Speed of Sound and Apparent Molal Volume Approach to Study the Ion-Solvent Interactions in Pharmaceutically Important Liquid Systems

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Article history Received: 17-02-2020 Revised: 14-04-2020 Accepted: 20-04-2020

Corresponding Author: Susmita Kamila Department of Chemistry, East Point College of Engineering and Technology, Bangalore – 49, India Bhubaneswar, Odisha. Email: sushkam@yahoo.co.in **Abstract:** Presence of zinc ion and lactose in different pharmaceutical compositions have been inspired to investigate the ion-solvation and related interactions among the components in solution media. This study has been carried out by the measurements of ultrasonic velocity, viscosity and density for the binary solutions of zinc sulphate (ZnSO₄) in lactose-water mixed solvent systems at 303.15, 308.15, 313.15 K temperature and at atmospheric pressure. Different thermo-acoustic parameters such as isentropic compressibility, apparent molal volume, solvation number, free volume, internal pressure etc., have been analyzed in the light of molecular interactions from their variations against concentration as well as temperature. Presence of ion-solvation and structure promoting nature of solute are the major findings in the present investigation. Besides, it also reveals that the ion-solvation decreases as the temperature increases.

Keywords: Solute-Solvent Interaction, Ion-Solvation, Thermo-Acoustic Parameters, Apparent Molal Compressibility

Introduction

Certain metal ions are essential for different types of critical functions in human organs. Scarcity of such metal ions can lead to many complicated diseases. Few alkali metals including some transition metals are currently known to be required for normal biological functions in humans and even other living organisms (Lane and Morel, 2006; Staiger and Pietak, 2006). Zinc is an essential and most important trace element in biological systems (Coleman, 1967). It is the second most abundant transition metal in organisms after iron and it is the only metal which appears in all enzyme classes (Wapnir, 1990; Broadley et al., 2007). Zinc metal is mostly present in the brain, muscle, bones, kidney and liver (Pfeiffer and Braverman, 1982). Deficiency of zinc produces growth retardation, skin lesions, poor appetite and loss of body hair etc. Even the deficiency of this metalion affects adversely chronic diseases like neurological disorders, Alzheimer's disease and other age-related diseases (Evans, 1986). The unique properties of zinc may have significant therapeutic benefits in these diseases. Besides, zinc ions along with other metal ions such as magnesium, potassium, sodium etc., have their importance in pharmaceutics for the preparations of various drugs. Especially, zinc sulfate is used to treat and to prevent zinc deficiency and also it is used as an alternative to, or in a rotation with, copper sulphate for controlling digital dermatitis. It is supposed to be a better supplement for chronic diarrhea (Khan and Sellen, 2011).

In continuation of our earlier works on ion solvation (Kamila *et al.*, 2017; Dash and Kamila, 2017), presently the inter-molecular interactions of zinc sulphate are studied with a saccharide like lactose in water systems. Generally, carbohydrates are supposed to be the major component of the entire living organisms in energy production and other protein synthesis processes. Various saccharides are known to have their functions as stabilizing agents for the native state of proteins and enzymes (Dash and Kamila, 2015; Kannappan and Vinayagam, 2006). They also show their importance in the field of chemicals, bio-chemicals, foods and pharmaceutical industries. Besides, it is exclusively present in milk apart from medicines and culture media. Absorption of lactose is dependent upon the enzyme



lactase that enhances the absorption of calcium, magnesium and zinc. It also promotes the growth of lactobacilli and provides galactose, which is essential for the formation of cerebral galactolipids and hence development of the brain. Review of literature shows that zinc sulphate mixed with lactose of different potencies is used in homeopathy for the biological activity (Chauhan *et al.*, 2013).

Number of studies has been reported for various electrolytes with mixed solvent systems for the intermolecular association studies (Kamila *et al.*, 2017; Dash and Kamila, 2017; 2015; Kannappan and Vinayagam, 2006; Jones and Dole, 1929; Nain *et al.*, 2013a). Besides, interaction studies have also been reported using different carbohydrates as major reacting components in the binary systems (Gupta, 1991; Timasheff and Arakawa, 1989; Nithiyanantham and Palaniappan, 2013; Kannappan and Palani, 2007). But, however, studies involving transition metal ion with a disaccharide in mixed solvent system is scarce in literature. So an attempt has been made to study the solvation of zinc sulphate in lactose-water mixed solvent system at three different temperatures.

Material and Methods

Analytical grade zinc sulphate heptahydrate and lactose were procured from Qualigen Chemicals (more than 99.9% purity). Stock solutions of lactose were prepared by taking accurate weight of solute in freshly prepared double distilled water (w/V ratio). The waterlactose mixed solvent system varies from 0% to 10% (w/V). Similarly, different ZnSO₄ solutions in lactosewater mixed solvents were prepared by taking exact amount of ZnSO4 in mixed solvents and the concentration of zinc sulphate varies from 0.01 to 0.1 mole dm⁻³. The solutions were kept for half an hour in a thermostat to acquire different temperature with an accuracy of 0.01K. The densities of all solutions were measured by a bicapillary pyknometer with deionized double distilled water with 0.9960×10^3 kg m⁻³ as its density at different temperatures. The precision of density measurement was within ±0.0003 kg m⁻³. The viscosity measurements of the solutions were carried out by using a calibrated Ostwald viscometer. The constant temperature was maintained by immersing the viscometer in a water bath maintained within ±0.01K. The ultrasonic velocity of all the solutions were measured at 303.15K, 308.15K, 313.15K temperature and atmospheric pressure by a Mittal make (India) singlecrystal variable-path ultrasonic interferometer operating at 3 MHz by circulating water from a thermostatically regulated bath maintained within ±0.01K around the sample holder for constant temperature. The velocity measurements were précised to $\pm 0.5 \text{ ms}^{-1}$.

