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Research Article

PARTIAL MOLAL VOLUME AND PARTIAL MOLAL ISENTROPIC COMPRESSIBILITY OF L-ISOLEUCINE IN AQUEOUS K_2SO_4, KNO_3 AND KCL SOLUTIONS AT THE TEMPERATURES (303.15, 308.15, 313.15, 318.15 AND 323.15 K)

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ABSTRACT

The apparent molal volume (ϕ_v) and apparent molal isentropic compressibility (ϕ_k) of amino acid; L-isoleucine in aqueous solutions of 0.5M K_2SO_4,1M KNO_3 and 1.5M KCl have been calculated at several temperatures by using density and isentropic compressibility values. These parameters values have been used to derive the apparent molal volume at infinite dilution i.e. partial molal volume, (ϕ_v^0) and apparent molal isentropic compressibility at infinite dilution i.e. partial molal compressibility, (ϕ_k^0) for amino acid in the said aqueous solution of electrolytes. The observed ϕ_v^0 values for the studied amino acid in aqueous electrolyte medium have been found to be larger in magnitude than those of corresponding values in water. The negative values with few positive apparent isentropic compressibility values have been found in all the system under investigation. The negative apparent isentropic compressibility values indicate that the water molecules around the amino acid molecules are less compressible than the water molecules in the bulk solvent. This behavior has been discussed in terms of electrostatic forces operative between zwitter-ions and ions of solutions.

Keywords: L-isoleucine; K_2SO_4; KNO_3; KCl; Partial molal volume; Partial molal compressibility

INTRODUCTION

Thermodynamic properties of amino acid-water-electrolyte systems have been extensively studied in order to obtain a better understanding of the behavior of intra-molecular and intermolecular associations, complex formation and related structural changes in liquid systems. Volumetric studies in particular have largely been undertaken in order to probe molecular interactions in solutions [1-3]. There have been many studies on the amino acid–water-salt systems, the volumetric studies have been recognized as a quantity that is sensitive to structural changes occurring in solutions. A number of studies have been made for the determination of partial molal volumes of amino acids in aqueous electrolytes [4-14]. The study of these interactions provide important information about physiological systems and showed that the presence of an electrolyte drastically affects the behavior of amino acids in solutions which can be used for their separation and purification. The study of volumetric properties of solutes have proved to be of great importance in understanding the intermolecular/interionic interactions in solutions and has been found to be sensitive to the degree and nature of solute salvation [15-18]. In continuation of my previous work,[19] I recorded in the present work the measurements of apparent molal volume (ϕ_v) and apparent molal isentropic compressibility (ϕ_k), and their corresponding partial molal volume (ϕ_v^0) and partial molal isentropic compressibility (ϕ_k^0) of ternary systems (L-isoleucine + K_2SO_4 + water, L-isoleucine + KNO_3 + water, L-isoleucine + KCl + water).

MATERIALS AND METHODS

Experimental Details

The amino acid: L-isoleucine used in this work is obtained from SRL (India). The salts: potassium sulphate, potassium nitrate and potassium chloride were purchased from E. Merck (India). All the chemicals were of ≥99% purity. The amino acid was dried at ~110° and kept in vacuum desiccator over P_2O_5 for several hours before use. The salts were recrystallized twice in triply distilled water, dried in a vacuum oven and then kept over P_2O_5 in a vacuum desiccator at room temperature for a minimum of 24 hours before use. All the solutions were made by weight using a balance having a resolution of ±0.1 mg. Stock solutions of 0.5M concentration of K_2SO_4, 1M KNO_3 and 1.5M KCl were prepared in triply distilled water and were used as solvents for the preparation of solutions. The specific conductivity of the water used was less than $18 \times 10^{-6}\ \Omega^{-1}cm^{-1}$. Solutions of amino acid of different molal concentration were prepared in aqueous solutions of 0.5M K_2SO_4, 1M KNO_3 and 1.5M KCl. An ultrasonic interferometer based on variable-path principle was used for the measurement of speed of sound at a frequency of 4 MHz in the temperature range: 303.15 – 323.15 K by a method described elsewhere [20]. An average of 10 readings was taken as a final value of ultrasound velocity. Water from ultra-thermostat (Type U-10) was circulated through the brass jacket surrounding the cell and the quartz crystal. The jacket was well insulated and the temperature of the solution under study was maintained to an accuracy of ±0.01°. The instrument was calibrated with the triply distilled water. The speed of sound values of water at different temperatures were taken from literature for calibration purpose [21]. The densities of solutions were measured by pyknometer using a method described elsewhere [20]. The densities of pure water at various required temperatures were taken from literature for calibration purpose [22]. Thermostated water/paraffin bath was maintained at a desired temperature (±0.01°) for about 30 minutes prior to recording of readings at each temperature of study. Average of close readings was recorded at each temperature.

