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MOLECULAR INTERACTION STUDIES OF GLYCYLGLYCINE IN AQUEOUS SODIUM HALIDE SOLUTIONS AT 303, 308 AND 313K

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ABSTRACT

Ultrasonic velocity (U), density (ρ) and viscosity (η) have been measured for glycylglycine in aqueous sodium halide (NaF, NaCl, NaBr) mixture (0.5m) at 303, 308 and 313K. Using the experimental values, the adiabatic compressibility (β), hydration number (n_H), apparent molal compressibility (φ_K), apparent molal volume (φ_V), limiting apparent molal compressibility (φ_K^0), limiting apparent molal volume (φ_V^0) and their constants (S_K, S_V), transfer volumes at infinite dilution ($\Delta\varphi_V^0$) and viscosity B-coefficient of Jones-Dole equation were calculated. These parameters have been interpreted the molecular interactions in terms of solute-co-solute and solute-solvent interaction present in the given solutions.

Keywords: glycylglycine, sodium halide mixture, ultrasonic velocity, apparent molal compressibility, apparent molal volume.

INTRODUCTION

Ultrasonic investigation in aqueous solutions of electrolytes and non-electrolytes with amino acids provides useful information in understanding the behaviour of liquid systems, because intramolecular and inter molecular association, complex formation and related structural changes affect the compressibility of the system which in turn produces corresponding variation in the ultrasonic velocity. During the last two decades, the ultrasonic study has been carried out to investigate hydration of proteins through volumetric and ultrasonic measurements, since these properties are sensitive to the degree and nature of hydration [1,2]. Due to the complex molecular structure of proteins, direct study is somewhat difficult. Therefore, the useful approach is to study simpler model compounds, such as amino acids which are building blocks of proteins. Most of the studies on amino acids [3,4] and biomolecules [5] have been carried out in pure and mixed aqueous solutions. Amino acids and peptides are the fundamental structural units of protein. The investigations on volumetric and thermodynamic properties of amino acids and peptides in aqueous and mixed aqueous solvents have been the area of interest of a number of researchers [6-8].

Most of the previous studies on oligopeptides have been restricted to water [9,10]. However, biological fluids are not pure water. Investigating the influences of solvents is therefore important. The properties of protein such as their structure, solubility, denaturation activity of enzymes, etc. are greatly influenced by electrolytes [11,12]. Though there are some researcher's invetigating this property of glycylglycine in aqueous NaCl solutions [13,14], no systematic studies exists on the volumetric properties of oligopeptides in electrolyte solutions. For complete understanding of the influence of electrolytes on oligopeptides, we present in this study, the values of densities, viscosities and ultrasonic velocities in aqueous sodium halides solution at 303, 308 and 313K. Only sodium salts have been chosen for this part of the isolated study to observe the relative effects of the change of anion of the electrolyte.

However, the ultrasound velocity data as such do not provide significant information about the native and relative strength of various types of intermolecular or interionic interactions between the components. Hence their derived parameters such as adiabatic compressibility (β), hydration number (n_H), apparent molal compressibility (φ_K) apparent molal volume (φ_V), limiting apparent molal compressibility (φ_K^0), limiting apparent molal volume (φ_V^0), transfer volumes at infinite dilution ($\Delta\varphi_V^0$) and their constants (S_K, S_V), and viscosity B-coefficient have been obtained to shed more light on such interactions.

MATERIALS AND METHODS

Analytical reagent (AR) and spectroscopic reagent (SR) grades with minimum assay of 99.9% of glycylglycine, sodium fluoride (NaF), sodium chloride (NaCl) and sodium bromide (NaBr) were obtained from E-Merck, Germany and SdFine chemicals, India, which are used as such without further purification. Water used in the experiment was deionised, distilled and was degassed prior to making solutions. Aqueous solutions of sodium halides (0.5 mol·kg^{-1}) were prepared by mass and used on the day they were prepared. Solution of glycylglycine in the concentration range of 0.02-0.1 mol·kg^{-1} were made by mass on the molality concentration scale with 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 ± 0.01 kgm^{-3}. An Ostwald's viscometer (10 ml) was used for the viscosity measurement. Efflux time was determined using a digital chronometer within ±0.01s. An ultrasonic interferometer having the frequency of 3 MHz (MITTAL Enterprises, New Delhi, Model: F-81) with an overall

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accuracy of ± 0.1% has been used for velocity measurement. An electronically digital operated constant temperature bath (Raaga Industries) has been used to circulate water through the double walled measuring cell made up of steel containing the experimental solution at the desired temperature. The accuracy in the temperature measurement is ± 0.1 K.

