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Research Article

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Excess Thermodynamic Properties of Binary Liquid Mixture of 1,3-dioxolane with 1-Alkanols (C_5-C_{10}) at 298.15K

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ABSTRACT

The work presented in this paper deals with the study of thermodynamic properties of liquid and liquid mixtures. With the help of measured values of ultrasonic velocity (u), density (ρ) and viscosity (η) excess acoustic and thermodynamic properties like excess enthalpy (H^E), excess available volume (V_a^E), excess relaxation strength ($ı^E$) and excess surface tension (S^E) have been calculated for the binary mixture of cyclic diether with 1-alknols at 298.15K by using ultrasonic interferometer technique. The graphs were plotted against the concentration of the mixture at 298.15K, from that the strength of interaction between like and unlike molecules has been discussed.

Keywords: Surface Tension, Available Volume, Excess Relaxation Strength, Enthalpy, Free Volume, Density, Viscosity, Binary Mixture.

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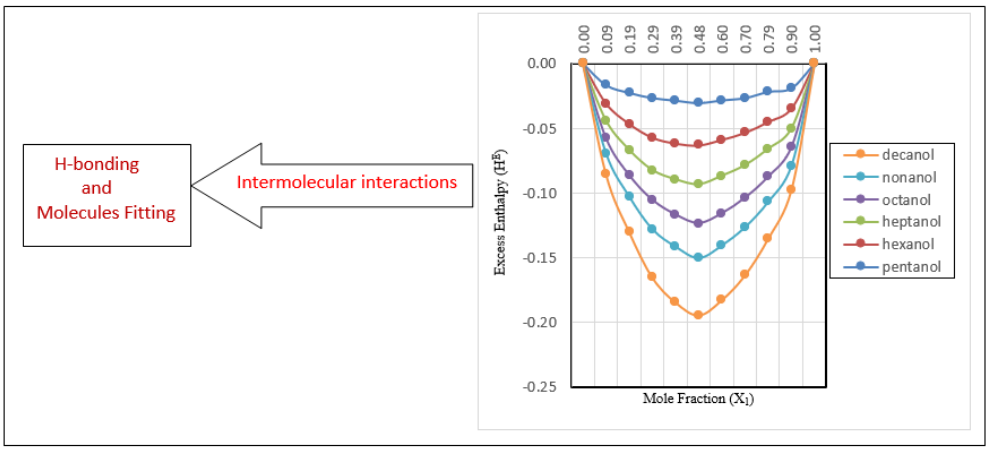
Graphical Abstract



Introduction

Thermodynamic properties derived from the measurement of density, ultrasonic velocity and viscosity for binary mixtures are useful in understanding the nature and type of inter molecular interactions between the component molecules [1, 2]. The estimation of different thermodynamic properties is essential for understanding the interactions occurring in components of unlike molecules. In recent years, there have been considerable interests in theoretical and experimental investigation of the excess thermodynamic properties of polymers [3-5]. The measurement of ultrasonic velocity has been used to elaborate the molecular associations in pure liquids and liquid mixtures. Ultrasonic techniques are highly sensitive to molecular interactions and can be used to accommodate qualitative information about the physical nature and

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strength of molecular interaction in liquid mixtures. The excess thermodynamic and acoustic properties are very essential for understanding the physicochemical behavior and solute–solvent interactions of the binary and multi-component liquid mixtures. The study of molecular associations in liquid mixtures having an alcohol as one component is of particular interest since 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 attractions [6-7]. Alcohols exist in the form of aggregates. When they are mixed with other non-polar molecules, the aggregates of alcohols dissociate and form intermolecular complexes with unlike molecules. Excess thermodynamic properties provide an information for the nature of inter molecular and intra molecular interactions in the liquid mixture. The negative excess values indicate the strong interactions and the positive values show weak interactions between the component's liquid. Excess thermodynamic and thermos acoustic parameters were discussed in terms of the difference in molecular structure and intermolecular forces between the components of the system. Thus, an attempt has been made to explicate the molecular interaction between the binary liquid mixtures of 1,3-dioxolane with 1-alkanols. Alkanols 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 anesthetic agent. The experimental results have been used to discuss the nature of interaction between unlike molecules in terms of hydrogen bonding [8]. The nature and relative strength of the molecular interaction between the components of the liquid mixtures have been successfully investigated by the ultrasonic methods. Thermodynamic and transport properties of binary liquid mixtures with different organic liquids have been studied by many workers.

