. These results have been fitted to the Redlich-Kister polynomial equation.

II. EXPERIMENTAL PROCEDURE

2.1 Chemicals.The source and purity of the chemical compound are shown in table-1.The substances density, viscosity and ultrasonic velocity is compared with the literature data (Table-2) to ascertain the purity, and a good agreement between the experimental data and literature data [13-24] was observed.

2.2 Apparatus and Procedure: All six binary liquid mixtures were prepared by weighing appropriate amounts of pure liquids on a digital electronic balance (Citizen Scale (I) PVT. LTD. Mumbai, India.) with a precision ± 0.1. The experimental uncertainty in mole fractions did not exceed \pm 0.0005. All the solutions were prepared by mass ratios and stored in the air-tight stopper measuring flasks.

Component	Formula	CAS Reg. No.	Supplier	Mass Fraction Purity (%)	Method Purity analysis method
1,3-Dioxolane	C_3H6O_2	646-06-0	CDH Delhi	99.7	Chromatography by the supplier
Pentanol	$C_5H_{12}O$	$71-41-0$	CDH Delhi	99.7	Chromatography by the supplier
Hexanol	$C_6H_{14}O$	$111 - 27 - 3$	CDH Delhi	99.5	Chromatography by the supplier
Heptanol	C ₇ H16O	$111 - 70 - 6$	CDH Delhi	99	Chromatography by the supplier
Octanol	$C_8H_{18}O$	111-87-5	CDH Delhi	99.7	Chromatography by the supplier
Nonanol	$C_9H_{20}O$	143-08-8	CDH Delhi	99	Chromatography by the supplier
Decanol	$C_{10}H_{22}O$	$112 - 30 - 1$	CDH Delhi	99	Chromatography by the supplier

Table 1. CAS Registry Number, Mass Fraction Purity of the chemicals

2.3 Measurements:

Density: Densities of pure components and liquid-liquid mixtures were measured with a 25-ml specific gravity bottle by relative measurement method with an accuracy of \pm 0.01 kg.m⁻³. The specific gravity bottle with the experimental mixture was immersed in the temperature-controlled water bath (MSI Goyal scientific, Meerut, U.P. India.), operating in the temperature range of -10⁰C to 85⁰C with an accuracy \pm 0.1⁰C. Double distilled water used for the calibration of the specific gravity bottle. At least three times for each composition in experimental were generally repeated and the results were treatment.

Sound velocity:

The speed of sound (u) was measured at a frequency 3 MHz in these solutions using the interferometric method with a (Model F-80D, Mittal Enterprise, New Delhi, India) at 298.15K. The interferometer cell was filled with the test liquid, and water was circulated around the measuring cell from a water bath. The uncertainty was estimated to be ± 0.1 %. The measured values of ultrasonic velocities of pure1,3-dioxolane with pentanol. Hexanol, heptanol, octanol, nonanol and decanol compare well with the corresponding literature values.

Viscosity:

The viscosities of pure liquids and their binary mixtures were measured by using a Ostawald's viscometer. The viscometer was calibrated with doubly distilled water and benzene, liquid was allowed to stand for about 30 minutes in a thermostatic water bath so that the thermal fluctuations in viscometer were minimized. The accuracy in viscosity data was \pm 0.0005 mPa.s. The flow time of pure liquids and liquid mixtures were repeated for five times. The efflux Time was measured with an electronic stopwatch (Racer) with a time resolution (± 0.015) , and an average of at least five flow time readings was taken. Glass stopper was placed at the opening of the viscometer to prevent the loss due to evaporation during measurements. The measured values of viscosities of pure 1,3-dioxolane with Pentanol, Hexanol, Heptanol, Octanol, Nonanol and Decanol compare well with the corresponding literature values.

III. THEORETICAL

The experimentally measured ultrasonic velocity (u), density (ρ) and viscosity (η) are used to evaluate derived parameters like molar volume (V_m), adiabatic compressibility (β_{ad}), and acoustic impedance (Z) using well established relations.

The molar volume (V_m) of binary liquid mixtures were calculated by using a following equation:

$$
V_m = \frac{(X_1 M_1 + X_2 M_2)}{\rho} \tag{1}
$$

The adiabatic compressibility(β_{ad}) has been calculated from the ultrasonic velocity (u) and density (ρ) of the medium using the equation as

$$
\beta_{ad} = \frac{1}{u^2 \rho} \tag{2}
$$

The acoustic impedance is the parameter related to elastic properties of the medium and calculated by using the expression

$$
Z = \rho. U \tag{3}
$$

where ρ is the density and u is the sound velocity.

