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EXCESS THERMODYNAMIC PARAMETERS OF 1-[2-(DIMETHYLAMINO)-1-(4-METHOXYPHENYL) ETHYL] CYCLOHEXAN-1-OL IN AQUEOUS NA_2SO_4

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ABSTRACT:
Density(ρ), Viscosity(η), Ultrasonic Velocity(U) and Surface Tension(γ) of an aqueous-consolute, Na_2SO_4 solution of [2-(dimethylamino)-1-(4-methoxyphenyl) ethyl] cyclohexan-1-ol (Venlafaxine) 0.0201,0.0402,0.0804,0.1608 mol/kg were measured at 293.15, 303.15 and 313.15K. The resulting data were used to calculate various acoustical parameters, acoustic impedance (Z), adiabatic compressibility(β), Intermolecular free length (L_f), Wada's Constant (W), Rao's Constant (R), free volume (V_f), Relative Association (R_A) were calculated which provides valuable information regarding drug-electrolyte (Na_2SO_4) interaction. The excess parameters viz. partial molar volume and excess adiabatic compressibility (β^E), excess inter molecular free length (L_f^E), excess free volume (V_f^E) were also calculated. These calculations help in predicting the intermolecular interactions.

Key words: - Venlafaxine*; Acoustical parameters; Inter molecular interaction; Excess Parameters; Partial molar volume.*

INTRODUCTION:

Ultrasonic velocity measurements of liquid and mixtures allow compressibility calculations and, therefore, structural information can be obtained (Akhilesh *et.al.,* 2012, Ananthanarayan *et.al.*, 2003, Anderson *et. al.,*1968, Bauer *et.al.*,1966). In turn, sound speed data can be examined by using the theory of Jacobins free length. Dipole-dipole, dipole-inductive dipole and other dispersing force interactions were attributed with the deviations found in free length and other parameters (Biswas *et. al.*, 2002, Cruickshank *et al.,*1975, Dubey *et. al.*, 2003).

Thermodynamic and transport properties derived from density, sound speed and viscosity measurement of liquid mixtures are useful for understanding the nature of intermolecular interaction and provide important data for molecular liquid structures (Ali *et.al.*, 2002, Rama Rao *et.al.,*2004, Badran *et.al.,*2014). The intermolecular interactions among various species were widely used in the study. The excess thermodynamic functions (Djekic *et. al.,*2011) are sensitively dependent not only on intermolecular forces differences but also on molecule size differences. Excess properties give us a better understanding of intermolecular interactions of a molecular system and their nature and scope Excess functions are overly thermodynamic in comparison to the ideal solution of the same temperature, pressure and composition. For the description of solvent non-ideal compliance, excess functions are useful for mixing different components. Roy *et. al.,* 2003, Dash *et. al.,* 2013, Sreekant *et.al.,* 2011, Sravana *et. al.,* 2007 have measured sound density, viscosity

and speed for a wide range of alcohol-containing binary blends, and these features have been interpreted for specific or unspecific interactions (Almasi *et. al.,* 2011, Anuradha *et. al*, 2005.Kagathara *et.al.,* 2000)

In the study of molecular interactions and arrangements, excess thermodynamic properties of mixtures are helpful. The change in temperature and mixture composition of these properties that include polar molecules and hydrogen components can be complicated by a decrease and an increase in the interaction between hydrogen binding due to mixing. Various researchers have examined the various molecular interactions on the basis of sign and the excess parameters (Venkatramana *et. al.,* 2014, Ramadevi *et. al.,* 1995) in binary systems. The importance of excess parameters in the design calculations such as chemical processes, chemical separations, mass transfers (adsorption, precipitation evaporating), heat transfer and fluid flow was identified by the chemical industries (Venkatesu *et. al.,* 2006) This drug is [2-(dimethylamino)-1-(4-methoxyphenyl) ethyl] cyclohexan-1-ol (DCH) known as venlafaxine and used an antidepressant medicine that belongs to a drug group of serotonin and norepinephrine.

