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Viscosity and Apparent Molar Volume of Univalent Electrolytes in DMF + Water Mixed Solvents at 293.15 K

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ABSTRACT: Near the end of the 19th century, the properties of electrolyte solutions were investigated extensively by the early workers in physical chemistry. A suggestion of Svante August Arrhenius, a Swedish chemist, that salts of strong acids and bases (for example, sodium chloride) are completely dissociated into ions when in aqueous solution received strong support from electrical-conductivity measurements and from molecular-weight studies (freezing-point depression, boiling-point elevation, and osmotic pressure). These studies showed that the number of solute particles was larger than it would be if no dissociation occurred. For example, a 0.001 molal solution of a uni-univalent electrolyte (one in which each ion has a valence, or charge, of 1, and, when dissociated, two ions are produced) such as sodium chloride, Na^+Cl^- , exhibits colligative properties corresponding to a nonelectrolyte solution whose molality is 0.002; the colligative properties of a 0.001 molal solution of a univalent-divalent electrolyte (yielding three ions) such as magnesium bromide, $\text{Mg}^{2+}\text{Br}_2^-$, correspond to those of a nonelectrolyte solution with a molality of 0.003. At somewhat higher concentrations the experimental data showed some inconsistencies with Arrhenius' dissociation theory, and initially these were ascribed to incomplete, or partial, dissociation. In the years 1920–30, however, it was shown that these inconsistencies could be explained by electrostatic interactions (Coulomb forces) of the ions in solution. The current view of electrolyte solutions is that, in water at normal temperatures, the salts of strong acids and strong bases are completely dissociated into ions at all concentrations up to the solubility limit. At high concentrations Coulombic interactions may cause the formation of ion pairs, which implies that the ions are not dispersed uniformly in the solution but have a tendency to form two-ion aggregates in which a positive ion seeks the close proximity of a negative ion and vice versa. While the theory of dilute electrolyte solutions is well advanced, no adequate theory exists for concentrated electrolyte solutions primarily because of the long-range Coulomb forces that dominate in ionic solutions.

The equilibrium properties of electrolyte solutions can be studied experimentally by electrochemical measurements, freezing-point depressions, solubility determinations, osmotic pressures, or measurements of vapour pressure. Most electrolytes, such as salts, are nonvolatile at ordinary temperature, and, in that event, the vapour pressure exerted by the solution is the same as the partial pressure of the solvent. The activity coefficient of the solvent can, therefore, be found from total-pressure measurements, and, using the Gibbs-Duhem equation, it is then possible to calculate the activity coefficient of the electrolyte solute. This activity coefficient is designated by γ_{\pm} to indicate that it is a mean activity coefficient for the positive and negative ions. Since it is impossible to isolate positive ions and negative ions into separate containers, it is not possible to determine individual activity coefficients for the positive ions and for the negative ions. The mean activity coefficient γ_{\pm} is so defined that it approaches a value of unity at very low

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molality where the ions are so far apart that they exert negligible influence on one another. For small concentrations of electrolyte, the theory of Peter Debye, a Dutch-born American physical chemist, and Erich Hückel, a German chemist, relates γ_{\pm} to the ionic strength, which is the sum of the products of the concentration of each ion (in moles per litre) and the square of its charge; the equation predicts that γ_{\pm} decreases with rising ionic strength in agreement with experiment at very low ionic strength; at higher ionic strength, however, γ_{\pm} rises, and in some cases γ_{\pm} is greater than 1. The derivation of the Debye-Hückel theory clearly shows that it is limited to low concentrations. Many attempts have been made to extend the Debye-Hückel equation to higher electrolyte concentrations. One of the more successful attempts is based on the idea that the ions are solvated, which means that every ion is surrounded by a tight-fitting shell of solvent molecules.

The concept of solvation is often used to explain properties of aqueous solutions; one well-known property is the salting-out effect, in which the solubility of a nonelectrolyte in water is decreased when electrolyte is added. For example, the solubility of ethyl ether in water at 25° C is 0.91 mole percent, but, in an aqueous solution containing 15 weight percent sodium chloride, it is only 0.13 mole percent. This decrease in solubility can be explained by postulating that some of the water molecules cannot participate in the dissolution of the ether because they are tightly held (solvated) by sodium and chloride ions.

