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Study of Excess Parameters and Partial Molar Volume for the Molecular Interactions of an aqueous 2-(tert-butylamino)-1-(3-chlorophenyl) Propan-1-one and NaCl Solution at Different Temperatures

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Abstract: **Density(ρ),Viscosity(η),Ultrasonic Velocity(U) and Surface Tension(γ) of an aqueous-consolute, NaCl solution of 2-(tert-butylamino)-1-(3-chlorophenyl)Propan-1-one (0.0201, 0.0402, 0.0804,0.1608 mol/kg)were measured at 293, 303 and 313K. These data were used to calculate various acoustical parameters viz., adiabatic compressibility(β), acoustic impedance (Z), relative association(RA), molar Compressibility (W), Rao's Constant (R), free volume(V_f^E), Intermolecular free length (L_f), and partial molar volume which provides valuable information regarding drug-electrolyte (NaCl) interaction. The excess parameters viz. excess adiabatic compressibility (β_E), excess inter molecular free length (L_f^E) and excess free volume (V_f^E) were also calculated. These calculations help in predicting the intermolecular interactions.**

Index Terms: **Acoustical parameters, Excess parameters, Inter molecular interaction, Partial molar volume.**

I. INTRODUCTION

Recently ultrasonic studies in aqueous solution of various drugs yield a piece of essential information about the strength of the intermolecular interactions and physical nature (Baluja, Solanki & Kacchadia, 2007; Murshed, Castro, Lourenco, Lopes & Santos, 2011). The interpretation of the nature of molecular interactions depends upon knowledge of the basics of different experimental aspects of the binary fluid system and the hypothetical analysis of ultrasonic velocity. There is thus much practical use to study and understand the experimental aspects of certain aqueous electrolytic solution through the exposure of them to the ultrasound (Blandamer, 1973; Hwrzfeld & Litovitz, 1959). Extended applications can be found in the ultrasonic studies as the ultrasounic velocity in liquids and mixtures are closely related to several parameters which determine the physicochemical behavior of liquid and liquid soltions. Mixtures of solvents are extremely important in most chemical and industrial processes rather than single pure liquids because they give ample information of more component mixtures and it ensures that the derived characteristics of the medium can be continually adjusted. Relevant information on pharmaceutical and medicinal chemistry is provided through physicochemical behavior and inter-molecular interaction of the drug with an electrolyte. Multiple authors have examined interactions in between the molecules of various binary liquid mixtures with an electrolyte at varying temperatures. NaCl has specialized property like low melting but a high boiling point(Begaum, Sandhya, Karunakumar & Rambabu, 2013).These electrolytes are insoluble in ether but soluble in water (Begaum, Sandhya, Karunakumar & Rambabu, 2013; Gurung & Roy, 2006; ; Kharat, 2008; Nain, 2008; Thanuja, Kanagam, Sreedevi, 2011;Thirumaran & Sabu, 2012]. The interactions of the molecules of the drug and electrolytic solvent and its dependence on the temperature play a key role in understanding drug action (Dhondge, Zodape & Parwate, 2012a; Dhondge, Paliwal, Bhave & Pandhurnekar, 2012b; Syal, Chauhan & Chauhan, 2005; Thakur & Chauhan, 2011) Therefore, it can be fascinating to analyze the deviation of these parameters with temperature to understand drug-electrolytic solvent mechanisms (Baluja, Solanki & Kachhadia, 2007; Baragi & Maganur, 2013;

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Sonar & Pawar, 2010; Yadav & Yadav, 2005;) Thermodynamic studies of the binary solution with an electrolyte have attracted much attention. These experimental data from various scientist for the different systems can now be assessed and published (Ali &Navi, 2008; Chauhan, Singh, Kumar, Neelakantan et. al , 2016; Nain, 2007).