The accuracies in measurements of the speed of sound and density were ascertained by comparing the measured values of these parameters for water with the corresponding literature values at different temperatures. For instance, the measured values of the speed of sound of water were found to be 1519.9 and 1536.4 ms^{-1}, at 308.15 and 318.15 K, respectively (corresponding literature values[23] are: 1519.808, 1536.409 ms^{-1}); the experimental values of density of water were 0.9942, 0.9903 and 0.9879 $g.cm^{-1}$ at 308.15, 318.15 and 323.15K, respectively (corresponding literature values[22] are: 0.994032, 0.990213, 0.988036 $g.cm^{-1}$). The uncertainties in speed of sound and density measurements were found to be within ±0.2 m/s and ±0.0001 gm/cm^3, respectively.

RESULTS AND DISCUSSION

The data of the measured density (ρ) and speed of sound (u) of ternary systems (amino acid + salt + water): L-isoleucine in the three solvents: 0.5M K_2SO_4, 1M KNO_3 and 1.5M KCl solutions as functions of amino acid concentration and temperature (303.15, 308.15, 313.15, 318.15 and 323.15 K) is given in Table 1 and 2.

Table 1: Density values ($\rho/10^{-3}$ kg. m^{-3}) as functions of concentration and temperature.

(i) L-isoleucine in 0.5 M aqueous K_2SO_4 solution

m/(mol. kg^{-1})	T/K				
	303.15	308.15	313.15	318.15	323.15
0.0000	1.0593	1.0579	1.0561	1.0542	1.0519
0.0284	1.0597	1.0582	1.0564	1.0544	1.0521
0.0474	1.0600	1.0584	1.0565	1.0545	1.0522
0.0665	1.0603	1.0586	1.0567	1.0546	1.0523
0.0857	1.0605	1.0588	1.0569	1.0547	1.0524
0.1049	1.0608	1.0590	1.0570	1.0548	1.0525
0.1243	1.0611	1.0592	1.0571	1.0549	1.0526
0.1437	1.0613	1.0593	1.0573	1.0551	1.0527
0.1633	1.0617	1.0596	1.0575	1.0552	1.0528

(ii) L-isoleucine in 1.0 M aqueous KNO_3 solution

m/(mol. kg^{-1})	T/K				
	303.15	308.15	313.15	318.15	323.15
0.0000	1.0529	1.0511	1.0490	1.0467	1.0442
0.0285	1.0534	1.0515	1.0493	1.0470	1.0445
0.0477	1.0536	1.0517	1.0496	1.0472	1.0447
0.0669	1.0540	1.0520	1.0497	1.0474	1.0448
0.0862	1.0542	1.0523	1.0500	1.0476	1.0450
0.1056	1.0546	1.0524	1.0502	1.0477	1.0451
0.1250	1.0548	1.0527	1.0504	1.0479	1.0454
0.1446	1.0552	1.0529	1.0506	1.0482	1.0456
0.1642	1.0555	1.0532	1.0508	1.0483	1.0457

(iii) L-isoleucine in 1.5 M aqueous KCl solution

m/(mol. kg^{-1})	T/K				
	303.15	308.15	313.15	318.15	323.15
0.0000	1.0614	1.0597	1.0578	1.0556	1.0533
0.0285	1.0618	1.0600	1.0580	1.0558	1.0535
0.0474	1.0622	1.0603	1.0583	1.0560	1.0537
0.0665	1.0624	1.0606	1.0585	1.0562	1.0539
0.0856	1.0628	1.0607	1.0586	1.0563	1.0540
0.1049	1.0631	1.0610	1.0589	1.0565	1.0542
0.1243	1.0633	1.0612	1.0590	1.0567	1.0544
0.1437	1.0637	1.0615	1.0592	1.0569	1.0546
0.1632	1.0639	1.0616	1.0595	1.0571	1.0548

Table 2: Ultrasonic velocity values (u/m. s^{-1}) as functions of concentration and temperature

