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Thermo acoustical study of intermolecular interactions in binary liquid mixtures of 1,3-dioxolane with1-alkanols at 298.15K and 3 MHz.

Dhirendra Kumar Sharma[1] and Arpit Babu Shukla[2]
[1,2]Department of Chemistry, Institute of Basic Science, Bundelkhand University, Jhansi (U.P), India.

Abstract: *Ultrasonic velocity and density of binary liquid mixtures of 1,3-dioxolane with1-alkanols has been measured using ultrasonic interferometer and 30 ml gravity bottle respectively at frequency 3MHz and constant temperature of 298.15K. Experimental values then obtained are used for determining various thermo acoustical parameters like adiabatic compressibility and intermolecular free length. Graphs are plotted of each parameter against mole fraction. The linear variation in most of the acoustical parameters showed that there is no complex formation in the mixture. The weak interaction between the molecules of the binary mixture is founded. Acoustic parameters provide important information in understanding the solute-solvent interaction in a solution. These results emphasize the value of excess parameters in describing solute–solvent interactions by confirming that the degree and kind of departures from ideality are significantly impacted by the polarity and structural features of the constituents involved.*

Keywords: *Density, sound velocity, adiabatic compressibility, free length, hydrogen bonding, 1,3-dioxolane, molecular interaction.*

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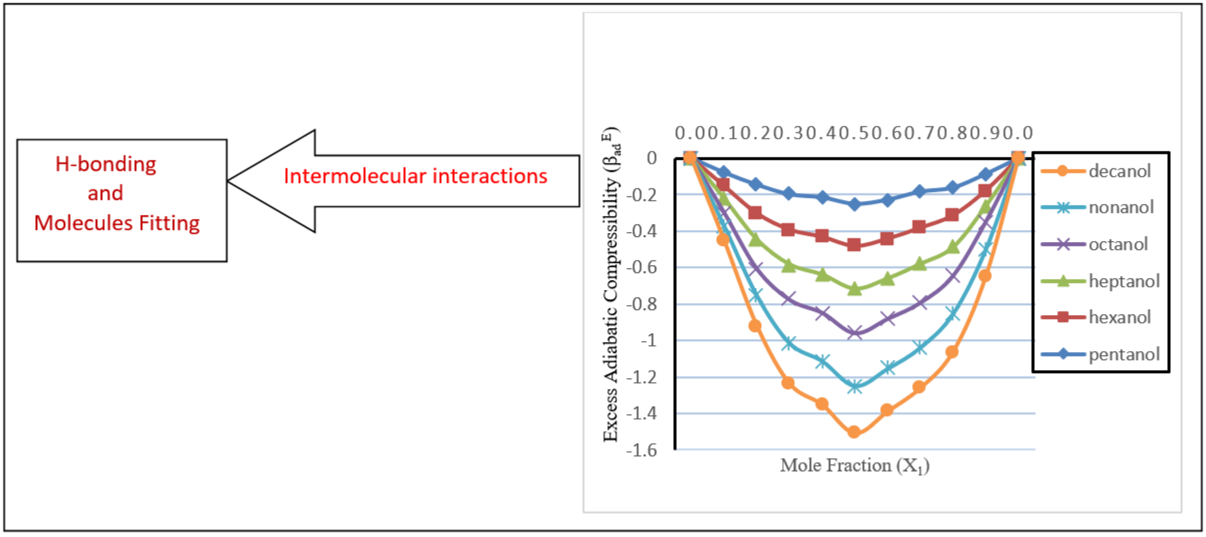
Graphical abstract



Nomenclature
ρ, Densities of liquid
u, Ultrasonic velocity
β_{ad}, Adiabatic compressibility
β_{ad}^E, excess adiabatic compressibility
(L_f), Inter molecular free length
(L_f^E), Excess inter molecular free length
X_1, Mole fraction of 1,3-Dioxolane
Y^E, Thermodynamic excess function