Thermodynamic studies of binary liquid mixtures have attracted much attention of scientists. These excess values of enthalpy (H^E), available volume (V_a^E), relaxation strength (r^E) and surface tension (S^E) in binary liquid mixture are useful in understanding the solute-solvent interactions [9, 10]. In recent years ultrasonic study of liquid and liquid mixtures has gained much importance during the last four decades in assessing the nature of molecular interaction and investigating the physiochemical behavior of system. In the present paper we have report the results of study on binary liquid mixture of 1,3-dioxolane over the entire range of composition at 298.15K. By using the experimental data of sound velocity (u), viscosity (η) and density (ρ), various acoustical parameters like excess enthalpy (H^E), excess available volume (V_a^E), excess relaxation strength (r^E) and excess surface tension (S^E) have been calculated the mixture. The binary liquid mixtures studied in this paper are as follows:

(i) 1,3-dioxolane- 1-pentanol
(ii) 1,3-dioxolane- 1-hexanol
(iii) 1,3-dioxolane- 1-heptanol
(iv) 1,3-dioxolane- 1-octanol
(v) 1,3-dioxolane- 1-nonanol
(vi) 1,3-dioxolane- 1-decanol

Materials and Methods

The chemicals used are of A.R grade and used without further purification. The liquid mixtures were prepared by mixing the calculated value of molar concentration in an air tight glass bottles to minimize the evaporation and contamination of the solvent. The velocities of ultrasonic waves in the liquid samples have been measured using an ultrasonic interferometer working at a fixed frequency of 3 MHz supplied by Mittal Enterprises New Delhi. The temperature of the liquids in the measuring cell is stabilized by a water circulation thermostat which maintains the temperature within the accuracy of ± 0.2 C. The density is measured using a 5 ml specific gravity bottle with experimental liquid immersed in a temperature-controlled water bath. The viscosities of the pure and liquid mixture are measured using an Ostwald's Viscometer. The chemicals used were of analytical grade and 1,3-dioxolane, pentanol, hexanol, heptanol, octanol, nonanal, decanal purchased from CDH New Delhi, India. 1,3-dioxolane (CDH New Delhi, India) was supplied with purity ≥ 99.7 %, pentanol (CDH New Delhi, India) with ≥ 99.7 %, hexanol (CDH New Delhi India,) with ≥ 99.5 %, heptanol (CDH New Delhi, India) with ≥ 99 %, octanol (CDH New Delhi India,) with ≥ 99.7 %, nonanal (CDH New Delhi, India) with ≥ 99 %, decanal (CDH New Delhi, India) with ≥ 99 %, respectively. All the liquids were used after double distillation [11]. All chemicals were purified by the method described by Zhao et al. 1,3-dioxolane was dried over K2CO3, filtered and distilled were discarded [12]. The measured density, viscosity and sound velocity of the pure component at 298.15 K with the available literature [13-24] as shown in Table 1. The reported experimental values of density (ρ), sound velocity (u) and viscosity (η) conform closely to their corresponding literature values.

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]
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[23]
		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[24]		5.9443[24]
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[24]		9.0200[24]
Decanol	0.8292	0.8267[15]	1378	1380[15]	11.8027	11.825[15]
		0.8264[19]		1379[24]		11.829[15]

Results and Discussion

The experimental values of speed of sound (u), viscosities (η) and densities (ρ) of 1,3-dioxolane with 1-alkanol mixtures at 298.15K are listed in Table 2. From these values, we have computed enthalpy (H), available volume (V_a), relaxation strength (r) and surface tension (S), are presented in table 2.

