ULTRASONIC STUDY OF SOME AMINO ACIDS IN AQUEOUS SALT SOLUTION OF KNO_3 AT 303.15K

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Abstract

Apparent molar compressibilities (φ_k) and apparent molar volume (φ_v) of L-valine, L-leucine andL-phenylalanine in aqueous solution of KNO_3 at different concentrations have been determined at the temperature 303.15K using a standard ultrasonic frequency of 2 MHz. The above parameters can be evaluated from precise density, ultrasonic velocity and time flow measurements using a specific gravity bottle, ultrasonic single frequency interferometer and Ostwald type capillary viscometer respectively. With the help of these results various ultrasonic derived parameters such as limiting apparent molar compressibilities (φ_k^o), limiting apparent molar volumes (φ_v^o), and their constants (S_k, S_v), viscosity A & B-coefficients and the corresponding transfer parameters ($\Delta\varphi_k^o$, $\Delta\varphi_v^o$ and ΔB), have been evaluated. The results have been interpreted in the light of intermolecular interactions between solute and solvent.

Keywords: *Amino acid, apparent molar compressibility, apparent molar volume, transfer parameter, viscosity B-coefficients.*

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1. INTRODUCTION

Interaction of proteins with their surrounding environment plays an important role in their conformational characteristics. These interactions are mainly those between the protein molecules and the solvent ions. Most of these interactions, such as hydrogen bonding and electrostatic interactions, have non-covalent nature. The study of these interactions provides important insight into the conformational stability and folding / unfolding behavior of globular proteins [1].To get a better understanding of these interactions, various low molecular weight model compounds, such as amino acids and peptides, have been studied because of the complexities of proteins and infeasibility of direct thermodynamic studies.

Amino acids are among the simplest biomolecules that contain intramolecular hydrogen bonds. They serve as building blocks on more complex peptides and proteins. In aqueous medium amino acids exist as dipolar ions manifesting a unique hydration behaviour which is linked to the vital biological phenomenon. Due to this linkage, the study of amino acids is considerably important in unfolding the role of dipolar ions in the biological system[2-3]. Amino acids also play a significant role in metabolism and in many neurochemical response mechanism such as, memory, appetite control, and pain transmission. 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 thermodynamical properties of the aqueous amino acids 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 solution have been employed to understand the nature of interactions operative in solution[4-6]. 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].

Potassium nitrate is used as a diuretic in medicine. It also include as an ingredient in tooth paste. It makes the teeth less sensitive to pain, by interfering with the transmission of pain signalsinthe nerves of teeth. It is also added to drugs for back pain and joint pain. Potassium nitrate affects nuclei acid synthesis is in the greening cucumber cotyledons[8] and the stability of tropomyosin[9]. Phenylalanine is found naturally in the breast milk of mammals. It is used in the manufacture of food and drink products and sold as a nutritional supplement for its reputed analgesic and antidepressant effects. Valine is an essential amino acid, hence it must be ingested, usually as a component of proteins. It is synthesized in plants via several steps starting from pyruvic acid. Leucine is the only dietary amino acid that has the capacity to stimulate muscle protein synthesis. As a dietary supplement, leucine has been found to slow the degradation of muscle tissue by increasing the synthesis of muscle proteins in aged rats[10].

This study mainly focuses on the measurement of density, viscosity, and ultrasonic velocity values of some amino acids such as L-valine, L-leucine and L-phenylalanine in aqueous KNO_3 solutions as functions of molar concentration at 303.15K. The measured density and ultrasonic velocity data

have been used to compute the apparent molar compressibility (φ_k), limiting apparent molar compressibility (φ^o_k), apparent molar volume (φ_v),limiting apparent molar volume (φ^o_v), transfer adiabatic compressibility ($\Delta\varphi^o_k$) and transfer volume ($\Delta\varphi^o_v$) values with the view to understand the zwitterion-ion, zwitterion-water dipole, ion-ion, and ion-water diploe interactions operative in the solutions.