KEYWORDS: univalent, electrolytes, viscosity, molar volume, solvents, apparent

I. INTRODUCTION

Electrolyte solutions have long been of interest in industry since many common inorganic chemicals are directly obtained, or else separated, by crystallization from aqueous solution. Further, many important chemical and metallurgical products (e.g., aluminum) are obtained or refined by electrochemical processes that occur in liquid solution. In recent years there has been renewed interest in electrolyte solutions because of their relevance to fuel cells as a possible source of power for automobiles.[1,2]

The properties of electrolyte solutions also have large importance in physiology. Many molecules that occur in biological systems bear electric charges; a large molecule that has a positive electric charge at one end and a negative charge at the other is called a zwitterion. Very large molecules, such as those of proteins, may have numerous positive and negative charges; such molecules are called polyelectrolytes.[3,4] In solution, the conformation (i.e., the three-dimensional structure) of a large, charged molecule is strongly dependent on the ionic strength of the dissolving medium; for example, depending on the nature and concentration of salts present in the solvent, a polyelectrolyte molecule may coagulate into a ball, it may stretch out like a rod, or it may form a coil or helix. The conformation, in turn, is closely related to the molecule's physiological function. As a result, improved understanding of the properties of electrolyte solutions has direct consequences in molecular biology and medicine.[5,6]

Since the dissolution of one substance in another can occur only if there is a decrease in the Gibbs energy, it follows that, generally speaking, gases and solids do not dissolve in liquids as readily as do other liquids. To understand this, the dissolution of a solid can be visualized as occurring in two steps: in the first, the pure solid is melted at constant temperature to a pure liquid, and, in the second, that liquid is dissolved at constant temperature in the solvent. Similarly, the dissolution of a gas can be divided at some fixed pressure into two parts, the first corresponding to constant-temperature condensation of the pure gas to a liquid and the second to constant-temperature mixing of that liquid with solvent. In many cases, the pure liquids (obtained by melting or by condensation) may be hypothetical (i.e., unstable and, therefore, physically unobtainable), but usually their

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properties can be estimated by reasonable extrapolations.[7,8] It is found that the change in Gibbs energy corresponding to the first step is positive and, hence, in opposition to the change needed for dissolution. For example, at -10°C , ice is more stable than water, and, at 110°C and one atmosphere, steam is more stable than water. Therefore, the Gibbs energy of melting ice at -10°C is positive, and the Gibbs energy of condensing steam at one atmosphere and 110°C is also positive. For the second step, however, the change in Gibbs energy is negative; its magnitude depends on the equilibrium composition of the mixture. Owing to the positive Gibbs energy change that accompanies the first step, there is a barrier that makes it more difficult to dissolve solids and gases as compared with liquids.

For gases at normal pressures, the positive Gibbs energy of condensation increases with rising temperature, but, for solids, the positive Gibbs energy of melting decreases with rising temperature. For example, the change in energy, ΔG , of condensing steam at one atmosphere is larger at 120°C than it is at 110°C , while the change in energy of melting ice at -5°C is smaller than it is at -10°C . [9,10] Thus, as temperature rises, the barrier becomes larger for gases but lower for solids, and therefore, with few exceptions, the solubility of a solid rises while the solubility of a gas falls as the temperature is raised.

For solids, the positive Gibbs energy “barrier” depends on the melting temperature. If the melting temperature is much higher than the temperature of the solution, the barrier is large, shrinking to zero when the melting temperature and solution temperature become identical.