Table 2: Experimental Values of speed of sound (u), viscosities (η) and densities (ρ) and derived parameter Enthalpy (H), Available volume (V_a), Relaxation Strength (r) and Surface Tension (S) 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}	Viscosity (η) / mPas	Sound velocity (u) / ms^{-1}	Surface Tension (S) × 10^3 / N.m^{-1}	Available volume (V_a) × 10-^{3}/ m^{3}mol^{-1}	Enthalpy (H) × 10^6	Relaxation Strength (r)
			1,3-Dioxolane + 1-Pentanol				
0	0.8124	3.3978	1198	0.2155	0.0272	0.3156	0.0631
0.0939	0.8276	2.3973	1284	0.2436	0.0207	0.3450	0.0390
0.1942	0.8436	1.8970	1290	0.2501	0.0196	0.3468	0.0375
0.2941	0.8640	1.4437	1296	0.2579	0.0184	0.3384	0.0361
0.3942	0.8836	1.1866	1300	0.2650	0.0175	0.3341	0.0351
0.4787	0.9068	1.0904	1304	0.2732	0.0165	0.33.8	0.0342
0.5999	0.9316	0.9311	1310	0.2826	0.0154	0.3262	0.0328
0.6972	0.9596	0.7717	1318	0.293.8	0.0143	0.3236	0.0310
0.7928	0.9876	0.7171	1324	0.3045	0.0134	0.3201	0.0297
0.9035	1.0260	0.6489	1332	0.3192	0.0123	0.3166	0.0280
1.0000	1.0616	0.5885	1340	0.3332	0.0113	0.3135	0.0264
			1,3-Dioxolane +1-Hexanol				
0	0.8176	4.6091	1306	0.2469	0.0229	0.4163	0.0337
0.0912	0.8252	3.3826	1317	0.2524	0.0213	0.4112	0.0312
0.1955	0.8432	2.3306	1320	0.2588	0.0200	0.4003	0.0306
0.2923	0.8584	1.9839	1322	0.2640	0.0190	0.3899	0.0301
0.3982	0.8792	1.5720	1325	0.2713	0.0177	0.3787	0.0295
0.4942	0.8992	1.3059	1327	0.2781	0.0167	0.3683	0.0291
0.6059	0.9264	1.0343	1330	0.2875	0.0155	0.3567	0.0284
0.6976	0.9508	0.9131	1332	0.2958	0.0145	0.465	0.0280
0.8018	0.9836	0.7680	1335	0.3070	0.0133	0.3352	0.0274
0.8914	1.0168	0.7304	1337	0.3181	0.0124	0.3254	0.0270
1.0000	1.0616	0.5885	1340	0.3332	0.0113	0.3135	0.0264
			1,3-Dioxolane + 1-Heptanol				
0	0.8196	5.9066	1325	0.2529	0.0243	0.4838	0.0295
0.0928	0.8304	4.3181	1334	0.2589	0.0224	0.4725	0.0276
0.1905	0.8412	3.2577	1334	0.2623	0.0213	0.4552	0.0252
0.2939	0.8592	2.5895	1335	0.2682	0.0200	0.4373	0.0274
0.3894	0.8740	1.9926	1335	0.2728	0.0188	0.4201	0.0273
0.4818	0.8916	1.5315	1336	0.2786	0.0177	0.4042	0.0272
0.6021	0.9184	1.2190	1337	0.2873	0.0162	0.3835	0.0270

