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Investigation of Experimental Density, Apparent Molar Volume, Refractive Index, and Transport Properties of Binary Involving aq. Copper (II) Salts at 308.15K

B. R. Bhosale
Department of Chemistry, Shri Vasantrao Naik Mahavidyalaya Dharni Dist. Amravati

Yogesh I. Biradar
Department of Chemistry, Indira Mahavidyalaya, Ralegaon, dist. Amravati

S.B. Waghmare
Department of Chemistry, Gopikabai Sitaram Gawande Mahavidyalaya, Umarkhed Dist. Yavatmal.
Corresponding author: bhosaleb700@gmail.com

Abstract
The present investigation deals with the experimental and acoustical study of aqueous copper (II) sulfate monohydrate, $CuSO_4 \cdot H_2O$ (CSM), and copper (II) nitrate trihydrate, $Cu(NO_3)_2 \cdot 3H_2O$ (CNT), at 308.15 K over the concentration range of 0.01–0.10 mol kg^{-1}. The density (ρ), ultrasonic velocity (U) and refractive index (n) of the studied aqueous electrolyte solutions were determined experimentally. Using the measured density and ultrasonic velocity data, the apparent molar volume (Φ_v), apparent molar isentropic compressibility, acoustic impedance (Z), and intermolecular free length (L_f) were evaluated. The concentration dependence of these physicochemical and acoustical parameters was analysed to investigate the nature and strength of solute–solvent and solute–solute interactions in the aqueous medium. The observed variation in ultrasonic velocity and derived acoustical parameters is attributed to changes in the hydration environment and molecular packing around the dissolved copper(II) ions and their respective counter-ions. The apparent molar volume and isentropic compressibility provide information concerning volumetric and compressibility effects, whereas acoustic impedance and intermolecular free length reflect changes in the structural characteristics of the solutions. Refractive-index measurements provide complementary information regarding the optical response and molecular interactions of the systems. The comparative behaviour of CSM and CNT solutions provides insight into the influence of sulfate and nitrate ions on the structural and acoustical properties of aqueous copper (II) electrolyte solutions.
Keywords: *- Ultrasonic velocity, viscosity, refractive index Density, apparent molar compressibility, free length.*

1.Introduction

Studying the physicochemical properties of electrolyte solutions provides valuable information about the nature and strength of solute–solvent interactions in aqueous media. Such investigations are particularly important for understanding the structural and dynamical behaviour of ions in solution, which is relevant to coordination chemistry, solution chemistry, electrochemistry, and various industrial and biological processes. Among the different experimental techniques available for investigating molecular interactions in liquids, density, ultrasonic velocity, and refractive-index measurements are simple and sensitive methods for obtaining information about the microscopic structure and intermolecular interactions present in solution [1-3]. Ultrasonic measurements have been extensively employed to investigate the structural characteristics of electrolyte solutions because the propagation of ultrasonic waves through a liquid is strongly influenced by intermolecular forces, compressibility, molecular arrangement, and solute–solvent interactions [4-5]. The experimentally determined ultrasonic velocity, together with density, can be used to evaluate several derived acoustical parameters, such as apparent molar volume, apparent molar isentropic compressibility, acoustic impedance, and intermolecular free length.The variation of these parameters with concentration provides valuable insights into the nature of ion-solvent and ion-ion interactions, as well as the structural changes occurring in the solvent medium that surrounds the ions [6].Density is an important thermodynamic property of liquid mixtures and electrolyte solutions. The variation of density with concentration is indicative of the variations of the molecular packing, hydration and interactions of the issolved species with water molecules [7]. The apparent molar volume is particularly useful for examining the volumetric behaviour of solutes in aqueous solutions and can provide information concerning solute–solvent interactions and hydration effects [8-9]. In the same way, the apparent molar isentropic compressibility calculated from the ultrasonic velocity and density measurements gives information regarding the compressibility and

