

Research Article

Some Experimental and Theoretical Studies of Molecular Interactions Prevailing in Cyclic Diether + 1-alkanols Binary Liquid Mixtures Through Ultrasonic Measurements

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Abstract

The density, viscosity and sound velocity for binary mixtures of 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 have been measured at the temperature 298.15 K, are conducted at atmospheric pressure. From these experimental values, various thermodynamic and excess thermodynamic properties were calculated. The 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), excess enthalpy (H^E), free volume (V_f), excess free volume (V_f^E), internal pressure (P_i), excess internal pressure (P_i^E) have been investigated from density (ρ), viscosity (η) and sound velocity (u) measurements of six binary liquid mixtures of 1,3-Dioxolane with pentanol, hexanol, heptanol, octanol, nonanol and decanol over the entire composition range of mole fractions at 298.15K. An excess values of adiabatic compressibility (β_{ad}^E), inter molecular free length (L_f^E), excess enthalpy (H^E), excess free volume (V_f^E) and excess internal pressure (P_i^E) were plotted against the mole fraction of 1,3-dioxolane over the whole composition range. The excess properties are found to be negative depending on the molecular interactions and the nature of the liquid mixtures. The systems studied exhibit very strong cross association through hydrogen bonding.

Keywords

Binary Mixtures, Adiabatic Compressibility, Enthalpy, Free Volume, Internal Pressure, Hydrogen Bonding, Molecular Interaction

1. Introduction

The use of cyclic diethers and 1-alcohols in many industrial processes such as pharmaceutical and cosmetics have greatly stimulated the need for extensive information on the thermodynamic, acoustic and transport properties of cyclic diethers, 1-alcohols and their mixtures. [1-6] In recent years, many research scholars have discussed thermodynamic and

excess thermodynamic properties in liquid mixture and interpreted these properties using the interactions among the molecules [7]. In our systematic investigation of these properties, we have reported the viscosities, densities, speeds of sound, isentropic compressibilities, molar volume and excess molar volumes of many binary systems in previous papers.

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[8-10] There has been a recent upsurge of interest in the study of thermodynamic properties of binary liquid mixtures, which have been used extensively to obtain information on intermolecular interactions and stereo chemical effects in these systems.

Therefore, ultrasonic techniques mainly used to explore and understand the characteristics and dynamics of such molecular interactions. The excess thermodynamic property of binary liquid mixtures of 1,3-dioxolane with 1-alkanols is of great importance both from practical and theoretical point of view. We can interpret the interactions and predict the application of the liquid mixture using the thermodynamic and physical properties of liquid and liquid mixtures. The ultrasonic velocity, density and viscosity of liquid mixtures are used to understand the theory of a mixture in liquid state. The intermolecular forces among the molecules in a liquid mixture alter the physical and chemical properties like dipole moment in the heat of mixing [11]. The experimental values of sound velocity (u), density (ρ) and viscosity (η) are useful in evaluating thermodynamic properties such as adiabatic compressibility (β_{ad}), inter molecular free length (L_f), enthalpy (H), free volume (V_f), internal pressure (P_i) and several excess parameters which will be very much useful in concerning the nature of intermolecular forces between the component molecules. Over the last four decades, research has been focused on measuring the ultrasonic velocity of liquid system and interpreting their molecular structures. In the present paper, sound velocity (u), density (ρ) and viscosity (η) of six binary liquid mixtures of 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, have been studied at 298.15 K over the entire composition range of mole fractions. From these experimental values, adiabatic compressibility (β_{ad}), inter molecular free length (L_f), enthalpy (H), free volume (V_f), internal pressure (P_i) and their deviations excess adiabatic compressibility (β_{ad}^E), excess inter molecular free length (L_f^E), excess enthalpy (HE), excess free volume (V_f^E) and excess internal pressure (p_i^E) have been calculated and interpreted in term of molecular interaction between the components of

the binary liquid mixtures. We know that excess thermodynamic properties such as excess adiabatic compressibility (β_{ad}^E), excess inter molecular free length (L_f^E), excess enthalpy (HE), excess free volume (V_f^E) and excess internal pressure (p_i^E) good information provide a understanding the intermolecular interaction between component molecules of the liquid mixtures. The cyclic diether and the 1-alcohols have both a proton donor and a proton acceptor group. It is expected that there will be a significant degree of H-bonding leading to self-association in the pure state in addition to mutual association in their binaries. [12] In addition, this work also provides a test of various empirical equations to correlate viscosity and ultrasonic sound data of binary mixtures containing both polar components.

This work is the first to report a combined study of sound velocity (u), density (ρ) and viscosity (η) of six binary liquid mixtures of practical importance in many industrial processes such as pharmaceutical and cosmetics have greatly stimulated the need for extensive information on the thermodynamic, acoustic and transport properties of 1,3-dioxolane, 1-alcohols and their mixtures. The 1,3-dioxolane and the 1-alcohols have both a proton donor and a proton acceptor group. It is expected that there will be a significant degree of H-bonding leading to self-association in the pure state in addition to mutual association in their binaries.

2. Materials and Methods

2.1. Materials

1,3-dioxolane (CDH New Delhi) was supplied with purity $\geq 99.7\%$, pentanol (CDH New Delhi) with $\geq 99.7\%$, hexanol (CDH New Delhi) with $\geq 99.5\%$, heptanol (CDH New Delhi) with $\geq 99\%$, octanol (CDH New Delhi) with $\geq 99.7\%$, nonanol (CDH New Delhi) with $\geq 99\%$, decanol (CDH New Delhi) with $\geq 99\%$, respectively with corresponding literature values [13-23]. Since the agreement with the literature values is very good.

Table 1. CASRegistryNumber, Mass Fraction Purity of the chemicals.

Component	Formula	CAS Reg. No.	Supplier	Mass Fraction Purity (%)	Method Purity analysis method
1,3-Dioxolane	C ₃ H ₆ O ₂	646-06-0	CDH New Delhi	99.7	Chromatography by the supplier
Pentanol	C ₅ H ₁₂ O	71-41-0	CDH New Delhi	99.7	Chromatography by the supplier
Hexanol	C ₆ H ₁₄ O	111-27-3	CDH New Delhi	99.5	Chromatography by the supplier
Heptanol	C ₇ H ₁₆ O	111-70-6	CDH New Delhi	99	Chromatography by the supplier
Octanol	C ₈ H ₁₈ O	111-87-5	CDH New Delhi	99.7	Chromatography by the supplier
Nonanol	C ₉ H ₂₀ O	143-08-8	CDH New Delhi	99	Chromatography by the supplier

Component	Formula	CAS Reg. No.	Supplier	Mass Fraction Purity (%)	Method Purity analysis method
Decanol	C ₁₀ H ₂₂ O	112-30-1	CDH New Delhi	99	Chromatography by the supplier

Table 2. Density (ρ), sound velocity (u) and viscosity (η) of pure Components at $T=298.15\text{K}$.