Results

These measured values of ultrasonic velocity, density and viscosity were used to evaluate different acoustic parameters such as isentropic compressibility, β_s and intermolecular free length, L_f , using standard formulae (Franks *et al.*, 1972; Kaulgud, 1976; Kamila *et al.*, 2005). In addition, parameters like free volume, V_f , internal pressure, π_i , relaxation time, τ and relative association, R_A were calculated using following relations (Kamila *et al.*, 2017):

$$V_f = \left(M_{eff}U/K\right)^{3/2} \tag{1}$$

$$\pi_{i} = bRT \left(\frac{K}{U}\right)^{\frac{1}{2}} \frac{\rho^{2/s}}{M_{eff}^{7/6}}$$
(2)

$$\tau = 4/3\rho U^2 \tag{3}$$

$$RA = \left(\rho / \rho_0\right) \left(\frac{U_o}{U}\right)^{\frac{1}{3}} \tag{4}$$

where, *b* is the space packing factor generally 2 for liquids, *R* is the gas constant, *T* is absolute temperature and *K* is a constant equal to 4.28×10^9 , independent of temperature for all types of liquids. M_{eff} is the effective molecular mass. Similarly, Parameters like apparent molal volume, ϕ_v and apparent molal compressibility, ϕ_k can be determined by using the following formulae (Kamila and Dash, 2012; Kamila *et al.*, 2004; Nain *et al.*, 2013b; Wadi and Ramasami, 1997):

$$\phi_{v} = \frac{1000(\rho_{0} - \rho)}{c\rho\rho_{0}} + \frac{M}{\rho}$$
(5)

$$\phi_k = \frac{1000\left(\beta_s \rho_0 - \beta_s^0 \rho\right)}{c\rho\rho_0} + \frac{\beta_s M}{\rho}$$
(6)

where, ρ_0 and β_s^0 are the density and isentropic compressibility of the solvent (aqueous-lactose), ρ and β_s are those of solution, respectively. *c* is the molarity of the solution, *M* is the molar mass of the solute (zinc sulphate), n_0 and n_i are the moles of solvent and solute, respectively.

The limiting apparent molal volume ϕ_{ν}^{0} was obtained using the method of linear regression from Masson's equation (Masson, 1929) based on Debye-Huckel theory as follows (Debye and Huckel, 1923):

$$\phi_{v} = \phi_{v}^{0} + S_{v} c^{1/2} \tag{7}$$

where, ϕ_{ν}^{0} and S_{ν} are the intercept and slope in the plot of ϕ_{ν} against $c^{1/2}$. Similarly, the limiting apparent molal

compressibility, ϕ_k^0 , is obtained from Gucker's limiting law (Gucker Jr., 1933), which states ϕ_k as:

$$\phi_k = \phi_k^0 + S_k c^{1/2} \tag{8}$$

where, S_k is the slope related to ion-ion interaction and ϕ_k^0 is the intercept related to ion-solvent interaction. The slope and intercept are obtained from the plot of ϕ_k against $c^{1/2}$. Solvation number, S_n , suggested by Passynski (1938) was calculated to assess the mode of association:

$$S_n = \frac{n_0}{n_i} \left(1 - \frac{\beta_s}{\beta_s^0} \right) \tag{9}$$

where, n_0 and n_i are the moles of solvent and solute, respectively. The limiting solvation number, S_n^0 , is determined from the variation of the solvation number with molar concentration of solute. It can be calculated from the following relation:

$$\lim_{c \to 0} \phi_k^0 = -S_n^0 V_1^0 \beta_s^0$$
(10)

where, V_1^0 is the molar volume of the solvent having moles of solvent as n_1 .

The measured values of density, viscosity, ultrasonic velocity and computed values of other relevant acoustic parameters of zinc sulphate in water and different proportions of lactose-water systems at 303.15 K, 308.15 K and 313.15 K have been summarized in Table 1 to 5 whereas some parameters are graphically represented as figures.

Discussion

From the observations obtained in the present study, it reveals that the sound velocity, density and viscosity increase with the increase in the concentration of both zinc sulphate and lactose. As the concentration increases, the number of molecules in the medium increases. This makes the medium denser and reduces compressibility resulting in the increment of the sound velocity. The velocity was also found to increase with temperature indicating the structural rearrangement due to solvation (Singh and Banipal, 2008; Palani and Jayachitra, 2008). According to Thirumaran and Kannappan (2009), the increase in ultrasonic velocity with solute concentration and temperature is due to the cohesion brought about by ionic hydration. The increase in components increases the frictional resistance between the layers of the liquids and hence there is enhancement of coefficient of viscosity (Table 1). This decreases with temperature due to reduction in frictional forces among the particles.