2. MATERIALS AND METHODS

The amino acids L-valine, L-leucine and L-phenylalanine and the salt potassium nitrate of high purity has been used in the present studies, are purchased from E-Merck and SD fine chemicals (India). These chemicals were dried in a vacuum desiccator over P_2O_5 for 72 h before use. The triplicate distilled water (with specific conductivity of 1.29 X 10-6 Ω^{-1} cm^{-1}) was used for preparing all the amino acids and the stock solutions. All the solutions were stored in special airtight bottles to avoid exposure of solutions to air and evaporation.

Densities of the mixed solvent (salt + water) and amino acids solutions were measured using a specific gravity bottle by relative measurement method with an accuracy of ± 0.01kgm^{-3}. The specific gravity bottle was kept for about 30 minutes in a thermo – stated water bath for minimizing the thermal fluctuation in density. The ultrasonic velocities in mixed solvent and amino acids solutions were measured by using a single-crystal variable-path multi-frequency ultrasonic interferometer (Model: M-84, Mittal Enterprises, Delhi, India) with stainless steel sample cell operating at 2 MHz with an accuracy of ± 0.01 has been used for velocity measurements. The temperature of test solution during the measurement was maintained to an accuracy of ± 0.02K in an electrically controlled thermostatic water bath obtained from Raaga Industries. 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± 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 ± 0.001m.Pa.s.

3. RESULTS AND DISCUSSION

The experimentally calculated values of density (ρ), viscosity (η), ultrasonic velocity (U), adiabatic compressibility (β), apparent molar compressibility (φ_k), apparent molar volume (φ_v), limiting apparent molar compressibility (φ^o_k), limiting apparent molar volume (φ^o_v), transfer adiabatic compressibility ($\Delta\varphi^o_k$), transfer volume ($\Delta\varphi^o_v$), and viscosity coefficient of Jones-Dole equation for different molar concentration of the three amino acids such as L-leucine, L-valine and L-phenylalanine in aqueous potassium nitrate solutions are given in Tables 1, 2 and 3 Further, the Figs.1 & 2 show the variation of transfer adiabatic compressibility and transfer volume at 303.15 K.

Adiabatic compressibility

$$\beta = \frac{1}{U^2\rho} \qquad (1)$$

The apparent molar compressibility has been calculated from relation,

$$\varphi_k = \frac{1000}{M\rho_0}(\rho_o\beta - \rho\beta o) + \left(\frac{\beta o M_w}{\rho_o}\right) \qquad (2)$$

Where, β, ρ and βo, ρ_o are the adiabatic compressibility and density of solution and solvent respectively, Mis the molar concentration of the solute and M_w the molecular weight of the solute. φ_k is the function of Mas obtained by Gucker (1993)[11] from Debye Huckel[12] and is given by

$$\varphi_k = \varphi^o_k + S_k M^{1/2} \qquad (3)$$

Where, φ^o_k is the limiting apparent molar compressibility at infinite dilution and S_k is a constant. $\varphi^o_k$and S_k of equation 3 have been evaluated by least square method.

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

$$\varphi_v = \left(\frac{M_w}{\rho}\right) - \left(\frac{1000(\rho-\rho_0)}{M\rho\rho_0}\right) \qquad (4)$$

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

$$\varphi_v = \varphi^o_v + S_v M^{1/2} \qquad (5)$$

Where, $\varphi^o_v$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[13].

$$\left(\frac{\eta}{\eta_0}\right) = 1 + AM^{1/2} + BM \qquad (6)$$

Where, η and η_o are the viscosities of the solution and solvent respectively and M are the molar concentration of the solute. A is determined by the ionic attraction theory of Falkenhagen – Vernon[14] and therefore also called Falkenhagen coefficient B or Jones-Dole coefficient is an empirical constant determined by ion - solvent interactions.