0.6952	0.9420	1.0959	1337	0.2947	0.0151	0.3667	0.0268
0.7892	0.9756	0.9903	1338	0.3055	0.0139	0.3505	0.0267
0.9006	1.0156	0.7057	1339	0.3184	0.0125	0.3309	0.0266
1.0000	1.0616	0.5885	1340	0.3332	0.0113	0.3135	0.0264
1,3-Dioxolane + 1-Octanol							
0	0.8296	7.1508	1350	0.2633	0.0247	0.5619	0.0244
0.0885	0.8296	5.6095	1350	0.2645	0.0235	0.5363	0.0243
0.1967	0.8464	3.9321	1349	0.2683	0.0220	0.5100	0.0246
0.2998	0.8560	3.2616	1348	0.2711	0.0208	0.4845	0.0248
0.3902	0.8712	2.4284	1348	0.2759	0.0195	0.4629	0.0247
0.4963	0.8876	1.9058	1348	0.2811	0.0181	0.4375	0.0246
0.6008	0.9140	1.3631	1347	0.2891	0.0166	0.4117	0.0250
0.6925	0.9340	1.1376	1348	0.2958	0.0154	0.3905	0.0248
0.7975	0.9676	0.9141	1348	0.3064	0.0139	0.3652	0.0247
0.8940	1.0104	0.7652	1348	0.3200	0.0124	0.3421	0.0246
1.0000	1.0616	0.5885	1340	0.3332	0.0113	0.3135	0.0264
1,3-Dioxolane +1-Nonanol							
0	0.8248	8.9258	1366	0.2665	0.0255	0.6291	0.0213
0.0876	0.8336	6.8601	1366	0.2693	0.0242	0.6020	0.0210
0.1913	0.8404	5.8531	1363	0.2706	0.0230	0.5684	0.0219
0.2942	0.8504	4.4022	1359	0.2726	0.0218	0.5347	0.0226
0.3963	0.8692	3.1558	1355	0.2774	0.0205	0.5014	0.0234
0.4959	0.8844	2.3340	1352	0.2813	0.0191	0.4697	0.0240
0.6050	0.9092	1.7321	1349	0.2883	0.0175	0.4354	0.0246
0.6947	0.9332	1.3334	1346	0.2949	0.0162	0.4072	0.0252
0.7993	0.9648	0.9642	1343	0.3039	0.0146	0.3744	0.0258
0.9013	1.0084	0.8031	1340	0.3165	0.0130	0.3402	0.0262
1	1.0616	0.5885	1340	0.3332	0.0113	0.3135	0.0264
1,3-Dioxolane +1-Decanol							
0	0.8292	11.8027	1378	0.2714	0.0264	0.6990	0.0192
0.0881	0.8364	8.5615	1374	0.2726	0.0254	0.6634	0.0199
0.191	0.8396	7.8207	1370	0.2724	0.0243	0.6226	0.0206
0.2921	0.8560	5.5340	1366	0.2765	0.0228	0.5827	0.0213
0.3937	0.8672	4.2319	1362	0.2789	0.0214	0.5429	0.0221
0.4956	0.8824	3.4173	1358	0.2826	0.0199	0.5035	0.0228
0.604	0.9076	2.5370	1353	0.2890	0.0182	0.4615	0.0238
0.7129	0.9308	1.5262	1348	0.2948	0.0166	0.4198	0.0248
0.7983	0.9616	1.1637	1344	0.3032	0.0151	0.3871	0.0256
0.8971	1.0040	0.8623	1340	0.3151	0.0133	0.3505	0.0260
1	1.0616	0.5885	1340	0.3332	0.0113	0.3135	0.0264

The excess parameters such as excess surface tension (S^E), excess available volume (V_a^E), excess relaxation strength (r^E) and excess enthalpy (H^E) have been calculated using the following equations.

Surface Tension $S = 6.4 \times 10^{-3}. \rho. u^{1/2}$ …(1)

Available volume $V_a = (M/\rho)\,[1-(U/U_\infty]$ …(2)

Where M is the molecular weight of the solution which can be calculated according to the equation [$M = M_1X_1 + M_2X_2$] and U_∞ = 1600 m/s.

Relaxation strength $r = 1-(U/ U_{\infty})^2$ …(3)

Enthalpy $H = V_m \times P_i$ …(4)

$Y^E = Y_{exp} - (X_1 Y_1 + X_2 Y_2)$ …(5)

Y^E refer to (S^E), (V_a^E), (r^E), (H^E) and (V_f^E) where as Y_{exp} is measured property. Y_1, Y_2, are any acoustic parameter, X_1 and X_2 are mole fraction of 1,3-Dioxolane and 1-alkanol.

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 [25]. 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 calculated excess surface tension (S^E), values for the binary liquid mixture listed in Figure 1. A perusal of curves in figure-1 indicates the values of excess surface tension (S^E), data for binary mixtures of 1,3-Dioxolane with 1-alkanols are negative. The excess surface tension (S^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. Excess values of surface tension are negative over the entire composition range at 298.15K, indicating weak interaction between the components of the mixture. In all the cases surface tension increases with increase in mole fraction of 1,3-Dioxolane, which indicate reduction in the intermolecular interactions. For all studied systems, excess surface tension (S^E), were negative over the entire mole fraction range of 1,3-Dioxolane at 298.15 K. Among the six systems, 1,3-Dioxolane with 1-alkanols containing system showed more negative.

