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Research Article

EVALUATION OF THERMO DYNAMICAL ACOUSTIC PARAMETERS OF BINARY MIXTURE OF CYCLIC ETHER WITH 1-ALKANOLS AT 303.15 K AND AT 3 M HZ. FREQUENCIES

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A B S T R A C T

Ultrasonic investigation of molecular interactions in a binary mixture of cyclic ether (1,4-dioxane) with 1-alkanols at 303.15K over the frequency 3 MHz. The ultrasonic velocity, density and viscosity of the 1, 4- dioxane with 1-alkanols system have been measured experimentally. From these three values, we have calculated thermodynamic and excess thermodynamic parameters such as adiabatic compressibility, deviation in adiabatic compressibility, molar volume, excess molar volume, free length, excess free length, deviation in viscosity, internal pressure, excess internal pressure, acoustic impedance, excess acoustic impedance, excess sound velocity, Rao's Constant, Wada Constant, relaxation time and relaxation strength of binary liquid mixture to interpret the nature of the interactions taking place in the binary liquid mixtures. The results are analyzed in the sight of molecular interaction between the components. The interaction resulting in the interstitial accommodation of 1,4-dioxane in to 1-alkanol are predominant factor over dipole-dipole interaction.

INTRODUCTION

Ultrasonic velocity in a medium is fundamentally related to the binding forces between the constituent molecules. Ultrasonic velocities of liquid mixture are of considerable importance in studying intermolecular interaction between component molecules and used to compute various physical and chemical parameters which have wide applications in several industrial and engineering processes [1-5]. The present work is a continuation of systematic experimental studies on thermodynamic properties of binary liquid mixtures of 1,4-Dioxane with 1-alkanols at 303.15K

In recent years, there has been considerable development in the experimental investigation of the thermodynamic properties of liquid and liquid mixtures are used to study the molecular interactions between the various components of the mixtures. An ultrasonic technique has become a powerful tool for studying the molecular behavior of liquid mixtures. 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]. The 1,4-dioxane is selected as a solvent in the present work since it finds a variety of application. Alcohols play an important role in many chemical reactions due to the ability to undergo self-association with internal structures. 1,4-Dioxane cyclic ether is used as a degreasing agent, as a component of paints and varnish removers and as a wetting and dispersion agent in the textile industry. 1,4- Dioxane is also used as a solvent in the chemical synthesis. Ultrasonic studies of the solution of the alcohols with 1,4- dioxane have yielding valuable information regarding the association between the momomers of alcohols and free oxygen of dioxane through hydrogen bonding.

Ultrasonic propagation parameters yield valuable information regarding the behaviour of liquid systems because intermolecular and intermolecular association, complex formation, dipolar interactions and related structural charges effect 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 interaction that influence the structured arrangement along with the shape of the molecules[9-12]. In the present study, density, viscosity and ultrasonic velocity of binary mixture 1,4-Dioxane with 1-alkanols at 303.15K at frequencies 3MHz. These measured values are used to calculate different parameters like impedance (Z), adiabatic compressibility (βad), intermolecular free length (L_f), Rao's constant (R), Wada's constant (W), molar volume (V_m), internal pressure (pi), viscous relaxation time (τ) and relaxation strength (r).

The excess parameters of ultrasonic velocity (u^E), molar volume (V^E), acoustic impedance (Z^E), adiabatic compressibility ($\Delta\beta ad$), intermolecular free length ($L_f{}^E$),

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excess internal pressure (p_i^E), are computed, which are highly useful in understanding the nature and strength of molecular interactions, internal structure and the aggregation behavior.

MATERIALS AND METHODS

The chemicals used in the present work were high purity laboratory reagent grade samples of 1,4-dioxane, 1-methanol, 1-ethanol, 1-propanol, 1-butanol, 1-hexanol, 1-octanol purchased from Merck Chem. Ltd India. All chemicals was stored over sodium hydroxide pellets for several days. All the chemicals were kept in tightly sealed bottles to minimize the absorption of atmospheric moisture. The purity of the solvent was ascertained by comparing the measured density, dynamic viscosities and sound velocity of the pure component at 303.15K with the available literature [13-24] as shown in Table 2.

Table 1 Provenance and purity of the materials used

Component	CAS number	Source	Initial mass fraction purity
1,4-Dioxane	17647-74-4	Merck Chem. Ltd India	0.995
1-Methanol	67-56-1	Merck Chem. Ltd India	0.995
1-Ethanol	64-17-5	Merck Chem. Ltd India	0.995
1-Propanol	71-23-8	Merck Chem. Ltd India	0.995
1-Butanol	71-36-3	Merck Chem. Ltd India	0.995
1-Hexanol	111-27-3	Merck Chem. Ltd India	0.995
1-Octanol	111-87-5	Merck Chem. Ltd India	0.995

Measurements

Six binary system viz. 1,4-dioxane + 1-methanol, 1,4-dioxane + 1-ethanol, 1,4-dioxane + 1-propanol, 1,4-dioxane + 1-butanol, 1,4-dioxane + 1-hexanol and 1,4-dioxane +1-octanol were studied. Each sample mixture was prepared, on mass basis, by mixing the calculated volume of liquid components in specially designed glass stoppered bottles. All binary mixtures were prepared by weight covering the entire mole fraction range. The components of binary mixtures were injected by means of syringe in to the glass vials of sealed with rubber stopper in order to check evaporation losses during sample preparation. The mass measurements were carried out using an single pan analytical balance (Model K-15 Deluxe, K Roy Instruments Pvt. Ltd.) with an accuracy of ± 0.00001×10^{-3} kg as described elsewhere[25]. 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.

