INVESTIGATION OF THERMO-ACOUSTIC EXCESS PARAMETERS OF BINARY LIQUID MIXTURE USING ULTRASONIC NON DESTRUCTIVE TECHNIQUE

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Abstract: The acoustic parameters for binary liquid mixture namely n-hexane - tetrahydrofuran has been determined at four different temperatures. The excess acoustical parameters such as excess ultrasonic velocity (u^E) , excess viscosity (η^E) , excess adiabatic compressibility (β_a^E) , excess intermolecular free length (L_f^E) , excess free volume (V_f^E) , excess acoustic impedance (Z^E) and excess internal pressure (Π_i) has been computed using the measured values of ultrasonic velocity, viscosity and density. The extent of interaction existing between component molecules has been found out in n-hexane and tetrahydrofuran system. The interaction parameters values have been out to be negative suggesting the presence of dipole - induced dipole interaction with increase in temperature.

Keywords: Binary mixture, ultrasonic velocity, adiabatic compressibility, intermolecular free length, and internal pressure.

Introduction:

The importance of liquid mixtures rather than single component liquid system ^[1, 2], has gained much importance during the last two decades in assessing the nature of molecular interaction and investigation of liquid mixtures consisting of polar and non-polar components is of considerable importance in understanding inter molecular interaction between the component molecules and they find applications in several industrial and technological process ^[3-5]. Such studies as a function of concentration are useful in gaining insight into the stracture and bonding of associated molecular components and other molecular processes. The measurement of ultrasonic speed and parameters derived from it has been used in understanding the nature of intermolecular interaction in liquid mixtures ^[6-11]. Though a number of investigations were carried out in mixtures having n-hexane as one of the components ^[12-13], binary system with tetrahydrofuran as one of the component at constant frequency are scarcely reported. As tetrahydrofuran is a secondary ether it is used as a solvent for resins, waxes, oils, dyes and surface coatings.

Experimental:

The chemicals n-hexane and tetrahydrofuran were used of analytical grade and obtain from Merck chemicals private Ltd. (Purity 99.5%). Special attention was given to avoid the vaporization of solution. Comparing their density with literature values checked the purity of chemicals. The mixtures of various concentrations in mole fraction were prepared.

(i) Velocity Measurement:-

Measurements

The velocity of ultrasonic wave in the binary mixture have been measured using ultrasonic interferometer with an high degree of accuracy operating at 5 MHz frequencies (Model F-81) supplied by M/s Mittal Enterprises, New Delhi. The measuring cell of interferometer is a specially designed double walled vessel with provision for temperature constancy. An electronically operated digital constant temperature bath supplied by M/s Mittal Enterprises, New Delhi, operating in the temperature range 5°c to 99.9°c with an accuracy of ± 0.1 °c has been used to circulate water through the outer jacket of the double walled measuring cell containing the experimental liquid.

(ii) Density Measurement:-

The densities of the mixture were measured using a 25ml specific gravity bottle. The specific gravity bottle with the experimental mixture was immersed in a temperature controlled water bath. The density was measured using the formula

$$\rho_2 = \left(\frac{W_2}{W_1}\right)\rho_1$$

Where, w_1 = weight of distilled water, w_2 = Weight of experimental liquid, ρ_1 = Density of water, ρ_2 = Density of experimental liquid

(iii) Viscosity measurement:-

The viscosities of the ternary mixture were measured using an Oswald's viscometer calibrated with double distilled water. The Oswald's viscometer with the experimental mixture was immersed in a temperature controlled water bath. The time of flow was measured using a digital racer stop watch with an accuracy of 0.1 sec. The viscosity was determined using the relation,

$$\eta_2 = \eta_1 \left(\frac{t_2}{t_1}\right) \left(\frac{\rho_2}{\rho_1}\right)$$

Where, η_1 = Viscosity of water, η_2 = Viscosity of mixture, ρ_1 = Density of water, ρ_2 = Density of mixture, t_1 = Time of flow of water, t_2 = Time of flow of mixture.