I. Introduction:

Ultrasonic technique offers a rapid non-destructive methods for characterizing material [1]. The ultrasonic velocity in a liquid is fundamentally related to the binding forces between atoms or molecules and has been successfully employed in the field of interactions and structural aspect studies, for characterizing the physicochemical behavior of liquid mixtures [2-5]. The ultrasonic velocity along with density and viscosity furnish wealth of information about the interaction between ions, dipoles, hydrogen bonding, multi-polar and dispersive forces [6]. Ultrasonic propagation parameters yield valuable information regarding the behaviour of liquid systems because intermolecular and intra molecular association, complex formation, dipolar interactions and related structural changes affect the compressibility of the system which in turn produces corresponding variations in ultrasonic velocity [7]. The different acoustical parameters interpret the nature and strength of molecular interaction that exist in the system [8]. The intermolecular interactions influence the structural arrangement along with the shape of the molecules [9, 10]. The study of liquids under ultrasonic conditions is crucial for determining the type and intensity of molecular interactions. Numerous studies investigating how molecules interact in liquid mixtures have been conducted using a variety of techniques, including the ultrasonic method, nuclear magnetic resonance, the Raman effect, infrared, dielectric constant, and ultraviolet. The ultrasonic method has recently emerged as a potent instrument for supplying data about the physiochemical characteristics of liquid systems [11,12].

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 study, an attempt has been made to investigate the behaviour of binary mixtures of 1,3-dioxolane with 1-pentanol, 1-hexanol, 1-heptanol, 1-octanol, 1-nonanol and 1-decanol by measuring ultrasonic velocity and density at 298.15K. From the measured data various derived acoustical parameters are computed and the results are interpreted in the light of molecular interaction. In the present paper several parameters such as adiabatic compressibility (β_{ad}), and free length (L_f) of a binary system 1,3-dioxolane with 1-pentanol, 1-hexanol, 1-heptanol, 1-octanol, 1-nonanol and 1-decanol have been reported using the experimental values of sound velocity (u) and density (ρ) 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 ± 0.0005. All the solutions were prepared by mass ratios and stored in the air-tight stopper measuring flasks.

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

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_7H16O	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 2. Comparison of Experimental and Literature density (ρ) and sound velocity (u) of pure Components with Available Literature Values at T = 298.15K.

Compound	ρ (g.cm^{-3})		u (m.s^{-1})	
	This work	**Literature**	**This work**	**Literature**
1,3-Dioxolane	1.0616	1.0577[17]	1340	1338[17]
		1.0586[17]		1338[18]
Pentanol	0.8124	0.8108[13]	1198	1197[16]
		0.8107[13]		1268[22]
Hexanol	0.8176	0.8187[13]	1306	1304[15]
		0.8152[15]		1303[15]
Heptanol	0.8196	0.8187[13]	1325	1327[15]
		0.8197[19]		1327.37[24]
Octanol	0.8236	0.8216[13]	1350	1348[14]
		0.8218[13]		1347[22]
Nonanol	0.8248	0.8244[15]	1366	1365[15]
		0.824224[15]		1364[24]
Decanol	0.8292	0.8267[15]	1378	1380[15]
		0.8264[19]		1379[24]

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 ± 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 -10^{0}C to 85^{0}C with an accuracy ± 0.1^{0}C. 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 1-pentanol. 1-hexanol, 1-heptanol, 1-octanol, 1-nonanol and 1-decanol compare well with the corresponding literature values.

Theoretical:

The experimentally measured ultrasonic velocity (u) and density (ρ) are used to evaluate derived parameters like adiabatic compressibility (β_{ad}) and free length (L_f) using well established relations.

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

$$\beta_{ad} = \frac{1}{u^2\rho} \qquad (1)$$

In the year 1952, Jacobson, suggested an empirical relation for calculating the free length (L_f) of liquids.

Intermolecular free length (L_f), can be calculated from the adiabatic compressibility(β_{ad}) by the relation given below

$$L_f = K\,\beta_{ad}^{1/2} \qquad (2)$$

Where K is temperature dependent constant and β_{ad} is the adiabatic compressibility, which is given by the relation