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]
Pentanol	0.8124	0.8108 [13]	1198	1197 [16]	3.3978	3.5411 [13]
		0.8107 [13]		1268 [22]		3.5424 [13]
Hexanol	0.8176	0.8187 [13]	1306	1304 [15]	4.6091	4.5924 [8]
		0.8152 [15]		1303 [15]		4.5932 [20]
Heptanol	0.8196	0.8187 [13]	1325	1327 [15]	5.9066	5.9443 [13]
		0.8197 [19]		1327 [23]		5.9443 [23]
Octanol	0.8236	0.8216 [13]	1350	1348 [14]	7.1508	7.6605 [13]
		0.8218 [13]		1347 [22]		7.5981 [13]
Nonanol	0.8248	0.8244 [15]	1366	1365 [15]	8.9258	9.0230 [21]
		0.8242 [15]		1364 [23]		9.0200 [23]
Decanol	0.8292	0.8267 [15]	1378	1380 [15]	11.8027	11.825 [15]
		0.8264 [19]		1379 [23]		11.829 [15]

2.2. Methods

2.2.1. Apparatus and Procedure

Air tight stopper bottles were used for the preparation of the mixtures and were placed in the dark place. The losses in the mixtures were kept to minimum, as evidenced by repeated measurements of physical properties over an interval of 2-3 days during in which before use time no change in physical properties was observed. The mixtures were well mixed by shaking before use. Binary mixtures were prepared by mass, using an electronic analytical balance (Model K-15 Deluxe, K Roy Instruments Pvt. Ltd.) with an accuracy of $\pm 0.00001 \times 10^{-3}$ kg as described elsewhere. The possible error in the mole fraction was estimated to be less than 1×10^{-4} . Five samples were prepared for one system, and their density and sound velocity were measured on the same day.

2.2.2. Density

Densities of pure liquids and their binary mixtures were determined by using a R. D. Bottle with a 25 cm³ is used to

measure the densities (ρ) of pure liquids and binary mixtures. Calibration was done at 298.15K with triply distilled water and purified methanol using density values from the literature. The R. D. Bottle is calibrated by using conductivity water (having specific conductance less than 1×10^6 ohm⁻¹) with 0.9970 and 0.9940 gcm⁻³ as its densities at $T = 298.15\text{K}$, respectively. The R. D. Bottle filled with air bubbles free liquids is kept in a thermostate water bath (MSI Goyal Scientific, Meerut, India) controlled with a thermal equilibrium. The precision of the density measurements was estimated to be ± 0.0002 g cm⁻³.

2.2.3. Sound Velocity

The ultrasonic velocity were measured using a multi-frequency ultrasonic interferometer (Model F80D, Mittal Enterprise, New Delhi, India) working at 3 M.Hz. The meter was calibrated with water, methanol, and benzene at 298.15 K. The details of the methods and techniques have been described previously.[8, 10] Measurement of sound velocity through medium was based on the accurate determination of the wavelength of ultrasonic waves of known frequency pro-

duced by quartz crystal in the measuring cell. 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 ms^{-1} . The measured values of ultrasonic velocities of pure 1,3-dioxolane, pentanol, hexanol, heptanol, octanol, nonanol and decanol at 298.15K were 1340, 1198, 1306, 1325, 1350, 1366 and 1378 m.s^{-1} respectively, which compare well with the corresponding literature values.

2.2.4. Viscosity

The viscosity of pure liquids and their binary mixture were measured using suspended Ostwald viscometer having a capacity of about 15 ml and the capillary having a length of about 90 mm and 0.5 mm internal diameter has been used to measure the flow time of pure liquids and liquid mixtures and it was calibrated with triply distilled water, methanol and benzene at 298.15 K. The details of the methods and techniques have been described by researchers [1, 4]. The efflux time was measured with an electronic stop watch (Racer) with a time resolution (± 0.015), and an average of at least four 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 two bulbs reservoir, one at the top and other at the bottom of the viscometer

linked to each other by U type facilitate the free full of liquid at atmospheric pressure. Viscosity values (η) of pure liquids and their binary mixtures are calculated using the solution.

$$\frac{\eta}{\rho} = at - \frac{b}{t}$$

Where t is the efflux time and a and b are viscometric constants.

The measured viscosities have reproducibility within $\pm 0.002 \text{ mPa.s}$. The measured values of viscosities of pure 1,3-dioxolane, pentanol, hexanol, heptanol, octanol, nonanol and decanol at 298.15K were 0.5885, 3.3978, 4.6091, 5.9066, 7.1508, 8.9258 and 11.8027 mPas. Which compare well with the corresponding literature values.

3. Results and Discussion

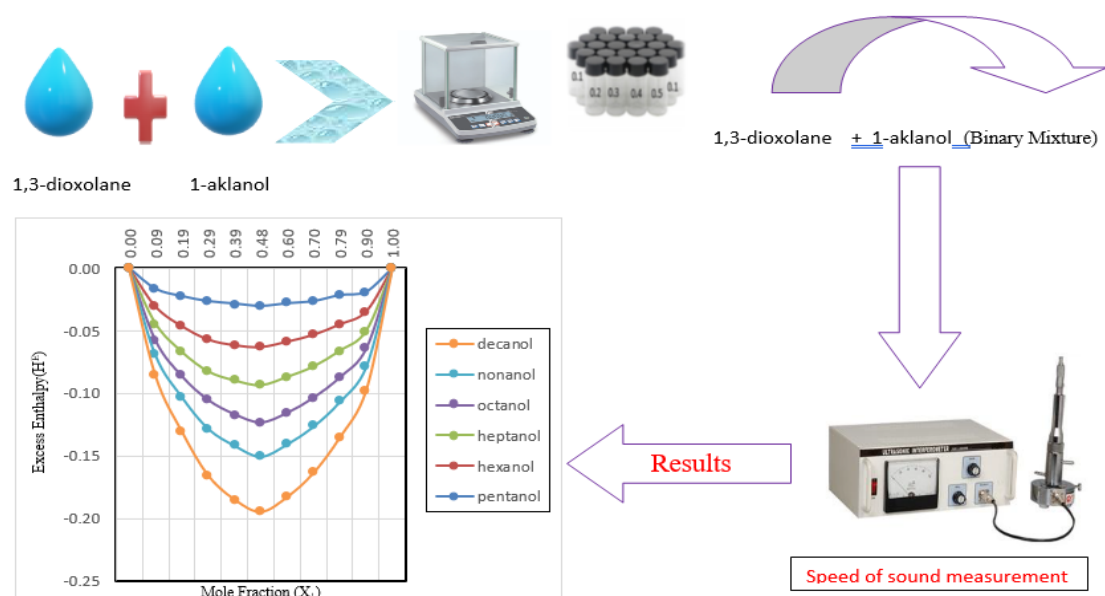
The experimental values of ultrasonic velocity (u), density (ρ) and viscosity (η) of 1,3-dioxolane with 1-alkanol mixtures at 298.15K are listed in Table 3. From these values, we have computed Intermolecular free length (L_f), adiabatic compressibility (β_{ad}), Enthalpy (H), internal pressure (P_i) and Free Volume (V_f) are presented in Table 3.