Theory:

The adiabatic compressibility is the fractional decrease of volume per unit increase of pressure, when no heat flows in or out. It is calculated from sound velocity 'u' and the density (ρ) of the medium using the equation of Newton Laplace as,

$$\beta = \frac{1}{u^2 \rho} \tag{1}$$

Intermolecular free length (L_f) has been determined by the equation.

$$L_f = K_T \sqrt{\beta} \tag{2}$$

Where K_T is a Jacobsen's constant.

The free volume $L_{\rm f}$ in terms of ultrasonic, velocity (u) and the viscosity (η) of a liquid is

$$V_f = \left(\frac{M_{eff} u}{k\eta}\right)^{3/2} \tag{3}$$

Where M_{eff} is the effective molecular weight

$$M_{eff} = \sum m_i x_i \tag{4}$$

In which $m_i \& x_i$ are the molecular weights and mole fraction of individual constituents respectively and K is a temperature dependent constant equal to 4.28×10^9 for all liquids in MKS system.

Relaxation time is the time taken for the excitation energy to appear as translational energy and it depends on temperature and on impurities. The dispersion of the ultrasonic velocity in binary mixture reveals information about the Characteristic time of the relaxation process that causes dispersion. The relaxation time (τ) can be calculated from the relation.

$$\tau = \frac{4}{3} \left(\beta \,\eta\right) \tag{5}$$

Where ' β ' is the adiabatic compressibility and ' η ' is the viscosity of the mixture.

An excess value of ultrasonic related parameters has been calculated by using the relations.

$$\beta^E = \beta_{expt} - \sum x_i \beta_i \tag{6}$$

$$V^E = V_{expt} - \sum x_i V_i \tag{7}$$

Where β_a^E and V_f^E are the excess values of adiabatic compressibility, and free volume respectively.

Result and Discussion:

. Ultrasonic velocity (*u*), density (ρ), adiabatic compressibility (β_a) and other related excess thermodynamic parameters like excess adiabatic compressibility (β_a^E), excess acoustic impedance (Z^E), excess free volume (V_f^E) and excess free length (L_f^E) are evaluated for binary mixture tetrahydrofuran in n-hexane over whole concentration at 301.15 K, 305.15K, 309.15K and 313.15K and presented in Table- I, Table- II, Table- III and Table- IV respectively.

Mole	Excess	Excess	Excess	Excess	Excess	Excess	Excess	Excess
Fraction of	Adiabatic	Free	Free	Acoustic	Viscosity	Velocity	Relaxatio	Internal
Triethylamin	Compressib	Length	Volume	Impedanc	$\eta^{E} \times 10^{-3}$	u^E	n Time	Pressure
e (<i>x</i>)	ility	$L_{f}^{E} \times 10^{-11}$	$V_{f}^{E} \times 10^{-3}$	e	(Nsm ⁻²)	(<i>m/s</i>)	$T \times 10^{-13}$	$\Pi_i \times 10^5$
	$\beta^{E} \times 10^{-10}$	(m)	(M ³ mole	Z^{E} (Kg ⁻			(s)	(Pas)
	(m^2n^{-1})		1)	2_{ms}^{-1})				
0.0	0.0000	0.0000	0.0000	0.0000	0.0000	00.0000	0.0000	0.0000
0.1	-1.40181	-0.3075	0.7900	-0.0032	0.0000	34.7100	-0.5315	-0.1614
0.2	-2.69162	-0.6380	1.5200	0.0124	-0.0100	73.7500	-1.0769	-0.2582
0.3	-2.76943	-0.6525	2.0400	0.0027	-0.0200	78.1300	-1.2233	-0.3584
0.4	-2.38424	-0.5373	1.4900	-0.0129	-0.0200	57.3100	-0.9763	-0.3589
0.5	-2.40245	-0.5628	1.4100	-0.0077	-0.0200	64.5100	-0.9725	-0.3746
0.6	-1.91116	-0.4308	1.0800	-0.0201	-0.0200	47.0000	-0.7495	-0.3667
0.7	-1.56367	-0.3572	0.7600	-0.0196	-0.0100	42.6300	-0.5768	-0.3374
0.8	-1.13478	-0.2711	0.5900	-0.0137	-0.0100	32.8100	-0.4395	-0.2683
0.9	-0.53709	-0.1170	0.3900	-0.0190	-0.0100	15.2200	-0.2311	-0.1888
1.0	0.0000	0.0000	0.0000	0.0000	0.0000	00.0000	0.0000	0.0000