In the present study, an effort has been made to study the interaction between the drug, 2-(tert-butylamino)-1-(3-chlorophenyl) propan-1-one and the aqueous NaCl solution to determine the nature of the molecular environment. This drug is used as an anti-depressant drug (Fig.1)

II. Experimental Methods and Materials

The solutions of different concentration (mol/kg) of 2-(tert-butylamino)-1-(3-chlorophenyl) propan-1-one (2-TBP) (Molar mass = 276.201 g/mol) were prepared in double distilled water (DW) as the solvent. The densities (ρ) of these binary solutions were determined accurately using 25 ml density bottle in an electronic balance with ±0.01mg accuracy. The basic parameter ultrasonic velocity (U) had been measured on Digital Ultrasonic Pulse Echo Velocity Meter (Vi Microsystems Pvt. Ltd. Model VCT-70 with single frequency of 2 MHz having an accuracy of 0.1%. Ostwald's viscometer calibrated with doubly distilled water was used to determine the viscosities (η) of the solutions with an accuracy of ±0.001 Pa.sec. Stalagmometer was used to determine the surface tension of the solutions. These basic parameter U, η and ρ of various solutions of drug viz.,0.0201,0.0402, 0.0804, 0.1608mol/kg were measured at 293, 303 and 313 K. Thermostatically controlled water circulation system is used to maintain the temperature with an accuracy of 0.05^0C. For all studied solutions and pure components, measurements were repeated thrice. The various acoustical parameters were evaluated from U, η and ρ values by using following formulae.

1. Adiabatic compressibility (β) $= \frac{1}{U^2\rho}$ $Kg^{-1}ms^2$

   U= velocity; ρ= Density of liquid

2. Specific Acoustic impedance (Z)=U*ρ $Kg^{-1}ms^{-3}$

3. Relative association (RA) $= [\frac{\rho}{\rho_0}][\frac{U_0}{U}]^{1/3}$

Where ρ and ρ_0 are the densities of solution and solvent respectively. U and U_0 are the ultrasonic velocities of solution and solvent respectively.

4. Molar compressibility (W) $= \frac{(M.\beta)^{\frac{1}{7}}}{\rho}$

   Where ρ =density, M=Molecular weight,

   β = adiabatic compressibility

5. Rao's constant (R) $= \frac{M}{\rho} . [U]^{\frac{1}{2}}$

   Where, M = Molecular Weight,

   ρ = density

6. Free volume (V_f) $= [M_{eff}. \frac{U}{K\eta}]^{1/2}$ m^3,

   Where M_{eff}= effective molecular weight,

   K-temperature independent constant

   K = 4.28 $x10^9$ for all liquid) (Nori, Babavali & Srinivasu, 2019).

7. Intermolecular Free Length (L_f) =KT x $\sqrt{\beta}$

   Where, Jacobson's Constant,

   KT = (93.875 + 0.375 x T)

8. Surface Tension: $\gamma=(S/6.3\ X10^{-4}\rho)^{2/3}$ N/m

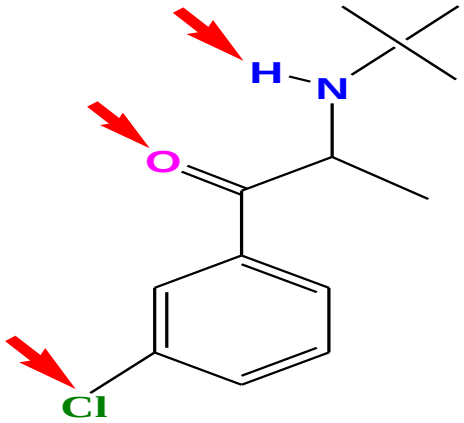

Fig.1-Structure of 2-(tert-butylamino)-1-(3- chlorophenyl) propan-1-one (arrow shows the interacting sites)

III. RESULT AND DISSCUSSION

The experimental data of density (ρ), viscosity(η), ultrasonic velocity (U) and Surface tension (γ) and the derived acoustic parameters of the aqueous 2-(tert-butylamino)-1-(3-chlorophenyl) propan-1-one (2-TBP)-NaCl solution at various studied concentrations at different temperature are reported in the table 1-6 .

