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RESEARCH ARTICLE

SOLUTE – SOLVENT AND SOLUTE – SOLUTE INTERACTIONS OF TETRABUTYLAMMONIUM IODIDEE IN DIMETHYLSULPHOXIDE – ACETONE SYSTEMS AT DIFFERENT TEMPERATURES

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ABSTRACT

Ultrasonic velocity, viscosity and density studies on solution of tetrabutylammonium iodide (Bu$_4$NI) in Dimethylsulphoxide (DMSO) and Dimethylsulphoxide + Acetone solvent mixtures containing 50,60,70,80,90 and 100 mol % of DMSO at 298 and 308K have been reported. From the velocity, viscosity and density data values, various parameters namely, the adiabatic compressibility (β), apparent molar compressibility (ϕ_k), apparent molar volume (ϕ_v), limiting apparent molar compressibility (ϕ_k^o), limiting apparent molar volume (ϕ_v^o), free volume (V_f), internal pressure (π_i), relaxation time (τ) and viscosity B-coefficient have been calculated. All these parameters have been discussed separately to throw light on the solute-solvent and solvent-solvent interactions.

INTRODUCTION

Ultrasonic studies have gained importance in recent years in the fields of medicine, engearing, agriculture and industry (**Johri and Mishra, 1984 and Hautman *et al.*, 1984**). In chemistry, such studies are used to investigate various chemical processes that play a major role in synthetic work (**Johri and Mishra, 1984 and Hautman *et al.*, 1984**). In solution chemistry, electrolytic solutions are, however, investigated in the light of ultrasonic studies with special reference to ion-solvent interaction (Marcus and John Wiley, 1985). Studies of densities, viscosities and ultrasonic speeds of electrolytic solutions are of great use in characterizing the structure and properties of solutions (**Chauhan *et al.*, 2013; Syal *et al.*, 1995; Syal *et al.*, 1998; Chauhan *et al.*, 2007; Mehra and Gaur, 2008 and Hemalatha *et al.*, 2013**). Various types of interactions exist between the solutes in the solutions, and these solute-solute and solute-solvent interactions are of current interest in all branches of chemistry. These interactions provide a better understanding of the nature of the solute and solvent i.e., weather the solute modifiesor distorts the structure of the solvent. Recently, ion-ion and io-solvent interactions for the tetraalkylammonium salts have been reported (**Hemalatha *et al.*, 2013; Nikam *et al.*, 2004 and Nikam *et al.*, 2002**). The present investigation reports the ultrasonic velocity, density and viscosity studies of tetrabutylammoniumIodide (Bu$_4$NI) in Dimethylsulphoxide (DMSO) and Dimethylsulphoxide (DMSO) – Acetone (Ac) mixtures at 298 and 308 K. From these experimental data, the number of thermodynamic parameters viz. the adiabatic compressibility (β), apparent molar compressibility(ϕ_k), apparent molar volume (ϕ_v), limiting apparent molar compressibility (ϕ_k^o), limiting apparent molar volume (ϕ_v^o), free volume (V_f), internal pressure (π_i), relaxation time (τ) and viscosity B-coefficient have been calculated. These parameters have been used to interpret various molecular interactions occurring in solutions at different temperatures.

Experimental: Dimethyl sulphoxide (extra pure, Sisco Research Laboratories Pvt. Ltd., Mumbai) was kept over Cao and distilled (**Syal *et al.*, 1988 and Syal *et al.*, 2005**). Acetone (Ac) of 99.5% purity (BDH, AR) was dried over 4Å molecular sieves and distilled (**Syal *et al.*, 2005 and Reddick *et al.*, 1986**). Tetraprobutylammonium iodide (Bu$_4$NI from Fluka, was dried and used as described earlier (**Syal *et al.*, 2000; Patial, 2015; Patial, 2014 and Patial *et al.*, 2002**). Viscosity measurements were carried out as described elsewhere (**Patial, 2014; Patial *et al.*, 2002; Syal *et al.*, 2002 and Syal *et al.*, 1977). Viscosity values were found to be good agreement with those reported in literature (Syal *et al.*, 2000; Patial, 2015; Patial, 2014; Patial *et al.*, 2002; Syal *et al.*, 1999; Syal *et al.*, 1977**). The densities of pure solvent, solvent system and various electrolytic solutions were measurement with the help