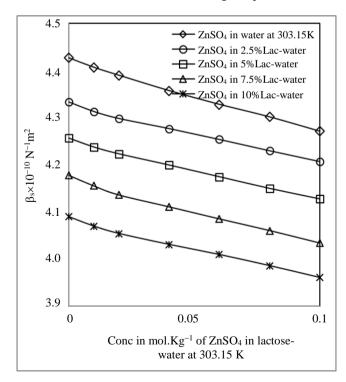


Fig. 1: Variation of isentropic compressibility, $\beta_s vs$. Concentration of ZnSO₄ in water and Lactose-water mixed solvent at 303.15K

Table 1: Experimentally determined density, ρ , viscosity, η and ultrasonic velocity, U of ZnSO₄ in water, 2.5%, 5.0%, 7.5% and 10% lactose-water mixed solvent at 303.15, 308.15 and 313.15K temperature

		ρ×10 ⁻³ kg.r			313.15K tempo η×10 ³ kg.m		U m.s ⁻¹			
C mol.kg ⁻¹	303.15K	308.15K	313.15K	303.15K	308.15K	313.15K	303.15K	308.15K	313.15K	
				ZnSO ₄	in Water					
0.00	0.9964	0.9950	0.9937	0.7975	0.7234	0.6610	1505.8	1517.1	1525.4	
0.01	0.9987	0.9973	0.9959	0.8054	0.7307	0.6663	1507.5	1517.3	1527.1	
0.02	1.0008	0.9995	0.9976	0.8091	0.7390	0.6709	1508.8	1518.7	1529.7	
0.04	1.0044	1.0024	1.0009	0.8185	0.7474	0.6801	1511.7	1522.0	1534.7	
0.06	1.0073	1.0045	1.0040	0.8285	0.7549	0.6890	1514.6	1525.1	1537.8	
0.08	1.0107	1.0070	1.0062	0.8399	0.7647	0.6956	1516.7	1528.8	1539.7	
0.10	1.0136	1.0096	1.0089	0.8490	0.7729	0.7034	1519.9	1532.1	1542.0	
			Z	nSO4 in 2.5%	6 Lactose-Wat	er				
0.00	1.0060	1.0046	1.0030	0.8482	0.7672	0.6986	1514.7	1523.2	1533.1	
0.01	1.0079	1.0066	1.0050	0.8557	0.7747	0.7057	1516.8	1525.2	1534.8	
0.02	1.0096	1.0085	1.0065	0.8624	0.7817	0.7127	1518.1	1527.0	1536.5	
0.04	1.0129	1.0117	1.0096	0.8782	0.7907	0.7214	1519.4	1530.5	1538.2	
0.06	1.0162	1.0151	1.0123	0.8884	0.8000	0.7311	1521.0	1532.1	1541.1	
0.08	1.0191	1.0184	1.0152	0.8980	0.8089	0.7395	1523.1	1534.0	1542.6	
0.10	1.0222	1.0214	1.0181	0.9133	0.8187	0.7489	1524.9	1536.0	1544.8	
			2	ZnSO ₄ in 5%	Lactose-Wate	r				
0.00	1.0151	1.0143	1.0125	0.8993	0.8118	0.7423	1521.4	1533.0	1540.9	
0.01	1.0172	1.0164	1.0145	0.9070	0.8253	0.7495	1523.2	1534.6	1542.7	
0.02	1.0192	1.0181	1.0163	0.9216	0.8335	0.7578	1524.3	1536.4	1544.3	
0.04	1.0224	1.0215	1.0199	0.9364	0.8484	0.7663	1526.2	1538.6	1547.2	
0.06	1.0259	1.0249	1.0234	0.9527	0.8641	0.7820	1528.1	1540.0	1549.2	
0.08	1.0286	1.0278	1.0266	0.9632	0.8729	0.7966	1530.6	1542.2	1551.4	
0.10	1.0309	1.0293	1.0281	0.9763	0.8867	0.8166	1533.0	1544.6	1553.6	
			Z	nSO4 in 7.5%	6 Lactose-Wat	er				
0.00	1.0238	1.0218	1.0197	0.9633	0.8678	0.7908	1529.0	1540.0	1547.0	
0.01	1.0262	1.0241	1.0220	0.9720	0.8762	0.7948	1531.4	1542.0	1548.8	
0.02	1.0284	1.0262	1.0242	0.9865	0.8845	0.8077	1533.3	1543.8	1550.2	
0.04	1.0323	1.0302	1.0284	1.0028	0.8938	0.8285	1535.1	1544.9	1553.0	
0.06	1.0353	1.0341	1.0322	1.0174	0.9088	0.8380	1537.7	1547.2	1554.7	
0.08	1.0385	1.0369	1.0352	1.0279	0.9186	0.8475	1540.0	1549.2	1557.6	
0.10	1.0416	1.0391	1.0376	1.0500	0.9346	0.8572	1542.8	1552.4	1560.0	
					Lactose-Wate					
0.00	1.0327	1.0311	1.0302	1.0411	0.9324	0.8434	1538.8	1546.1	1555.9	
0.01	1.0356	1.0334	1.0325	1.0497	0.9408	0.8517	1540.4	1548.2	1557.6	
0.02	1.0370	1.0356	1.0345	1.0585	0.9494	0.8601	1542.3	1549.7	1559.1	
0.04	1.0404	1.0395	1.0383	1.0743	0.9654	0.8754	1544.2	1553.1	1562.0	
0.06	1.0435	1.0421	1.0413	1.0892	0.9771	0.8843	1546	1555.3	1563.8	
0.08	1.0468	1.0455	1.0446	1.1064	0.9902	0.8999	1548.1	1557.4	1565.6	
0.10	1.0495	1.0477	1.0467	1.1235	1.0018	0.9164	1551.0	1559.0	1569.0	

compressibility, βs , an Isentropic acoustic parameter, derived from the velocity data is known to be a sensitive indicator of the elasticity as well as of the molecular interactions of the system (Katrinak et al., 2012). In the present study, the compressibility decreases as the concentration of both zinc sulphate and lactose increases (Fig. 1). When the solute ion enters into the bulk of the solvent, the solvent molecules are attracted towards this ion because of electrostrictive force resulting in the decrease of solvent molecules for the incoming ions. This condition of the system is compression. Therefore, compressibility decreases with the increase in volumetric concentration of the solution. Moreover, there is breakage in the molecular structure of water with the addition of solute and making a more compact structure with the water molecules surrounded by the solute in mixed solvent. Santosh *et al.* (2011) also have reported similar type of observations and which was depicted as the existence of strong solute– solvent interactions through dipole–dipole and acceptor-donor interactions (Rao and Verrall, 1987; Nikam *et al.*, 2000). This behaviour increases the inter molecular association and decreases βs (Savaroglu and Ozdemir, 2008; Fort and Moore, 1965). There is also decrease in compressibility with temperature due to thermal expansion of the liquid systems (Fig. 2).

