



ULTRASONIC VELOCITY AND BULK PROPERTIES OF MULTI-CHARGED ELECTROLYTES (POTASSIUM FERRI AND FERRO CYANIDES) IN AQUEOUS DEXTROSE SOLUTIONS AT DIFFERENT TEMPERATURES.

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ABSTRACT

The ultrasonic velocity (U) of potassium ferricyanide and ferrocyanide has been measured at 298.15 K only. The density (d) and viscosity (η) of both the electrolytes have been measured in the concentration range of 1×10^{-2} to 1×10^{-1} mol dm⁻³ in aqueous binary mixtures containing dextrose (5, 10, 15% (w/w)) at different temperatures ranging from 298.15 to 313.15K. Various important parameters, such as apparent molar volume (V_ϕ), limiting apparent molar volume (V_ϕ^0), apparent molar expansibility (E_ϕ), isentropic compressibility (K_s), solvation number (S_n) and other viscosity parameters have been computed using experimentally determined values of ultrasonic velocity, density and viscosity. In the light of solute-solvent and solute-solute interactions, the variation of these parameters has been enlightened. The viscosity data have been analyzed on the basis of Jones-Dole equation to know the molecular interactions.

KEYWORDS: Aqueous dextrose solutions, ultrasonic velocity, viscosity, molecular interactions.



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INTRODUCTION

Dextrose is a monosaccharide with molecular formula, $C_6H_{12}O_6$ and acts as a source of instant energy, and is available in world market under different brand names. Because of its solubility in water, it is of great interest in many fields of chemistry and molecular biology. The study of the effect of ion-solvent interaction involving multi-charged electrolytes is also of much interest to solution chemists. The study of viscosity and ultrasonic velocity measurements on ferricyanide and ferrocyanide ions in different compositions of water + dextrose mixtures can be useful for obtaining information about the behavior of ions in solutions and the interactions of water and dextrose with each other and with the solute. Analysis of partial molar quantities is of special importance as they give a lot of information regarding the ion-solvent interactions in various solutions. Since these quantities cannot be directly determined from the experiments, it is difficult to study the molecular interaction in ternary mixtures. However, these quantities are related to the corresponding apparent molar quantities which are directly determined. In this article, the values of density (d), ultrasonic velocity (U) and viscosity coefficient (η) of the solutions of potassium ferrocyanide and ferricyanide have been measured in different compositions (5, 10 and 15% (w/w)) of dextrose + water mixture at temperatures ranging from 298.15K to 313.15K at an interval of 5K to elucidate the effect of the medium and particularly the role of ion-ion and ion-solvent interactions on the acoustic, volumetric and viscometric parameters in binary aqueous solutions.

Theoretical Calculations

The apparent molar volume, V_ϕ was calculated from the density data by the standard equation⁷.

$$V_\phi = 1000(cd_0)^{-1} (d_0 - d) + Md_0^{-1} \quad (1)$$

MATERIALS AND METHODS

All chemicals used were of G.R., B.D.H., or AnalaR grades. Conductivity water (Sp. cond. $\sim 10^{-6} \text{ Scm}^{-1}$) was used for preparing water + dextrose (5.0, 10.0 and 15.0 % (w/w)) mixtures. The dextrose content in the mixed solvents was accurate within $\pm 0.01\%$. The salt solutions were prepared on the molal basis and conversion of molality to molarity was done by using the standard expression¹ considering the density values at the respective temperatures. Solutions were kept for 2 hours in a water thermostat maintained at the required temperature accurate to within $\pm 0.05\text{K}$ before use for density measurements. Density measurements were done using a specific gravity bottle (25 ml. capacity) as described earlier². At least five observations were taken and differences in any two readings did not exceed $\pm 0.02\%$. Viscosity measurements were made using an Ostwald viscometer³⁻⁶ in a water thermostat whose temperature was controlled to $\pm 0.05\text{K}$. The values of viscosity so obtained were accurate to within $\pm 0.3 \times 10^{-3} \text{ cP}$. Viscosity values of water at the experimental temperatures were obtained from literature¹. Ultrasonic velocity measurements on the solutions of the salts were made by using an Ultrasonic Interferometer (Mittal Enterprises, New Delhi, India) at a frequency of 2MHz at 298.15K only. The accuracy of the sound velocity measurement is within $\pm 0.5 \text{ m/s}$. The salt content of the solutions varied over a concentration range of 1.0×10^{-2} to $1.0 \times 10^{-1} \text{ M}$ for all these measurements.

