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#### **RESEARCH ARTICLE**

## Ultrasonic studies on molecular interactions in strong electrolytes-Metal chlorides in aqueous medium at different temperatures and 2MHz frequency

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#### Abstract

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#### Key words:

Apparent isentropic compressibility, Relative association, Solvation number, Ultrasonic attenuation, Gibb's free energy

\*Corresponding Author Bidyadhar Swain. Values of ultrasonic velocity (U), density (d) and viscosity  $(\eta)$  have been measured in aqueous solutions of magnesium chloride, zinc chloride, strontium chloride, cadmium chloride and barium chloride in different concentrations over the temperature range 298.15K to 313.15K at 5K interval. The thermo-acoustical parameters, such as acoustic impedance (Z), isentropic compressibility (K<sub>s</sub>), apparent isentropic compressibility (K<sub>s, $\phi$ </sub>), intermolecular free length ( $L_f$ ), relative association ( $R_A$ ), relaxation time ( $\tau$ ), Gibb's free energy change ( $\Delta G$ ), solvation number (S<sub>n</sub>), ultrasonic attenuation  $(\alpha/f^2)$ , internal pressure  $(\pi_i)$ , free volume  $(V_f)$  and van der Walls constant (b) have been computed to assess the ion-solvent and ion-ion interactions in these solutions. It is found that the ion-solvent and ion-ion interactions depend on concentration, temperature, ionic size, ionic field strength and nature of the ion. The structural arrangement of molecules in electrolyte solutions has been discussed on the basis of electrostatic field (ionic field) of ion. The qualitative intermolecular elastic forces between the solute and solvent molecules are explained in terms of compressibility. The variation of solvation number with respect to temperature and concentration of electrolyte solution has been explained in the light of dipolar interaction between solute and solvent.

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#### Introduction:-

Ultrasonic method is a versatile non-destructive technique, which provides useful information for understanding the physico-chemical characteristics and the structural properties of electrolyte solutions (Johnson et al., 2001, Osamu et al., 1978). Since ultrasonic wave is low amplitude and high frequency wave, due to its penetrating nature through liquid medium, it is of interest to study its interaction with matter to throw light into the molecular interaction and molecular kinetic properties of the materials. Hence, the bulk properties of solutions like density, viscosity and ultrasonic velocity provides insight into the intermolecular arrangement of the components in solutions and assists to understand the thermo-acoustics and thermodynamics properties of the electrolyte solutions (Ranga Nayakulu et al., 2005, Kannappan et al., 2005, Ravichandran et al., 2010). It is desirable that any discussion about thermo-acoustical and thermodynamic parameters of electrolytes is connected to ions as ions are playing important role in an electrolyte solution. The effect of concentrations and temperatures of electrolyte solutions help to understand ion-ion (electrostatic) and ion-solvent (solvation) interactions.

In continuous of our previous work in aqueous medium (Swain et al., 2016), the present investigation aims at studying various thermo-acoustical parameters, such as acoustic impedance (Z), isentropic compressibility ( $K_{s,\phi}$ ), apparent isentropic compressibility ( $K_{s,\phi}$ ), intermolecular free length ( $L_f$ ), solvation number ( $S_n$ ), relative association ( $R_A$ ), relaxation time ( $\tau$ ), Gibb's free energy change ( $\Delta G$ ), ultrasonic attenuation ( $\alpha/f^2$ ), internal pressure ( $\pi_i$ ), free

volume ( $V_f$ ) and van der Walls constant (b) in different concentrations and at different temperatures ranging from 298.15K to 313.15K at 5K interval to examine the ion-ion and ion-solvent interactions in the aqueous solutions of magnesium chloride, zinc chloride, strontium chloride, cadmium chloride and barium chloride, which have wide applications in pharmaceutical, medicinal, agricultural, environmental, industry, etc.

#### Materials and methods:-

All electrolytes like MgCl<sub>2</sub>.6H<sub>2</sub>O (Merck, India), ZnCl<sub>2</sub> (Merck, India), SrCl<sub>2</sub>.6H<sub>2</sub>O (Merck, India), CdCl<sub>2</sub> (Merck, Germany) and BaCl<sub>2</sub>.2H<sub>2</sub>O (Merck, India) used were of GR or AR grades and dried over anhydrous CaCl<sub>2</sub> in desiccator before use. All solutions were prepared in conductivity water (Sp. cond. ~10<sup>-6</sup> S.cm<sup>-1</sup>). The solutions were prepared on the molal basis and conversion of molality to molarity was done by using standard expression (Robinson et al., 1955) and using the density data at the corresponding temperature. The solute content of the solutions varied over a concentration range of  $6.0 \times 10^{-3}$  to  $8.0 \times 10^{-2}$  mol.dm<sup>-3</sup> for all measurements.

#### Ultrasonic velocity measurement:-

Ultrasonic velocity measurements in the electrolyte solutions at different concentrations were made by using a multi frequency ultrasonic interferometer (M-84, Mittal enterprises, New Delhi, India) operating at a frequency of 2MHz at different temperatures ranging from 298.15K to 313.15K at 5K intervals.

The measuring cell of interferometer is a specially designed double-walled vessel with provision for temperature constancy. A constant temperature water bath (TH-012, Thermotech, India) with an accuracy of  $\pm 0.05$ K was used to circulate water through the outer jacket of double-walled measuring cell containing the experimental solution. The cell was allowed to equilibrate for 30 minutes prior to making the ultrasonic velocity measurements. The interferometer was calibrated with water and the experimental value is in good agreement with literature value (Jahagirdar et al.,  $2000_a$ ). The precision of the ultrasonic velocity measurements was within  $\pm 0.5$  ms<sup>-1</sup>.

#### **Density measurement:-**

The densities of solutions were measured (Swain et al.,  $2015_a$ ) by relative measurement method using a specific gravity bottle (25ml capacity, Borosil glass, India) and digital balance (PGB 200, WENSAR, India) with an accuracy of  $\pm 10^{-3}$ gm. At least five observations were taken and the differences in any two readings did not exceed  $\pm 0.02\%$ . Sufficient care was taken to avoid any entrapment of air bubble.