Density, ultrasonic velocity, viscosity and surface tension are the essential parameters to know the strength of intermolecular interactions. Increase in these parameters with increased concentration shows that there is strong interaction between the drug and solvent and these interaction found to increase with the increase in temperature. Cohesive strength and thus molecular association in the solution is often dependable for observed values of unionized solute particles. It is also clearly observed that density, ultrasonic velocity, surface tension and viscosity rise with the increase in the aqueous co solute NaCl solution. This is may be due to the association of the aqueous co solute solution with the drug in consequence of dipole interaction between the Na^+ and Cl^- ions and the polar solvent.

Acoustic Impedance (Z) increases with an increase of drug concentration and the co solute, NaCl concentration. But Z was found to be decrease with the increase in the temperature which may be due to decrease in the interaction between the co-solute and solvent. Adiabatic compressibility (β) is measure of an inter-molecular association or dissociation measurement. The structure of the molecules affects their adiabatic compressibility and it also determines their orientation around the liquid molecules. Decrease in the adiabatic compressibility with the increase in the temperature and the increase in the co solute concentration concludes the structural changes in the drug- polar solvent interaction reflecting the system reduces compressibility. Relative association (RA) increases with concentration indicate that the solvation of the solute is existing over the breakdown of 2-TBP-aqueous solution aggregates (Table 6). The values of molar compressibility (W) and Rao's Constant (R) observed in the entire system are growing with increasing concentration at any temperature indicating that there are more components available in the region, thereby enhancing the medium's interactions in close packaging. Free Volume (V_f) is easily determined and is fundamental to the distinctly different interactions in the solution. The gradual raise in the free volume with the rise in the drug co solute NaCl mole fraction confirms the structural interaction between Na^+ and Cl^- ions and solvent.

The free length (L_f) of the neighboring molecules is the distance between them. The deviation of the ultrasonic speed in a solution depends on the mixing intermolecular free length. In the current study L_f decreases with a concentration of an aqueous drug solution signify that the extensive interaction between drug and solvent molecules. But the reduce in free length in the aqueous 2-TBP- co solute NaCl solution reveals the structural promoting behaviour between co solute and the drug.

Partial molar volume of an aqueous drug solution and 2-TBP-NaCl solutions were calculated by using the equation(Deosarkar, Tawde & Kalyankar, 2018; Arsule, Sawale & Deosarkar, 2019)-

$$\text{Partial molar volume} = \frac{M}{\rho} - \frac{\rho - \rho_0}{m\,\rho\,\rho_0}$$

Where, ρ and ρ_0 are the density of solvent and solution ($kg.m^{-3}$), m is the molality of solution ($mol{\cdot}kg^{-1}$). M is the molar mass of drug ($kg{\cdot}mol^{-1}$). Fig 2-3 shows the plot of partial molar volume against the concentration of the drug and the effect of co-solute NaCl.

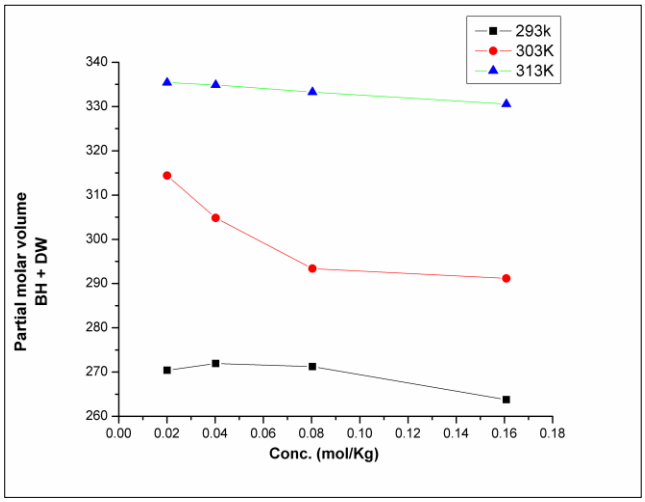

Fig. 2- Partial molar volume of 2-TBP + DW at different temperature.

