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ULTRASONIC INVESTIGATIONS OF $MnSO_4$, $NiSO_4$ AND CuSO4 AQUEOUS IN POLYVINYL ALCOHOL SOLUTION AT 303K

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ABSTRACT

Ultrasonic velocities and densities have been measured for the aqueous solution of manganese sulphate, nickel sulphate and copper sulphate in polyvinyl alcohol solution, in different concentration at 303K. Adiabatic compressibility, intermolecular free length, acoustic impedance, surface tension and other acoustical parameters have been calculated to assess the polymer-ion interaction. It is seen that the ultrasonic velocity increases initially with the increase in the concentration of manganese sulphate, nickel sulphate and copper sulphate salts in polyvinyl alcohol solution. The increase in velocity with concentrations suggests the increase in cohesive forces due to polymer-solvent interactions. The ion-solvent interaction is relatively weak in the case of polyvinyl alcohol solution containing Mn^{+2} and copper ions. Further study may give more details about complex ion formation.

Key Words: Ultrasonic velocity-$MnSO_4$-$NiSO_4$-$CuSO_4$-Acoustical parameters-Pressure

INTRODUCTION

Recently, a new class of polyelectrolyte materials having great potential for a variety of important applications has been developed. The physical-chemical property of these polymers in solutions depends on the dissociated ionic groups, solvent quality, salt concentration and polymer-substrate interactions. Knowledge of the interaction process of metal ions with macromolecular ligands is of importance for understanding of their physico-chemical behavior. Number of researchers has studied the characteristics of inorganic salts in polymer solution. Effect of inorganic salts on viscosity of acrylonitrile-*N*-vinylpyrrolidone copolymer solutions have studied by Chen Hou *et al.* [1]. Zana studied the ultrasonic characteristics of some polyelectrolyte [2]. Additional investigations of polyelectrolyte solution have been reported by Hiroshi Asai [3], Stefan Köstler *et al.* [4] and Mende Mandy *et al.* [5].
Ultrasonic investigation in aqueous solutions of electrolytes and non-electrolytes with polyvinyl alcohol provides useful tool in understanding the physico-chemical properties of the interacting components [6]. A number of researchers have studied the acoustical behavior of electrolyte solution containing metal ion [7-9]. The ion-dipole interaction depends mainly on ion-size and polarity of the solvent. The strength of ion-dipole attraction is directly proportional to size of the ion, charge and the magnitude of the dipole; but inversely proportional to the distance between the ion and the dipolar molecule [10]. A review on acoustical studies [11-14] of polymer solutions have shown that the ultrasonic velocity and its allied parameters provide more information on molecular interactions which are of the utmost importance for process involving polymer production and their uses [15]. In the present paper, an attempt has been made to determine the densities and ultrasonic velocities of manganese sulphate, nickel sulphate and copper sulphate in polyvinyl alcohol solution at 303K. The data so obtained have been utilized to calculate various acoustic properties for the study of various interactions.

EXPERIMENTAL

Sample preparation and Experimental Techniques

Manganese sulphate (MW=169.10g/mol), Nickel sulphate, (MW= 262.86g/mol, Qualigen's fine chemicals) and Copper sulphate (MW=249.68 g/mol, Fisher Scientific Chemicals) were used to prepare the salt solutions. The above salts are dissolved in required volume of distilled water to prepare the 0.25 N of salt solution.

Polyvinyl alcohol (MW= 125000g/mol Sd fine chemicals Ltd., Mumbai) 1% of polyvinyl alcohol was prepared by dissolving 5 gm of PVA in 500 ml of distilled water. All the chemicals were kept in special air-tight bottles. The weighing was done on electronic balance with the accuracy of 0.01gm.(Digital balance,Model:AD5OB) Polyvinyl alcohol and salt solutions were mixed in different concentrations in the ratio of 90:10, 80:20, 70:30 etc and the ultrasonic velocity was measured. The total volume was maintained as 100ml. The densities of pure liquids and of their binary mixtures were measured using a specific gravity bottle and are accurate to three decimal places.

