

Electrical Conductances of Some 2:2 Electrolytes in Ethanol-Water Mixtures at Different Temperatures

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Abstract

The electrical conductances of the solutions of manganese sulphate (MnSO_4), nickel sulphate (NiSO_4), and zinc sulphate (ZnSO_4) in binary aqueous mixtures containing 5%, 10%, 15% and 20% (w/w) of ethanol have been measured at 25, 30, 35, and 40 °C. The conductance data were analyzed by the Fuoss-Onsager equation in terms of the limiting equivalent conductance (Λ_0), and the association constant (K_A). These electrolytes were found to build up ionic association in the mixed solvent system. The Λ_0 values were found to be increased with temperature and it was decreased with increase of the organic portion in the mixtures. The K_A values were found to be increased with increase of organic content. It was also found that K_A values were increased with increase of temperatures.

Keywords: Electrolytes, ethanol, mixed solvent, ion association, conductance, limiting conductance, ion association constant

Introduction

In 1926, Bjerrum was first introduced the ion-association concept of electrolytes in solution [1]. According to this concept an electrolyte solution is considered to be a mixture of free ions and ion pairs. The ion-association phenomenon has an important effect on thermodynamic, transport, dielectric and other properties of electrolyte solutions [2]. It is already reported that precise conductivity measurements provide important information about ion-ion and ion-solvent interactions [3]. The ion-association of electrolytes on the basis of conductance measurements has been extensively investigated. Various conductance measurements have been reported in the literature in aqueous medium, studies in mixed solvents are relatively rare [4]. Recently considerable attention is received on ion-association process and a number of studies have been reported in the literature which shown the nature of the spherical ions, having a large variation in size in mixed solvent of water and alcohols [5].

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Reported data of ion-association are typically available for 1:1 electrolytes, mainly alkali halides in water-organic mixed solvents [6, 7]. Very few works is reported on the ion-association of polyvalent symmetrical electrolytes in mixed solvent [8, 9]. Baker group systematically studied the conductivity of some electrolytes of alkaline earth metal in water-dioxane mixed solvent with different solvent composition and temperatures [10]. The studies of divalent symmetrical electrolytes solution in ethanol-water mixed solvent are capable to provide important information about the ion-solvent interaction which is crucial for us to know about the influence of solvent structures. The conductivity measurements of divalent electrolytes solution in mixed solvent with variation of solvent compositions and temperature will be able to give very important information regarding equivalent conductance and ion-association constant values. Thermodynamic parameters like Gibbs free energy, enthalpy and entropy which estimated on the basis of ion-association constant provide significant information about ion-ion, ion-solvent and solvent-solvent interactions. Shonzu Katayama measured the electrical conductance of aqueous solutions of various 2:2 electrolytes at different temperatures and estimated the ion association constants (K_A) and the thermodynamic parameters. He determined the association constants by using the revised Fuoss-Onsager equation which provides a better fit to the conductivity data [11]. The conductance studies over a wide range of solvent composition and temperature are able to provide useful information regarding some specific short-range interactions [12, 13].

A lot of investigations were performed for better understanding about extraordinary characteristics of water as liquid and as well as solvent [14, 15]. Monohydric alcohols offer most favorable conditions among a variety of components that have been added to water for such studies due to the amphiphilic character of alcohol molecules. Alcohol molecules interact strongly with water through hydrogen bonds. It seemed interesting to study the ion-association of the symmetrical electrolytes in aqueous-organic solvent mixtures since it will be given us a useful information about ion-solvent interactions in solutions. Thus, conductance studies of some 2:2 electrolytes in water-ethanol mixed solvent were carried out to see the change in ion-association with the variation of temperatures and the solvent composition of the mixtures. The aim of this present study is to determine

the effect of solvent compositions and temperatures on the values of equivalent conductances and ion-association constants.

In this paper, we report equivalent conductance of manganese sulphate (MnSO_4), nickel sulphate (NiSO_4), and zinc sulphate (ZnSO_4) solution in ethanol-water solvent mixtures at various temperatures. The composition and temperature dependence of the limiting equivalent conductances and ion-association constants of these electrolytes in various solvent compositions is determined from these conductance data.