Transfer adiabatic compressibility ($\Delta\varphi^o_k$) and transfer volume ($\Delta\varphi^o_v$) of each amino acid from water to aqueous cadmium chloride solutions have been calculated as:

$\Delta\varphi^{o}_{y} = \varphi^{o}_{y}$ (in aqueous cadmium chloride solution) - φ^{o}_{y}(in water) (7)

Where, φ^{o}_{y} denotes limiting apparent molar compressibility φ^{o}_{k}, limiting apparent molar volume φ^{o}_{v} and viscosity coefficient B.

The values of density, viscosity and ultrasonic velocity are found to increase with increase in molar concentration of amino acids as well as potassium nitrate (KNO_3) content (Table 1). This increasing trend suggests a moderate strong electrolytic nature in which the solutes (amino acids) tend to attract the solvent (aqueous potassium nitrate) molecules. Thus molecular interaction is responsible for the observed increase in density, viscosity and ultrasonic velocity in these mixtures. The factors apparently responsible for such behavior may be due to the presence of interactions caused by the proton transfer reactions of amino acids in aqueous potassium nitrate mixtures. The increasing trend of viscosity indicates the existence of ion-solvent interaction occurring in these systems. The increase in the ultrasonic velocity may be attributed to the cohesion brought about by the ionic hydration[15].

Table - 1: Values of density (ρ), viscosity (η) and ultrasonic velocity (U) of amino acids in aqueous potassium nitrate solutions at 303.15K

M/(mol.dm^{-3})	ρ / (kgm^{-3})			η / (x 10^{-3} Nsm^{-2})			U / (m.s^{-1})		
	Water + potassium nitrate (KNO_3)								
	0.0 M	0.5M	1.0 M	0.0M	0.5M	1.0M	0.0M	0.5M	1.0M
water + potassium nitrate + L-valine									
0.00	994.8	1011.7	1027.6	0.7210	0.7468	0.7723	1516.2	1520.9	1524.1
0.02	997.1	1016.2	1031.7	0.7319	0.7636	0.7898	1517.4	1522.6	1525.9
0.04	999.6	1020.6	1035.6	0.7428	0.7789	0.8013	1518.5	1524.3	1527.7
0.06	1002.3	1025.4	1039.5	0.7546	0.7914	0.8175	1519.6	1526.1	1529.5
0.08	1004.9	1030.2	1043.4	0.7661	0.8065	0.8351	1520.9	1527.4	1531.3
0.10	1007.6	1034.9	1047.5	0.7773	0.8231	0.8496	1522.2	1529.1	1533.1
Water + potassium nitrate + L-leucine									
0.00	994.5	1011.7	1027.6	0.7210	0.7468	0.7723	1516.2	1520.9	1524.1
0.02	998.1	1017.6	1033.1	0.7388	0.7712	0.7978	1512.6	1524.1	1527.3
0.04	1001.3	1021.3	1037.2	0.7516	0.7916	0.8213	1522.4	1527.3	1530.3
0.06	1004.1	1026.7	1040.9	0.7674	0.8161	0.8437	1524.9	1530.4	1533.6
0.08	1006.9	1031.5	1044.8	0.7832	0.8388	0.8632	1527.1	1533.3	1536.5
0.10	1009.8	1036.1	1049.1	0.8012	0.8611	0.8867	1530.4	1533.7	1539.3
Water + potassium nitrate + L-phenylalanine									
0.00	994.5	1011.7	1027.6	0.7210	0.7468	0.7723	1516.2	1520.9	1524.1
0.02	998.9	1019.1	1032.4	0.7423	0.7816	0.8086	1520.9	1526.3	1529.1
0.04	1002.7	1022.9	1038.7	0.7615	0.8154	0.8436	1525.1	1529.1	1532.4
0.06	1006.5	1028.1	1042.1	0.7807	0.8463	0.8759	1528.7	1532.1	1535.3
0.08	1009.3	1032.9	1045.9	0.7989	0.8776	0.9088	1531.1	1535.6	1538.1
0.10	1012.6	1037.6	1050.7	0.8181	0.9084	0.9416	1533.6	1537.6	1514.6

The values of adiabatic compressibility from Table-2 decreases with increase in concentration of solute (amino acids) as well as increase in concentration of aqueous KNO_3. 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^+, NO_3^-) called electrostriction. The magnitudes of β values are larger in L-valine than in other two amino acids. The larger β value shows molecular interactions are greater in L- valine than the other two amino acids. Amino acids 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.

