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Thermo-acoustic Investigations on L-valine in Aqueous Salt

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ABSTRACT

The ultrasonic velocity (u), density (ρ) and viscosity (η) of an amino acid L-Valine of various concentrations in 2% of NaCl solutions have been measured. From these experimentally measured parameters thermodynamic parameters such as adiabatic compressibility (β_{α}), intermolecular free length (L_f), acoustic impedance (Z), internal pressure (π_i), relaxation time (τ), Gibb's free energy (ΔG), free volume (V_f), Vander Waal's constant (b), Rao constant (R), Wada constant (W), molecular radius (r_o) absorption coefficient (a/f^2), relaxation strength (r), available volume (Va), volume expansivity (α), reduced volume (\tilde{V}) and enthalpy (ΔH) were calculated. It was found that there is certain degree of variation in these parameters with change in concentration and temperature. The variations of acoustic parameters with concentration and temperature indicate the existence of intermolecular interaction in the present systems.

Keywords: L-valine, Ultrasonic velocity, Thermodynamic parameters, Molecular interactions

INTRODUCTION

Proteins are the most abundant bio-molecules of the living system of all organic compounds. They are needed for the most important biological processes like cell growth and their maintenance, movement and defense. Proteins are complex molecules and their behavior in solutions is governed by a combination of many specific interactions [1,2]. These interactions are mainly those between the protein molecules and the solvent ions. Most of these interactions such as hydrogen bonding, electrostatic interactions, hydrophobic interaction etc. are non-covalent. The study of these interactions provides an important insight into the conformational stability and unfolding behavior of globular proteins.

Amino acid serves as basic building blocks of proteins and belongs to an important family of bio-molecules. The direct study of protein-electrolyte is very difficult as proteins are very large complex molecules. Therefore the study of interaction of some compound that constitutes the part of protein structure such as amino acids [3,4] is significant. Solution studies of amino acids with their surrounding environment play an important role in conformational characteristics of proteins [5]. Salts have large effects on the structure and properties of proteins [6]. Many researchers have reported the physico-chemical properties of amino acids [7-12] but very scanty work on solution of amino acid in aqueous and aqueous electrolyte solutions. Thus, it will be of great importance to examine the solution behavior of L-Valine amino acid in aqueous electrolytes. Moreover the physico-chemical study of liquid solutions finds direct application in chemical, biochemical and medical industry.

L-Valine is an essential non-polar amino acid. It is found in muscle tissue. It is a branched chain proteinogenic amino acid [13]. It helps to remove extra nitrogen from liver and also stimulate muscle growth. It also stimulates the central nervous system. It helps in repairing damaged tissues, promotes normal growth and provides energy to cells. It regulates blood sugar level. It is non-polar in nature and found inside a protein structure to protect it from water. Deficiency of L-Valine causes degenerative nerve diseases [14]. The molecular structure of L-Valine [15] is as shown in Figure 1.



Figure 1: Molecular structure of L-valine

The ultrasonic velocity is highly sensitive to structure and a molecular interaction in liquid mixture as it is fundamentally related to the binding forces between the components of liquid mixtures. The measurement of ultrasonic velocity of low amplitude in liquid solution enables us to study the properties of liquid solutions with ease and without much complicated instrumentation. Therefore the ultrasonic technique has nowadays become an important tool for examining physico-chemical properties of the liquid solutions and is preferred among various techniques such as dielectric relaxation, infrared spectroscopy, nuclear magnetic resonance etc.

MATERIALS AND METHODS

The compound L-Valine CAS No. 72-18-4, molecular weight 117.146 was obtained from HIMEDIA India Ltd. The purity of compound is 99%. It was used as supplied. Initially 2% aqueous NaCl stock solution was prepared by using double distilled water. The digital balance having an accuracy of ± 0.1 mg was used for the measurement of weights. The fresh L-Valine solutions under the study of different concentration in the given solvent were prepared. The densities of aqueous solvent and solution of different concentration (0.02 M to 0.12 M) at different temperature range T=(288.15 to 303.15) K were measured by pycnometer method. During the measurement of densities temperature was kept constant by using digital constant temperature water bath having an accuracy of ± 0.1 K. Owing to the importance of Ostwald's viscometer for the measurement of viscosity, the viscosity of solvent and solution under the study was measured. The ultrasonic velocity of solvent and solution of different concentration at different temperature range (288.15 K to 303.15 K) was measured by using digital ultrasonic interferometer at frequency 2 MHz (VI Microsystems Pvt. Ltd. Perungudi, Chennai) with an accuracy of $\pm 0.1\%$. The temperature of solution was kept constant during each measurement.

Physical parameters

Using experimentally measured basic parameters such as density, viscosity and ultrasonic velocity various thermodynamic parameters are evaluated by using the following mathematical equations. Adiabatic compressibility (β):

$$\beta = \frac{1}{\alpha U^2}$$

$$\beta = \frac{1}{\rho U^2} \tag{1}$$

Where ρ and U are the density and ultrasonic velocity of solution. Intermolecular free length (L_f):

$$L_f = K \beta^{1/2} \tag{2}$$

Where, K is Jacobson's constant.