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of a sealable type of pycnometer (supplied by M/s. Harsh and Co., Ambala Cantt.) of 20 cm^3 capacity). The value of ultrasonic velocity for the conductivity water was found to be 1490 m/s at 298.15 K at 1 MHz, which is agreed well with literature value (**Syal *et al.*, 1995**). Ultrasonic velocity were measured using interferometer (Model-81, supplied by Mittal Enterprises, New-Delhi) operating at a frequency of 1 MHz, which is a direct and simple device for measuring ultrasonic velocity in liquids. The viscosities and densities of the above electrolyte in DMSO and DMSO +Ac solvent systems were measured at 298 and 308 K. The overall accuracy of the viscosity and density measurements in this study was estimated to be ± 0.2% and ± 0.1% respectively. Different parameters such as the adiabatic compressibility (β), apparent molar compressibility (ϕ_k), apparent molar volume (ϕ_v), limiting apparent molar compressibility (ϕ_k^o), limiting apparent molar volume (ϕ_v^o), free volume (V_f), internal pressure (π_i), relaxation time (τ) viscosity B-coefficient have been calculated at different temperatures, with the help of ultrasonic velocity (u), density (ρ) and viscosity (η) values using the following relations (**Hemalatha *et al.*, 2013; Syal *et al.*, 2005; Kumar *et al.*, 2014 and Eyring and Kincaid, 1928**):

Adiabatic compressibility (β)

The adiabatic compressibility values for various compositions of the binary solvent mixtures have been calculated from the measured ultrasonic velocities (u) and densities (ρ):

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

Apparent molar compressibility (ϕ_k)

The apparent molar compressibility (ϕ_k) have been calculated using the relation:

$$\phi_k = \frac{1000}{c\rho_o}(\rho_o\beta - \beta_o\rho) + \frac{\beta_o M}{\rho_o}$$

Where β,ρ and β_o, ρ_o are the adiabatic compressibility and density of the solution and solvent, respectively. M is the molar mass of the solute, and c is the molar concentration.

Liming apparent molar compressibility (ϕ_k^o)

The values of the limiting apparent molar compressibility ϕ_k^o of solute solution are obtained by the use of least square treatment to the plots of the use of ϕ_k, apparent molar compressibilty of solution versus $C^{1/2}$ in accordance with Masson's empirical (**Masson, 1929**):

$$\phi_k = \phi_k^o + S_k^* + C^{1/2}$$

Where ϕ_k^o is the limiting apparent molar compressibility at infinite dilution and S_k is a constant.

Limiting apparent molar volume (ϕ_v^o)

The values of the limiting apparent molar volume ϕ_v^o of solute solution are obtained by the use of least square treatment to the plots of the use of ϕ_v, apparent molar volume of solution versus $C^{1/2}$ in accordance with Masson's empirical (**Masson, 1929**):

$$\phi_k = \phi_k^o + S_k^* + C^{1/2}$$

Where ϕ_v^o is the limiting apparent molar compressibility at infinite dilution and S_v is a constant.

The apparent molar volume (ϕ_V)

The apparent molar volume, ϕ_v is calculated from the density data by using flowing expression:

$$\phi_V = 1000\frac{(\rho_o - \rho)}{c\rho_o} + \frac{M}{\rho_o}$$

Where ρ_o and ρ are densities of solvent and solution, respectively; M is the molar mass of the solute, and c is the molar concentration.