#### Viscosity measurement:-

Viscosity measurements were made (Swain et al.,  $2015_b$ ) using an Ostwald viscometer (10ml capacity, Borosil glass, India) in a water thermostat whose temperature was controlled to  $\pm 0.05$ K and efflux time was determined using a digital stop clock (RACER, India) with an accuracy of  $\pm 0.01$ s. An average of three sets of flow times for each solution was taken for the calculation of viscosity. The values of viscosity so obtained were accurate to within  $\pm 0.3 \times 10^{-3}$  cP.

#### **Theoretical aspects:-**

The experimentally measured values of ultrasonic velocity (U), density (d) and viscosity ( $\eta$ ) in aqueous solutions of magnesium chloride, zinc chloride, strontium chloride, cadmium chloride and barium chloride at different temperatures have been used to compute the values of different parameters, such as acoustic impedance (Z), isentropic compressibility (K<sub>s</sub>), apparent isentropic compressibility (K<sub>s, $\phi$ </sub>), intermolecular free length (L<sub>f</sub>), solvation number (S<sub>n</sub>), relative association (R<sub>A</sub>), relaxation time ( $\tau$ ), Gibb's free energy change ( $\Delta G$ ), ultrasonic attenuation ( $\alpha/f^2$ ), internal pressure ( $\pi_i$ ), free volume (V<sub>f</sub>) and van der Walls constant (b) from the following relations (Moharatha et al., 2011, Palani et al., 2010, Das et al., 2013)

$$Z = Ud \tag{1}$$
$$K_s = 1/U^2 d \tag{2}$$

$$\mathbf{K}_{s,\Phi} = 1000 \,\mathbf{K}_s \, c^{-1} - \mathbf{K}_s^0 \, d_0^{-1} (1000 \, c^{-1} \, d - M) \quad (3)$$

$$L_f = \mathbf{K}_T \sqrt{\mathbf{K}_s} \tag{4}$$

$$S_n = n_1 n_2^{-1} \left( 1 - \mathbf{K}_s / \mathbf{K}_s^0 \right)$$
 (5)

$$R_A = (d/d_0) (U_0/U)^{1/3}$$
(6)

$$\tau = 4\eta/3U^2d\tag{7}$$

$$\Delta G = k_B T \ln \left( k_B T \tau / h \right) \tag{8}$$

$$\alpha / f^2 = 4\pi^2 \tau / 2U \tag{9}$$

$$\pi_i = b' RT \left( K\eta / U \right)^{1/2} \left( d^{2/3} / M_{eff}^{7/6} \right)$$
(10)

$$V_f = \left( M_{eff} U / K \eta \right)^{3/2} \tag{11}$$

$$b = \frac{M_{eff}}{d} \left[ 1 - \left( \frac{RT}{M_{eff} U^2} \right) \left\{ \left( 1 + \frac{M_{eff} U^2}{3RT} \right)^{1/2} - 1 \right\} \right]$$
(12)

where  $M_{eff} = \sum m_i x_i$  is the effective molecular weight,  $m_i$  is the effective molecular weight of the individual constituent,  $x_i$  is the mole fraction of the individual constituent,  $K_T$  is the temperature dependent Jacobson's constant {(93.875+0.375T)x10<sup>-8</sup>}, M is the molecular mass of the solute, c is the molar concentration,  $d_o$  and d are the densities of pure solvent and solution, respectively,  $U_0$  and U are the velocities of pure solvent and solution, respectively,  $n_1$  and  $n_2$  are the number of moles of solvent and solute, respectively,  $K_s$  and  $K_s^0$  are the isentropic compressibility of the solution and solvent, respectively, b' is the cubic packing factor having value '2' for all liquids, T is the temperature in Kelvin,  $\alpha$  is the attenuation co-efficient and 'f' is the frequency of ultrasonic wave (2MHz),  $k_B$  is the Boltzmann's constant (1.38x10<sup>-23</sup>JK<sup>-1</sup>), h is the Planck's constant (6.626x10<sup>-34</sup>Js), R is the Universal gas constant (8.3143JK<sup>-1</sup>.mol<sup>-1</sup>), K is the dimensionless constant (4.281x10<sup>9</sup>) independent of temperature and nature of liquid.

#### **Results and discussions:-**

As previously (Swain et al., 2016), the sound velocity (U) increases with concentration of aqueous solutions of magnesium chloride, zinc chloride, strontium chloride, cadmium chloride and barium chloride. It also increases with increase in temperature. Typical plot of ultrasonic velocity versus concentration of magnesium chloride electrolyte at different temperatures are shown in Fig. 1.

As observed, the values of sound velocity in aqueous solutions of all the metal chlorides are in the order:  $BaCl_2 > CdCl_2 > ZnCl_2 > MgCl_2 > SrCl_2$  at all temperatures. The values of U were fitted to an equation of the form,  $U = U_0 + A'c + B'c^{3/2} + C'c^2$  (13)

where  $U_0$  is the sound velocity in water, c is the molar concentration, and A', B' and C' are the empirical constants. These constants are given in Table 1. The variation of  $(U-U_0)/c$  vs  $c^{1/2}$  are parabolic for lower concentration and decreases linearly for the higher concentration in aqueous solutions of all the metal chlorides at all temperatures agreeing fairly well with the Eq. (13). Typical plots are shown in Fig. 2 over the concentration range at 298.15K.