Density

Densities of pure liquids and their binary mixtures were determined by using a double-arm pycnometer [26] with a bulb of 25 cm^3 and a capillary of an internal diameter of about 1 mm is used to measure the densities (ρ) of pure liquids and binary mixtures. The pycnometer is calibrated by using conductivity water (having specific conductance less than 1×106 ohm^{-1}) with 0.9970 and 0.9940 g cm^{-3} as its densities at T = 303.15 K, respectively. The pycnometer 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}. The observed values of densities of pure 1,4-dioxane, 1- methanol, 1-ethanol, 1-propanol, 1-butanol, 1-hexanol and 1-octanol at 303.15K were 1.0108, 0.7840, 0.7720, 0.8070, 0.8040, 0.8128 and 0.8242g.cm^{-3} which compare well with corresponding literature values ofrespectively.

Sound velocity

The ultrasonic velocities were measured using a multifrequency ultrasonic interferometer (Model F-80D, Mittal Enterprise, New Delhi, India) working at 3 M.Hz. The meter was calibrated with water and benzene at 303.15K. The measured values of ultrasonic velocities of pure 1,4-dioxane, 1- methanol, 1-ethanol, 1-propanol, 1-butanol, 1-hexanol and 1-octanol at 303.15K were 1322.3, 1084, 1141, 1182, 1196, 1298 and 1327 $m.s^{-1}$ respectively, which compare well with the corresponding literature values.

Viscosity

The viscosity of pure liquids and their binary mixture were measured using suspended ubbelohde type viscometer [27-28] 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 303.15 K. The details of the methods and techniques have been described by researchers [29-30]. 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. The measured values of viscosities of pure 1,4-dioxane,1-methanol, 1-ethanol, 1-propanol, 1-butanol, 1-hexanol and 1-octanol at 303.15 K were 1.0303, 0.4949, 1.1399,1.5477, 2.2045, 4.5642 and 7.8512 C.P. which compare well with the corresponding literature values.

Table 2 Physical properties of pure components at 303.15K

component	Density (ρ) g-m^{-3}		Ultrasonic Velocities (u) $m.s^{-1}$		Viscosity (η) CP	
	Observed	Literature	Observed	Literature	Observed	Literature
1,4-dioxane	1.0108	1.0229 [23]	1348	1322.3 [24]	1.0303	1.0690 [17]
Methanol	0.7840	0.7817 [14]	1084	1084.0 [21]	0.4949	0.5040 [15]
Ethanol	0.7720	0.7807 [13]	1141	1144.3 [15]	1.1399	1.3560 [13]
Propanol	0.8070	0.8003 [20]	1182	1182.6 [15]	1.5477	1.6626 [15]
Butanol	0.8040	0.8020 [15]	1196	1196.6 [15]	2.2045	2.2740 [16]
Hexanol	0.8128	0.8118 [15]	1298	1282.0 [22]	4.5642	4.5930 [18]
Octanol	0.8242	0.8187 [19]	1327	1330.8 [19]	7.8512	7.6630 [18]

Theoretical

The ultrasonic velocity (u), density (ρ) and viscosity (η) in pure liquids and liquid mixtures of various concentrations have been measured at 303.15 K. Thermodynamic and acoustical parameters such as adiabatic compressibility (β), free length (L_f), acoustic impedance (Z), relaxation strength (r), Rao's constant (R), molar compressibility or wada constant (W), relaxation time (τ) and internal pressure (p_i) were determined using the observed values of velocity, density and viscosity using the standard relations given below.

The adiabatic compressibility (β_{ad}) has been determined by using experimentally measured ultrasonic velocity (u) and density (ρ) by using the following relation:

$$= \quad 1 / \rho u^2 \qquad (1)$$

The mean molar volume (V) of binary liquid mixtures at a given molefraction is given by:

$V = M / \rho$ (2)

The excess volume (V^E) at a given mole fraction is the different between mean molar volume and the average volume is calculated using the given formula:

$$V^E = V - (V_1 x_1 + V_2 x_2) \quad (3)$$

Where V_1 and V_2 are the mean molar volumes of pure liquids 1 & 2 respectively.

The excess compressibility (β_{ad}^{E}) at a given mole fraction is the different between adiabatic compressibility (β_{ad}) and the sum of the fractional contributes of the two liquids is:

$$= \beta_{ad} - (\beta_{ad1} x_1 + \beta_{ad2} x_2) \quad (4)$$

Where β_{ad_1} and β_{ad_2} are the individual β_{ad}, values of pure liquid in thebinary mixtures at that Temperature.

The molar sound velocity or Rao's constant has been calculated usingformula:

$$R = (M/\rho)\, u^{1/3} \quad (5)$$

Where M is the molecular weight of the solution which can be calculatedaccording to the equation

$$M = M_1 x_1 + M_2 x_2 \quad (6)$$

Where x_1 and x_2 are mole fraction of solvent and solute, respectively. M_1 and M_2 are the molecular weights of the solvent and solute respectively.