This compactness among the particles also reduces the intermolecular free length. According to literature, such type of behaviour is indicative of structurepromoting tendency of added solute as well as presence of significant ion-solvent interaction (Dash and Kamila, 2017; Kannappan and Vinayagam, 2007).

The apparent molal compressibility shows negative variation through out the concentration range indicating specific interaction involving ion-solvent type (Fig. 3 and 4) (Palani *et al.*, 2008). According to Dhanalakshmi and Vasantharani (1999), the negative values of apparent compressibility are indicative of electrostrictive solvation of ions. The limiting apparent compressibility ϕ_k^0 is the compressibility of solute at infinite dilution and it reflects the effects of solute-solvent interactions. The negative variation of this parameter indicates ion-

solvent interaction and positive values of S_k represent ion-ion interaction (Pal and Chauhan, 2011).

The dissociation occurs during the mixing of components resulting in the increases of free volume whereas association between the components will reduce the free volume. So the decreased values of free volume with the lactose concentration in the present study (Fig. 5) suggests the association due to Hbonding with the water molecules (Table 2). Again with the increase in solute concentration the strength of interaction among the components increases and as a result there would be decrease in free volume.

Table 2: Calculated values of isentropic compressibility, β_s , intermolecular free length, L_f, free volume, V_f, of ZnSO₄ in water, 2.5%, 5.0%, 7.5% and 10% lactose-water mixed solvent at 303.15, 308.15 and 313.15K temperature

C mol.kg ⁻¹					$L_f \times 10^{11} M$		$V_{\rm f} \times 10^8 {\rm m}^3.{ m mol}^{-1}$			
	303.15K	308.15K	313.15K	303.15K	308.15K	313.15K	303.15K	308.15K	313.15K	
				ZnSO ₄ i	in Water					
0.00	4.4262	4.3667	4.3249	4.3655	4.3778	4.3984	2.2407	2.6228	3.0276	
0.01	4.4061	4.3554	4.3058	4.3556	4.3722	4.3887	2.2205	2.5947	3.0087	
0.02	4.3892	4.3378	4.2838	4.3472	4.3634	4.3775	2.2171	2.5650	2.9976	
0.04	4.3567	4.3066	4.2419	4.3311	4.3476	4.3560	2.2030	2.5507	2.9754	
0.06	4.3276	4.2801	4.2118	4.3166	4.3342	4.3405	2.1871	2.5410	2.9507	
0.08	4.3011	4.2488	4.1922	4.3034	4.3184	4.3304	2.1646	2.5218	2.9380	
0.10	4.2707	4.2197	4.1685	4.2881	4.3035	4.3182	2.1535	2.5096	2.9187	
				nSO4 in 2.5%	Lactose-Wate					
0.00	4.3326	4.2904	4.2419	4.3191	4.3394	4.3560	2.1361	2.5043	2.9104	
0.01	4.3125	4.2706	4.2241	4.3090	4.3294	4.3469	2.1211	2.4829	2.8830	
0.02	4.2978	4.2525	4.2084	4.3017	4.3202	4.3388	2.1076	2.4639	2.8569	
0.04	4.2765	4.2197	4.1862	4.2910	4.3035	4.3274	2.0703	2.4501	2.8330	
0.06	4.2537	4.1968	4.1594	4.2796	4.2918	4.3135	2.0545	2.4308	2.8075	
0.08	4.2299	4.1728	4.1394	4.2676	4.2796	4.3031	2.0423	2.4147	2.7864	
0.10	4.2071	4.1497	4.1159	4.2561	4.2677	4.2908	2.0105	2.3949	2.7617	
			Z	2nSO ₄ in 5%	Lactose-Wate	r				
0.00	4.2560	4.1952	4.1596	4.2808	4.2910	4.3136	2.0420	2.4083	2.7760	
0.01	4.2372	4.1778	4.1418	4.2713	4.2821	4.3043	2.0277	2.3626	2.7520	
0.02	4.2228	4.1610	4.1259	4.2640	4.2735	4.2960	1.9899	2.3414	2.7221	
0.04	4.1991	4.1353	4.0959	4.2520	4.2603	4.2804	1.9624	2.3034	2.7063	
0.06	4.1744	4.1141	4.0714	4.2395	4.2494	4.2676	1.9313	2.2622	2.6517	
0.08	4.1498	4.0908	4.0472	4.2270	4.2373	4.2549	1.9201	2.2511	2.6056	
0.10	4.1276	4.0722	4.0298	4.2157	4.2276	4.2457	1.9011	2.2217	2.5362	
			Zı	nSO4 in 7.5%	Lactose-Wate	er				
0.00	4.1780	4.1266	4.0978	4.2413	4.2558	4.2814	1.9243	2.2754	2.6342	
0.01	4.1552	4.1067	4.0790	4.2297	4.2455	4.2716	1.9106	2.2561	2.6294	
0.02	4.1360	4.0887	4.0629	4.2200	4.2362	4.2631	1.8796	2.2373	2.5804	
0.04	4.1107	4.0670	4.0318	4.2071	4.2250	4.2468	1.8520	2.2226	2.5107	
0.06	4.0850	4.0397	4.0081	4.1939	4.2107	4.2343	1.8317	2.1902	2.4922	
0.08	4.0602	4.0184	3.9817	4.1811	4.1996	4.2203	1.8225	2.1771	2.4773	
0.10	4.0335	3.9933	3.9602	4.1673	4.1865	4.2089	1.7841	2.1452	2.4607	
					Lactose-Wate					
0.00	4.0894	4.0572	4.0097	4.1961	4.2198	4.2351	1.7936	2.1318	2.5019	
0.01	4.0695	4.0372	3.9921	4.1859	4.2094	4.2258	1.7812	2.1160	2.4793	
0.02	4.0540	4.0208	3.9767	4.1779	4.2009	4.2177	1.7696	2.0987	2.4564	
0.04	4.0308	3.9882	3.9474	4.1660	4.1838	4.2021	1.7480	2.0700	2.4184	
0.06	4.0095	3.9670	3.9270	4.1549	4.1727	4.1912	1.7293	2.0541	2.4057	
0.08	3.9860	3.9434	3.9056	4.1427	4.1603	4.1798	1.7063	2.0340	2.3666	
0.10	3.9609	3.9271	3.8809	4.1297	4.1516	4.1665	1.6856	2.0181	2.3293	