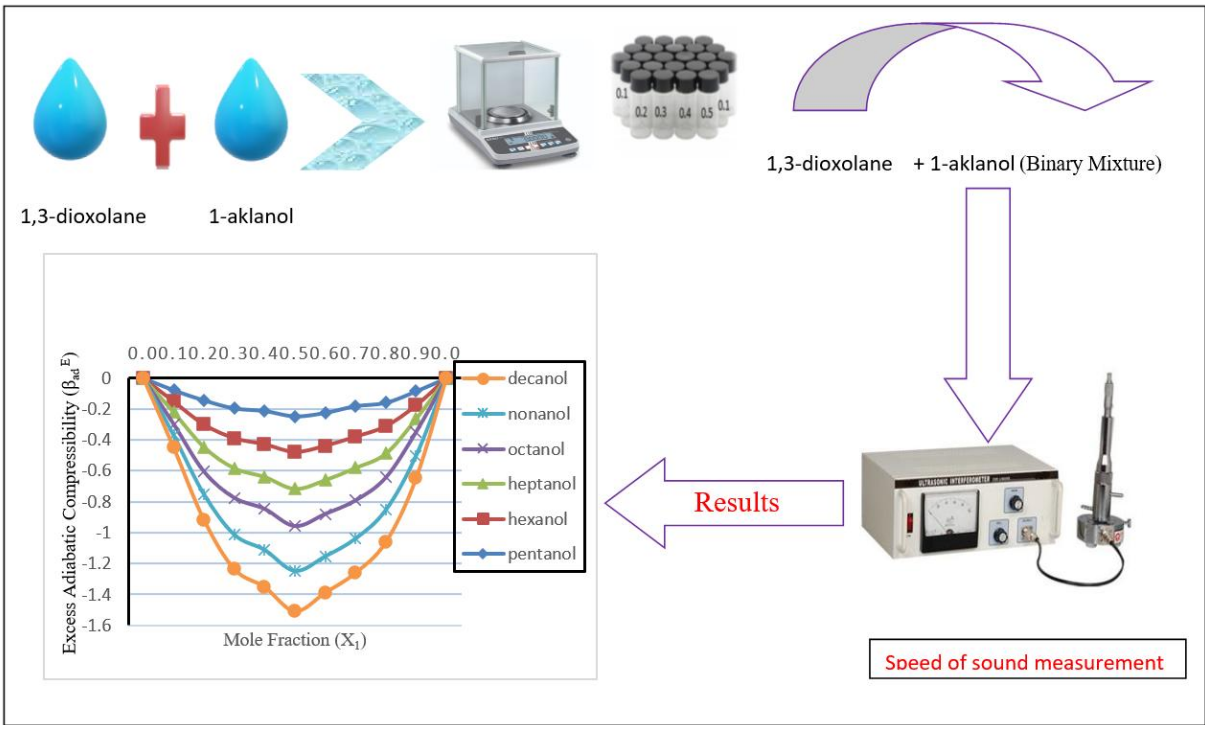
The excess value of ultrasonic related parameters has been calculated by using the following relation

$$A^E = A_{exp.} - (X_1 A_1 + X_2 A_2) \qquad (3)$$

Where A represents the parameter such as intermolecular free length, molar volume, isentropic compressibility, viscosity and internal pressure and X_1 andX_2 is the mole fractions of components whose parameters.

III. Result and Discussion:

The experimentally determined values of density (ρ) and sound velocity (u) and derived parameters adiabatic compressibility(β_{ad}) and free length (L_f) at 298.15K for the binary liquid system 1,3-Dioxolane with 1-pentanol, 1-hexanol, 1-heptanol, 1-octanol, 1-nonanol and 1-decanol are listed in table-2. The same excess values for the binary liquid mixtures 1,3-Dioxolane with 1-pentanol, 1-hexanol, 1-heptanol, 1-octanol, 1-nonanol and 1-decanol are presented in table-2. The data related to excess adiabatic compressibility and excess free length for the binary liquid system 1,3-Dioxolane with1-pentanol, 1-hexanol, 1-heptanol, 1-octanol, 1-nonanol and 1-decanol were graphically represented in figures 1 to 2 at 298.15K respectively.



Scheme 1. Interactions between1,3-dioxolane with 1-alkanols at 298.15K

From the table-2, it was observed that the density and ultrasonic velocity increase with increasing mole fraction of 1,3-Dioxolane. This may be due to association of a very strong dipole- induced dipole interaction between the component molecules.

Table 3. Experimental Values of density (ρ), sound velocity (u) and viscosity (η), derived parameters adiabatic compressibility(β_{ad}), free length (L_f), excess intermolecular free length (L_f) and excess adiabatic compressibility (β_{ad}^E) for the binary mixtures of 1,3-Dioxolane(1) + 1-Alkanols (2) at 298.15K.