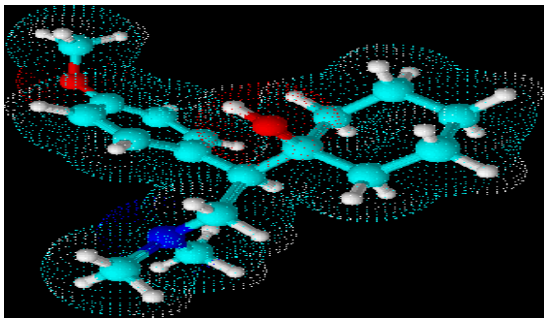

Fig. 1 Structure of -[2-(dimethylamino)-1-(4-methoxyphenyl) ethyl] cyclohexan-1-ol

MATERIAL & METHODS:

The solutions of different concentration (mol/kg) of 2-(dimethylamino)-1-(4-methoxyphenyl) ethyl] cyclohexan-1-ol Venlafaxine (Molar mass = 276.201 g/mol) were prepared in double distilled water (DW) as the solvent. The densities (ρ) of these binary solutions were measured accurately using 25 ml specific gravity bottle in an electronic balance with an accuracy of ±0.0001g. 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%. The viscosities (η) of solutions were determined by using Ostwald's viscometer by calibrating with doubly distilled water with an accuracy of ±0.001 Pa.sec. Surface tension was measured by using Stalagmometer. The basic parameter U, η and ρ of various concentrations of drug viz.,0.0201, 0.0402, 0.0804, 0.1608 mol/kg were measured at 293.15, 303.15 and 313.15K. Thermostatically controlled water circulation system is used to maintain the temperature with an accuracy of 0.05 0C. For all solutions and pure components, triplicate measurements were performed. The various acoustical parameters were calculated from U, η and ρ values by using standard formulae as shown below-

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 $Kg^{-1}ms^{-2}s^{-1}$

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

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

4. Absorption coefficient or Attenuation (α/f2) = $8\pi i^2 \eta f^2 /3\rho U^3$ $NPm^{-1}s^2$

   Where f is the frequency of ultrasonic wave

5. Molar compressibility (W) = $[M.\beta^{1/7}]/\rho$

Where ρ =density, M=Molecular weight,

β = adiabatic compressibility

6. Rao's constant or molar sound velocity (R) $=M/\rho\ [U]^{1/2}$

Where, M = Molecular Weight,

ρ = density

U = Ultrasonic Velocity

7. Free volume (Vf) = $[M_{eff}\ U/K\eta]^{1/2}.m^3$

Where M_{eff}= effective molecular weight,

K is a temperature independent constant (4.28 x109 for all liquid)

8. Intermolecular Free Length (L_f) =KT x $\beta^{1/2}$

Where, KT = The Temperature dependent constant known as Jacobson's Constant

KT = (93.875 + 0.375 x T),

T=Absolute Temp. in Kelvin.

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

Where, U=Ultrasonic Velocity,

ρ =Density of solution

RESULT AND DISCUSSION:

Table (1-6) gives the density (μ), viscosity (μ), ultrasonic-velocity (U) and surface tension (ϒ) of an aqueous drug and Na_2SO_4 solution and the derived acoustic parameters are shown at different concentrations at different temperatures.

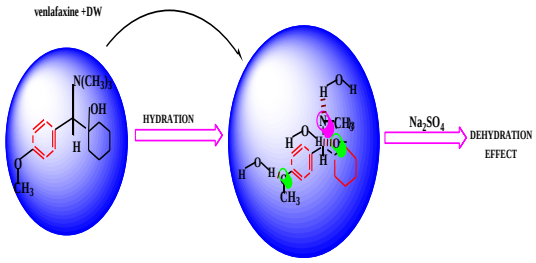

Fig.2- Strong solute-solvent interactions and hydrogen bonding between DCH +water

The essential parameters for understanding the strength of molecular interactions include density, ultrasound speed, viscosity and surface tension. Increased concentration of these parameters show a strong interaction between the medicine and the solvent and these interactions increased as the temperature increased. The observed values of unionized solute particles are often the result of cohesive strength and thus molecular association in the solution. It is also evident that with the increase in the aqueous solution Na_2SO_4, density, ultrasound velocities, viscosity and surface tension increase. This may be because the aqueous co-solute solution is associated to the drug due to dipole interaction between Na^{++} and SO^{4-} -and the polar solvent.

