



Thermo-acoustical Studies on Interionic Interactions of Some α -amino Acids in Aqueous Sucrose Solution at Varying Mass Percentages

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ABSTRACT

The present study deals with the structure-making and breaking behaviour of some alpha-amino acids in aqueous sucrose (disaccharide) solution at 301.15K. Experimental values of density, viscosity and speed of sound were carried out on the ternary mixtures of water +sucrose + amino acids namely (L-asparagine, L-glutamine L-serine and L-threonine) at 301.15K. The binary solvent mixtures was prepared by taking sucrose at different mass percentages, say at 0%, 5%, 10%, 15%, 20%, and 25% was added with water. The given amino acids under study were added with aqueous solvent under different molarities at normal atmospheric pressure. The related and relevant parameters correlated to our present study such as 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 associated constants (S_K , S_V), partial transfer volume ($\Delta\phi_V^0$) from water to aqueous solution and viscosity B-Coefficient of Jones-Dole equations were meticulously evaluated and eventually this present study predicts dominance of hydrophilic –ionic interactions in the solution. Besides, our investigation also explores about the presence of possible interionic interactions such as solute-solvent, solute-solute, ion-solvent and ion-ion in the solution.

Key words: molar hydration number, solute-solvent, solute-solute, ion-solvent, viscosity B-Coefficient.

INTRODUCTION

It is well known that various substances cause changes in the conformation of proteins when present in aqueous-protein solutions. The complex conformational and conformational factors affecting the structure of proteins in solution make the direct study of protein interactions difficult. Therefore,

one useful approach is to investigate interactions of the model compounds of proteins, i.e., amino acids in aqueous and mixed-aqueous solution¹⁻⁵. The physicochemical properties of amino acids in aqueous solutions provide valuable information on solute-solute and solute-solvent interactions that are important in understanding the stability of proteins, and are implicated in several biochemical

and physiological processes in a living cell⁶. Water is chosen for preparing mixed solvent because its presence gives rise to hydrophobic forces⁷, which are of prime importance in stabilizing the native globular structure of proteins. The interactions of water with the various functional groups of proteins are important factors in determining the conformational stability of proteins²⁻⁴. The stabilization of native conformations of biological macromolecules (proteins) is related to several non-covalent interactions including hydrogen-bonding, electrostatic and hydrophobic interactions⁹.¹⁰. These interactions are affected by the surrounding solute and solvent molecules; for this reason, the physicochemical properties of proteins are strongly affected by the presence of these solutes. Because of direct solute-solvent interactions and/or alteration of the water structure, these solutes can change many properties of globular proteins, such as their hydration, solubility and the activity of enzymes^{1, 4, 5, 11 and 12}. These interactions are important in understanding the stability of proteins, and are implicated in several biochemical and physiological processes in a living cell¹³⁻¹⁶. The protein-carbohydrate interactions are important for immunology, biosynthesis, pharmacology, medicine, and cosmetic industry¹⁷⁻¹⁸. Thus, the properties of amino acids in aqueous-carbohydrate solutions are essential for understanding the chemistry of biological systems¹⁹⁻²².

Direct study of solute/solvent interactions is difficult due to complex conformation of biological macromolecules⁵. Therefore, the convenient approach is to study simpler model compounds i.e. amino acids and peptides, which are the basic components of proteins^{5,6}. When dissolved in water amino acids convert into zwitterionic forms due to the ionization of their carboxyl ("COOH) and amino groups (NH₂). In physiological media, this dipolar character of amino acids has an important bearing on biological functions. Amino acids differ from each other in size, charge, hydrogen-bonding capacity, hydrophobicity and chemical reactivity. Hence, these side chains contribute to the structure and function of proteins, individually and collectively⁷.

The protein-carbohydrate interactions are important for immunology, biosynthesis, pharmacology, medicine and cosmetic industry²³.¹⁷. Thus, the properties of amino acids in

aqueous-carbohydrate solutions are essential for understanding the chemistry of biological systems^{18,19,24} have studied the volumetric, ultrasonic, and viscometric behavior of L-histidine in aqueous glucose solutions²⁵ have studied the density, viscosity, and speed of sound of diglycine in aqueous xylose, L(-)-arabinose, and D(-)-ribose solutions; in aqueous glucose, galatose, and fructose solutions¹⁹ of L-alanine in aqueous fructose solutions² and the density of L-alanine and L-valanine in aqueous sucrose solutions²⁶ have studied the density, viscosity and ultrasonic velocity of L-serine, L-glutamine, and L-asparagine in aqueous glucose solutions.

To the best of our knowledge, no volumetric, ultrasonic, and viscometric studies have been reported on amino acids with polar uncharged side chain in aqueous carbohydrate solutions, except the study by⁴ who investigated volumetric and viscometric properties of arginine in aqueous-carbohydrate solutions. These considerations led us to undertake the study of α -amino acids (with polar uncharged R group) in aqueous-disaccharide solutions. In recent years, a number of workers have determined the various thermodynamic properties of these model compounds in aqueous solutions containing simple electrolytes having hydrophilic nature^{25,3}. But very few studies have been done in aqueous disaccharide solutions^{26,4}.

In the present study, we report the densities, ρ ultrasonic speeds, u and viscosities, η of solutions of α -amino acids in aqueous sucrose solvents taken at regular intervals 0%, 5%, 10%, 15%, 20% and 25% of sucrose, at 301.15 K and at normal atmospheric pressure. An attempt has been carried out in the light following aspects:

Determination of adiabatic compressibility (β), molar hydration number (n_{H}) as a function of molar concentration of α -amino acids in aqueous sucrose solutions at 301.15K.

Determination of apparent molar compressibility (ϕ_k), apparent molar volume (ϕ_v), Limiting apparent molar compressibility (ϕ_k^0) and its related constants (S_k) limiting apparent molar volume and its related constants (S_v), partial transfer volume ($\Delta\phi_v^0$) from water to aqueous disaccharide solution.

To shed more details on the viscometric study, viscosity B-coefficient of Jones–Dole equation has also been evaluated.