Table 1:- Density (ρ), ultrasonic velocity (U) and Viscosity (η) and surface tension of drug at 293K, 303K and 313K in distilled water

Molar Conc.	Density(ρ) $Kg.m^{-3}$			Velocity(U) $m.s^{-1}$			Viscosity(η) $N.s.m^{-2}$			Surface tension (σ) X 10^5 N/m		
Temp.	293K	303K	313K	293K	303K	313K	293K	303K	313K	293 K	303K	313K
0.0201	1.0804	0.954	0.9034	1218.2	1212.4	1202.2	7.7175	5.958	6.040	6.1522	5.3556	4.9447
0.0402	1.1024	0.994	0.9125	1228.6	1216.9	1210.6	7.9331	6.279	6.114	6.4394	5.6446	5.1000
0.0804	1.1208	1.040	0.9213	1235.9	1221.2	1222.4	7.9269	6.589	6.265	6.6645	5.9685	5.3012
0.1604	1.1604	1.054	0.9312	1241.3	1225.2	1226.3	8.3094	6.708	6.352	6.9917	6.1062	5.4094

Table 2:- Acoustic impedance (Z) Adiabatic compressibility (β) and free length (L_f) and Wada's constant (W) of drug at 293K , 303K and 313K in distilled water.

Molar Conc.	Acoustic impedance(z) $x10^4 g/s/m^{-2}$			Adiabatic compressibility (βad)x10^{-7} $m^2 s^{-2}$			Free path length (L_f) x10^{-7}			Wada's constant (W)		
Temp.	293K	303K	313K	293K	303 K	313K	293K	303K	313K	293K	303K	313K
0.0201	1316.1	1156.6	1086.1	6.2373	7.1323	7.8476	6.4376	7.3564	8.0313	1.7924	1.6593	1.6256
0.0402	1354.4	1210.8	1104.7	6.0095	6.7945	7.4864	6.2028	7.0723	7.8613	1.7832	1.6682	1.6408
0.0804	1385.2	1270.5	1126.2	5.8415	6.4594	7.2695	6.0291	6.7119	7.6323	1.7943	1.6884	1.6812
0.1604	1440.4	1291.3	1141.9	5.5929	6.3243	7.1454	5.7728	6.5894	7.5945	1.7348	1.7018	1.6956

Table 3 :- Rao's constant (R), free volume (Vf) and Relatively association of drug at 293K , 303K and 313K in distilled water.

Molar Conc.	**Rao's constant (R) x101m$^{10/3}$s$^{-1/3}$Mol^{-1}**			**Free volume (V_f) x 10^{-8}**			**Relatively Association (R_A)**		
Temp	**293K**	**303K**	**313K**	**293K**	**303K**	**313K**	**293K**	**303K**	**313K**
0.0201	442503.7	498825.1	522304.3	4.7382	3.6445	3.6696	0.04262	0.03782	0.03612
0.0402	437492.9	480335.3	520702.9	4.9121	3.8594	3.7378	0.04312	0.03927	0.03723
0.0804	432894.8	460720.5	520762.2	4.9375	4.0616	3.8616	0.04359	0.04095	0.03822
0.1608	419843.4	456264.9	516886.5	5.1984	4.1419	3.9315	0.04493	0.04135	0.0395

Table 4:- Density (ρ), ultrasonic velocity (U), Viscosity (η) and surface tension(γ) of drug at 293K, 303K and 313K in aq. NaCl solution