The excess value of ultrasonic related parameters has been calculated by using the following relation $A^E = A_{exp.} - (X_1 A_1 + X_2 A_2)$ (4)

Where A represents the parameter such as intermolecular free length, molar volume, isentropic compressibility, viscosity and internal pressure and X_1 and X_2 is the mole fractions of components whose parameters.

IV. RESULT AND DISCUSSION

The experimentally determined values of density (ρ), sound velocity (u) and viscosity (η) and derived parameters adiabatic compressibility(β_{ad}), molar volume (V_m) and acoustic impedance (Z) at 298.15K for the binary liquid system 1,3-Dioxolane with Pentanol, Hexanol, Heptanol, Octanol, Nonanol and Decanol are listed in table-3. The same excess values for the binary liquid mixtures 1,3-Dioxolane with Pentanol, Hexanol, Heptanol, Octanol, Nonanol and Decanol are presented in table-4. The data related to excess adiabatic compressibility, excess viscosity and excess acoustic impedance for the binary liquid system 1,3-Dioxolane with Pentanol, Hexanol, Heptanol, Octanol, Nonanol and Decanol were graphically represented in figures 1 to 3 at 298.15K respectively.

From the table-3, it was observed that the density and ultrasonic velocity increase with increasing mole fraction of 1,3-Dioxolane while the viscosity decrease. This may be due to association of a very strong dipoleinduced dipole interaction between the component molecules.

Excess Molar Volume (V_m^E)

The excess molar volume (V_m^E) data of all the binary mixtures of 1,3-Dioxolane with Pentanol, Hexanol, Heptanol, Octanol, Nonanol and Decanol are presented in table-4 at 298.15K.

A perusal of table-4 indicates that the values of excess molar volume (V_m^E) data for the binary mixtures of 1,3-Dioxolane with Pentanol, Hexanol, Heptanol, Octanol, Nonanol and Decanol are positive. On other hand, an inversion in sign for the binary mixtures containing 1,3-Dioxolane with Pentanol, Hexanol, Heptanol, Octanol, Nonanol and Decanol was observed. Further, it is observed the magnitude of positive excess molar volume (V_m^E) values decrease with increase in composition of 1,3-Dioxolane. According to Marcus [25], the molecules of alkanols are associated through hydrogen bonding in pure state. Mixing these alcohol molecules with polar molecule like 1,3-Dioxolane would induce mutual dissociation of the hydrogen-bonded structure present in pure alcohols with subsequent formation of inter molecular hydrogen bonds (O-----OH) between the oxygen atom of ether group of 1,3-Dioxolane molecule and hydrogen atom of hydroxyl group of alcohols. The positive excess molar volume (V_m^E) values suggest that the higher alcohols less proton donating ability than the lower alcohols. Hence hetro association affects decrees in the binary liquid mixtures with an increase of chain length of linear alcohols. The algebraic values of excess molar volume (V_m^E) for the mixtures of 1,3-Dioxolane with Pentanol, Hexanol, Heptanol, Octanol, Nonanol and Decanol fall in the order,

Pentanol < Hexanol < Heptanol < Octanol < Nonanol < Decanol

The order suggests that dipole-dipole interaction between unlike molecules decrease with increasing in chain length of alcohols as consequences of these decreases the degree of polymerization in the pure state. These experimental results suggest that the positive excess molar volume (V_m^E) indicate that, the breaking up off self-associated structure of the components of the mixtures is dominant over the effect of H-bonding and dipoledipole interaction between unlike molecules.

Excess adiabatic compressibility (β_{ad}^E)

The excess adiabatic compressibility (β_{ad}^E) data of all the binary mixtures of 1,3-Dioxolane with Pentanol, Hexanol, Heptanol, Octanol, Nonanol and Decanol are graphically presented Figures 1 at 298.15 K. An examination of curves in Figure 1 shows that the values of excess adiabatic compressibility (β_{ad}^E) data for 1,3-Dioxolane with Pentanol, Hexanol, are negative and for the remaining binary mixtures excess adiabatic compressibility (β_{ad}^E) is positive over the entire composition range at 298.15 K.

The excess adiabatic compressibility (β_{ad}^E) values were ascribed according to Sri Devi et al [26] the negative excess values have been due to the closely packed molecules which account for existence of strong molecular interaction where as positive excess values are due to prevailing of dispersion forces between unlike molecules. The sign of excess adiabatic compressibility (β_{ad}^E) and deviation in intermolecular free length (ΔL_f) play a vital role in assessing the compactness due to molecular interaction in liquid mixtures through hydrogenbonding, charge-transfer complex formation and dipole-dipole interactions and dipole-induced dipole interactions, interstitial accommodation and orientational ordering [27] leading to more compact structure making negative excess isentropic compressibility and excess intermolecular free length values.