With increased drug concentration and co-solute concentration Na_2SO_4, acoustic impedance (Z) increases. But Z decreased when the temperature was increased, which could be due to the reduction of the interaction between the solvent and the solvent. The measurement of an inter-molecular association or dissociation is adiabatic compressibility (β). It also affects the structures of the molecules and their orientation around the liquid molecules. The reduction in adiabatic compression with an increase in the temperature and the increase in co solute concentration indicates that, due to structural changes in the drug-polar solvent interaction, the system is less compressible. The increased concentration relative association (RA) indicates that the salvation of solute over the breakdown of solvent-DCH aggregates is prevalent (Table 6). With increasing levels of concentration at any temperature, the molar compression values (W) and the Rao's constant (R) found throughout the system show that additional compositions are available in the region thus enhancing the interaction of the medium with close packaging. Free volume (V_f) is easy to determine and is critical for the clearly different solution

interactions. The gradual increase in the free volume of the Na_2SO_4 mole fraction co-solution drug confirms the structural interaction between Na^{++} and $SO4^{--}$ ion and solvent. A decrease in its value with rising temperatures could result in a steric impediment to the component molecules and dispersion forces. The distance between the neighbouring molecules is their free length (L_f).

The variation of the ultrasonic velocity depends on the free length of the mixing system. In the present study, L_f declines with an aqueous drug solution concentration means that there is extensive interaction between drug and solvent molecules. But the free-length decline in the aqueous solution DCH and co-solute Na_2SO_4 reveals structural support between co-solute and drug. The partial molar volume of a water-based solution for the drug and solution DCH - Na_2SO_4 were determined by using the equation (Redlich, *et.al.*, 1948).

$$\text{Partial molar volume} = \frac{M}{\rho} - \frac{\rho - \rho 0}{m \rho \rho 0}$$

Where, ρ and ρ0 are the densities 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.3 shows the plot of partial molar volume against the molality of the drug and the effect of co-solute Na_2SO_4.

The increase in the partial molar volume of the Na_2SO_4 drug-co-solute as opposed to that of the aqueous drug concentration is due to the dipole relationship between the co-solute ions and the solvent.

The excess properties represent the difference between real values at the same thermodynamic stage (experientially measured) and ideal values. The excess properties depend on molecular geometry (size and shape) and interactions between mixture and component. The following relation can be used to calculate different excess molar properties for binary mixtures (Deosarkar *et al.,* 2018).

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

Where, Y^E is the excess thermos-acoustical parameter. x_1 and x_2 are the mole fraction of solute and solvent. Y_{Exp} is a parameter which is determined experimentally. The values of the coefficients Ai are calculated using regression analysis are summarised in Table 7. These calculated excess parameters may be used to determine molecular interactions among the mixture components. The following Redlich-Kister Polynomial is fitted with these excess parameters: (Redlich, *et.al.,*1948).

$$Y^E = x_1(1\text{-}x1) \sum_{i=0}^{n} Ai\ (2xi - 1)^{i-1}$$

Y^E is the excessive parameter of thermo acoustics. The mole fraction of the solution and the solvent are $\mathbf{x_1}$ and $\mathbf{x_2}$. Table 1 summarizes the values of the $\mathbf{A_i}$ coefficients with regression analysis.

In the Fig 4 are represented the variations of excess free volume (V_f^E) and mole fraction of DCH and electrolyte in aqueous solution. V_f^E values in the aqueous solution are negative over the selected mole fraction range. The component molecules are therefore more closely linked in a liquid than in pure liquids, indicating that strong attractive interactions between component molecules like dipole-dipole interactions and other specific interactions between molecules other than those of the system are operational. At the end of the interaction, V_f^E values are reduced by temperature (Tarlok, *et. al.2004).*

From table 1 it was observed that for the entire mole fraction range of binary liquid mixtures the values of excess intermolecular free length (L_f^E) were found to be negatives. The negative values of excess intermolecular free length show strong interactions between liquid mixing components and the support for the Kerr effect (Venkatesu, *et. al.* 2006). In addition, L_f^E values show a negative trend, with a decrease in temperature interaction.