The tables give the solubilities of some common gases and the solubility of (solid) naphthalene in a few typical solvents. These solubilities illustrate the qualitative rule that “like dissolves like”; thus naphthalene, an aromatic hydrocarbon, dissolves more readily in another aromatic hydrocarbon such as benzene than it does in a chlorinated solvent such as carbon tetrachloride or in a hydrogen-bonded solvent such as methyl alcohol. By similar reasoning, the gas methane, a paraffinic hydrocarbon, dissolves more readily in another paraffin such as hexane than it does in water. In all three solvents, the gas hydrogen (which boils at -252.5°C) is less soluble than nitrogen (which boils at a higher temperature, -195.8°C).[11,12]

Dissolution, apparent diffusion and phase potentials of nine univalent electrolytes, viz. NaCl, NaBr, NaI, KCl, KBr, KI, NH_4Cl , NH_4Br and NH_4I are measured in methanol and ethanol. The sign of diffusion and phase potentials are explained in the light of ionic mobility and ion—solvent interaction respectively. The sign and relative magnitude of true dissolution potential are explained on the basis of unequal rate of removal of cations and anions from the crystal lattice due to the difference in their solvation energies.[13,14]

II. DISCUSSION

Viscosity measurements of a series of univalent electrolytes in water have been performed with an automatic dual viscometer system, covering the temperature range of 5 to 95°C . Results are discussed in terms of Jones-Dole B and D coefficients. The B coefficients of the salts are divided into their ionic contributions according to $B(\text{K}^+) = B(\text{Cl}^-)$ at all temperatures. On a simple model intrinsic and structural contributions in B are calculated for the different ions. The structural term depends exponentially on the temperature in a unique manner, independent of the ion (except for Li^+).[15,16]

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Molar Enthalpy of Solution of Univalent Electrolytes at 25 °C

| Mol. form. | Name | CAS Reg. No. | Mol. wt. | State | $\Delta_{\text{sol}}H/\text{kJ mol}^{-1}$ |
|-------------------------------------|-----------------------------|--------------|----------|---------|---|
| <i>Continued on next page...</i> | | | | | |
| <i>Acids</i> | | | | | |
| HF | Hydrogen fluoride | 7664-39-3 | 20.006 | gas | -61.50 |
| HCl | Hydrogen chloride | 7647-01-0 | 36.461 | gas | -74.84 |
| HBr | Hydrogen bromide | 10035-10-6 | 80.912 | gas | -85.14 |
| HI | Hydrogen iodide | 10034-85-2 | 127.912 | gas | -81.67 |
| HClO ₄ | Perchloric acid | 7601-90-3 | 100.459 | liquid | -88.76 |
| HClO ₄ ·H ₂ O | Perchloric acid monohydrate | | 118.474 | crystal | -32.95 |
| HIO ₃ | Iodic acid | 7782-68-5 | 175.910 | crystal | 8.79 |
| HNO ₃ | Nitric acid | 7697-37-2 | 63.013 | liquid | -33.28 |
| HCOOH | Formic acid | 64-18-6 | 46.026 | liquid | -0.86 |
| CH ₃ COOH | Acetic acid | 64-19-7 | 60.052 | liquid | -1.51 |
| <i>Silver Compounds</i> | | | | | |
| AgClO ₄ | Silver(I) perchlorate | 7783-93-9 | 207.319 | crystal | 7.36 |
| AgNO ₂ | Silver(I) nitrite | 7783-99-5 | 153.874 | crystal | 36.94 |
| AgNO ₃ | Silver(I) nitrate | 7761-88-8 | 169.873 | crystal | 22.59 |
| <i>Cesium Compounds</i> | | | | | |
| CsBr | Cesium bromide | 7787-69-1 | 212.809 | crystal | 25.98 |
| CsBrO ₃ | Cesium bromate | 13454-75-6 | 260.807 | crystal | 50.46 |
| CsCl | Cesium chloride | 7647-17-8 | 168.358 | crystal | 17.78 |
| CsClO ₄ | Cesium perchlorate | 13454-84-7 | 232.356 | crystal | 55.44 |
| CsF | Cesium fluoride | 13400-13-0 | 151.903 | crystal | -36.86 |
| CsF·H ₂ O | Cesium fluoride monohydrate | | 169.918 | crystal | -10.46 |

The study of the physical properties of three mixed solvents, which are: a) Ethylene glycol-water (EG-H₂O); b) glycerol-water (GL-H₂O), and c) Urea-water (urea-H₂O). The study covered the measurements of the specific conductances, densities, viscosities and dielectric constants of the solvent mixtures at four different temperatures in