The sensitive tool for understanding the molecular interactions in solution is the volumetric properties such as apparent molar compressibility (φ_k) and apparent molar volume (φ_v).

Table - 2 : Values of adiabatic compressibility (β), apparent molar compressibility (φ_k) and apparent molar volume (φ_v) of amino acids in aqueous potassium nitrate solutions at 303.15 K.

M/ (mol.dm^{-3})	β / (x 10^{-10} m^2 N^{-1})			-φ_k / (x 10^{-7} m^2 N^{-1})			- φ_v / (x 10^{-3} m^3 mol^{-1})		
	Water + potassium nitrate								
	0.0M	0.5M	1.0M	0.0M	0.5M	1.0M	0.0M	0.5M	1.0M
Water + potassium nitrate + L-valine									
0.00	4.3727	4.2731	4.1894	--	--	--	--	--	--
0.02	4.3557	4.2447	4.1629	1.4577	2.0626	2.3423	127.57	215.36	184.08
0.04	4.3385	4.2170	4.1374	1.4375	2.0883	2.3703	126.17	218.73	185.55
0.06	4.3206	4.1874	4.1122	1.4178	2.0952	2.3805	125.24	219.98	187.81
0.08	4.3021	4.1608	4.0872	1.3825	2.1154	2.3839	120.55	221.75	189.75
0.10	4.2832	4.1327	4.0617	1.3555	2.1608	2.3927	115.81	224.46	193.24
Water + potassium nitrate + L-leucine									
0.00	4.3727	4.2731	4.1894	--	--	--	--	--	--
0.02	4.3388	4.2305	4.1496	2.0899	2.5415	2.8365	166.03	286.41	258.87
0.04	4.3094	4.1976	4.1170	2.1043	2.5665	2.8986	160.50	240.47	224.97
0.06	4.2829	4.1586	4.0848	2.1780	2.6470	2.9140	155.03	236.97	207.07
0.08	4.2587	4.1236	4.0542	2.2858	2.7884	2.9625	150.85	232.20	200.07
0.10	4.2282	4.0925	4.0229	2.4203	3.111	3.3759	149.19	230.57	199.27
Water + potassium nitrate + L-phenylalanine									
0.00	4.3727	4.2731	4.1894	--	--	--	--	--	--
0.02	4.3279	4.2121	4.1427	3.1410	4.0814	4.6127	206.03	358.63	259.74
0.04	4.2878	4.1811	4.0998	2.9905	3.4826	4.0713	197.73	270.33	226.04
0.06	4.2515	4.1437	4.0710	2.8771	3.3111	3.9585	194.53	262.53	225.44
0.08	4.2264	4.1057	4.0415	2.6254	3.2117	3.4813	180.33	253.34	213.74
0.10	4.1989	4.0765	4.0048	2.5204	3.1134	3.1877	176.70	246.54	212.64

The following observations have been made on φ_k and $\varphi_v$from Table-2 of the three amino acids in aqueous potassium nitrate solutions at 303.15 K.

(i) The values of φ_k and φ_v are negative over the entire range of the molarity.
(ii) The negative values of φk and φv increase with the increase in concentration of amino acids, but it is
(iii) Found to decrease with increasing the contents of KNO3.
(iv) The magnitude of φk is in the order: L-valine > L-leucine > L-phenylalanine.

The negative values of φ_k and φ_v in all systems indicate the presence of ion-solvent interactions. Since more number of water molecules is available at lower concentration of potassium nitrate, the chances for the penetration of solute molecules into the solvent molecules are highly favored. The decrease in φ_v may be due to strong ion-ion interaction and vice-versa. The negative values of φ_v indicate electrostrictive solvation of ions [16]. From the magnitude of φ_k, it can be concluded that stronger molecular association is found in L-valine than in other two amino acids. Hence L- valine can be considered as an effective structure making in compared to the other two amino acids.

