

Ionic Interaction Studies of Some Amino Acids in Aqueous Cadmium Chloride Solutions at 308.15K.

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ABSTRACT

Apparent molar compressibilities (ϕ_{κ}) and apparent molar volume (ϕ_V) of L-serine, L-asparagine and L-lysine in aqueous and aqueous solutions of cadmium chloride at different concentrations have been determined at the temperature 308.15K from precise density, ultrasonic velocity and time flow measurements using a specific gravity bottle, ultrasonic single frequency interferometer and Ostwald type capillary viscometer respectively. From these data limiting apparent molar compressibilities (ϕ_{κ}°), limiting apparent molar volumes (ϕ_V°), and their constants (S_{κ} , S_V), viscosity A & B-coefficients and the corresponding transfer parameters ($\Delta\phi_{\kappa}^{\circ}$, $\Delta\phi_V^{\circ}$ and ΔB), and hydration number (n_H) have been calculated. Transfer parameters have been discussed in terms of ion-solvent and ion-ion interactions in the studied mixtures.

INTRODUCTION

Amino acids are the building blocks of all living organisms and incorporate structural features of proteins, their physico-chemical and thermodynamical properties in aqueous solutions are found to provide valuable informations that are important in understanding the stability of proteins. Some of these interactions are found implicated in several biochemical and physiological process in a living cell^[1]. Measurements of ultrasonic velocity and density values of amino acids in aqueous electrolytes have been of interest with a view to improve the comprehension about the stability native proteins and equilibrium process between "folded" versus "unfolded" forms of proteins^[2]. Studies on the solubility and stability of proteins have generated a great interest for a long time, but because of complications involved in dealing with these complex molecules, various low molecular weight model compounds are generally taken for investigations^[3]. Therefore, physico-chemical properties of amino acids, peptides, and their derivatives which mimic some specific aspects of proteins structure in aqueous solutions have been extensively studied to gain better understanding of solute-solvent interactions and their role in the conformational stability of proteins^[4].

Salts solutions are known to produce remarkable effects on the conformation and properties of proteins^[5]. The interactions between solvent and various constituent groups of a protein such as the amino acids chain and the peptide backbone group play a central role in the structure, the conformation, and the function of proteins in aqueous solutions^[6]. It has been established that metal ions play crucial role in various biological processes. They are generally involved in enzyme regulation, stabilization of structure of reactive molecules, transportations to transmembrane channels and so forth^[7]. Cadmium ions are mainly found in the biological systems in the form of Zn/Cd metallothionein (cysteine-rich protein) complex. Metallothionein are the only biomolecules known to normally contain cadmium. Therefore cadmium ion is found in association with proteins whose functions are probably involved with essential trace element (zinc) metabolism^[8]. The effect of ions of salt on the solution behaviour of amino acids can be studied through the acoustic volumetric and viscometric properties. Such studies on the solutions behaviour of amino acids are of great relevance because all biological fluids are not pure water and additions have a significant impact on protein stability and functions^[9]. In this paper we report the densities, ultrasonic velocities, and viscosities of L-serine, L-asparagine and L-lysine in aqueous and aqueous CdCl₂ solutions of (0.5 and 1.0) mol.dm⁻³ at 308.15K. From these data, various transfer parameters $\Delta\phi_{\kappa}^{\circ}$, $\Delta\phi_V^{\circ}$, ΔB , and hydration number n_H have been calculated. These results have been discussed in terms of various interactions operating in these systems.