Table 1: Excess Values of ultrasonic Velocity, Viscosity, Adiabatic Compressibility, Free Length, Free Volume, Acoustic Impedance at 301.15K.

Table 2: Excess Values of Ultrasonic Velocity, Viscosity, Adiabatic Compressibility, Free Length, Free Volume, Acoustic Impedance at 305.15K.

Molo	Excoss	Excoss	Excoss	Excoss	Excoss	Excoss	Excoss	Excoss
Fraction of	Adiabatia	Excess	Excess	Accustic	Viscosity	Valaaitu	Delevatio	Intorn
Traction of	Adiabatic	Fiee	Free	Acoustic	VISCOSILY		Relaxatio	Intern
Iriethylamin	Compressib	Length	Volume	Impedanc	$\eta^{E} \times 10^{-5}$	<i>u</i> ⁻	n I ime	al
e (<i>x</i>)	ility	$L_{f}^{L} \times 10^{-11}$	$V_f^L \times 10^{-5}$	e	(Nsm ⁻²)	(<i>m/s</i>)	$T \times 10^{-13}$	Pressu
	$\beta^{E} \times 10^{-10}$	(m)	(M [°] mole	Z^{E} (Kg ⁻			(s)	re
	(m^2n^{-1})		1)	2 -1				$\Pi_i \times 10^3$
				2ms^{-1}				(Pas)
0.0								
0.0	0.0000	0.0000	0.0000	0.0000	0.0000	00.0000	0.0000	0.0000
0.1								-
0.1	-0.8360	-0.1626	0.5264	-0.0114	-0.0046	17.5530	-0.2650	0.1382
								-
0.2	-2.3509	-0.5456	1.7333	0.0073	-0.0172	62.0960	-0.9294	0.2555
								_
0.3	-2 1934	-0 4884	1 8451	-0.0083	-0.0188	55 0690	-0.9043	0 3321
	2.1751	0.1001	1.0151	0.0005	0.0100	55.0070	0.2013	0.3521
0.4	1 0768	0 4226	1 /156	0.0170	0.0101	42 2020	0.7506	0 3300
	-1.9708	-0.4220	1.4150	-0.0179	-0.0191	42.2020	-0.7390	0.5599
0.5	1 55 47	0 2011	1 0022	0.0242	0.0210	26 1450	0 5 (9 9	-
	-1.5547	-0.3011	1.0922	-0.0342	-0.0210	26.1450	-0.3088	0.3009
0.6	1 201 5	0.000.0	0.00.00	0.0000	0.01.60	1.5.5.400	0.0054	-
	-1.2815	-0.2336	0.6050	-0.0380	-0.0160	15.7480	-0.3854	0.3257
0.7								-
	-0.9779	-0.1653	0.6211	-0.0401	-0.0190	9.6010	-0.3283	0.3198
0.8								-
0.8	-0.9067	-0.1895	0.6318	-0.0203	-0.0155	19.7340	-0.3542	0.2672
0.0								-
0.9	-0.2427	-0.0154	0.1174	-0.0001	-0.0080	4.9130	-0.0430	0.1575
1.0								
1.0	0.0000	0.0000	0.0000	0.0000	0.0000	00.0000	0.0000	0.0000