(i)L-isoleucine in 0.5 M aqueous K_2SO_4 solution

m/(mol. kg^{-1})	T/K				
	303.15	308.15	313.15	318.15	323.15
0.0000	1571.2	1578.8	1587.6	1592.6	1597.6
0.0284	1573.8	1580.4	1589.2	1593.6	1598.2
0.0474	1575.6	1582.0	1590.6	1595.2	1599.4
0.0665	1578.0	1584.8	1592.2	1596.4	1600.4
0.0857	1579.6	1587.7	1594.4	1598.4	1602.8
0.1049	1581.8	1590.7	1593.4	1596.2	1601.2
0.1243	1582.1	1591.7	1595.8	1600.4	1605.0
0.1437	1585.0	1592.0	1598.8 1600.8	1604.4	1608.4
0.1633	1586.0	1593.8		1605.7	1609.4

(ii) L-isoleucine in 1.0 M aqueous KNO_3 solution

m/(mol. kg^{-1})	T/K				
	303.15	308.15	313.15	318.15	323.15
0.0000	1541.8	1550.2	1557.4	1563.6	1568.4
0.0285	1542.6	1551.2	1558.2	1564.4	1568.8
0.0477	1543.8	1552.4	1559.2	1564.8	1569.2
0.0669	1545.2	1553.4	1560.0	1565.6	1569.8
0.0862	1549.3	1556.8	1561.0	1566.0	1571.0
0.1056	1551.6	1558.8	1563.2	1569.7	1573.8
0.1250	1553.2	1559.4	1564.8	1573.2	1576.4
0.1446	1554.0	1560.2	1567.8	1574.4	1578.0
0.1642	1556.4	1563.4	1568.8	1575.8	1579.0

(iii) L-isoleucine in 1.5 M aqueous KCl solution

m/(mol. kg^{-1})	T/K				
	303.15	308.15	313.15	318.15	323.15

0.0000	1584.0	1590.5	1595.8	1601.6	1606.2
0.0284	1585.9	1591.5	1596.0	1602.4	1607.0
0.0474	1586.2	1592.0	1596.4	1603.0	1607.8
0.0665	1587.2	1593.2	1597.4	1604.4	1608.4
0.0856	1588.8	1595.2	1599.4	1605.2	1609.2
0.1049	1592.1	1598.5	1602.6	1606.0	1610.4
0.1243	1593.3	1600.0	1604.6	1608.0	1613.5
0.1437	1596.0	1600.8	1606.8	1612.0	1615.8
0.1632	1598.4	1603.2	1608.2	1614.0	1618.0

Using the data of density (ρ) and speed of sound (u), the apparent molal volume, ϕ_v and apparent molal isentropic compressibility, have been calculated using the following relations:

$\phi_v = (M/\rho) - \{1000\ (\rho - \rho_o)/m\rho\rho_o$ [1]

$\phi_k = [\{1000\ (\kappa_s - \kappa_o)\}/m\rho_o] + \kappa_s\phi_v$ [2]

Where M denotes the molar mass, m is the molality and ρ and ρ_o are the densities of solution and that of the solvent, respectively. The respective values of κ_s and κ_o denote the isentropic compressibilities of solution and solvent that can be related to the density (ρ) of the solution and the speed of sound (u) according to the relation

$K_S = 1/\rho u^2$ [3]

The measured experimental data are correlated using the following linear functions:

$\phi_v = \phi_v^o + S_v m$ [4]

$\phi_k = \phi_k^o + S_k m$ [5]

Where ϕ_v^o and ϕ_k^o are infinite dilution apparent molal volume and apparent molal isentropic compressibility of the said solutions. S_v and S_k are the slopes of the lines obtained from fitting Eqs. (4) and (5) to the experimental data.

The apparent molal volume, ϕ_v and apparent molal isentropic compressibility, ϕ_k of amino acid solution with different electrolyte concentrations that are presented in Table-3 and 4

Table 3: Apparent molal volume ($\phi_v x10^6$, $m^3\ mol^{-1}$) as functions of concentration and temperature

(i) L-isoleucine in 0.5M aqueous K_2SO_4 solution

m/(mol . kg^{-1})	T/K				
	303.15	308.15	313.15	318.15	323.15
0.0284	111.22	114.55	114.69	118.06	118.31
0.0474	110.59	114.52	116.59	118.70	118.95
0.0665	110.32	114.53	116.05	118.97	120.59
0.0857	111.32	114.51	115.74	118.06	119.37
0.1049	110.93	114.54	116.41	119.21	119.46
0.1243	110.73	114.50	116.88	119.28	119.53
0.1437	111.22	115.14	116.58	118.69	119.58
0.1633	110.48	114.50	116.36	118.80	119.61