Experimental

Materials

The reagent grade ethanol (E. Merck, India) was used for the experimental works which was fractionally distilled for further purification. The distilled water was purified to get deionized (DI) water with the help of a double Barnstead ion-exchange column. The purified DI water was specific conductance of less than $10^{-6} \text{ S cm}^{-1}$ and was used for the preparation of the mixed solvents. Analytical grade electrolytes (E. Merck, Germany) like, manganese sulphate (MnSO_4), nickel sulphate (NiSO_4) and zinc sulphate (ZnSO_4) were used in our present study. These electrolytes were recrystallized by using DI water and dried at room temperature to get more pure electrolytes.

Density and Viscosity Measurement

Various solvent mixtures (w/w) of ethanol and DI water were prepared. The densities of the pure and mixed solvents were measured by weighing an accurate volume of the pure and mixed solvents in a density bottle using analytical balance. Ostwald viscometer was used to measure the viscosity of each solvent mixture. The values of dielectric constants of the solvent mixtures were interpolated from the published data [16,17]. The physical properties of the mixed solvents are summarized in Table 1. It is observed that the values of viscosity increase with increasing ethanol content at a given temperature and decrease with increase of temperature in a given solvent mixtures. It is also observed that the dielectric constant of the medium decreased at a given temperature with increasing ethanol content and it also decreased at a given solvent mixture with increase of

temperature. This trend is also found in literature [18] for methanol content in water.

Conductance measurement

The conductance measurements were performed with the help of the digital conductance meter (WTW LF 521, Germany) with a cell constant of 0.620 cm^{-1} at several temperatures. The conductance cell was calibrated by the help of reported method [19] using dilute potassium chloride solutions. Thermostated water bath with high precision was used to carry out this conductance measurements. Solutions of each electrolyte were prepared with mixed solvents for the conductance measurement and various independent solutions were prepared to ensure the reproducibility of the conductance data. The reproducibility of the measurements was examined by repeating each experimental run several times. Solvent's specific conductance was subtracted from the specific conductance to get exact conductance of the electrolytic solution.

Results and Discussion

The calculated equivalent conductance (Λ) of the studied three 2:2 electrolyte solutions in the ethanol-water mixed solvent (5, 10, 15 and 20, wt.% ethanol) at different temperature (25, 30, 35 and 40°C) are summarized in Table 2.1 to 4.2. It showed that the equivalent conductance of the studied electrolytes decreased with the increased of molar concentration, as expected from conductivity equations. It also showed that the equivalent conductances decreased with increasing ethanol content in the solvent mixtures. These results are in agreement with other studies in the literature [20, 21]. A plot of Λ vs. \sqrt{c} is shown in Figure 1 which indicate the evidence of ion-pair formation [22]. The conductance data were treated with the Fouss-Onsager method [23,24].

$$\Lambda' = \Lambda_0 - S(c\gamma)^{1/2} + E c \gamma \log c \gamma + J c \gamma - K_A c \gamma f_{\pm}^2 \Lambda \quad (1)$$

Where S is the Onsager limiting slope, c is the molar concentration, γ is the degree of dissociation, E and J are the theoretical coefficients, Λ_0 is the limiting equivalent conductance, K_A is the ion-association constant and f_{\pm} is the mean ionic activity coefficient.

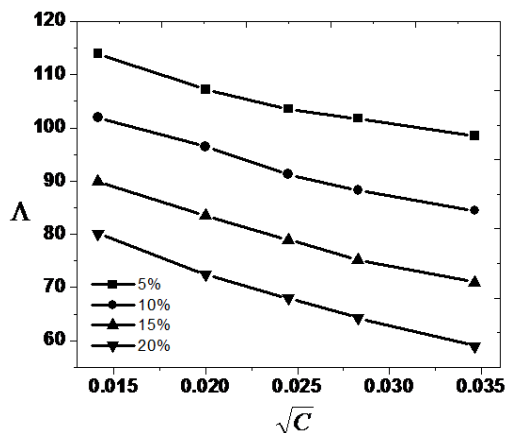


Figure 1: Variation of equivalent conductance (Λ) with square root of molar concentration (\sqrt{C}) for NiSO_4 solution at 25°C .