Table 3: Calculated values of Internal pressure, π_i , viscous relaxation time, τ , relative association, R_A of ZnSO₄ in water, 2.5%, 5.0%, 7.5% and 10% lactose-water mixed solvent at 303.15, 308.15 and 313.15K temperature

		$\pi_I \times 10^9 Pa$			$\tau \times \! 10^{13} s$		RA				
C mol.kg ⁻¹	303.15K	308.15K	313.15K	303.15K	308.15K	313.15K	303.15K	308.15K	313.15K		
				ZnSO ₄	in Water						
0.00	2.5955	2.5010	2.4208	4.7065	4.2118	3.8117					
0.01	2.6027	2.5094	2.4250	4.7315	4.2434	3.8252	1.0019	1.0018	1.0016		
0.02	2.6029	2.5182	2.4264	4.7351	4.2742	3.8320	1.0037	1.0037	1.0027		
0.04	2.6053	2.5187	2.4290	4.7547	4.2916	3.8466	1.0067	1.0059	1.0049		
0.06	2.6073	2.5163	2.4320	4.7805	4.3081	3.8692	1.0090	1.0073	1.0074		
0.08	2.6128	2.5178	2.4303	4.8166	4.3321	3.8881	1.0119	1.0090	1.0091		
0.10	2.6131	2.5173	2.4314	4.8345	4.3485	3.9095	1.0141	1.0109	1.0114		
			Z	nSO4 in 2.5%	Lactose-Wate	er					
0.00	2.6121	2.5158	2.4290	4.8999	4.3887	3.9512					
0.01	2.6169	2.5218	2.4356	4.9202	4.4113	3.9746	1.0014	1.0016	1.0016		
0.02	2.6207	2.5269	2.4409	4.9419	4.4323	3.9991	1.0028	1.0030	1.0027		
0.04	2.6326	2.5279	2.4440	5.0075	4.4487	4.0266	1.0058	1.0055	1.0055		
0.06	2.6355	2.5311	2.4468	5.0386	4.4766	4.0546	1.0087	1.0085	1.0075		
0.08	2.6363	2.5331	2.4488	5.0646	4.5005	4.0815	1.0112	1.0114	1.0101		
0.10	2.6462	2.5361	2.4521	5.1231	4.5299	4.1099	1.0138	1.0139	1.0125		
					Lactose-Wate						
0.00	2.6253	2.5244	2.4436	5.1033	4.5408	4.1169					
0.01	2.6303	2.5395	2.4495	5.1242	4.5972	4.1390	1.0017	1.0017	1.0016		
0.02	2.6456	2.5454	2.4569	5.1890	4.6243	4.1688	1.0034	1.0030	1.0030		
0.04	2.6539	2.5558	2.4587	5.2427	4.6779	4.1849	1.0061	1.0059	1.0059		
0.06	2.6646	2.5676	2.4722	5.3026	4.7400	4.2451	1.0092	1.0089	1.0090		
0.08	2.6648	2.5674	2.4829	5.3295	4.7612	4.2986	1.0113	1.0113	1.0116		
0.10	2.6682	2.5720	2.4988	5.3731	4.8144	4.3877	1.0130	1.0122	1.0126		
					Lactose-Wate						
0.00	2.6499	2.5437	2.4582	5.3662	4.7748	4.3207					
0.01	2.6556	2.5502	2.4590	5.3851	4.7977	4.3227	1.0018	1.0018	1.0019		
0.02	2.6692	2.5562	2.4736	5.4403	4.8219	4.3755	1.0036	1.0035	1.0037		
0.04	2.6796	2.5594	2.4942	5.4963	4.8468	4.4538	1.0070	1.0072	1.0072		
0.06	2.6850	2.5692	2.4976	5.5414	4.8950	4.4784	1.0093	1.0105	1.0106		
0.08	2.6854	2.5697	2.4983	5.5647	4.9217	4.4993	1.0119	1.0128	1.0129		
0.10	2.7004	2.5768	2.4989	5.6469	4.9762	4.5263	1.0143	1.0142	1.0147		
0110	2.7001	210700			Lactose-Wate		110110	110112	110117		
0.00	2.6845	2.5731	2.4774	5.6767	5.0439	4.5091					
0.01	2.6911	2.5788	2.4842	5.6957	5.0642	4.5334	1.0025	1.0018	1.0019		
0.02	2.6945	2.5850	2.4907	5.7215	5.0898	4.5605	1.0034	1.0036	1.0035		
0.04	2.7017	2.5941	2.5008	5.7737	5.1336	4.6074	1.0063	1.0066	1.0065		
0.04	2.7070	2.5956	2.5009	5.8228	5.1682	4.6302	1.0089	1.0087	1.0000		
0.08	2.7150	2.6004	2.5109	5.8802	5.2064	4.6862	1.0116	1.0115	1.0119		
0.10	2.7211	2.6015	2.5185	5.9334	5.2455	4.7419	1.0136	1.0133	1.0132		