structural environment of the solution [10]. The concentration dependence of these parameters can therefore be used to understand the changes in the local solvent structure around hydrated ions [11]. Another important acoustical parameter is the acoustic impedance, which is a measure of the resistance offered by a medium to the propagation of an ultrasonic wave [12]. It depends on both density and ultrasonic velocity and is consequently sensitive to changes in the composition and intermolecular interactions of the solution. The intermolecular free length is related to the average distance between the surfaces of neighboring molecules and the compressibility of the medium.Its variation with concentration can provide additional evidence concerning the association, hydration, and structural rearrangement of the constituent species [13]. Refractive index is an optical property that is also sensitive to the composition and molecular structure of a solution. Measurements of refractive index at different concentrations provide information about the interaction of light with the molecular constituents of the medium [14]. When considered together with density and ultrasonic measurements, refractive-index data can provide complementary information about changes in molecular packing and solute–solvent interactions [15].Copper(II) salts are important inorganic electrolytes because their aqueous solutions exhibit significant ion–solvent interactions arising from the hydrated Cu^{2+} ion and the corresponding counter-ions. Copper(II) sulphate and copper(II) nitrate differ in the nature, size, hydration behaviour, and interaction characteristics of their anions [16]. Consequently, aqueous solutions of these salts provide a useful system for investigating how the identity of the counter-ion influences the volumetric, acoustic, and optical properties of electrolyte solutions. Comparative studies of sulfate- and nitrate-containing copper(II) electrolytes can therefore contribute to a better understanding of the influence of ionic composition on solution structure [17-18].

In the present investigation, the density (ρ), ultrasonic velocity (U), and refractive index (n) of aqueous copper (II) sulphate monohydrate, $CuSO_4 \cdot H_2O$ (CSM), and copper (II) nitrate trihydrate, $Cu(NO_3)_2 \cdot 3H_2O$ (CNT), solutions have been measured at 308.15 K over the concentration range 0.01–0.10 mol kg^{-1}. From the experimentally measured density and ultrasonic velocity, the apparent molar volume (Φ_v), apparent molar isentropic compressibility, acoustic impedance (Z), and intermolecular free length (L^f) have been evaluated. The concentration dependence of these physicochemical parameters has been analysed to obtain insights into the nature of solute–solvent and solute–solute interactions in the investigated aqueous systems. A comparative assessment of CSM and CNT solutions is expected to provide useful information on the role of the sulphate and nitrate counter-ions in modifying the structural and acoustical properties of aqueous copper (II) electrolyte solutions.

Materials and Methods

2.Chemicals and Reagents

All weighing operations were carried out using Anamed analytical balance (Model AA-2200) with a precision of ±0.0001 g. The uncertainty of the molality of $CuSO_4 \cdot 5H_2O$ and $Cu(NO_3)_2 \cdot 3H_2O$ was estimated to be within $\pm 5 \times 10^{-4}$ mol·kg^{-1} [19–20]. The solutions were prepared carefully and kept in sealed containers to reduce the uptake of moisture from the atmosphere and loss of solvent by evaporation. The density of copper sulfate and copper nitrite in aqueous solutions of CSM and CNT was determined by using a calibrated pycnometer having an approximate bulb volume of 10 cm^3 [21-22]. The pycnometer was fitted with standardized marks and had a standard bore, sealed with a snug-fitting Teflon stopper. The standard uncertainty of the density measurements was less than ±0.08 kg·m^{-3}. Each solution was measured three times to ensure the accuracy and reproducibility of the densities. A quartz crystal-based ultrasonic interferometer (Model M-F05, Mittal Enterprises) was used to measure the velocity of ultrasonic waves with a stainless-steel sample cell and a digital micrometer[23].

The instrument was operated at a frequency of 2 ± 0.0001 MHz.For each solution, at least 20 readings were taken, and the average micrometer value was used for calculation of ultrasonic velocity. The uncertainty in ultrasonic velocity measurement was estimated to be ±1.09 m·s^{-1} .

All measurements were conducted at a controlled temperature maintained at 308.15 K using an electronically regulated thermostatic water bath (accuracy ±0.1 K). The interferometer was calibrated using triple-distilled water. All measurements were carried out at a constant temperature using a thermally controlled water bath at 308.15 K. (**35** ± 0.1°C) [24-25]

3. Results and Discussion

The experimental volumetric and ultrasonic parameters of aqueous copper nitrate trihydrate and copper sulphate solutions were investigated at 308.15 K over the concentration range of 0.010–0.110 mol cm^{-3}. The measured/calculated parameters include density, apparent molar volume, adiabatic compressibility, apparent molar adiabatic compressibility, acoustic impedance, relative association, free length, Rao's constant and Wada's constant. These parameters provide information regarding the changes in the structure of the aqueous medium and the interactions operating between solute and solvent molecules.