Table 1: Physical properties of ethanol-water mixtures at different temperature

$T^\circ\text{C}$	5 wt.% Ethanol		10 wt.% Ethanol		15 wt.% Ethanol		20 wt.% Ethanol	
	D	η (m N s m ⁻²)	D	η (m N s m ⁻²)	D	η (m N s m ⁻²)	D	η (m N s m ⁻²)
25	74.38	0.9664	69.85	1.0722	65.33	1.1780	60.80	1.2720
30	72.65	0.8694	68.20	0.9511	63.75	1.0451	59.30	1.1300
35	70.68	0.7826	66.35	0.8532	62.03	0.9281	57.70	1.0110
40	69.05	0.7038	64.80	0.7705	60.55	0.8381	56.30	0.9110

Table 2.1: Equivalent conductance (Λ) of NiSO_4 in 5% and 10% (w/w) ethanol-water mixture at different temperature

Conc. mol dm ⁻³	25°C	30°C	35°C	40°C	25°C	30°C	35°C	40°C
	5% (w/w)				10% (w/w)			
	Λ in S cm ² equ ⁻¹							
2.00×10^{-4}	113.95	127.98	141.87	155.99	101.95	113.98	126.10	138.10
4.00×10^{-4}	107.20	124.18	137.21	151.15	96.50	109.90	120.08	132.85
6.00×10^{-4}	103.53	118.01	131.05	142.35	91.25	105.05	114.60	124.98
8.00×10^{-4}	101.75	113.10	125.75	137.05	88.30	101.00	109.80	118.10
12.00×10^{-4}	98.50	109.50	122.08	131.92	84.50	96.00	103.00	112.90

Table 2.2: Equivalent conductance (Λ) of NiSO₄ in 15% and 20% (w/w) ethanol–water mixture at different temperature

Conc. mol dm ⁻³	25°C	30°C	35°C	40°C	25°C	30°C	35°C	40°C
	15% (w/w)				20% (w/w)			
	Λ in S cm ² equ ⁻¹							
2.00x10 ⁻⁴	89.96	99.98	112.15	122.75	80.15	88.10	99.50	107.90
4.00x10 ⁻⁴	83.50	95.87	107.00	116.98	72.50	82.90	93.00	102.10
6.00x10 ⁻⁴	79.00	90.00	98.75	108.25	68.00	74.70	83.25	93.30
8.00x10 ⁻⁴	75.25	83.15	93.10	102.85	64.30	67.12	78.50	85.50
12.00x10 ⁻⁴	71.00	78.50	87.00	95.00	59.00	62.80	72.00	77.10

Table 3.1: Equivalent conductance (Λ) of MnSO₄ in 5% and 10% (w/w) ethanol–water mixture at different temperature

Conc. mol dm ⁻³	25°C	30°C	35°C	40°C	25°C	30°C	35°C	40°C
	5% (w/w)				10% (w/w)			
	Λ in S cm ² equ ⁻¹							
2.00x10 ⁻⁴	116.51	130.18	140.32	151.54	104.50	118.05	127.25	138.25
4.00x10 ⁻⁴	109.08	124.73	134.25	144.75	98.05	111.70	120.50	130.75
6.00x10 ⁻⁴	105.25	117.10	126.05	138.10	93.29	104.85	112.50	121.60
8.00x10 ⁻⁴	100.10	112.25	122.20	133.08	90.00	100.19	107.00	115.30
12.00x10 ⁻⁴	96.05	108.00	117.00	128.25	84.50	94.00	98.00	106.10

Table 3.2: Equivalent conductance (Λ) of MnSO₄ in 15% and 20% (w/w) ethanol–water mixture at different temperature

Conc. mol dm ⁻³	25°C	30°C	35°C	40°C	25°C	30°C	35°C	40°C
	15% (w/w)				20% (w/w)			
	Λ in S cm ² equ ⁻¹							
2.00x10 ⁻⁴	91.51	101.08	110.85	124.05	80.15	90.25	100.23	110.75
4.00x10 ⁻⁴	84.10	92.73	102.15	113.60	72.52	81.54	90.75	99.50
6.00x10 ⁻⁴	79.24	87.30	96.10	107.08	66.50	75.50	84.10	92.00
8.00x10 ⁻⁴	75.09	83.00	91.23	101.75	63.75	73.25	81.50	89.10
12.00x10 ⁻⁴	71.25	79.15	87.40	96.10	61.24	70.50	78.00	86.05