where c is the molar concentration, d_0 is the density of the solvent, d is the density of the solution and M is the molar mass of the solute. The V_ϕ data were fitted to the Masson equation⁸ by least squares method⁷ and the limiting apparent volume, V_ϕ^0 was determined from the equation

$$V_\phi = V_\phi^0 + S_V c^{1/2} \quad (2)$$

where S_V is the slope of the V_ϕ versus $c^{1/2}$ plot.

The apparent molar expansibility E_ϕ was calculated by using Eqn (3)⁷,

$$E_\phi = \alpha_0 V_\phi + (\alpha - \alpha_0) 1000 c^{-1} \quad (3)$$

The E_ϕ data were fitted into the Masson equation to get the limiting apparent expansibility (E_ϕ^0) and the slope (S_E)

$$E_\phi = E_\phi^0 + S_E c^{1/2} \quad (4)$$

In Eqn (13), α and α_0 are the coefficient of expansion of the solution and solvent respectively and were obtained from the usual relations^{7,8}. The experimental ultrasonic velocities, U of the solutions of potassium ferri and ferrocyanide in mixed solvents at different concentrations were fitted into an equation⁹ of the form

$$U = U_0 + Fc + Gc^{3/2} + Hc^2 \quad (5)$$

where U_0 is the sound velocity in pure solvent and U is the sound velocity in solutions of electrolytes of different concentrations. F , G and H are the constants. U and U_0 have been found experimentally and F , G and H were calculated by iteration method. From the ultrasonic velocity and density data, the values of adiabatic isentropic compressibility, K_s have been calculated by using the relation (6)⁸⁻¹¹.

$$K_s = \left(\frac{1}{U^2 d} \right) \quad (6)$$

where U = ultrasonic velocity of the solution and d = density of the solution The values of K_s obtained for solutions of different concentrations were fitted to an equation of the form

$$K_s = K_s^0 + Ac + Bc^{3/2} + Cc^2 \quad (7)$$

where A , B and C are the constants. The apparent molar isentropic compressibility, $K_{s,\phi}$ has been computed from equation

$$K_{s,\phi} = 1000 K_s c^{-1} - K_s^0 d_0^{-1} (1000 c^{-1} d - M) \quad (8)$$

The $K_{s,\phi}$ data were fitted to an equation

$$K_{s,\phi} = K_{s,\phi}^0 + F'c^{1/2} + G'c \quad (9)$$

to obtain $K_{s,\phi}^0$ (the limiting apparent molar compressibility). The solvation number, S_n of the solute can be calculated from the isentropic compressibility by the equation

$$S_n = n_1 n_2^{-1} [1 - V K_s (n_1 V_1^0 K_s^0)^{-1}] \quad (10)$$

where

V = Volume of the solution containing n_2 moles of the salt of molecular mass, M

V_1^0 = molar volume of the solvent,

n_1 = number of moles of the solvent

K_s^0 = the apparent isentropic molar compressibility at infinite dilution.

The empirical equation of Jones-Dole¹² relates the relative viscosity of an electrolytic solution to the concentration c of the electrolyte as follows

$$\frac{\eta}{\eta_0} = 1 + A_F c^{\frac{1}{2}} + B_J c \quad (11)$$

where

η = Viscosity coefficient of the solution

η_0 = Viscosity coefficient of the solvent

A_F = Falkenhagen constant, characteristic of the solute

B_J = Jones-Dole coefficient

The constants A_F and B_J were determined from the intercept and slope of the linear plots of $(\eta/\eta_0 - 1)/c^{1/2}$ versus $c^{1/2}$, respectively.