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

Mole fraction 1,3-Dioxolane (x1)	Density (ρ) / g.cm^{-3}	Sound velocity (u) / ms^{-1}	Viscosity (η) / mPas.	Intermolecular free length (L_f) $\times 10^{-4}/\text{m}$	adiabatic compressibility (β_{ad}) $\times 10^{-7}/\text{Pa}^{-1}$	Internal pressure (P_i) $\times 10^9/\text{Nm}^{-2}$	Free Volume (Vf) $\times 10^{-7}$ $\text{M}^3\text{mol}^{-1}$	Enthalpy (H) $\times 10^6$
1,3-Dioxolane + Pentanol								
0.0000	0.8124	1198	3.3978	2.6732	8.5770	2.9099	1.9568	0.3156
0.0939	0.8276	1284	2.3973	2.2842	7.3290	3.2892	3.5817	0.3450
0.1942	0.8436	1290	1.8970	2.2201	7.1233	3.3763	4.9996	0.3468
0.2941	0.8640	1296	1.4437	2.1477	6.8909	3.4821	9.9265	0.3384
0.3942	0.8836	1300	1.1866	2.0872	6.6966	3.5776	11.0374	0.3341
0.4787	0.9068	1304	1.0904	2.0213	6.4853	3.6885	10.8499	0.33.8
0.5999	0.9316	1310	0.9311	1.9495	6.2551	3.8155	13.4125	0.3262
0.6972	0.9596	1318	0.7717	1.8697	5.9991	3.9663	17.4788	0.3236
0.7928	0.9876	1324	0.7171	1.8003	5.7762	4.1099	17.4788	0.3201
0.9035	1.0260	1332	0.6489	1.7121	5.4934	4.3085	19.1422	0.3166
1.0000	1.0616	1340	0.5885	1.6350	5.2460	4.4982	21.7624	0.3135
1,3-Dioxolane+Hexanol								
24.74130	0.8176	1306	4.6091	2.2349	7.1709	3.3333	1.7591	0.4163
0.0912	0.8252	1317	3.3826	2.1775	6.9867	3.4069	2.7275	0.4112

Mole fraction 1,3-Dioxolane (x1)	Density (ρ) / g.cm ⁻³	Sound veloci- ty (u) / ms ⁻¹	Viscosity (η) / mPas.	Intermolecu- lar free length (Lf) $\times 10^{-4}$ /m	adiabatic com- pressibility (β_{ad}) $\times 10^{-7}$ / Pa ⁻¹	Internal pressure (Pi) $\times 10^9$ /Nm ⁻²	Free Vol- ume (Vf) $\times 10^{-7}$ M ³ mol ⁻¹	Enthalpy (H) $\times 10^6$
0.1955	0.8432	1320	2.3306	2.1214	6.8065	3.4931	4.5760	0.4003
0.2923	0.8584	1322	1.9839	2.0775	6.6657	3.5642	5.5951	0.3899
0.3982	0.8792	1325	1.5720	2.0192	6.4786	3.6629	7.5845	0.3787
0.4942	0.8992	1327	1.3059	1.9619	6.3154	3.7548	9.5968	0.3683
0.6059	0.9264	1330	1.0343	1.9019	6.1024	3.8815	12.9396	0.3567
0.6976	0.9508	1332	0.9131	1.8475	5.9279	3.9927	14.9307	0.465
0.8018	0.9836	1335	0.7680	1.7779	5.7045	4.1444	18.3980	0.3352
0.8914	1.0168	1337	0.7304	1.7147	5.5018	4.2939	18.9465	0.3254
1.0000	1.0616	1340	0.5885	1.6350	5.2460	4.4982	21.7624	0.3135
1,3-Dioxolane+Heptanol								
0.0000	0.8196	1325	5.9066	2.1660	6.9497	3.4147	1.5030	0.4838
0.0928	0.8304	1334	4.3181	2.1091	6.7671	3.4949	2.3075	0.4725
0.1905	0.8412	1334	3.2577	2.0820	6.6802	3.5404	3.3296	0.4552
0.2939	0.8592	1335	2.5895	2.0353	6.5304	3.6202	4.4224	0.4373
0.3894	0.8740	1335	1.9926	2.0009	6.4199	3.6826	6.1746	0.4201
0.4818	0.8916	1336	1.5315	1.9584	6.2837	3.7609	8.6425	0.4042
0.6021	0.9184	1337	1.2190	1.8984	6.0912	3.8784	11.2315	0.3835
0.6952	0.9420	1337	1.0959	1.8509	5.9387	3.9780	12.3322	0.3667
0.7892	0.9756	1338	0.9903	1.7845	5.7255	4.1245	13.4017	0.3505
0.9006	1.0156	1339	0.7057	1.7116	5.4918	4.2985	20.4381	0.3309
1.0000	1.0616	1340	0.5885	1.6350	5.2460	4.4982	21.7624	0.3135
1,3-Dioxolane+Octanol								
0.0000	0.8296	1350	7.1508	2.0764	6.6622	3.5546	1.3767	0.5619
0.0885	0.8296	1350	5.6095	2.0614	6.6139	3.5585	1.8692	0.5363
0.1967	0.8464	1349	3.9321	2.0235	6.4923	3.6225	2.9529	0.5100
0.2998	0.8560	1348	3.2616	2.0038	6.4291	3.6596	3.6234	0.4845
0.3902	0.8712	1348	2.4284	1.9688	6.3168	3.7245	5.2656	0.4629
0.4963	0.8876	1348	1.9058	1.9324	6.2002	3.7947	6.9577	0.4375
0.6008	0.9140	1347	1.3631	1.8794	6.0301	3.9032	10.5160	0.4117
0.6925	0.9340	1348	1.1376	1.8364	5.8921	3.9930	12.7180	0.3905
0.7975	0.9676	1348	0.9141	1.7726	5.6875	4.1367	15.9753	0.3652
0.8940	1.0104	1348	0.7652	1.6975	5.4466	4.3197	18.9060	0.3421
1.0000	1.0616	1340	0.5885	1.6350	5.2460	4.4982	21.7624	0.3135
1,3-Dioxolane+Nonanol								
0.0000	0.8248	1366	8.9258	2.0251	6.4976	3.5970	1.1714	0.6291
0.0876	0.8336	1366	6.8601	2.0037	6.4289	3.6354	1.6286	0.6020
0.1913	0.8404	1363	5.8531	1.9963	6.4051	3.6530	1.8990	0.5684

