

Thermodynamic properties of binary liquid mixtures formed by 1,3-dioxolane and 1-alkanols at 298.15K: By Ultrasonic Velocity Measurements.

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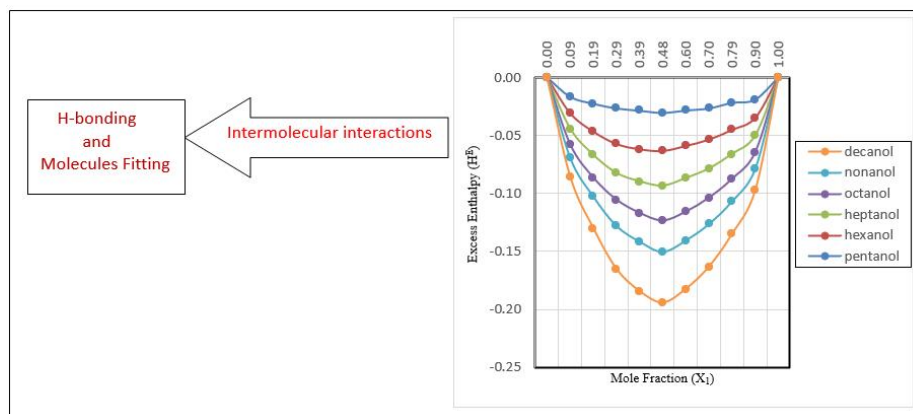
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Abstract

We measured the speed of sound, density, and viscosity for six binary mixtures of 1,3-dioxolane with successive 1-alkanols (C₅–C₁₀): 1-pentanol, 1-hexanol, 1-heptanol, 1-octanol, 1-nonanol, and 1-decanol. All measurements were carried out at 298.15 K. The experimental data were used to evaluate derived properties (e.g., adiabatic compressibility (β_{ad}), excess adiabatic compressibility (β_{ad}^E), inter molecular free length (L_f), excess inter molecular free length (L_f^E), enthalpy (H) and excess enthalpy (H^E) and to assess molecular interactions across increasing alkyl-chain length. The excess adiabatic compressibility (β_{ad}^E), excess intermolecular free length (L_f^E), and excess enthalpy (H^E) values were correlated using the Redlich–Kister polynomial equation. The excess properties were found to be negative, reflecting the influence of molecular interactions and the nature of the liquid mixtures. The negative values of the excess properties suggest significant specific interactions between the components, dominated by molecular association via hydrogen bonding. The extent of these interactions depends on the nature and structure of the liquid mixtures. These results indicate the occurrence of molecular association through hydrogen bonding in the binary systems.

Keywords: Density, 1,3-dioxolane, binary liquid mixture, Molecular interactions, hydrogen bonding and Ultrasonic velocity.