The ultrasonic velocity in the liquid mixtures have been measured using an Ultrasonic interferometer (Mittal Enterprises, New Delhi) working at 2 MHz frequency with accuracy ± 0.1 ms^{-1}. The density and viscosity are measured using a gravity bottle and an Ostwald's viscometer of accuracy of ± 0.1kgm^{-3} and ± 0.001$mNsm^{-2}$ respectively.

RESULTS AND DISCUSSION

Ultrasonic investigations have been carried out on aqueous solutions of manganese sulphate, nickel sulphate and copper sulphate in polyvinyl alcohol at 303 K. The ultrasonic velocity was measured in a wide range of concentration to investigate the ion-solvent interactions. The experimental values of densities and ultrasonic speed were used to calculate the values of compressibility, intermolecular free length, acoustic impedance, relative association and surface tension.

The solution property parameters namely densities, ultrasonic velocities, adiabatic compressibility, intermolecular free length and surface tension of manganese sulphate, nickel sulphate and copper sulphate in polyvinyl alcohol solution have been measured at different concentrations. Table 1 presents the acoustical parameters for aqueous solution of manganese sulphate in polyvinyl alcohol solution. Figure.1 shows a plot of ultrasonic velocity against the concentration of salt solution. The variation of ultrasonic velocity in a solution depends upon the increase or decrease of intermolecular free length (L_f) after mixing the component. Variations of ultrasonic velocity in the aqueous solution nickel sulphate in polyvinyl alcohol were shown in Table 2. It is seen that the ultrasonic velocity increases initially with the increase in the concentration of manganese sulphate, nickel sulphate and copper sulphate salts in polyvinyl alcohol solution. The increase in velocity with concentrations suggests the increase in cohesive forces due to polymer-solvent interactions [16-17.] Increase in velocity also shows the intermolecular attractions, macromolecular motion in solution. Non-linear variations of ultrasonic velocity were observed at higher concentration of salt solutions in polyvinyl alcohol. It indicates the critical characteristics at a particular composition [18]. A sudden decrease of velocity in the solution of manganese sulphate with polyvinyl alcohol at a concentration of 20:80 indicates the complex formation. Similarly, the dips observed in the solution of copper sulphate in polyvinyl alcohol at a concentration of 30:70 may be due to the presence of dipole-ion interaction in the system [19-20]. The experimental values of ultrasonic velocity, density, adiabatic compressibility, intermolecular free length for the mixture of copper sulphate in polyvinyl alcohol solution were presented in Table III. The observed decrease/increase in adiabatic compressibility, intermolecular free length, acoustic impedance and relative association with composition is an evidence of significant interaction between the component molecules in the binary mixtures.

Adiabatic compressibility (β_{ad}) of the solution was calculated using the formula-

$$\beta_{ad} = 1/(U^2\rho) \qquad (1)$$

Where U is the ultrasonic velocity and ρ is the density of the solution. Adiabatic compressibility is inversely proportional to the square of the ultrasonic velocity. The deviation in adiabatic compressibility can be explained by taking into consideration of the following factors-

1. Loss of di-polar association and difference in size and shape of the component molecules leads to decrease in velocity and increase in compressibility.
2. Dipole-dipole interaction or hydrogen bonded complex formation between unlike molecules leads to increase in sound velocity and decrease of compressibility. The actual deviation depends on the resultant effect.

The compressibility data indicate an ordering interaction leading to the formation of ionic complex. The complex formation in an ionic system is due to ionic association. In this case, the adiabatic compressibility decreases with the increase in concentration and it is given in Figure 2. And it attains sudden increase at higher concentration of salt solution. It means ion-solvent interaction increases at a given composition [21.]