Mole fraction 1,3-Dioxolane (x1)	Density (ρ) / g.cm ⁻³	Sound velocity (u) / ms ⁻¹	Viscosity (η) / mPas.	Intermolecular free length (Lf) $\times 10^{-4}$ /m	adiabatic compressibility (β_{ad}) $\times 10^{-7}$ / Pa ⁻¹	Internal pressure (Pi) $\times 10^9$ /Nm ⁻²	Free Volume (Vf) $\times 10^{-7}$ M ³ mol ⁻¹	Enthalpy (H) $\times 10^6$
0.2942	0.8504	1359	4.4022	1.9844	6.3671	3.6802	2.6620	0.5347
0.3963	0.8692	1355	3.1558	1.9529	6.2662	3.7449	3.9924	0.5014
0.4959	0.8844	1352	2.3340	1.9279	6.1859	3.7978	5.7014	0.4697
0.6050	0.9092	1349	1.7321	1.8837	6.0439	3.8913	7.9725	0.4354
0.6947	0.9332	1346	1.3334	1.8434	5.9145	3.9807	10.6902	0.4072
0.7993	0.9648	1343	0.9642	1.7910	5.7466	4.1018	15.3683	0.3744
0.9013	1.0084	1340	0.8031	1.7213	5.5228	4.2372	17.3683	0.3402
1.0000	1.0616	1340	0.5885	1.6350	5.2460	4.4982	21.7624	0.3135
1,3-Dioxolane+Decanol								
0.0000	0.8292	1378	11.8027	1.9794	6.4976	3.6639	0.8971	0.6990
0.0881	0.8364	1374	8.5615	1.9638	6.4289	3.6797	1.3454	0.6634
0.191	0.8396	1370	7.8207	1.9578	6.4051	3.6977	1.4040	0.6226
0.2921	0.8560	1366	5.5340	1.9413	6.3671	3.7331	2.1400	0.5827
0.3937	0.8672	1362	4.2319	1.9374	6.2662	3.7654	2.8863	0.5429
0.4956	0.8824	1358	3.4173	1.9153	6.1859	3.8145	3.5598	0.5035
0.604	0.9076	1353	2.5370	1.8759	6.0439	3.9018	4.8971	0.4615
0.7129	0.9308	1348	1.5262	1.8427	5.9145	3.9793	9.1301	0.4198
0.7983	0.9616	1344	1.1637	1.7943	5.7466	4.0927	12.1810	0.3871
0.8971	1.0040	1340	0.8623	1.7288	5.5228	4.2541	16.4668	0.3505
1.0000	1.0616	1340	0.5885	1.6350	5.246	4.4982	21.7624	0.3135



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

Table 4. Excess thermodynamic parameters (β_{ad}^E , L_f^E , V_f^E , p_i^E and HE) for binary mixture of 1,3-dioxolane (1) + 1-alkanol (2) at 298.15K.

Mole fraction 1,3-Dioxolane (x1)	Excess adiabatic com- pressibility $\beta_{ad}^E \times 10^{-7} / \text{Pa}^{-1}$	Excess intermolecular free length $L_f^E \times 10^{-4} / \text{m}$	Excess Free ume $V_f^E \times 10^{-7} \text{M}^3 \text{mol}^{-1}$	Excess Internal pressure $p_i^E \times 10^9 / \text{Nm}^{-2}$	Excess Enthal- py HE $\times 10^6$
1,3-Dioxolane + Pentanol					
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.0939	-0.0782	-0.0115	-0.5145	-0.0630	-0.0165
0.1942	-0.1248	-0.0515	-1.2381	-0.0879	-0.0224
0.2941	-0.1467	-0.0802	-1.8168	-0.1151	-0.0264
0.3942	-0.2136	-0.1767	-2.4698	-0.1616	-0.0283
0.4787	-0.2487	-0.1849	-2.5138	-0.2183	-0.0302
0.5999	-0.2260	-0.1709	-2.2127	-0.1972	-0.0281
0.6972	-0.1823	-0.0997	-1.7633	-0.1609	-0.0264
0.7928	-0.1603	-0.0798	-1.2781	-0.1192	-0.0216
0.9035	-0.0857	-0.0231	-0.7801	-0.0764	-0.0189
1.0000	0.0000	0.0000	0.0000	0.0000	0.0000
1,3-Dioxolane+Hexanol					
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.0912	-0.07423	-0.0127	-1.1275	-0.0526	-0.0142
0.1955	-0.1874	-0.037	-1.8761	-0.0679	-0.024
0.2923	-0.1345	-0.0795	-2.8817	-0.1096	-0.0306
0.3982	-0.197	-0.1632	-3.3261	-0.1543	-0.0333
0.4942	-0.2321	-0.198	-3.7201	-0.2042	-0.0328
0.6059	-0.2147	-0.1825	-3.3444	-0.1876	-0.0306
0.6976	-0.1987	-0.0971	-2.8519	-0.1632	-0.0269
0.8018	-0.1524	-0.069	-1.789	-0.1229	-0.0233
0.8914	-0.0914	-0.0245	-0.830	-0.0878	-0.0157
1.0000	0.0000	0.0000	0.0000	0.0000	0.0000
1,3-Dioxolane+Heptanol					
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.0928	-0.0685	-0.0176	-1.3519	-0.0603	-0.0134
0.1905	-0.1745	-0.0472	-2.615	-0.0807	-0.0204
0.2939	-0.1426	-0.0854	-3.9103	-0.1129	-0.0255
0.3894	-0.189	-0.1717	-4.3773	-0.1540	-0.0281
0.4818	-0.2367	-0.1842	-4.4567	-0.2258	-0.0305
0.6021	-0.2203	-0.1791	-4.2632	-0.2087	-0.0282
0.6952	-0.2004	-0.0944	-3.3260	-0.1754	-0.0253
0.7892	-0.1766	-0.0776	-2.4409	-0.1453	-0.0211
0.9006	-0.0765	-0.0238	-1.4933	-0.0920	-0.0155
1.0000	0.0000	0.0000	0.0000	0.0000	0.0000

Mole fraction 1,3-Dioxolane (x1)	Excess adiabatic com- pressibility $\beta_{ad}^E \times 10^{-7} / \text{Pa}^{-1}$	Excess intermolecular free length $L_f^E \times 10^{-4} / \text{m}$	Excess Free ume $V_f^E \times 10^{-7} \text{M}^3 \text{mol}^{-1}$	Excess Internal pressure $p_i^E \times 10^9 / \text{Nm}^{-2}$	Excess Enthal- py $H^E \times 10^6$
1,3-Dioxolane+Octanol					
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.0885	-0.077	-0.01241	-1.575	-0.0835	-0.0136
0.1967	-0.109	-0.0439	-3.6097	-0.1177	-0.0195
0.2998	-0.192	-0.0797	-4.7580	-0.1779	-0.0229
0.3902	-0.207	-0.1646	-5.6279	-0.1983	-0.0271
0.4963	-0.241	-0.1751	-6.0149	-0.2282	-0.0299
0.6008	-0.219	-0.1682	-5.5981	-0.2183	-0.0289
0.6925	-0.211	-0.0967	-4.6387	-0.2150	-0.0251
0.7975	-0.155	-0.0742	-3.0346	-0.1704	-0.0214
0.8940	-0.051	-0.0257	-1.3586	-0.0785	-0.0144
1.0000	0.0000	0.0000	0.0000	0.0000	0.0000
1,3-Dioxolane+Nonanol					
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.0876	-0.071	-0.0128	-1.6075	-0.0705	-0.0115
0.1913	-0.147	-0.0458	-3.781	-0.1168	-0.0163
0.2942	-0.238	-0.0741	-5.4436	-0.1819	-0.0225
0.3963	-0.265	-0.1685	-6.5197	-0.2092	-0.0246
0.4959	-0.309	-0.1863	-7.1582	-0.2461	-0.0268
0.6050	-0.304	-0.1796	-7.0586	-0.2209	-0.0247
0.6947	-0.286	-0.0993	-5.9552	-0.1924	-0.0226
0.7993	-0.249	-0.0777	-3.6424	-0.1555	-0.0193
0.9013	-0.153	-0.0378	-1.6685	-0.1121	-0.0144
1.0000	0.0000	0.0000	0.0000	0.0000	0.0000
1,3-Dioxolane+Decanol					
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.0881	-0.0794	-0.01247	-1.6524	-0.0977	-0.0164
0.191	-0.2059	-0.0642	-4.0473	-0.1456	-0.0275
0.2921	-0.2325	-0.0725	-5.7219	-0.1745	-0.0374
0.3937	-0.3002	-0.1936	-7.4983	-0.2269	-0.0431
0.4956	-0.3418	-0.2066	-9.1545	-0.2630	-0.0442
0.604	-0.3352	-0.1905	-8.4018	-0.2560	-0.0421
0.7129	-0.3492	-0.1188	-5.7655	-0.2294	-0.0371
0.7983	-0.2882	-0.0741	-3.7509	-0.1972	-0.0284
0.8971	-0.1873	-0.0584	-1.8209	-0.1583	-0.0186
1.0000	0.0000	0.0000	0.0000	0.0000	0.0000