Experimental methods

The α -Amino acids (SRL India, mass fraction purity > 0.99) was used after re-crystallization from ethanol–water mixture and dried in vacuum over P_2O_5 at room temperature for 72 h. Sucrose (E. Merck, Germany, mass fraction purity > 0.998) was used as such without further purification, except drying in oven for 24 h. The aqueous sucrose solutions 0%, 5%, 10%, 15%, 20% and 25% mass percentage of sucrose, were prepared using triple distilled water (conductivity less than $1 \times 10^{-6} \text{ S.cm}^{-1}$) and these were used as solvents and amino acids was added of six different molar concentrations (ranging from 0 to 0.1 in step of 0.02). The chemicals were weighed in an electronic digital balance (SHIMADZU AX-200, Japan Make) with a least count of 0.0001g. The density was determined using a 5ml specific gravity bottle by relative measurement method with an accuracy of $\pm 0.01 \text{ kgm}^{-3}$. An Ostwald's viscometer of 10ml capacity was used for the viscosity measurement. Efflux time was determined using a digital chronometer within $\pm 0.01 \text{ s}$. An Ultrasonic interferometer having the fixed frequency of 2MHz (Mittal Enterprises, New Delhi-Model: F-81) with an overall accuracy of $\pm 0.1 \text{ ms}^{-1}$ has been used for velocity measurement. An electronically digital operated constant temperature bath (RAAGA industries, Chennai) has been used to circulate water through the double walled measuring cell made up of steel containing the experimental solution at desired temperature, whose accuracy is maintained at 0.1K.

RESULTS AND DISCUSSION

The values of density (ρ), viscosity (η) and speed of sound (U) of α -amino acids in aqueous sucrose solution at 301.15K are shown in Table 1. The related parameters such as adiabatic compressibility (β), molar hydration number (η_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 associated constants (S_k, S_v), partial molar transfer volume ($\Delta\varphi_v^0$) and viscosity A and B-coefficient of Jones-Dole equation were meticulously evaluated and are reported in Table (1-6).

In the all four amino acid systems studied, (from Table-1) our present values of density increase with increase in molar concentration of amino acids as well as mass percentage of sucrose content. The other measured parameter ultrasonic velocity (U) which is also found (from Table-2) to be increased with increase in same concentration of amino acids as well as aqueous sucrose content. Such an observed increase in the ultrasonic velocity in these solutions may be attributed to the cohesion brought about by the ionic hydration; which may also be due to the overall increase of cohesion brought about by solute-solute and solute-solvent interaction in solution.

Incidentally, the density (ρ) which is a measure of solute-solvent interactions, which can be attributed as increase of density with concentration indicates the increase in solute-solvent interactions, whereas the decrease in density indicates the lesser magnitude of solute-solvent interactions. Increase in density with concentration is due to the shrinkage in the volume which in turn is due to the presence of solute molecules. As observed in Table-1, an increasing trend of density values may be interpreted to the structure-making behavior of the solvent due to the added solute²⁹.

In the present study, the measured pH values of aqueous solution were found to be less than 8.4 (Table-2) however, these pH values decreases, while increasing molarities of amino acids as well as mass percentage of the sucrose. This can be inferred that at the pH of the solution, the side-chains of the acidic amino acids studies are considered to exist mainly as Zwitterions in aqueous sucrose solution, having mainly the –OH groups. Sugar molecules such as glucose or sucrose is a polyhydroxy compound and its addition to amino acid (with short alkyl chains) aqueous solutions were found to enhance the zwitterions ($-\text{CHCOO}^-$ and NH_3^+) hydrophilic – OH group interactions and almost eliminate the of hydrophilic – hydrophobic interactions between the –OH group of sucrose or water and non-polar alkyl chain of amino acids, at higher concentrations. Sucrose molecules have a ring structure and have several –OH groups. It may be inferred that when amino acids are added with aqueous sucrose solution, the terminal groups of zwitterions of amino acids, NH_3^+ and COO^- are hydrated in an

Table 1: Values of Density (ρ) and Viscosity (η) of amino acids in aqueous Sucrose solution at 301.15K

Molarity M/(mol.dm ⁻³)	Density(ρ)(kg/m ³)						Viscosity (η)($\times 10^{-3}$ Nsm ⁻²)					
	0%	5%	10%	15%	20%	25%	0%	5%	10%	15%	20%	25%
System-I (Water + Sucrose + L-glutamine)												
0.00	996.59	1007.48	1026.13	1042.46	1061.30	1080.41	0.8836	1.3967	1.5598	1.9232	2.3428	2.7624
0.02	999.94	1009.37	1028.91	1045.29	1065.33	1086.18	0.9099	1.3156	1.5737	1.9366	2.3609	2.7786
0.04	1001.03	1010.98	1029.81	1046.40	1066.54	1087.29	0.9162	1.3326	1.5862	1.9497	2.3734	2.7834
0.06	1002.19	1012.03	1030.89	1047.51	1067.76	1088.47	0.9223	1.3442	1.5992	1.9623	2.3853	2.7997
0.08	1003.43	1013.17	1031.96	1048.76	1068.96	1089.54	0.9291	1.3561	1.6106	1.9750	2.3978	2.8056
0.10	1004.52	1014.26	1032.99	1049.87	1069.99	1090.62	0.9389	1.3681	1.6227	1.9882	2.4108	2.8175
System-II (Water + Sucrose + L-asparagine)												
0.00	996.59	1007.48	1026.13	1042.46	1061.30	1080.41	0.8836	1.2967	1.5598	1.9232	2.3428	2.7624
0.02	999.21	1008.96	1028.04	1044.48	1064.93	1085.38	0.9149	1.3291	1.5832	1.9465	2.3668	2.7893
0.04	1000.56	1010.01	1029.09	1045.62	1065.99	1086.59	0.9255	1.3411	1.5955	1.9590	2.3792	2.7984
0.06	1001.79	1011.11	1029.99	1046.70	1067.08	1087.66	0.9360	1.3531	1.6070	1.9706	2.3911	2.8023
0.08	1002.92	1012.26	1030.93	1047.98	1068.02	1088.74	0.9469	1.3652	1.6199	1.9824	2.4043	2.8167
0.10	1003.89	1013.98	1031.96	1048.95	1068.98	1089.82	0.9581	1.3773	1.6313	1.9948	2.4170	2.8245
System-III (Water + Sucrose + L-serine)												
0.00	996.59	1007.48	1026.13	1042.46	1061.30	1080.41	0.8836	1.2967	1.5598	1.9232	2.3428	2.7624
0.02	998.69	1007.01	1027.64	1043.04	1062.18	1081.27	0.9210	1.3484	1.6018	1.9660	2.3868	2.8012
0.04	1000.12	1008.69	1028.79	1044.71	1063.74	1083.21	0.9317	1.3595	1.6135	1.9779	2.3989	2.8124
0.06	1000.83	1010.96	1029.67	1046.13	1065.89	1085.29	0.9424	1.3714	1.6234	1.9901	2.4310	2.8218
0.08	1001.99	1011.94	1030.24	1047.37	1066.99	1087.72	0.9532	1.3829	1.6379	2.0018	2.4228	2.8329
0.10	1002.96	1012.86	1031.01	1048.39	1068.19	1088.78	0.9640	1.3944	1.6496	2.0140	2.4355	2.8423
System-IV (Water + Sucrose + L-threonine)												
0.00	996.59	1006.48	1026.13	1042.46	1061.30	1080.41	0.8836	1.2967	1.5598	1.9232	2.3428	2.7624
0.02	998.99	1008.64	1027.99	1043.96	1064.23	1082.21	0.9121	1.3387	1.5925	1.9560	2.3771	2.7984
0.04	1000.87	1010.39	1028.91	1044.89	1065.35	1084.27	0.9222	1.3417	1.6034	1.9679	2.3883	2.8075
0.06	1001.24	1011.37	1029.89	1046.33	1066.14	1086.39	0.9379	1.3613	1.6155	1.9801	2.4015	2.8154
0.08	1002.36	1012.15	1030.78	1047.94	1067.41	1088.70	0.9435	1.3730	1.6278	1.9923	2.4139	2.8247
0.10	1003.19	1013.06	1031.14	1048.67	1068.49	1089.68	0.9591	1.3846	1.6401	2.0045	2.4258	2.8313