MATERIALS AND METHODS

Analytical reagent (AR) and spectroscopic reagent (SR) grades which minimum assay of 99.9% of L-serine, L-asparagine, L-lysine and cadmium chloride were obtained from E-Merck, Germany and SD fine chemicals, India, which are used as such without further purification. Doubly distilled degassed water with specific conductance less than $1.29 \times 10^{-6} \Omega^{-1} \text{cm}^{-1}$ was used for the preparation of all solutions. Solutions of cadmium chloride (0.5 and 1.0 mol. dm^{-3}) were prepared by volume and used on the day they were prepared. Solutions of amino acids in the concentration range of (0 - 0.1 mol. dm^{-3}) were made by volume on the molarity concentration scale with a precision of $\pm \times 1 \times 10^{-4} \text{g}$ on an electronic digital balance (Model: SHIMADZU AX -200). The density was determined using a specific gravity bottle by relative measurement method with an accuracy of $\pm 0.01 \text{ kgm}^{-3}$. An ultrasonic interferometer having the frequency of 3 MHz (Mittal Enterprises, New Delhi, Model: F-81) with an overall accuracy of $\pm 0.01\%$ has been used for velocity measurements. An electronically digital operated constant temperature bath (Raaga Industries) has been used to circulate water through the double walled measuring cell made up of steel containing the experimental solution at the desired temperature. The accuracy in the temperature measurement is $\pm 0.1 \text{ K}$. Solution viscosities were measured by Ostwald type capillary Viscometer, which was placed in a water thermostat having temperature stability. Flow time measurements were performed using digital chronometer within $\pm 0.01 \text{ s}$ (Model: CASIO HS -10W). The average of at least six readings was used as the final efflux time. The measured viscosity values have an uncertainty of $\pm 0.001 \text{ m.Pa.s}$.

THEORY AND CALCULATIONS

Using the measured data, the following volumetric, compressibility and transport parameter have been calculated using the standard relations.

$$\text{Adiabatic compressibility } \beta = \frac{1}{U^2 \rho} \quad (1)$$

Molar hydration number has been computed using the relation

$$n_H = \left(\frac{n_1}{n_2} \right) \left(1 - \frac{\beta}{\beta_0} \right) \quad (2)$$

Where, β and β_0 are adiabatic compressibilities of solution and solvent respectively, n_1 and n_2 are number of moles of solvent and solute respectively.

The apparent molar compressibility has been calculated from relation,

$$\varphi_k = \frac{1000}{M \rho_0} (\rho_0 \beta - \rho \beta_0) + \left(\frac{\beta_0 M_w}{\rho_0} \right) \quad (3)$$

Where, β , ρ and β_0 , ρ_0 are the adiabatic compressibility and density of solution and solvent respectively, M is the molar concentration of the solute and M_w the molecular weight of the solute. φ_k is the function of M as obtained by Gucker (1993)^[10] from Debye Huckel^[11] and is given by

$$\varphi_k = \varphi_k^0 + S_k M^{1/2} \quad (4)$$

where, φ_k^0 is the limiting apparent molar compressibility at infinite dilution and S_k is a constant. φ_k^0 and S_k of equation 4 have been evaluated by least square method.

The apparent molar volume φ_V has been calculated using the relation:

$$\varphi_V = \left(\frac{M_w}{\rho} \right) - \left(\frac{1000(\rho - \rho_0)}{M \rho \rho_0} \right) \quad (5)$$

The apparent molar volume φ_V has been found to differ with concentration according to empirical relation as:

$$\varphi_V = \varphi_V^0 + S_v M^{1/2} \quad (6)$$

where, φ_V^0 is the limiting apparent molar volume at infinite dilution and S_v is a constant and these values were determined by least square method.

The viscosity A and B coefficients for the amino acids in aqueous cadmium chloride solutions were calculated from the Jones-Dole equation^[12].

$$\left(\frac{\eta}{\eta_0}\right) = 1 + AM^{1/2} + BM \quad (7)$$

where, η and η_0 are the viscosities of the solution and solvent respectively. A is determined by the ionic attraction theory of Falkenhagen –Vernon^[13] and therefore also called Falkenhagen coefficient. B or Jones-Dole coefficient is an empirical constant determined by ion-solvent interactions.

Transfer adiabatic compressibility ($\Delta\phi_k^0$), transfer volume ($\Delta\phi_v^0$) and transfer viscosity coefficient (ΔB) of each amino acid from water to aqueous cadmium chloride solutions have been calculated as:

$$\Delta\phi_y^0 = \phi_y^0 \text{ (in aqueous cadmium chloride solution)} - \phi_y^0 \text{ (in water)} \quad (8)$$

where, ϕ_y^0 denotes limiting apparent molar compressibility ϕ_k^0 , limiting apparent molar volume ϕ_v^0 and viscosity coefficient B.