From Table 2, it is observed that the acoustic impedance (Z), which assesses the absorption of sound wave in a medium and determines the elastic behaviour (i.e., the bulk modulus of elasticity) of the medium increases with increase in concentration of electrolytes (Moharatha et al., 2011). This is well agreeing with the theoretical requirement as density and ultrasonic velocity increase with increase in the concentration of electrolytes. The increase in Z values with concentration of electrolytes at all given temperatures may be attributed to the effective solute-solvent interactions. The Z values also increase with increase in temperature due to structural properties of electrolyte in the solution and there occurs a structural rearrangement as a result of hydration (Solvation) leading to a comparatively more ordered state (Singh et al., 2008, Palani et al., 2008). Therefore, ultrasound speed increases with increase in temperature and the resistance offered by the solution to the sound velocity increases resulting in an increase in Z.

The values of isentropic compressibility, K<sub>s</sub> as calculated by Eq. (2) were fitted to an equation of the form,

$$K_s = K_s^0 + A''c + B''c^{3/2} + C''c^2$$
(1)

where A", B" and C" are constants and  $K_s^0$  is the isentropic compressibility of water. The values of the constants A", B" and C" are given in Table 1 for the aqueous solutions of all the metal chlorides at 298.15K. Typical plots of  $(K_s - K_s^0)/c$  versus  $c^{1/2}$  are shown in Fig. 3.

As observed, the value of  $K_s$  decreases with increase in concentration of the solute at all temperatures. The reason is that, when an electrolyte (solute) dissolves in water (solvent) some of the surrounding solvent molecules are closely attached to the ions due to the influence of electrostatic field of the ions. Since the solvent molecules are oriented in the ionic field (electrostatic field), the solvent molecules are more compactly packed in the primary solvation shell as compared to the compactness in the absence of the ions. Thus, the electrostatic field of the ion causes compactness of the medium due to ion-solvent interaction giving rise to a phenomenon called electrostriction. The interstitial spaces in water are occupied by the solute molecules making the medium harder to compress, i.e., providing greater electrostriction. The medium does not respond to further application of pressure. So, the

compressibility and internal pressure increase. Hence, isentropic compressibility as well as internal pressure describes the molecular arrangement in the electrolyte solutions (Moharatha et al., 2011). Also the values of K<sub>c</sub> decrease with increase in temperature for all concentrations of aqueous solutions of the metal chlorides due to the fact that, with increase in temperature, the compression of the medium becomes more prominent, resulting a decrease in  $K_s$  values. As observed, the isentropic compressibility values of all the metal chlorides solution are in the order:  $BaCl_2 < CdCl_2 < ZnCl_2 < SrCl_2 < MgCl_2$  at all temperatures.

The values of apparent molar isentropic compressibility,  $K_{s,\phi}$  computed by means of Eq. (3) were fitted into Eq. (15)

 $K_{s,\phi} = K_{s,\phi}^0 + A''c^{1/2} + B''c$ (15) to obtain  $K_{s,\phi}^0$ , the limiting apparent molar isentropic compressibility, A'' and B''' are constants, and are given in Table 1.

As observed, the values of apparent isentropic compressibility  $(K_{s,\phi})$  initially decrease (for lower concentrations) and then increase (for higher concentrations) with increase in concentration of the solutions. The negative values of  $K_{s,\phi}$  and  $K_{s,\phi}^0$  are due to loss of compressibility of surrounding solvent molecules due to strong electrostrictive forces in the vicinity of the ions causing electrostrictive solvation of the ions. As seen, the values of  $K_{s,\phi}$  of all the metal chlorides solution follow the same order as of K<sub>s</sub> discussed earlier.

From Table 3, it is observed that the intermolecular free length  $(L_i)$  decreases with increase in concentration of solution at all temperatures in all the electrolytes. It indicates that, there is a significant interaction between solute and solvent suggesting the structure promoting behaviour on addition of electrolytes (Kanhekar et al., 2010). The increase of temperature increases the thermal energy of the system and decreases the intermolecular forces thereby causing an expansion in volume and decrease in density, and hence, free length increases. Therefore, intermolecular free length increases with increase in temperatures. The intermolecular free length of one aqueous electrolyte is less than the other is due to the less isentropic compressibility value of former than the latter.

The number of solvent molecules surrounded by solute and taking part in the formation of primary shell with the central ion is termed as solvation number (Kannappan et al., 2004). In the present investigation, it is observed that the solvation number  $(S_n)$  computed for all the electrolytes is positive in aqueous medium at all the experimental temperatures indicating the appreciable solvation of solute or structure forming tendency of the solute. The solvation number increases with increase in concentration and attains maximum value at specific concentration at all experimental temperatures indicating increased dipolar interaction between solute and water molecule (Thirumaran et al., 2011). The maximum value of solvation number at specific concentration indicates stronger ion-solvent interaction. Further, it starts to decrease with increase in concentration suggesting increased ion-ion interaction. The resultant values of the  $S_n$  depends upon solute-solvent and solute-solute interaction occurring in the solution (Ali et al., 1999, Kagathara et al., 2000). With rise in temperature, the  $S_n$  values decrease in aqueous solutions of BaCl<sub>2</sub>,  $CdCl_2$  and  $SrCl_2$  but increase in MgCl\_2 and ZnCl\_2. As seen, the values of  $S_n$  are in the order:  $BaCl_2 > CdCl_2 > ZnCl_2$ > SrCl<sub>2</sub> > MgCl<sub>2</sub>. Typical plots of S<sub>n</sub> versus c are shown in Fig. 4.

The relative association  $(R_A)$  is another important property (Eyring et al., 1938) of the electrolyte solution which can be studied to understand the ion-ion or ion-solvent interactions. It is influenced by two factors: (i) breaking up of the associated solvent molecules on addition of the solute and (ii) the solvation of solute molecules. The former leads to a decrease and the latter to an increase of relative association. In the present study, RA increases with increase in concentration (Jahagirdar et al., 2000b) due to the decrease in intermolecular free length and also increases in electrostatic interaction for all the electrolyte solutions. It suggests that the solvation of the chloride salt predominate over the breaking-up of the solvent structure. As observed,  $R_A$  values of all the chlorides increase linearly with concentration for all experimental temperatures. At very dilute solution the decrease is due to breaking up of the solvent on addition of metal chlorides. But, increase in temperature weakens the intermolecular bond strength and release more solvent molecules leading to increase in R<sub>A</sub> in most of the electrolyte solutions (Mehra et al., 2014). Typical plots of  $R_A$  versus c are shown in Fig. 5.