Molar Conc.	**Density(ρ) Kg.m^{-3}**			**Velocity(U) m.s^{-1}**			**Viscosity(η) N.s.m^{-2}**			**Surface tension (σ) X 10^5 N/m**		
Temp.	**293K**	**303K**	**313K**	**293K**	**303K**	**313K**	**293K**	**303K**	**313K**	**293K**	**303K**	**313K**
0.0201	1.3856	1.3244	1.3228	1210.3	1214.2	1216.4	9.9466	7.8821	7.6268	9.9466	7.8821	7.6268
0.0402	1.4356	1.3832	1.3748	1215.9	1219.2	1223.2	10.282	8.4190	8.0431	10.2802	8.4190	8.0438
0.0804	1.4636	1.4272	1.4023	1218.6	1222.3	1225.3	10.853	8.8634	8.5043	10.8513	8.8634	8.5048
0.1608	1.5072	1.4748	1.4488	1220.3	1224.5	1230.2	10.970	8.3455	8.3735	10.9704	8.3455	8.3455

Table 5:- Acoustic impedance(Z) Adiabatic compressibility (β) and free length (L_f) and Wada's constant (W) of drug at 293K, 303K and 313K in NaCl.

Molar Conc.	**Acoustic impedance(z) x10^4g/s/m^{-2}**			**Adiabatic compressibility (β_{ad})x$10^{10}10^3$Kg^{-1}ms^{-2}**			**Free path length (L_f) x10^{-7} m**			**Wada's constant (W)**		
Temp.	**293K**	**303K**	**313K**	**293K**	**303K**	**313K**	**293K**	**303K**	**313K**	**293K**	**303K**	313K
0.0201	1676.9	1620.0	1600.0	4.92694	5.1204	5.1234	5.0213	5.3334	5.3723	1.6238	1.6593	1.6654
0.0402	1745.5	1686.3	1640.6	4.71162	4.8634	4.8607	4.8631	5.0713	5.1163	1.7308	1.6782	1.6848
0.0804	1783.5	1744.4	1717.8	4.60103	4.6984	4.7507	4.749E	4.8893	4.9934	1.7328	1.6884	1.6832
0.1608	1839.2	1805.8	1782.3	4.45557	4.5234	4.5627	4.5988	4.7133	4.7947	1.7624	1.6813	1.7343

Table 6:- Rao's constant (R), free volume (V_f) and Relatively association of drug at 293K, 303K and 313K in aq. NaCl.

Molar Conc.	**Rao's constant (R) x10^1m$^{10/3}$s-$^{1/3}$Mol^{-1}**			**Free volume (V_f) x 10^{-8}**			**Relatively association (R_A)**		
Temp	**293K**	**303K**	**313K**	**293K**	**303K**	**313K**	**293K**	**303K**	**313K**
0.0201	3428	359850.2	360938.3	6.0671E+16	4.83E+16	4.67E+16	0.055	0.052	0.052
0.0402	3324	345971.8	349227.7	6.2996E+16	5.19E+16	4.94E+16	0.056	0.054	0.054
0.0804	3268	336158.2	343040.3	6.6644E+16	5.47E+16	5.20E+16	0.057	0.056	0.054
0.1608	3177	325883.2	333286.7	6.7469E+16	5.38E+16	5.15E+16	0.059	0.057	0.056

Table 7:- The Redlich –Kister coefficients of the calculated Excess parameters of the BH+DW+ NaCl solutions at 293K, 303K and 313K.

Excess Parameters	**Coefficients**	**Temperature**		
		293 K	**303K**	**313K**
` Excess free volume (V_f^E) X 10^{-7}m^3/mol	A_0	5.4729	8.1156	8.4977
	A_1	-1.4735	-1.6174	-1.4003
	A_2	1.2101	1.7866	1.5350
	A_3	-3.0169	-2.9170	-2.8133

	A_4	1.3901	1.2135	1.1974
	A_5	5.4729	8.1156	8.4977
Excess intermolecular free length (L_f^E) $X10^{-2}$ A^0	A_0	-1.1595	0.4124	6.1833
	A_1	-1.2524	-1.9753	-7.8452
	A_2	-9.6057	-1.4549	-2.0016
	A_3	2.8377	5.3632	1.1361
	A_4	-2.8193	-5.5252	-1.2973
	A_5	8.9856	1.7272	4.3866
Excess adiabetic compressibility X $10^{-11}m^2/N$	A_0	-1.4735	-1.6174	-1.4003
	A_1	-1.9155	-3.2341	-4.1245
	A_2	2.9072	5.9135	7.2749
	A_3	-3.6933	-7.1228	-9.352
	A_4	1.3588	2.4863	3.3714
	A_5	0.07297	0.1394	0.2123