The derived excess parameters such as (L_f^E), (β_{ad}^E), (HE), (p_i^E) and (V_f^E) at the above temperature 298.15K are summarized in Table 4. These parameters have been calculated using the following equations.

In the year 1952, Jacobson [24, 25], suggested an empirical relation for calculating the free length (L_f) of liquids. Inter molecular free length (L_f), can be calculated from the adiabatic compressibility (β_{ad}) by the relation given below

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

Where K is temperature dependent constant and β_s is the isentropic compressibility, which is given by the relation.

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

Where u is the sound velocity and ρ is the density of liquid.

The adiabatic compressibility (β_{ad}) has been calculated from the ultrasonic velocity (u) and density (ρ) of the medium using the equation as [26, 27].

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

$$\beta_{ad}^E = \beta_{ad} - X_1 \beta_{ad,1} - X_2 \beta_{ad,2} \quad (3)$$

where β_{ad} , $\beta_{ad,1}$ and $\beta_{ad,2}$ are the isentropic compressibilities of the mixture, pure component 1 and pure component 2, respectively.

According to Bingham [28] and Macleod [29] free volume (V_f) can be calculated from the sound velocity and viscosity by the relation given below

$$V_f = (M U / k \eta)^{3/2} \quad (4)$$

Where M is the molecular weight (gm)

U is the sound velocity (cm/sec)

η is the viscosity (poise)

k is the constant, equal to 4.28×10^9 , independence of temperature and V_f , the free volume is in milliliters per mole.

On the basis of dimensional analysis, using free volume concept, the following expression can be used for calculating internal pressure. The internal pressure of a liquid can also be evaluated by using the equation proposed by Suryanarayana [30, 31].

$$p_i = bRT \left(\frac{k\eta}{u} \right)^{\frac{1}{2}} \frac{\rho^{\frac{2}{3}}}{M_{eff}^{\frac{6}{5}}} \quad (5)$$

where b is the packing fraction of the liquid which is taken equal to 2 for most of the liquids, k is a constant equal to 4.28×10^9 , M_{eff} ($= x_1 M_1 + x_2 M_2$) is the effective molecular mass and M is the molar mass of the mixture of pure liquid and T is absolute temperature.

Enthalpy (H) can be calculated by the following equation [32].

$$H = V_m \times P_i \quad (6)$$

Where V_m is molar volume and P_i is internal pressure.

The molar volume V_m calculated from the measured values of density (ρ), molar volume (V_m) was calculated using there-lation.

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

Where X_1 , X_2 and M_1 , M_2 are the mole fraction and molecular weight of the component 1 and 2 respectively.

The excess thermodynamic function (Y^E) provide a way to represent directly the deviation of a solution from ideal behaviour. The difference between the thermodynamic function of mixing for a real system and the value corresponding to a perfect solution at the same temperature, pressure and composition is called the thermodynamic excess function, denoted by Y^E . Excess values for all the parameters are computed using the general formula.

$$Y^E = Y_{exp} - (X_1 Y_1 + X_2 Y_2) \quad (7)$$

Where Y represents the parameter such as intermolecular free length, free volume, internal pressure, adiabatic compressibility and entropy and X_1 and X_2 are the mole fractions of components whose parameters.

A perusal of Table 2 shows the mole fraction (X_1) of 1,3-dioxolane increases, density and ultrasonic velocity increase, while viscosity decreases. This trend can be explained by molecular interactions in the system [33]. When 1,3-Dioxolane is added, it likely leads to closer packing of molecules due to molecular interactions, such as dipole-induced dipole forces.

The experimental values of ultrasonic velocity (u), density (ρ) at temperatures 298.15K along with the calculated values of excess intermolecular free length L_f^E , are given in Table 4 for 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 mixtures respectively. The values of L_f^E are plotted against the mole fraction of 1,3-dioxolane are shown in Figure 1. The important effects which are expected to contribute to the value of excess functions in the present work are arbitrarily divide into physical, chemical and structural contributions: (1) Physical contributions comprise non-specific physical interactions. (2) Chemical effects occur due to the breaking up of the liquid order of associated species. (3) Structural effects take place due to the geometrical fitting of 1,3-dioxolane and 1-alkanols into the voids created by each other and also due to differences in molar and free volumes of these components. Each factor

makes positive or negative contribution to the resultant values of excess function and the magnitude of the contribution is dependent on mole fraction range. The sign and magnitude of excess intermolecular free length L_f^E play an important role in assessing the molecular interactions between the component molecules in the liquid mixtures. Excess intermolecular free length L_f^E has been found to be negative for all six binary mixtures, over the whole mole fraction range (Figure 1).

Negative values of L_f^E suggest that the structure is less compressible than the corresponding ideal mixture, suggesting that there may be intermolecular hydrogen bonding between ether and 1-alkanols. This is in accordance with a view proposed by Fort and Moore [34] according to which liquids of different molecular size usually mix with decrease in volume yielding negative L_f^E values.

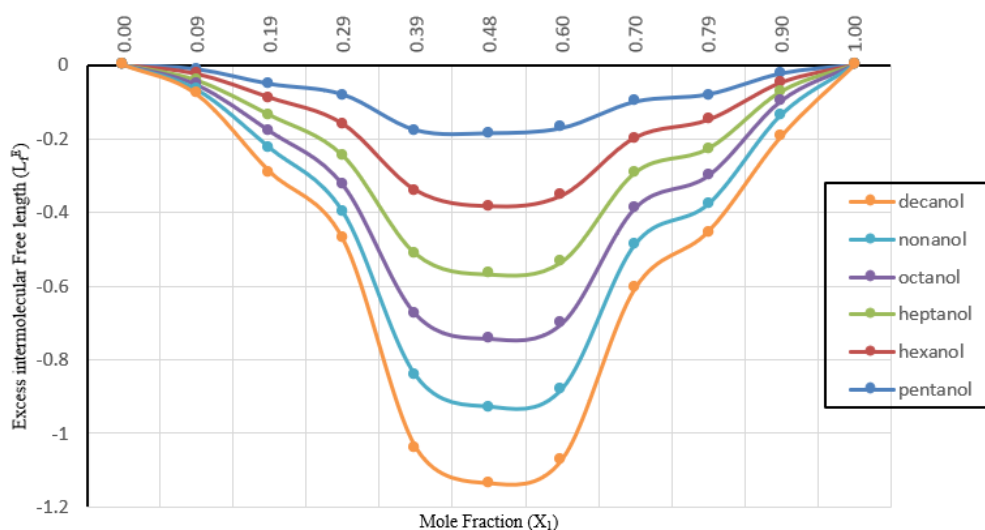


Figure 1. Variation of excess intermolecular free length (L_f^E) with mole fraction(x_1) of 1,3-dioxolane with 1-alkanols at 298.15K.