Graphical Abstract



Abbreviations

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

Introduction: The analysis of thermodynamic properties of mixtures containing cyclic ethers and 1-alkanols is of considerable interest, not only because they represent a class of technically important compounds widely used as solvents in the chemical industry, but also because such studies provide valuable insight into the structure and behavior of highly non ideal liquid mixtures. Thermodynamic studies of cyclic ether–1-alkanol mixtures are important both for their industrial relevance as solvent systems and for understanding the structural characteristics of highly non ideal liquids. 1,3-Dioxolane, a five-membered cyclic ether, possesses distinctive physicochemical properties arising from its molecular structure. Its increasing use in chemical applications has prompted extensive investigation, particularly in combination with other solvents such as 1-alkanols, to better understand the thermodynamic and molecular interaction behavior of the resulting mixtures. Owing to its multifunctional nature and broad applicability, 1,3-dioxolane plays a pivotal role in modern chemical processes, including pharmaceuticals and energy storage. Investigating its behavior in binary mixtures with 1-alkanols is therefore essential for understanding the molecular interactions that govern its performance in practical applications. It is widely employed as a solvent in the synthesis of active pharmaceutical ingredients, where it plays a key role in drug formulation and delivery. The present work extends our ongoing investigations into the excess thermodynamic properties of binary mixtures comprising cyclic ethers and 1-alkanols [1–4]. In this work, we report experimental values of excess adiabatic compressibility (β_{ad}^E), excess intermolecular free length (L_f^E) and excess enthalpy (H^E) for binary mixtures of 1,3-dioxolane with 1-alkanols. Owing to their growing application in industries such as cosmetics and pharmaceuticals, 1,3-dioxolane and 1-alkanols have attracted considerable interest, creating a demand for detailed data on their thermodynamic, acoustical, and transport properties, both in pure form and in mixtures [5–10]. Excess properties provide essential information on the molecular interactions and bulk behavior of liquid mixtures, while also enabling the evaluation and refinement of thermodynamic models for predicting fluid-phase equilibria. Excess adiabatic compressibility (β_{ad}^E) provides insight into molecular orientation and serves as a sensitive indicator of the strength and nature of intermolecular interactions in liquid mixtures. The thermodynamic properties of alkanols have been widely examined in different solvents. In recent years, interest has increased in binary liquid mixtures, which provide valuable insight into intermolecular interactions and stereochemical effects [13]. In this work, we investigate binary mixtures of 1,3-dioxolane (component 1) with six 1-alkanols (component 2): 1-pentanol, 1-hexanol, 1-heptanol, 1-octanol, 1-nonanol, and 1-decanol, as part of our continuing study of cyclic ether–alkanol systems. From the experimental measurements, we determined adiabatic compressibility (β_{ad}), excess adiabatic compressibility (β_{ad}^E), intermolecular free length (L_f), excess intermolecular free

length (L_f^E), enthalpy (H), and excess enthalpy (H^E). Given that both 1,3-dioxolane and the 1-alkanols contain proton donor and proton acceptor groups, a substantial degree of molecular association is expected in these binary mixtures.

Experimental

The chemicals used were: 1,3-dioxolane (purity $\geq 99.7\%$), 1-pentanol ($\geq 99.7\%$), 1-hexanol ($\geq 99.5\%$), 1-heptanol ($\geq 99.0\%$), 1-octanol ($\geq 99.7\%$), 1-nonanol ($\geq 99.0\%$), and 1-decanol ($\geq 99.0\%$), all supplied by CDH (New Delhi, India). All liquids were subjected to double distillation prior to use [11]. Purification was carried out according to the method reported by Zhao et al. [12]. For 1,3-dioxolane, the liquid was dried over anhydrous K_2CO_3 , filtered, and distilled, with the first and last portions of the distillate being discarded. Table 1 presents the experimentally measured density, viscosity, and sound velocity of the pure components at 298.15 K, together with literature values for comparison [13–24]. The measured values of density (ρ), sound velocity (u), and viscosity (η) closely match the corresponding literature data.

Table 1. Density (ρ), sound velocity (u) and viscosity (η) of pure Components at T = 298.15K.

Compound	ρ (g.cm ⁻³)		u (m.s ⁻¹)		η (mPa s)	
	Observed	Literature	Observed	Literature	Observed	Literature
1,3-Dioxolane	1.0616	1.0577 ¹⁷	1340	1338 ¹⁷	0.5885	0.5878 ¹⁷
		1.0586 ¹⁷		1338 ¹⁸		0.5873 ¹⁷
1-Pentanol	0.8124	0.8108 ¹³	1198	1197 ¹⁶	3.3978	3.5411 ¹³
		0.8107 ¹³		1268 ²²		3.5424 ¹³
1-Hexanol	0.8176	0.8187 ¹³	1306	1304 ¹⁵	4.6091	4.5924 ²³
		0.8152 ¹⁵		1303 ¹⁵		4.5932 ²⁰
1-Heptanol	0.8196	0.8187 ¹³	1325	1327 ¹⁵	5.9066	5.9443 ¹³
		0.8197 ¹⁹		1327 ²⁴		5.9443 ²⁴
1-Octanol	0.8236	0.8216 ¹³	1350	1348 ¹⁴	7.1508	7.6605 ¹³
		0.8218 ¹³		1347 ²²		7.5981 ¹³
1-Nonanol	0.8248	0.8244 ¹⁵	1366	1365 ¹⁵	8.9258	9.0230 ²¹
		0.8242 ¹⁵		1364 ²⁴		9.0200 ²⁴
1-Decanol	0.8292	0.8267 ¹⁵	1378	1380 ¹⁵	11.8027	11.825 ¹⁵
		0.8264 ¹⁹		1379 ²⁴		11.829 ¹⁵