The excess adiabatic compressibility (β^E) values were found to be negative over the whole concentration range for the aqueous temperature range. This illustrates that, due to dipole–dipole dipole interactions between the dipole and dipole, the intermolecular attraction in component molecules represents a stronger indication of strong hetero-molecular interaction in the fluid mixtures. In the current study, excess compression is negative in both liquid mixtures. The increase in temperatures also means weaker interactions, and β^E values are decreasing negatively.

CONCLUSION

In an aqueous 2-(dimethylamino)-1-(4-methoxyphenyl)ethyl]cyclohexan-1-ol and co-solute Na_2SO_4 solution, ultra-sonic velocity, density, viscosity and surface tension are measured at 293.15, 303.15 and 313.15K. The excess thermo-acoustic parameters, such as excess freely intermolecular (Lf^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 (Venkatesu, *et. al.* 2006). Thus it may be concluded that the hydration sphere of the drug releases water molecules due to the electrolyte Na_2SO_4 and solvent interactions. 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 Na_2SO_4 solutions the strength of intermolecular interactions is observed to decline with temperature.

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Table 1:- Values of Density (ρ), ultrasonic velocity (U) and Viscosity (η) of drug at 293.15, 303.15K and 313.15K in distilled water.

Molar Conc.	Density(ρ) Kg.m^{-3}			Velocity(U) m.s^{-1}			Viscosity(η) N.s.m^{-2}			Surface tension (N/m	
Temp.(K)	293.15	303.15	313.15	293.15	303.15	313.15	293.15	303.15	313.15	293.15	303.15
0.0201	1.3036	1.1664	1.0712	1174.7	1226.5	1237.4	7.7175	5.958	6.040	6.6566	6.7789
0.0402	1.3284	1.2285	1.0924	1217.5	1234.7	1247.9	7.9331	6.279	6.114	7.5517	7.2336
0.0804	1.3316	1.2444	1.1164	1224.1	1256.8	1257.2	7.9269	6.589	6.265	7.6937	7.7816
0.1604	1.3324	1.2524	1.1247	1267.2	1269.3	1270.4	8.3094	6.708	6.352	8.5404	8.0619

Table 2:- Values of Acoustic impedance (Z) Adiabatic compressibility (β) and free length (L_f) and Wada's constant (W) of drug at 293.15K, 303.15K and 313.15K in distilled water

Molar Conc.	Acoustic impedance(Z) x10^{4}g/s/m^{-2}			Adiabatic compressibility (β_{ad}) x10 $^{-11}$ m ^{2}/ N			Free path length (L_f) x10^{-13} m			Wada's constant (W) X10^8		
Temp.(K)	293.15	303.15K	313.15	293.15	303.15	313.15	293.15	303.15	313.15	293.15	303.15	313.15
0.0201	1531.33	1430.59	1325.50	5.5592	5.7239	6.1349	5.7378	5.9913	6.3583	1.5576	1.6963	1.7273
0.0402	1617.37	1506.33	1363.20	5.0745	5.3819	5.8827	5.2417	5.6524	6.1273	1.6693	1.7185	1.7598
0.0804	1630.01	1563.96	1403.03	5.0185	5.0993	5.6785	5.1729	5.3534	5.9434	1.6828	1.7796	1.7754
0.1604	1688.41	1589.29	1427.93	4.6857	4.9674	5.5194	4.8241	5.2134	5.7413	1.8915	1.8145	1.8123

Table 3 :- Rao's constant (R), free volume (V_f) and Relatively association of drug at 293.15K , 303.15K and 313.15K in distilled water