Acoustic impedance (Z) was calculated using the formula

$$(Z) = (U\rho) \tag{2}$$

The acoustic impedance (Z) also shows the same trend of relative association. The variation of acoustic impedance and mole fraction of manganese sulphate, nickel sulphate and copper sulphate salt solution in polyvinyl alcohol is shown in Table 1-3. In the aqueous mixed solution, acoustic impedance increases with increasing the concentration of salt solutions suggesting that the ion-solvent interaction increases, but there is a sudden decrease only in the solution of manganese sulphate in polyvinyl alcohol. It may be due to the complex formation in the solution and on the basis of the interaction between solute and solvent molecules [22.]

Intermolecular free length(L_f) were calculated using the formula-

$$(L_f) = K\sqrt{\beta_{ad}} \tag{3}$$

Where K is the Jocobson's constant, β_{ad} is the adiabatic compressibility of the mixed solution. Intermolecular free length in binary liquid mixtures can be used to access the attraction between the component molecules. Increase in concentration leads to decrease in gap between two species and it is referred as intermolecular free length. The intermolecular free length has an inverse behavior of ultrasonic velocity. The intermolecular free length is found to be a predominant factor in determining the nature of sound velocity variation in liquid mixtures [23.] The intermolecular free length (L_f) decreases with increase in ultrasonic velocity and is given in Figure. 3. This indicates that, there is a strong interaction between the ion and solvent molecules. Also it suggests that a new structure may be formed due to the addition of salts with polyvinyl alcohol solution. The greater interaction between complex species and solvent molecule is due to the size of the molecule compared to the size of the ligand. This also indicates the increase in number of free ions, showing the occurrence of ion-polymer interaction. This supports the close packing of molecules at higher concentration. It shows the ion-solvent interaction increases with concentration of a good solvent [24]. This observation is in accordance with those reported in the case of evaluation of thermo-acoustic parameters of some binary liquid mixtures [25].

Relative association (R_A) of the solution were calculated using the formula

$$(R_A) = (\rho/\rho_o)\,(U_o/U)^{1/3} \tag{4}$$

Where ρ_o and U_o are the density and ultrasonic velocity of the solvent and ρ and U are the density and ultrasonic velocity of solution. Figure 4 shows the variation of relative association of aqueous solution. The relative association is influenced by the breaking up of the solvent molecules on addition of solute to it and subsequent salvation of ions by the free solvent molecules. The former effect results in a decrease while the latter increase the values of relative association. In this present study, the relative association increase with concentration of salt solution in polyvinyl alcohol solution suggest that ion-solvent interaction is varied and it is maximum at higher concentration of salts [26].

The solvent-solute interaction will not only depend upon the structure, resonance stabilization, concentration of solutions, dielectric constant, polarity, hydrogen bonding in the solvents. It also depends upon the nuclear charge of metal ion, tendency of metal ion, ionic size, ionic radius and nuclear repulsion between metal ion-polymer molecules. These factors also influence the interaction between solute and solvent molecules and show changes in other acoustical parameters.

Surface tension (σ) were calculated using the formula-

$$(\sigma) = (6.3\times10^{-4})\,\rho U^{3/2} \qquad (5)$$

Where U is the ultrasonic velocity and ρ is the density of the solution. Surface tension is used to study the surface composition of aqueous solution of the mixtures. A variation of surface tension shows the attractive interactions between the two solutions. The surface tension of a liquid mixture is not a simple function of the surface tension of the pure liquids. At the interface, there is migration of the species having the lowest surface tension, or free energy per unit area, at the temperature of the system. This migration at the interface results in a liquid-phase rich in the component with the highest surface tension and a vapor phase rich in the component with the lowest surface tension. The variation of surface tension is given in Figure 5. It shows the interaction between the solute and solvent molecules [27-28].