Molar compressibility or Wada's constant (W) can be calculated by thefollowing equation

$$W = (M/\rho)\, \beta^{1/7} \quad (7)$$

Intermolecular free length (L_f) is calculated using the standardexpression

$$L_f = K \beta^{1/2} \quad (8)$$

Where K is Jacobson's constant which is temperature dependent parameter. The excess viscosity (η^E) at a given mole fraction is the difference between viscosity (η) and the sum of the fraction contribution of the liquid are givenby Fort and Moore is:

$$\eta^E = \eta_{mix} - (\eta_1 x_1 + \eta_2 x_2) \quad (9)$$

Where η_{mi}, η_1 & η_2 are the individual viscosity values of pure liquid in the binary mixture and viscosity of mixture.

The excess mean free length (L_f^E) at a given mole fraction is the difference between mean free length and sum of the fraction contribution of the two liquid are given by:

$$L_f^E = L_{f\,mix} - (L_{f1} x_1 + L_{f2} x_2) \quad (10)$$

Where $L_{f_{mix}}$, L_{f1} and L_{f2} are the individual mean free length values of pure liquid in the binary mixture and mean free length of mixture.

Specific acoustic impedance (Z) is also calculated using the below relation:

$$Z = u.\, \rho \quad (11)$$

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

Excess acoustic impedance (Z^E) is difference between the ideal acousticimpedance and acoustic impedance of the pure components i.e.

$$Z^E = Z_{real} - Z_{idea} \quad (12)$$

Excess velocity (u^E) is difference between the ideal velocity and velocity of the pure components.

$$u^E = u_{mix.} - (u_2 x_1 + u_2 x_2) \quad (13)$$

(6)

Relaxation strength (r) has been calculated by the following relation:

$$r = 1 - u^2 / u_\infty \quad (14)$$

Where u_∞ = 1600 m/sec

RESULT AND DISCUSSION

The experimental values of density (ρ), viscosity (η) and sound velocity (u) for the binary liquid mixtures of 1,4-dioxane with 1-alkanols in different mole fraction at constant temperature 303.15 K over the frequency 3 M Hz are given in Table -3.

From the standard parameters the values of derived parameters adiabatic compressibility (β_{ad}), molar volume (V_m), intermolecular free length (L_f), internal pressure (p_i), Rao's constant acoustic impedance (Z), Wada constant (W), Viscous relaxation time (τ) and relaxation strength (r) are shown in Table-4.

Table 3 Values of density, sound velocity and viscosity for binary liquids mixtures of 1,4-dioxane + methanol, 1,4-dioxane + ethanol, 1,4-dioxane + propanol, 1,4-dioxane + butanol, 1,4-dioxane + hexanol and 1,4-dioxane +octanol at 303.15 K.

Mole fraction of 1,4-dioxane (x_1)	Density (ρ) g.m^{-3}	Sound velocity (u) m.s^{-1}	Viscosity(η) c. p.
1,4-dioxane + 1-Methanol			
0.00000	0.7840	1084.0	0.4949
0.09770	0.82748	1092.0	0.5704
0.20043	0.86692	1130.0	0.6005
0.28674	0.89696	1155.0	0.6339
0.38010	0.91716	1176.0	0.6703
0.49857	0.94224	1240.0	0.7168
0.59198	0.96312	1266.0	0.7802
0.70860	0.98772	1289.0	0.8441
0.80020	0.99876	1306.0	0.9426
0.90362	1.00892	1330.0	1.0244
1.00000	1.0108	1348.0	1.0303

1,4-dioxane + 1-Ethanol			
0.00000	0.7720	1141.0	1.1399
0.09885	0.80944	1150.0	1.1038
0.20465	0.8426	1170.0	1.0986
0.29964	0.8652	1189.0	1.0814
0.39745	0.8990	1217.0	1.0740
0.50220	0.92016	1285.0	1.0710
0.59502	0.94108	1288.0	1.0697
0.69003	0.9720	1298.0	1.0593
0.79934	0.98624	1310.0	1.0485
0.89342	0.99356	1340.0	1.0406
1.00000	1.0108	1348.0	1.0303
1,4-dioxane + 1-Propanol			
0.00000	0.80708	1182.0	1.5477
0.10006	0.82064	1202.0	1.4964
0.12264	0.84796	1215.0	1.2610
0.29821	0.87008	1248.0	1.1498
0.40573	0.89936	1264.0	1.1319
0.50439	0.91672	1270.0	1.1274
0.60251	0.93908	1275.0	1.1097
0.69410	0.9558	1284.0	1.0881
0.79626	0.98084	1290.0	1.0711
0.89926	1.0054	1312.0	1.0534
1.00000	1.0108	1348.0	1.0303
1,4-dioxane + 1-Butanol			
0.00000	0.8040	1196.0	2.2045
0.09734	0.8136	1203.0	1.7804
0.19759	0.84252	1209.0	1.4627
0.30443	0.86264	1221.0	1.3458
0.40480	0.87696	1268.0	1.1934
0.49442	0.90224	1282.0	1.1939
0.59768	0.92744	1287.0	1.1879
0.68628	0.95028	1297.0	1.1044
0.79076	0.97392	1315.0	1.0953
0.89091	0.99836	1334.0	1.0728
1.00000	1.0108	1348.0	1.0303
1,4-dioxane + 1-Hexanol			
0.00000	0.8128	1298.0	4.5642
0.09108	0.83796	1302.0	3.2904
0.19485	0.85408	1311.0	2.7369
0.29842	0.86024	1314.0	2.2727
0.40439	0.88144	1320.0	1.9013
0.45430	0.88996	1334.0	1.7643
0.60286	0.92576	1338.0	1.4365
0.69974	0.9486	1340.0	1.2914
0.80182	0.9684	1342.0	1.2103
0.88834	0.99384	1346.0	1.1144
1.00000	1.0108	1348.0	1.0303
1,4-dioxane + 1-Octanol			
0.00000	0.8242	1327.0	7.8512
0.09780	0.8284	1329.0	5.1466
0.20653	0.83708	1330.0	4.6513
0.29810	0.85292	1332.0	3.2294
0.40275	0.85956	1334.0	2.5625
0.49229	0.88528	1336.0	2.3806
0.60068	0.90304	1338.0	1.8916
0.69888	0.92664	1339.0	1.4950
0.79610	0.95648	1341.0	1.3490
0.89749	0.98596	1345.0	1.1845
1.00000	1.0108	1348.0	1.0303