From Table 4, it is clear that, relaxation time ( $\tau$ ) increases with increase in concentration of electrolytes (MgCl<sub>2</sub>,  $ZnCl_2$  and  $SrCl_2$ ) at all temperatures suggesting the rearrangement of molecules due to co-operation process and reinforcement of H-bonds (Ali et al., 2000), but it decreases with concentration of electrolytes (CdCl<sub>2</sub> and BaCl<sub>2</sub>) which may be due to less stronger molecular interaction. Further, with rise in temperature, H-bonds become weak due to thermal vibration resulting in structure breaking effect that predominates over H-bond formation, and hence,  $\tau$  decreases in all the electrolyte solutions (Wadekar, 2013).

Gibb's free energy change ( $\Delta G$ ) increases with increase in concentration of MgCl<sub>2</sub>, ZnCl<sub>2</sub> and SrCl<sub>2</sub>, but it decreases with concentration of CdCl<sub>2</sub> and BaCl<sub>2</sub> at all temperatures. The former suggests shorter time and latter suggests longer time for rearrangement of molecules. Moreover,  $\Delta G$  decreases with increase in temperature as given in Table 4 due to increase in kinetic energy of the molecules by thermal energy and takes longer time for rearrangement of molecules for a given concentration (Fort et al., 1965).

Ultrasonic attenuation  $(\alpha/f^2)$  is a measure of spatial rate of decrease in the intensity level of the ultrasonic wave. Attenuation co-efficient ( $\alpha$ ) is characteristic of the medium which depends on the external conditions like temperature, pressure and frequency of the measurement. Ultrasonic attenuation decreases with increase in concentration as well as rise in temperature (except in few cases). The decrease in isentropic compressibility or increase in velocity of ultrasonic wave indicates that the wave is less attenuated in the electrolyte solutions.

The free volume ( $V_t$ ) is the effective volume accessible to the centre of a molecule in a liquid. The structure of a liquid is determined by strong repulsive forces in the liquid with the relatively weak attractive forces providing the internal pressure which held the liquid molecules together. The free volume seems to be conditional by repulsive forces whereas the internal pressure is more sensitive to attractive forces. These two factors together uniquely determine the entropy of the system. Thus, the internal pressure, free volume and temperature seem to be the thermodynamic variables that describe the liquid system of fixed composition (Moharatha et al., 2011). From Table 5, it is observed that the free volume ( $V_t$ ) decreases with increase in concentration of electrolyte and internal pressure ( $\pi_i$ ) changes in a manner opposite to that of free volume at all experimental temperatures. The decrease of  $V_f$  (or increase of  $\pi_i$ ) at a given temperature indicates the formation of hard and/or tight solvation layer around the ion (Syal et al., 1998) due to more ion-solvent interaction. With increase in temperature, thermal energy of the

molecules increase, hence available free volume (V<sub>f</sub>) increases (or  $\pi_i$  decreases).

It is also found that van der Walls constant (b) is increasing almost linearly with concentrations at all the experimental temperatures as well as with rise in temperature which indicates the binding forces between the solute and solvent in the solution becomes stronger and there exist a strong molecular interaction and binding forces between the solute and solvent molecules (Nithiyanantham et al., 2009).

#### **Conclusion:-**

The results of the present study reveal that specific ion-ion and ion-solvent interactions play an important role for explaining the different thermo-acoustical parameters of strong electrolytes in aqueous medium at four different temperatures 298.15K, 303.15K, 308.15K and 313.15K. These interactions result in attractive forces which promote the structure forming tendency. It is also noticed that the strength of molecular interaction weakens with rise of temperature which may be due to weak intermolecular forces and thermal energy of the system. However, any deviation from the usual behaviour is probably due to characteristic structural changes in the system.

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D of Eq.(10													
Electrolytes	10A'	10 <sup>2</sup> B'	10 <sup>4</sup> C'	10 <sup>13</sup> A''	10 <sup>14</sup> B''	10 <sup>17</sup> C''	$10^{10} { m K}^{0}_{s, \phi}$	10 <sup>11</sup> A'''	10 <sup>13</sup> B'''				
MgCl <sub>2</sub>	6.38	-5.50	-8.02	-4.90	4.27	40.10	-6.0	5.24	3.33				
ZnCl <sub>2</sub>	4.17	-8.32	399.7	-6.87	5.97	21.32	-8.28	6.94	0.42				
SrCl <sub>2</sub>	4.54	-3.33	-12.59	-4.37	3.17	61.99	-6.03	4.33	4.94				
CdCl <sub>2</sub>	18.61	-19.08	13.75	-13.03	12.99	-98.59	-15.05	14.63	-12.91				
BaCl <sub>2</sub>	20.84	-21.75	14.25	-13.60	13.37	-89.96	-14.86	13.77	-9.37				