The inter-molecular free length values decrease with the increase in ultrasonic velocity. The free length is the distance between the surfaces of the neighboring molecules. It indicates significant interaction between solute and solvent molecules due to which the structural arrangements in the neighborhood of constituent ions is considerably affected [29]. The closer packing molecule indicates the dipole-dipole interaction between polyvinyl alcohol and salt solutions. The structures of solvents and solutes are significantly modified in solutions due to interactions causing wide variation in their properties. The resultant interaction in a solution is not solely due to molecular structure of components of solution but also influenced by other factors like dispersion forces, dipole-dipole interaction, dipole-induced-dipole interaction, hydrogen bonding, charge transfer interaction and or complex formation. In these systems, the slight decrease in U in a higher concentration suggests that, the transition metal ion behave as structure breakers. It indicates the complex formation between unlike molecules [30-31]. It shows the occurrence of ionic dissociation due to weak interaction.

It is found that the plot is steeper for Ni^{+2} indicating stronger ion-dipole interactions in the polyvinyl alcohol solution. It may be due to size effect, smaller charge density and uniform distribution of nickel ions and hence there is strong possibility of interactions between these ions [32.] The ion-solvent interaction is relatively weak in the case of polyvinyl alcohol solution containing Mn^{+2} and copper ions. The polarizibility values of manganese, copper and nickel ions are concentration dependent. At lower concentration, the polarizibility values are almost the same in the above three systems indicating the complete ionization at the dilute solutions, while at higher concentration, it forma complex at a particular concentrations. Copper ion forms the complexes that tend to give planar complex. It has been pointed out that, the added salt in polymer solution interacts with polymer and is termed as image-charge force [33]. In polymer solutions containing salts, the polymer structure is affected by addition of salts solution, either through direct interaction with various polar and ionized groups or indirectly due to the electrostatic effect on the solvent structure [33]. Surface tension increases with addition of metal ions in aqueous solution. The observation is in accordance with the change in mean free length [34]. The cations of the more electropositive metals form weaker Lewis acids that form fewer and less stable complex. Although they are hydrated in solution, the interaction with water is much weaker than in the case of transition of metal ions.

Nonlinearity parameter of salts-polyvinyl alcohol mixed solution calculated by Harymann and Ballou relation:

In the last few years [35-36] a number of theoretical methods have been proposed for estimating the non-linearity parameter (B/A) for pure liquids and liquid mixtures. This parameter has been further correlated with other thermo acoustical parameters [37-38] which is used to deduce the available volume and intermolecular free length of liquid mixtures. General formulation for non-linearity in terms of acoustical parameters of liquids has been made using the experiment for sound velocity (U) and introducing the contribution due to isobaric acoustic parameters (k) and the isothermal acoustic parameter (k”). The expression for B/A is given by [35].

$$B/A = 2k + 2\gamma k''$$

Computations of k and k” require only the knowledge of thermal expansion co-efficient. Detailed method of calculation is given in [37] Hartmann and Balizer [39] obtained the following relation for B/A

$$B/A = 2 + (0.98 \times 10^{4} / U) \qquad (6)$$

Empirical relation proposed by Ballou [40] is given by-

$$B/A = -0.5 + (1.2 \times 10^{4} / U) \qquad (7)$$

Where U is the velocity in m/s. Table 4 shows the variations of the B/A values calculated from Hartmann and Ballou relation and it shows decreased trend with increase in concentration. The B/A values for the liquids have been interpreted as the quantity representing the magnitude of the hardness of liquids. The B/A values are concenerned with interactions between the components of the binary systems. The interaction between the components of the binary mixtures is stronger at higher concentration, while it is weaker at lower concentration of salts [41].