The values of excess viscosity (η^E), excess molar volume (V^E), excess adiabatic compressibility (β_{ad}), excess internal pressure ($p_i{}^E$), and excess free length (L_f^E) are listed in Table -5.

Various type of interaction which are possible and which can operate in the binary liquid mixtures containing 1,4- dioxane are hydrogen bonded complex formation, that can produce negative deviation in excess viscosity, excess molar volume, excess adiabatic compressibility, excess internal pressure and excess free length. This may be fact that O-H hydrogen bonded complex formation.

Table 4 Calculated values of adiabatic compressibility (β_{ad}), Molar volume (V_m), intermolecular free length (L_f), internal pressure (p_i), acousticimpedance (Z), Wada constant (W), Rao's constant (R), viscous relaxation time (τ) and relaxation strength (r) at 303.15 K.

X1	$\beta_{ad}\times10^{-8}$ (m^2N^{-1})	V $m\times10$ m^3mol^{-1}	$L_f\times10^{-9}$m	($p_i\times10^4$)	R (m^3/mol) (m/s)$^{1/3}$	Z × 10^4 (kg/m^2s)	W ($m^3/mol_{Pa}1/7$)	τ ×10^{-10} sec	r
				1,4-dioxane + 1-Methanol					
0.0000	108.54	40.8673	2.22022	1.85827	0.41883	84.9856	0.29074	7.86277	0.54099
0.0977	101.34	45.3365	2.14527	1.55867	0.46571	90.3520	0.32567	7.70821	0.53419
0.2004	90.336	49.9160	2.02542	1.29924	0.59827	97.9597	0.36451	7.23391	0.50121
0.2867	83.572	53.6390	1.94811	1.13200	0.56143	103.591	0.39608	7.06461	0.47889
0.3801	78.838	58.1676	1.89214	0.98575	0.61242	107.850	0.43307	7.04625	0.45977
0.4985	69.023	63.6649	1.77043	0.83121	0.68238	116.832	0.48318	6.59746	0.39937
0.5919	64.781	67.7222	1.71517	0.75747	0.73093	121.928	0.51867	6.43909	0.37392
0.7086	60.934	72.6602	1.66346	0.67723	0.78879	127.314	0.56127	6.40819	0.35096
0.8002	58.701	76.9994	1.63271	0.63839	0.83960	130.430	0.59800	6.3725	0.33373
0.9036	56.032	81.9704	1.59515	0.58824	0.89927	134.183	0.64087	6.2519	0.30902
1.0000	54.444	87.1685	1.57239	0.52793	0.96060	136.255	0.68433	7.47987	0.29019
				1,4-dioxane + 1-Ethanol					
0.0000	99.497	59.6761	2.12563	1.48567	0.62212	88.0852	0.42985	15.1222	0.49145
0.0988	95.038	62.0470	2.0596	1.33256	0.64849	93.081	0.45094	13.7489	0.48339
0.2046	90.272	64.8839	1.98420	1.17588	0.68207	98.5842	0.47663	12.7004	0.46527
0.2996	85.991	67.8040	1.92682	1.02250	0.71657	102.872	0.50225	11.7887	0.44776
0.3974	81.590	69.8318	1.84677	0.92291	0.74369	109.408	0.52354	10.7556	0.42144
0.5022	76.861	73.0067	1.72881	0.81416	0.79149	118.240	0.55761	9.39907	0.35499
0.5950	72.681	75.5312	1.70551	0.75046	0.79374	121.200	0.56093	9.13595	0.35197
0.6900	68.402	77.2383	1.66523	0.69393	0.84046	126.165	0.59647	8.62480	0.34491
0.7993	63.484	80.7854	1.63802	0.62849	0.88178	129.192	0.62683	8.26058	0.32964
0.8934	59.246	84.1700	1.59544	0.57388	0.92563	133.129	0.65799	7.77771	0.29859
1.0000	54.444	87.1685	1.57239	0.52793	0.96060	136.255	0.68433	7.47987	0.29019
				1,4-dioxane + 1-Propanol					
0.0000	88.684	74.4563	2.00681	1.12536	0.78535	95.3874	0.54517	18.3009	0.45424
0.1000	84.340	76.6390	1.95705	1.02842	0.81289	98.6361	0.56518	16.8286	0.43562
0.1226	79.886	77.9160	1.90466	0.94383	0.79744	103.019	0.55675	13.4319	0.42334
0.2982	73.792	78.6654	1.83058	0.79892	0.84487	108.576	0.59129	11.3130	0.3916
0.4057	69.594	79.4542	1.77774	0.74945	0.85694	113.671	0.60221	10.5034	0.3759