Table 1. Values of constants A', B' and C' of Eq.(13), A'', B'' and C'' of Eq.(14),  $K_{s,\phi}^0$  (m<sup>3</sup>.mol<sup>-1</sup>.Pa<sup>-1</sup>), A''' and B''' of Eq.(15) for aqueous metal chlorides at 298.15K

cx10 <sup>-3</sup>	Zx10 <sup>-3</sup>				K <sub>s</sub> x10 <sup>10</sup>				$\mathbf{K}_{s,\phi} \mathbf{x} 10^{10}$			
(mol.m <sup>-3</sup> )						Tempera	ature(K)					
	298.15	303.15	308.15	313.15	298.15	303.15	308.15	313.15	298.15	303.15	308.15	313.15
			I.	I.	Magne	sium chlo	ride	I.				
0.006	1495.3	1504.1	1510.9	1516.6	4.464	4.409	4.360	4.318	-3.62	-3.46	-2.75	-2.30
0.008	1498.0	1507.5	1514.5	1520.9	4.450	4.390	4.341	4.298	-4.69	-5.14	-4.67	-4.75
0.009	1499.3	1508.7	1515.8	1522.5	4.444	4.384	4.335	4.290	-5.03	-5.31	-4.99	-5.25
0.010	1500.6	1510.0	1517.1	1524.1	4.437	4.377	4.328	4.283	-5.31	-5.52	-5.24	-5.65
0.030	1503.7	1512.5	1520.4	1528.4	4.422	4.364	4.313	4.262	-2.373	-2.34	-2.37	-2.68
0.050	1509.0	1517.9	1526.0	1534.0	4.398	4.340	4.289	4.239	-2.05	-2.02	-2.05	-2.23
0.080	1514.4	1523.0	1532.6	1540.6	4.373	4.317	4.259	4.210	-1.67	-1.63	-1.75	-1.85
					Zin	c chloride	9					
0.006	1497.3	1506.1	1515.1	1520.5	4.455	4.397	4.342	4.301	-5.57	-5.41	-6.80	-6.00
0.008	1500.6	1509.7	1518.4	1523.8	4.437	4.378	4.324	4.284	-6.63	-6.70	-7.47	-6.83
0.009	1501.9	1511.0	1519.7	1525.1	4.430	4.371	4.318	4.277	-6.76	-6.80	-7.47	-6.90
0.010	1503.5	1512.7	1521.0	1526.8	4.422	4.364	4.311	4.270	-7.05	-7.08	-7.47	-7.13
0.030	1509.0	1518.2	1525.6	1532.0	4.400	4.342	4.293	4.250	-3.42	-3.43	-3.36	-3.35
0.050	1516.8	1526.0	1533.4	1540.2	4.366	4.308	4.259	4.213	-2.96	-2.95	-2.90	-2.92
0.080	1528.2	1536.5	1544.3	1550.4	4.319	4.265	4.216	4.175	-2.67	-2.59	-2.57	-2.53
					Stront	ium chloi	ride					
0.006	1495.6	1504.1	1512.1	1517.9	4.466	4.409	4.356	4.314	-3.90	-3.46	-3.97	-3.51
0.008	1498.2	1506.7	1514.5	1520.2	4.453	4.395	4.344	4.302	-4.88	-4.52	-4.64	-4.27
0.009	1499.6	1508.3	1515.8	1521.5	4.446	4.388	4.337	4.296	-5.21	-5.08	-4.96	-4.63
0.010	1501.2	1510.0	1517.1	1522.8	4.438	4.380	4.331	4.289	-5.64	-5.53	-5.22	-4.91
0.030	1508.0	1517.0	1523.3	1528.7	4.412	4.354	4.308	4.267	-3.22	-3.21	-2.92	-2.73
0.050	1514.4	1522.9	1530.4	1537.2	4.385	4.329	4.279	4.235	-2.69	-2.60	-2.55	-2.60
0.080	1524.4	1532.3	1541.9	1547.7	4.347	4.293	4.236	4.196	-2.41	-2.30	-2.40	-2.35
					Cadm	ium chlor	·ide					
0.006	1503.4	1510.2	1517.1	1522.5	4.423	4.373	4.328	4.290	-11.6	-9.46	-8.73	-7.87
0.008	1506.9	1513.7	1520.6	1526.0	4.404	4.355	4.309	4.272	-11.2	-9.58	-9.05	-8.37
0.009	1508.2	1516.6	1522.7	1528.1	4.397	4.341	4.298	4.260	-10.8	-10.4	-9.37	-8.75
0.010	1510.5	1519.3	1524.1	1529.7	4.384	4.327	4.291	4.253	-11.1	-10.9	-9.18	-8.79
0.030	1518.0	1524.8	1531.9	1537.3	4.354	4.306	4.261	4.224	-5.16	-4.68	-4.53	-4.33
0.050	1525.8	1532.6	1539.8	1545.2	4.320	4.273	4.228	4.191	-3.99	-3.70	-3.61	-3.47
0.080	1536.2	1543.7	1550.9	1557.2	4.280	4.231	4.187	4.146	-3.24	-3.09	-3.02	-2.99
					Bari	um chlori	de					
0.006	1503.0	1511.0	1518.4	1523.5	4.421	4.368	4.321	4.285	-11.2	-10.2	-9.98	-8.81
0.008	1506.6	1515.3	1522.7	1527.1	4.402	4.347	4.301	4.266	-11.0	-10.8	-10.5	-9.15
0.009	1509.2	1517.6	1525.0	1529.7	4.388	4.335	4.289	4.253	-11.5	-11.0	-10.8	-9.77
0.010	1510.9	1519.0	1526.4	1531.4	4.380	4.328	4.282	4.246	-11.3	-10.7	-10.5	-9.71
0.030	1519.6	1527.0	1534.2	1539.6	4.347	4.297	4.252	4.215	-5.47	-5.11	-4.97	-4.75

# Table 2. Values of Z (kg.m<sup>-2</sup>s<sup>-1</sup>), K<sub>s</sub> (m<sup>2</sup>N<sup>-1</sup>) and K<sub>s, $\phi$ </sub> (m<sup>3</sup>.mol<sup>-1</sup>.Pa<sup>-1</sup>) of aqueous electrolytes at different temperatures

0.050	1530.0	1536.9	1543.3	1549.0	4.306	4.258	4.216	4.178	-4.48	-4.17	-3.99	-3.89
0.080	1543.0	1549.9	1558.2	1562.2	4.258	4.211	4.162	4.130	-3.72	-3.52	-3.52	-3.33