Table 2: Values of Ultrasonic velocity (U) and pH of amino acids in aqueous sucrose solution at 301.15K

Molarity	U/(m/s)										pH				
	0%	5%	10%	15%	20%	25%	0%	5%	10%	15%	20%	25%			
System-I (Water + Sucrose + L-Glutamine)															
0.00	1502.7	1514.8	1529.4	1546.6	1561.6	1578.3	7.0	7.5	7.7	7.9	8.1	8.4			
0.02	1504.9	1525.2	1536.1	1554.4	1569.4	1586.4	5.3	6.2	6.3	6.4	6.5	6.7			
0.04	1506.3	1527.6	1538.9	1556.9	1571.7	1588.4	5.1	6.1	6.2	6.3	6.4	6.6			
0.06	1508.9	1529.3	1540.3	1558.4	1573.9	1590.2	5.0	6.0	6.1	6.2	6.3	6.5			
0.08	1510.2	1531.1	1542.1	1560.1	1575.8	1592.4	4.9	5.9	6.0	6.1	6.2	6.3			
0.10	1512.6	1533.9	1544.9	1562.7	1577.9	1594.7	4.8	5.8	5.9	6.0	6.1	6.2			
System-II (Water + Sucrose + L-asparagine)															
0.00	1502.7	1514.8	1529.4	1546.6	1561.6	1578.3	7.0	7.5	7.7	7.9	8.1	8.4			
0.02	1504.1	1522.1	1534.8	1551.3	1566.8	1584.0	5.9	6.0	6.2	6.3	6.4	6.5			
0.04	1506.9	1524.7	1536.1	1553.6	1568.9	1586.1	5.8	5.9	6.1	6.2	6.3	6.4			
0.06	1508.8	1526.3	1538.5	1555.1	1570.6	1588.0	5.7	5.8	6.0	6.1	6.2	6.3			
0.08	1510.3	1528.4	1540.7	1557.8	1572.8	1590.4	5.6	5.7	5.9	6.0	6.1	6.2			
0.10	1512.2	1530.9	1542.3	1559.2	1574.5	1592.3	5.5	5.6	5.8	5.9	6.0	6.1			
System-III (Water + Sucrose + L-serine)															
0.00	1502.7	1514.8	1529.4	1546.6	1561.6	1578.3	7.0	7.5	7.7	7.9	8.1	8.4			
0.02	1503.4	1519.2	1531.3	1548.1	1564.7	1580.1	6.1	5.9	6.1	6.2	6.3	6.4			
0.04	1505.1	1521.3	1533.8	1550.4	1566.9	1582.9	5.9	5.8	6.0	6.1	6.2	6.3			
0.06	1507.5	1523.1	1536.3	1552.8	1568.9	1585.9	5.8	5.7	5.9	6.0	6.1	6.2			
0.08	1509.3	1525.8	1538.2	1554.7	1570.7	1588.9	5.7	5.6	5.8	5.9	6.0	6.1			
0.10	1511.0	1527.9	1540.7	1557.9	1572.8	1590.6	5.6	5.5	5.7	5.8	5.9	6.0			
System-IV (Water + Sucrose + L-threonine)															
0.00	1502.7	1514.8	1529.4	1546.6	1561.6	1578.3	7.0	7.5	7.7	7.9	8.1	8.4			
0.02	1503.7	1520.1	1533.6	1549.7	1565.1	1583.4	5.8	6.5	6.6	6.7	6.8	6.9			
0.04	1506.6	1522.3	1535.3	1551.3	1567.6	1585.9	5.6	6.4	6.5	6.6	6.7	6.8			
0.06	1508.4	1524.7	1537.9	1553.4	1569.2	1587.3	5.5	6.3	6.4	6.5	6.6	6.7			
0.08	1510.1	1526.9	1539.4	1555.1	1571.7	1589.1	5.4	6.2	6.3	6.4	6.5	6.6			
0.10	1511.7	1528.3	1541.1	1557.7	1573.2	1591.8	5.3	6.1	6.2	6.3	6.4	6.5			

Table 3: Values of Adiabatic compressibility (β) and Molar hydration number (n_H) of amino acids in aqueous sucrose solution at 301.15K