RESULTS AND DISCUSSION

The experimental values of density (ρ), viscosity (η) and ultrasonic velocity (u) for different molarity composition of each of the three amino acids viz., L-serine, L-asparagine and L-lysine in aqueous and aqueous cadmium chloride solutions (0.5 and 1.0 mol.dm⁻³) at 308.15K are shown in the Table-1. Further, the values of adiabatic compressibility (β), apparent molar compressibility (ϕ_k), apparent molar volume (ϕ_v), limiting apparent molar compressibility (ϕ_k^0), limiting apparent molar volume (ϕ_v^0), and their constants (S_k, S_v), transfer adiabatic compressibility ($\Delta\phi_k^0$), transfer volume ($\Delta\phi_v^0$), viscosity A&B coefficients and ΔB are shown in Tables 2-3.

In all the three amino acids system the values of density (Table-1) increases with increase in molar concentration of amino acids as well as cadmium chloride (CdCl₂) content. This increasing trend suggests a moderate strong electrolytic nature in which the solutes (amino acids) tend to attract the solvent (aqueous cadmium chloride) molecules. Further, it is observed from the Table-1 that the values of ultrasonic velocity increases with increase in concentration of amino acids but it is found to decrease with increasing the CdCl₂ content in all the three systems studied. The factors apparently responsible for such behaviour may be the presence of interactions caused by the proton transfer reactions of amino acids in water and cadmium chloride mixtures. Molecular interaction is thus responsible for the observed increase in density and ultrasonic velocity in these mixtures. The increase in ultrasonic velocity in these mixtures may be attributed to the cohesion brought about by the ionic hydration.

In all the three systems the value of adiabatic compressibility (Table 2) decreases with increase in concentration of amino acids as well as CdCl₂. The decrease in adiabatic compressibility is attributed to the influence of the electrostatic field of ions on the surrounding solvent molecules (Cd²⁺, Cl⁻) so called electrostriction. The magnitude of β values is larger in L-serine than other two amino acids. The larger β value which shows molecular associations/interactions is greater in L-serine than that of other two amino acids. Amino acid molecules in the neutral solution exist in the dipolar form and thus have stronger interaction with the surrounding water molecules. The increasing electrostrictive compression of water around the molecules results in a large decrease in the compressibility of the solutions^[14]. The interaction between the solute and water molecules in the solvent is termed hydration. From the Table-2 it is observed that the values of n_H are positive in all the systems studied and such positive values of n_H indicate an appreciable solvation of solutes. These values are found to decrease with increasing the content of amino acids as well as cadmium chloride in all the three systems studied. The decreasing values of n_H which indicate the increase in solute-co-solute interaction. The decreasing behaviour of n_H shows that cadmium chloride has a dehydration effect on the amino acids.

The negative values of ϕ_k and ϕ_v Table-2 are increases with increase in concentration of amino acids as well as CdCl₂. The negative values of ϕ_k and ϕ_v in all systems indicate the presence of ion-solvent interactions. The limiting apparent molar compressibility ϕ_k^0 provides information regarding ion-solvent interactions and S_k , that of ion-ion interactions in the solution. From the Table-3 it is observed that ϕ_k^0 values are negative and it increases with increasing the concentration of cadmium chloride in all the systems studied. Appreciable negative values of ϕ_k^0 and its behaviour for all the systems reinforce our earlier view that existence of ion-solvent interaction in the mixtures. The magnitude of ϕ_k^0 is in the order L-serine > L-asparagine > L-lysine. The values of S_k exhibits positive value and it decreases with increasing the concentration of cadmium chloride in all the three amino acids. The positive values of S_k suggest that ion-ion interactions are relatively stronger. It is well known that solutes causing electrostriction

lead to decrease in the compressibility of the solution. This is reflected by the negative values of ϕ_k^o of the amino acids.