THEORY AND CALCULATIONS
Various acoustical and thermodynamical parameters are calculated from the measured data such as adiabatic Compressibility

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

The molal hydration number has been computed using the relation,

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

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

The apparent molal compressibility has been calculated from relation,

$$\varphi_K = \frac{1000}{m\rho_0}\left(\rho_0\beta - \rho\beta_0\right) + \left(\frac{\beta_0 M}{\rho_0}\right) \qquad \dots (3)$$

Where β, ρ and β_0, ρ_0 are the adiabatic compressibility and density of solution and solvent respectively, m is the molal concentration of the solute, and M the molecular mass of the solute. φ_K is the function of m as obtained by Gucker [15] from Debye Huckel theory [16] and is given by

$$\varphi_K = \varphi_K{}^0 + S_K m^{½} \qquad \dots (4)$$

Where $\varphi_K{}^0$ is the limiting apparent molal compressibility at infinite dilution and S_K is a constant. $\varphi_K{}^0$ and S_K of equation (4) have been evaluated by the least square method.

The apparent molal volume φ_V has been calculated using the relation

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

The apparent molal volume φ_V has been found to differ with concentration according to Masson [17] empirical relation as

$$\varphi_V = \varphi_V^0 + S_V m^{½} \qquad \dots (6)$$

Where φ_V^0 is the limiting apparent molal volume at infinite dilution and S_V is a constant and these values were determined by least square method.

Transfer volumes ($\Delta\varphi_V^0$) of glycylglycine in aqueous sodium halide solution at 303, 308 and 313K, $\Delta\varphi_V^0$ from water to aqueous sodium halide solution have been calculated by the equation

$$\Delta\varphi_V^0 = \varphi_V^0 \text{ (in aqueous sodium halide solution)} - \varphi_V^0 \text{ (in water)} \qquad \dots (7)$$

Where φ_V^0 denotes limiting apparent molal volume.

The viscosity A and B coefficients for the amino acids in aqueous sodium halide solutions were calculated from the Jones-Dole equation [18].

$$\frac{\eta}{\eta_0} \;=\; 1 + Am^{½} + Bm \qquad \dots (8)$$

Where, η and η_0 are the viscosities of the solution and solvent respectively and m is the molal concentration of the solute. A is determined by the ionic attraction theory of Falkenhagen-Vernon and therefore also called Falkenhagen coefficient [19] B or Jones-Dole coefficient is an empirical constant determined by ion-solvent interactions.

RESULTS AND DISCUSSIONS
The experimental values of ultrasonic velocity (U), density (ρ) and viscosity (η) for different molal composition of glycylglycine in aqueous sodium halides (NaF, NaCl, NaBr) solution (0.5m) at 303, 308, 313K are shown in Table-1. The values of adiabatic compressibility (β), hydration number (n_H), apparent molal compressibility (φ_K) apparent molal volume (φ_V), limiting apparent molal compressibility (φ_K^0), limiting apparent molal volume (φ_V^0) and their constants (S_K, S_V), transfer volumes at infinite dilution ($\Delta\varphi_V^0$) and viscosity B-coefficient of Jones-Dole equation are shown in Tables 2 to 3.

In all the three systems (Table-1) the values of density increases with increase in molal concentration of glycylglycine, but it decreases with rising of temperature. The increasing trend suggests a strong electrolytic nature in which the solutes (glycylglycine) tend to attract the solvent (Sodium halides) molecules. The decreases in density with elevation of temperature in all the three systems indicate decrease in inter molecular forces due to increase in the thermal energy of the system, which causes increase in volume expansion. Further it is observed that the value of ultrasonic velocity increases (Table-1) with increase in concentration of glycylglycine as well as increasing of temperature. The existence of molecular interaction between solute and solvent molecules is thus

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responsible for the observed increase in ultrasonic velocity in these mixtures. The increase in ultrasonic velocity in these solutions may be attributed to the cohesion brought about by the ionic hydration.