The internal pressure (Pi) is the fundamental liquid property and it is calculated using the non-linear parameter of a solution [42]. It is the resultant of the forces of attraction and repulsion between the molecules in a solution. As the degree of cohesion differs from liquid to liquid, internal pressure provides useful information about the molecular interactions in the liquid systems. Its measurement is significant in the evaluation of thermodynamic properties of liquids because it is closely related to velocity and density in liquid pressure. The calculated values of internal pressure of binary mixtures are shown in Table 5. As the repulsive forces become predominant, the internal pressure gets relatively lowered. From the Figure.6.it is observed that, the addition of salts increase the internal pressure, while a slight decrease in pressure at higher concentration of salts, which confirms the weak induced ionic-induced ionic interaction between the solute and solvent molecules [43]. In the case of manganese sulphate the internal pressure increases with concentration. It has been reported that when the internal pressure increases and the adiabatic compressibility decreases, there is a significant solvent solute interaction [44]. This irregular behavior in different solutions in this binary mixture suggests the existence of different molecular interactions in these solvents.

Table-1: Experimental values of acoustical parameters of manganese sulphate in polyvinyl alcohol aqueous solution in different concentration at 303K

Composition PVA:$MnSO_4$	Mole fraction of $MnSo_4$	Velocity (U) ms^{-1}	Density (ρ) kgm^{-3}	Adiabatic compressibility $(\beta_{ad})10^{-10}$ $kg^{-1}ms^{2}$	Acoustic impedance $(Z)\times10^{6}$ $kgm^{-2}s^{-1}$	Int. Mole Free length (L_f) $10^{-11}m$	Relative Association (R_A)	Surface tension(σ) Nm^{-1}
100:00	0.00000	1517.70	1003	4.3299	1.5217	4.1617	1.000	37,347
80:20	0.99760	1520.89	1009	4.2830	1.5352	4.1391	1.006	37,717
60:40	0.99949	1529.41	1016	4.2073	1.5541	4.1342	1.011	38,290
50:50	0.99968	1531.40	1019	4.1840	1.5607	4.0910	1.013	38,478
40:60	0.99978	1531.00	1023	4.1708	1.5661	4.0845	1.017	38,604
35:65	0.99983	1531.22	1025	4.1627	1.5688	4.0805	1.019	38,677
30:70	0.99986	1533.60	1026	4.1430	1.5739	4.0709	1.020	38,831
25:75	0.99989	1538.60	1028	4.1085	1.5819	4.0539	1.021	39,092
20:80	0.99992	1532.45	1029	4.1371	1.5773	4.0680	1.023	38,900
10:90	0.99997	1547.10	1033	4.0461	1.5975	4.0230	1.023	39,587
00:100	1.00000	1537.80	1038	4.0742	1.5961	4.0369	1.031	39,438

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Table-2: Experimental values of acoustical parameters of nickel sulphate in polyvinyl alcohol aqueous solution at different concentration at 303K.

Composition PVA:$NiSO_4$	Mole fraction of $NiSO_4$	Velocity (U) ms^{-1}	Density (ρ) kg m^{-3}	Adiabatic Compressibility (β_{ad}) $\times 10^{-10}$ $kg^{-1}ms^2$	Acoustic Impedance (Z)$\times 10^6$ kg $m^{-2}s^{-1}$	Int.MoleFree length (L_f) 10^{-11}m	Relative Association (R_A)	Surface tension (σ) Nm^{-1}
100:00	0.00000	1517.70	1003	4.3299	1.5217	4.1617	1.000	37,347
90:10	0.99713	1516.85	1006	4.3219	1.5254	4.1578	1.003	37,428
80:20	0.99872	1518.57	1010	4.2929	1.5340	4.1439	1.007	37,660
70:30	0.99925	1522.77	1014	4.2519	1.5445	4.1240	1.013	37,970
60:40	0.99952	1525.60	1017	4.2252	1.5514	4.1111	1.013	38,175
50:50	0.99968	1528.02	1024	4.1840	1.5642	4.0910	1.019	38,520
40:60	0.99974	1530.32	1024	4.1699	1.5671	4.0841	1.019	38,620.
30:70	0.99986	1533.24	1029	4.1358	1.5770	4.0673	1.022	38,902
20:80	0.99992	1534.45	1032	4.1173	1.5828	4.0583	1.025	39,063
10:90	0.99997	1536.00	1035	4.0956	1.5896	4.0475	1.028	39,249
00:100	1.00000	1533.06	1039	4.0966	1.5923	4.0480	1.032	39,278

Table-3: Experimental values of acoustical parameters of copper sulphate in polyvinyl alcohol aqueous solution at different concentration at 303K.