Table 1. Experimental data for ultrasonic velocity, density, adiabatic compressibility, and apparent molar volume of copper(II) nitrate trihydrate [$Cu(NO_3)_2$]·$3H_2O$ and copper (II) sulphate [$CuSO_4$·$5H_2O$] at 308.15 K

[$Cu(NO_3)_2$]·$3H_2O$				
Mol kg^{-1}	**ρ $Kg.m^{-3}$**	**$V_{2\varphi}$ ×10^{-6} m^3·mol^{-1}**	**Ks x 10^{10} Pa^{-1}**	**Ks2φx10^{-15} m^3 mol^{-1} Pa^{-1}**
0.010	0.99786	40.14	4.398	-7.13
0.030	1.00186	40.22	4.391	-7.84
0.050	1.00585	40.53	4.384	-7.79
0.070	1.00981	40.94	4.377	-7.77
0.090	1.01375	41.31	4.370	-7.64
0.110	1.01769	41.48	4.364	-7.46
$CuSO_4$·$5H_2O$				
0.010	0.99743	91.17	4.342	-9.64
0.030	1.00058	91.44	4.331	-9.56
0.050	1.00370	91.70	4.320	-9.39
0.070	1.00680	91.99	4.310	-8.97
0.090	1.00988	92.23	4.300	-8.42
0.110	1.01292	92.66	4.289	-7.78

3.1 Density

The density of the aqueous copper nitrate trihydrate solution increases gradually from 0.99786 to 1.01769 with an increase in concentration from 0.010 to 0.110 mol cm^{-3}. A similar increase is observed for copper sulphate, for which the density increases from 0.99743 to 1.01292 over the same concentration range. The progressive increase in density is primarily due to adding electrolyte to the aqueous medium. As concentration increases, the amount of dissolved solute per unit volume increases, resulting in an increase in the mass density of the solution. In addition to this concentration effect, interactions between the dissolved ions and water molecules influence the packing of the solution. The continuous variation of density indicates that the structure of the aqueous medium is progressively modified with increasing electrolyte concentration. The difference between the density values of the two systems at corresponding concentrations also indicates that the nature of the dissolved salt influences the volumetric behavior of the solution.

Table no.3 ultrasonic velocity (μ) for copper(II) nitrate trihydrate [$Cu(NO_3)_2$]·$3H_2O$ and copper (II) sulphate [$CuSO_4$·$5H_2O$] at 308.15 K

μ $m.s^{-1}$	μ $m.s^{-1}$
1522.09	1520.99
1522.65	1523.30
1523.01	1525.57
1523.48	1527.49
1523.86	1528.98
1524.11	1530.00

3.2 Apparent Molar Volume

For copper nitrate trihydrate, the apparent molar volume increases as the concentration increases. This is calculated by using the formula.

$$V_{2,}\,\Phi = \frac{M}{\rho} + \frac{[1000(\rho_0 - \rho)]}{(m\,\rho\,\rho_0)} \qquad (1)$$

Where, M is molar mass of copper (II) salts, ρ is the density of copper salts Kg m^{-3} and m is the molality of the copper salts mol. Kg^{-1} The observed increase in apparent molar volume suggests that the effective volume contribution of the electrolyte changes with concentration. At low concentration, the ions are surrounded predominantly by water molecules and solute–solvent interactions make an important contribution to the apparent volume. As the concentration increases, the hydration environment changes and interactions between solute species become increasingly relevant.For copper sulphate, the apparent molar volume varies from 91.17 to 92.66 $cm^3\ mol^{-1}$. The magnitude is substantially higher than that obtained for copper nitrate trihydrate. This difference demonstrates that the volumetric contribution of the dissolved species depends strongly on the chemical identity of the electrolyte. The positive concentration dependence of AMV in both systems therefore indicates that the apparent volume of the solute is concentration dependent and that the surrounding aqueous structure changes progressively with increasing electrolyte concentration.

3.3 Adiabatic Compressibility

The adiabatic compressibility of copper nitrate trihydrate decreases from 4.398 to 4.364 with increasing concentration. Copper sulphate shows a similar but somewhat larger decrease, from 4.342 to 4.289. The decrease in adiabatic compressibility indicates that the ability of the solution to undergo volume changes under the ultrasonic pressure wave decreases as the electrolyte concentration increases.

$$\kappa_s = \frac{1}{(u^2\,\rho)} \qquad (2)$$

The presence of dissolved ions modifies the arrangement and mobility of water molecules, thereby changing the compressional response of the solution.The decrease observed for both systems indicates that increasing electrolyte concentration produces a progressively more resistant or compact acoustic medium. The larger change observed for copper sulphate indicates that its addition produces a different magnitude of change in the compressional properties of water compared with copper nitrate trihydrate.