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the range 293.15 -323.15 K and at a number of compositions . The deviation of the solvent mixtures from an ideal volume mixture relation was examined from the dielectric properties of the mixtures and of the individual constituents.[17,18] The volumetric behaviours of EG-H₂O and GL-H₂O mixtures have been investigated in terms of the excess molar volume. The variation of the dielectric constant deviation function (ΔD) with the volume fraction of the organic component of the mixture was interpreted in terms of the change in the degree of alignment of the dipoles with changing the composition as a consequence of the destruction caused by EG or GL of the three-dimensional hydrogen-bonded water structure. The variation in the relative excess molar volumes of the mixtures was due to a volume contraction (negative ΔV) on mixing the constituents of the mixture. The results were rationalized in terms of the two models for liquid water; one maximally hydrogenbonded and voluminous, the other non-hydrogen-bonded and dense. The effect of the added organic co-solvent to water was to shift this equilibrium in one direction other The possibility of ion-association in the solvent mixtures was examined at a number of solvent compositions and temperatures.[19,20] The results have been expressed in terms of the association constant (K_A)of the electrolytes (AgNO₃ , KC1, KBr, KI, NaCl and LiCl)-It was found that the factors which increase the value of K_A involved the (a) law dielectric constant of the medium in which the electrolyte is dissolved, (b) the small ionic radii which lead to a small value of the ion-size parameter, and (c) the large charges which are carried by the ions. The ideas have first been expressed on Bjerrum's picture of ion-pair formation. The mean activity coefficients (f_{\pm}) of the 1:1 electrolytes at various concentrations could be estimated from the extended Deby-Huckel equation. Values of K_A , for the same electrolytes have also been determined from the experimentally measured conductance data and compared with those derived from Bjerrum picture of ion-pair formation. The agreement of the values obtained by the two sources was substantially high. The association constant and the limiting molar conductance in all cases have been iteratively calculated by a least-square treatment with an appropriate computer programe using Shedlovsky method. The results indicated a good agreement between the values of K_A which have been derived from Bjerrum equation and those obtained by inserting the experimentally measured. conductances, dielectric constant and viscosity date into a set of equations using Shedlovsky method.[21,22] The possibility of the formation of triple-ions in the electrolyte solutions in the mixed solvents has also been examined with the aid of Fouss and Kraus plots. The results indicated the formation of such ions at certain compositions of the solvent mixtures. The thermodynamic functions for ion-association in KCl solutions at each experimental temperatures have also been determined. Ionassociation was found to be quite unfeasible on hermodynamic grounds. 3- The cation transference numbers (t_{\pm}) of several alkali halides in urea-H₂ O mixtures at two different compositions have been determined from E.M.F. measurements of cells with and without transference. [23,24]Attempts have also been made to determine the liquid-junction potential values in cells with transference where two similar electrolytes of different concentrations were in contact. 4- The transference numbers of a number of alkali halides as well as of AgNO₃ in the various solvent mixtures have been r determined from the potential measurements using cells with ransference. The dependence of the transference number on concentration of each electrolyte was also investigated in an attempt to elucidate the values of the limiting transference numbers, [25,26] The Longworth method has been used in many cases of the extrapolation of cation transference numbers in the solvent mixtures. The cation (and anion) Longworth function was then calculated by the method reported . Using the values of the limiting transference number and the appropriate values of the limiting molar conductance it was possible to determine the correspodng values of the limiting ionic conductances. The Walden product for each type of ions in the different solvents could then be obtained from the product of the limiting ionic conductance and the viscosity of the solvent mixture.[27,28]

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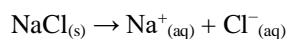
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III. RESULTS AND CONCLUSIONS

Electrolyte solutions are normally formed when salt is placed into a solvent such as water and the individual components dissociate due to the thermodynamic interactions between solvent and solute molecules, in a process called "solvation". For example, when table salt (sodium chloride), NaCl, is placed in water, the salt (a solid) dissolves into its component ions, according to the dissociation reaction[29,30]



It is also possible for substances to react with water, producing ions. For example, carbon dioxide gas dissolves in water to produce a solution that contains hydronium, carbonate, and hydrogen carbonate ions.[31,32]

Molten salts can also be electrolytes as, for example, when sodium chloride is molten, the liquid conducts electricity. In particular, ionic liquids, which are molten salts with melting points below 100 °C, are a type of highly conductive non-aqueous electrolytes and thus have found more and more applications in fuel cells and batteries.