Molarity M/(mol.dm ⁻³)	Adiabatic compressibility β ($\times 10^{-10}$ m ² N ⁻¹)					Molar hydration number (n_H)						
	0%	5%	10%	15%	20%	25%	0%	5%	10%	15%	20%	25%
System-I (Water + Sucrose + L-Glutamine)												
0.00	4.4436	4.3256	4.1663	4.0103	3.8638	3.7156	-	-	-	-	-	-
0.02	4.4158	4.2588	4.1182	3.9594	3.8110	3.6582	17.36	43.75	31.53	35.12	37.72	41.15
0.04	4.4028	4.2387	4.1003	3.9425	3.7956	3.6453	13.28	28.30	21.79	23.24	24.24	25.94
0.06	4.3825	4.2249	4.0886	3.9308	3.7807	3.6331	12.77	21.90	17.15	18.23	19.73	20.34
0.08	4.3696	4.2102	4.0748	3.9175	3.7673	3.6195	12.64	20.78	16.71	17.57	18.97	19.62
0.10	4.3510	4.1904	4.0560	3.9004	3.7537	3.6055	11.81	17.11	14.63	15.21	15.78	16.39
System-II (Water + Sucrose + L-Asparagine)												
0.00	4.4436	4.3257	4.1663	4.0103	3.8638	3.7156	-	-	-	-	-	-
0.02	4.4237	4.2779	4.1293	3.9784	3.8258	3.6720	14.72	31.24	24.62	22.06	27.70	32.38
0.04	4.4013	4.2589	4.1182	3.9623	3.8111	3.6582	13.19	21.84	15.99	16.58	18.83	21.31
0.06	4.3849	4.2454	4.1017	3.9505	3.7990	3.6459	12.22	17.53	14.30	13.77	15.45	17.25
0.08	4.3712	4.2289	4.0863	3.9320	3.7850	3.6313	11.29	15.84	13.29	13.45	14.09	15.65
0.10	4.3560	4.2080	4.0738	3.9214	3.7735	3.6190	10.93	15.09	12.30	12.28	12.91	14.35
System-III (Water + Sucrose + L-Serine)												
0.00	4.4436	4.3256	4.1663	4.0103	3.8638	3.7156	-	-	-	-	-	-
0.02	4.4301	4.3026	4.1499	4.0003	3.8453	3.7042	8.40	13.98	10.93	9.28	13.23	8.46
0.04	4.4138	4.2836	4.1317	3.9821	3.8289	3.6845	10.57	14.04	11.49	9.99	12.45	11.52
0.06	4.3966	4.2639	4.1148	3.9644	3.8115	3.6635	10.28	14.36	11.42	10.56	12.47	12.89
0.08	4.3811	4.2447	4.1023	3.9500	3.7988	3.6415	9.93	13.35	10.62	10.27	12.86	13.74
0.10	4.3670	4.2292	4.0860	3.9300	3.7844	3.6302	9.61	12.70	10.67	11.08	11.35	12.67
System-IV (Water + Sucrose + L-Theronine)												
0.00	4.4436	4.3257	4.1665	4.0107	3.8637	3.7153	-	-	-	-	-	-
0.02	4.4270	4.2906	4.1360	3.9886	3.8360	3.6855	26.46	17.03	20.38	15.21	20.15	22.72
0.04	4.4017	4.2708	4.1232	3.9768	3.8197	3.6670	15.35	13.91	14.36	11.60	15.79	18.02
0.06	4.3896	4.2532	4.1053	3.9606	3.8091	3.6533	13.91	12.93	13.50	25.25	13.03	15.32
0.08	4.3748	4.2377	4.0938	3.9459	3.7925	3.6373	12.29	12.79	12.04	11.12	12.60	14.53
0.10	4.3620	4.2261	4.0833	3.9300	3.7814	3.6218	11.23	12.00	11.02	11.09	11.77	13.98

Table 4: Values of Apparent molar compressibility (ϕ_k) and apparent molar volume (ϕ_v) of amino acids in aqueous sucrose solution at 301.15K

Molarity M/(mol.dm ⁻³)	Adiabatic compressibility ϕ_k ($\times 10^{-10}$ m ² N ⁻¹)					Apparent molar volume ϕ_v (\times m ³ mol ⁻¹)						
	0%	5%	10%	15%	20%	25%	0%	5%	10%	15%	20%	25%
System-I (Water + Sucrose + L-glutamine)												
0.00	-	-	-	-	-	-	-	-	-	-	-	-
0.02	-2.7011	-3.7441	-2.9362	-2.7064	-2.6500	-2.4953	-167.926	-143.424	-135.31	-87.632	-81.366	-81.778
0.04	-1.9382	-2.5480	-2.0227	-1.8826	-1.8221	-1.6678	-111.23	-111.630	-89.514	-70.366	-76.183	-66.506
0.06	-1.5970	-2.0036	-1.6173	-1.5222	-1.5198	-1.3835	-93.506	-91.759	-77.170	-64.610	-65.190	-62.495
0.08	-1.4640	-1.7471	-1.4390	-1.3673	-1.3482	-1.2541	-85.645	-82.941	-70.876	-63.411	-62.992	-59.217
0.10	-1.4302	-1.6431	-1.3764	-1.3081	-1.2599	-1.3186	-79.424	-77.153	-66.710	-61.348	-61.013	-57.342
System-II (Water + Sucrose + L-asparagine)												
0.00	-	-	-	-	-	-	-	-	-	-	-	-
0.02	-1.5320	-2.7011	-2.2397	-1.6036	-1.5811	-1.8779	-131.315	-123.070	-92.939	-68.796	-38.978	-73.460
0.04	-1.4977	-1.9382	-1.5036	-1.3139	-1.2566	-1.3774	-99.457	-87.550	-71.986	-58.673	-46.751	-61.891
0.06	-1.3641	-1.5970	-1.3368	-1.1407	-1.0705	-1.1682	-86.830	-76.538	-62.566	-53.673	-43.532	-56.183
0.08	-1.2563	-1.4648	-1.2432	-1.1435	-1.0160	-1.0939	-79.263	-71.653	-58.343	-51.872	-42.511	-52.982
0.10	-1.2001	-1.4044	-1.1616	-1.0628	-0.9527	-0.9016	-73.117	-74.385	-56.686	-44.537	-40.203	-51.709
System-III (Water + Sucrose + L-serine)												
0.00	-	-	-	-	-	-	-	-	-	-	-	-
0.02	-1.1391	-1.2643	-1.1280	-0.6416	-0.8660	-0.6480	-105.239	-56.017	-43.461	-20.258	-13.079	-30.433
0.04	-1.1137	-1.2876	-1.1355	-0.7300	-0.8791	-0.8112	-88.432	-59.992	-47.441	-29.862	-29.097	-36.681
0.06	-1.0965	-1.3495	-1.0980	-0.8688	-0.9222	-0.9000	-70.788	-61.316	-49.950	-41.613	-42.131	-41.694
0.08	-1.0954	-1.3589	-1.1077	-0.8939	-0.8774	-0.9425	-67.611	-67.692	-57.381	-46.770	-42.170	-44.085
0.10	-1.0490	-1.2798	-1.1096	-0.9549	-0.8222	-0.9047	-63.798	-68.437	-64.690	-47.177	-35.598	-45.150
System-IV (Water + Sucrose + L-threonine)												
0.00	-	-	-	-	-	-	-	-	-	-	-	-
0.02	-1.3610	-2.2167	-1.8916	-0.9927	-1.1287	-1.4672	-120.305	-107.201	-90.529	-23.880	-34.292	-36.925
0.04	-1.5226	-1.7911	-1.3600	-0.8801	-1.0730	-1.2050	-107.260	-97.016	-67.627	-34.193	-43.479	-42.941
0.06	-1.1627	-1.5568	-1.2701	-0.9494	-0.9476	-1.0271	-77.659	-89.150	-60.968	-45.784	-42.301	-42.787
0.08	-1.1567	-1.4032	-1.1419	-0.9451	-0.9434	-0.9940	-72.266	-70.314	-56.542	-48.821	-40.221	-40.536
0.10	-1.0811	-1.2772	-1.0325	-0.9061	-0.8624	-0.9854	-66.120	-65.271	-48.721	-49.877	-38.438	-37.680

electrostatic manner whereas, hydration of R group depends on its nature, which may be hydrophilic, hydrophobic or amphiphilic; and the overlap of hydration co-spheres of terminal NH_3^+ and COO^- groups and of adjacent groups results in volume change. This may increase due to reduction in the electrostriction at terminals, whereas it decreases due to disruption of side group hydration by that of the charged end³⁰.