### Table 3. Values of $L_f(m)$ , $S_n$ and $R_A$ of aqueous electrolytes at different temperatures

10-3		L <sub>f</sub> x	10 <sup>11</sup>			S	'n		R <sub>A</sub>			
$cx10^{\circ}$ (mol m <sup>-3</sup> )						Temper	ature(K)					
(1101.111 )	298.15	303.15	308.15	313.15	298.15	303.15	308.15	313.15	298.15	303.15	308.15	313.15
	•				Magne	sium chlo	ride	•				
0.006	4.353	4.358	4.373	4.391	34.7	27.7	23.4	20.6	1.0007	1.0015	1.0010	1.0007
0.008	4.346	4.349	4.364	4.380	47.4	50.4	47.6	48.2	1.0006	1.0011	1.0008	1.0009
0.009	4.342	4.346	4.360	4.377	51.5	53.5	51.6	53.4	1.0006	1.0010	1.0008	1.0011
0.010	4.339	4.342	4.357	4.373	54.8	56.4	54.8	57.5	1.0006	1.0010	1.0008	1.0013
0.030	4.332	4.336	4.349	4.363	24.4	24.3	24.5	27.8	1.0009	1.0009	1.0012	1.0015
0.050	4.320	4.324	4.337	4.350	20.6	20.5	20.8	22.7	1.0017	1.0018	1.0022	1.0026
0.080	4.308	4.312	4.322	4.335	16.6	16.4	17.7	18.8	1.0027	1.0025	1.0031	1.0034
	•				Zinc	chloride		•				
0.006	4.348	4.352	4.364	4.382	52.8	52.1	61.1	56.2	1.0011	1.0010	1.0021	1.0016
0.008	4.339	4.343	4.355	4.373	68.6	69.2	74.3	70.4	1.0006	1.0008	1.0016	1.0011
0.009	4.336	4.340	4.352	4.370	70.3	70.8	75.3	71.8	1.0006	1.0008	1.0016	1.0011
0.010	4.332	4.336	4.348	4.366	72.9	73.3	76.1	74.0	1.0008	1.0010	1.0016	1.0013
0.030	4.321	4.325	4.339	4.356	33.3	33.5	33.1	33.2	1.0027	1.0029	1.0028	1.0030
0.050	4.304	4.308	4.322	4.337	28.4	28.5	28.2	29.2	1.0043	1.0045	1.0045	1.0040
0.080	4.281	4.286	4.300	4.317	25.0	24.6	24.5	24.4	1.0074	1.0070	1.0072	1.0072
					Stront	ium chloi	ride					
0.006	4.354	4.358	4.371	4.389	30.2	27.7	31.1	28.4	1.0017	1.0015	1.0019	1.0016
0.008	4.347	4.351	4.365	4.383	44.0	41.9	42.9	40.7	1.0017	1.0014	1.0016	1.0013
0.009	4.344	4.348	4.362	4.380	48.5	48.0	47.4	45.4	1.0017	1.0017	1.0016	1.0013
0.010	4.340	4.344	4.358	4.376	53.2	52.8	51.0	49.2	1.0019	1.0019	1.0016	1.0013
0.030	4.327	4.331	4.347	4.365	28.4	28.5	26.8	25.7	1.0046	1.0048	1.0039	1.0034
0.050	4.313	4.318	4.332	4.349	23.9	23.4	23.2	23.6	1.0063	1.0060	1.0060	1.0064
0.080	4.295	4.301	4.310	4.328	20.7	20.1	21.3	21.0	1.0102	1.0096	1.0100	1.0098
					Cadm	ium chloi	ride					
0.006	4.332	4.341	4.357	4.377	120.7	101.6	91.3	80.0	1.0007	1.0003	1.0008	1.0011
0.008	4.323	4.331	4.348	4.367	119.7	105.2	97.6	89.0	1.0003	0.9999	1.0004	1.0008
0.009	4.320	4.324	4.342	4.362	115.6	113.4	103.1	95.4	1.0003	1.0001	1.0001	1.0005
0.010	4.313	4.318	4.338	4.358	119.5	118.6	101.0	95.2	1.0001	1.0001	1.0001	1.0007
0.030	4.298	4.307	4.323	4.343	52.4	48.4	46.4	44.1	1.0024	1.0020	1.0026	1.0030
0.050	4.282	4.290	4.306	4.326	39.7	37.3	36.1	34.7	1.0040	1.0036	1.0043	1.0047
0.080	4.262	4.269	4.285	4.302	31.0	29.8	29.1	28.9	1.0073	1.0073	1.0080	1.0081
					Bari	um chlori	de					
0.006	4.331	4.338	4.354	4.374	123.9	112.4	105.2	91.9	0.9995	0.9999	1.0008	1.0009
0.008	4.322	4.328	4.343	4.364	122.9	117.1	111.4	98.4	0.9993	1.0001	1.0009	1.0007
0.009	4.315	4.321	4.337	4.358	127.8	121.3	116.1	105.8	0.9993	0.9999	1.0007	1.0007
0.010	4.311	4.318	4.334	4.354	124.5	117.4	112.8	104.5	0.9995	0.9999	1.0007	1.0009

0.030	4.295	4.302	4.318	4.338	55.4	52.2	50.3	47.9	1.0026	1.0026	1.0033	1.0036
0.050	4.275	4.283	4.300	4.319	43.4	40.9	39.2	38.0	1.0059	1.0055	1.0057	1.0063
0.080	4.251	4.259	4.273	4.294	34.4	32.9	33.0	31.5	1.0109	1.0105	1.0110	1.0105

