CHAPTER - VII

PROBING SOLUTE-SOLVENT INTERACTIONS OF SOME BIO-ACTIVE SOLUTES IN AQUEOUS BARIUM NITRATE SOLUTION ON THE BASIS OF PHYSICO-CHEMICAL CONTRIVANCES

7.1. Introduction

Amino acids are the bioactive compounds playing central roles both as building blocks of proteins and as intermediates in metabolism. They are very much useful in food technology and industry.

Limiting apparent molar volume (ϕ_V^0), experimental slopes (S_V^*), and viscosity *B*-coefficients, molar refraction (*R*) and limiting apparent molar adiabatic compressibility (ϕ_K^0) of a solute reflect the cumulative effects [1-5] of the interactions like ion-ion, ion-solvent, and solvent-solvent interactions in solutions. In this paper we have attempted to study these properties for three amino acids namely Glycine, L-Alanine, and L-Valine in aqueous Barium Nitrate solutions at 298.15 K to understand the solute-solute and solute-solvent interactions.

7.2. Experimental Methods

7.2.1. Source and Purity of Samples

The mass fraction of purity of Barium Nitrate of puriss grade (Sigma-Aldrich, Germany) was ≥ 0.99. It was recrystallised from aqueous Ethanol solution after being dried under vacuum at T = 348 K and it was stored over P_2O_5 kept in a desiccators [6]. The three amino acids namely Glycine, L-Alanine and L-Valine (S.D. Fine Chemicals, >99%) were taken without any further purification. Barium Nitrate solution was prepared with freshly distilled conductivity water. Table 1. depicts the physical properties of different mass fraction of aqueous Barium Nitrate solution.

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7.2.2. Apparatus and Procedure

Density (ρ) was measured with Vibrating-tube Anton Paar Density-Meter (DMA 4500M) with a uncertainty of 0.00005 g.cm^{-3} after being calibrated by double-distilled water and dry air [7]. The temperature was kept fixed within ± 0.01 K.

Ubbelohde type viscometer was used to measure the viscosity (η) whilst calibrated at 298.15 K with distilled water and pure Methanol. A thoroughly cleaned and dried viscometer containing the experimental liquid was placed vertically in a glass-walled thermostat (Bose Panda Instruments Pvt. Ltd.). The flow times were accurate to ±0.1s. Airtight-stopper bottles were used for the preparation of the mixtures. A triplicate measurement was taken into account with sufficient precautions to minimize the evaporation loss. A Mettler AG-285 electronic balance with a uncertainty of ±0.01 mg was used to measure the mass. The uncertainty of density measurements was ± 3·10^{-4} g·cm^{-3}.

Viscosity of the solution is evaluated using the appropriate equation as reported earlier [8]. The uncertainty in viscosity measurements was within ± 0.003 mPa·s.

Refractive index was calculated by means of Digital Refractometer Mettler Toledo. The light source was LED, λ=589.3nm. The calibration of refractometer was done twice using distilled water and being checked after every few measurements. The uncertainty of refractive index measurement was ± 0.0002 units.

An ultrasonic interferometer (Model M-83 from Mittal enterprises) was utilized to measure the ultrasonic velocities, u (ms^{-1}). The interferometer (2 MHz) is based on the principle used by Freyer et al. [9] and Kiyoharo et al. [10,11]. The obtained velocities were corrected for diffraction errors as given by Subrahmayan et al.[12]. The maximum uncertainty in the velocity was ±0.5 m s^{-1}. The temperature control were carried out within ±0.01 K using a Lauda thermostat for velocity measurements.

The solutions under investigation were prepared by mass and the conversion of molarity into molality was accomplished [4] from the studied

density values. The experimental values of densities *(ρ)*, viscosities (η), refractive indices (n_D) and ultrasonic speeds (*u*) of solutions are reported in Table 2 and the derived parameters are listed in Table 3 and Table 4.