The adiabatic compressibility (β) of the solute can be expressed as the extent to which hydration around the solute molecule can be expressed. The perusal of Table-3 exhibits the values of adiabatic compressibility (β), which are found to be decreased with increase in molar concentration of solute (aminoacids) as well as mass percentage of sucrose content. Such a decrease in adiabatic compressibility observed in solvent (aqueous

sucrose solution) may be attributed to weakening of hydrogen bond in the solution. It is well known 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 interaction. Since, the solvent molecules are oriented in the ionic field; these molecules are more compactly packed in the primary solvation shells as compared to the packing in the absence of the ions. This is the reason, why the solvent is compressed by the introduction of the ions. Thus, the electrostatic field of the ions causes the compression of the medium giving rise to a phenomenon called '*Electrostriction*'. Since the water molecules are compressed, they do not respond to a further application of pressure. So the solutions become harder to compress. Consequently, this will lead to decrease in compressibility values. It may also be inferred that weakening of hydrogen bond strength formed by the solute and solvent

Table 5: Values of Limiting apparent molar compressibility (φ^0_{κ}) and limiting apparent molar volume φ^0_v and their constant S_{κ} and S_v of amino acids in aqueous sucrose at 301.15 K

Amino acids	Water + sucrose	$\varphi^0_{\kappa} (\times 10^{-8} \text{ m}^2 \text{ N}^{-1})$	$S_{\kappa} (\times 10^{-8} \text{ N}^{-1} \text{ m}^1 \text{ mol}^{-1})$	$\varphi^0_v (\times \text{m}^3 \text{ mol}^{-1})$	$S_v (\times 10^{-8} \text{ N}^{-1} \text{ m}^1 \text{ mol}^{-1})$
L-glutamine	0%	-17.34	87.58	-246.74	584.84
	5%	-57.21	142.17	-232.37	462.49
	10%	-44.07	106.26	-195.63	452.57
	15%	-42.52	97.54	-113.64	185.45
	20%	-39.44	93.45	-108.01	161.29
	25%	-35.58	81.29	-106.25	171.39
L-asparagine	0%	-18.84	21.62	-188.12	395.42
	5%	-39.32	88.71	-167.40	339.35
	10%	-31.77	70.58	-128.93	253.90
	15%	-21.47	37.55	-88.04	138.07
	20%	-22.18	43.83	-45.15	11.59
	25%	-27.99	63.65	-781.20	245.17
L-serine	0%	-12.87	7.74	-150.80	300.96
	5%	-13.75	0.47	46.76	-66.94
	10%	-11.90	10.27	61.58	-12.57
	15%	18.60	-5.31	72.86	-178.61
	20%	9.75	4.26	4.61	-155.56
	25%	-15.41	-4.75	-87.48	-18.78
L-threonine	0%	-18.98	26.94	-185.08	404.80
	5%	-29.52	65.17	-159.66	310.41
	10%	-27.05	57.35	-129.81	272.83
	15%	-5.76	12.48	-86.25	192.17
	20%	-14.70	20.13	-35.30	19.76
	25%	-35.30	19.76	-8.49	-18.42

molecules may also be the reason for decrease in compressibility³¹. The present investigation finds that β values are larger in L-serine system comparing other amino acid systems, advocating the strong molecular association in this system.

The Molar hydration number (n_H) has been evaluated from

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

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

The hydration number (n_H) reflecting the electrostriction effect of the charged centre of the

amino acid and in the vicinity of the water molecules. From Table-3, one can observe that the values of n_H are positive in all the four liquid systems and such positive values of n_H indicate an appreciable solvation of solutes³⁰. It can be taken as an added support for the structure making nature of solutes as well as the presence of dipolar interactions between the solutes and water molecules. This sensitive parameter also suggests that the compressibility of the solution will be less than that of the solvent, resulting in solutes will gain more mobility and have a more probability of contacting solvent molecules, which in turn may enhance the interaction between the solute solvent molecules. Our present study finds that the values of n_H which are decreasing with increasing molar concentration of solute (amino acids) and however, found to be increased with elevation of mass percentage of sucrose. This may suggest that an enhancement of solute-co solute

Table 6 Values of Partial transfer volume ($\Delta\phi_v^0$), A and B co-efficient of Jones - Dole equation of amino acids in aqueous sucrose solution at 301.15 K

Amino acids	Water + Sucrose	$\Delta\phi_v^0$ ($\times 10^{-3} \text{ m}^3 \text{ mol}^{-1}$)	A ($\text{dm}^{3/2} \text{ mol}^{-1/2}$)	B ($\text{dm}^3 \text{ mol}^{-1}$)
L-glutamine	0%	-	0.2277	-0.1552
	5%	14.37	-0.7251	2.2387
	10%	51.11	0.0058	0.3901
	15%	133.10	-0.0018	0.3465
	20%	138.73	0.0219	0.2175
	25%	140.49	0.0182	0.1362
L-asparagine	0%	-	0.2444	0.0164
	5%	20.72	0.1622	0.0823
	10%	59.19	0.0729	0.2195
	15%	100.08	0.0597	0.1739
	20%	142.97	0.0485	0.1551
	25%	146.29	0.0375	0.1213
L-serine	0%	-	0.3154	-0.1407
	5%	197.56	0.3271	-0.3386
	10%	212.38	0.2002	-0.0949
	15%	226.66	0.1615	-0.0625
	20%	155.41	0.1416	-0.0910
	25%	63.32	0.1087	-0.0708
L-threonine	0%	-	0.1952	0.1944
	5%	25.42	0.2197	0.0554
	10%	55.27	0.1362	0.0587
	15%	98.83	0.1102	0.0563
	20%	149.78	0.0956	0.0367
	25%	176.59	0.1065	0.1026

interactions in the mixture. The decreasing values of n_H may be presumed as that the strength of the interactions may be weakened among the solute-co solute molecules.

