Molecular Interaction Studies of some Amino Acids with Aqueous K₂SO₄solution at 308.15 K

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Abstract - **Apparent molar compressibilities (φ_K) and apparent molar volumes (φ_V), of L-valine, L-asparagine and L-arginine in aqueous and aqueous potassium sulphate solutions (0, 0.5, and 1.0 mol.dm^{-3}) at 308.15 K have been determined from precise density and ultrasonic velocity measurements. Limiting apparent molar compressibilities (φ_K^0) and limiting apparent molar volumes (φ_V^0) and their constants (S_K, S_V) of these amino acids at infinite dilution is evaluated. Transfer adiabatic compressibilities ($\Delta\varphi_K^0$), transfer volumes ($\Delta\varphi_V^0$) at infinite dilution from water to aqueous potassium sulphate solutions and molar hydration number (n_H) have been calculated. Viscosity A and B coefficients of Jones-Dole equation have been determined from viscositymeasurements. These parameters have been interpreted in terms of ion-solvent and ion-ion interactions in the studied solutions.**

Keywords – **Viscosity coefficients, apparent molarcompressibility, apparent molar volume, ultrasonic velocity, hydration number.**

I. INTRODUCTION

In recent years, the studies of acoustical properties of amino acids in aqueous mixed electrolytic solutions have been found to be useful in understanding the specific ion-ion and ion-solvent interaction in solutions. A number of researchers have employed the ultrasonic technique to evaluate the thermodynamic properties of amino acids, peptides and proteins. The accurate measurement of density, viscosity, ultrasonic velocity and hence the derived parameters will give significant information regarding the state of affairs in a solution [1]. Viscosity is one of the most important factors affecting the mobility of ions clusters [2]. Proteins, which have enzymatic, structural, and regulatory functions, carry out the vast majority of all biological processes in cells. Amino acids and peptides have been taken up as model compounds for understanding the behavior of more complex protein molecules in solutions. It is well-known that electrolytes influence the stability of proteins [3]. Metal ions are essential for the biological function of many proteins. At least one-third of all proteins encoded in the human genome appear to contain metal ions which perform a wide range of specific functions [4]. Salt-induced precipitation is an extensively used method as an initial step to purify proteins because of its selectivity and low cost. Thus, the study of the interactions of ions and proteins is useful to understand the physiological systems and in the separation and purification processes of proteins. As amino acids are the building blocks of the proteins, their study provides important information which can be related to the behavior of larger biomolecules such as proteins. Amino acids also play a significant role in metabolism and in many neurochemical response mechanisms, such as memory,

appetite control, and pain transmission [5,6]. They are also used as food additives and have many applications in the pharmaceutical industries. Studies of the effect of concentration of salt and temperature on the thermodynamic properties of aqueous amino acid solutions have been proven to be very useful in elucidating the various interactions that occur in these solutions. The volumetric and compressibility studies of amino acids in aqueous salt solutions have been employed to understand the nature of interactions operative in solutions. K_2SO_4 influences the unfolding of protein [7].Thus, the study of interactions in amino acid/aqueous K_2SO_4 systems is significant with a view to understanding the interactions in protein-aqueous K_2SO_4 systems.This study focuses on the measurements of density (ρ) viscosity (η) and ultrasonic velocity (U) values of L-valine, L-asparagine and L-arginine in aqueous and aqueous potassium sulphate (0, 0.5, and 1.0 mol.dm^{-3}) solution at 308.15 K. The measured density, viscosity and ultrasonic velocity data have been used to compute the adiabatic compressibility, molar hydration number, apparent molar compressibility, apparent molar volume, limiting apparent molar compressibility, limiting apparent molar volume, and their constants,transfer adiabatic compressibility, transfer volume and viscosity A and B coefficient of Jones-Dole equation values with the view to understanding the various types of interactions operative in the solutions.