7.3. Results & Discussion

7.3.1. Density Calculation

The determination of Apparent molar volumes (ϕ_V) were done from the solution densities using the following equation [13].

$$\phi_V = M / \rho - 1000(\rho - \rho_o) / m\rho\rho_o \quad (1)$$

where *M* is the molar mass of the solute, *m* is the molality of the solution ρ_0 and ρ are the densities of the solution and the mixture respectively. The limiting apparent molar volume ϕ_V^0 was obtained using a least-square treatment to the plot of ϕ_V versus $\sqrt{m}$ with the help of Masson equation [14].

$$\phi_V = \phi_V^0 + S_V^* m^{1/2} \quad (2)$$

where ϕ_V^0 is the limiting apparent molar volume at infinite dilution and S_V^* is the experimental slope. The plots of ϕ_V against square root of molal concentration ($\sqrt{m}$) were found to be linear. Values of ϕ_V^0 and S_V^* are reported in Table 4.

A perusal of Table 4 reveals that ϕ_V^0 values for amino acids are positive and increase with an increase in concentrations of aqueous Barium Nitrate mixture, indicating the presence of strong solute-solvent interactions and these interactions are further strengthened with the increase of the mass fraction of $Ba(NO_3)_2$ in the solvent mixture. The trend in the solute-solvent interaction is

Glycine < L-Alanine < L-Valine

This indicates that the solute-solvent interaction increases with the increase in the number of carbon atoms in the three amino acids.

The S_V^* values of the amino acid solution (Table 4) follow a decreasing order with the increase in the number of carbon atoms of the above mentioned amino

acids and also the mass fraction of Barium Nitrate in its aqueous mixture. Thus solute-solute interaction is minimized in the higher analogs.

The predominance of solute-solvent interactions over solute-solute interactions was cemented from the higher values of ϕ_V^0 than S_V^* obtained from Figure 1. The same result was obtained for each of the studied amino acids as well as mass fraction of Barium Nitrate in the mixture.

7.3.2. Viscosity Calculation

The viscosity data has been analyzed using Jones-Dole equation [15]

$$\left(\eta / \eta_0 - 1\right) / \ m^{1/2} = A + Bm^{1/2} \qquad (3)$$

where η_0 and η are the viscosities of the solvent and solution respectively. *A* and *B* are the viscosity co-efficient estimated by a least-squares method and are reported in Table 4. The values of the *A* co-efficient are found to decrease with the increase in the number of carbon atoms of amino acids (starting from Glycine to L-Valine) and with the increase in mass fraction of Barium Nitrate in solvent mixture. The results reveal the presence of very weak solute-solute interactions. These results are in excellent agreement with those obtained from S_V^* values discussed earlier.

The effects of solute-solvent interactions on the solution viscosity can be inferred from the *B*-coefficient [16,17]. The viscosity *B*-coefficient is an important tool to provide information concerning the solvation of the solutes and their effects on the structure of the solvent. From Table 4 and Figure 2 it is evident that the values of the *B*-coefficient are positive for all the amino acids and increase with the increase of the size of the side chains. This is due to the greater structure breaking tendency of L-valine as compared to L-alanine [18]. The *B*-coefficients reflect the net structural effects of the charged groups and the hydrophobic CH_2- groups on the amino acids. Thus solute-solvent interaction predominates and increases with the increase of mass fraction of $Ba(NO_3)_2$ in the

solvent mixture. This observation is in a fine agreement with the results obtained from ϕ_V^0 values discussed earlier.

7.3.3. Refractive Index Calculation

The molar refraction, R has been evaluated from Lorentz-Lorenz relation [19]

$$R = \left\{(n_D^2\text{-}1)/(n_D^2 + 2)\right\}(M/\rho) \quad (4)$$

where R, n_D, ρ and M are the molar refraction, the refractive index, density and the molar mass of the solution respectively. According to Born et al.[20] the greater the refractive index of a substance, the more the light is refracted and it was further supported by Deetlefs et al.[21].