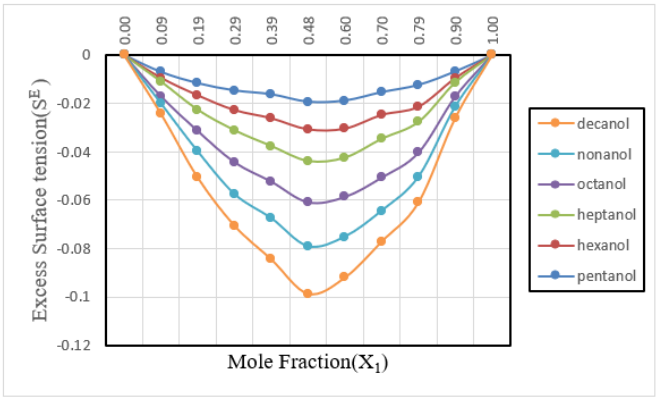

Figure 1: Variation of excess surface tension (S^E) with mole fraction (x_1) of 1,3-dioxolane with 1-alkanols at 298.15K.

Figure 2 depicts the variation of the excess available volume (V_a) of the chosen system. We clearly see from fig. 2 that the non-linear variation of excess available volume (V_a) with mole fraction of 1,3-dioxolane. The negative deviation of excess available volume (V_a) reaching a minimum at 0.48 mole fraction of 1,3-dioxolane shows the molecular interactions between the molecules [26]. It is evident from figure- 2 that the value of excess available volume (V_a^E) are negative for all binary liquid system at 298.15 K. The value of V_a^E are plotted against the mole fraction of 1,3- dioxolane and are shown in Figure 2. Similarly, results were also obtained by D. Bala Karuna Kumar et al. for binary mixtures of N-methyl-2-pyrrolidone [27]. The de-polymerisation of hydrogen bonded alcohols aggregates and decrease in dipolar association of component molecules leads to expansion of volume dominating interstitial accommodation which causes negative value of excess available volume (V_a^E) but it is compensated by charge transfer complex formation between hydrogen of alcohols and oxygen of 1,3- dioxolane. From the plots of (V_a^E) Vs X_1 (Figure- 2) it may see that excess available volume is negative over the whole mole fraction.

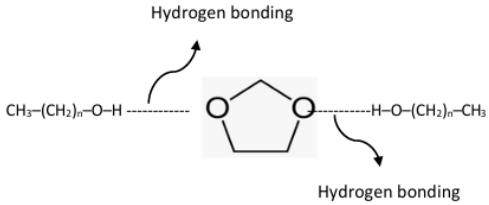

Figure: Hydrogen bonding present in 1,3-dioxolane n-alkanols.

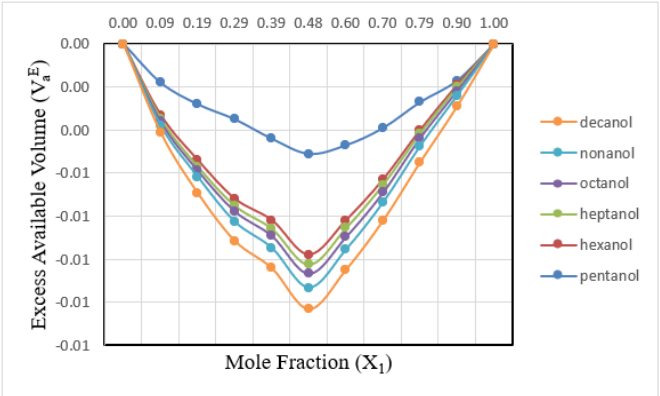

Figure 2: Variation of excess available volume (V_a^E) with mole fraction (x_1) of 1,3-dioxolane with 1-alkanols at 298.15K.