II. MATERIALS AND METHODS

Analytical reagent (AR) and spectroscopic reagent (SR) grades with minimum assay of 99.9% of L-valine, L-asparagine, L-arginine and potassium sulphate 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 $10^{-6}\Omega^{-1}$cm^{-1} was used for the preparation of all solutions. Aqueous solutions of potassium sulphate (0.5 and1.0 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 $\pm 1 \times 10^{-4}$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 $\pm$ 0.01kgm^{-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, Chennai, India. Model: ULTRA COLD CHAMBER - 437) 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 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 $\pm$ 0.01s (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 $\pm$ 0.001m.Pa.s.

III. THEORY AND CALCULATIONS

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

$$\beta = \frac{1}{U^2\rho} \tag{1}$$

Molar hydration number has been computed using the relation

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

Where, β and βo 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,

$$\varphi_K = \frac{1000}{M\rho_0}(\rho_o\beta\text{-}\rho\beta o) + \left(\frac{\beta o M_w}{\rho_o}\right) \tag{3}$$

where, β, ρ and βo, ρ_o 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)[8] from Debye Huckel[9] and is given by

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

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:

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

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

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

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 potassium sulphate solutions were calculated from the Jones-Dole equation [10].

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

Where, η and η_o are the viscosities of the solution and solvent respectively. A is determined by the ionic attraction theory of Falkenhagen – Vernon[11] and therefore also called Falkenhagen coefficient. B or Jones-Dole coefficient is an empirical constant determined by ion-solvent interactions.

Transfer adiabatic compressibility ($\Delta\varphi_K^0$) and transfer volume ($\Delta\varphi_V^0$) of each amino acid from water to aqueous potassium sulphate solutions have been calculated as:

$$\Delta\varphi_y^0 = \varphi_y^0 \text{ (in aqueous potassium sulphate) - } \varphi_y^0 \text{ (in water)} \qquad (8)$$

Where φ_y^0 denotes, limiting apparent molar compressibility φ_K^0, limiting apparent molar volume φ_V^0.

IV. RESULTS AND DISCUSSION

The experimental values of density (ρ), viscosity (η) and ultrasonic velocity (U) for different molar composition of each of the three amino acids viz., L-valine, L-asparagine and L-arginine in aqueous and aqueous potassium sulphate solutions are shown in Table I. The values of adiabatic compressibility (β), molar hydration number (n_H), 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 ($\Delta\varphi_K^0$), transfer volume ($\Delta\varphi_V^0$) and viscosity A and B coefficient of Jones-Dole equation were calculated and the results are given in Tables II and III. Further, the Figs.1-2 shows the variation of transfer adiabatic compressibility and transfer volume of L-valine, L-asparagine and L-arginine in aqueous potassium sulphate solutions at 308.15 K and the curves are drawn using least square fitting.

In all the three amino acids system (Table I) the values of density and ultrasonic velocity increases with increase in molar concentration of amino acids as well as potassium sulphate (K_2SO_4) content. This increasing trend suggests a moderate strong electrolytic nature in which the solutes (amino acids) tend to attract the solvent (aqueous potassium sulphate) molecules. Molecular interaction is thus responsible for the observed increase in density and ultrasonic velocity in these mixtures. The factors apparently responsible for such behaviour may be due to the presence of

interactions caused by the proton transfer reactions of amino acids in water + potassium sulphate mixtures. The increase in ultrasonic velocity in these solutions may be attributed to the cohesion brought about by the ionic hydration [12].

In all the three systems the value of adiabatic compressibility (Table II) decreases with increase in concentration of solute (amino acids) as well as increase in concentration of aqueous K_2SO_4. The decrease in adiabatic compressibility is attributed to the influence of the electrostatic field of ions (NH_3^+ and COO^-) on the surrounding solvent molecules (K^+, SO_4^{2-}) so called electrostriction. 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. This result in the present study generally confirms the conclusions drawn earlier from the velocity data.