Table 4. Values of  $\tau$  (s),  $\Delta G$  (kJ.mol<sup>-1</sup>) and  $(\alpha/f^2)$  (Np.m<sup>-1</sup>) of aqueous electrolytes at different temperatures

cx10 <sup>-3</sup>	$\tau x 10^{13}$				ΔGx10 <sup>20</sup>				$(\alpha/f^2)$ x10 <sup>15</sup>				
$(\text{mol.m}^{-3})$						Temper	ature(K)						
	298.15	303.15	308.15	313.15	298.15	303.15	308.15	313.15	298.15	303.15	308.15	313.15	
-	I.		I.		Magnes	sium chlo	ride	I.					
0.006	5.107	4.556	4.099	3.633	0.4880	0.4420	0.4113	0.3728	6.731	5.965	5.331	4.697	
0.008	5.222	4.659	4.150	3.673	0.4890	0.4514	0.4166	0.3775	6.873	6.088	5.387	4.740	
0.009	5.285	4.723	4.167	3.701	0.4900	0.4571	0.4183	0.3808	6.952	6.167	5.406	4.773	
0.010	5.324	4.739	4.218	3.723	0.4920	0.4585	0.4235	0.3834	6.999	6.184	5.469	4.798	
0.030	5.406	4.760	4.296	3.779	0.4983	0.4603	0.4312	0.3899	7.097	6.203	5.562	4.861	
0.050	5.435	4.815	4.329	3.809	0.5005	0.4651	0.4345	0.3933	7.121	6.262	5.594	4.890	
0.080	5.469	4.841	4.327	3.811	0.5031	0.4674	0.4344	0.3935	7.151	6.284	5.577	4.880	
					Zine	c chloride	•						
0.006	5.311	4.696	4.186	3.768	0.4910	0.4547	0.4202	0.3886	6.995	6.141	5.437	4.866	
0.008	5.306	4.682	4.186	3.758	0.4906	0.4534	0.4202	0.3874	6.975	6.109	5.427	4.844	
0.009	5.304	4.686	4.191	3.752	0.4904	0.4538	0.4208	0.3868	6.968	6.111	5.430	4.833	
0.010	5.307	4.701	4.196	3.752	0.4906	0.4551	0.4213	0.3867	6.966	6.127	5.433	4.829	
0.030	5.310	4.701	4.195	3.751	0.4929	0.4551	0.4212	0.3866	6.961	6.118	5.425	4.822	
0.050	5.362	4.751	4.242	3.769	0.4949	0.4595	0.4259	0.3887	7.011	6.167	5.471	4.830	
0.080	5.373	4.782	4.233	3.768	0.4958	0.4623	0.4250	0.3886	7.003	6.188	5.441	4.816	
					Stronti	um chlor	ide						
0.006	5.473	4.773	4.293	3.802	0.5033	0.4615	0.4309	0.3925	7.218	6.250	5.583	4.917	
0.008	5.468	4.770	4.292	3.797	0.5030	0.4613	0.4309	0.3919	7.202	6.238	5.575	4.904	
0.009	5.465	4.768	4.291	3.797	0.5028	0.4610	0.4308	0.3919	7.194	6.230	5.570	4.901	
0.010	5.462	4.765	4.290	3.797	0.5025	0.4608	0.4307	0.3919	7.185	6.223	5.566	4.897	
0.030	5.518	4.853	4.302	3.841	0.5067	0.4685	0.4319	0.3968	7.249	6.329	5.573	4.946	
0.050	5.536	4.854	4.325	3.840	0.5081	0.4685	0.4341	0.3967	7.258	6.317	5.592	4.936	
0.080	5.559	4.877	4.360	3.882	0.5097	0.4705	0.4376	0.4015	7.273	6.336	5.623	4.978	
					Cadmi	um chlor	ide						
0.006	5.272	4.693	4.166	3.712	0.4879	0.4544	0.4182	0.3821	6.921	6.120	5.401	4.788	
0.008	5.259	4.677	4.160	3.706	0.4869	0.4530	0.4176	0.3814	6.890	6.087	5.383	4.770	
0.009	5.254	4.671	4.151	3.699	0.4865	0.4524	0.4167	0.3805	6.879	6.071	5.364	4.754	
0.010	5.244	4.668	4.154	3.703	0.4860	0.4522	0.4170	0.3810	6.857	6.059	5.364	4.757	
0.030	5.248	4.691	4.136	3.689	0.4858	0.4542	0.4151	0.3794	6.849	6.081	5.330	4.730	
0.050	5.225	4.666	4.127	3.672	0.4842	0.4520	0.4142	0.3774	6.800	6.033	5.304	4.695	
0.080	5.210	4.649	4.131	3.687	0.4831	0.4504	0.4146	0.3792	6.763	5.995	5.296	4.700	
					Bariu	m chlorio	le						
0.006	5.264	4.625	4.154	3.662	0.4873	0.4483	0.4170	0.3762	6.906	6.027	5.382	4.719	
0.008	5.247	4.614	4.140	3.652	0.4860	0.4473	0.4156	0.3750	6.869	6.001	5.353	4.697	

0.009	5.242	4.607	4.134	3.652	0.4856	0.4466	0.4150	0.3750	6.855	5.984	5.339	4.691
0.010	5.239	4.605	4.139	3.657	0.4854	0.4465	0.4155	0.3756	6.846	5.978	5.342	4.695
0.030	5.227	4.595	4.144	3.675	0.4849	0.4456	0.4160	0.3778	6.817	5.953	5.337	4.709
0.050	5.236	4.627	4.148	3.666	0.4848	0.4485	0.4164	0.3767	6.810	5.979	5.329	4.685
0.080	5.229	4.638	4.140	3.684	0.4846	0.4495	0.4155	0.3788	6.783	5.978	5.301	4.692