Molar compressibility studies

The apparent molar compressibility (ϕ_K) of the solute is given by the relation

$$\kappa = \frac{1000}{\rho_0} (\beta_0 - \beta) + \left(\frac{M}{\rho} \right)$$

in which, m is the molarity of the solution ($\text{mol} \cdot \text{dm}^{-3}$), ρ_0 is the density of the solvent ($\text{kg} \cdot \text{m}^{-3}$) and ρ is the density of the solute, β_0 and β are the adiabatic compressibility of the solvent and solution, M is the molar mass of the solute ($\text{kg} \cdot \text{mol}^{-1}$) respectively.

The apparent molar volume (ϕ_V) of amino acids has been calculated from the density values of solvent and solution using the relation.

$$v = \frac{1000}{\rho_0} (\rho - \rho_0) + \left(\frac{M}{\rho} \right)$$

Where m is the molarity of the solution ($\text{mol} \cdot \text{dm}^{-3}$), M is the molar mass of the solute ($\text{kg} \cdot \text{mol}^{-1}$) and, ρ_0 and ρ are the density values of the solvent and solution respectively.

The following observations are noticed from Table- 4 on apparent molar compressibility (ϕ_K) and apparent molar volume (ϕ_V) of α -amino acids in aqueous sucrose at 301.15K.

- (i) The values of the apparent molar compressibility (ϕ_K) and apparent molar volume (ϕ_V) are all negative over the entire molarity range of amino acids (solute).
- (ii) The values of apparent molar compressibility (ϕ_K) are found to be increased with increasing molarity (m) of solute (amino acids).
- (iii) It is also interesting to note that apparent

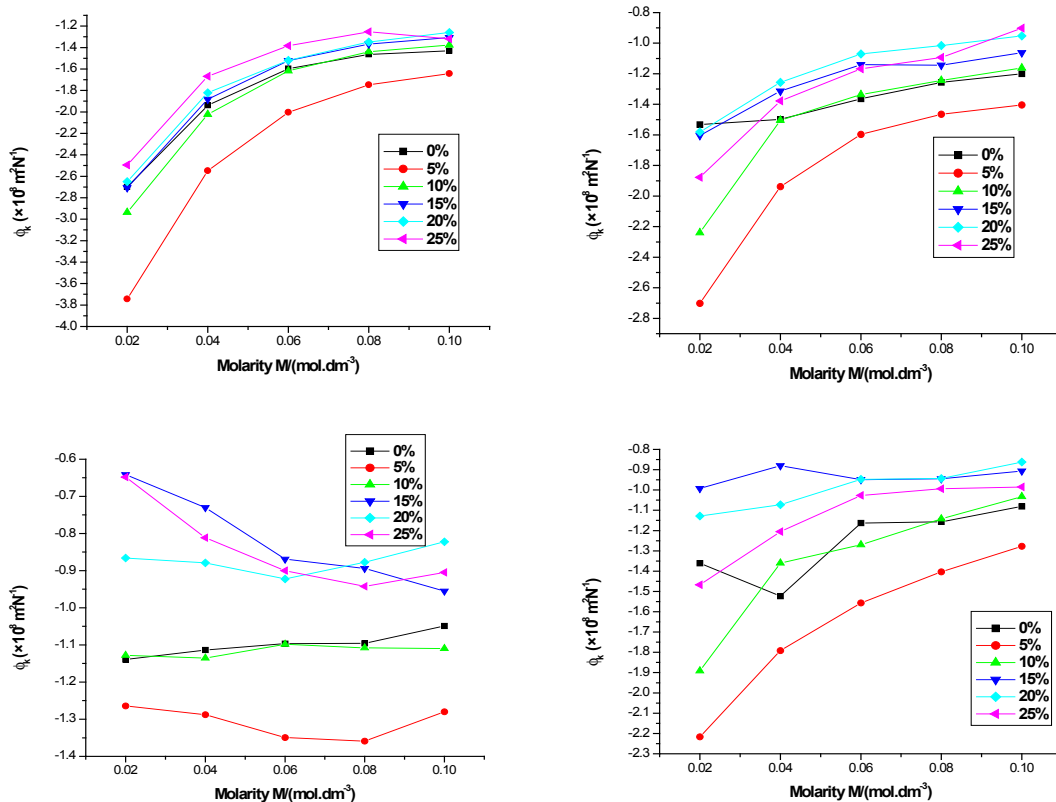


Fig. 1: Apparent molar compressibility (ϕ_K) Vs Molarity (M)

- molar volume (ϕ_v) too exhibits the same trend as that of the apparent molar compressibility (ϕ_k).
- (iv) However, one can notice from the Table-4, both the values of ϕ_k as well as ϕ_v in L-serine system decreases with molar concentration of the amino acids, and the same increase with increase of mass percentage of the sucrose content.
 - (v) Our present investigation observes the values of apparent molar compressibility (ϕ_k) and apparent molar volume (ϕ_v) are higher in L-serine system over the other amino acids, indicating the molecular association is more pronounced in this system.

occurring in these systems. Since, more number of water molecules are available at lower concentration of aqueous sucrose, the chances for the penetration of solute molecules in the solvent molecules are highly³¹ favored. The decreasing values of apparent molar compressibility and molar volume in L-serine system with increasing molar concentration of solute and solvent content in aqueous medium reveal the strengthening of the solute-solvent interaction.

The values of Limiting apparent molar compressibility ϕ_k^0 in the present study, have been fitted by Least-squares method with the equation

$$\phi_k = \phi_k^0 + S_k m^{3/2}$$

Where ϕ_k^0 is the limiting apparent molar compressibility at infinite dilution and S_k is its associated constant.