The interaction between the solute and the water molecules present in the solvent can be termed as hydration. From Table II it is observed that the positive values of n_H indicate an appreciable solvation of solutes [13]. This is an added support not only for the structure promoting tendency of the solutes but also for the presence of appreciable dipole-dipole interactions between solute and water molecules. This also leads further suggestion that the compressibility of the solution will be less than that of the solvent. As a result, solutes will gain mobility and hence there will be more probability of conducting solvent molecules. This may further enhance the interaction between solute and solvent molecules. The values of n_H decreases with increase in solute content in an aqueous medium as well as in potassium sulphate solutions. The decreasing behaviour of n_H shows the strength of interaction gets weakened between the ion-solvent molecules, but however it increases the ion-ion interaction in the

mixtures [14]. The following observations have been made on φ_K and φ_V (Table II) of the three amino acids in aqueous potassium sulphate solutions at 308.15 K.

(i) The values of φ_K and φ_V are all negative over the entire range of the molarity.

(ii) The negative values of φ_K and φ_V are vary nonlinearly with the increase in concentration of amino acids as well as K_2SO_4 contents.

(iii) The magnitude of φ_V is in the order:

L-valine> L-asparagine > L-arginine.

The above observations clearly suggests that the negative values of φ_K and φ_V in all systems indicate the presence of ion-solvent interactions. The observed behaviour of φ_K and φ_V reveals that strengthening of the ion-solvent interaction in all systems studied. The negative values of φ_V indicate electrostrictive solvation of ions [15]. From the magnitude of φ_V, it can be concluded that stronger molecular association is found in L-arginine than other two amino acids and hence L-arginine is a more effective structure maker.

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 Table III, it is observed that φ_K^0 values are negative and it increases with increasing the concentration of potassium sulphate in all systems studied.

Appreciable negative values of φ_K^0, and its behaviour for all systems reinforce our earlier view regarding existence of ion-solvent interaction in the mixtures. The values of S_K exhibits positive value and it decreases with increasing the concentration of potassium sulphate in all the three amino acids. This behaviour indicates the existence of ion-ion interaction with

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increase in potassium sulphate content and suggests structure making / breaking effect of the amino acids.

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 of the amino acids.

The volume behaviour of a solute at infinite dilution is satisfactorily represented by φ_V^0 which is independent of the ion-ion interactions and provides information concerning ion-solvent interactions. Table III reveals that the values of φ_V^0 are negative in all the three studied amino acids. The values of φ_V^0 decreases with the addition of potassium sulphate contents with respect to aqueous medium in all the systems studied. The decrease in φ_V^0 may be attributed to the increased hydrophobicity / non-polar character of the side chain of the amino acids. The magnitude of φ_V^0 is in the order: L-arginine < L-asparagine < L-valine. It is evident from the Table III that S_V is positive in all the three systems suggesting the presence of strong ion-ion interactions.

The values of transfer adiabatic compressibility $\Delta\varphi_K^0$ and transfer volume $\Delta\varphi_V^0$ (Table III) are negative and it increases with increase in the concentration of potassium sulphate in all the three amino acid systems which suggest the existence of ion-solvent interactions in the mixtures. The magnitude of $\Delta\varphi_V^0$ is in order: L-arginine < L-asparagine < L-valine. Generally, the interactions between amino acids and potassium sulphate can be classified as:

(i) ion-ion interactions among the   ,   , and
    (and   ) zwitterionic end groups.

(ii) Ion-hydrophilic interactions between ions and hydrophilic groups ($-CONH_2$, $-CONH$) of amino acids.

(iii) Ion-nonpolar group interactions occurring between ions and the nonpolar groups ($-CH_2/-CH_3$) of amino acids.