Relaxation Time (τ)

Relaxation can be calculated from viscosity coefficient (η), density and ultrasonic velocity of binary mixtures and given by

$$\tau = \frac{4\eta}{3\rho u^2}$$

Free Volume (v_f)

The free volume of binary mixture is given by

$$V_f = \left[\frac{M_{eff}u}{K\eta}\right]^{3/2}$$

where K is time independent constant whose value is 4.28 x 10^9 in MKS system and M_{eff} effective molecular weight of the liquid is given by

$$M_{eff} = X_1M_1 + X_2M_2$$

Where $X_1$and X_2 are the mole fraction of first and second components and $M_1$and M_2 are the molecular weights of first and second components respectively.

Internal Pressure (π_i)

Internal pressure is given by

$$\pi_i = \frac{bRT[K'\eta]}{M_{eff}^{\frac{7}{6}}}\rho^{\frac{2}{3}}$$

Where, b is the cubic packing factor which is assumed to be 2 in liquid systems.
K = 4.28 x 10^9 and is independent to the nature of liquid.
R is gas constant.
η is the viscosity and ρ is the density of solution.

RESULTS AND DISCUSSION

The experimental values of density, viscosity and ultrasonic velocity of Bu_4NI, have been measured in DMSO and DMSO

+ Ac mixtures containing 100,90,80,70,60 and 50 mol% of DMSO in the concentration range (0.1-0.5) mol dm^{-3} at 298 and 308 K. The calculated values of adiabatic compressibility, apparent molar volume and apparent molar adiabatic compressibility are reported in Table 1. The value of limiting apparent molar compressibility, limiting molar volume, and the constants S_k and S_v for tetrabutylammonium iodide (Bu_4NI) are presented in Table 2. The values of A- and B- coefficients are given in Table 3 and the calculated values of free volume, internal pressure and relaxation time in Table 4. It is clear from the Table 1 that the adiabatic compressibility (β) values of solution decrease with increase in the concentration of electrolyte at all compositions. β – values for a particular solute concentration increase with the increase of a particular solute concentration increase with the increase of acetone content to DMSO + Ac mixture. With increase of temperature, β – values of solution increase, indicates temperature dependence of β and increase of solute-solvent interactions.

Table 1. Adiabatic compressibility (β), Apparent molar compressibility (φ_k) and apparent molar volume (φ_v) of Bu_4NI in DMSO- Ac mixtures

C mol dm^{-3}	βx10^{-10} (Kg^{-1}m s^{-1})		ϕ_k x 10^4 (atm^{-1}m^{3}mol^{-1})		ϕ_v (m^{3}mol^{-1})	
	298K	308K	298K	308K	298K	308K
			Pure DMSO			
0.00	4.20	4.40	--	--	--	--
0.10	4.18	4.39	113.95	126.51	319.95	321.01
0.20	4.15	4.37	109.62	124.19	318.48	319.91
0.30	4.13	4.35	106.86	122.87	316.35	319.10
0.40	4.09	4.32	102.91	118.75	314.68	317.88
0.50	4.06	4.30	101.38	117.37	313.57	317.45
			90%DMSO			
0.00	4.48	4.75	--	--	--	--
0.10	4.44	4.71	106.00	116.83	317.02	318.79
0.20	4.40	4.67	102.76	112.43	314.64	317.76
0.30	4.36	4.63	99.96	110.10	312.51	316.42
0.40	4.31	4.59	95.03	108.32	310.90	315.74
0.50	4.27	4.54	93.62	103.66	309.10	315.15
			80%DMSO			
0.00	4.83	5.12	--	--	--	--
0.10	4.77	5.07	98.16	110.03	312.84	316.52
0.20	4.72	5.01	93.84	104.58	309.98	314.63
0.30	4.66	4.95	91.21	101.09	308.40	313.77
0.40	4.60	4.90	86.86	98.17	306.15	312.68
0.50	4.55	4.84	84.46	96.70	305.27	311.94
			70%DMSO			
0.00	5.20	5.53	--	--	--	--
0.10	5.14	5.46	91.95	100.18	308.73	313.44
0.20	5.07	5.39	87.93	95.71	306.45	312.23
0.30	5.00	5.31	84.71	91.66	304.16	311.42
0.40	4.93	5.24	80.51	89.37	302.41	310.49
0.50	4.86	5.17	78.41	86.11	300.24	309.53
			60%DMSO			
0.00	5.63	6.00	--	--	--	---
0.10	5.54	5.91	85.70	90.89	305.57	310.42
0.20	5.46	5.82	79.73	86.62	302.91	308.88
0.30	5.38	5.73	77.22	82.21	301.01	307.78
0.40	5.30	5.64	74.69	79.21	299.05	307.44
0.50	5.22	5.56	72.18	77.93	296.39	307.22
			50%DMSO			
0.00	6.09	6.54	--	--	--	--
0.10	6.00	6.43	78.29	82.33	302.76	306.48
0.20	5.90	6.32	73.32	75.41	300.91	305.21
0.30	5.80	6.21	69.76	71.15	299.12	304.04
0.40	5.71	6.10	68.33	69.41	296.48	303.25
0.50	5.63	6.00	65.84	67.35	293.50	303.18