Mole fraction 1,3-Dioxolane (x_1)	Density (ρ) g.cm^{-3}	Sound velocity (u) ms^{-1}	Intermolecular free length (L_f) × 10^{-4} /m	Adiabatic compressibility (β_{ad}) × 10^{-7} Pa^{-1}	Excess intermolecular free length(L_f) × 10^{-4} / m	Excess adiabatic compressibility (β_{ad}^E)× 10^{-7}Pa^{-1}
1,3-Dioxolane + Pentanol						
0	0.8124	1198	2.6732	8.5770	-	-
0.0939	0.8276	1284	2.2842	7.3290	0.2915	-0.9352
0.1942	0.8436	1290	2.2201	7.1233	0.2515	-0.8068
0.2941	0.8640	1296	2.1477	6.8909	0.2202	-0.7064
0.3942	0.8836	1300	2.0872	6.6966	0.1767	-0.5673
0.4787	0.9068	1304	2.0213	6.4853	0.1549	-0.4975
0.5999	0.9316	1310	1.9495	6.2551	0.1009	-0.3236
0.6972	0.9596	1318	1.8697	5.9991	0.0797	-0.2555
0.7928	0.9876	1324	1.8003	5.7762	0.0498	-0.1599
0.9035	1.0260	1332	1.7121	5.4934	0.0231	-0.0740
1.0000	1.0616	1340	1.6350	5.246	-	-
1,3-Dioxolane + Hexanol						
0	0.8176	1306	2.2349	7.1709	-	-
0.0912	0.8252	1317	2.1775	6.9867	0.0027	0.0086
0.1955	0.8432	1320	2.1214	6.8065	0.0037	0.0119
0.2923	0.8584	1322	2.0775	6.6657	0.0179	0.0574
0.3982	0.8792	1325	2.0192	6.4786	0.0232	0.0742
0.4942	0.8992	1327	1.9619	6.3154	0.0298	0.0958
0.6059	0.9264	1330	1.9019	6.1024	0.0305	0.0978
0.6976	0.9508	1332	1.8475	5.9279	0.0310	0.0998
0.8018	0.9836	1335	1.7779	5.7045	0.0239	0.0769
0.8914	1.0168	1337	1.7147	5.5018	0.0145	0.0468
1.0000	1.0616	1340	1.6350	5.2460	-	-
1,3-Dioxolane + Heptanol						
0	0.8196	1325	2.1660	6.9497	-	-
0.0928	0.8304	1334	2.1091	6.7671	0.0076	0.0245
0.1905	0.8412	1334	2.0820	6.6802	0.0172	0.0551

0.2939	0.8592	1335	2.0353	6.5304	0.0254	0.0814
0.3894	0.8740	1335	2.0009	6.4199	0.0417	0.1336
0.4818	0.8916	1336	1.9584	6.2837	0.0482	0.1548
0.6021	0.9184	1337	1.8984	6.0912	0.0521	0.1673
0.6952	0.9420	1337	1.8509	5.9387	0.0441	0.1734
0.7892	0.9756	1338	1.7845	5.7255	0.0376	0.1204
0.9006	1.0156	1339	1.7116	5.4918	0.0238	0.0765
1.0000	1.0616	1340	1.6350	5.2460	-	-
1,3-Dioxolane + Octanol						
0	0.8296	1350	2.0764	6.6622	-	-
0.0885	0.8296	1350	2.0614	6.6139	0.0241	0.0770
0.1967	0.8464	1349	2.0235	6.4923	0.0339	0.1087
0.2998	0.8560	1348	2.0038	6.4291	0.0597	0.1915
0.3902	0.8712	1348	1.9688	6.3168	0.0646	0.2072
0.4963	0.8876	1348	1.9324	6.2002	0.0751	0.2409
0.6008	0.9140	1347	1.8794	6.0301	0.0682	0.2188
0.6925	0.9340	1348	1.8364	5.8921	0.0657	0.2106
0.7975	0.9676	1348	1.7726	5.6875	0.0482	0.1547
0.8940	1.0104	1348	1.6975	5.4466	0.0157	0.0505
1.0000	1.0616	1340	1.6350	5.2460	-	-
1,3-Dioxolane + Nonanol						
0	0.8248	1366	2.0251	6.4976	-	-
0.0876	0.8336	1366	2.0037	6.4289	0.0128	0.0409
0.1913	0.8404	1363	1.9963	6.4051	0.0458	0.1469
0.2942	0.8504	1359	1.9844	6.3671	0.0741	0.2377
0.3963	0.8692	1355	1.9529	6.2662	0.0825	0.2646
0.4959	0.8844	1352	1.9279	6.1859	0.0963	0.3089
0.6050	0.9092	1349	1.8837	6.0439	0.0946	0.3035
0.6947	0.9332	1346	1.8434	5.9145	0.0893	0.2864
0.7993	0.9648	1343	1.7910	5.7466	0.0777	0.2494
0.9013	1.0084	1340	1.7213	5.5228	0.0478	0.1533
1	1.0616	1340	1.6350	5.2460	-	-
1,3-Dioxolane + Decanol						
0	0.8292	1378	1.9794	6.4976	-	-
0.0881	0.8364	1374	1.9638	6.4289	0.0247	0.0794
0.191	0.8396	1370	1.9578	6.4051	0.0642	0.2059
0.2921	0.8560	1366	1.9413	6.3671	0.0725	0.2325
0.3937	0.8672	1362	1.9374	6.2662	0.0936	0.3002
0.4956	0.8824	1358	1.9153	6.1859	0.1066	0.3418
0.604	0.9076	1353	1.8759	6.0439	0.1005	0.3352
0.7129	0.9308	1348	1.8427	5.9145	0.9088	0.3492
0.7983	0.9616	1344	1.7943	5.7466	0.0898	0.2882
0.8971	1.0040	1340	1.7288	5.5228	0.0584	0.1873
1	1.0616	1340	1.6350	5.246	-	-