The $\Delta\varphi_V^0$ values can also be explained on the basis of co-sphere overlap model [16] in terms of solute–co-solute interactions. According to this model, ion-ion group interactions and ion-hydrophilic interactions contribute positively, whereas ion–non-polar group interactions contribute negatively to the $\Delta\varphi_V^0$ values.
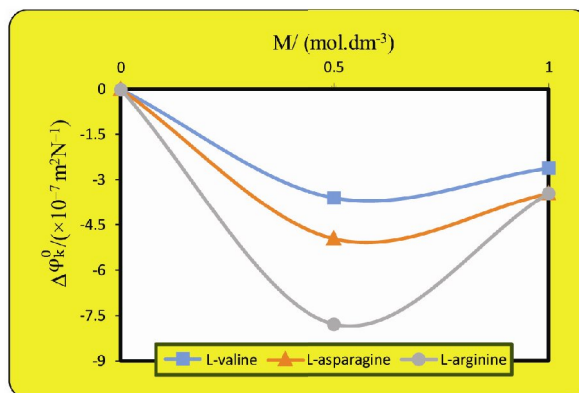

Fig.1 Variation of transfer adiabatic compressibility (Δ) of some amino acids with molarity of aqueous potassium sulphate solutions at 308.15 K
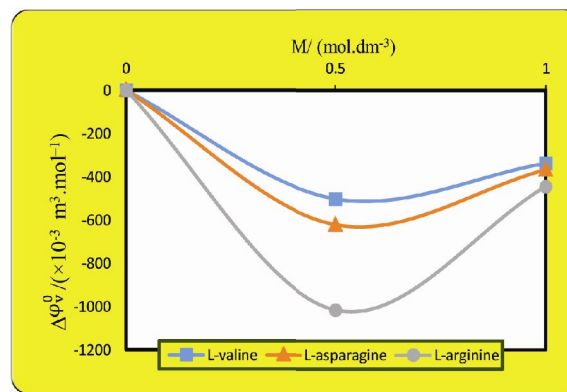

Fig. 2 Variation of transfer volume (Δ) of some amino acids with molarity of aqueous potassium sulphate solutions at 308.15 K

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Therefore, from Figs. 1-2, the negative $\Delta\varphi_K^0$ and $\Delta\varphi_V^0$ values observed in all the three amino acids suggest that the interaction contribution of type (iii) is stronger than that of type (i) and (ii).

Viscosity is another important relation in understanding the structure as well as molecular interactions occurring in the mixtures. From Table I, it is observed that the values of viscosity increases with increase in molar concentration of amino acids as well as potassium sulphate content. This increasing trend indicates the existence of ion-solvent interaction occurring in these systems. In order to shed more light on this, the role of viscosity coefficients have been obtained. From the Table III, it is observed that the values of A coefficient is positive for all the system indicating the presence of ion-ion interactions [17]. Further, the values of the B-coefficient are positive in all systems studied. B-coefficient is also known as measure of order and disorder introduced by the solute into the solvent. It is also a measure of ion-solvent interaction and relative size of the ion and solvent molecules. The behaviour of B-coefficient in all the three systems suggests the existence of strong ion-solvent interaction.

The magnitude of B values is in the order L-arginine > L-asparagine > L-valine. This conclusion is in excellent agreement with that drawn from φ_V^0 and $\Delta\varphi_V^0$.

V. CONCLUSION

In the present work, volumetric, compressibility and transport parameters of L-valine, L-asparagine and L-arginine in aqueous and aqueous potassium sulphate solutions (0, 0.5 and 1.0 mol.dm^{-3}) at 308.15 K were obtained using density, viscosity and ultrasonic velocity data and the results have been used to study the existence of ion-solvent interactions. From the

magnitude of φ_V^0, $\Delta\varphi_V^0$ and the values of B-coefficient it can be concluded that L-arginine possess strong ion-solvent interaction than the other two amino acids. The transfer adiabatic compressibility $\Delta\varphi_K^0$ and transfer volume $\Delta\varphi_V^0$ data suggest that ion-nonpolar group interactions are dominating over the ion-ion and ion-hydrophilic group interactions.