The viscosity data have been analyzed on the basis of transition state theory from the relation,

$$\Delta\mu_2^{0*} = \Delta\mu_1^{0*} + \left(\frac{RT}{\bar{V}_1^0}\right) 1000 B_J - (\bar{V}_1^0 - \bar{V}_2^0) \quad (12)$$

where

$\Delta\mu_2^{0*}$ = contribution per mole of the solute to free energy of activation for viscous flow of the solution.

$$\Delta\mu_1^{0*} = 2.303 RT \log\left(\frac{\eta_0 \bar{V}_1^0}{hN}\right)$$

$\Delta\mu_1^{0*}$ = contribution per mole of the solvent to free energy of activation for viscous flow of the solution)

$$\bar{V}_1^0 = \frac{M_{\text{solvent}}}{d} \quad \text{and} \quad \bar{V}_2^0 = V_\phi^0$$

RESULTS

1. The values of V_ϕ^0 and S_v as obtained from Eqn (2) are given in Table 1.

Table 1

Values of parameters, V_{ϕ}° ($m^3 mol^{-1}$), S_v ($m^{9/2} mol^{-3/2}$), E_{ϕ}° ($m^3 mol^{-1} K^{-1}$), S_E ($m^{9/2} mol^{-3/2} K^{-1}$) for $K_3[Fe(CN)_6]$ and $K_4[Fe(CN)_6]$ in aqueous dextrose solutions at different temperatures.

Wt % D	Temp /K	$V_{\phi}^{\circ} \times 10^{-7}$	$S_v \times 10^{-8}$	$E_{\phi}^{\circ} \times 10^{-3}$	$S_E \times 10^{-4}$
Potassium Ferricyanide					
5.0	298.15	-2.1	-5.2	-7.41	-18.12
	303.15	-4.0	-4.4	-14.20	-15.29
	308.15	-12.5	-1.26	-44.03	-4.42
	313.15	-19.1	1.09	-66.96	3.82
10.0	298.15	-6.2	-4.0	-22.58	-14.56
	303.15	-16.0	-0.66	-58.59	-2.4
	308.15	-13.0	-1.8	-47.09	-6.7
	313.15	-16.0	-0.23	-57.83	-0.85
15.0	298.15	-8.7	-2.7	-29.73	-9.35
	303.15	-8.5	-2.7	-29.02	-9.24
	308.15	-7.8	-2.9	-26.46	-9.87
	313.15	-8.4	-2.8	-28.64	-9.68
Potassium Ferrocyanide					
5.0	298.15	-35.3	3.66	-123.8	12.85
	303.15	-29.9	2.28	-105.2	8.02
	308.15	-30.4	2.27	-106.8	7.98
	313.15	-30.5	2.40	-106.9	8.43
10.0	298.15	-34.6	3.50	-126.9	12.83
	303.15	-40.6	5.65	-148.7	20.72
	308.15	-33.3	3.48	-122.1	12.76
	313.15	-32.0	3.22	-117.2	11.82
15.0	298.15	-49.9	8.96	-169.9	30.53
	303.15	-38.8	4.97	-132.2	16.91
	308.15	-27.0	0.94	-91.84	3.19
	313.15	-33.8	3.64	-115.1	12.41

As observed, the positive and large values of S_v for potassium ferrocyanide in aqueous dextrose solutions at the experimental temperatures indicate the presence of strong ion-ion interactions varying with change in temperature and weight percentage of the dextrose in water. But the reverse is the case in case of potassium ferricyanide in aqueous dextrose solutions. The V_{ϕ}° values are negative for both the salts in all solvents at the experimental temperatures. Since V_{ϕ}° is a measure of ion-solvent interaction, negative value indicates weak ion-solvent interaction or strong ion-ion interaction¹⁴ as evidenced and also provides evidence of electrostriction from viscosity measurements (Table 4). As