The volume behaviour of a solute at infinite dilution is satisfactorily represented by ϕ_v^o which is independent of the ion-ion interactions and provides information concerning ion-solvent interactions. Table-3 reveals that the negative values of ϕ_v^o increases with the addition of cadmium chloride contents in all the systems studied. The increase in ϕ_v^o may be attributed to the increased hydrophilicity polar character of the side chain of the amino acids. The magnitude of ϕ_v^o is in the order of L-serine > L-asparagine > L-lysine.

It is evident from the Table-3 that the positive S_v indicates the presence of strong ion-ion interactions and less complex ion formation taking place in the ternary mixtures^[15]. The negative S_v values are associated with hydrophobic solutes^[16]. Generally, the interactions between amino acids and cadmium chloride can be classified into: (i) ion-charged group interactions occurring between Zwitter ions (NH_3^+ and COO^-) of amino acids and cations (Cd^{2+}) and anions (Cl^-) of CdCl_2 and (ii) ion-non polar group interactions occurring between ions of cadmium chloride (Cd^{2+} , Cl^-) and the hydrophobic side chain of amino acids. The $\Delta\phi_k^o$ and $\Delta\phi_v^o$ values can also be explained on the basis of co-sphere overlap model^[17] in terms of solute-cosolute interactions. According to this model, ion-charged group interactions contribute positively, whereas ion-non polar interactions contribute negatively to the $\Delta\phi_k^o$ and $\Delta\phi_v^o$ values. Therefore, the presently observed positive $\Delta\phi_k^o$ and $\Delta\phi_v^o$ (Figures 1 & 2) values in the presence of CdCl_2 on the whole indicate that ion-charged group interactions dominate over the ion-nonpolar group interactions in these solutions.

Table 1: Values of density (ρ), viscosity (η) and ultrasonic velocity (U) of each amino acids in aqueous cadmium chloride solutions at 308.15K.

M/(mol.dm ⁻³)	$\rho / (\text{kgm}^{-3})$			$\eta / (x 10^{-3} \text{Nsm}^{-2})$			U / (m.s ⁻¹)		
Water + cadmium chloride (CdCl_2)									
	0.0M	0.5M	1.0M	0.0M	0.5M	1.0M	0.0M	0.5M	1.0M
System - I : Water + cadmium chloride + L-serine									
0.00	994.0	1064.7	1125.4	0.7190	0.7999	0.8844	1527.3	1524.2	1521.0
0.02	996.7	1065.8	1127.0	0.7238	0.8056	0.8986	1529.2	1527.4	1523.4
0.04	997.5	1066.9	1128.3	0.7245	0.8063	0.9044	1530.4	1528.8	1525.8
0.06	998.4	1067.7	1129.1	0.7273	0.8168	0.9071	1532.6	1530.4	1528.8
0.08	998.8	1069.2	1130.4	0.7292	0.8218	0.9096	1534.8	1532.2	1530.6
0.10	999.6	1069.9	1131.2	0.7312	0.8265	0.9240	1537.2	1535.6	1533.8
System - II : Water + cadmium chloride + L-asparagine									
0.00	994.0	1064.7	1125.4	0.7190	0.7999	0.8844	1527.3	1524.2	1521.0
0.02	997.5	1066.6	1127.6	0.7240	0.8092	0.9018	1531.8	1528.4	1526.4
0.04	998.6	1067.9	1129.3	0.7249	0.8100	0.9064	1533.0	1531.2	1528.8
0.06	999.4	1068.9	1130.1	0.7283	0.8187	0.9102	1537.2	1534.2	1531.2
0.08	1001.2	1070.3	1131.0	0.7323	0.8249	0.9129	1541.4	1535.4	1534.9
0.10	1003.1	1071.8	1132.0	0.7364	0.8397	0.9252	1543.8	1539.0	1536.6
System - III : Water + cadmium chloride + L-lysine									
0.00	994.0	1064.7	1125.4	0.7190	0.7999	0.8844	1527.3	1524.2	1521.0
0.02	998.2	1067.1	1129.1	0.7345	0.8146	0.9164	1532.6	1531.0	1528.2
0.04	999.3	1068.4	1131.4	0.7436	0.8200	0.9224	1534.2	1532.0	1530.4
0.06	1000.1	1069.7	1132.4	0.7504	0.8289	0.9285	1539.4	1537.2	1535.4
0.08	1002.8	1071.2	1133.7	0.7638	0.8422	0.9383	1544.2	1539.6	1537.6
0.10	1004.0	1072.9	1134.9	0.7649	0.8479	0.9474	1547.3	1543.2	1540.4