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TABLE I

Values of density (ρ), viscosity (η), and ultrasonic velocity (U) of amino acids in aqueous potassium sulphate solutions at 308.15K

M/(mol.dm^{-3})	ρ / (kgm^{-3})			η / (x 10^{-3} Nsm^{-2})			U / (m.s^{-1})		
	water + potassium sulphate (K$_2$SO$_4$)								
	0.0 M	0.5M	1.0 M	0.0M	0.5M	1.0M	0.0M	0.5M	1.0M
System - I : water + potassium sulphate + L-valine									
0.00	994.0	1008.2	1040.3	0.7190	0.7775	0.8309	1528.6	1584.9	1614.9
0.02	995.6	1016.5	1046.9	0.7235	0.7845	0.8399	1529.2	1585.0	1615.9
0.04	997.2	1017.9	1047.1	0.7257	0.7930	0.8439	1531.8	1587.6	1616.5
0.06	999.8	1019.7	1048.3	0.7325	0.7961	0.8461	1532.5	1590.8	1617.5
0.08	1001.2	1021.8	1051.2	0.7394	0.7997	0.8503	1534.2	1593.3	1618.9
0.10	1003.5	1022.5	1052.4	0.7486	0.8069	0.8541	1535.6	1594.2	1619.4
System – II : water + potassium sulphate + L-asparagine									
0.00	994.0	1008.2	1040.3	0.7190	0.7775	0.8309	1528.6	1584.9	1614.9
0.02	996.2	1020.2	1049.3	0.7242	0.7952	0.8404	1530.3	1587.1	1618.9
0.04	998.6	1023.1	1052.6	0.7294	0.8019	0.8452	1532.5	1589.5	1620.4
0.06	1001.1	1028.6	1058.4	0.7370	0.8102	0.8717	1534.2	1591.4	1621.9
0.08	1003.2	1034.1	1063.2	0.7416	0.8190	0.8808	1535.7	1594.3	1623.2
0.10	1004.6	1038.9	1069.6	0.7594	0.8267	0.8900	1538.2	1596.2	1624.1
System - III : water + potassium sulphate + L-arginine									
0.00	994.0	1008.2	1040.3	0.7190	0.7775	0.8309	1528.6	1584.9	1614.9
0.02	998.4	1025.2	1050.6	0.7337	0.7961	0.8452	1532.2	1588.2	1619.5
0.04	1002.6	1028.6	1056.5	0.7452	0.8023	0.8487	1534.4	1590.5	1621.4
0.06	1009.8	1032.4	1062.3	0.7549	0.8140	0.8795	1536.6	1593.7	1623.3
0.08	1019.4	1036.5	1069.8	0.7695	0.8208	0.8884	1539.2	1595.9	1625.2
0.10	1024.8	1040.9	1074.8	0.7781	0.8303	0.8988	1541.4	1598.5	1627.9

TABLE II

Values of adiabatic compressibility (β), apparent molar compressibility (φ_K), hydration number (n_H) and apparent molar volume (φ_V) of amino acids in aqueous potassium sulphate solutions at 308.15 K