0.5043	67.632	80.9651	1.75251	0.70945	0.87468	119.253	0.61606	10.1672	0.36996
0.6025	65.505	81.9650	1.72473	0.67187	0.88665	119.722	0.62669	9.69287	0.36499
0.6941	67.174	82.6892	1.74656	0.63513	0.90222	122.724	0.63393	9.20750	0.3916
0.7962	61.266	84.0102	1.66799	0.60302	0.91233	126.523	0.64851	8.75025	0.34996
0.8992	57.782	84.8285	1.61987	0.56923	0.92636	131.908	0.66027	8.11630	0.3276
1.0000	54.444	87.1685	1.57239	0.52793	0.96060	136.255	0.68433	7.47987	0.29019
				1,4-dioxane + 1-Butanol					
0.0000	86.952	92.1902	1.98712	0.93886	0.97625	96.1584	0.67695	25.5582	0.44124
0.0973	84.929	92.7720	1.96386	0.82365	0.80086	97.8760	0.68353	20.1612	0.43468
0.1975	81.202	91.2552	1.92029	0.73831	0.96979	101.858	0.67662	15.8375	0.42903
0.3044	77.756	90.8593	1.87911	0.69325	0.96859	105.323	0.67784	13.9535	0.41764
0.4048	70.922	90.9767	1.79462	0.62881	0.98231	111.190	0.68774	11.2858	0.37194
0.4944	67.437	89.8172	1.74998	0.62110	0.97330	115.662	0.68385	10.7355	0.35799
0.5976	65.096	88.9342	1.71934	0.61152	0.96505	119.356	0.68060	10.3109	0.35298
0.6862	62.555	88.1010	1.68545	0.58231	0.95848	123.251	0.67807	9.21223	0.34288
0.7907	59.377	87.4632	1.64208	0.56878	0.95590	128.067	0.67818	8.67223	0.32452
0.8909	56.286	86.7254	1.59876	0.55292	0.95237	133.173	0.67761	8.04942	0.30486
1.0000	54.444	87.1685	1.57239	0.52793	0.96060	136.255	0.68433	7.47987	0.29019
				1,4-dioxane + 1-Hexanol					
0.0000	73.024	125.691	1.82102	0.76533	1.36781	105.501	0.94625	44.4396	0.34187
0.0910	70.397	120.389	1.78797	0.67619	1.31145	109.102	0.91162	30.8850	0.33781
0.1948	68.123	116.409	1.75886	0.63767	1.27107	111.959	0.88515	24.8602	0.32862
0.2984	67.327	113.884	1.74855	0.59775	1.24438	113.030	0.86736	20.4021	0.32554
0.4043	65.111	109.455	1.71954	0.56874	1.19781	116.344	0.83762	16.5067	0.31937
0.4543	63.141	107.619	1.69333	0.55520	1.18186	118.712	0.82719	14.8541	0.30486
0.6028	60.337	101.202	1.65530	0.53273	1.11250	123.858	0.78294	11.5574	0.30068
0.6997	58.709	97.3310	1.63281	0.53284	1.07047	127.112	0.75594	10.1091	0.29859
0.8018	57.337	93.8597	1.61362	0.52919	1.03281	129.959	0.73144	9.25342	0.29649
0.8883	55.538	90.2338	1.58810	0.52758	0.99389	133.770	0.70640	8.25288	0.29229
1.0000	54.444	87.1685	1.57239	0.52793	0.96060	136.255	0.68433	7.47987	0.29019
				1,4-dioxane + 1-Octanol					
0.0000	68.901	158.005	1.76887	0.66872	1.73215	109.371	1.19933	72.1360	0.31213
0.0978	68.345	152.230	1.76172	0.57272	1.66968	110.094	1.15754	47.0410	0.31006
0.2065	67.535	145.177	1.75120	0.58289	1.59276	111.321	1.10251	41.8837	0.30902
0.2981	66.082	137.962	1.73230	0.51855	1.51433	113.606	1.05355	28.4547	0.30694
0.4027	65.375	131.769	1.72301	0.49443	1.44707	114.657	1.00780	22.3197	0.30486
0.4922	63.285	123.674	1.69526	0.51382	1.35892	118.262	0.95034	20.0882	0.30277
0.6006	61.855	116.193	1.67599	0.49788	1.27729	120.821	0.89573	15.6014	0.30068
0.6988	60.190	108.770	1.65328	0.48131	1.19599	124.071	0.84177	11.9981	0.29964
0.7961	58.138	101.096	1.62486	0.49999	1.11216	128.253	0.78628	10.4573	0.29754
0.8974	56.065	93.7423	1.59562	0.51467	1.03228	132.603	0.73287	8.85491	0.29334
1.0000	54.44	87.1685	1.57239	0.52793	0.96060	136.255	0.68433	7.47987	0.29019

Plot of excess viscosity versus mole fraction in Figure -1.