The dipole interaction between the co solute ions and the solvent may be the reason for the rise in the partial molar volume of the drug -co solute NaCl than in the aq. drug concentration. These interactions were found to increase with the rise in the temperature.

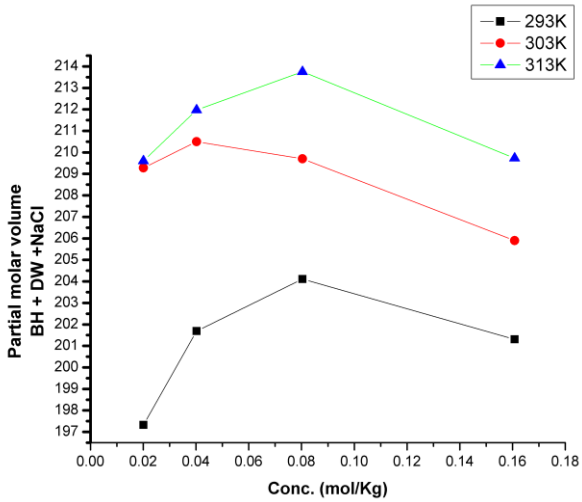

Fig.3- Partial molar volume of 2-TBP+ DW+ NaCl at different temperature.

The divergence between the experientially measured values and the ideal values at the particular thermodynamic stage are called as the excess properties. These properties are dependent on molecular size as well as shape and the mixture-component interactions. The various excess molar properties for studied solutions were evaluated using the relation given below- (Deosarkar, Tawde& Kalyankar, 2018).

$$Y^E = Y_{Exp} - (x_1Y_1+x_2Y_2) \text{ ----------(1)}$$

Where, Y^E is the excess thermoacoustical parameter. Mole fraction of solute and solvent are given by x_1 and x_2 respectively. Y_{Exp} is experimentally determined parameter. Molecular interactions among the components were discussed by using these calculated excess parameters. These excess parameters are built-in to the Redlich-Kister polynomial given below (Redlich & Kister, 1948).

$$Y^E = x1(1\text{-}x_1) \sum_{i=1}^{n} A_i \, (2x_i - 1)^{i-1} \text{ ----------(2)}$$

Y^E is the excess thermoacoustical parameter. are the Mole fraction of solute and solvent are given by x_1 and x_2 respectively. The regression analysis method was used to calculate the values of the coefficients A_i. These values are summarized in Table 7.

The variations of excess free volume (V_f^E) with the concentration of 2-TBP and electrolyte in the aqueous solution are represented in Fig.4. For the studied mole fraction range the values of V_f^E are negative for the given concentrations of 2-TBP. Thus it is concluded from these observations that these component molecules are more close proximity in the liquid mixture than in pure liquids. This reflects the strong dipole–dipole interactions between component molecules. The values of the excess free volume depend on the temperature. From Fig.4 it is also observed that the temperature effect is not uniform. With the initial 10K rise in the temperature from 293 to 303K there is no considerable change in the excess free volume but with further rise of 10K shows measurable change in the its values. This implies that the decreases in the interaction with increase in temperature, which is credited to more aggressive thermal movement and the decline in the solvation of solute ions (Chauhan, Singh, Kumar, Neelakantan & Kumar, 2016; Roy, Jha & Dey.2001)

From Fig. 5, excess intermolecular free length (L_f^E) values are observed to be negative for the full mole fraction range of binary liquid mixutre. The negative trend of L_f^E indicates the sturdy interactions between liquid mixtures and this is supported by the Kerr effect as well (Deosarkar, Tawde& Kalyankar, 2018).