Molar Conc.	Rao's constant (R) x10²m $^{10/3}$ s$^{-1/3}$Mol^{-1}			Free volume (V_f) x m³			Relatively Association (R_A)		
Temp. (K)	293.15	303.15	313.15	293.15	303.15	313.15	293.15	303.15	313.15
0.0201	35368.6	41274.5	453409.1	4.6403E+16	3.7924E+16	3.5254E+16	0.0533	0.0457	0.0416
0.0402	35971.7	39729.4	448382.7	5.0544E+16	4.1184E+16	3.9985E+16	0.0524	0.0474	0.042
0.0804	36082.7	39641.2	442171.7	5.3315E+16	4.4482E+16	4.2254E+16	0.0522	0.0475	0.042
0.1608	37332.8	39772.5	443634.1	5.6748E+16	4.57482E+16	4.5183E+16	0.0505	0.0474	0.0423

Table 4:- Values of Density (ρ), ultrasonic velocity (U) , Viscosity (η) and surface tension(γ) of drug at 293.15K, 303.15K and 313.15K in aq. Na_2SO_4 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. (K)	293.15	303.15	313.15	293.15	303.15	313.15	293.15	303.15	313.15	293.15	303.15	313.15
0.0201	1.3024	1.1268	1.0624	1210.8	1231	1255.4	8.0452	5.2254	5.4279	7.28236	6.6211 2	6.62132
0.0402	1.324	1.1668	1.0936	1230.7	1265.6	1272.6	8.7094	5.6036	5.6761	7.77419	7.4506 8	7.09977
0.0804	1.3488	1.1956	1.1244	1246.6	1272.4	1287.9	9.1581	5.9394	5.9975	8.23075	7.7583 1	7.56620
0.1608	1.3588	1.2384	1.1564	1255.2	1274.1	1297.2	9.7542	6.6020	6.5437	8.46457	8.0683 0	7.95132

Table 5 :- Values of Acoustic impedance(Z) Adiabatic compressibility (β) and free length (L_f) and Wada's constant (W) of drug at 293.15K, 303.15K and 313.15K in Na_2SO_4

Molar Conc.	Acoustic impedance(z) x10⁴g/s/m^{-2}			Adiabatic compressibility (β_{ad}) x10^{-11}m² /N			Free path length (L_f) x10-^{13} m			Wada's constant (W) X10⁸		
Temp. (K)	293.15	303.15	313.15	293.15	303.15	313.15	293.15	303.15	313.15	293.15	303.15	313.15
0.0201	1576.9	1387.01	1333.7	5.2373	5.8603	5.9714	5.4057	6.1596	6.2237	1.54752	1.68701	1.71713
0.0402	1629.4	1476.7	1391.7	4.9866	5.3519	5.6552	5.1469	5.6273	5.8813	1.66234	1.70964	1.74639
0.0804	1681.4	1521.2	1448.1	4.7708	5.1725	5.3658	4.9243	5.4313	5.5838	1.68041	1.77139	1.77252
0.1608	1705.5	1577.8	1500.0	4.6710	4.9717	5.1442	4.8213	5.2392	5.3583	1.80083	1.80595	1.80993

Table 6:- Rao's constant (R), free volume (V_f) and Relatively association of drug at 293.15K, 303.15K and 313.15K in aq. Na_2SO_4

Molar Conc.	Rao's constant (R) $x10^{-2}m^{10/3}s^{-1/3}Mol^{-1}$			Free volume (V_f) x m^3			Relatively association (R_A)		
Temp. (K)	293.15	303.15	313.15	293.15	303.15	313.15	293.15	303.15	313.15
0.0201	364904.1	428807.1	463815	4.6403E+16	3.7984E+16	3.5272E+16	0.051697	0.043993	0.040673
0.0402	364850.5	425746.2	456755.9	5.05446E+16	4.1184E+16	3.9528E+16	0.051705	0.044309	0.041301
0.0804	362769.1	417723.1	449585.2	5.33115E+16	4.4864E+16	4.2282E+16	0.052002	0.04516	0.04196
0.1608	362583.6	403825.1	440300.9	5.65748E+16	4.5798E+16	4.5193E+16	0.052028	0.046715	0.042845