The values of adiabatic compressibility (Table-2) decreases with increasing the concentration of the glycylglycine as well as temperatures in all systems studied. The decreasing adiabatic compressibility observed in glycylglycine in aqueous sodium halide mixture at all temperature generally confirms that conclusion drawn from the velocity data. The increasing electrostrictive compression of water around the molecules results in a large decrease in the compressibility of solutions. The decrease in compressibility implies that there is enhanced molecular association in these systems an increase solute content, as the new entities (formed due to molecular association) become compact and less compressible [20].

The interaction between the solute and the water molecules in the solvent is termed as hydration. The positive value of hydration number (Table-2) indicates an appreciable solvation of solutes [21]. This is added support for the structure promoting nature of the solute as well as the presence of appreciable dipole-dipole interaction between solute and water molecules. This also suggests that compressibility of the solution will be less than that of the solvent. As a result, solutes will gain mobility and have more probability of contacting solvent molecules. This may enhance the interaction between solute and solvent molecules.

Further from Table-2, it is observed that the values of φ_K and φ_V are all negative and vary non-linearly over the entire range of molality of solutes. This observation clearly suggests that the negative values of φ_K and φ_V indicates ionic and hydrophilic interactions occurring in these systems. A non linear variation of φ_K and φ_V values in all the systems studied which indicates an existence of solute-solvent interaction.

φ_K^0 provides information regarding solute-solvent interaction and S_K that of solute–solute interaction in the mixtures. From the Table-3, it is found that the φ_K^0 values are negative and decreases with rising of temperature in all the systems studied. Appreciable negative values of φ_K^0 for all systems reinforce our earlier view that existence of solute-solvent interaction. The magnitude of φ_K^0 is in the order of NaF > NaCl > NaBr. The values of S_K exhibits negative for aqueous NaF and NaCl, but it found to be positive in aqueous NaBr solution. But these values are varies non-linearly with rise in temperature. This behaviour indicates the existence of ion-ion / solute-solute interactions present in the systems.

The volume behaviour of a solute at infinite dilution is satisfactorily represented by φ_v^0 which is independent of the solute-solute interactions and provides information concerning solute-solvent interactions. Table-3 reveals that the values of φ_v^0 are both positive and negative in aqueous NaF and NaCl but it is found to be negative in aqueous NaBr solution. Large positive values suggesting the presence of strong solute-solvent interactions and vice-versa. F⁻, Cl⁻ and Br⁻ have the same charge, but Br⁻ has the biggest ionic radius, and F⁻ has the smallest ionic radius. The ionic radius is bigger; the interaction between the charged centres of glycylglycine and ions (F⁻, Cl⁻ and Br⁻) is weaker. On the condition of the same molality of sodium halides, the weaker interaction causes the smaller φ_v^0 of glycylglycine. So φ_v^0 of glycylglycine are the biggest in aqueous NaF solution and the ones in aqueous NaBr solution are smallest. The φ_v^0 are decreases with rise in temperature due to the increase in the electrostriction at the terminals also it makes disruptions of side group hydration by that of the charged end. The S_V values (Table-3) for NaF and NaCl are found to be negative and positive for NaBr. The negative values of S_V indicate the presence of stronger solute-solute interaction and less complex ion formation taking place in the systems.

Generally, speaking the types of interactions occurring between the charged centres of glycylglycine and ions (Na⁺, F⁻, Cl⁻ and Br⁻) can be classified as follows:

i. ions of co-solutes (Na⁺, F⁻, Cl⁻ and Br⁻) and hydrophilic part of charged centre of glycylglycine; and

ii. ions of co-solute and hydrophobic parts of non-polar side group of glycylglycine.