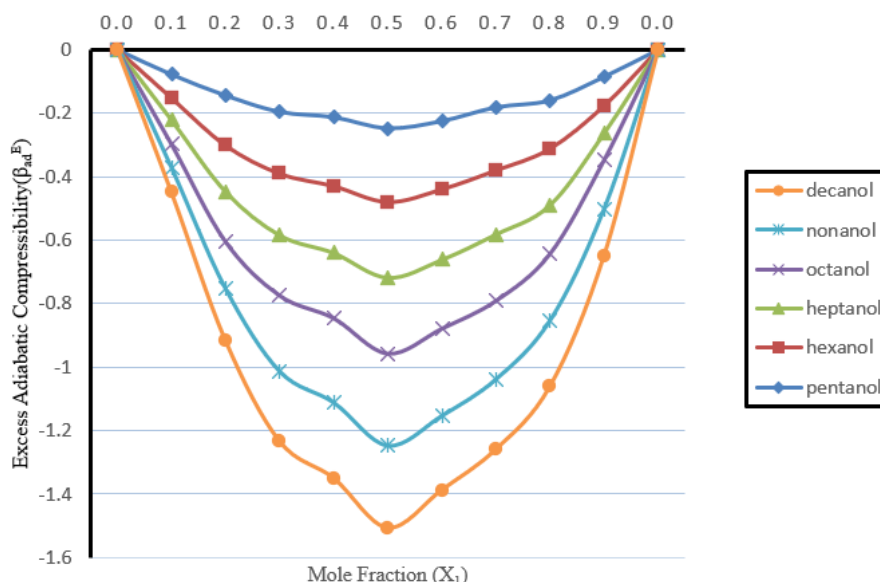


Figure 2. Variation of excess adiabatic compressibility (β_{ad}^E) with mole fraction (x_1) of 1,3-dioxolane with 1-alkanols at 298.15K.

We have calculated excess adiabatic compressibility (β_{ad}^E), at 298.15K for the binary mixtures of 1,3- dioxolane (1) with the 1-alkanols (2). The variations of the excess properties over the entire range of compositions for the binary mixtures are depicted in Figure 2. The value of the excess adiabatic

compressibility (β_{ad}^E), was found to be negative for the 1,3-dioxolane and 1-alkanols mixture, but the magnitude of negative value decreases with increasing chain length of the alcohols in the series (Figure 2). The trend it follows is,

Pent.OH < Hex.OH < Hept.OH < Oct. OH < Non.OH < Dec.OH

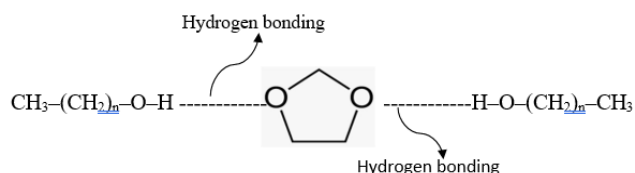


Figure 3. Hydrogen bonding present in 1,3-dioxolane– *n*-alkanols.

The negative excess adiabatic compressibility (β_{ad}^E), values indicate the presence of strong molecular interactions between the components of the mixture. Several effects may contribute to the value of (β_{ad}^E), such as [35-37] (a) dipolar interactions, (b) interstitial accommodation of one component into the other and (c) possible hydrogen-bond interactions between unlike molecules. The actual volume change, therefore, depends on the relative strength of these three effects. It is known fact that as the number of C-atoms of the alkyl group increases, the electron releasing ability (+I effect) increases, thereby decreasing the polarity of the O---H bond of the 1-alkanols. Consequently, Pent.OH having the highest polarity achieves the most favorable intermolecular H-bonded interactions with the cyclic diether molecules. Moreover, its simple structure and smaller size leads to interstitial accommodation with 1, 3-dioxolane molecules more easily compared to the higher 1-alkanols that have greater structural complexity. Similar results have been reported earlier.[38, 39].

The concept of free volume is an extension of the idea that each molecule is enclosed by its neighbor in a cell. The free volume per molecules may be regarded as the effective volume accessible to the centers of a molecule in a liquid. It is however, evident from the consideration of the liquid state theories that the concept of free volume varies with the specific model chosen for the liquid. The excess free volume (V_f^E) is another important parameter through which molecular interactions can be explained. A perusal of Figure 4 shows that the value of excess Free Volume (V_f^E), are negative for the all binary liquid system 1,3-dioxolane with 1-alkanols at 298.15 K. In the present investigation the negative excess free volume (V_f^E) for binary mixtures of ethyl acetate with alkanols may be attributed to hydrogen bond formation through dipole-dipole interaction between alkanol and 1,3-dioxolane molecule or to structural contributions arising from the geometrical fitting of one component (alkanol) into the other (1,3-dioxolane) due to difference in the free volume between components. In the present investigation the negative excess free volume (V_f^E), for binary mixtures of 1,3-dioxolane with 1-alkanols may be attributed to hydrogen bond formation through dipole-dipole interaction between 1-alkanol and 1,3-dioxolane molecule or to structural contributions arising from the geometrical fitting of 1-alkanol into the 1,3-dioxolane due to difference in the free volume between components.

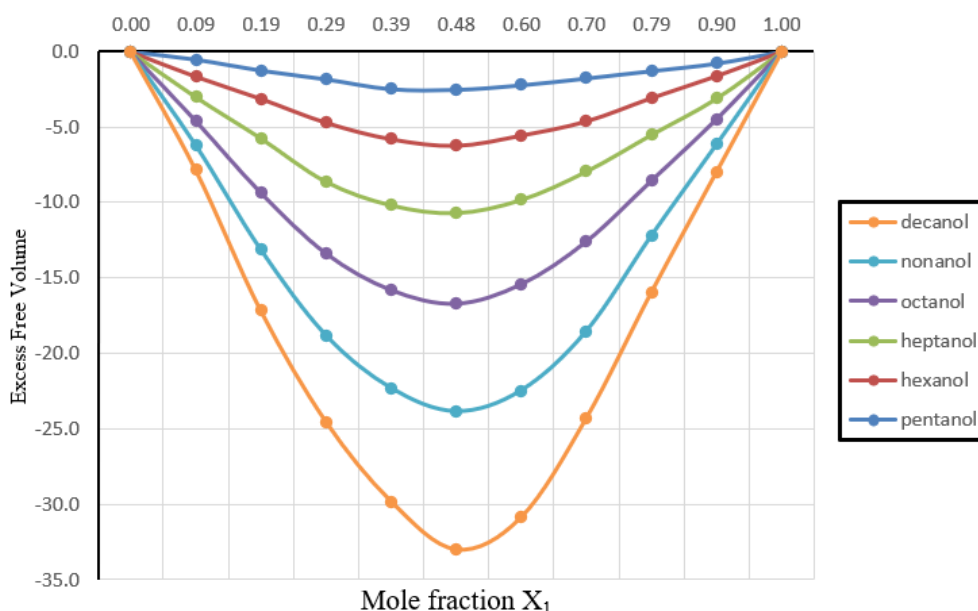


Figure 4. Variation of excess Free Volume (V_f^E), with mole fraction (x_1) of 1,3-dioxolane with 1-alkanols at 298.15K.