Table 2: Values of adiabatic compressibility (β), molar hydration number (n_H), apparent molar compressibility (Φ_k) and apparent molar volume (Φ_V) of each amino acids in aqueous cadmium chloride solutions at 308.15 K.

M / (mol.dm ⁻³)	$\beta / (x 10^{-10} \text{ m}^2 \text{ N}^{-1})$			n_H			$\Phi_k / (x 10^{-7} \text{ m}^2 \text{ N}^{-1})$			$\Phi_V / (x 10^{-3} \text{ m}^3 \text{ mol}^{-1})$		
Water + cadmium chloride (CdCl ₂)												
	0.0M	0.5M	1.0M	0.0M	0.5M	1.0M	0.0M	0.5M	1.0M	0.0M	0.5M	1.0M
System - I : Water + cadmium chloride + L-serine												
0.00	4.3128	4.0429	3.8409	--	--	--	--	--	--	--	--	--
0.02	4.2905	4.0218	3.8234	14.36	13.29	10.56	1.7007	1.2639	1.1480	136.16	48.37	62.98
0.04	4.2803	4.0103	3.8070	10.47	10.28	10.23	1.1922	1.0239	1.0949	88.14	48.32	57.00
0.06	4.2642	3.9989	3.7894	10.43	9.24	10.36	1.1282	0.9232	1.0688	73.79	43.89	48.44
0.08	4.2503	3.9839	3.7761	10.06	9.29	9.78	1.0416	0.9511	1.0233	60.33	49.31	49.04
0.10	4.2336	3.9637	3.7577	10.20	9.98	10.64	1.0350	0.9895	1.0789	56.26	45.55	45.47
System - II : Water + cadmium chloride + L-asparagine												
0.00	4.3128	4.0429	3.8409	--	--	--	--	--	--	--	--	--
0.02	4.2725	4.0135	3.8063	25.95	18.53	20.87	2.7743	1.8307	2.1054	176.35	83.52	86.55
0.04	4.2611	3.9940	3.7887	16.65	15.41	15.75	1.7915	1.5263	1.6378	115.71	70.22	76.58
0.06	4.2344	3.9747	3.7742	16.83	14.33	13.42	1.6972	1.4025	1.3790	90.45	61.37	61.46
0.08	4.2039	3.9633	3.7530	17.53	12.54	13.19	1.7518	1.2608	1.3314	90.29	61.29	54.86
0.10	4.1959	3.9392	3.7414	15.06	13.07	12.01	1.5638	1.2262	1.2203	91.12	62.09	51.68
System - III : Water + cadmium chloride + L-lysine												
0.00	4.3128	4.0429	3.8409	--	--	--	--	--	--	--	--	--
0.02	4.2651	3.9980	3.7923	30.71	28.29	29.32	3.2962	2.7956	3.0614	211.47	105.45	145.43
0.04	4.2515	3.9879	3.7738	19.74	17.33	20.25	2.1074	1.7262	2.1894	133.21	81.15	117.64
0.06	4.2194	3.9562	3.7459	20.05	18.21	19.11	1.9979	1.6614	1.9815	102.09	72.99	91.39
0.08	4.1820	3.9383	3.7309	21.06	16.48	16.60	2.1123	1.6160	1.7291	110.17	71.07	81.16
0.10	4.1602	3.9138	3.7134	19.66	16.27	15.39	1.9599	1.6024	1.5992	100.02	71.61	74.22

The $\Delta\phi_k^0$ values increase in the order L-serine > L-asparagine > L-lysine at all concentration of CdCl₂. The positive $\Delta\phi_k^0$ values may be attributed to a decrease in the shrinkage volume in the presence of CdCl₂ solutions. This may be due to stronger interactions between amino acids and ions of co-solute.