Mole	Excess	Excess	Excess	Excess	Excess	Excess	Excess	Excess
Fraction of	Adiabatic	Free	Free	Acoustic	Viscosity	Velocity	Relaxatio	Internal
Triethylamin	Compressibil	Length	Volume	Impedanc	$\eta^E \times 10^{-3}$	u^E	n Time	Pressur
e (<i>x</i>)	ity	$L_{f}^{E} \times 10^{-11}$	$V_{f}^{E} \times 10^{-3}$	e	(Nsm ⁻²)	(<i>m/s</i>)	$T \times 10^{-13}$	e
	$\beta^{E} \times 10^{-10}$	(m)	(M ³ mole	Z^{E} (Kg ⁻			(s)	$\Pi_i \times 10^3$
	(m^2n^{-1})		1)	2_{ms}^{-1})				(Pas)
0.0	0.0000	0.0000	0.0000	0.0000	0.0000	00.0000	0.0000	0.0000
0.1	-1.2061	-0.2482	0.4541	-0.0034	-0.0015	23.7760	-0.3277	-0.1249
0.2	-2.8526	-0.6588	2.2686	0.0165	-0.0152	70.1720	-1.1693	-0.2710
0.3	-2.6938	-0.6064	1.7937	0.0038	-0.0144	64.8380	-1.0040	-0.3231
0.4	-2.5684	-0.5741	1.6210	-0.0011	-0.0168	59.5040	-0.9505	-0.3490
0.5	-2.2632	-0.4980	1.4896	-0.0088	-0.0200	51.6700	-0.8571	-0.3753
0.6	-1.8772	-0.4049	1.2149	-0.0160	-0.0197	43.3360	-0.7025	-0.3816
0.7	-1.3366	-0.2688	0.8056	-0.0227	-0.0186	25.0020	-0.4867	-0.3309
0.8	-0.8736	-0.1647	0.5402	-0.0213	-0.0164	12.0880	-0.3265	-0.2535
0.9	-0.4279	-0.0745	0.1951	-0.0150	-0.0081	4.3340	-0.1358	-0.1448
1.0	0.0000	0.0000	0.0000	0.0000	0.0000	00.0000	0.0000	0.0000

Table 3: Excess Values of Ultrasonic Velocity, Viscosity, Adiabatic Compressibility, Free Length, Free Volume, Acoustic Impedance at 309.15K.

Table 4: Excess Values of Ultrasonic Velocity, Viscosity, Adiabatic Compressibility, Free Length, Free Volume, Acoustic Impedance at 313.15K.

Mole	Excess	Excess	Excess	Excess	Excess	Excess	Excess	Excess
Fraction of	Adiabatic	Free	Free	Acoustic	Viscosit	Velocity	Relaxatio	Interna
Triethylamin	Compressi	Length	Volume	Impedanc	У	u^E	n Time	1
e (<i>x</i>)	bility	$L_{f}^{E} \times 10^{-11}$	$V_{f}^{E} \times 10^{-3}$	e	$\eta^E \times 10^-$	(<i>m/s</i>)	$T \times 10^{-13}$	Pressur
	$\beta^{E} \times 10^{-10}$	(m)	(M ³ mole	Z^{E} (Kg ⁻	$^{3}(Nsm^{-2})$		(s)	е
	(m^2n^{-1})		-1)	21				$\Pi_i \times 10^5$
				-ms +)				(Pas)
0.0								
	0.0000	0.0000	0.0000	0.0000	0.0000	00.0000	0.0000	0.0000
0.1								-
	-1.8162	-0.3602	0.5414	-0.0006	0.0002	30.3930	-0.4690	0.1404
0.2								-
	-4.4564	-1.0028	2.3879	0.0329	-0.0072	96.3260	-1.5765	0.2889
0.3								-
	-4.5273	-1.0259	2.7165	0.0292	-0.0139	102.4690	-1.6706	0.3732
0.4								-
	-4.3957	-1.0100	2.6453	0.0284	-0.0166	103.8520	-1.6402	0.4137
0.5								-
	-3.8749	-0.8917	2.5258	0.0199	-0.0213	95.1150	-1.5054	0.4512
0.6								-
	-3.2658	-0.7547	2.0925	0.0134	-0.0209	83.3780	-1.2780	0.4457
0.7								-
	-2.5497	-0.5911	1.5373	0.0090	-0.0183	65.6410	-0.9991	0.3880
0.8								-
	-1.6450	-0.3709	1.0665	-0.0003	-0.0177	39.9040	-0.6809	0.3093
0.9								-
	-0.6950	-0.1389	0.4062	-0.0098	-0.0099	13.1670	-0.2651	0.1803