The decrease of β with increase of solute concentration can be attributed due to the electrostatic effect of solute on the surrounding solvent molecules, which results in increase of internal pressure and thus solution becomes harder to compress. The decrease of β with increase of concentration of solute in a particular system is indicative of presence of solute-solvent interactions. Similar observations were made by Syal *et al.* (2005) and Kumar *et al.* (2008). The increase in β with increase of Ac content in DMSO in DMSO + Ac mixture may be interpreted to the presence of dipole-dipole interaction/ association between DMSO and Ac molecules and presence of solute-solvent interaction between electrolyte and solvent system.

The apparent molar adiabatic compressibility (ϕ_k) values for Bu_4NI are positive at all the compositions and decrease with increase of Ac content to DMSO + Ac mixtures. These values show a general decrease with the increase of concentration for a particular composition showing the presence of interactions. From Table 1, it is evident that ϕ_k values increase with increase of temperature for Bu_4NI in all the composition. A similar trend shown by **Syal *et al.* (2005)** in EMK+DMF solvent systems. The limiting apparent molar compressibility ϕ_k^o and S_k for each of electrolytic solutions have computed by least-squares method. From Table 2, the observed ϕ_k^o values are large and positive for Bu_4NI, which are found to be similar as reported earlier (**Syal *et al.*, 2000**). ϕ_k^o values decrease with decrease in DMSO content in DMSO + Ac mixtures. Large positive values of ϕ_k^o are indicative of solute-solvent and intermolecular interactions. These ϕ_k^o values show little temperature dependence showing slight increase in the values with increased in temperature. The corresponding S_k values which indicative the solute-solute interactions (**Muhuri *et al.*, 1996**) decrease with an increase in temperature.

Table 2. Limiting apparent molar compressibility (ϕ_k^o), Limiting apparent molar volume (ϕ_v^o) and constant S_k and S_v of Bu_4NI in DMSO- Ac mixtures

X_{DMSO}	ϕ_k^o 10^4 (m^{3}atm^{-1}mol^{-1})		S_k 10^4		ϕ_v^o (m^{3}mol^{-1})		S_v	
	298K	308K	298K	308K	298K	308K	298K	308K
1.00	124.11	135.08	-33.01	-24.11	323.61	325.19	-19.69	-23.16
0.90	116.84	127.11	-33.21	-29.14	321.52	323.88	-18.12	-2163
0.80	109.10	119.76	-.35.08	-34.22	318.94	320.04	-17.69	-18.56
0.70	103.00	111.23	-35.14	-35.10	315.82	316.62	-17.59	-17.81
0.60	95.08	102.32	-33.22	-35.22	313.02	314.77	-16.71	-16.89
0.50	86.92	93.14	-22.14	37.88	309.01	310.68	-16.12	-16.16