A perusal of Table-3 shows that in all the three systems the values of A and B coefficients are positive and these values are increases with increasing the concentration of CdCl₂. Since A is a measure of ionic interaction, it is evident that the positive A-coefficient has strong ion-ion interaction whereas negative A-coefficient has weak ion-ion interactions in the mixtures studied [18]. The viscosity B-coefficient reflects the effect of ion-solvent interactions on the solution viscosity. The positive B values suggest the presence of strong ion-solvent interaction. Further, ΔB values show in Figure 3 which supports the result obtained from $\Delta\phi_k^0$ and $\Delta\phi_V^0$ values discussed above. The magnitude of ΔB is in the order L-serine > L-asparagine > L-lysine.

TABLE 3: Values of limiting apparent molar compressibility (ϕ_k^0), limiting apparent molar volume (ϕ_v^0) and their constants S_k and S_v , transfer adiabatic compressibility ($\Delta\phi_k^0$), transfer volumes ($\Delta\phi_v^0$), A and B coefficients of Jones-Dole equation and transfer viscosity coefficient (ΔB) of each amino acids in aqueous cadmium chloride solution at 308.15 K.

Amino acids	CdCl ₂ M/ (mol.dm ⁻³)	$-\phi_k^0$ /($\times 10^{-7}$ m ² N ⁻¹)	$-\phi_v^0$ /($\times 10^{-3}$ m ³ .mol ⁻¹)	S_k /($\times 10^{-7}$ N ⁻¹ m ⁻¹ .mol ⁻¹)	S_v /($\times 10^{-3}$ m ³ L ^{1/2} .mol ^{-3/2})	$\Delta\phi_k^0$ /($\times 10^{-7}$ m ² N ⁻¹)	$\Delta\phi_v^0$ /($\times 10^{-3}$ m ³ .mol ⁻¹)	A /(dm ^{3/2} . mol ^{-1/2})	B /(dm ^{3/2} mol ⁻¹)	ΔB /(dm ^{3/2} mol ⁻¹)
L-serine	0.0	2.07	188.93	3.60	447.13	--	--	0.0391	0.0287	--
	0.5	1.40	49.75	1.56	11.22	0.67	139.18	0.0119	0.0369	0.3408
	1.0	1.20	76.47	0.51	100.76	0.87	112.46	0.0942	0.0869	0.0582
L-asparagine	0.0	2.47	226.41	2.32	479.30	--	--	0.0176	0.1663	--
	0.5	1.58	97.65	5.60	126.36	0.89	128.76	0.0032	0.4162	0.2499
	1.0	2.02	116.57	2.04	212.37	0.45	109.84	0.1311	0.2111	0.0448
L-lysine	0.0	3.87	273.77	6.62	600.59	--	--	0.1034	0.3436	--
	0.5	3.35	125.37	6.19	189.46	0.52	148.4	0.0610	0.4008	0.0572
	1.0	3.37	159.73	8.03	243.66	0.50	114.04	0.2622	0.3654	0.0218

Figure 1: Variation of transfer adiabatic compressibility ($\Delta\phi_k^0$) of some amino acids with molarity of aqueous cadmium chloride solutions at 308.15 K.