The above observations clearly suggesting the existence of solute-solvent interaction in the solution. The negative values of apparent molar compressibility indicate the hydrophilic interactions

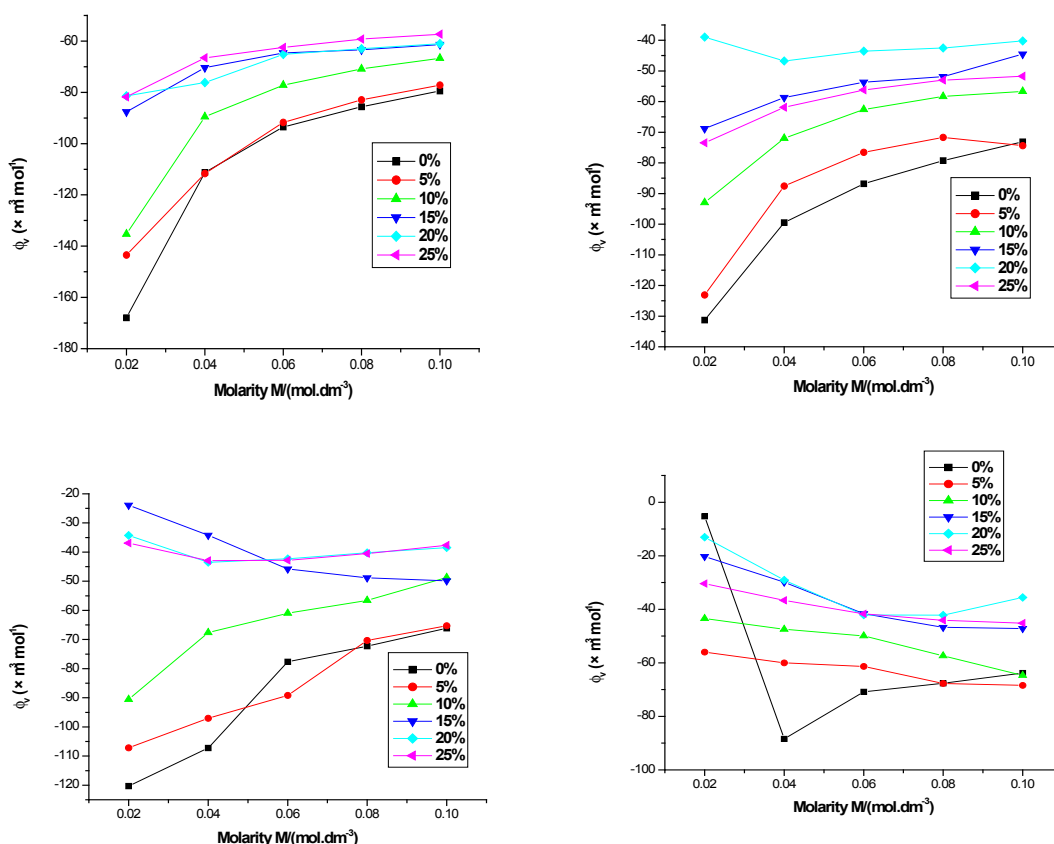


Fig. 2: Apparent molar volume (ϕ_v) Vs Molarity (M)

The evaluated former parameter Limiting apparent molar compressibility (ϕ_k^0) which is concerned with the ion-solvent interactions and its later related constant (S_k) of the ion-ion interactions in the solution, which are systematically tabulated in Table-5. It is noticed that the ϕ_k^0 values are negative in all the four liquid systems and increase with increase of mass percentage of sucrose content. From fig-III, the negative values of ϕ_k^0 for all the systems, and their increasing trend in the present study suggesting the existence of strong ion-solvent interactions. The related constant (S_k) whose values are positive in all the systems and however decrease with elevation of sucrose content. Such a decreasing trend of positive values of S_k indicates the weakening of ion-ion interactions in the solution. The apparent molar compressibility (ϕ_k^0) values are found to be higher in L-serine system indicating that molecular association is more pronounced in this system.

The limiting apparent molar volume (ϕ_v^0) in the present study have been fitted by method of Least-squares with the equation

$$\phi_v = \phi_v^0 + S_v m^{3/2}$$

Where ϕ_v^0 is the limiting apparent molar volume at infinite dilution and S_v is its associated constant.

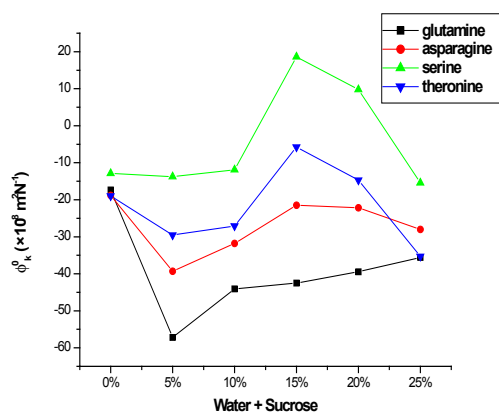


Fig. 3: Limiting apparent molar compressibility (ϕ_k^0) Vs mass percentage of sucrose at 301.15K

The perusal of Table-5 depicts the values of limiting apparent molar volume (ϕ_v^0) furnishing negative values in all the systems except in system-III, where as reverse trend is observed. And however all the systems increase with increase of mass percentage of sucrose content. Our present study notices that the large values of ϕ_v^0 observed in L-serine system comparing the other amino acid systems. The electrostriction at Zwitterionic terminals due to ionic-hydrophilic interactions between the Zwitterionic centre of L-serine and the -OH group of sucrose molecules decreases, which in turn, causing the larger values of ϕ_v^0 of L-serine in aqueous solution. Such large values of ϕ_v^0 in L-serine in aqueous sucrose solution is due to the strong hydrophilic-ionic interactions in the solution, as sucrose molecule contains a larger number of -OH groups than other disaccharides. Further, it is evident from the same Table-5 that the increasing trend of ϕ_v^0 values, suggesting the strong ion-solvent interaction in the solution.

The possession of such large of ϕ_v^0 in L-serine system clearly establishing that it serves as an effective structure-maker in the mixture. As seen from the table-5, the values of S_v are positive in all liquid systems and except in L-serine system, where it is found to be decreased with increase of mass percentage of sucrose. Such a decreasing of positive values of S_v clearly indicates the presence of weak ion-ion interactions in the solution^{34,24}.

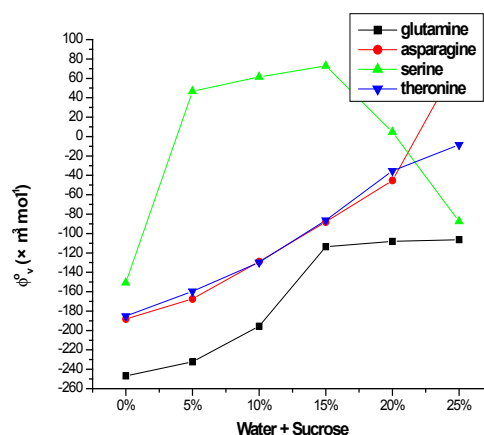


Fig. 4: Limiting apparent molar volume (ϕ_v^0) Vs mass percentage of sucrose at 301.15K

Partial transfer volume studies

The Partial molar transfer volume ($\Delta\phi_v^0$), of amino acids from water to aqueous sucrose solution at a given temperature quoted as.

$$\Delta\phi_v^0 = \phi_v^0 = (\text{in aqueous solution}) - (\text{in water})$$

Where $\Delta\phi_v^0$ denotes the transfer volume.