More tightly packed substance i.e. more denser solution has higher refractive index. So refractive index value rises with the increase of mass fraction of Barium Nitrate in solvent mixture. Hence from Table 2 & Table 3 it is clear that for L-Valine the refractive index and the molar refraction values are higher than the other two amino acids. Thus solute-solvent interaction predominates over solute-solute interaction. This fact also supports the results obtained from density and viscosity parameters discussed above. The studied amino acids in aqueous Barium Nitrate solution obey the same trend of solute solvent interaction i.e.

Glycine < L-Alanine < L-Valine.

7.3.4. Ultrasonic Speed Calculation

The adiabatic compressibility (β) was evaluated with the help of the following equation:

$$\beta = 1/u^2\rho \quad (5)$$

where u is the sound speed in the solution and ρ is the solution density. The apparent molal adiabatic compressibility (ϕ_K) of the solutions was analyzed from the relation.

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$$\phi_K = M\beta / \rho + 1000\left(\beta\ \rho_o - \beta_o \rho\right)\ / m\ \rho\ \rho_o \qquad (6)$$

where β and β_0 are the adiabatic compressibility of the solution and solvent respectively and *m* is the molality of the solution. Limiting partial molal adiabatic compressibilities (ϕ_K^0) and experimental slopes (S_K^*) were achieved by fitting ϕ_K against the square root of molality of the electrolyte ($\sqrt{m}$) using the least-squares method.

$$\phi_K = \phi_K^0 + S_K^* m^{1/2} \qquad (7)$$

The values of β and ϕ_K are enlisted in Table 3 and the values of ϕ_K^0 and S_K^* are presented in Table 4. The values of ϕ_K^0 and S_K^* also represent the extent of solute-solvent and solute-solute interactions respectively. An inspection of Table 4 and Figure 3 confirms that the ϕ_K^0 values are in an excellent agreement with those drawn from the values of ϕ_V^0 discussed earlier.

Owing to the greater +I effect of alkyl chain in L-Valine in comparison with other two studied amino acids negative charge density becomes maximum on the oxygen atom of the carboxylic part. So the solute-solvent interaction of L-Valine with Ba^{2+} ion is highest among the three and thereby supporting the order of interactions mentioned above.

7.4. Conclusions

The values of the limiting apparent molar volume (ϕ_V^0), viscosity *B*-coefficients and limiting partial isentropic compressibility (ϕ_K^0) indicate the presence of strong solute-solvent interactions which increases with the increase in the number of carbon atoms of the amino acids and also with the increase of mass fraction of Barium Nitrate in the aqueous mixture. The refractive index and the molar refraction values indicate that molecules of L-Valine are more tightly packed in the solution resulting higher solute-solvent interaction than the other two amino acids.

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Table 1. The values of Density (ρ), Viscosity (η), Refractive index (n_D), and Speed of sound (u) in different mass fraction of Barium Nitrate at 298.15K and Experimental Pressure 0.1MPa.

Mass-fraction of Barium Nitrate	[a] ρ x 10^{-3} (kg m^{-3})	[b] η (mPa s)	[c] n_D	[d] u (ms^{-1})
w_1 = 0.01	0.99737	0.904	1.3327	1512.7
w_1 = 0.03	0.99848	0.910	1.3334	1516.4
w_1 = 0.05	0.99956	0.916	1.3342	1519.3

[a] uncertainty in the density values: ±0.01 kg m^{-3}

[b] uncertainty in the viscosity values: ±0.003 mPas

[c] uncertainty in the refractive index values: ±0.0002 units

[d] uncertainty in the speed of sound: ± 0.2 $m.s^{-1}$

Table 2. Experimental values of Molality (m), Density (ρ), Viscosity (η), Refractive Index (n_D) and Ultrasonic Speed (u) of Glycine, L-Alanine and L-Valine in different mass fraction of Barium Nitrate in the solvent mixture [$Ba(NO_3)_2$+ Water] at 298.15K and Experimental Pressure 0.1MPa.