The limiting apparent molar compressibility φ_k° provides information regarding ion-solvent interaction and S_k to that of ion-ion interactions in the solution. From Table-3 it is observed that φ_k° values are negative and they increase with increasing the concentration of potassium nitrate in all systems studied. The negative values of φ_k°(loss of compressibility of the medium) indicate that the water molecules surrounding the amino acids molecules present a greater resistance to compression that the bulk [17]. The values of S_k exhibit positive value and they decrease with increasing the concentration of potassium nitrate in all the three amino acids. This behavior indicates the existence of ion-ion interaction with increase in potassium nitrate content. It is well known fact 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.

Table - 3: Values of limiting apparent molar compressibility (φ^o_k), limiting apparent molar volume (φ^o_v) and their constants S_k and S_v, transfer adiabatic compressibility ($\Delta\varphi^o_k$), transfer volumes ($\Delta\varphi^o_v$) , and A and B co- efficient of Jones-Dole equation of amino acids in aqueous potassium nitrate solution at 303.15 K.

Amino acids	Molarity (M) (mol.dm^{-3})	$-\varphi^o_k$ ($\times10^{-7}$ m^2 N^{-1})	$-\varphi^o_v$ ($\times10^{-3}$ m^3 mol^{-1})	S_k ($\times10^{-7}$ N^{-1} m^{-1} mol^{-1})	S_v ($\times10^{-3}$ m^3 L^{½} mol$^{-3/2}$)	$\Delta\varphi^o_k$ ($\times10^{-7}$ m^2 N^{-1})	$\Delta\varphi^o_v$ ($\times10^{-3}$ m^3 mol^{-1})	A (dm$^{3/2}$ mol$^{-1/2}$)	B (dm^3 mol^{-1})
L-valine	0.0	2.32	106.82	0.5885	168.52	--	--	0.0075	0.8053
	0.5	2.21	213.36	0.1298	149.97	0.11	54.43	0.0302	0.9052
	1.0	1.97	158.93	0.4333	125.71	0.35	106.54	0.0158	0.9382
L-leucine	0.0	3.62	180.11	1.9629	344.83	--	--	0.0209	1.0141
	0.5	3.50	306.48	2.6054	256.33	0.12	43.55	0.0180	1.4674
	1.0	3.08	264.93	3.2523	172.49	0.42	126.36	0.0529	1.3062
L-phenylalanine	0.0	4.67	231.95	3.6471	587.39	--	--	0.0343	1.2357
	0.5	4.24	417.52	3.9291	149.39	0.43	119.59	0.0508	2.0099
	1.0	3.99	297.93	3.7398	272.49	0.68	185.57	0.0467	2.0471

The volume behavior of a solute at infinite dilution is satisfactorily represented by φ^o_v which is independent of the ion-ion interactions and provides information concerning ion-solvent interactions. Similarly it is also observed from the Table-3 that the values of φ^o_v are negative in all the three amino acids. The values of φ^o_v increase with the addition of potassium nitrate contents in all the systems The increase in φ^o_v may be attributed to the increased hydrophilicity / polar character of the side chain of the amino acids. It is evident from the Table 3 that S_v is positive in all the three systems suggesting the presence of strong ion-ion interactions.

The values of transfer adiabatic compressibility $\Delta\varphi^o_k$ and transfer volume $\Delta\varphi^o_v$ (Table 3) are positive and they increase with increase in the concentration ofpotassium nitrate in all the three amino acid systems which suggest the existence of strong ion-solvent interactions in the mixtures.

Generally, the interaction between amino acids and potassium nitrate can be classified as:
(i) ion-ion interaction among the K^+, and NO_3^- ions and (COO^- , NH_3^+) Zwitter ionic end groups,
(ii) ion-hydrophilic interactions between ions and hydrophilic groups (-$CONH_2$,-CONH) of amino acids,
(iii) ion-nonpolar group interaction occurring between ions and the nonpolar groups (-CH_2 / -CH_3) of amino acids.