Table 4.1: Equivalent conductance (Λ) of ZnSO_4 in 5% and 10% (w/w) ethanol–water mixture at different temperature

Conc. mol dm^{-3}	25°C	30°C	35°C	40°C	25°C	30°C	35°C	40°C
	5% (w/w)				10% (w/w)			
	Λ in $\text{S cm}^2 \text{equ}^{-1}$							
2.00×10^{-4}	118.25	131.70	141.00	152.50	106.50	119.75	128.00	139.25
4.00×10^{-4}	110.50	125.50	135.25	145.75	99.00	112.50	121.20	132.10
6.00×10^{-4}	106.00	119.20	127.00	139.25	94.10	106.25	116.50	126.00
8.00×10^{-4}	102.06	113.00	122.20	134.50	91.00	101.30	110.15	120.20
12.00×10^{-4}	97.15	109.00	118.00	129.30	86.30	96.00	106.00	116.50

Table 4.2: Equivalent conductance (Λ) of ZnSO_4 in 15% and 20% (w/w) ethanol–water mixture at different temperature

Conc. mol dm^{-3}	25°C	30°C	35°C	40°C	25°C	30°C	35°C	40°C
	15% (w/w)				20% (w/w)			
	Λ in $\text{S cm}^2 \text{equ}^{-1}$							
2.00×10^{-4}	91.50	101.00	111.50	123.75	80.10	90.25	100.30	110.75
4.00×10^{-4}	84.00	92.75	102.10	113.60	72.50	81.50	90.75	99.50
6.00×10^{-4}	79.20	87.30	96.00	107.00	66.50	75.50	84.10	92.00
8.00×10^{-4}	75.00	83.00	91.00	101.75	64.75	73.25	81.50	89.10
12.00×10^{-4}	71.25	79.15	87.40	96.00	61.25	70.50	78.00	86.05

Table 5: Limiting equivalent conductance (Λ_0) of MnSO_4 , NiSO_4 and ZnSO_4 in ethanol–water mixtures at different temperature

T/°C	5 wt.% Ethanol			10 wt.% Ethanol			15 wt.% Ethanol			20 wt.% Ethanol		
	NiSO_4	MnSO_4	ZnSO_4	NiSO_4	MnSO_4	ZnSO_4	NiSO_4	MnSO_4	ZnSO_4	NiSO_4	MnSO_4	ZnSO_4
25	129.93	132.21	135.62	119.15	122.52	125.37	108.97	110.73	113.51	101.18	103.13	104.11
30	146.82	148.94	152.67	132.95	138.83	141.45	120.92	122.87	125.42	112.94	114.95	116.18
35	165.11	162.95	165.74	148.24	152.53	155.20	135.84	137.13	138.15	127.69	128.18	129.42
40	183.40	177.25	174.48	163.17	166.98	170.13	149.78	152.18	148.35	139.64	142.98	138.63

Table 6: Ion-association constant (K_A) of $MnSO_4$, $NiSO_4$ and $ZnSO_4$ at different temperature in ethanol–water mixtures

T ^o C	5 wt.% Ethanol			10 wt.% Ethanol			15 wt.% Ethanol			20 wt.% Ethanol		
	NiSO ₄	MnSO ₄	ZnSO ₄	NiSO ₄	MnSO ₄	ZnSO ₄	NiSO ₄	MnSO ₄	ZnSO ₄	NiSO ₄	MnSO ₄	ZnSO ₄
25	263	241	225	358	292	280	426	351	359	553	538	516
30	300	278	252	396	314	306	454	384	371	565	551	533
35	329	310	289	419	335	327	468	399	397	573	561	554
40	356	332	311	445	368	358	505	425	437	599	588	573

The limiting equivalent conductance (Λ_0)