The apparent molal volume, ϕ_v shows positive value and increasing trends with lactose content in the solution (Fig. 6 and 7). Similarly, the limiting apparent molal volume, ϕ_v^0 provides information about ion-solvent interactions, which shows positive variation. Positive variations for both ϕ_v and ϕ_v^0 (Table 4 and 5) are indicative of strong solute-solvent interactions in the systems (Parmar and Thakur, 2006; Rathore and Singh, 2006; Ali *et al.*, 2002). Pinto *et al.* (2015) and Naseem and Ashraf (2016) have also reported similar type of observation for their systems. The constant S_v , is indicative of ion-ion interaction and the values are positive but smaller in comparison to ϕ_v^0 . According to Ali and Shahjahan (2016) such type of variations are due to weak ion-ion interactions.

The parameter, internal pressure is the cohesive force, which is due to the attractive and repulsive forces between the molecules (Kannappan *et al.*, 2009). In the present systems, there is increase in internal pressure with the concentration of both $ZnSO_4$ as well as lactose (Fig 8). This can be explained due to the strengthening of cohesive forces obtained during the structure making of solvents in presence of solute (Suryanarayan and Kuppu Samy, 1981). This force, however decreases with the increase in temperature because of dispersion of solute molecules (Table. 3).

Table 4: Calculated values of apparent molal volume, ϕ_{v} , apparent molal compressibility, ϕ_K , solvation number, S_n of ZnSO₄ in water, 2.5%, 5.0%, 7.5% and 10% lactose-water mixed solvent at 303.15, 308.15 and 313.15K temperature

		$\phi_v \times 10^3 dm$	n^3 .mol ⁻¹		$-\phi_{\rm K} \times 10^{10} { m N}$	J ⁻¹ .dm ⁵ .mol ⁻¹	S _n			
C mol.kg ⁻¹	303.15K	308.15K	313.15K	303.15K	308.15K	313.15K	303.15K	308.15K	313.15K	
				ZnSO ₄	in Water					
0.00	5 4 50		<i>cc</i> 11	1 55 1	2 0 2 2	2 275	25.1256	22 52 15	20.2004	
0.01	56.78	56.53	66.41	1.771	2.032	2.375	25.1256	23.7347	29.3004	
0.02	66.69	61.44	91.52	1.561	1.756	2.042	23.0942	22.9536	28.6195	
0.04	86.43	101.36	106.30	1.366	1.361	1.820	21.6402	21.2529	27.5655	
0.06	104.45	127.83	114.32	1.197	1.094	1.538	20.4054	19.6480	24.6731	
0.08	106.99	135.83	129.49	1.109	1.046	1.218	19.3730	19.5751	21.5296	
0.10	113.37	139.46	133.38	1.075	1.003	1.091	19.3124	19.3362	20.2581	
0.00			Z	nSO ₄ in 2.5%	Lactose-Wat	er				
0.00										
0.01	97.90	87.87	87.70	1.578	1.585	1.397	25.2809	24.9889	22.6716	
0.02	107.58	92.64	112.33	1.265	1.486	1.190	21.8511	23.9419	21.3532	
0.04	114.59	109.57	121.86	0.903	1.294	0.874	17.5687	22.3002	17.7248	
0.06	116.66	111.65	131.38	0.811	1.082	0.822	16.4294	19.6393	17.4554	
0.08	122.40	113.73	133.46	0.758	0.987	0.723	15.9940	18.4660	16.2233	
0.10	123.75	117.78	134.55	0.726	0.910	0.701	15.6774	17.7222	15.9961	
			Z	ZnSO4 in 5%	Lactose-Wate	r				
0.00										
0.01	79.30	79.20	88.75	1.515	1.377	1.394	23.7169	22.1231	22.9116	
0.02	83.97	98.43	98.28	1.281	1.269	1.260	20.9153	21.7693	21.6570	
0.04	105.39	107.76	102.77	0.958	1.027	1.151	17.8755	19.0415	20.4225	
0.06	107.43	110.61	105.64	0.891	0.875	1.022	17.0482	17.1507	18.8050	
0.08	117.92	117.89	110.52	0.818	0.802	0.940	16.5900	16.5159	17.9279	
0.10	127.93	135.67	129.81	0.736	0.659	0.758	16.0750	15.5836	16.5781	
			Zi	nSO4 in 7.5%	Lactose-Wat	er				
0.00										
0.01	51.76	60.97	60.64	2.013	1.701	1.582	28.8530	25.4406	24.0218	
0.02	61.15	70.38	65.30	1.796	1.566	1.439	26.5470	24.1978	22.3282	
0.04	77.47	79.61	72.19	1.323	1.133	1.325	21.2278	18.9912	21.1066	
0.06	96.90	84.04	80.63	1.118	1.078	1.140	19.4972	18.4219	19.0898	
0.08	104.05	99.15	94.21	1.015	0.925	1.047	18.4746	17.1513	18.4840	
0.10	109.13	113.78	107.94	0.971	0.849	0.920	18.1764	16.9314	17.5600	
			Z	nSO4 in 10%	Lactose-Wate	er				
0.00										
0.01	64.92	62.39	62.25	1.900	1.680	1.462	25.3409	25.4828	22.7820	
0.02	76.51	66.94	76.21	1.403	1.490	1.299	22.4961	23.2169	21.3307	
0.04	97.20	80.68	87.62	1.026	1.348	1.165	18.5729	21.9901	20.0928	
0.06	108.51	105.30	103.68	0.854	1.038	0.930	16.8327	19.1039	17.7268	
0.08	111.64	108.05	108.00	0.806	0.951	0.841	16.2965	18.0373	16.6933	
0.10	118.97	120.78	121.69	0.773	0.789	0.778	16.2361	16.5242	16.5473	