Composition PVA:$CuSO_4$	Mole fraction of $CuSO_4$	Velocity (U) ms^{-1}	Density (ρ) kgm^{-3}	Adiabatic Compressibility (β_{ad})$\times 10^{-10}$ $kg^{-1}ms^2$	Acoustic Impedance (Z)$\times 10^6$ $kgm^{-2}s^{-1}$	Int.Mole Free length (L_f) 10^{-11}m	Relative Association (R_A)	Surface tension (σ) Nm^{-1}
100:00	0.00000	1517.70	1003	4.3299	1.5217	4.1617	1.000	37,347
90:10	0.99710	1517.63	1007	4.3126	1.5279	4.1534	1.004	37,499
80:20	0.99870	1522.90	1010	4.2685	1.5383	4.1321	1.006	37,821
70:30	0.99925	1521.65	1015	4.2550	1.5445	4.1255	1.011	37,957
60:40	0.99952	1520.93	1018	4.2449	1.5489	4.1206	1.015	38,056
50:50	0.99968	1522.49	1023	4.2191	1.5568	4.1081	1.018	38,269
40:60	0.99979	1531.10	1025	4.1626	1.5690	4.0805	1.019	38,679
30:70	0.99986	1525.49	1029	4.1749	1.5702	4.0865	1.025	38,636
20:80	0.99992	1524.49	1035	4.1577	1.5777	4.0781	1.030	38.809
10:90	0.99996	1528.51	1036	4.1313	1.5836	4.0651	1.031	39,005
00:100	1.00000	1533.27	1047	4.0630	1.6052	4.0314	1.041	39,599

CONCLUSION

The ultrasonic study of metal sulphate aqueous solutions in polyvinyl alcohol confirmed the presence of strong ion-dipole interactions. Nickel sulphate solutions interact strongly with polyvinyl alcohol whereas manganese sulphate and copper sulphate solutions is slightly weak interact with polyvinyl alcohol solution. It shows some complex formation at the concentration of 20:80 of mixed solution of polyvinyl alcohol with manganese sulphate and it is very weak interaction with copper sulphate salt solutions due to the repulsion caused by the electron pairs of the ligands. In conclusion, the concentration, nature of solvent, nature of the solute and nature of the substituent and its position play an important role in determining the interactions occurring in the solutions. Further study may give more details about complex ion formation.

Table-4: Non-linearity (B/A) values of binary mixtures using Hartmann and Balizer equation and Ballou relation

Composition PVA: Salts	Using equation1 MnSo$_4$	Using equation 2 MnSo$_4$	Using equation 1 NiSo$_4$	Using equation 2 NiSo$_4$	Using equation 1 CuSo$_4$	Using equation 2 CuSo$_4$
100:00	8.4571	7.407	8.4571	7.407	8.4571	7.407
90:10	8.4436	7.390	8.4608	7.411	8.4574	7.407
80:20	8.4077	7.346	8.4534	7.402	8.4351	7.380
70:30	8.3994	7.336	8.4356	7.380	8.4404	7.386
60:40	8.4011	7.338	8.4237	7.366	8.4434	7.390
50:50	8.4001	7.337	8.4135	7.353	8.4368	7.382
40:60	8.3902	7.325	8.4039	7.342	8.4006	7.338
30:70	8.3694	7.299	8.3917	7.327	8.4242	7.366
20:80	8.3950	7.331	8.3867	7.320	8.4284	7.372
10:90	8.3344	7.256	8.3802	7.312	8.4115	7.351
00:100	8.3727	7.303	8.3924	7.328	8.3916	7.326