(ii) L-isoleucine in 1.0 M aqueous KNO_3 solution

m/(mol. kg^{-1})	T/K				
	303.15	308.15	313.15	318.15	323.15
0.0285	108.72	112.07	115.46	115.67	115.94
0.0477	111.26	113.34	113.54	115.69	115.96
0.0669	109.63	112.52	115.46	115.71	117.32
0.0862	110.84	112.06	114.39	115.68	117.02
0.1056	109.88	113.51	114.58	116.56	117.70
0.1250	110.67	113.04	114.72	116.43	116.68
0.1446	109.99	113.33	114.81	115.68	116.58
0.1642	110.03	112.99	114.89	116.25	117.07

(iii) L-isoleucine in 1.5 M aqueous KCl solution

m/(mol. kg^{-1})	T/K				
	303.15	308.15	313.15	318.15	323.15
0.0284	111.02	114.33	117.68	117.91	118.15
0.0474	108.51	112.44	114.52	116.64	116.88
0.0665	110.12	111.63	114.51	116.09	116.33
0.0856	108.93	113.27	115.57	116.85	117.09
0.1049	109.02	112.61	114.53	116.46	116.70
0.1243	109.81	112.87	115.24	116.20	116.43
0.1437	109.14	112.43	115.14	115.99	116.23
0.1632	109.73	113.21	114.51	115.85	116.08

Table 4: Apparent molal isentropic compressibility ($\phi_k x10^{11}$, $bar^{-1}m^3mol^{-1}$) as functions of concentration and temperature

(i) L-isoleucine in 0.5 M aqueous K_2SO_4 solution

m/(mol. kg^{-1})	T/K				
	303.15	308.15	313.15	318.15	323.15

0.0284	-4.41	14.19	14.21	26.04	32.29
0.0474	-5.46	9.10	16.29	17.73	25.26
0.0665	-9.98	-1.21	9.44	16.89	24.62
0.0857	-7.38	-17.89	4.63	11.48	15.43
0.1049	-8.95	-1.07	15.88	27.21	27.20
0.1243	-3.16	-0.24	11.40	14.52	16.00
0.1437	-6.48	-1.54	5.81	5.45	9.09
0.1633	-4.61	0.70	4.32	6.33	13.06

(ii) L-isoleucine in 1.0 M aqueous KNO_3 solution

m/(mol. kg^{-1})	T/K				
	303.15	308.15	313.15	318.15	323.15
0.0285	23.28	22.27	28.10	34.74	34.69
0.0477	18.43	17.85	21.88	29.38	33.36
0.0669	12.73	16.35	22.82	27.09	32.43
0.0862	-4.10	2.02	20.56	28.05	27.76
0.1056	-7.71	0.77	14.34	14.37	18.25
0.1250	-6.28	4.20	12.37	5.19	11.34
0.1446	-3.51	6.54	6.30	5.58	10.09
0.1642	-5.70	0.83	7.53	6.17	11.31

(iii) L-isoleucine in 1.5 M aqueous KCl solution

m/(mol. kg^{-1})	T/K				
	303.15	308.15	313.15	318.15	323.15
0.0284	6.54	21.92	38.23	28.85	28.83
0.0474	14.29	22.71	33.39	27.30	25.44
0.0665	14.71	18.38	28.34	21.36	25.31
0.0856	10.27	13.41	21.17	21.93	25.00
0.1049	0.89	3.41	10.33	21.66	22.51
0.1243	2.41	3.69	8.21	17.25	14.23
0.1437	-1.85	5.50	5.59	7.96	10.44
0.1632	-3.51	3.38	5.36	6.26	7.93

The values of the infinite dilution apparent molal volume and isentropic compressibility of L-isoleucine in K_2SO_4, KNO_3, and KCl solutions are presented in Table 5.