[e] m (mol kg^{-1})	[f] ρ x 10^{-3} (kg m^{-3})	[g] η (mPas)	[h] n_D	[i] u (m s^{-1})	[e] m (mol kg^{-1})	[f] ρ x 10^{-3} (kg m^{-3})	[g] η (mPas)	[h] n_D	[i] u (m s^{-1})
w_1 = 0.01					w_1 = 0.03				
Glycine					Glycine				
0.1001	0.99763	0.907	1.3328	1517.3	0.1001	0.99867	0.914	1.3339	1521.8
0.1584	0.99806	0.910	1.3332	1531.8	0.1583	0.99902	0.917	1.3343	1537.8
0.2004	0.99851	0.913	1.3335	1551.9	0.2003	0.99941	0.920	1.3346	1560.9
0.2351	0.99898	0.915	1.3338	1578.6	0.2350	0.99984	0.923	1.3349	1589.9
0.2653	0.99947	0.918	1.3341	1609.4	0.2652	1.00028	0.926	1.3352	1625.1
0.2925	0.99998	0.920	1.3344	1647.6	0.2923	1.00075	0.929	1.3355	1668.1

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		L-Alanine					L-Alanine		
0.1002	0.99759	0.909	1.3330	1516.5	0.1001	0.99861	0.918	1.3340	1520.7
0.1584	0.99797	0.917	1.3334	1529.8	0.1584	0.99892	0.926	1.3344	1535.4
0.2005	0.99838	0.923	1.3337	1549.1	0.2004	0.99929	0.933	1.3347	1556.4
0.2352	0.99884	0.930	1.3340	1573.9	0.2351	0.99972	0.940	1.3350	1583.9
0.2655	0.99932	0.937	1.3344	1604.6	0.2654	1.00018	0.948	1.3354	1616.9
0.2927	0.99984	0.944	1.3346	1641.3	0.2925	1.00068	0.955	1.3356	1658.2
		L-Valine					L-Valine		
0.1002	0.99750	0.913	1.3332	1515.5	0.1001	0.99854	0.925	1.3341	1519.8
0.1585	0.99779	0.926	1.3337	1528.0	0.1584	0.99878	0.939	1.3347	1533.3
0.2006	0.99814	0.939	1.3341	1546.3	0.2005	0.99911	0.953	1.3351	1553.1
0.2354	0.99852	0.951	1.3343	1570.2	0.2353	0.99952	0.967	1.3356	1578.9
0.2658	0.99899	0.963	1.3348	1598.9	0.2656	0.99995	0.982	1.3361	1612.6

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0.2931	0.99947	0.976	1.3354	1633.6	0.2929	1.00044	0.996	1.3365	1650.2
w_1 = 0.05									
		Glycine							
0.1000	0.99974	0.920	1.335	1525.2					
0.1582	1.00007	0.924	1.3354	1542.9					
0.2002	1.00045	0.928	1.3357	1568.9					
0.2349	1.00086	0.931	1.3361	1601.3					
0.2651	1.00130	0.935	1.3364	1642.0					
0.2922	1.00176	0.938	1.3367	1690.2					
		L-Alanine							
0.1001	0.99967	0.925	1.3351	1524.3					
0.1583	0.99994	0.933	1.3355	1540.7					
0.2003	1.00031	0.941	1.3358	1565.1					

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0.2350	1.00074	0.949	1.3362	1596.3
0.2652	1.00122	0.957	1.3365	1634.9
0.2924	1.00173	0.965	1.3368	1683.2
		L-Valine		
0.1001	0.99960	0.932	1.3352	1523.5
0.1583	0.99983	0.948	1.3358	1539.5
0.2004	1.00020	0.963	1.3363	1563.1
0.2352	1.00064	0.978	1.3368	1594.2
0.2655	1.00116	0.995	1.3372	1634.1
0.2927	1.00175	1.010	1.3376	1680.3

[e] uncertainty in the molality: ±0.0001 mol kg^{-1}

[f] uncertainty in the density values: ±0.01 kg m^{-3}

[g] uncertainty in the viscosity values: ±0.003 mPas

[h] uncertainty in the refractive index values: ±0.0002 units

[i] uncertainty in the speed of sound: ± 0.2 $m.s^{-1}$

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Table 3. Molality (m), apparent molar volume (ϕ_V),$(\eta/\eta_0-1)/m^{1/2}$, molar refraction (R), adiabatic compressibility (β) and apparent molal adiabatic compressibility (ϕ_K) of Glycine, L- Alanine and L-Valine in Barium Nitrate at 298.15K and Experimental Pressure 0.1MPa.