3.4 Apparent Molar Adiabatic Compressibility

The apparent molar adiabatic compressibility values for copper nitrate trihydrate are negative throughout the investigated concentration range. The values vary from approximately −7.13 to −7.84. Negative apparent molar compressibility is generally associated with significant solute–solvent interactions. In an aqueous electrolyte solution, the electric field associated with hydrated ions can influence the orientation and arrangement of neighbouring water molecules. Such changes in the hydration environment can contribute to the negative apparent molar compressibility. For copper sulphate, the apparent molar adiabatic compressibility values range from approximately −9.64 to −7.78. The negative values throughout the concentration range similarly indicate appreciable interaction between the dissolved species and the surrounding aqueous medium. The difference in magnitude between the two electrolytes suggests that their influence on the hydration structure of water is not identical. In particular, the more negative values observed for copper sulphate at lower concentrations indicate a stronger contribution to the compressional response of the hydrated solution under those conditions.

Table-2 Experimental data of Acoustic impedance, Relative association, Free length Rao's constant, and Wada's constant for copper (II) nitrate trihydrate [Cu $(NO_3)_2$]·$3H_2O$ and copper (II) sulphate [$CuSO_4 \cdot 5H_2O$] at 308.15 K

$[Cu(NO_3)_2]\cdot 3H_2O$					
Mol kg^{-1}	**Z**	**RA**	**Lf**	**Rao**	**Wada**
0.010	1.519	1.003	4.352	2785.11	5256.56
0.030	1.525	1.007	4.348	2774.31	5236.71
0.050	1.532	1.011	4.345	2763.53	5217.19
0.070	1.538	1.015	4.341	2752.98	5197.87
0.090	1.545	1.019	4.338	2742.51	5178.86
0.110	1.551	1.023	4.335	2732.03	5159.87
$CuSO_4 \cdot 5H_2O$					
0.010	1.5171	1.002	4.324	2878.79	5444.77
0.030	1.5242	1.006	4.318	2871.19	5429.50
0.050	1.5312	1.009	4.313	2863.68	5414.60
0.070	1.5379	1.013	4.308	2856.06	5399.72
0.090	1.5441	1.016	4.303	2848.27	5385.12
0.110	1.5498	1.019	4.297	2840.36	5370.80

3.5 Acoustic Impedance

The acoustic impedance of copper nitrate trihydrate increases from 1.519 to 1.551 as concentration increases. For copper sulphate, it increases from 1.5171 to 1.5498.Acoustic impedance is dependent on the density and ultrasonic velocity of the medium. Therefore, its systematic increase with concentration reflects changes in the propagation of ultrasonic waves through the solution.

$$Z = \rho u \qquad (3)$$

The increase in acoustic impedance is consistent with the observed increase in density and the changes in compressibility. The addition of electrolyte modifies the physical structure of the aqueous medium, thereby changing its resistance to the propagation of an acoustic disturbance.The similar concentration dependence for both electrolytes indicates a common structural response of the aqueous medium, although the magnitude of the response differs slightly between the two systems.

3.6 Relative Association

The relative association of copper nitrate trihydrate increases from 1.003 to 1.023 with increasing concentration. For copper sulphate, it increases from 1.002 to 1.019.The systematic increase in relative association indicates progressive changes in the association behaviour of the solute and solvent species. The increase suggests that the structure of the aqueous solution changes continuously as the amount of dissolved electrolyte increases.The slightly larger values obtained for copper nitrate trihydrate at corresponding concentrations indicate that its concentration-dependent association behaviour differs from that of copper sulphate.

3.7 Free Length

The free length decreases continuously with increasing concentration. For copper nitrate trihydrate, it decreases from 4.352 to 4.335, while for copper sulphate it decreases from 4.324 to 4.297. The decrease in free length indicates a reduction in the effective intermolecular free space in the solution.

$$L_{(f)} = K \sqrt{\kappa_s} \qquad (4)$$

With increasing electrolyte concentration, the interactions between ions and water molecules alter the local arrangement of the solvent molecules. Consequently, the distance associated with free molecular movement decreases. This behavior is consistent with the decrease in adiabatic compressibility. Both parameters indicate that the acoustic properties of the aqueous medium are progressively altered by the presence of electrolyte.