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Table-5: Calculated values of internal pressures (P$_i$) of binary mixtures in different Concentrations at 303K

Concentration of PVA: Salts mixed solution	Internal Pressure(Pi) in PVA:MnSo$_4$ 10^8 Pa	Internal Pressure(Pi) in PVA:NiSo$_4$ 10^8 Pa	Internal Pressure(Pi) in PVA: CuSo$_4$ 10^8 Pa
100:00	2.442	2.442	2.442
90:10	2.472	2.446	2.452
80:20	2.527	2.464	2.483
70:30	2.543	2.493	2.490
60:40	2.550	2.512	2.495
50:50	2.566	2.539	2.512
40:60	2.571	2.550	2.555
30:70	2.598	2.575	2.542
20:80	2.573	2.588	2.551
10:90	2.648	2.603	2.572
00:100	2.619	2.599	2.621

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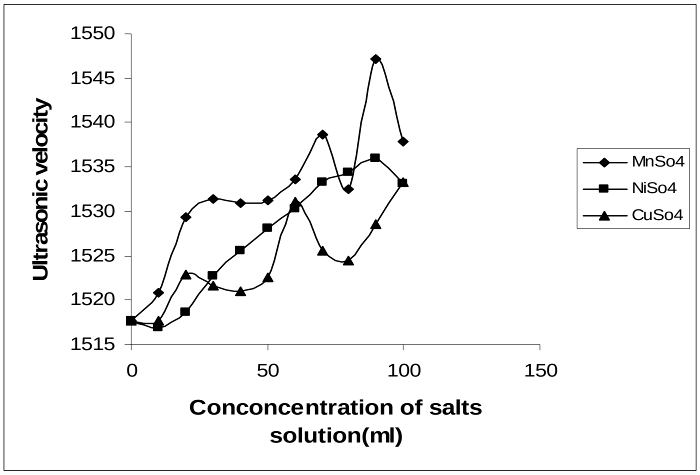

Fig.-1: Variation of ultrasonic velocity with concentration of salts solution in polyvinyl alcohol at 303K

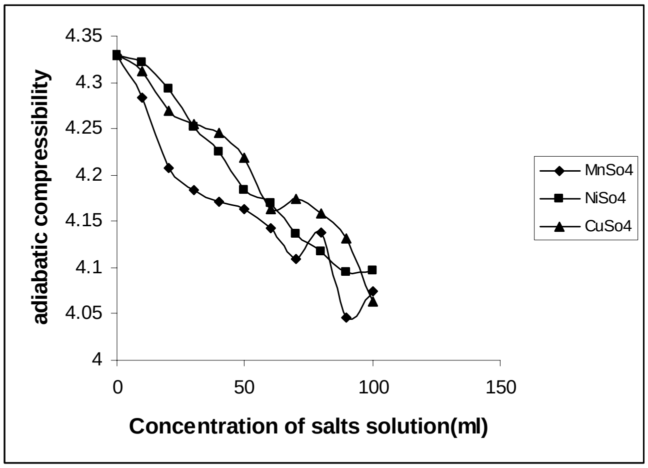

Figure 2 Variation of compressibility with concentration of salts solution in polyvinyl alcohol at 303K.