observed, the V_{ϕ}° values show an irregular trend as the temperature increases for both the salts in all solvents. The values of E_{ϕ}° and S_E as obtained from Eqn (4) are also given in Table 1. The E_{ϕ}° values are negative for both the salts in all solvents at the experimental temperatures. Since E_{ϕ}° gives an indication of caging or packing effect¹⁵, the large negative values of E_{ϕ}° of potassium ferricyanide as compared to those of potassium ferrocyanide in all weight percentages of dextrose points to the fact that the structure making effect is not favored in the former than in the latter in aqueous dextrose solutions. The ultrasonic velocities of potassium ferricyanide and ferrocyanide in aqueous binary mixture of

dextrose was found to increase as the proportion of dextrose as well as the concentration of potassium salts increase in water. The values of U were fitted to an Eqn (5)

and constants F , G and H calculated for different weight percentages of dextrose, are given in Table 2.

Table 2
Values of U_0 (m/s) and the constants F , G and H at 298.15K.

Electrolyte	Wt% of Dextrose	U_0 (m/s)	F	G	H
$K_3[Fe(CN)_6]$	5	1514	381.2	-100.3	100.4
	10	1528	552.8	-758.1	-645.1
	15	1540	550.8	-678.2	-398.2
$K_4[Fe(CN)_6]$	5	1514	1140.5	-2736.1	1238.7
	10	1528	1426.1	-3495.4	655.4
	15	1540	783.6	-543.6	-884.7

The variation of $(U-U_0)/c$ vs $c^{1/2}$ at very low concentration of dextrose is parabolic and thereafter increases as the concentration of the medium increases with slight deviation in 5wt% dextrose + potassium ferricyanide. This trend is graphically represented in Figure 1 which is in good agreement with Eqn (5).

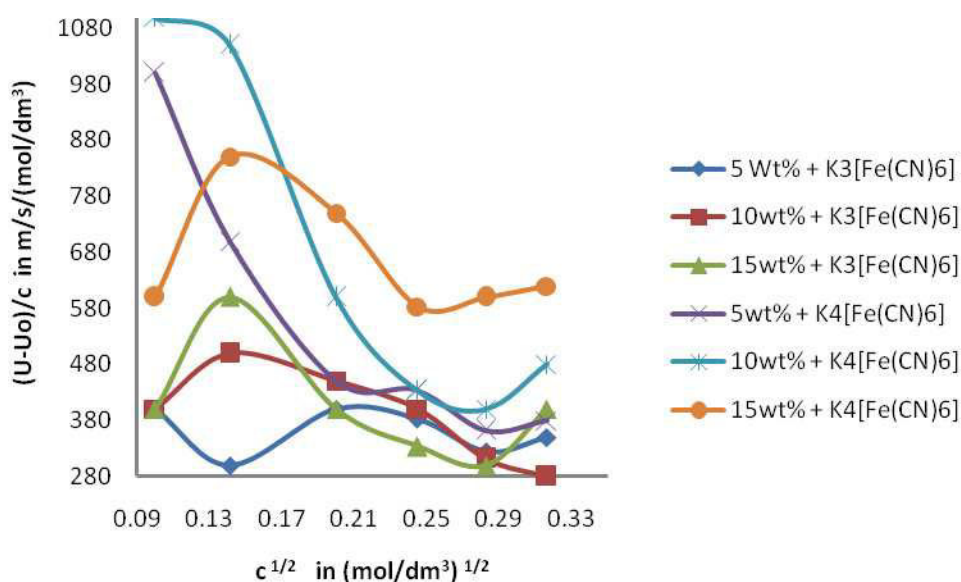


Figure 1 Plot of $(U-U_0)/c$ vs $c^{1/2}$

The values of isentropic compressibility, K_s were calculated by Eqn (6) and were fitted to Eqn (7). The values of the constants A , B and C of Eqn (7) are shown in Table 3. As observed, the K_s^0 values decrease with the increase of the weight percentage of the dextrose and the concentration of the potassium salts in water. The limiting apparent molar compressibility ($K_{s,\phi}^0$) was obtained from Eqn (9) and are given in the Table 3.

Table 3
Values of $K_{s,\phi}^0$ (pa^{-1}) and the constants A, B and C at 298.15K.