 Table 5: Acoustic Parameters of ZnSO4 in water and different proportions of lactose water mixture at 303.15, 308.15 and 313.15K temperature

	$\phi_{v}^{0} \times 10^{3} (\mathrm{dm^{3}.mol^{-1}})$			$-\phi_k^0 \times 10^9 (N^{-1}.dm^5.mol^{-1})$			S_v			$S_k \times 10^9 (N^{-1}.dm^{13/2}.mol^{-3/2})$		
Solvent	303.15K	308.15K	313.15K	303.15K	308.15K	313.15K	303.15K	308.15K	313.15K	303.15K	308.15K	
ZnSO ₄ in Water	275.5	436.0	295.6	3.264	4.962	5.891	30.08	10.31	43.55	2.045	2.445	2.943
2.5% Lactose-Water	114.1	141.5	201.6	3.880	3.293	3.246	89.35	75.21	77.01	1.838	1.929	1.646
5% Lactose-Water	224.2	218.7	153.3	3.509	3.33	2.722	55.60	61.39	73.11	1.785	1.714	1.670
7.5% Lactose-Water	282.4	224.3	209.1	5.102	4.083	2.997	22.89	36.59	35.34	2.465	2.083	1.884
10% Lactose-Water	252.9	283.5	262.8	5.012	4.104	3.253	42.10	29.93	36.93	2.200	2.095	1.776

In the present study, the relaxation time, τ , is in the order of 10^{-13} and according to Kinsler and Rray (1989) this is due to structural relaxation process, which again suggests these molecules to be rearranged due to supportive process (Sawhney *et al.*, 2014) (Fig. 9). The relative association, R_A is the parameter that depends on the structure breaking

behaviour of solute when is added to solvent and the solvation of ions by the free solvent molecules (Palani and Geeta, 2007). However, the increase in R_A is associated with the later effect as per the literature (Ali *et al.*, 2001). Therefore in the present investigation, the solvation of solute predominates over the breaking of the solvent

aggregates. Jahagirdar and Shankarwar (2000) have also reported similar kind of observations.

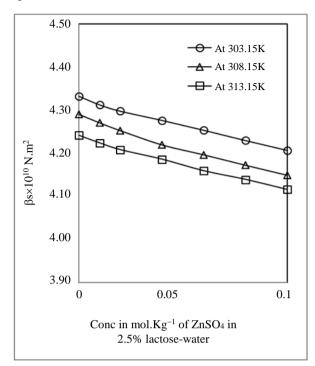


Fig. 2: Variation of isentropic compressibility, β_s of ZnSO₄ in 2.5% Lactose-water *vs.* temperature

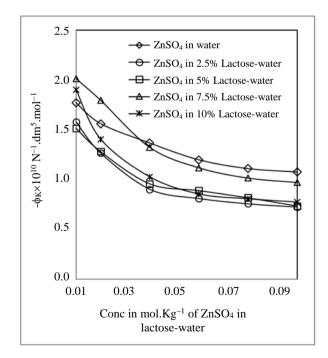


Fig. 3: Variation of apparent molal compressibility, φk vs. Concentration of ZnSO₄ in water and Lactose-water mixed solvent at 303.15K

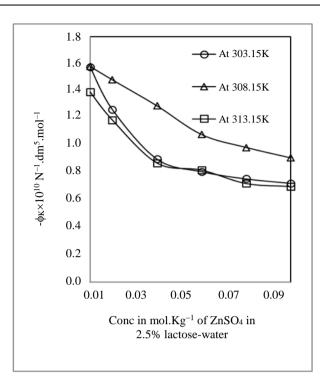


Fig. 4: Variation of apparent molal compressibility, ϕ_k of ZnSO₄ in 2.5% Lactose-water *vs*. Temperature

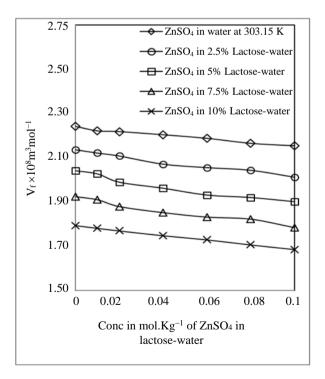


Fig. 5: Variation of free volume, V_f vs. Concentration of ZnSO₄ in water and Lactose-water mixed solvent at 303.15K

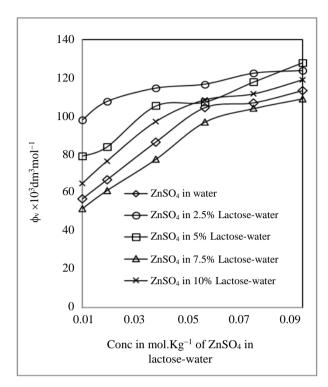


Fig. 6: Variation of apparent molal volume, ϕ_v vs. Concentration of ZnSO₄ in water and Lactose-water mixed solvent at 303.15K