M/ (mol.dm^{-3})	β / (x 10^{-10} m^2 N^{-1})			n_H			$-\varphi_k$/ (x 10^{-7} m^2 N^{-1})			$-\varphi_V$/(x 10^{-3} m^3 mol^{-1})		
	water + potassium sulphate (K$_2$SO$_4$)
	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 + potassium sulphate + L-valine**
0.00	4.3055	3.9487	3.6860	--	--	--	--	--	--	--	--	--
0.02	4.2952	3.9159	3.6582	6.65	21.27	17.68	0.8615	3.2654	2.5593	80.68	404.83	302.89
0.04	4.2738	3.8977	3.6548	10.23	16.54	9.92	1.1390	2.2248	1.3823	80.58	236.18	155.95
0.06	4.2588	3.8752	3.6461	10.04	15.89	8.46	1.1971	1.9757	1.1374	97.15	186.32	122.15
0.08	4.2434	3.8551	3.6297	10.02	15.18	8.95	1.1661	1.8358	1.1865	90.32	164.91	124.48
0.10	4.2260	3.8481	3.6234	10.26	13.05	7.96	1.2065	1.5661	1.0547	95.12	138.60	110.41
**System – II : water + potassium sulphate + L-asparagine**
0.00	4.3055	3.9487	3.6860	--	--	--	--	--	--	--	--	--
0.02	4.2865	3.8914	3.6363	12.26	37.16	31.60	1.4265	5.2147	4.0795	110.94	583.19	412.11
0.04	4.2639	3.8687	3.6182	13.42	25.94	21.55	1.5381	3.4589	2.7845	115.71	360.98	280.67
0.06	4.2438	3.8388	3.5917	13.27	23.75	19.99	1.5401	3.1632	2.6405	118.77	327.71	273.84
0.08	4.2267	3.8045	3.5698	12.71	23.38	18.47	1.4831	3.0705	2.4668	115.18	310.38	258.66
0.10	4.2071	3.7779	3.5445	12.70	22.15	17.99	1.4431	2.9104	2.4532	106.00	292.96	263.18
**System - III : water + potassium sulphate + L-arginine**
0.00	4.3055	3.9487	3.6860	--	--	--	--	--	--	--	--	--
0.02	4.2664	3.8671	3.6291	25.23	52.94	36.19	2.9079	7.4092	4.6698	221.51	822.19	471.04
0.04	4.2364	3.8431	3.6004	22.29	34.24	27.21	2.6588	4.6375	3.5750	215.56	491.62	368.33
0.06	4.1941	3.8136	3.5724	23.96	29.21	24.08	2.9973	3.8245	3.1925	262.18	387.33	331.63
0.08	4.1406	3.7881	3.5390	26.60	26.04	23.37	3.4365	3.3930	3.1441	313.17	338.35	331.18
0.10	4.1071	3.7598	3.5109	25.60	24.50	22.27	3.3181	3.1697	2.9734	302.19	311.60	308.39

TABLE III

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 ($\Delta\varphi_K^0$), transfer volumes ($\Delta\varphi_V^0$) and A and B co-efficients of Jones – Dole equation of each amino acids in aqueous potassium sulphate solution at 308.15 K

Amino acids	K_2SO_4 M/ (mol.dm⁻³)	$-\varphi_K^0$/(×10⁻⁷ m²N⁻¹)	$-\varphi_V^0$/(×10⁻³ m³.mol⁻¹)	S_k/(×10⁻⁷N⁻¹ m⁻¹.mol⁻¹)	S_v/(×10⁻³ m³ L^½.mol⁻³/²)	$\Delta\varphi_K^0$/(×10⁻⁷ m²N⁻¹)	$\Delta\varphi_V^0$/(×10⁻³ m³.mol⁻¹)	A /(dm³/². mol⁻¹/²)	B /(dm³/² mol⁻¹)
L-valine	0.0	0.69	67.31	1.77	90.51	--	--	0.0417	0.5116
	0.5	4.31	569.01	9.00	1446.23	-3.62	-501.7	0.0339	0.2634
	1.0	3.33	404.60	7.85	1018.40	-2.64	-337.29	0.0666	0.0569
L-asparagine	0.0	1.48	117.16	0.03	16.21	--	--	0.0509	0.6489
	0.5	6.45	739.24	12.17	1536.32	-4.97	-622.08	0.8270	0.2747
	1.0	4.95	484.90	8.72	789.70	-3.47	-367.74	0.0646	0.9515
L-arginine	0.0	2.23	124.29	3.50	584.80	--	--	0.0461	0.6822
	0.5	10.03	1139.64	23.38	2823.86	-7.80	-1015.35	0.1436	0.2807
	1.0	5.69	568.54	9.21	870.82	-3.46	-444.25	0.0289	0.9607