These tendencies can also be explained using the co-sphere overlap model [9,22]. According to this model, hydrophilic-ionic group interactions contribute positively, where as ionic-hydrophobic group interactions contribute negatively to the $\Delta\varphi_v^0$ values. Therefore, from Figure-1 the positive values of $\Delta\varphi_v^0$ indicates that the interaction between the charged centres of glycylglycine (hydrophilic) and ions (co-solutes) dominates other forms interactions. The $\Delta\varphi_v^0$ values are small in NaBr than NaCl and NaF. Because the Br⁻ has the small charge and big ion radius, the electrostriction is weaker. So comparing the $\Delta\varphi_v^0$ in aqueous NaCl and NaF solutions, the value of $\Delta\varphi_v^0$ in aqueous NaBr solution is the smallest. Further, in all systems the $\Delta\varphi_v^0$ values are decreasing with rise of temperature indicates the weakening of ion-ion interaction.

Viscosity is another important parameter in understanding the structure as well as molecular interactions occurring in the mixtures. From the Table-1, it is observed that the values of viscosity increases with increase in concentration of glycylglycine in aqueous NaF, NaCl and NaBr (0.5m) at 303, 308 and 313K and

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decreases with increasing of temperature. This increasing trend indicates the existence of molecular interaction occurring in these systems studied. In order to shed more light on this, the role of viscosity B-coefficient has been obtained. From the Table-3, it is observed that the values of A are both negative and positive in all systems studied and B-coefficient are positive. Since A is measure of ionic interaction[23] and it is evident that the positive value of A indicates there is a strong solute-solute interaction and vice-versa in the systems. B-coefficient is also known as

measure of solute-solvent interaction and the relative size of the solute 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 in the order of NaF > NaCl > NaBr. This conclusion is an excellent agreement with that drawn from S_V and φ_v^0 data and the larger values of B indicate structure making capacity of the solute.

Table-1. Values of density (ρ), viscosity (η) and ultrasonic velocity (U) of glycylglycine in aqueous sodium halides (0.5m) at 303, 308 and 313K for.

M/ (mol·dm^{-3})	ρ/(kg.m^{-3})			η/($\times 10^{-3}$ Nsm^{-2})			U/(ms^{-1})		
	303 K	308 K	313 K	303 K	308 K	313 K	303 K	308 K	313 K
System - I glycylglycine + Water + NaF									
0.00	891.6	889.2	888.9	0.8559	0.7832	0.7199	1542.8	1548.9	1554.8
0.02	893.2	890.7	889.6	0.8668	0.7909	0.7342	1550.7	1556.1	1564.1
0.04	895.2	892.4	890.8	0.8755	0.7992	0.7396	1559.4	1566.2	1570.4
0.06	898.7	894.0	891.9	0.8899	0.8054	0.7462	1564.5	1572.5	1576.8
0.08	899.9	895.3	892.5	0.8977	0.8134	0.7526	1566.0	1574.8	1581.1
0.10	911.8	896.7	893.4	0.9153	0.8213	0.7577	1590.4	1600.8	1602.0
System - II glycylglycine + Water + NaCl									
0.00	892.8	891.6	889.5	0.8777	0.7956	0.7283	1553.1	1554.2	1562.9
0.02	894.1	893.0	890.7	0.8843	0.8043	0.7396	1559.1	1564.2	1572.6
0.04	895.9	893.3	892.9	0.8908	0.8102	0.7499	1560.8	1567.8	1580.0
0.06	899.0	895.1	893.8	0.8999	0.8177	0.7563	1570.8	1579.7	1585.2
0.08	902.5	896.9	894.5	0.9088	0.8228	0.7632	1574.7	1580.2	1590.9
0.10	912.7	897.7	895.8	0.9239	0.8320	0.7702	1609.5	1612.8	1620.7
System - III glycylglycine + Water + NaBr									
0.00	907.69	905.52	902.98	0.8980	0.8167	0.7488	1564.2	1571.4	1567.7
0.02	907.75	906.54	905.48	0.9024	0.8244	0.7550	1588.2	1595.1	1597.9
0.04	908.65	907.94	906.87	0.9104	0.8315	0.7629	1602.1	1606.9	1609.6
0.06	909.36	908.27	907.57	0.9160	0.8374	0.7686	1621.9	1627.3	1634.1
0.08	911.09	910.86	907.82	0.9232	0.8450	0.7753	1633.9	1638.2	1643.4
0.10	913.89	911.30	908.21	0.9304	0.8509	0.7830	1642.2	1650.0	1658.4

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Table-2. Values of adiabatic compressibility **(**β), hydration number **(** n_H), apparent molal compressibility **(** φ_K) and apparent molal volume **(** φ_V) of glycylglycine in aqueous sodium halides (0.5m) at 303, 308 and 313K for.