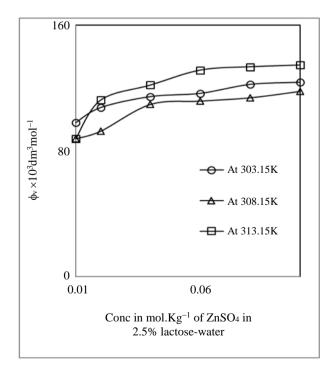


Fig. 7: Variation of apparent molal volume, ϕ_v of ZnSO₄ in 2.5% Lactose-water vs temperature

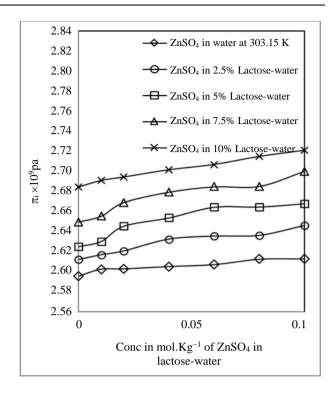


Fig. 8: Variation of internal pressure, π_i vs. Concentration of ZnSO₄ in water and Lactose-water mixed solvent at 303.15 K

Solvation number explains the interaction of solute with the solvent molecules present in the solutions. The positive values of solvation number in the present study indicate appreciable solvation of solutes (Mehraand and Sajnami, 2000). This is indicative of structure-promoting nature of soulte and the positive values also suggest the compressibility to be less in solution in comparison to solvent. This increases the ion mobility and so also the probability to interact with the solvent molecules. The solvent molecules are attached to the ions by strong covalent bonds in the primary sheath of solvation and in the secondary sheath, there are weak forces of attraction between solute and solvent species (Dash and Kamila, 2017). In the present investigation, the solvation number decreases as the concentration of solute increases (Fig. 10). This is attributed to the decrease in the size of the secondary sheath of solvation. More over, S_n decreases with increase in temperature, which shows weakening of solute-solvent interaction (Fig. 11). According to Marcus (2005), this number depends on both the ion and solvent involved as well as on the concentration through the interactions of this ion with other ions. However, the non-linear variation of solvation number with ZnSO₄, indicates the increase in ion-solvent interactions (Palani et al., 2001).

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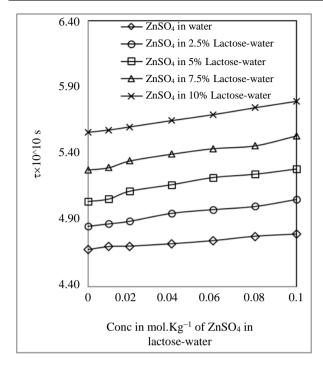


Fig. 9: Variation of viscus relaxation time, τvs . Concentration of ZnSO₄ in water and Lactose-water mixed solvent at 303.15 K

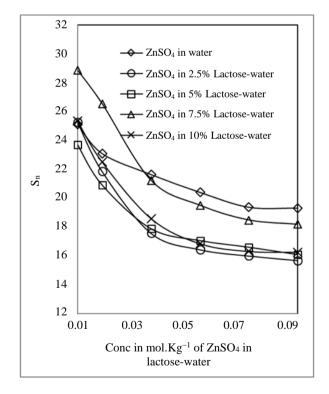


Fig. 10: Variation of Solvation number, Sn vs. Concentration of ZnSO4 in water and Lactose-water mixed solvent at 303.15 K

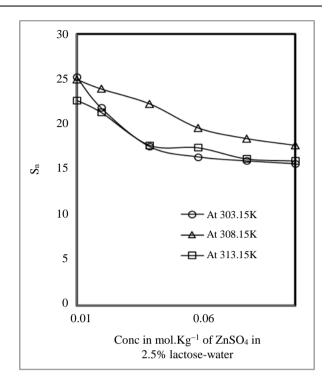


Fig. 11: Variation of Solvation number, S_n of ZnSO₄ in 2.5% Lactose-water vs. temperature

Conclusion

Acoustic studies involving ultrasonic velocity measurement are non-destructive techniques to access the molecular association among the components through ion-solvation. Besides, different other supporting parameters such as isentropic compressibility, internal pressure and apparent molal volume etc., have been evaluated at three different temperatures. The present study reports the existence of solute-solvent and solute-solute interaction between ZnSO₄ and aqueous lactose solution. From the Results and discussion, it reveals the structure-promoting tendency of added solute. The electrostrictive solvation of ions is supported by partial molal volume. The breaking of solvent structure was also confirmed from internal pressure as well as relative association, where the solvation of solute predominates over the breaking of the solvent aggregates. S_V and S_K values show weak ion-ion interaction in the systems. The solvation number indicates appreciable ion solvation along with structure promoting nature of solute. However, the ion solvation decreases as the temperature increases. So overall, it can be concluded that the strength of interaction increases with increase in lactose as well as ZnSO₄ concentration. The decrease in solvation number with the increase in lactose concentration is indicative of the dehydrating effect of co-solute on ZnSO₄. This result would be of great importance to pharmaceutical industries for the

drugs related to lactose and ZnSO₄ for designing and delivery systems.

Acknowledgement

One of the authors (GDN) is thankful to Visvesvaraya Technological University (VTU), Belagavi, Karnataka for giving the opportunity to work for his Ph.D. programme.

Author's Contributions

Ganesh Durgachalam Natraj: Ganesh is the first author and did all the experimental work along with data collection.

Vinay Sathyanarayan Bhat: Vinay's contributions involve the evaluation and calculation part of different parametrs.

Susmita Kamila: Susmita is the supervisor and the corresponding author. Her contribution includes Research planning, organizational designing and framework of the manuscript.

Conflict of Interest

The authors declares that they have no conflict of interest.

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