A perusal of curves in Figures-1 shows that the excess adiabatic compressibility (β_{ad}^E) negative value decreases may be attributed to hetero association complexes decrease with increasing chain length, probably due to less proton-donating ability of higher alcohols. Experimental results suggest that the negative values of excess

adiabatic compressibility (β_{ad}^E) and deviation in intermolecular free length (ΔL_f) for binary mixtures attributed to the dipole-dipole interactions through formation of complexes between the molecules of mixing components and the positive values of excess adiabatic compressibility (β_{ad}^E) and deviation in intermolecular free length (ΔL_f) for binary mixtures may be due to the domination of dispersion forces over formation of complexes between unlike molecules.

The excess adiabatic compressibility (β_{ad}^E) values of 1,3-Dioxolane with alcohols fall in the order:

Decanol < Nonanol < Octanol < Heptanol < Hexanol < Pentanol

Excess Viscosity (η^E)

The excess viscosity (η^E) data of all the binary mixtures of 1,3-Dioxolane with Pentanol, Hexanol, Heptanol, Octanol, Nonanol and Decanol are graphically presented Figures 2 at 298.15 K. An examination of curves in Figure 2 shows that the values of excess viscosity (η^E) data for 1,3-Dioxolane with Pentanol, Hexanol, Heptanol, Octanol, Nonanol and Decanol are negative over the entire composition range at 298.15 K**.**

The measurement of viscosity in binary liquid mixture provides some reliable information in the study of molecular interaction. Table- 3 shows that the viscosity decrease with increase in concentration of 1,3- Dioxolane molecule. More insight about molecular interaction can be obtained by excess viscosity (η^E) values.

According to Fort and Moore, the excess viscosity gives the strength of the molecular interaction between the interacting molecules. The excess value of viscosity at the six binary mixtures 1,3-Dioxolane + Pentanol, 1,3-Dioxolane + Hexanol, 1,3-Dioxolane + Heptanol, 1,3-Dioxolane + Octanol, 1,3-Dioxolane + Nonanol and 1,3-Dioxolane + Decanol at the 298.15 K are reported in Table- 4. The Figure -2 represents the variation of excess viscosity (η^E) is found to be negative for all six binary liquid mixtures over the entire composition range at the 298.15 K. Which suggest the presence of weak intermolecular interactions. For systems where dispersion, induction and dipolar forces are operating, the values of excess viscosity are found to be negative, whereas the existence of specific interaction leading to the formation of complexes in mixtures tends to make positive. The excess viscosity is negative through the whole range of concentration in all the studied systems. The large negative values of excess viscosity for all systems can be attributed to the presence of dispersion, induction and dipolar forces between the components

The negative excess viscosity (η^E) for all the six binary liquid mixtures (1,3-Dioxolane + Pentanol, 1,3-Dioxolane + Hexanol, 1,3-Dioxolane + Heptanol, 1,3-Dioxolane + Octanol, 1,3-Dioxolane + Nonanol and 1,3- Dioxolane + Decanol) studied are indicative of the predominance of dispersion forces and further their magnitudes increase from pentanol to decanol (C_5-C_{10}) , hence suggesting an increase in dispersion forces in the same order . Alcohols are good solvent that can dissolve both the polar and non-polar components. The hydrophilic –OH group of alcohols can dissolve the polar whereas the short hydrophobic hydrocarbon group can dissolve the non-polar. Alcohols are strongly self-associated liquids with a three dimensional network of hydrogen bonds and can be associated with any other group having some degree of polar attraction. The associative alcohols molecule act as proton donor enabling hydrogen bonding with 1,3-Dioxolane molecule. In

the system studied, the complex formation is likely to occur between $H^{\delta+}$ of alcohol and $O^{\delta-}$ of ether group of 1,3-Dioxolane. Hence in the present study there is existence of solute-solvent interactions. The algebraic values of excess viscosity for binary mixtures of 1,3-Dioxolane with Pentanol, Hexanol, Heptanol, Octanol, Nonanol and Decanol fall in the order:

Decanol < Nonanol < Octanol < Heptanol < Hexanol < Pentanol

In the alkanol mixture, the 1,3-Dioxolane is completely dissolved and so no changes of hydrogen bond raptures and only the interaction with the 1,3-Dioxolane ring and the active group of alkanols, which are mostly dispersive in nature. The increase in mole fraction of 1,3-Dioxolane increase the net dispersive interaction and hence the velocity continuously increases as observed. As the mole fraction of 1,3-Dioxolane increases, the hydrogen bond repture of the boat form is of considerable extent and they leads to additional dipole type interaction. 1,3-Dioxolane being non-polar the predominant dispersive type interactions with temporary dipolar type are existing as a net result of intermolecular forces in all systems.