The chemicals used were procured from CDH, New Delhi, India, and purified using standard procedures. All samples were stored in tightly sealed bottles to minimize atmospheric moisture absorption. The binary liquid mixtures were prepared by accurately weighing appropriate amounts of the pure liquids on an electronic balance (Citizen Scale (I) Pvt. Ltd., Mumbai, India) with a precision of ± 0.1 . The experimental uncertainty in mole fraction measurements did not exceed ± 0.0005 . All solutions were prepared by mass ratios and stored in airtight, stoppered measuring flasks. Four to five samples were prepared each day, and their speed of sound, viscosity, and density were measured on the same day. The density was determined at the experimental temperature using a 25 mL capacity specific gravity bottle immersed in a thermostatic bath. The volume of the bottle at the experimental

temperature ($T= 298.15$ K) was calibrated using distilled water. The sound velocity was determined using a multi-frequency interferometer (Model F-80D, Mittal Enterprise, New Delhi, India) operating at 3 MHz and $T = 298.15$ K. A fixed-frequency generator working at 3 MHz was used; at its resonant frequency, the crystal undergoes rapid mechanical oscillations, generating ultrasonic waves. These ultrasonic waves propagate through the liquid in the vessel, potentially inducing phenomena such as cavitation, acoustic streaming, and enhanced mixing. The viscosity was measured using an Ostwald viscometer. The instrument was calibrated with distilled water at $T = 298.15$ K, and five replicate measurements were taken for each sample to ensure accuracy. The uncertainty in viscosity measurement was $\pm 0.005 \times 10^{-3}$ mPa·s, indicating high precision.

Results and Discussion

The experimental values of speed of sound (u), viscosity (η), and density (ρ) for 1,3-dioxolane + 1-alkanol mixtures at $T = 298.15$ K are listed in Table 2. From these data, the adiabatic compressibility (β_{ad}), excess adiabatic compressibility (β_{ad}^E), intermolecular free length (L_f), excess intermolecular free length (L_f^E), enthalpy (H), and excess enthalpy (H^E) were computed and are also presented in Table 2.

Table 2. Density (ρ), ultrasonic velocity (u), and viscosity (η), Intermolecular free length(L_f), adiabatic compressibility(β_{ad}) and enthalpy (H)of binary mixture of 1,3-dioxolane (1) + 1-alkanol (2) at 298.15K

Mole fraction 1,3-Dioxolane (x_1)	Density (ρ) / g.cm ⁻³	Sound velocity (u) / ms ⁻¹	Viscosity (η) / mPas.	Intermolecular free length(L_f) $\times 10^{-4}$ /m	adiabatic compressibility (β_{ad}) $\times 10^{-7}$ / Pa ⁻¹	Enthalpy (H) $\times 10^6$
1,3-Dioxolane + Pentanol						
0.0000	0.8124	1198	3.3978	2.6732	8.5770	0.3156
0.0939	0.8276	1284	2.3973	2.2842	7.3290	0.3450
0.1942	0.8436	1290	1.8970	2.2201	7.1233	0.3468
0.2941	0.8640	1296	1.4437	2.1477	6.8909	0.3384
0.3942	0.8836	1300	1.1866	2.0872	6.6966	0.3341
0.4787	0.9068	1304	1.0904	2.0213	6.4853	0.33.8
0.5999	0.9316	1310	0.9311	1.9495	6.2551	0.3262
0.6972	0.9596	1318	0.7717	1.8697	5.9991	0.3236
0.7928	0.9876	1324	0.7171	1.8003	5.7762	0.3201
0.9035	1.0260	1332	0.6489	1.7121	5.4934	0.3166
1.0000	1.0616	1340	0.5885	1.6350	5.2460	0.3135
1,3-Dioxolane + Hexanol						
0.0000	0.8176	1306	4.6091	2.2349	7.1709	0.4163
0.0912	0.8252	1317	3.3826	2.1775	6.9867	0.4112
0.1955	0.8432	1320	2.3306	2.1214	6.8065	0.4003
0.2923	0.8584	1322	1.9839	2.0775	6.6657	0.3899
0.3982	0.8792	1325	1.5720	2.0192	6.4786	0.3787
0.4942	0.8992	1327	1.3059	1.9619	6.3154	0.3683
0.6059	0.9264	1330	1.0343	1.9019	6.1024	0.3567
0.6976	0.9508	1332	0.9131	1.8475	5.9279	0.465
0.8018	0.9836	1335	0.7680	1.7779	5.7045	0.3352
0.8914	1.0168	1337	0.7304	1.7147	5.5018	0.3254
1.0000	1.0616	1340	0.5885	1.6350	5.2460	0.3135
1,3-Dioxolane + Heptanol						
0.0000	0.8196	1325	5.9066	2.1660	6.9497	0.4838