The present investigation observes that the partial transfer volume ($\Delta\phi_v^0$) exhibit positive deviations in all the four liquid systems and increase with elevation of mass percentage of sucrose content except in L-serine, (From Table-6) where it shows a reverse trend.

The increasing behavior of partial transfer volume values ($\Delta\phi_v^0$) shows stronger and more extensive interactions between amino acids (co-solute) and sucrose (solute) and vice versa. It is interesting to note that large value of $\Delta\phi_v^0$ are observed in L-serine system. The electrostriction at zwitterionic terminals due to ionic-hydrophilic interactions between the zwitterionic centre of amino acids and the -OH group of sucrose molecules decrease, which in turn, cause the larger values of " ϕ_v^0 " are dominant in L-serine system over the other co-solutes, which is attributed due to the existence of strong hydrophilic-ionic interaction

The partial transfer volume ($\Delta\phi_v^0$) can be explained on the basis of Co-Sphere Overlap Model³³. The overlap of ions of co-solute (amino acids) and solute (sucrose) comes into play because of the interaction between (i) ions of

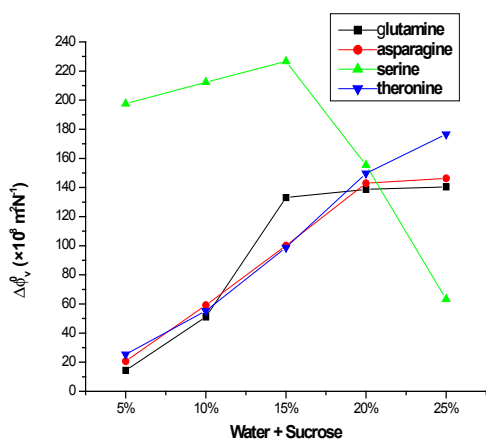


Fig. 5: Transfer volume ($\Delta\phi_v^0$) Vs mass percentage of sucrose at 301.15 K

solute (amino acids) and hydrophilic, -OH sites of disaccharide (sucrose) molecules, and (ii) ions of co-solutes (disaccharide) and the hydrophobic parts/groups of amino acid molecules. Out of which, the former type of interactions contributes positively, and later type of interactions to negatively to $\Delta\phi_v^0$ values. The possession of positive values of $\Delta\phi_v^0$ indicates the dominance of hydrophilic-ionic interactions. This may be further interpreted as the overlap of hydration co-spheres of two ionic species results in an enhanced volume as some electrostricted water molecules return to the bulk water with a higher volume contribution than electrostricted water molecules; whereas, overlap of hydration co-spheres of hydrophobic- hydrophobic groups and ion- hydrophobic/hydrophilic- hydrophobic groups and results in a net volume decrease. This may due to greater hydrophilic- ionic groups and hydrophilic- hydrophilic groups interactions, with the presence of more hydroxyl groups in sucrose molecules. Incidentally, the larger values of $\Delta\phi_v^0$ are observed in L-serine system comparing its other co-solutes. Hence it is very ambiguous that L-serine identified as an effective structure maker in aqueous sucrose solution as it enhances all possible interionic interactions such as solute-solvent, ion-solvent, solute-co solute etc (except ion-ion) in the mixture. Hence it is very obvious that of all the amino acids, L-serine is an effective structure maker in the present system of the mixture. Similar trend is already noticed by earlier workers, supports our present study^{25, 33}.

In general, the following types of interaction occurring between amino acids and sucrose can be classified as follows³⁶⁻³⁹

1. The hydrophilic- ionic interaction between OH groups of sucrose and zwitterions of amino acids.
2. Hydrophilic- hydrophilic interaction between OH groups of sucrose and OH groups in the side chain of amino acids.
3. Hydrophobic- hydrophilic interaction between OH groups of sucrose molecule and polar (-CH₂) in the side chain of amino acid molecule.
4. Hydrophobic- hydrophobic group interaction between the non-polar groups of sucrose and polar (-CH₂) in the side chain of amino acid molecule.

Viscometric studies

We have incorporated the viscometric study by employing the Jones-Dole equation⁴⁰.

$$\eta/\eta_0 = 1 + Am^{1/2} + Bm$$

Where η and η_0 are the viscosities of the solution and solvent respectively, m is the molar concentration ($\text{mol}\cdot\text{dm}^{-3}$) of the solute. A and B are constants which predict for a solute-solvent system. A is known as the Falkenhagen co-efficient which characteristics the ionic interaction in the solution and B is the Jones-Dole (or) viscosity B co-efficient, which depends on the size of the solute and the nature of the solute-solvent interactions.

Viscosity is one of the key transport properties of the solutions. Accurate viscosity data give useful information regarding ion-solvent interactions (long-range electrostatic interaction), which are the controlling force in dilute solutions. Viscosity data are necessary to calculate the physical parameters of Jones-Dole equation, which apply to analyze the experimental data⁴⁰⁻⁴¹. In the present study, the values of B for all studied system are positive and the positive values of the B co-efficient is attributed with structure making (ordering) ions. Our present study finds that the viscosity B co-efficient values are decreasing with increase mass percentage of sucrose content. However, in L-serine system, an increasing trend is observed. It is obvious that the increasing positive values of B in this system clearly attribute the dominance of solute-solvent interactions comparing the other three liquid systems.

For identifying the ionic interactions existing in the mixture, which is reflected by the values of A in the present investigation are all positive and decrease with increase of mass percentage of sucrose content (From Table-6), which clearly depicts the weakening of ion-ion interaction in the mixture. This is in well agreement with our earlier conclusion drawn from the associated constants of S_k and S_v .

CONCLUSION

The acoustical behaviors of α -amino acids in aqueous disaccharide (sucrose) at 301.15 K have been investigated and are summarized as,

1. It is obvious that L-serine serves as an effective structure - maker in the aqueous saccharide solution.
2. The addition of solute (amino acids) in the solvent enhances strong molecular interionic interactions such as ion-solvent, solute-solute, and solute-solvent etc., in the present systems of mixtures. and however a weak ion-ion interaction are observed.
3. Our viscometry study lends another fine support about the existence of strong solute-solvent interactions in the solution.
4. The trends of partial transfer volume studies predicting the dominance of ionic-hydrophilic interactions in the mixture.
5. Such predominating ionic-hydrophilic interactions between solute-cosolute are larger in L-serine system comparing the other co-solute systems.

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