Figure 4 indicate that (V_f^E), values are negative for 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 mixtures over the entire mole fraction range. The observed trends in (V_f^E), values for 1,3-dioxolane + 1-alkanols mixtures indicate the

presence of specific interactions between 1,3-dioxolane and 1-alkanols molecules in these mixtures. The magnitude of V_f^E values follow the sequence: Decanol > Nonanol > Octanol > Heptanol > Hexanol > Pentanol, which in turn indicate the interactions in the same order.

Here it is observed that the magnitude of negative values increases with increasing chain length of the alcohols. Alcohol molecules self-associate very strongly (OH---OH interaction), whereas the cyclic diether, 1,3-dioxolane, molecules self-associate rather marginally. This has a dramatic influence on the thermo-physical properties studied. The negative value for the 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 mixture indicates favorable intermolecular complexation through H-bonding, *i.e.*, O---O---H---O. This interaction can be considered as the H-bond formation between the alcohols as Lewis acids and the cyclic diether as a Lewis base [35, 40].

The internal pressure is a cohesive force, which is the result of attractive and repulsive forces between the molecules. The attraction forces mainly consist of hydrogen bonding, dipole-dipole and dispersion interactions. Repulsive forces acting over very small intermolecular distances play a minor role in the cohesion process under normal circumstances.

Figure 5 shows the variation of excess internal pressure (p_i^E) with mole fraction of 1,3-dioxolane at the temperature 298.15K. The excess internal pressure (p_i^E) is another important parameter through which molecular interactions can be explained. In the present investigation for the six binary systems it is observed that, as the mole fraction of 1,3-Dioxolane increase, the (p_i^E) values decrease. The values of (p_i^E) are almost negative. More over the (p_i^E) decrease with increase in X_1 . This situation is observed for all six binary system under study and can be viewed from plots Figure 5.

Excess internal pressure (p_i^E) is found to be negative for six the binary mixtures over the entire composition range at the temperature 298.15K, which suggest the presence of weak intermolecular interaction. It can be seen from Figure 5 that in all the six mixtures, absolute values of decreases as concentration raised of 1,3-Dioxolane. An increment of concentration (X_1) diminishes the self association of the pure component and also the hetero association between unlike molecules, because of the increase of the thermal energy. This lead to less negative values of as concentration is raised as observed in the present binary mixtures. Many workers [41, 42] have reported similar behaviour where negative values of indicates dispersive interactions.

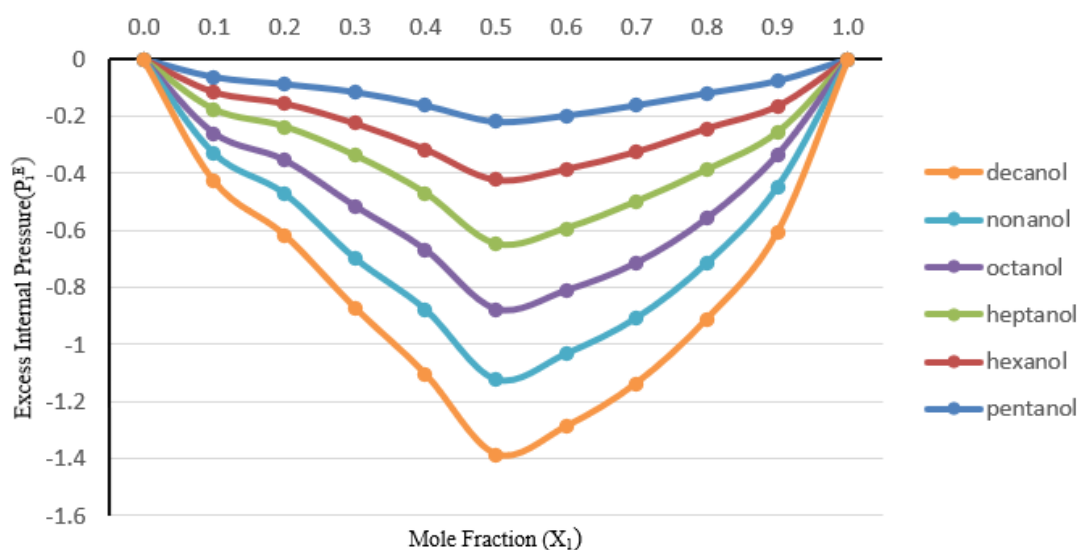


Figure 5. Variation of excess internal pressure (p_i^E) with mole fraction(x_1) of 1,3-dioxolane with 1-alkanols at 298.15K.

Excess enthalpy (H^E) is a crucial thermodynamic quantity that reflects the energetic changes occurring upon mixing. The value of H^E is inherently dependent on temperature, as thermal energy influences intermolecular interactions. Generally, the magnitude and sign of H^E change with temperature: attractive forces may weaken with increasing thermal agitation, leading to reduced exothermic effects or even shifts toward endothermic mixing. Conversely, certain mixtures may exhibit enhanced exothermicity at higher temperatures due to structural reorganization or association phenom-

ena. The sign and magnitude of H^E provide direct information about the nature of intermolecular forces in the mixture. In contrast, negative H^E values reflect exothermic mixing and imply that stronger intermolecular attractions, such as hydrogen bonding, dipole-dipole interactions, or specific complex formation, occur upon mixing. Thus, the analysis of H^E offers a powerful means to assess compatibility, miscibility, and interaction strength between molecular species. Excess molar enthalpy serves as a direct measure of the strength and nature of intermolecular interactions between

components in a binary liquid mixture. Attractive forces, such as hydrogen bonding or van der Waals interactions, contribute favorably to the excess enthalpy, while repulsive forces, arising from steric hindrance or electrostatic interactions, can lead to a decrease in the excess enthalpy. The temperature dependence of excess molar enthalpy offers a nuanced perspective on how these molecular interactions evolve with changing thermal conditions.