0.0928	0.8304	1334	4.3181	2.1091	6.7671	0.4725
0.1905	0.8412	1334	3.2577	2.0820	6.6802	0.4552
0.2939	0.8592	1335	2.5895	2.0353	6.5304	0.4373
0.3894	0.8740	1335	1.9926	2.0009	6.4199	0.4201
0.4818	0.8916	1336	1.5315	1.9584	6.2837	0.4042
0.6021	0.9184	1337	1.2190	1.8984	6.0912	0.3835
0.6952	0.9420	1337	1.0959	1.8509	5.9387	0.3667
0.7892	0.9756	1338	0.9903	1.7845	5.7255	0.3505
0.9006	1.0156	1339	0.7057	1.7116	5.4918	0.3309
1.0000	1.0616	1340	0.5885	1.6350	5.2460	0.3135

1,3-Dioxolane + Octanol

0.0000	0.8296	1350	7.1508	2.0764	6.6622	0.5619
0.0885	0.8296	1350	5.6095	2.0614	6.6139	0.5363
0.1967	0.8464	1349	3.9321	2.0235	6.4923	0.5100
0.2998	0.8560	1348	3.2616	2.0038	6.4291	0.4845
0.3902	0.8712	1348	2.4284	1.9688	6.3168	0.4629
0.4963	0.8876	1348	1.9058	1.9324	6.2002	0.4375
0.6008	0.9140	1347	1.3631	1.8794	6.0301	0.4117
0.6925	0.9340	1348	1.1376	1.8364	5.8921	0.3905
0.7975	0.9676	1348	0.9141	1.7726	5.6875	0.3652
0.8940	1.0104	1348	0.7652	1.6975	5.4466	0.3421
1.0000	1.0616	1340	0.5885	1.6350	5.2460	0.3135

1,3-Dioxolane + Nonanol

0.0000	0.8248	1366	8.9258	2.0251	6.4976	0.6291
0.0876	0.8336	1366	6.8601	2.0037	6.4289	0.6020
0.1913	0.8404	1363	5.8531	1.9963	6.4051	0.5684
0.2942	0.8504	1359	4.4022	1.9844	6.3671	0.5347
0.3963	0.8692	1355	3.1558	1.9529	6.2662	0.5014
0.4959	0.8844	1352	2.3340	1.9279	6.1859	0.4697
0.6050	0.9092	1349	1.7321	1.8837	6.0439	0.4354
0.6947	0.9332	1346	1.3334	1.8434	5.9145	0.4072
0.7993	0.9648	1343	0.9642	1.7910	5.7466	0.3744
0.9013	1.0084	1340	0.8031	1.7213	5.5228	0.3402
1.0000	1.0616	1340	0.5885	1.6350	5.2460	0.3135