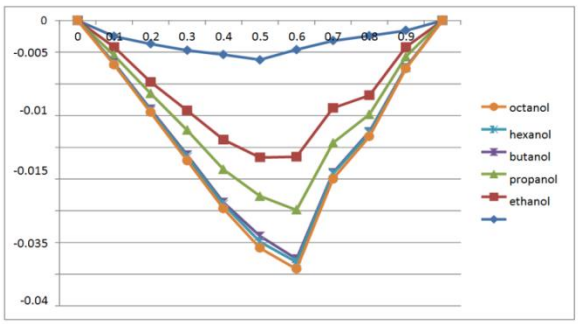

Figure 1 Plots of excess viscosity versus mole fraction of 1,4-dioxane (x_1) at 303.15 K for binary mixtures of 1,4-dioxane with methanol, ethanol, propanol, butanol, hexanol and octanol at 303.15 K.

It decrease with increase in concentration of 1,4-dioxane. It is observed that the converse on the behaviour of adiabatic compressibility and intermolecular free length from the Table – 4. This may be due to self-association of the solute molecules and very weak dipole- induced dipole interaction between the component molecules [31] The adiabatic compressibility decrease with increasing concentration which is due to the strong molecular interaction among the solute and solvent molecules shown in Figure -2.

Figure -3 depicts the variation in intermolecular free length. This decrease in free length in due to the decrease adiabatic compressibility which brings the molecules to a closer packing[32].

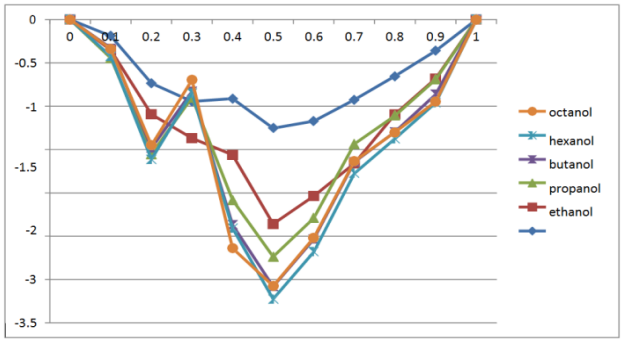

Figure 2 Plots of adiabatic compressibility versus mole fraction of 1,4- dioxane (x_1) at 303.15 K for binary mixtures of 1,4-dioxane with methanol, ethanol, propanol, butanol, hexanol and octanol at 303.15 K.

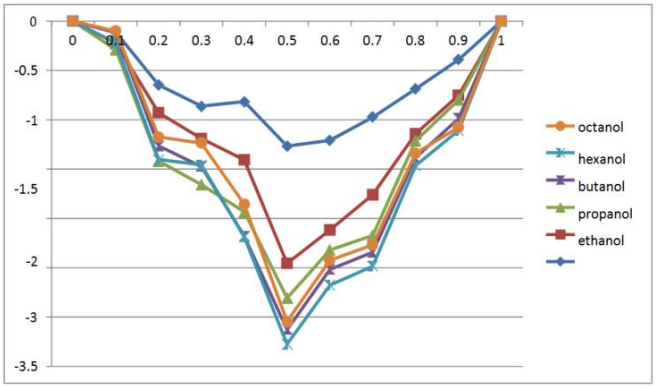

Figure 3 Plots of excess free length versus mole fraction of 1,4-dioxane (x_1) at 303.15 K for binary mixtures of 1,4-dioxane with methanol, ethanol, propanol, butanol, hexanol and octanol at 303.15 K.

Figure-4 it can been seen that molar volume of the liquid mixture increases. Increase in molar volume shows that presence of strong intermolecular forces.

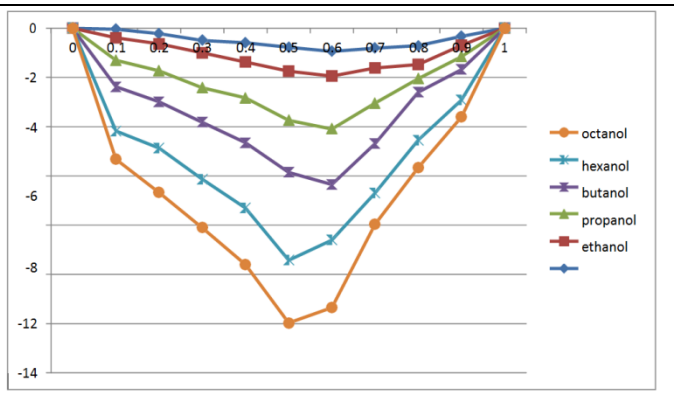

Figure 4 Plots of molar volume versus mole fraction of 1,4-dioxane (x_1) at 303.15 K for binary mixtures of 1,4-dioxane with methanol, ethanol, propanol, butanol, hexanol and octanol at 303.15 K.

The liquid variation in acoustic impedance indicates the absence of specific interaction like complex formation in the binary mixture are shown in Figure- 5.

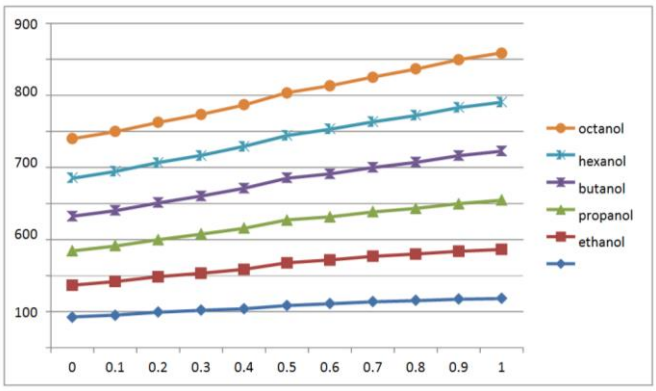

Figure 5 Plots of acoustic impedance versus mole fraction of 1,4-dioxane (x_1) at 303.15 K for binary mixtures of 1,4-dioxane with methanol, ethanol, propanol, butanol, hexanol and octanol at 303.15 K.