Initial Λ_0 values of all the three electrolytes were found from the graphical plot of Λ vs. \sqrt{c} and used as preliminary data in the iteration calculation method to get final Λ_0 values. The calculated Λ_0 values of electrolytes $NiSO_4$, $MnSO_4$ and $ZnSO_4$ in various mixed solvent at different temperatures are summarized in Table 5. It was found that the Λ_0 values were decreased with increase of ethanol content in the solvent mixtures. This decreasing tendency of Λ_0 values has good agreement with the reported data in methanol-water mixtures [21]. It was also increased with increase of temperature for all the electrolytes. It may be happened because of association or solvation of electrolytes in the mixed solvents. Among the three electrolytes, $ZnSO_4$ shows highest Λ_0 values than that of $NiSO_4$ and $MnSO_4$ and follow the order: $ZnSO_4 > MnSO_4 > NiSO_4$. It indicates that the ion-solvent interaction of $ZnSO_4$ is lower than the ion-solvent interaction of $NiSO_4$ and $MnSO_4$ [25]. It also point out that the ionic mobility of the zinc ion is higher than that of the manganese and nickel ions in all solutions.

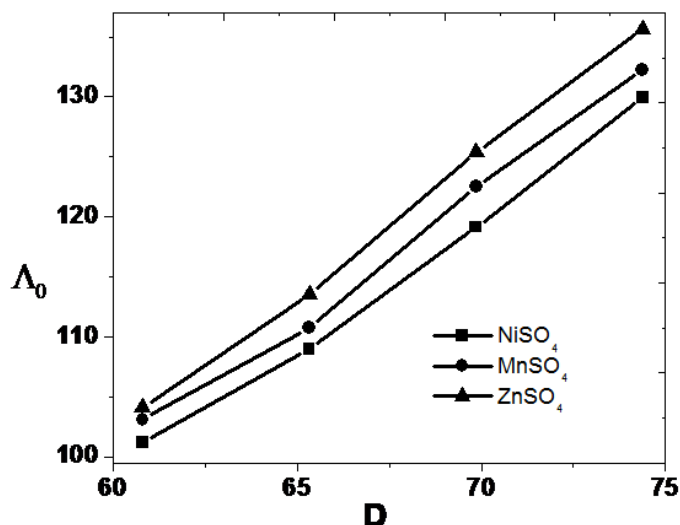


Figure 2: Variation of limiting equivalent conductance (Λ_0) of NiSO_4 , MnSO_4 and ZnSO_4 with dielectric constant (D) at 25°C .

The reported ionic radii of Ni^{+2} (0.78\AA) is smaller than that of Zn^{+2} (0.83\AA) ion and Mn^{+2} (0.91\AA) ion [26] and it follows the order: $\text{Mn}^{+2} > \text{Zn}^{+2} > \text{Ni}^{+2}$. According to our Λ_0 values, the existing relative sizes of these ions in solutions follow the order: $\text{Ni}^{+2} > \text{Mn}^{+2} > \text{Zn}^{+2}$. This result demonstrates that the electrostatic ion-solvent interaction [27] is strong for smaller (Ni^{+2}) ion in mixed solvent as the smaller ion with greater surface charge density should be solvated more than those of Zn^{+2} and Mn^{+2} ions and makes it a bigger ion compared to the zinc ions. This is same for Zn^{+2} ions with compare to Mn^{+2} ions which are remain unsolvated. In our study, Λ_0 values are increased with increase of temperature which indicates that ionic mobility of ions increase with temperature. This increasing trend of Λ_0 values is occurred because of ion-solvent interaction. The variation of Λ_0 of NiSO_4 , MnSO_4 and ZnSO_4 with dielectric constant (D) is shown in Figure 2.