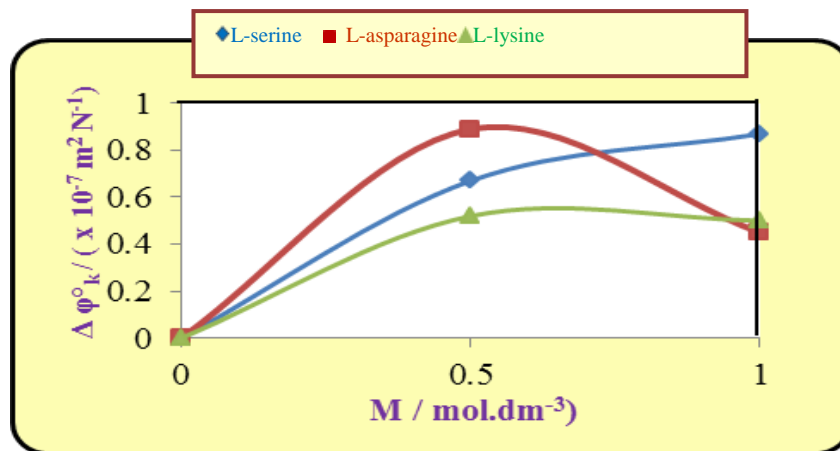


Figure 2: Variation of transfer volume ($\Delta\phi_v^0$) of some amino acids with molarity of aqueous cadmium chloride solutions at 308.15 K.

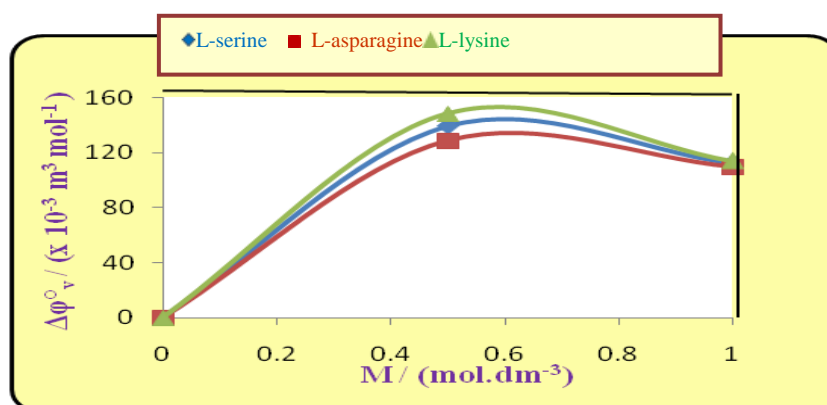
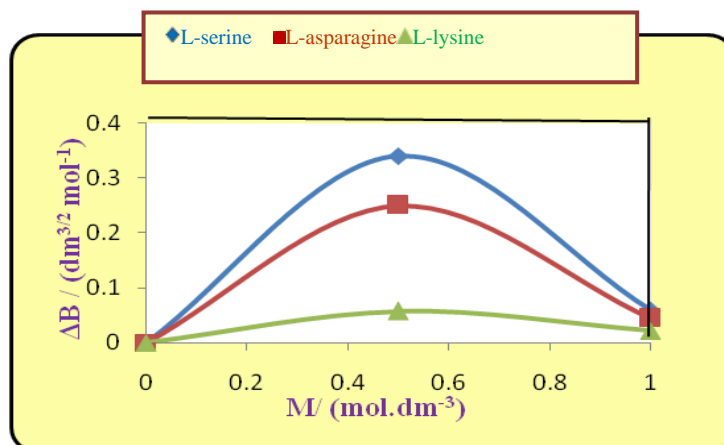


Figure. 3 Variation of transfer viscosity coefficient (ΔB) of some amino acids with molarity of aqueous cadmium chloride solutions at 308.15 K



CONCLUSION

In the summary, limiting apparent molar adiabatic compressibility ϕ_k^0 , limiting apparent molar volume ϕ_v^0 , viscosity B-coefficients of L-serine, L-asparagine and L-lysine in aqueous solutions of CdCl₂ have been determined at 308.15 K. The various transfer values ($\Delta\phi_k^0$, $\Delta\phi_v^0$ and ΔB) are positive in all cases and decreases with increases in the concentration of CdCl₂. From the magnitude of ϕ_k^0 , $\Delta\phi_k^0$ and ΔB it can be concluded that L-serine possess strong ion-solvent inter action than the other two amino acids. The transfer properties suggest that ion-charged group interactions are dominating over ion-non polar group interactions.

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