It can be seen from mathematical relation for acoustic impedance ($Z = \rho u$) and adiabatic compressibility ($\beta_a = 1 / \rho u^2$) that they must show opposite behaviour and adiabatic compressibility and intermolecular free length ($L_f = K \beta_{ad}^{1/2}$) should exhibit same behaviour which is in agreement with the experimental results [33].

Figure 6 & 7 illustrate the linear behaviour of relaxation time (τ) and relaxation strength (r) with mole fraction at the trend of these parameters is almost same with best results obtained between 0.2 to 0.8 range of mole fraction. As seem from the experimental results ultrasonic velocity increase which results in the decrease of relxation strength (r). The decreases in relaxation time (τ) indicate that the viscous forces has no effect on it.

Table 5 Excess values of viscosity (η^E), molar volume(V_m^E), adiabatic compressibility (β_{ad}^E), internal pressure (p_i^E) and free length (L_f^E) properties for binary liquids mixtures of 1,4-dioxane + methanol, 1,4-dioxane + ethanol, 1,4-dioxane + propanol, 1,4-dioxane + butanol, 1,4-dioxane +hexanol and 1,4-dioxane + octanol at 303.15 K.

0.10006	-0.0013	-0.91399	-0.09167	-0.05836	-0.06274
0.12264	-0.0018	-1.09620	-0.45983	-0.11371	-0.48849
0.29821	-0.0031	-1.42088	0.46806	-0.17610	-0.46664
0.40573	-0.0047	-1.45746	-0.51974	-0.18209	-0.52792
0.50439	-0.0061	-1.99903	-0.37808	-0.19038	-0.35163
0.60251	-0.0084	-2.14871	-0.25484	-0.16522	-0.20317
0.69410	-0.0055	-1.43194	+0.22566	-0.13077	-0.41304
0.79626	-0.0030	-0.56704	-0.01533	-0.09162	-0.07112
0.89926	-0.0015	-0.45833	-0.01109	-0.05613	-0.04737
1.00000	0.0000	0.00000	0.00000	0.00000	0.00000
		1,4-dioxane + 1-butanol			
0.00000	0.0000	0.00000	0.00000	0.00000	0.00000
0.09734	-0.0012	-1.07405	0.11421	-0.07519	-0.17137
0.19759	-0.0025	-1.25808	0.06737	-0.11934	-0.15147
0.30443	-0.0039	-1.39875	0.07017	-0.12050	-0.18268
0.40480	-0.0052	-1.82020	-0.28702	-0.14369	-0.24579
0.49442	-0.0063	-2.11069	-0.34414	-0.11458	-0.32065
0.59768	-0.0077	-2.25372	-0.24258	-0.08173	-0.19885
0.68628	-0.0047	-1.64205	-0.20861	-0.07453	-0.17024
0.79076	-0.0027	-0.55516	-0.18677	-0.04512	-0.17063
0.89091	-0.0017	-0.52992	-0.17039	-0.01983	-0.18852
1.00000	0.0000	0.00000	0.00000	0.00000	0.00000
		1,4-dioxane + 1-hexanol			
0.00000	0.0000	0.00000	0.00000	0.00000	0.00000
0.09108	-0.0001	-1.79251	-0.09343	-0.06751	-0.10397
0.19485	-0.0002	-1.87459	-0.12798	-0.08139	-0.13703
0.29842	-0.0003	-2.31017	-0.01519	-0.09673	-0.01738
0.40439	-0.0003	-2.65638	-0.03983	-0.10058	-0.00921
0.45430	-0.0009	-3.56962	-0.14408	-0.10227	-0.14722
0.60286	-0.0005	-2.26339	-0.14848	-0.08947	-0.15812
0.69974	-0.0002	-2.00314	-0.13132	-0.06636	-0.14214
0.80182	-0.0002	-1.94200	-0.07883	-0.04578	-0.08020
0.88834	-0.0001	-1.23491	-0.09800	-0.02685	-0.12028
1.00000	0.0000	0.00000	0.00000	0.00000	0.00000

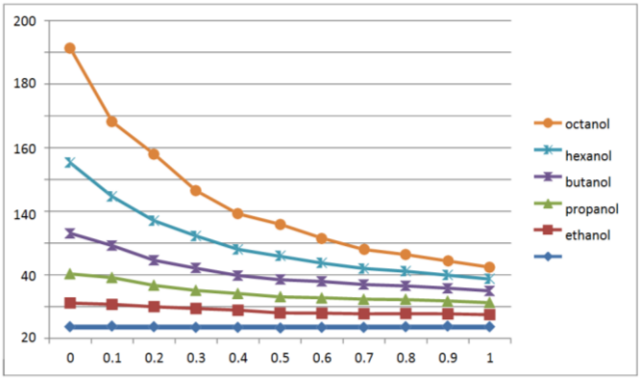

Figure 6 Plots of viscous relaxation time versus mole fraction of 1,4-dioxane (x_1) at 303.15 K for binary mixtures of 1,4-dioxane with methanol, ethanol, propanol, butanol, hexanol and octanol at 303.15 K.