Table 5: The infinite dilution apparent molal volume and isentropic compressibility at different temperatures

(i) L-isoleucine in 0.5 M aqueous K_2SO_4 solution

Temperature (K)	ϕ_v^0 x 10^6 (m^3 mol^{-1})	S_v x 10^6 (m^3 mol^{-2} kg)	σ x 10^6 (m^3 mol^{-1})
303.15	110.90	-0.675	0.4
308.15	114.40	1.891	0.2
313.15	115.29	9.117	0.6
318.15	118.30	4.437	0.5
323.15	118.90	5.383	0.6

(ii) L-isoleucine in 1.0 M aqueous KNO_3 solution

Temperature (K)	ϕ_v^0 x 10^6 (m^3 mol^{-1})	S_v x 10^6 (m^3 mol^{-2} kg)	σ x 10^6 (m^3 mol^{-1})
303.15	109.84	2.990	0.8
308.15	112.30	5.769	0.5
313.15	114.70	0.308	0.7
318.15	115.55	4.264	0.4
323.15	116.19	6.146	0.6

(iii) Isoleucine in 1.5 M aqueous KCl solution

Temperature (K)	ϕ_v^0 x 10^6 (m^3 mol^{-1})	S_v x 10^6 (m^3 mol^{-2} kg)	σ x 10^6 (m^3 mol^{-1})
303.15	109.93	-4.113	0.8
308.15	113.13	-2.947	0.8
313.15	116.26	-11.028	1.0
318.15	117.54	-10.945	0.5
323.15	117.79	-11.007	0.5

(i) L-isoleucine in 0.5 M aqueous K_2SO_4 solution

Temperature (K)	ϕ_k^0 x 10^{11} (bar^{-1} m^3 mol$^{-1)}$	S_k x 10^{11} (bar^{-1} m^3 mol^{-2} kg)	σ x 10^{11} (bar^{-1} m^3 mol^{-1})
303.15	-7.05	0.78	0.3
308.15	7.75	-7.85	0.9
313.15	16.42	-6.46	0.4
318.15	26.98	-11.80	0.6

323.15	33.91	-14.18	0.5

(ii) L-isoleucine in 1.0 M aqueous KNO_3 solution

Temperature (K)	ϕ_k^0 x 10^{11} (bar^{-1} m^3 mol^{-1})	S_k x 10^{11} (bar^{-1} m^3 mol^{-2} kg)	σ x 10^{11} (bar^{-1} m^3 mol^{-1})
303.15	25.37	-22.88	0.7
308.15	23.25	-14.98	0.5
313.15	32.05	-15.94	0.2
318.15	42.33	-24.46	0.4
323.15	43.22	-21.67	0.4

(iii) L-isoleucine in 1.5 M aqueous KCl solution

Temperature (K)	ϕ_k^0 x 10^{11} (bar^{-1} m^3 mol^{-1})	S_k x 10^{11} (bar^{-1} m^3 mol^{-2} kg)	σ x 10^{11} (bar^{-1} m^3 mol^{-1})
303.15	17.14	-12.22	0.5
308.15	27.47	-16.67	0.4
313.15	44.80	-27.20	0.5
318.15	34.87	-16.55	0.3
323.15	35.15	-15.91	0.3

These properties are important because at infinite dilution the interactions between the amino acid molecules are negligible and these properties solely reflect the interactions between the amino acid molecules and the mixed solvent.

Fig. 1-2: Depict the experimental results of the effect of the L-isoleucine concentrations on their apparent molar volumes and isentropic compressibilities in KCl solution

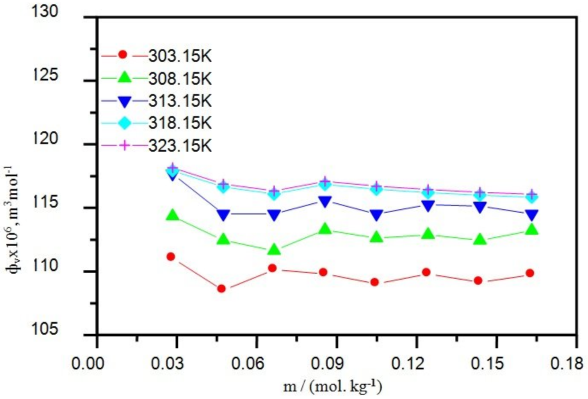

Fig. 1: Plots of apparent molal volume versus concentration of L-isoleucine in 1.5M aqueous KCl solution. At 303.15 K, 308.15 K, 313.15 K, 313.15 K, 318.15 K, 323.15 K