Excess adiabatic compressibility (β_{ad}^E)

The calculated excess adiabatic compressibility (β_{ad}^E), values for the binary liquid mixture listed in Figure 1. The change of this property has been shown in figure-1. The excess adiabatic compressibility (β_{ad}^E), values are negative over the entire mole fraction range and become more negative with increasing the mole fraction of second component for all binary mixtures. These results can be explained in term of molecular interactions and structured effects. The variation of excess adiabatic compressibility (β_{ad}^E), with volume fraction of 1,3-dioxolane (x_1) is represent in fig.1. Fort and Moore [25] have suggested that excess adiabatic compressibility (β_{ad}^E), is the result of several opposing effects. Strong molecular interactions occur through charge transfer, dipole- induced dipole and dipole-dipole interaction [26], interstitial accommodation and orientational ordering all lead to a more compact structure making excess adiabatic compressibility (β_{ad}^E) negative. The negative values of (β_{ad}^E), in these mixtures can be associated with a structure forming tendency.

The excess adiabatic compressibility $(\boldsymbol{\beta_{ad}^E})$ values were ascribed according to Sri Devi et al [27] 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 $(\boldsymbol{\beta_{ad}^E})$ and deviation in intermolecular free length (ΔL_f) play a vital role in assessing the compactness due to molecular interaction in liquid mixtures through hydrogen-bonding, charge-transfer complex formation and dipole-dipole interactions and dipole-induced dipole interactions, interstitial accommodation and orientational ordering 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 $(\boldsymbol{\beta_{ad}^E})$ 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. The excess adiabatic compressibility $(\boldsymbol{\beta_{ad}^E})$ values of 1,3-Dioxolane with alcohols fall in the order:

1-Decanol < 1-Nonanol < 1-Octanol < 1-Heptanol < 1-Hexanol < 1-Pentanol

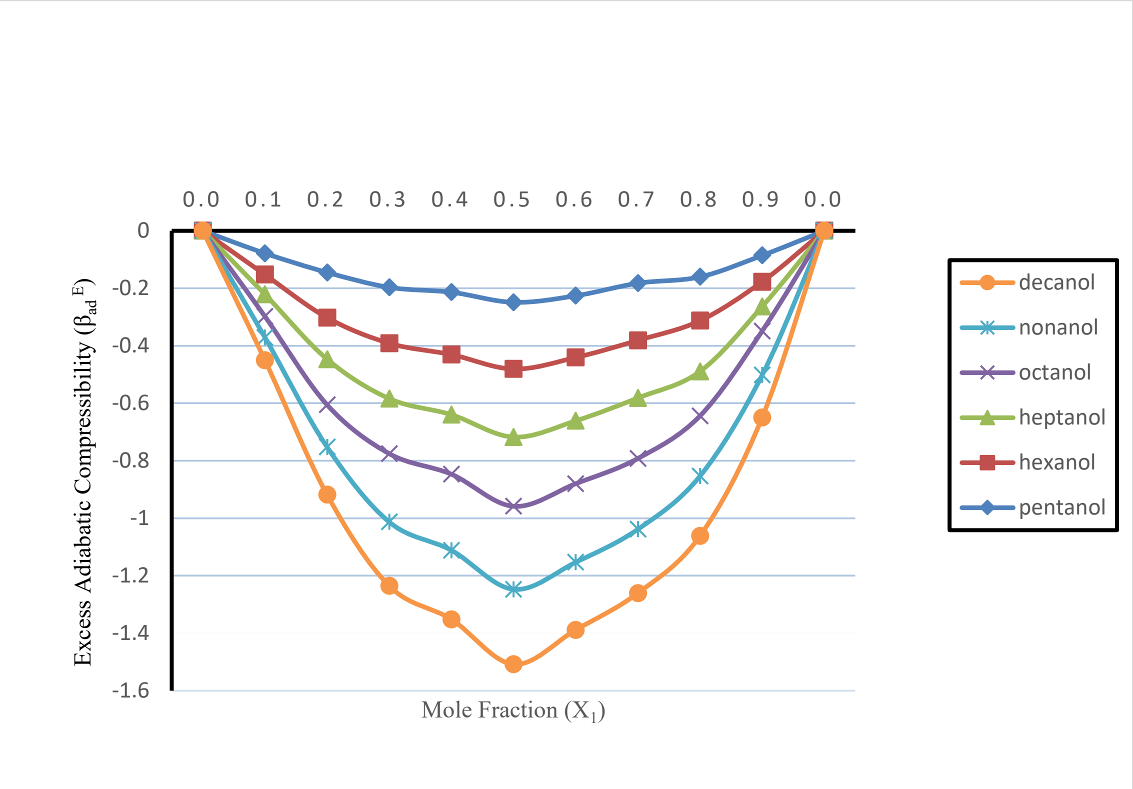


Figure 1: Excess adiabatic compressibility (β_{ad}^E) versus mole **fraction (X_1)** of the 1,3-dioxolane at experimental temperature.

A perusal of table 2 shows that the values of excess intermolecular free length for all the six binary system are negative. These negative values of excess intermolecular free length are shown in figure 2. From figure 2 shows that the excess intermolecular free length (L_f^E) values are negative for all binary systems but the magnitude of the negative values increase with increasing chain length of alcohols, the order are given below-

1-Pentanol < 1-Hexanol < 1-Heptanol < 1-Octanol < 1-Nonanol < 1-Decanol

The above order indicates the strength of interactions between component molecules decreases due to decrease in polarizability of 1-alkanol molecules.

The negative values of excess intermolecular free length (L_f^E) play a very important role in description of molecular interaction in liquid mixtures through dipole-dipole interaction and hydrogen bonding. Due to polar nature of 1,3-Dioxolane and 1-alkanols, the dipole-dipole interactions prevail in these mixtures. When the compounds are mixed the changes the occur in association equilibria are evidently rapture of the hydrogen bonds in pure 1,3-Dioxolane and 1-alkanols, dipole-dipole interactions and the formation of O − H----O hydrogen bonds between the components. This suggests the existence of strong interaction between the components in all the binary systems. The values of L_f^E suggest that strong specific interaction like the formation of H- bond association through weaker physical forcers of attraction [28

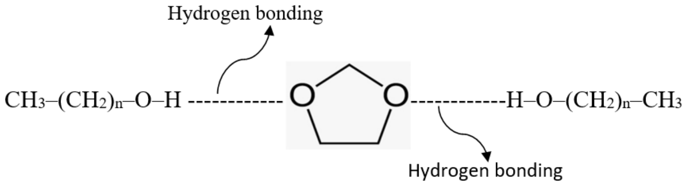


Figure: **Hydrogen bonding** present in 1,3-dioxolane **n-alkanols.**

Figure 2 depicts the variation of the excess intermolecular free length of the chosen system. We clearly see from fig. 2 that the non-linear variation of excess intermolecular free length with mole fraction of 1,3-dioxolane. The negative deviation of excess intermolecular free length reaching a minimum at 0.48 mole fraction of 1,3-dioxolane shows the molecular interactions between the molecules. The structural changes take place due to the variation in intermolecular free length (L_f) of the system. The excess intermolecular free length (shown in fig. 2) decrease with increase in the composition of 1,3-dioxolane till 0.48 mole fraction, reaches minimum at 0.48 mole fraction and beyond 0.48 mole fraction, it again increases. The existence of minimum free length indicates the squeezing of molecules in the system.