The ion-association constant (K_A)

The ion-association constant (K_A) values of the three electrolytes were calculated by using conductance data on the basis of Fuoss-Onsager equation [28] and summarized in Table 6. It shows that all the electrolytes

are associated in ethanol-water mixed solvent. The values of K_A in water were taken from the literature [12, 13]. A plot of $\log K_A$ vs. $1/D$ is shown in Figure 3. The plot demonstrates that the ion-association constant values of each electrolyte increase with increase of organic content in the mixed solvent. This is happened because of the regular decrease of dielectric constant values of the medium due to the increase of organic portion in the solvent mixtures. These results are in good agreement with many studies dealing with this subject [21,29,30]. The ion-association constant values of each electrolyte were also increased with increase of temperature for every solvent mixture. In our study, the ion-association constant values of NiSO_4

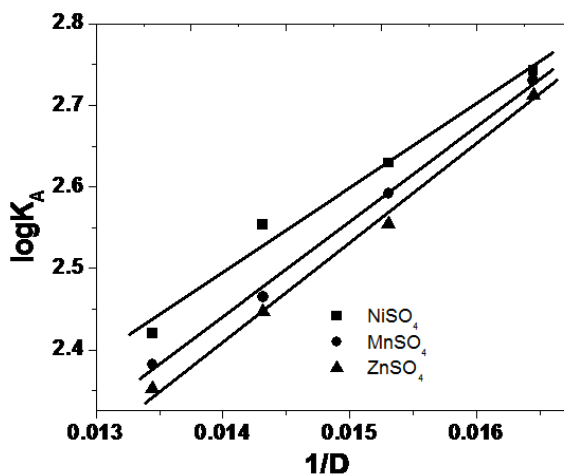


Figure 3: Variation of ion-association constant (K_A) of NiSO_4 , MnSO_4 and ZnSO_4 with dielectric constant (D) at 25°C .

are found greater than that of MnSO_4 and ZnSO_4 . The K_A values of our studied electrolytes follow the order: $\text{NiSO}_4 > \text{MnSO}_4 > \text{ZnSO}_4$. This result indicate that the cationic size of the Ni^{+2} ions in the solvent mixture is higher than that of Mn^{+2} ions and Zn^{+2} ions. Since electrostatic theory predict a stronger electric field to be associated with the ions of smaller radii and consequently, a larger association of solvent molecules [31]. The ion mobility will be dependent on its relationship with the solvent in terms of the solvent properties, such as viscosity, and on its association with the

solvent. Though, the reported crystallographic cation size follows the order: $\text{Mn}^{+2}(0.91) > \text{Zn}^{+2}(0.83) > \text{Ni}^{+2}(0.78)$. According to this ionic radii, Ni^{+2} ions should be least associated and Mn^{+2} ions should be most associated [32,33]. But our K_A values suggest that NiSO_4 is more associated in ethanol-water mixtures rather than MnSO_4 and ZnSO_4 . The plot of $\log K_A$ vs. $1/D$ should be linear according to the Fuoss equation

$$K_A = \frac{4\pi N_A^3}{3000} \frac{|z_+ z_-| e^2}{a D k T} \quad (2)$$

Linear behavior is also predictable according to the reported "sphere in continuum" model [34,35]. Figure 3 shows almost linear plot for MnSO_4 and ZnSO_4 . For NiSO_4 the plot of $\log K_A$ vs. $1/D$ shows slightly deviation from linearity compared to other two electrolytes. This phenomenon may be occurred because of specific ion-solvent interaction or the solvation of ions in ethanol-water mixed solvent [36]. Smaller ions have great opportunity to form ion-solvent complexes due to its higher charge density. Solution of electrolytes is considered as an example of highly charged solutions and it shows association up to some level [37] though the nature of this type of association is too complex. The existence of this association in this type of solution is found up to three equilibrium processes [38].

Conclusion

Equivalent conductance of the three electrolytes nickel sulphate, manganese sulphate and zinc sulphate solution in ethanol-water mixed solvent media have been presented as a function of solution concentration with different solvent composition at different temperatures. The equivalent conductances are found to decrease with increasing concentration of organic portion over the entire mixed solvent investigated and increase with increasing temperature. ZnSO_4 provided higher Λ_0 values than NiSO_4 as well as MnSO_4 which may reveals that the ion-solvent interaction for ZnSO_4 is lower than the ion-solvent interaction for NiSO_4 and MnSO_4 . The K_A values were found to increase with increase of organic content in the solvent mixtures. This phenomenon may be happened due to the decrease of dielectric constants of the solvent mixtures. The ion-association of ion pairs is found to be increased with increase of organic content in the solvent mixtures.

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