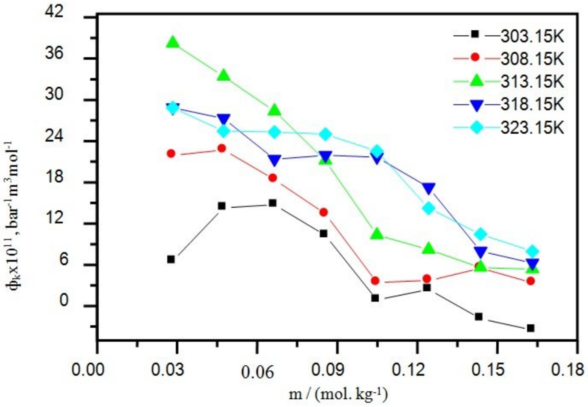

Fig. 2: Plots of apparent molal isentropic compressibility versus concentration of L-isoleucine in 1.5M aqueous KCl solution. At 303.15 K, 308.15 K, 313.15 K, 313.15 K, 318.15 K, 323.15 K

The solid lines are the results of the correlation obtained from Eqs. (4) and (5) with the parameters listed in table 5. The plots of ϕ_v with solute concentration exhibit apparently linear behavior as can be seen from the figures. An examination of the table 5 reveals that the partial molal volume and partial molal isentropic compressibility values of the said amino acid in aqueous electrolyte solutions at each temperature are higher than the corresponding values in aqueous medium. The positive values of ϕ_v^o indicate relatively stronger solute-solvent interactions. The partial molal volume values for the amino acid have been found to increase with increase in temperature from 303.15 to 323.15 K in all three aqueous electrolyte systems. The increase in ϕ_v^o values with temperature may be attributed to volume expansion of hydrated zwitter-ions of amino acid. It is noteworthy that the ϕ^{o}_{v} values in L-isoleucine-aqueous K_2SO_4, KNO_3, and KCl solutions have been found to be 107.54, 106.31, and 107.39, respectively at 298.15 K while in aqueous medium it is found to be 105.80 cm^3/mol [24]. Franks et al.[25] have shown that the partial molal volume of a non-electrolyte is a combination of two factors, viz., the intrinsic volume of solute and the volume changes due to its interactions with solvent. The intrinsic volume has been considered to be made up of two types of contributions [26, 27]

$$V_{intrinsic} = V_{vw} + V_{void} \qquad [6]$$

Where V_{vw} is Van der Waals volume [28, 29], and V_{void} is the volume associated with the voids or empty spaces present therein[30].

Shahidi et al.[27] have modified the above equation in order to evaluate the contribution of a solute molecule towards its partial molal volume as,

$$\phi^{0}_{v} = V_w + V_{void} - n\,\sigma_s \qquad [7]$$

Where σ_s is the shrinkage in volume produced by the interactions of hydrogen bonding groups present in the solute with water molecules and n is the potential number of hydrogen bonding sites in the molecule. For electrolytes and zwitter-ionic solutes, the shrinkage is caused by electrostriction and therefore the ϕ^{0}_{v} of an amino acid can be evaluated by employing the relation,

$$\phi^{0}_{v} = V_{vw} + V_{void} - V_{shrinkage\,.} \qquad [8]$$

It may be assumed that V_{vw} and V_{void} have the same magnitude in water and in mixed solvent for the same class of compounds [26].

The slope, S_v is a measure of solute-solute interactions. The values have been found to be positive for all the systems under investigation. The positive values of S_v indicate the stronger zwitter-ion-zwitter-ion and ion-ion interactions than those of apolar-apolar interactions. It has been observed that the apparent isentropic compressibility values for the said amino acid are negative in all aqueous electrolyte solutions. The values show irregular trend of variations with temperature as well as with concentration. The negative ϕ_k values exhibit strong interactions between the solute and solvent. The ion-zwitter-ion interactions seem stronger than ion-hydrophobic interactions in systems under investigation. The ϕ^{o}_{k} values are positive of smaller magnitude. These values apparently indicate a larger ordering effect of the solute molecules on those of the solvent. The values of S_k are found to be negative, which suggest the presence of essentially weak solute-solute interactions.

CONCLUSION

Finally an examination of the above computed parameters reveals that there is a significant increase in the values of ρ and u with successive increase in the concentration of amino acids and such values gives a better understanding of solute-solute and solute-solvent intermolecular/ interionic interactions. The partial molal volume and partial molal isentropic compressibility values of the said amino acid in aqueous electrolyte solutions at each temperature are higher than the corresponding values in aqueous medium. At infinite dilution the interactions between the amino acid molecules are negligible and these properties solely reflect the interactions between the amino acid molecules and the mixed solvent.

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