j m (mol kg^{-1})	ϕ_V x 10^6 (m^3 mol^{-1})	$(\eta/\eta_0-1)/m^{1/2}$ (kg$^{1/2}$ mol$^{-1/2}$)	R (m^3 mol^{-1})	β x 10^{10} (Pa^{-1})	ϕ_K x 10^{10} (m^3 mol^{-1} Pa^{-1})
w_1 = 0.01					
Glycine					
0.1001	49.1994	0.036	15.4698	4.3539	-2.6443
0.1584	47.5952	0.044	15.4800	4.2701	-4.2921
0.2004	46.6928	0.050	15.4857	4.1583	-5.4037
0.2351	45.9180	0.054	15.4910	4.0169	-6.4489
0.2653	45.1888	0.058	15.4961	3.8628	-7.2303
0.2925	44.4811	0.061	15.5008	3.6838	-8.0281
L-Alanine					
0.1002	67.2669	0.062	18.3697	4.3587	-2.0879
0.1584	65.2616	0.089	18.3827	4.2816	-3.7521
0.2005	64.0083	0.107	18.3902	4.1739	-4.9379
0.2352	62.5272	0.123	18.3967	4.0415	-5.9285
0.2655	61.3943	0.138	18.4078	3.8865	-6.8198
0.2927	60.1895	0.152	18.4082	3.7127	-7.6189
L-Valine					
0.1002	104.4246	0.100	24.1708	4.3649	-1.3100
0.1585	100.6146	0.153	24.1967	4.2925	-3.1623
0.2006	98.1582	0.192	24.2145	4.1901	-4.3847

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0.2354	96.4947	0.220	24.2184	4.0619	-5.4093
0.2658	94.2550	0.247	24.2399	3.9155	-6.2603
0.2931	92.6879	0.272	24.2676	3.7492	-7.0469
		w_1 = 0.03			
		Glycine			
0.1001	56.1554	0.034	15.5000	4.3237	-2.8432
0.1583	53.5514	0.044	15.5115	4.2327	-4.6445
0.2003	51.8989	0.052	15.5180	4.1068	-5.9740
0.2350	50.4194	0.058	15.5240	3.9566	-7.0214
0.2652	49.4308	0.063	15.5297	3.7854	-7.9215
0.2923	48.4377	0.067	15.5350	3.5911	-8.7771
		L-Alanine			
0.1001	76.2058	0.069	18.4009	4.3302	-2.1035
0.1584	71.5988	0.096	18.4152	4.2464	-4.0186
0.2004	68.9448	0.115	18.4234	4.1311	-5.2920
0.2351	66.6458	0.131	18.4304	3.9871	-6.3949
0.2654	64.9029	0.146	18.4419	3.8243	-7.2973
0.2925	63.3039	0.159	18.4426	3.6343	-8.2015
		L-Valine			
0.1001	111.3192	0.127	24.2048	4.3357	-1.4067
0.1584	105.3101	0.175	24.2384	4.2586	-3.3819
0.2005	101.5544	0.212	24.2567	4.1494	-4.6914
0.2353	98.3905	0.245	24.2795	4.0132	-5.7814
0.2656	96.2964	0.274	24.3019	3.8456	-6.8553
0.2929	94.2344	0.298	24.3162	3.6705	-7.6397
		w_1 = 0.05			
		Glycine			
0.1000	57.0951	0.036	15.5297	4.2999	-2.9682
0.1582	54.6941	0.047	15.5414	4.2004	-5.0326
0.2002	52.8433	0.055	15.5481	4.0608	-6.5580