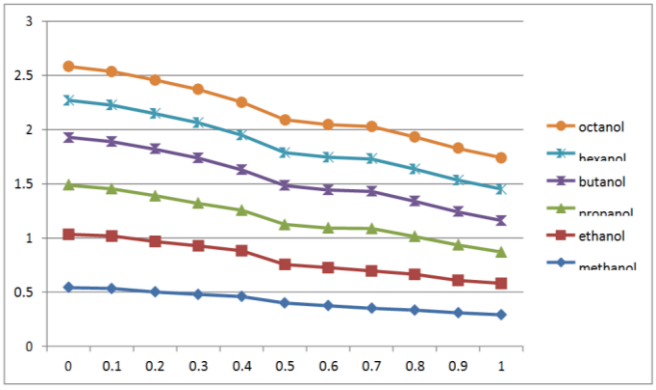

Figure 7 Plots of relaxation strength versus mole fraction of 1,4-dioxane (x_1) at 303.15 K for binary mixtures of 1,4-dioxane with methanol, ethanol, propanol, butanol, hexanol and octanol at 303.15 K.

All the two constant Wada constant and Rao's constant are increasing with increases in mole fraction and displayed in Figure 8 & 9 respectively. It was reported that if the variation in Wada constant and Rao's constant is linear then it shows that there is an absence of complex formation in two mixture and so is found in the present investigation which means that there is no complex formation in the mixture of 1,4-dioxane and 1-alkanols.

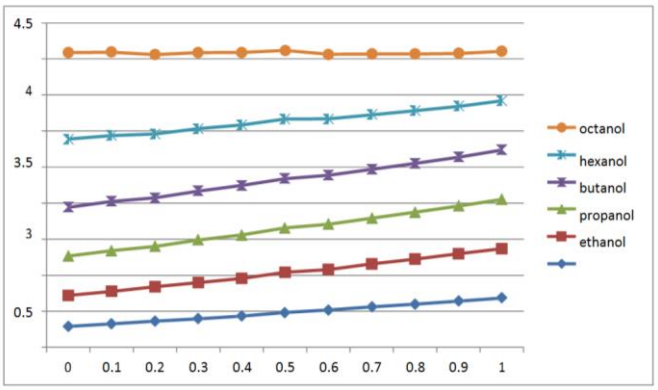

Figure 8 Plots of wada constant versus mole fraction of 1,4-dioxane (x_1) at 303.15 K for binary mixtures of 1,4-dioxane with methanol, ethanol, propanol, butanol, hexanol and octanol at 303.15 K.

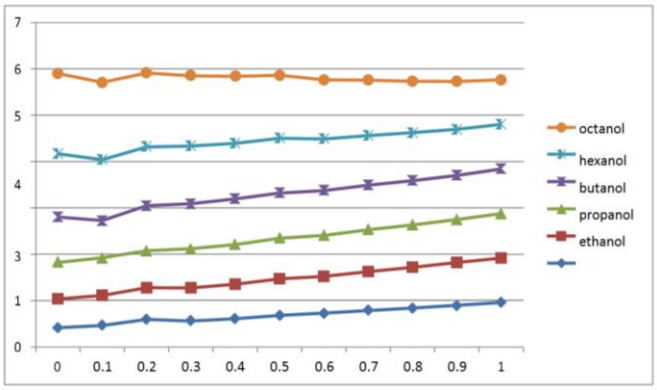

Figure 9 Plots of Rao's constant versus mole fraction of 1,4-dioxane (x_1) at 303.15 K for binary mixtures of 1,4-dioxane with methanol, ethanol, propanol, butanol, hexanol and octanol at 303.15 K.

The internal pressure give information regarding the nature and strength of force existing between the molecules. Due to decrease in internal pressure ofthe liquid mixture as shown in Figure - 4 which shows that the strength of interaction among the molecule decrease gradually with increase in concentration and hence weak interaction between the molecules is formed.

CONCLUSION

The ultrasonic velocity study of binary liquid mixtures of 1,4-Dioxane with 1- alkanols shows the presence of molecular interaction between the molecules of the mixture. The ultrasonic velocity increases with increase in concentration which is due to the decrease in intermolecular free length of the mixture. The density increases with increase in mole fraction. Adiabatic compressibility, intermolecular free length, internal pressure, relaxation time and relaxation strength decreases with increase in concentration. This decrease in acoustical parameters indicates that there is a weak interaction between the molecules of the mixture. Wada constant and Rao's constant show linear variation with increase in mole fraction which indicates the absence of complex formation in the mixture.

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Symbol and Description

ρ, Density of liquid
M, Molecular Weight
K, Jacobson's constant
u, Ultrasonic velocity
u^E, Excess ultrasonic velocity
η, Viscosity
η^E, Excess viscosity
pi, Internal pressure
pi^E, Excess internal pressure
X_1, Mole fraction of 1,4-Dioxane
Z, Acoustic impedance impedance
Z^E, Excess Acoustic impedance impedance
V_m, Molar Volume
V_m^E, Excess Molar Volume
β_{ad}, Adiabatic compressibility
β_{ad}^E, Excess adiabatic compressibility
R, Rao's constant
W, Wada constant
L_f, Intermolecular free length
τ, relaxation time
r, Relaxation strength

Declarations conflict of interest

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

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Data availability statement

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

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