Electrolyte	Wt% of Dextrose	$K_{s,\phi}^0 \times 10^{10}$ (pa^{-1})	A $\times 10^{10}$	B $\times 10^8$	C $\times 10^7$	$K_{s,\phi}^0 \times 10^7$ ($\text{m}^3/\text{mol}/\text{pa}$)	$S_n^0 \times 10^{-5}$
$K_3[\text{Fe}(\text{CN})_6]$	5	4.29	-2.53	-4.04	1.57	-3.00	38.28
	10	4.07	-1.54	-3.77	1.47	-0.93	37.14
	15	4.03	-2.85	-3.87	1.49	-5.11	36.10
$K_4[\text{Fe}(\text{CN})_6]$	5	4.25	-2.59	-4.02	1.56	-4.33	38.42
	10	4.13	-3.16	-3.96	1.53	-6.28	37.35
	15	3.97	-3.56	-3.73	1.45	-5.16	36.20

It was observed that apparent molar isentropic compressibility ($K_{s,\phi}$) increases with increase in concentration of the solution. The values of $K_{s,\phi}$ and $K_{s,\phi}^0$ were found to be negative. The negative $K_{s,\phi}^0$ values can be explained in terms of loss of compressibility of surrounding solvent molecules due to strong electrostrictive¹⁵⁻¹⁶ forces in the vicinity of ions causing electrostrictive solvation of the ions. As observed, the $K_{s,\phi}^0$ values increase with an increased proportion of dextrose in water. This indicates that electrostrictive solvation of ions decreases as the dextrose content increases. As observed, both the potassium salts show an irregular trend in $K_{s,\phi}^0$ with increase of dextrose

content in water. As seen in Table 3, the values of solvation number, S_n^0 of the salts are very high in water + dextrose mixtures. This is owing to the fact that the solvation layer formed around the ions is thick and /or hard in water +dextrose mixtures. However, any discrepancy in the values of $K_{s,\phi}^0$ and S_n^0 may be due to characteristic structural changes. S_n^0 values for the potassium ferrocyanide are higher than that of ferricyanide. However, it was observed that as the weight percentage of dextrose increases in water, the solvation number decreases. The reason could be that the ion-solute interaction is much stronger and bulky dextrose molecules are entering more and more in solvation cage leading to low hydration shell.

Table 4
Values of parameters, A_F ($\text{dm}^{3/2} \text{mol}^{-1/2}$) and B_J ($\text{dm}^{3/2} \text{mol}^{-1}$), of the Jones-Dole equation and \bar{V}_1^0 ($\text{m}^3 \text{mol}^{-1}$), \bar{V}_2^0 ($\text{m}^3 \text{mol}^{-1}$), $\Delta\mu_1^{0*}$ (kJ mol^{-1}) and $\Delta\mu_2^{0*}$ (kJ mol^{-1}) of $K_3[\text{Fe}(\text{CN})_6]$ and $K_4[\text{Fe}(\text{CN})_6]$ in aqueous dextrose solutions at different temperatures.

Wt % D	Temp/ K	A_F	B_J	$\bar{V}_1^0 \times 10^6$	$\bar{V}_2^0 \times 10^{-7}$	$\Delta\mu_1^{0*}$	$\Delta\mu_2^{0*} \times 10^{-7}$
Potassium Ferricyanide							
5.0	298.15	-0.045	0.658	18.54	-2.10	-116.20	8.80
	303.15	-0.022	0.687		-4.00	-118.45	9.35
	308.15	0.034	0.436		-13.00	-120.71	6.02
	313.15	0.055	0.338		-10.90	-122.89	4.74
10.0	298.15	-0.006	0.530	19.09	-6.20	-115.92	6.88
	303.15	0.007	0.425		-16.00	-118.30	5.59
	308.15	0.008	0.550		-13.00	-120.55	7.37
	313.15	-0.020	0.663		-16.00	-122.79	9.03