Furthermore, L_f^E values shows negative trend with temperature reveals that as the temperature rise interaction decreases.

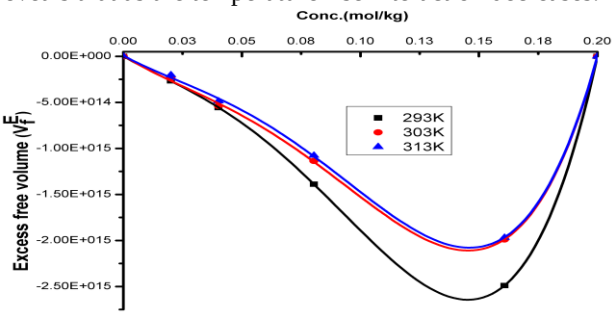

Fig.4- Excess free volume (V_f^E) of an aqueous 2-TBP +NaCl at different temperature.

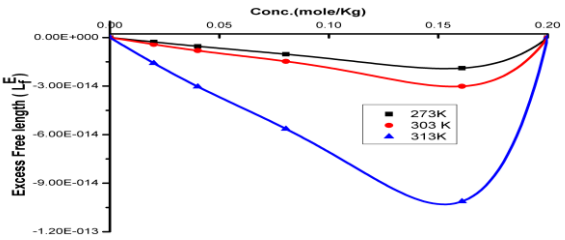

Fig. 5- Excess free length (L_f^E) of an aqueous 2-TBP + NaCl solution at studied temperature

Fig. 6 shows that over the entire concentration range, the β_E values for aqueous temperature range were found to be negative. This shows that the intermolecular attraction between the component molecules is a stronger indication of strong hetero-molecular interaction in the liquid mixtures. Excess compression of both the liquid mixtures is negative in the present study. It is also found that with the rise in temperatures interactions become weak, β_E values decrease negatively.

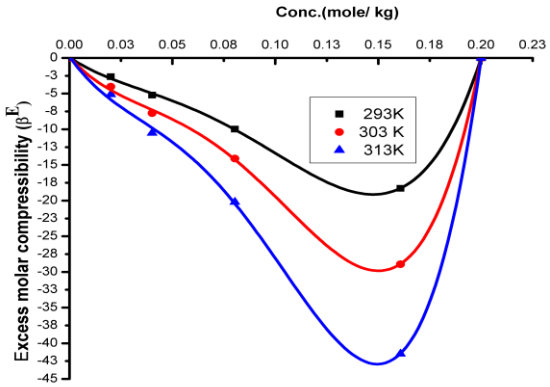

Fig. 6- Excess adiabatic compressibility (β^E) of an aqueous 2-TBP +NaCl solution at different temperatures.

CONCLUSION

In an aqueous 2-TBP and co solute NaCl solution, ultra-sonic velocity, density, viscosity and surface tension are measured at 293, 303 and 313K. The excess thermo-acoustic parameters, such as excess freely intermolecular (L_f^E), excess adiabatic compressibility (β_E) and excess free volume (V_f^E), are calculated for the whole mole fraction range. An interpretation of these data shows that the interaction between drug and solvent dominate over interaction between the solute-solute (Deosarkar, Tawde, Kalyankar, 2018). Thus it may be concluded that the hydration sphere of the drug releases water molecules due to the electrolyte NaCl and solvent interactions. 2-TBP has multiple interacting sites viz., secondary amine, carbonyl, cholo-phenyl group, shown by the red headed arrows (Fig.1). The deviation of the various studied parameters of an aq. NaCl solution of drug may be due to the intermolecular interactions at these sites. The anomalous relationship is observed in different excess parameters due to the characteristic property that strong dipolar interactions and directional interactions between the component molecules simultaneously act. Also in the aqueous co solute NaCl solutions the strength of intermolecular interactions is observed to decline with temperature.

CONFLICT OF INTERESTS

Authors declare that there is no conflict of interest regarding the publication of present research article.

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