The term relaxation Strength (r) is useful in prediction of molecular properties of liquid mixtures. The relaxation strength (r) decreases with increases in concentration of 1,3-dioxolane (X_1). Figure 3 shows the variation of excess relaxation strength (r^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 relaxation strength (r^E) values are negative and decreasing with the increase in mole fraction of 1,3-dioxolane up to the mole fraction (0.5) and the increase with increase in mole fraction. We clearly see from fig. 3 that the non-linear variation of excess relaxation strength (r^E) with mole fraction of 1,3-dioxolane. The negative deviation of excess relaxation strength (r^E) 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 relaxation strength (r) of the system. The excess relaxation strength (r^E) (shown in figure (3) 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 relaxation strength (r) indicates the squeezing of molecules in the system.

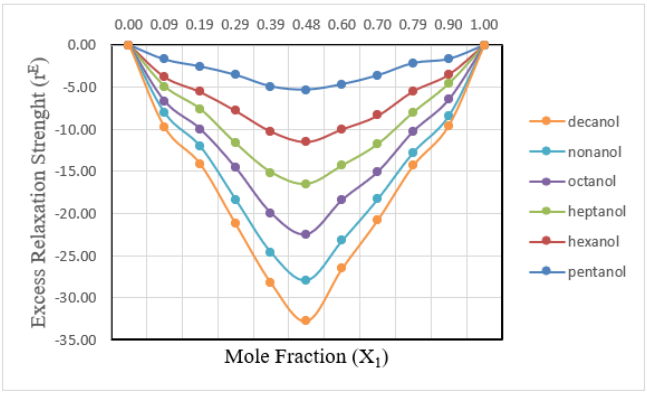

Figure 3: Variation of excess relaxation Strength (r^E) with mole fraction (x_1) of 1,3-dioxolane with 1-alkanols at 298.15K.

Figure 4 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 (0.5) and the increase with increase in mole fraction. In the present investigation for the six binary systems it is observe that, as the mole fraction of 1,3-dioxolane increase, the excess enthalpy (H^E) values decrease. This situation is observed for all six binary system under study and can be viewed from plots Figure 4. This suggests that dipole and dispersive force are operative in these systems, when the 1,3-dioxolane concentration low. When the concentration of 1,3-dioxolane increased, the corresponding decrease in concentration of 1,3-dioxolane leads to specific interactions i.e., the interactions move from weak to strong which supports the above arguments is case of other parameters. As a result, the free dipoles released from the 1-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 [27].

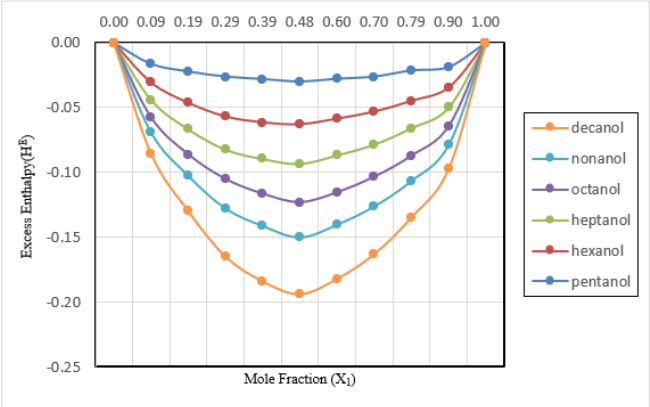

Figure 4: Variation of excess enthalpy (H^E) with mole fraction (x_1) of 1,3-dioxolane with 1-alkanols at 298.15K.

Conclusion

This research article reports experimental study of sound velocity, viscosity and density and its derived excess parameters. The existence of molecular interactions in solute-solvent is favored in the system, confirmed from the ultrasonic velocity (u), viscosity (η), density (ρ), surface tension (S), available volume (V_a), relaxation strength (r), enthalpy (H). We are concluded that there exists a molecular interaction between 1,3-dioxolane and 1-alkanols due to Hydrogen bonding and degree of complexation.

Acknowledgement

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Nomenclature

ρ, Densities of liquid
u, Ultrasonic velocity
η, Viscosity
X_1, Mole fraction of 1,3-Dioxolane
T, Temperature
S, surface tension
S^E, excess surface tension
(V_a), available volume
(V_a^E), excess available volume
(r), relaxation strength
(H), Enthalpy
(H^E), Excess enthalpy
Y^E, Thermodynamic excess function

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