The apparent molar volume (ϕ_v) behaves in a similar fashion to that of the apparent molar adiabatic compressibility in the salt solution. ϕ_v^o is regarded as a measure of solute –solvent interaction. It is evident from the Table 2 that ϕ_v^o values are positive for Bu_4NI in DMSO + AC mixtures, suggesting the presence of strong solute-solvent interactions. Small decrease in ϕ_v^o as the amount of Ac in the solution increases indicating the decreasing trend of solute-solvent interactions. The values of S_v^o are large and indicative the presence of weak solute-solute interaction in the solution. Moreover, the values of S_v^o become less negative with increase in Ac content in the system, suggesting increased solute-solute interaction in the present solvent system. In fact, negative S_v^o values are often obtain in solvents high dielectric constant (**Subha *et al.*, 1986**). This is attributed to fact that in solvents of high dielectric constant, like DMSO + Ac system, the salts remain completely ionized, even at fairly high concentrations. As a result, appreciable interionic penetration is likely to occur, giving rise to negative slope for ϕ_v versus C$^{1/2}$ plots (**Parmar *et al.*, 1989**).

Table 3. Values of A ($dm^{2/3}$ $mol^{-1/2}$) and B (dm^3mol^{-1}) parameters of Jones-Dole equation for Bu_4NI in DMSO- Acmixtures

X_{DMSO}	A x10^2		B	
	298K	308K	298K	308K
1.00	0.08	1.04	0.94	0.85
0.90	0.11	2.03	0.94	0.86
0.80	0.94	3.16	0.95	0.86
0.70	2.32	4.27	0.95	0.86
0.60	2.84	5.28	0.95	0.86
0.50	0.48	6.55	0.95	0.86

The viscosity data of the present solutions were analysed by using Jones-Dole equation (**Jone and Dole, 1929**),

$$\eta_r = \frac{\eta}{\eta_0} = 1 + AC^{1/2} + \mathrm{BC}$$

where η and η_o, respectively are the dynamical viscosities of solution and solvent, η_r is the relative viscosity of the solution. A is Falkenhagen coefficient[33] and is a measure of solute-solute interactions theoretically.

Table 4. Free volume (V_f), internal pressure (π_i) and relaxation time (τ) of Bu_4NI in DMSO- Acmixtures

c mol dm^{-3}	V_f x10^{-8} (m^3mol^{-1})		πix10^9 (Pa)		τx10^{12} (s)	
	298K	308K	298K	308K	298K	308K
Pure DMSO						
0.00	5.02	6.43	7.80	6.57	11.01	9.59
0.10	4.50	5.82	8.00	6.71	12.06	10.42
0.20	4.11	5.36	8.16	6.83	12.98.	11.27
0.30	3.78	4.97	8.30	6.93	13.88	11.93
0.40	3.48	4.61	8.45	7.03	14.72	12.71
0.50	3.26	4.33	8.54	7.10	15.68	13.44
90% DMSO						
0.00	6.64	8.30	7.10	6.02	9.35	8.30
0.10	5.95	7.50	7.28	6.16	10.12	9.01
0.20	5.43	6.92	7.43	6.26	10.90	9.63
0.30	4.99	10.32	7.56	6.36	11.65	10.32
0.40	4.60	10.99	7.69	6.45	12.41	10.99
0.50	4.32	11.54	7.78	6.52	13.13	11.54
80% DMSO						
0.00	8.25	10.14	6.59	5.63	8.23	7.42
0.10	7.37	9.08	6.77	5.77	8.99	7.13
0.20	6.73	8.37	6.91	5.87	9.61	8.68
0.30	6.20	7.78	7.04	5.96	10.61	9.22
0.40	5.72	7.24	7.16	6.05	11.05	9.77
0.50	5.37	6.81	7.25	6.11	11.65	10.32
70% DMSO						
0.00	10.53	12.53	6.06	5.24	7.29	6.67
0.10	9.45	11.24	6.23	5.38	7.85	7.25
0.20	8.58	10.41	6.37	5.46	8.49	7.73
0.30	7.92	9.66	6.48	5.54	8.97	8.29
0.40	7.30	8.94	6.61	5.63	9.51	8.68
0.50	6.91	8.43	6.67	5.69	10.02	9.10
60% DMSO						
0.00	13.40	15.43	5.59	4.87	6.36	5.95
0.10	11.98	13.82	5.75	5.00	6.83	6.47
0.20	10.93	12.71	5.87	5.10	7.32	6.91
0.30	10.08	11.83	5.98	5.18	7.82	7.31
0.40	9.34	11.04	6.08	5.25	8.39	7.72
0.50	8.76	10.42	6.17	5.30	8.70	8.19
50% DMSO						
0.00	16.54	18.13	5.20	4.60	5.63	5.50
0.10	14.68	16.18	5.35	4.74	6.14	6.09
0.20	13.40	14.88	5.47	4.83	6.57	6.49
0.30	12.36	13.83	5.57	4.91	7.09	6.76
0.40	11.44	12.95	5.67	4.97	7.46	7.13
0.50	10.74	12.22	5.74	5.03	7.86	7.47