M/ (mol·dm^{-3})	β/(×10^{-10} m^2 N^{-1})			n_H			- φ_K /(×10^{-8} m^2 N^{-1})			- φ_V /(10^{-3} m^3 mol^{-1})		
	303 K	308 K	313 K	303 K	308 K	313 K	303 K	308 K	313 K	303 K	308 K	313 K
System - I glycylglycine + Water + NaF												
0.00	4.7121	4.6876	4.6532	-	-	-	-	-	-	-	-	-
0.02	4.6558	4.6365	4.5949	52.66	29.47	33.87	32.38	29.50	30.98	100.41	94.65	37.89
0.04	4.5937	4.5682	4.5520	34.08	34.55	12.66	34.36	34.07	27.78	112.71	100.77	56.78
0.06	4.5460	4.5236	4.5095	31.76	31.53	27.83	33.94	31.55	26.57	147.63	100.59	60.91
0.08	4.5313	4.5038	4.4820	25.90	26.47	24.84	28.08	26.99	23.76	129.26	95.73	55.09
0.10	4.3360	4.3239	4.3182	43.08	41.88	38.86	48.29	43.38	39.36	248.43	165.68	138.67
System - II glycylglycine + Water + NaCl												
0.00	4.6435	4.6431	4.6025	-	-	-	-	-	-	-	-	-
0.02	4.6011	4.5768	4.5398	23.61	36.92	35.23	24.58	36.80	34.71	81.36	87.85	75.67
0.04	4.5819	4.5543	4.4863	17.28	24.91	32.88	19.43	24.41	33.58	96.83	53.30	106.95
0.06	4.5082	4.4769	4.4524	25.32	31.10	28.34	27.92	30.74	28.81	128.68	73.08	90.08
0.08	4.4685	4.4651	4.4171	24.50	24.92	26.19	28.18	25.70	26.47	150.42	82.78	78.49
0.10	4.3008	4.2826	4.2500	41.82	43.99	43.40	44.62	39.23	38.56	244.15	76.15	79.00
System - III glycylglycine + Water + NaBr												
0.00	4.5027	4.4724	4.4547	-	-	-	-	-	-	-	-	-
0.02	4.3676	4.3357	4.3253	67.02	68.28	64.89	67.55	70.82	70.87	59.56	60.80	152.76
0.04	4.2875	4.2656	4.2560	53.30	51.57	49.74	55.04	54.79	54.49	30.20	72.87	118.94
0.06	4.1802	4.1575	4.1262	53.17	19.97	54.75	55.15	54.79	58.53	34.21	56.63	93.43
0.08	4.1113	4.0907	4.0787	48.54	47.66	49.16	51.03	51.05	49.96	51.28	81.72	73.08
0.10	4.0574	4.0306	4.0035	44.23	44.18	46.89	47.60	47.04	47.69	74.63	70.18	63.29

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Table-3. Values of limiting apparent molal compressibility (φ_k^0), constant (S_K), limiting apparent molal volume (φ_v^0),

transfer volume ($\Delta \varphi_v^0$), constant (S_V) and A and B coefficients of Jones-Dole equation of glycylglycine in

aqueous sodium halides (0.5m) at 303, 308 and 313K for.