1,3-Dioxolane + Decanol

0.0000	0.8292	1378	11.8027	1.9794	6.4976	0.6990
0.0881	0.8364	1374	8.5615	1.9638	6.4289	0.6634
0.191	0.8396	1370	7.8207	1.9578	6.4051	0.6226
0.2921	0.8560	1366	5.5340	1.9413	6.3671	0.5827
0.3937	0.8672	1362	4.2319	1.9374	6.2662	0.5429
0.4956	0.8824	1358	3.4173	1.9153	6.1859	0.5035
0.604	0.9076	1353	2.5370	1.8759	6.0439	0.4615
0.7129	0.9308	1348	1.5262	1.8427	5.9145	0.4198
0.7983	0.9616	1344	1.1637	1.7943	5.7466	0.3871
0.8971	1.0040	1340	0.8623	1.7288	5.5228	0.3505
1.0000	1.0616	1340	0.5885	1.6350	5.246	0.3135

The excess parameters, namely excess intermolecular free length (L_f^E), excess adiabatic compressibility (β_{ad}^E), and excess enthalpy (H^E), were calculated using the following equations.

In 1952, Jacobson proposed an empirical relation for calculating the free length (L_f) of liquids. The intermolecular free length (L_f) can be obtained from the adiabatic compressibility (β_{ad}) using the following relation:

$$L_f = K \beta_{ad}^{1/2} \quad \dots(1)$$

where K is a temperature-dependent constant, and β_{ad} is the adiabatic compressibility, given by the following relation:

The adiabatic compressibility (β_{ad}) was calculated from the ultrasonic velocity (u) and density (ρ) of the medium using the following equation:

$$\beta_{ad} = u^{-2} \rho^{-1} \quad \dots(2)$$

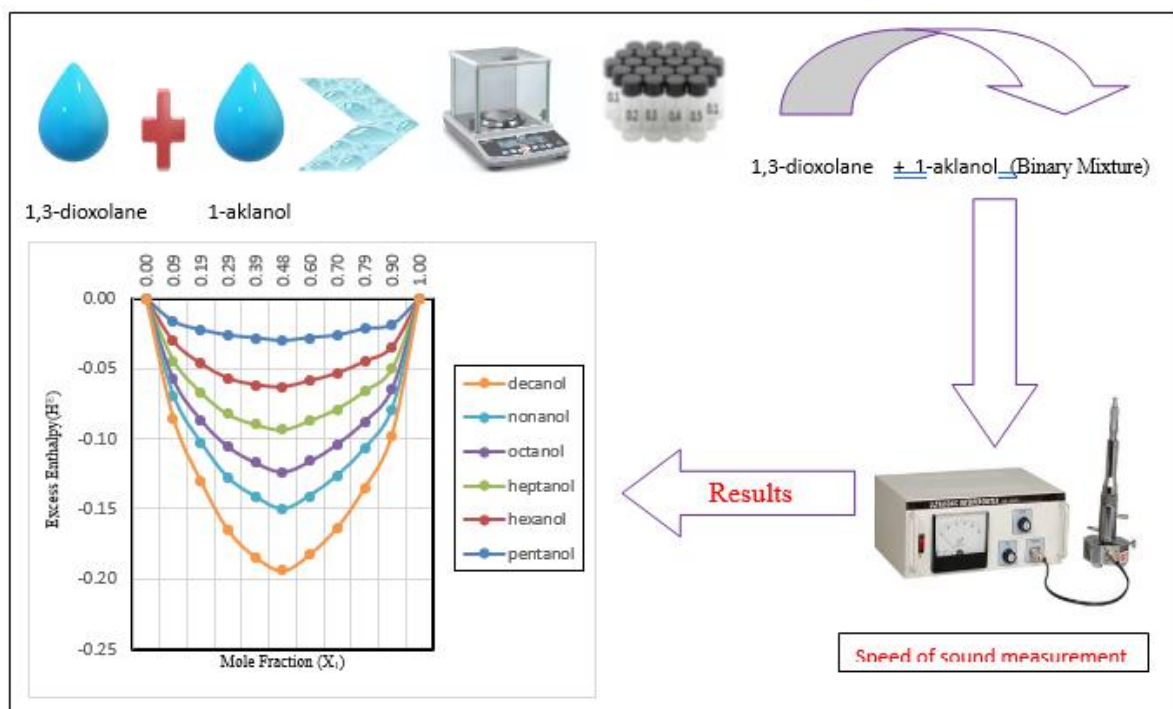
The enthalpy (H) was calculated using the following equation:

$$H = V_m \times P_i \quad \dots(3)$$

The excess values of the above acoustical parameters were calculated using the following relations:

$$Y^E = Y_{exp} - (X_1 Y_1 + X_2 Y_2) \quad \dots(4)$$

Here, Y^E refer to (L_f^E), (β_{ad}^E), (H^E), (p_i^E) and (V_f^E), whereas Y_{exp} is measured property under question. Y_1 , Y_2 , X_1 and X_2 refer to the properties and mole fractions of pure components 1 and 2 respectively.



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

A perusal of Table 2 shows that, as the mole fraction (X_1) of 1,3-dioxolane increases, the density and ultrasonic velocity increase, while the viscosity decreases. This trend can be attributed to molecular interactions in the system [25]. The addition of 1,3-dioxolane likely promotes closer packing of molecules through interactions such as dipole-induced dipole forces.

Figure 1 depicts the variation of excess intermolecular free length for the studied system. As seen in the figure, the excess intermolecular free length exhibits a non-linear variation with the mole fraction of 1,3-dioxolane. A negative deviation, reaching a minimum at $X_1 = 0.48$,

indicates significant molecular interactions between the components. These structural changes arise from variations in the intermolecular free length (L_f) of the system. The excess intermolecular free length decreases with increasing 1,3-dioxolane content up to $X_1=0.48$, after which it increases again. The observed minimum suggests close molecular packing (“squeezing” of molecules) at this composition.

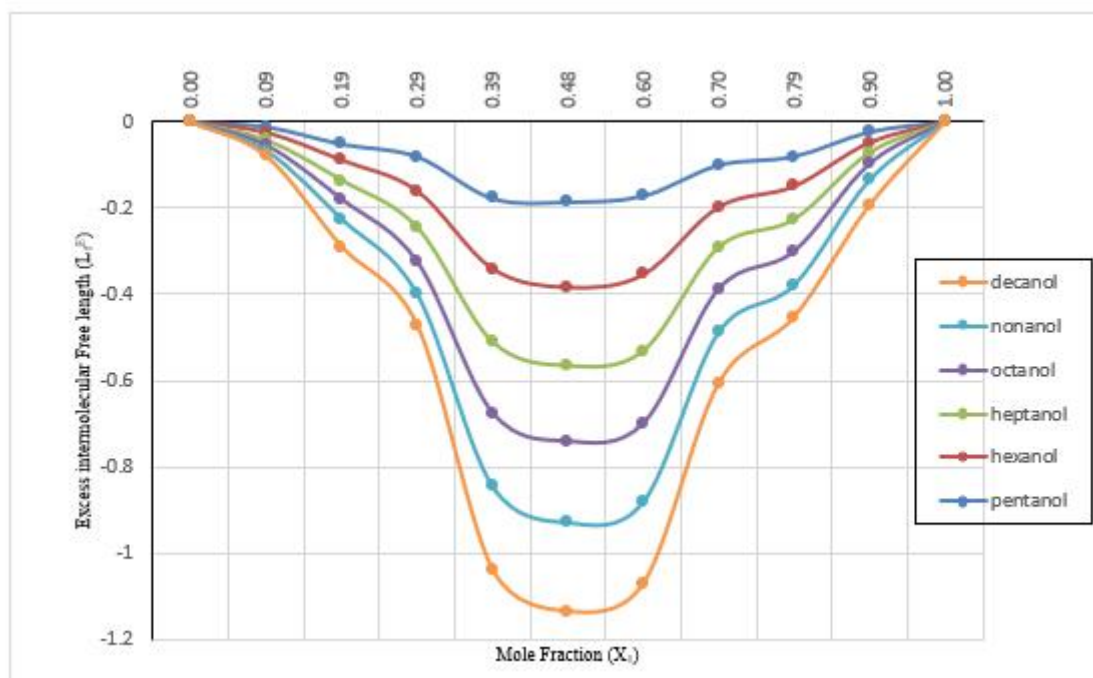


Figure 1: Variation of excess intermolecular free length (L_f^E) with mole fraction (x_1) of 1,3-dioxolane with 1-alkanols at 298.15K.