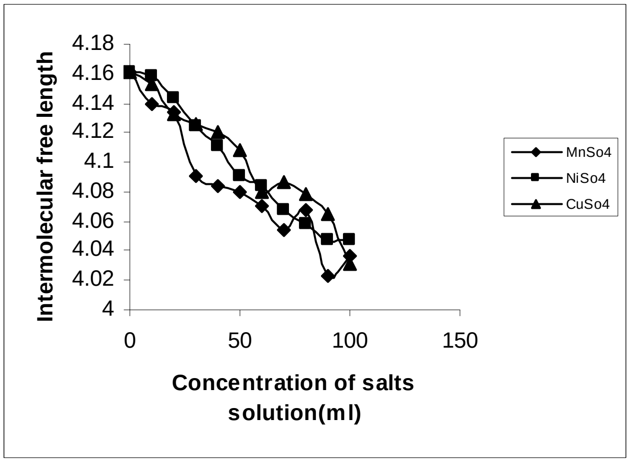

Fig.-3:Variation of intermolecular free length with concentration of salts solution in polyvinyl alcohol at 303K.

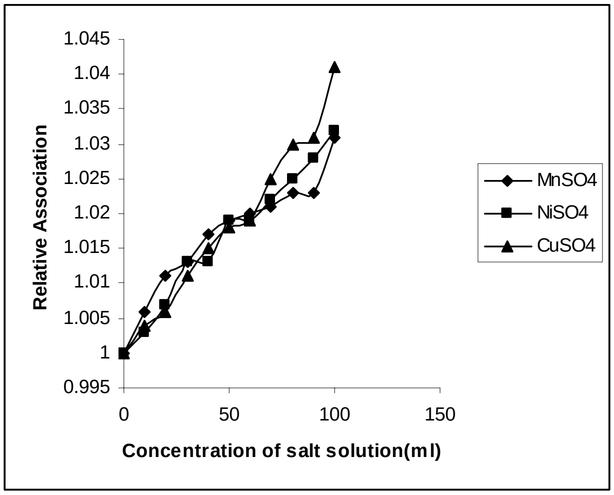

Fig.-4: Variation of relative association with concentration of salts solution in polyvinyl alcohol at 303K.

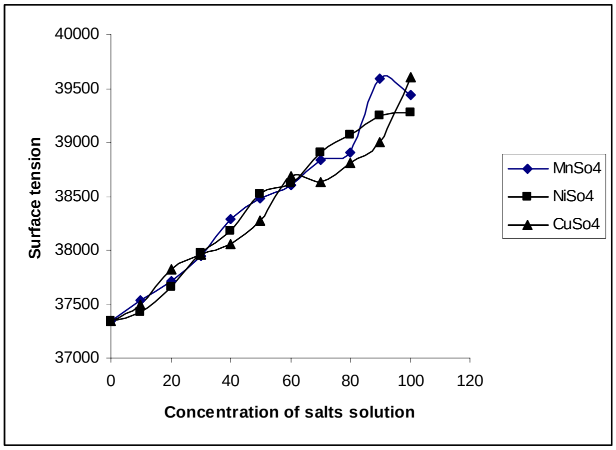

Fig.-5: Variation of surface tension with concentration of salts solution in polyvinyl alcohol at 303K.

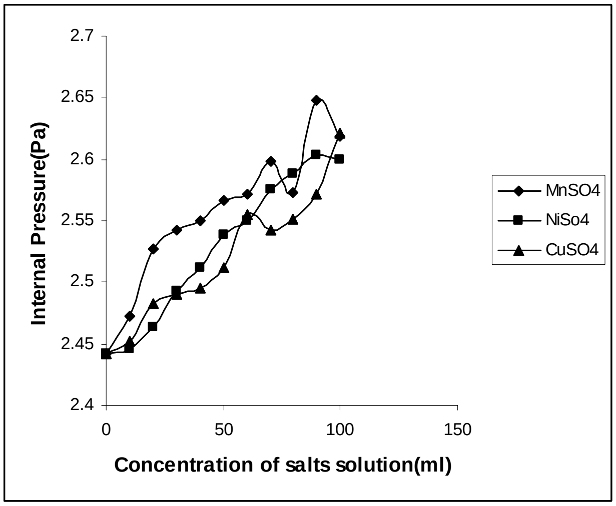

Fig.-6: Variation of internal pressure with concentration of salts solution in polyvinyl alcohol at 303K.

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