An electrolyte in a solution may be described as "concentrated" if it has a high concentration of ions, or "dilute" if it has a low concentration. If a high proportion of the solute dissociates to form free ions, the electrolyte is strong; if most of the solute does not dissociate, the electrolyte is weak. The properties of electrolytes may be exploited using electrolysis to extract constituent elements and compounds contained within the solution.[33,34]

Alkaline earth metals form hydroxides that are strong electrolytes with limited solubility in water, due to the strong attraction between their constituent ions. This limits their application to situations where high solubility is required.

In 2021 researchers have found that electrolyte can "substantially facilitate electrochemical corrosion studies in less conductive media".[35,36]

In physiology, the primary ions of electrolytes are sodium (Na⁺), potassium (K⁺), calcium (Ca²⁺), magnesium (Mg²⁺), chloride (Cl⁻), hydrogen phosphate (HPO₄²⁻), and hydrogen carbonate (HCO₃⁻). The electric charge symbols of plus (+) and minus (-) indicate that the substance is ionic in nature and has an imbalanced distribution of electrons, the result of chemical dissociation. Sodium is the main electrolyte found in extracellular fluid and potassium is the main intracellular electrolyte; both are involved in fluid balance and blood pressure control.

All known multicellular lifeforms require a subtle and complex electrolyte balance between the intracellular and extracellular environments. In particular, the maintenance of precise osmotic gradients of electrolytes is important. Such gradients affect and regulate the hydration of the body as well as blood pH, and are critical for nerve and muscle function. Various mechanisms exist in living species that keep the concentrations of different electrolytes under tight control.[37,38]

Both muscle tissue and neurons are considered electric tissues of the body. Muscles and neurons are activated by electrolyte activity between the extracellular fluid or interstitial fluid, and intracellular fluid. Electrolytes may enter or leave the cell membrane through specialized protein structures embedded in the plasma membrane called "ion channels". For example, muscle contraction is dependent upon the presence of calcium (Ca²⁺), sodium (Na⁺), and potassium (K⁺). Without sufficient levels of these key electrolytes, muscle weakness or severe muscle contractions may occur.

Electrolyte balance is maintained by oral, or in emergencies, intravenous (IV) intake of electrolyte-containing substances, and is regulated by hormones, in general with the kidneys flushing out excess levels. In humans, electrolyte homeostasis is regulated by hormones such as antidiuretic hormones, aldosterone and parathyroid