Acoustic impedance (Z)

The excess acoustic impedance (Z^E) data of all the binary mixtures of 1,3-Dioxolane with Pentanol, Hexanol, Heptanol, Octanol, Nonanol and Decanol are graphically presented Figures-3 at 298.15 K. An examination of curves in Figure- 3 shows that the values of excess acoustic impedance (Z^E) data for 1,3-Dioxolane with Pentanol, Hexanol, Heptanol, Octanol, Nonanol and Decanol are negative over the entire composition range at 298.15 K**.**

The variation of excess acoustic impedance (Z^E) with mole fraction of 1,3-Dioxolane at 298.15K are shown in figure-3.It is observed the excess acoustic impedance (Z^E) data for 1,3-Dioxolane with Pentanol, Hexanol, Heptanol, Octanol, Nonanol and Decanol are negative over the entire composition range at 298.15 K**.** This is in agreement with requirement as both ultrasonic velocity and density increase with increase in concentration of the solute and also effective due to solute – solvent interaction.

V. CONCLUSIONS

From experimental results, negative excess molar volume (V_m^E) and excess adiabatic compressibility (β_{ad}^E) values can be attributed to the dipole –dipole interactions between unlike molecules through hydrogen bonding and positive values indicate that the effect due to 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 The positive values of Excess molar volume (V_m^E) and excess adiabatic compressibility (β_{ad}^E) may be attributed to the formation of hydrogen bonding (O-H….O) resulting in the formation of complexes between the component molecules and negatives values suggesting breaking of the self-associated alcohols and

weak interactions between unlike molecules. From these data, several thermodynamic excess functions have been calculated and correlated using the Redlich – Kister type polynomial equation. The sign and magnitude of these quantities have been discussed in terms of hydrogen bond, electron-transfer complexes and dipole-dipole interactions between the component molecules.

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Data availability statement

All data generated or analyzed during this study are included in this published article.

Table 3. Experimental Values of density (ρ), sound velocity (u) and viscosity (), derived parameters adiabatic compressibility(β_{ad}), molar volume (V_m) and acoustic impedance (Z) for the binary mixtures of **1,3-Dioxolane(1) + Alkanols (2) at 298.15K.**

0.7975 | 0.9676 | 1348 | 0.9141 | 5.6875 | 0.0883 | 0.1304

0.8940	1.0104	1348	0.7652	5.4466	0.0792	0.1362	
1.0000	1.0616	1340	0.5885	5.2460	0.0697	0.1422	
		$1,3$ -Dioxolane + Nonanol					
Ω	0.8248	1366	8.9258	6.4976	0.1749	0.1126	
0.0876	0.8336	1366	6.8601	6.4289	0.1656	0.1138	
0.1913	0.8404	1363	5.8531	6.4051	0.1556	0.1145	
0.2942	0.8504	1359	4.4022	6.3671	0.1453	0.1155	
0.3963	0.8692	1355	3.1558	6.2662	0.1339	0.1177	
0.4959	0.8844	1352	2.3340	6.1859	0.1237	0.1195	
0.6050	0.9092	1349	1.7321	6.0439	0.1119	0.1226	
0.6947	0.9332	1346	1.3334	5.9145	0.1023	0.1256	
0.7993	0.9648	1343	0.9642	5.7466	0.0913	0.1295	
0.9013	1.0084	1340	0.8031	5.5228	0.0803	0.1351	
	1.0616	1340	0.5885	5.2460	0.0697	0.1422	
1,3-Dioxolane + Decanol							
Ω	0.8292	1378	11.8027	6.4976	0.1908	0.1142	
0.0881	0.8364	1374	8.5615	6.4289	0.1803	0.1149	
0.191	0.8396	1370	7.8207	6.4051	0.1693	0.1150	
0.2921	0.8560	1366	5.5340	6.3671	0.1561	0.1169	
0.3937	0.8672	1362	4.2319	6.2662	0.1442	0.1181	
0.4956	0.8824	1358	3.4173	6.1859	0.1320	0.1198	
0.604	0.9076	1353	2.5370	6.0439	0.1183	0.1227	
0.7129	0.9308	1348	1.5262	5.9145	0.1055	0.1254	
0.7983	0.9616	1344	1.1637	5.7466	0.0946	0.1292	
0.8971	1.0040	1340	0.8623	5.5228	0.0824	0.1345	
	1.0616	1340	0.5885	5.246	0.0697	0.1422	

Table-4 Excess adiabatic compressibility (β_{ad}^E) **, excess viscosity** (η^E) **, excess molar volume** (V_m^E) **and excess acoustic impedance (Z^E) for the binary mixtures of 1,3-Dioxolane (1) + Alkanols (2) at 298.15K.**

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