Figure 6 shows the variation of excess enthalpy (H^E) with mole fraction of 1,3-dioxolane at the temperature 298.15K. For the binary system 1,3-dioxolane with 1-alkanols, the excess enthalpy (H^E) values are negative and decreasing with the increase in mole fraction of 1,3-dioxolane up to the mole fraction and the increase with increase in mole fraction. The excess enthalpy (H^E) is another important parameter through which molecular interactions can be explained. In the present investigation for the six binary systems it is observed that, as the mole fraction of 1,3-dioxolane increases, the excess enthalpy (H^E) values decrease. This situation is observed for all six binary systems under study and can be viewed from plots Figure 6. 1,3-dioxolane is a polar solvent with a structure that is determined to a large extent by hydrogen bonding. In mixtures of 1,3-dioxolane with an organic liquid at least two effects must be taken into account: hydrogen bond formation between 1,3-dioxolane and the polar group of the solute molecule and structural modification of 1,3-dioxolane around the solute molecule [43]. According to Nakayama

and Shinoda [43], the dependence of the excess enthalpy for the present mixtures may be explained as a balance between positive contributions (hydrogen bond rupture or dispersive interactions between unlike molecules) and negative contributions (intermolecular dipolar interactions or geometrical fitting between components). The behavior of all considered systems containing 1-alkanols is governed by aggregate formation through hydrogen bonding, where 1-alkanols self-association and its complex formation with the proton donors compete. When 1-alkanols is diluted, there is a positive contribution to H^E from the breaking of hydrogen bonds between the alcohol molecules. This effect is relatively more important at low pentanol concentrations than at high concentrations, where the dissociation of the less associated molecules predominates. The negative values of excess enthalpy in the 1-alkanols-rich region are then essentially due to the hydrogen bonded interaction between the hydroxyl oxygen of 1-alkanols and the hydrogen of the other compound and due to the specific interaction between the hydroxyl hydrogen of 1-alkanols with the oxygen (1,3-dioxolane) of the other component.

As a result, the free dipoles released from the alkanols in association with 1,3-dioxolane molecules forming strong hydrogen bonds, hence stronger molecular association existing between the 1,3-dioxolane with 1-alkanols molecules through hydrogen bonding [44-46].

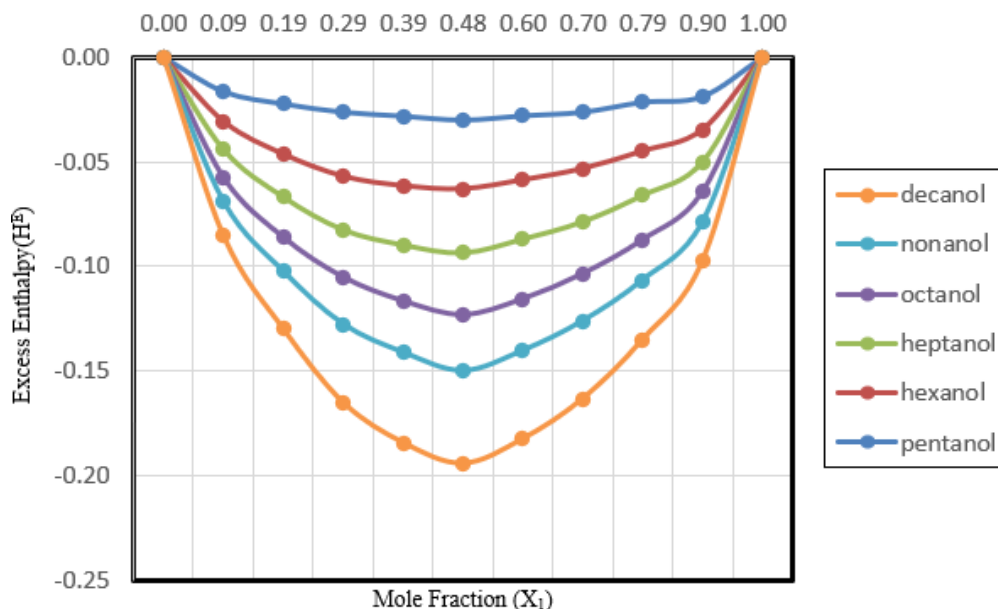


Figure 6. Variation of excess enthalpy (H^E) with mole fraction (x_1) of 1,3-dioxolane with 1-alkanols at 298.15K.

Figure 7 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, indicat-

ing the onset of specific interactions. This observation is consistent with the trends observed for the other parameters discussed above. As a result, forming of strong hydrogen bonds.

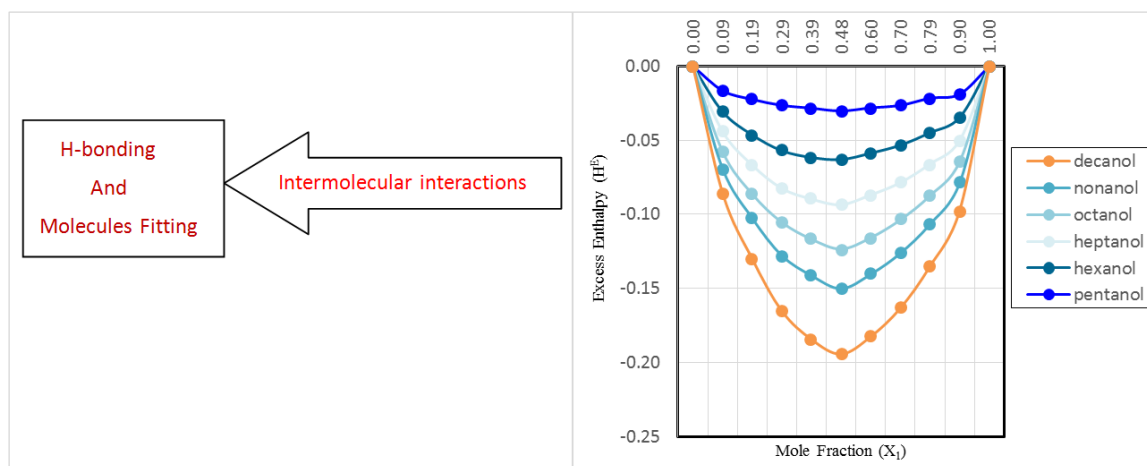


Figure 7. Show that Hydrogen bonding and Molecules fitting.

4. Conclusion

We measured sound velocity, density and viscosity of 1,3-dioxolane with 1-alkanols experimentally at 298.15K. 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), excess enthalpy (H^E), internal pressure (P_i), excess internal pressure (p_i^E), Free Volume (V_f) and excess Free Volume (V_f^E) strongly confirm the presence of strong molecular interactions between the unlike molecules through the hydrogen bonding. After a through study of the behavior of 1-alkanols and 1,3-dioxolane, we get a clear idea about the type and amount of molecular interactions between the components. In addition, molecular interactions are confirmed from the negative values of excess intermolecular free length (L_f^E), excess adiabatic compressibility (β_{ad}^E), excess enthalpy (H^E), excess internal pressure (p_i^E) and excess Free Volume (V_f^E). Hence it is concluded that there exist a molecular interaction between 1,3-dioxolane and 1-alkanols due to Hydrogen bonding.

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
(V_f)	Free Volume

(V_f^E)	Excess Free Volume,
(P_i)	Internal Pressure
(p_i^E)	Excess Internal Pressure
Y^E	Thermodynamic Excess Function

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Author Contributions

Dhirendra Kumar Sharma: Writing – original draft, Writing – review & editing

Suneel Kumar: Conceptualization, Data curation, Formal Analysis

Chandra Pal Prajapati: Investigation, Methodology, Software

Sandeep Sahu: Project administration, Validation, Visualization

Conflicts of Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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