The calculated excess adiabatic compressibility (β_{ad}^E), values for the binary liquid mixture are presented in Figure 2. The variation of this property with composition is illustrated in the figure-2. The excess adiabatic compressibility (β_{ad}^E), values are negative over the entire mole fraction range and become increasingly negative with the rising mole fraction of the second component in all binary mixtures. These results can be explained in terms of molecular interactions and structural effects.

The variation of excess adiabatic compressibility (β_{ad}^E), with the mole fraction of 1,3-dioxolane (X_1) is presented in Figure 2. Fort and Moore [26] suggested that excess adiabatic compressibility (β_{ad}^E) arises from several opposing effects. Strong molecular interactions, such as charge transfer, dipole-induced dipole, and dipole-dipole interactions [27], as well as interstitial accommodation and orientational ordering, lead to a more compact structure, thereby making (β_{ad}^E) negative. The negative values of (β_{ad}^E) in these mixtures can be attributed to a structure-forming tendency.

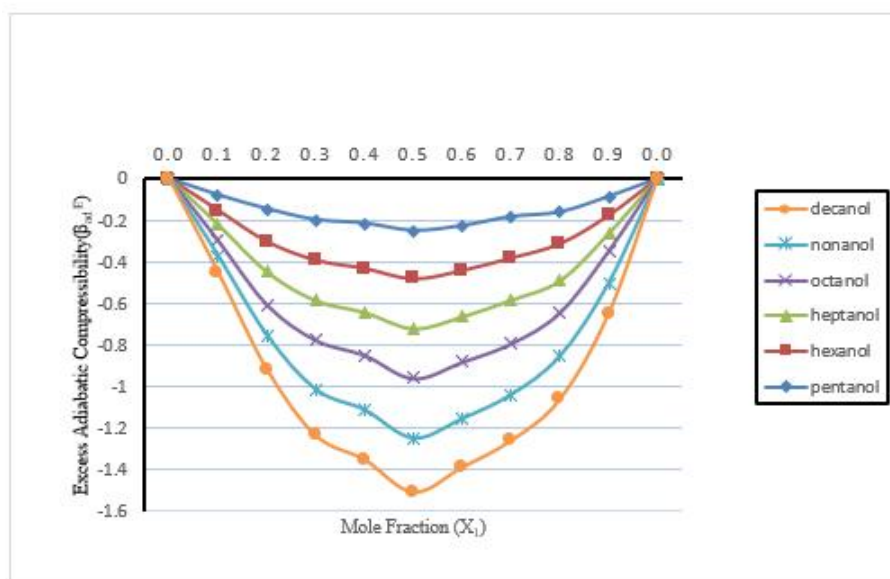


Figure 2. Variation of excess adiabatic compressibility (β_{ad}^E) with mole fraction (x_1) of 1,3-dioxolane with 1-alkanols at 298.15K.

Figure 3 shows the variation of excess enthalpy (H^E) with the mole fraction of 1,3-dioxolane at $T = 298.15$ K. For the binary system of 1,3-dioxolane with 1-alkanols, (H^E) values are negative, decreasing with increasing mole fraction of 1,3-dioxolane up to $X_1 \approx 0.5$, and then increasing with further addition of 1,3-dioxolane. Excess enthalpy (H^E) is an important parameter for interpreting molecular interactions. In the present investigation, for all six binary systems, it is observed that as the mole fraction of 1,3-dioxolane increases, the excess enthalpy (H^E) values decrease. This trend is consistent across all systems studied and can be seen in the plots presented in Figure 3. This suggests that dipole and dispersive forces are dominant in these systems at low 1,3-dioxolane concentrations. As the concentration of 1,3-dioxolane increases, the nature of the interactions shifts from weak to strong, indicating the onset of specific interactions. This observation is consistent with the trends observed for the other parameters discussed above. As a result, the free dipoles released from the 1-alkanols associate with 1,3-dioxolane molecules, forming strong hydrogen bonds. Consequently, a stronger molecular association exists between 1,3-dioxolane and 1-alkanol molecules through hydrogen bonding [28–31].