15.0	298.15	-0.008	0.4417	19.69	-8.70	-115.61	5.55
	303.15	0.008	0.3485		-8.50	-117.83	4.45
	308.15	-0.022	0.5020		-7.80	-120.11	6.52
	313.15	-0.007	0.5680		-8.40	-122.35	7.50
Potassium Ferrocyanide							
5.0	298.15	-0.004	0.592	18.54	-2.10	-116.20	7.92
	303.15	0.044	0.511		-4.00	-118.45	6.94
	308.15	0.096	0.279		-13.00	-120.71	3.85
	313.15	0.145	0.085		-10.90	-122.89	1.18
10.0	298.15	0.051	0.387	19.09	-6.20	-115.92	5.02
	303.15	0.056	0.331		-16.00	-118.30	4.36
	308.15	0.079	0.371		-13.00	-120.55	4.96
	313.15	0.043	0.516		-16.00	-122.79	7.02
15.0	298.15	0.034	0.358	19.69	-8.70	-115.61	4.50
	303.15	0.074	0.177		-8.50	-117.83	2.26
	308.15	0.042	0.355		-7.80	-120.11	4.62
	313.15	0.051	0.429		-8.40	-122.35	5.67

A perusal of table 4 shows that the values of A_F are positive in all weight percentages of dextrose excepting for 298.15K in case of potassium ferrocyanide. In case of potassium ferricyanide, the values of A_F show an irregular trend. Since A_F is considered to be a measure of ion-ion interaction, the low positive A_F values may indicate the presence of weak ion-ion interaction or very strong ion-solvent interactions in the solutions concerned. However, in the case of ionic interactions, it is evident that the interactions are dependent on the nature of the solute and also structure of the solvent. The co-efficient B_J is a measure of the effective solvodynamic volume of solvated ions, and is governed by ion-solvent interactions, i.e., the structural effect of the solvent in solution. It is a fact that when a solute dissolves in a solvent, some of the solvent molecules are attached to the ions (produced from the solutes) because of ion-solvent interactions, and this causes an increase in viscosity of the solution (a positive contribution to the co-efficient, B_J). On the other hand, these solvent molecules have to be

wrenched out of the bulk solvent and this breaking of the solvent structure causes a decrease in viscosity of the solution (a negative contribution to B_J). Thus, the value of B_J in the solution is the resultant of these two opposing factors. The values of the coefficient B_J for all the salts are positive at all temperatures indicating thereby the existence of ion-solvent interactions. The viscosity data were also analyzed on the basis of the transition state theory for relative viscosity of the potassium salt solutions as suggested by Feakins et. al¹⁷ using equation (11). The values of \bar{V}_1^0 and \bar{V}_2^0 and that of $\Delta\mu_1^{0*}$ and $\Delta\mu_2^{0*}$ are recorded in Table 4. The values of $\Delta\mu_1^{0*}$ are found to be negative for both the salts in all solvents at all temperatures showing negative contribution per mole of the solvent to free energy of activation for viscous flow of the solution. As observed, the values of $\Delta\mu_2^{0*}$ are positive showing greater contribution per mole of the solute to free energy of activation for viscous flow of the solution, and are in good agreement

with the B_J values. The values of $\Delta\mu_2^{0*}$ indicate that the behavior of $\Delta\mu_2^{0*}$ is quite similar to that of B_J in all cases. It follows that the formation of the transition state is accompanied by the rupture and distortion of the intermolecular forces in solvent structure¹⁸.

CONCLUSION

The ultrasonic velocities were found to increase for both the electrolytes with increase in proportion of dextrose as well as the concentration of electrolytes. The higher sound velocity value of potassium ferrocyanide is due to its higher mass in the mixed solvent. The isentropic compressibility, K_s^0 and solvation number, S_n^0 values decrease as the weight

percentage of the dextrose in water increases which may be due to occupation of the interstitial spaces of water by dextrose molecules thereby making the medium less compressible. Thus, it represents the degree of electrostrictive solvation. The negative value of limiting apparent molar volume (V_ϕ^0) indicates weak ion-solvent interaction and also provides evidence of electrostriction in all the solvents.

ACKNOWLEDGEMENT

The authors would like to thank Department of Chemistry, ITER, Siksha O Anusandhan University, Bhubaneswar, for their financial support and providing the research facilities.

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