0.2349	51.4563	0.062	15.5585	3.8965	-7.7019
0.2651	50.2350	0.069	15.5643	3.7041	-8.7591
0.2922	49.2093	0.074	15.5697	3.4942	-9.6492
		L-Alanine			
0.1001	78.1244	0.072	18.4363	4.3053	-2.3383
0.1583	73.9225	0.097	18.4513	4.2129	-4.4462
0.2003	70.3710	0.118	18.4594	4.0811	-5.9724
0.2350	67.6652	0.135	18.4714	3.9214	-7.1757
0.2652	65.4045	0.151	18.4775	3.7366	-8.2244
0.2924	63.5886	0.165	18.4830	3.5235	-9.2374
		L-Valine			
0.1001	113.1998	0.132	24.2513	4.3101	-1.7034
0.1583	106.3968	0.184	24.2851	4.2201	-4.0238
0.2004	101.1945	0.221	24.3089	4.0920	-5.5660
0.2352	97.5566	0.254	24.3309	3.9322	-6.8507
0.2655	94.3344	0.288	24.3445	3.7405	-8.0430
0.2927	91.4255	0.313	24.3563	3.5356	-8.9733

[j] uncertainty in the molality: ±0.0001 $mol.kg^{-1}$

Table 4. Limiting apparent molar volumes (ϕ_V^0), experimental slopes (S_V^*), *A*, *B* coefficients, limiting partial adiabatic compressibility (ϕ_K^0), and experimental slope (S_K^*) of Glycine, L-Alanine, and L-Valine in aqueous Barium Nitrate at 298.15K and Experimental Pressure 0.1MPa.

Salt	ϕ_V^0 x 10^6 (m^3 mol^{-1})	S_V^* x 10^6 (m^3 $mol^{-3/2}$ $kg^{1/2}$)	*A* (kg mol^{-1})	*B* ($kg^{1/2}$ $mol^{-1/2}$)	ϕ_K^0 x 10^{10} (m^3 mol^{-1} Pa^{-1})	S_K^* x 10^4 (m^3 $mol^{-3/2}$ Pa^{-1} $kg^{1/2}$)
			w_1 = 0.01			
Glycine	51.53	-24.01	0.023	0.131	0.151	-27.92
L-Alanine	71.05	-36.48	0.015	0.463	0.795	-28.68
L-Valine	110.30	-60.17	0.012	0.887	1.591	-29.61
			w_1 = 0.03			
Glycine	60.01	-40.08	0.011	0.181	0.229	-30.82
L-Alanine	82.47	-66.5	0.008	0.477	1.012	-31.46
L-Valine	119.60	-88.43	0.007	0.920	1.796	-32.35
			w_1 = 0.005			
Glycine	61.19	-41.27	0.009	0.197	0.478	-34.82
L-Alanine	85.84	-76.71	0.007	0.478	1.215	-35.72
L-Valine	124.30	-113.10	0.006	0.931	2.012	-37.72

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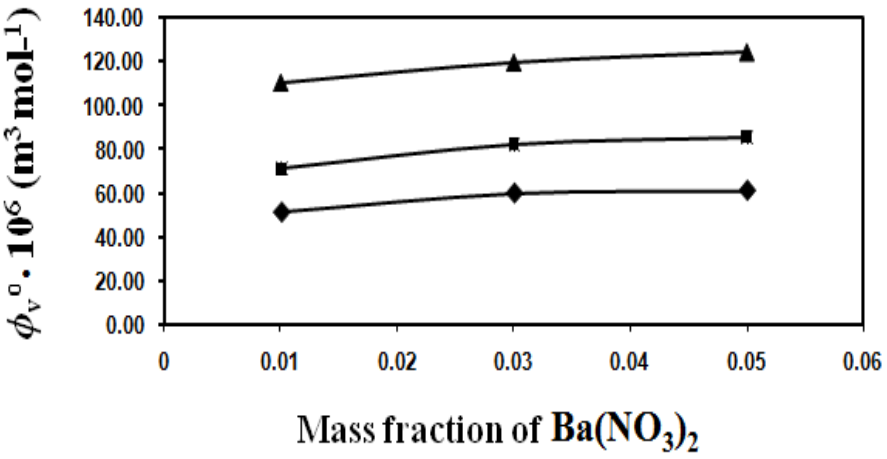

Figure 1. Plots of limiting apparent molar volumes (ϕ_V^0) for Glycine (—♦—), L-Alaline (—■—), L-Valine (—▲—) in different mass fractions (w_1) of $Ba(NO_3)_2$ in aqueous mixture at 298.15K.