The $\Delta\varphi^o_k$ and $\Delta\varphi^o_v$ values can also be explained on the basis of co-sphere overlap model [18] in terms of solute-co-solute interactions. According to this model, ion-ion and ion-hydrophilic group interactions contribute positively, whereas ion-non-polar group interactions contribute negatively to the $\Delta\varphi^o_k$ and $\Delta\varphi^o_v$ values.

Therefore, from Figs.1-2, the positive $\Delta\varphi^o_k$ and $\Delta\varphi^o_v$ values observed in all the three amino acids suggest that the interaction contribution of type (i) and (ii) is much stronger than that of type (iii). The magnitude of $\Delta\varphi^o_v$ is in order: L-valine < L-valine < L-phenylalanine.

From the Table.3 it is observed that the values of A-coefficient are positive for all the systems indicating the presence of ion-ion interactions [19]. Further, the values of the B-coefficient are positive in all systems. 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 behavior 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-valine > L-leucine >L-phenylalanine. This conclusion is in excellent agreement with that drawn from φ_k and $\Delta\varphi^o_v$.

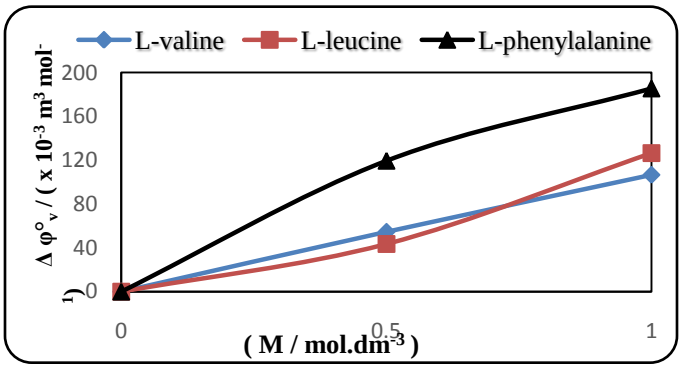

Fig.1 Variation of transfer adiabatic compressibility($\Delta\varphi^{o}_{k}$) of some amino acids with molarity ofaqueous potassium nitrate solutions at 303.15 K

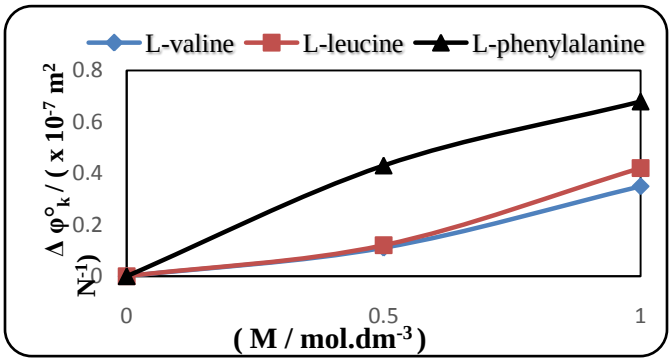

Fig.2 Variation of transfer volume ($\Delta\varphi^{o}_{v}$) of some amino acids with molarity of aqueous potassium nitrate solutions at 303.15K

4. CONCLUSIONS

In the present work volumetric, compressibility and transport parameters of L-valine, L-leucine and L-phenylalanine in aqueous potassium nitrate solutions at 303.15 K has been 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 $\varphi_{k,}$ $\Delta\varphi^{o}_{v}$ and the values of B-coefficient, it can be concluded that L-valine possesses strong ion-solvent interaction than the other two amino acids. The transfer adiabatic compressibility $\Delta\varphi^{o}_{k}$and transfer volume $\Delta\varphi^{o}_{v}$data suggest that ion-ion and ion-hydrophilic group interactions are dominating over the ion-non polar group interactions.

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