System	- φ_k^0 /($\times 10^{-7} m^2 N^{-1}$)			S_k/($\times 10^{-7} N^{-1} m^{-1}.mol^{-1}$)			φ_v^0 /($\times 10^{-3} m^3.mol^{-1}$)			$\Delta \varphi_v^0$ /($\times 10^{-3} m^3.mol^{-1}$)			S_V/($\times m^3 lt^{½}.mol^{-3/2}$)			-A / $\times 10^{-2} dm^{3/2} mol^{-1/2}$			B /$\times 10^{-2} dm^3 mol^{-1}$)		
	303 K	308 K	313 K	303 K	308 K	313 K	303 K	308 K	313 K	303 K	308 K	313 K	303 K	308 K	313 K	303 K	308 K	313 K	303 K	308 K	313 K
glycylglycine + aqueous NaF	22.88	15.37	24.48	-52.84	-74.78	-21.98	-14.39	-42.78	-33.32	62.37	34.34	44.17	-683.61	-289.80	-435.21	13.05	0.497	11.12	62.98	46.75	36.64
glycylglycine + aqueous NaCl	4.32	30.17	25.88	-103.86	-5.08	-27.62	-57.79	-74.55	-75.24	18.97	2.57	2.25	-836.19	-3.05	-34.58	-3.12	2.10	6.06	57.62	37.37	39.08
glycylglycine + aqueous Na Br	79.89	84.69	85.04	103.83	122.32	120.96	-42.24	-53.19	-65.47	34.52	23.93	12.02	360.58	64.30	549.17	-4.69	8.52	-0.56	52.82	39.44	27.07

Literature values of φ_v^0 / ($\times 10^{-3} m^3.mol^{-1}$) of glycylglycine in water:
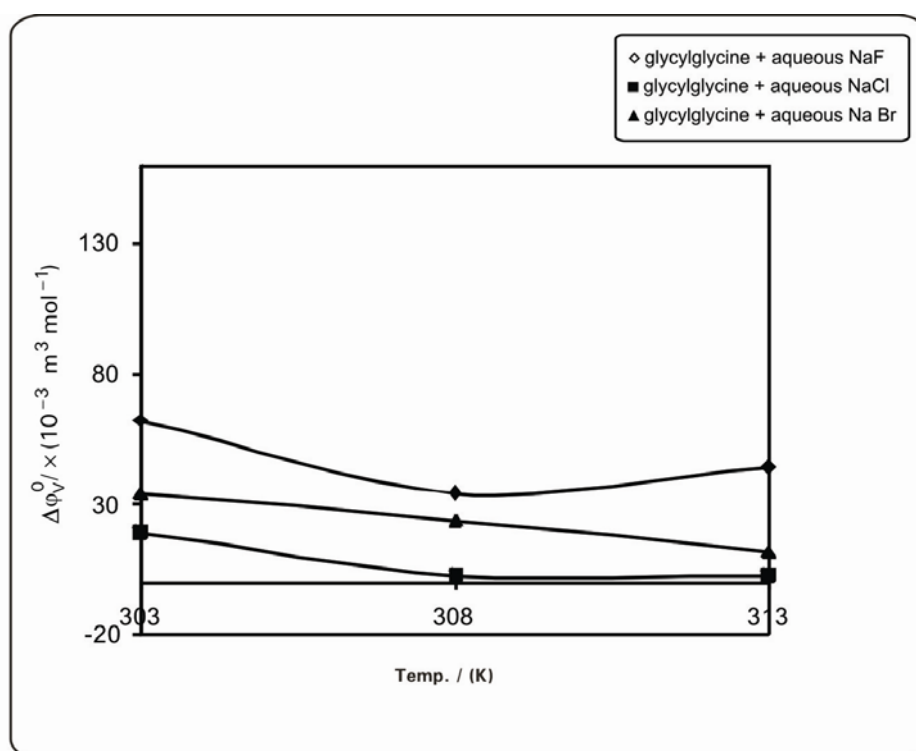-76.76; -77.12; and -77.49 at 303, 308 and 313K.

Fig. 1. Variation of transfer volume at infinite dilution with temperature of glycylglycine in aqueous sodium halide solution (0.5 m)

CONCLUSIONS

In summary volume and compressibility data have been determined for glycylglycine in aqueous sodium halides (NaF, NaCl and NaBr) solutions at 303, 308 and 313K and the results have been used to study the molecular interaction in the solutions. From the magnitude of φ_K^0 and B-coefficient values it can be concluded that the existence of molecular interaction is in the order of NaF > NaCl > NaBr. This suggests glycylglycine in aqueous NaF solution posses strong structure maker than other two systems. The positive $\Delta \varphi_v^0$ of glycylglycine from water to aqueous sodium halide solutions show that the interactions involving the charged centers of peptide as well as ions are dominating. Further, with rising of temperature the ion-solvent interaction gets weakened due to increase in the thermal energy of the system.

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