On the other hand, B, the Jones -Dole coefficient, is empirical and is a function of the solute-solvent interactions. The coefficients A – and B- were calculated by least square fitting of experimental η values in the Jones-Dole equation as given above. The values of A and B thus obtained are listed in Table 3. The values of the A-coefficient are found to be positive in all the cases, indicating strong solute-solute interactions in the concentration range investigated. Most of the studies in pure and mixed solvents have been positive (**Patial *et al*., 2002**). The values of B-coefficients for tetralkyl ammonium Iodide are positive.

This is identical with the general observation (**Patial, 2015 and Jone and Dole, 1929**) that B-coefficients are commonly large and almost always positive for salts in non-aqueous solvents. The positive B-coefficients value attributed to strong solute-solvent interaction in the system. From Table 3, it is clear that viscosity B-coefficients for $Bu_4NI$is decrease with rise in temperature suggesting the structure-making tendency of Bu_4NI in the studied solvent system. This found to be consistent with the work reported in literature (**Patial, 2015; Patial, 2014 and Patial *et al*., 2002**). From Table 4, it has been found that the viscous relaxation time (τ) values increase with increase in concentration of solute in the studied solvent systems at both temperatures. Acoustic relaxation time decreases with the rise in temperature, in accordance with the decrease of density, ultrasonic velocity and viscosity of solution system with the increase of temperature.

Relaxation time values decrease with the increase of acetone contents in DMSO + Ac mixture. This may be accounted for the decrease of dielectric constant of the medium, and change of intermolecular interactions between DMSO and Ac molecules. Increase of τ with increase of solute concentration may be attributed to the presence of solute-solvent interactions. Similar results have been reported for tetraalkylammonium salts by **Syal *et al.* (1999)**. Free volume can be defined as the average volume in which the central molecule can move inside the hypothetical cell due to repulsion of surrounding molecules. Free volume can also referred as the void space between the molecules i.e. volume present as holes of monomeric size, due to irregular packing of molecules. The calculated values of free volume (V_f) are given in Table 4 for studied electrolyte in all the compositions at both temperatures. V_f values in general decrease in magnitude with the increase of concentration of salt.

However, with the increase of Ac content in DMSO+ Ac mixture, V_f values increase. Increase of temperature also increases the magnitude of V_f. Internal pressure (π_i) is the resultant of forces of attraction and repulsion between solute and solvent molecules of the solution. It is evident from the Table 4 that π_i values increase with the increase of solute concentration and decrease with increase of temperature in all composition for $Bu4_4NI$. Increase of π_i with concentration indicates increase in intermolecular interactions due to the forming of aggregates of solvent molecules around the solute, which affects the structural arrangement of solution system. This may also attributed to the presence of solute-solvent interactions. Similar behavior has been reported for PVP polymer in DMSO+H_2O system (**Syal *et al*., 2005**).

Conclusion

Hence, it may be concluded that the studied parameters hint to the presence of strong solute-solvent interactions between the component molecules in the mixture at different temperatures. However, these interactions decrease with increase in temperatures. Solute solvent interactions are much pronounced in higher DMSO content as well as at lower concentration of salt. Solute-solute interactions are very apparent in higher DMF and higher salt concentration.

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