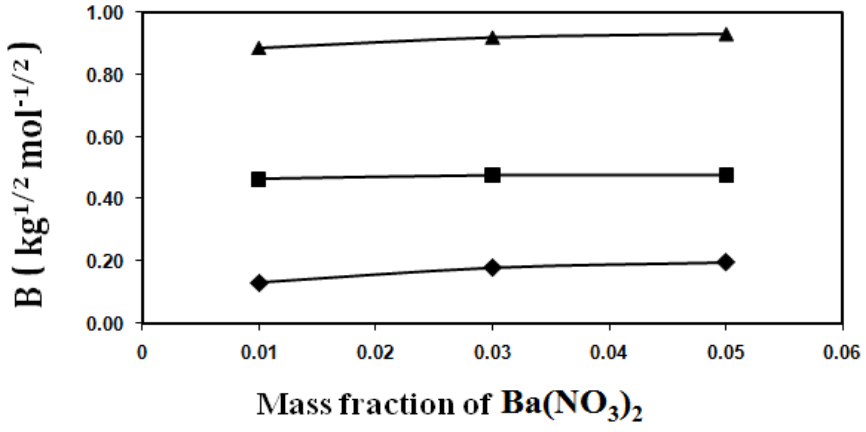

Figure 2. Plots of viscosity *B*-coefficient for Glycine (—♦—), L-Alaline (—■—), L-Valine (—▲—) in different mass fractions (w_1) of $Ba(NO_3)_2$ in aqueous mixture at 298.15K.

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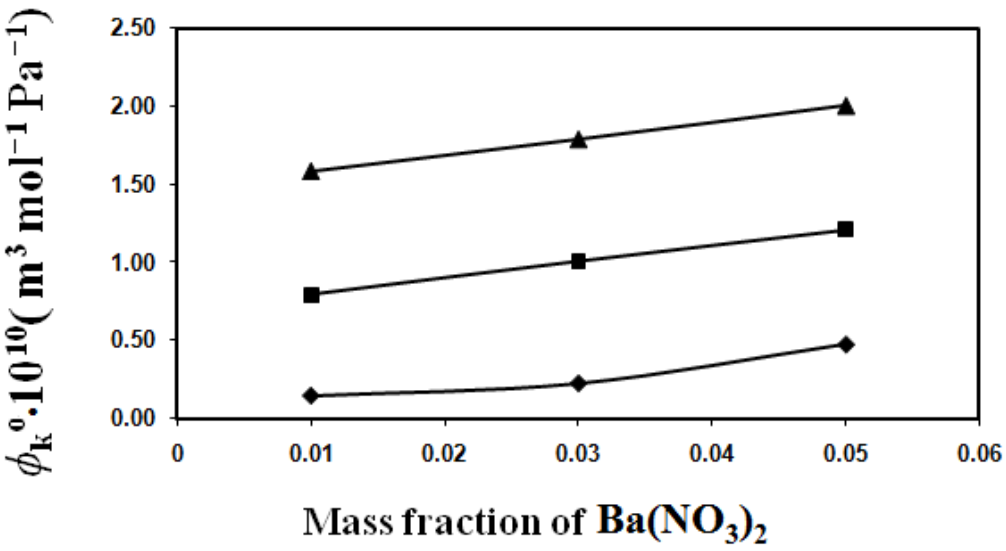

Figure 3. Plots of limiting partial adiabatic compressibility (ϕ_K^0) for Glycine (—♦—), L-Alanine (—■—), L-Valine (—▲—) in different mass fractions (w_1) of $Ba(NO_3)_2$ in aqueous mixture at 298.15K.