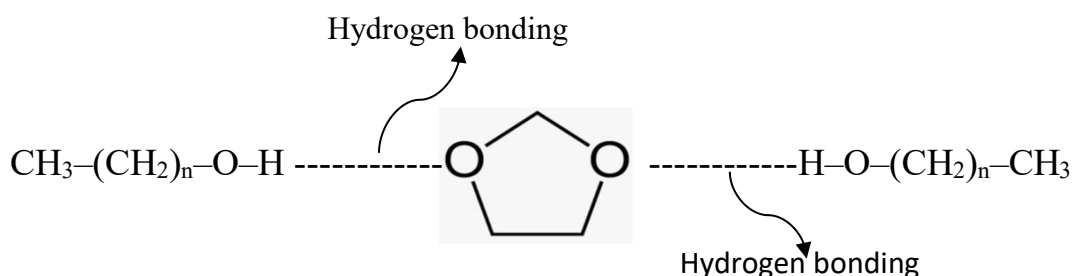


Figure 2.1: Hydrogen bonding present in 1,3-dioxolane-n-alkanols.

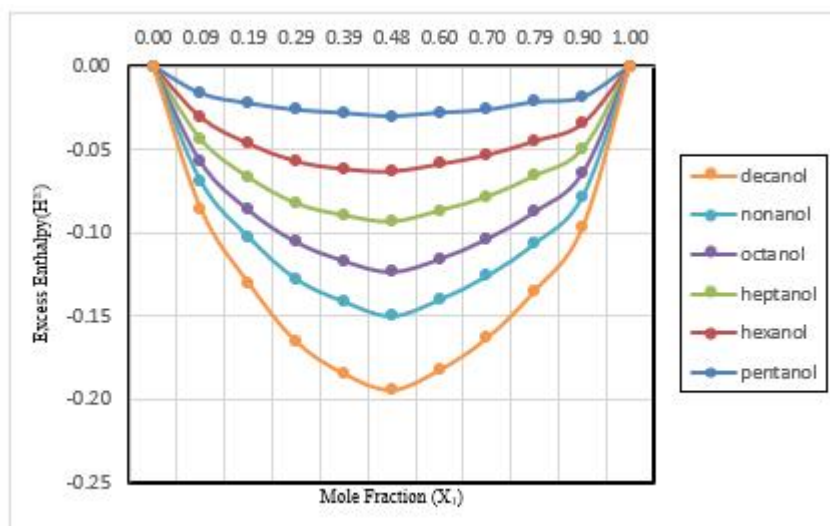


Figure 3- Variation of excess enthalpy (H^E) with mole fraction (x_1) of 1,3-dioxolane with 1-alkanols at 298.15K.

Conclusion

The sound velocity, density, and viscosity of 1,3-dioxolane with 1-alkanols were measured experimentally at $T = 298.15$ K. The calculated intermolecular free length (L_f), excess intermolecular free length (L_f^E), adiabatic compressibility (β_{ad}), excess adiabatic compressibility (β_{ad}^E), enthalpy (H), and excess enthalpy (H^E) strongly support the presence of strong molecular interactions between unlike molecules through hydrogen bonding. A thorough investigation of the behavior of 1-alkanols and 1,3-dioxolane provides a clear understanding of the nature and extent of molecular interactions between the components. In addition, molecular interactions are confirmed by the negative values of excess intermolecular free length (L_f^E), excess adiabatic compressibility (β_{ad}^E) and excess enthalpy (H^E). Hence, it is concluded that molecular interactions between 1,3-dioxolane and 1-alkanols arise due to hydrogen bonding.

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Author Contributions

Dhirendra Kumar Sharma: Methodology, Supervision, Writing – original draft

Gauri Khanwalkar: Software, Validation, methodology

Sandeep Sahu: Data curation, Investigation, Formal Analysis, Software

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