INVESTIGATION OF THERMO-ACOUSTIC EXCESS PARAMETERS OF BINARY LIQUID

1.0								
	0.0000	0.0000	0.0000	0.0000	0.0000	00.000	0.0000	0.0000

In the binary liquid systems under investigation, the variation of ultrasonic velocity (*u*), adiabatic compressibility (β_a), free volume (V_f), free length (L_f) and acoustic impedance (*Z*) are calculated. In Fig.-I, Fig.-II, Fig.-II, Fig.-IV, Fig.-V, Fig-VI, Fig-VII and Fig-VIII respectively represents the variation in excess ultrasonic velocity(u^E), excess viscosity (η^E), excess adiabatic compressibility (β_a^E), excess free length (L_f^E), excess free volume (V_f^E), excess acoustic impedance(Z^E), excess relaxation time (τ^E) and excess internal pressure (Π_i^E). These variation indicate the existence of molecular interaction between solvent and solvent. The excess adiabatic compressibility (β_a^E) is negative over whole concentration range and it becomes minimum at a concentration (0.2) of tetrahydrofuran in n-hexane at observed temperatures. This indicates that the intermolecular interaction at this optimum at this concentration and it may leads to formation weak hydrogen bonded complex in binary liquid mixture. The same effects are also observed in excess volume (V_f^E), excess free length and excess acoustic impedance. The excess values of internal pressure in table (1,2,3,4) at four various temperature and the behaviour of negative excess values shows that the strength of interaction decreases gradually as concentration of tetrahydrofuran increases in the mixture. The negative values of π_i^E indicate that only dispersion and dipolar forces are operating complete absence of specific interaction.



Fig.I: Plots of excess Ultrasonic Velocity Vs Mole Fraction of Tetrahydrofuran(x) for n-hexane -Tetrahydrofuran System at various Temperatures.

Fig.II: Plots of excess Viscosity Vs Mole Fraction of Tetrahydrofuran (x) for n-hexane -Tetrahydrofuran System at various Temperatures.



Fig.III: Plots of excess adiabatic compressibility Vs Mole Fraction of Tetrahydrofuran (x) for n-hexane - Tetrahydrofuran System at various Temperatures.



Fig.IV: Plots of excess free length Vs Mole Fraction of Tetrahydrofuran (x) for n-hexane - Tetrahydrofuran System at various Temperatures.



Fig.V: Plots of excess free volume Vs Mole Fraction of Tetrahydrofuran (*x*) for n-hexane - Tetrahydrofuran System at various Temperatures.



Fig.VI: Plots of excess acoustic impedance Vs Mole Fraction of Tetrahydrofuran (*x*) for n-hexane - Tetrahydrofuran System at various Temperatures.



Fig.VII: Plots of excess relaxation time Vs Mole Fraction of Tetrahydrofuran (x) for n-hexane – Tetrahydrofuran System at various Temperatures.



Fig.VIII: Plots of excess internal pressure Vs Mole Fraction of Tetrahydrofuran (x) for n-hexane -Tetrahydrofuran System at various Temperatures.

Conclusion:

Experimental data of the ultrasonic velocity, density and viscosity of n-hexane and tetrahydrofuran mixture have been measured over the entire composition range 301.15K, 305.15K, 309.15K and 313.15K. It has been observed that positive deviations of excess velocity, excess free volume, where as negative deviations were observed for excess free length at 301.15K, 305.15K, 309.15K and 313.15K. The present investigation shows that weak molecular interactions exist in the mixtures which may be due to the dominance of dispersion forces and dipolar interaction between the unlike molecules.

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