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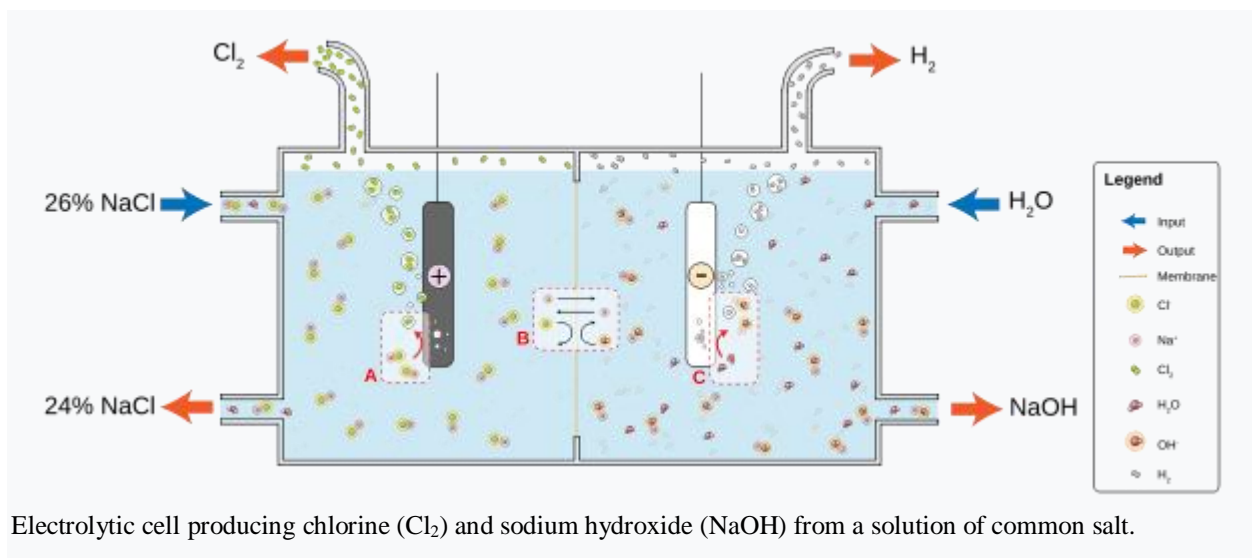
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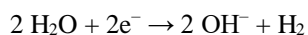
hormones. Serious electrolyte disturbances, such as dehydration and overhydration, may lead to cardiac and neurological complications and, unless they are rapidly resolved, will result in a medical emergency.[39,40]

When electrodes are placed in an electrolyte and a voltage is applied, the electrolyte will conduct electricity. Lone electrons normally cannot pass through the electrolyte; instead, a chemical reaction occurs at the cathode, providing electrons to the electrolyte. Another reaction occurs at the anode, consuming electrons from the electrolyte. As a result, a negative charge cloud develops in the electrolyte around the cathode, and a positive charge develops around the anode. The ions in the electrolyte neutralize these charges, enabling the electrons to keep flowing and the reactions to continue[41,42]



Electrolytic cell producing chlorine (Cl₂) and sodium hydroxide (NaOH) from a solution of common salt.

For example, in a solution of ordinary table salt (sodium chloride, NaCl) in water, the cathode reaction will be



and hydrogen gas will bubble up; the anode reaction is



and chlorine gas will be liberated into solution where it reacts with the sodium and hydroxyl ions to produce sodium hypochlorite - household bleach. The positively charged sodium ions Na⁺ will react toward the cathode, neutralizing the negative charge of OH⁻ there, and the negatively charged hydroxide ions OH⁻ will react toward the anode, neutralizing the positive charge of Na⁺ there. Without the ions from the electrolyte, the charges around the electrode would slow down continued electron flow; diffusion of H⁺ and OH⁻ through water to the other electrode takes longer than movement of the much more prevalent salt ions. Electrolytes dissociate in water because water molecules are dipoles and the dipoles orient in an energetically favorable manner to solvate the ions.[43,44]

In other systems, the electrode reactions can involve the metals of the electrodes as well as the ions of the electrolyte.

Electrolytic conductors are used in electronic devices where the chemical reaction at a metal-electrolyte interface yields useful effects.

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- In batteries, two materials with different electron affinities are used as electrodes; electrons flow from one electrode to the other outside of the battery, while inside the battery the circuit is closed by the electrolyte's ions. Here, the electrode reactions convert chemical energy to electrical energy.
- In some fuel cells, a solid electrolyte or proton conductor connects the plates electrically while keeping the hydrogen and oxygen fuel gases separated.
- In electroplating tanks, the electrolyte simultaneously deposits metal onto the object to be plated, and electrically connects that object in the circuit.[45,46]
- In operation-hours gauges, two thin columns of mercury are separated by a small electrolyte-filled gap, and, as charge is passed through the device, the metal dissolves on one side and plates out on the other, causing the visible gap to slowly move along.
- In electrolytic capacitors the chemical effect is used to produce an extremely thin dielectric or insulating coating, while the electrolyte layer behaves as one capacitor plate.
- In some hygrometers the humidity of air is sensed by measuring the conductivity of a nearly dry electrolyte.
- Hot, softened glass is an electrolytic conductor, and some glass manufacturers keep the glass molten by passing a large current through it.[47]

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