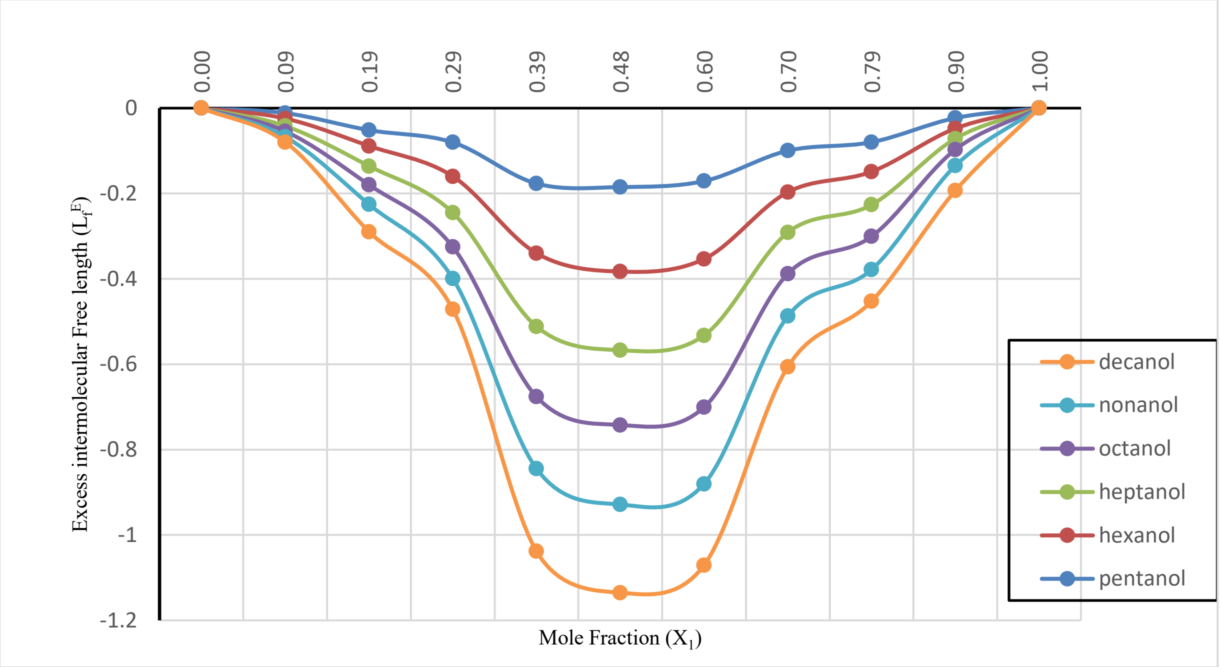


Figure 2: Excess intermolecular free length (L_f^E) versus mole **fraction (X_1)** of the 1,3-dioxolane at experimental temperature.

IV. Conclusion

In summary, the concentration dependencies of ultrasonic velocity and density, of 1,3-Dioxolane with1-pentanol, 1-hexanol, 1-heptanol, 1-octanol, 1-nonanol and 1-decanol binary liquid mixtures have been measured at temperature 298.15K at a constant 3 MHz frequency. The nonlinear variation of the related parameters such as adiabatic compressibility and intermolecular free length,was elaborated to understand the molecular interactions that lead to the process of complex formation between the solute and solvent molecules through π–cation, dipole–induced dipole, and ion–dipole interactions depending on the nature of the binary system. The negative variations of the excess values with concentration and temperature of the same acoustic parameters supported the presence of interaction between unlike molecules.

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Declarations conflict of interest

The authors have no competing interests to declare that are relevant to the content of this article.

Data availability statement

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

Author Contribution Statement

Dhirendra Kumar Sharma, Research design, Investigation, Writing-Original draft preparation and Manuscript correction.

Arpit Babu Shukla, Data Analysis, Mathematical Calculation, Tables and Graph preparation

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