Table 5. Values of  $\pi_i$  (Nm<sup>-2</sup>),  $V_f$  (m<sup>3</sup>.mol<sup>-1</sup>) and b (m<sup>3</sup>.mol<sup>-1</sup>) of aqueous electrolytes at different temperatures

cx10 <sup>-3</sup>	$\pi_{i} x 10^{-0}$					V <sub>f</sub> x	:10'		bx10°			
$(\text{mol.m}^{-3})$						Temper	ature(K)					
	298.15	303.15	308.15	313.15	298.15	303.15	308.15	313.15	298.15	303.15	308.15	313.15
	•	•		•	Magnes	sium chlo	ride					
0.006	2657.0	2557.6	2468.0	2362.4	0.1993	0.2345	0.2729	0.3252	16.354	16.366	16.399	16.432
0.008	2689.4	2589.6	2486.7	2379.6	0.1923	0.2260	0.2670	0.3186	16.351	16.365	16.397	16.422
0.009	2706.9	2608.1	2492.9	2390.4	0.1886	0.2212	0.2650	0.3146	16.350	16.366	16.395	16.417
0.010	2718.2	2613.8	2509.5	2399.3	0.1863	0.2198	0.2599	0.3113	16.348	16.365	16.394	16.412
0.030	2738.3	2617.6	2532.0	2417.7	0.1820	0.2183	0.2527	0.3038	16.364	16.386	16.407	16.427
0.050	2748.3	2635.2	2544.6	2430.0	0.1800	0.2140	0.2490	0.2993	16.367	16.389	16.408	16.428
0.080	2757.3	2642.1	2545.7	2431.9	0.1780	0.2119	0.2484	0.2982	16.382	16.407	16.421	16.442
Zinc chloride												
0.006	2711.7	2597.7	2499.4	2410.3	0.1876	0.2237	0.2634	0.3068	16.348	16.372	16.380	16.416
0.008	2713.1	2596.8	2501.8	2409.5	0.1873	0.2240	0.2627	0.3071	16.351	16.371	16.383	16.419
0.009	2713.7	2599.1	2504.4	2408.7	0.1872	0.2234	0.2619	0.3075	16.350	16.370	16.382	16.418
0.010	2716.2	2605.1	2507.0	2410.1	0.1868	0.2220	0.2611	0.3071	16.346	16.366	16.381	16.414
0.030	2718.6	2606.7	2506.4	2410.7	0.1863	0.2216	0.2609	0.3067	16.350	16.368	16.395	16.421
0.050	2735.9	2624.4	2524.2	2419.8	0.1828	0.2172	0.2555	0.3031	16.352	16.370	16.396	16.430
0.080	2745.5	2637.6	2526.1	2423.9	0.1812	0.2140	0.2551	0.3019	16.346	16.374	16.397	16.427
					Stronti	ium chlor	ide					
0.006	2751.1	2616.8	2527.1	2418.2	0.1797	0.2188	0.2545	0.3035	16.346	16.372	16.391	16.424
0.008	2752.1	2618.2	2528.4	2418.0	0.1795	0.2185	0.2540	0.3035	16.345	16.372	16.394	16.427
0.009	2752.6	2619.2	2529.2	2419.0	0.1795	0.2183	0.2538	0.3032	16.345	16.368	16.394	16.426
0.010	2753.5	2620.3	2530.0	2419.9	0.1794	0.2182	0.2536	0.3029	16.341	16.364	16.393	16.426
0.030	2770.8	2647.7	2534.8	2434.4	0.1762	0.2117	0.2521	0.2973	16.338	16.358	16.397	16.434
0.050	2776.4	2647.6	2543.7	2438.9	0.1749	0.2113	0.2494	0.2962	16.350	16.376	16.403	16.424
0.080	2786.1	2656.6	2559.6	2455.5	0.1733	0.2092	0.2452	0.2902	16.347	16.381	16.396	16.429
					Cadmi	um chlor	ide					
0.006	2707.6	2599.8	2493.3	2393.3	0.1886	0.2231	0.2649	0.3132	16.344	16.377	16.397	16.422
0.008	2706.8	2597.7	2493.9	2393.7	0.1888	0.2236	0.2647	0.3131	16.347	16.380	16.399	16.425
0.009	2706.7	2599.2	2492.6	2392.5	0.1888	0.2234	0.2650	0.3135	16.347	16.373	16.402	16.427
0.010	2705.9	2600.9	2494.4	2395.4	0.1890	0.2231	0.2645	0.3125	16.348	16.371	16.402	16.424
0.030	2708.7	2606.0	2491.0	2392.2	0.1883	0.2214	0.2655	0.3135	16.357	16.390	16.408	16.434
0.050	2703.7	2600.0	2489.3	2387.6	0.1890	0.2225	0.2657	0.3149	16.375	16.408	16.425	16.450
0.080	2701.0	2597.5	2492.8	2395.1	0.1893	0.2230	0.2643	0.3116	16.392	16.419	16.436	16.464
					Bariu	ım chlorio	le					

0.006	2703.0	2580.8	2490.7	2377.3	0.1892	0.2279	0.2657	0.3195	16.363	16.382	16.397	16.426
0.008	2701.4	2581.8	2490.3	2376.4	0.1895	0.2279	0.2661	0.3199	16.365	16.377	16.392	16.427
0.009	2702.7	2581.5	2490.1	2378.7	0.1893	0.2279	0.2662	0.3191	16.363	16.378	16.393	16.425
0.010	2703.5	2582.0	2492.6	2381.7	0.1893	0.2278	0.2654	0.3180	16.360	16.378	16.393	16.422
0.030	2703.2	2580.2	2494.6	2388.9	0.1893	0.2281	0.2644	0.3149	16.364	16.390	16.407	16.433
0.050	2710.1	2592.7	2497.7	2388.1	0.1880	0.2248	0.2632	0.3151	16.362	16.395	16.420	16.442
0.080	2712.6	2599.7	2501.2	2396.6	0.1876	0.2231	0.2624	0.3115	16.365	16.397	16.415	16.455



Fig. 1. Plot of ultrasonic velocity (U) vs c for aqueous magnesium chloride



Fig. 2. Plot of  $(U-U_0)/c$  vs  $c^{1/2}$  for aqueous metal chlorides at 298.15K



Fig. 3. Plot of  $(K_s-K_s^0)/c$  vs  $c^{1/2}$  for aqueous metal chlorides at 298.15K



Fig. 4. Plot of solvation number  $(S_n)$  vs c for aqueous metal chlorides at 298.15K



Fig. 5. Plot of relative association (R<sub>A</sub>) vs c for aqueous metal chlorides at 298.15K

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