3.8 Rao's Constant

Rao's constant decreases with increasing concentration for both electrolytes. For copper nitrate trihydrate, it decreases from 2785.11 to 2732.03, whereas for copper sulphate it decreases from 2878.79 to 2840.36.Because Rao's constant is derived from volumetric and ultrasonic properties, its variation reflects changes in the structural and acoustic characteristics of the solution. The observed decrease indicates that the addition of electrolyte modifies the molecular arrangement and acoustic behaviour of the aqueous medium.The higher values obtained for copper sulphate over the investigated concentration range demonstrate that the two electrolytes have different effects on the solution structure.

3.9 Wada's Constant

Wada's constant also decreases progressively with concentration. For copper nitrate trihydrate, the value decreases from 5256.56 to 5159.87, while for copper sulphate it decreases from 5444.77 to 5370.80. The systematic decrease in Wada's constant indicates that the compressional and structural characteristics of the solution vary with electrolyte concentration. Since Wada's constant is related to the acoustic properties of the solution, its concentration dependence provides additional evidence that the addition of electrolyte changes the arrangement and interaction of the constituent species.The higher Wada's constant values obtained for copper sulphate compared with copper nitrate trihydrate indicate a difference in their acoustic response in the aqueous medium.

3.10 Concentration Dependence of Molecular Interactions

The combined behaviour of the measured and calculated parameters provides a consistent picture of concentration-dependent changes in the aqueous electrolyte solutions.For both electrolytes, density increases, while adiabatic compressibility and free length decrease with increasing concentration. At the same time, acoustic impedance and relative association increase. These simultaneous changes indicate that increasing electrolyte concentration modifies the local structure of water and the interaction environment around the dissolved ions. The negative apparent molar adiabatic compressibility values provide additional evidence for appreciable solute–solvent interactions. At lower concentrations, ion–water interactions are expected to make a major contribution to the solution properties because the dissolved ions are surrounded by a relatively large amount of water. With increasing concentration, the average distance between dissolved species decreases, and ion–ion interactions can increasingly contribute to the observed properties. The experimental trends therefore reflect the combined effects of solute–solvent and solute–solute interactions.

3.11 Comparative Behavior of Copper Nitrate Trihydrate and Copper Sulphate

Both electrolyte systems show the same general trends with concentration, but the magnitudes of the calculated parameters are different. Copper nitrate trihydrate shows an increase in AMV from 40.14 to 41.48 $cm^3\ mol^{-1}$, whereas copper sulphate shows an increase from 91.17 to 92.66 cm^3mol^{-1}.The apparent molar adiabatic compressibility is negative for both systems, but copper sulphate shows more negative values at the lower concentrations. The acoustic parameters also exhibit systematic concentration dependence. Acoustic impedance increases for both systems, whereas free length, Rao's constant and Wada's constant decrease. Thus, although both electrolytes produce qualitatively similar changes in the aqueous medium, the numerical differences demonstrate that the magnitude of the interaction and structural response depends on the identity of the electrolyte.

3.12 Conclusion

The experimental results at 308.15 K demonstrate that the volumetric and ultrasonic properties of aqueous copper nitrate trihydrate and copper sulphate solutions are strongly concentration dependent. The systematic increase in density and acoustic impedance, together with the decrease in adiabatic compressibility and free length, indicates progressive modification of the aqueous solution structure. The negative values of apparent molar adiabatic compressibility are consistent with appreciable solute–solvent interactions and changes in the hydration environment around the dissolved species. The simultaneous changes in AMV, relative association, Rao's constant, and Wada's constant further demonstrate that the molecular

arrangement of the solution varies continuously with concentration. The comparison of the two electrolytes shows that the same general trends occur in both systems, but their magnitudes are different. Therefore, the experimental results indicate that both concentration and electrolyte identity play important roles in determining the volumetric, acoustic and interaction properties of aqueous electrolyte solutions. Overall, the combined volumetric and ultrasonic data provide evidence for concentration-dependent solute–solvent and solute–solute interactions in the investigated systems. The observed trends in the different parameters are mutually consistent and demonstrate the usefulness of volumetric and ultrasonic measurements for investigating molecular interactions in aqueous electrolyte solutions.

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