This constant is a temperature dependent parameter whose value at any temperature (T) is given by $(93.875+0.345T) \times 10^{-8}$. Acoustic impedance (Z):

$$Z=U\rho$$
 (3)

Internal pressure (π_i) :

$$\pi_{i} = bRT \left(\frac{K\eta}{U}\right)^{1/2} \left(\frac{p^{2/3}}{M_{eff}}\right)^{7/6}$$
(4)

Where, b stands for cubical packing, which is assumed to 2 for all the liquids;

K is a dimensionless constant independent of temperature;

T is the absolute temperature in Kelvin;

M_{eff} is the effective molecular weight;

R is gas constant.

Relaxation time
$$(\tau)$$

$$=\frac{4}{2}\eta\beta_a$$
 (5)

Gibbs free energy (ΔG)

$$\Delta G = -K_B T \ln\left(\frac{h}{\tau K_B T}\right)$$
(6)

Where 'h' is Planck's constant and K_B-is Boltzmann constant=1.3806 \times 10⁻²³ JK⁻¹

(9)

The free volume (V_f)

$$V_f = \frac{M_{eff} U}{K\eta} \tag{7}$$

Rao's constant (R)

 $R = V_m U^{1/3}$ (8)

Where, V_m is molar volume

Wada constant (W)

Molecular radius

Vander Waal's constant:

$$b = V_{\rm m} [1 - ({\rm RT}/{\rm M}_{\rm eff} {\rm U}^2) \{ \left[1 + ({\rm M}_{\rm eff} {\rm U}^2/3{\rm RT}) \right]^{\frac{1}{2}} - 1 \} \}$$
(10)

$$r_{o} = (3M_{eff}/16\pi\rho V_{m})^{1/3} \left[1 - \left(\gamma RgT/M_{eff}U^{2}\right) \left(\frac{M_{eff}U^{2}}{3\gamma RgT}\right) + 1\right) - 1\right)^{1/3}$$
(11)

Absorption coefficient

	$\left(\frac{\alpha}{f^2}\right) = \frac{8\eta\pi^2}{3\rho U^3}$	(12)
Available volume		
Relaxation strength	$V_a = \frac{f}{V_m}$	(13)
	$r=1-(\frac{U}{U_{m}})^{2}$	(14)
Volume expansivity	-	
	$\alpha = \frac{\pi i \beta i}{T}$	(15)
Reduced volume	1	
	$V = \{1 + [(\alpha T)/3(1 + \alpha T)]\}^3$	(16)
Enthalpy ΔH		
	$\Delta H = \pi_i \frac{V_m}{M_{eff}}$	(17)

 $W=\beta_a{}^{1/7}V_m$

RESULTS AND DISCUSSION

The experimentally measured values of solution of L-Valine in 2% aqueous solution of sodium salt of different concentration (0.02 to 0.12) mol dm⁻³ at different temperature range T=(288.15, 293.15, 298.15 and 303.15) K are as given in Table 1. For systematic study, the variations in density, viscosity and ultrasonic velocity of the solution under the study with concentration at different temperatures are plotted. These are as shown in Figure 2(a-c).

Table 1: Measured parameters of L-Valine in 2 % aqueous NaCl of different concentrations at temperature range T=(288.15, 293.15, 298.15and 303.15) K

	Temp. K	Value of parameters measured for concentrations (mol.dm ⁻³)							
Parameter		m=0	0.02	0.04	0.06	0.08	0.1	0.12	
ρ	288.15 K	1014.97	1014.11	1015.1	1016.32	1018.16	1018.77	1019.8	
(Kg m^{-3})	293.15 K	1013.06	1012.41	1013.28	1014.45	1016.56	1017.34	1018.81	
	298.15 K	1011.85	1011.38	1012.04	1013.48	1015.08	1015.93	1016.95	
	303.15 K	1010.28	1009.04	1010.49	1011.95	1013.38	1014.85	1015.59	
η	288.15 K	1.2666	1.2544	1.2778	1.3016	1.3151	1.3159	1.3284	
10^{-3}	293.15 K	1.0682	1.0572	1.0684	1.0799	1.0925	1.114	1.1259	
(Nsm ²)	298.15 K	0.9517	0.9317	0.9519	0.9532	0.9646	0.9753	0.9861	
	303.15 K	0.8564	0.8554	0.8661	0.8769	0.8877	0.889	0.8992	
U	288.15 K	1495.071	1498.473	1500.521	1503.26	1508.079	1510.846	1512.928	
(ms^{-1})	293.15 K	1504.414	1505.487	1507.565	1510.345	1515.235	1518.044	1520.157	
	298.15 K	1519.535	1515.345	1516.739	1520.235	1523.747	1527.293	1530.742	
	303.15 K	1529.876	1526.345	1527.755	1531.294	1534.849	1536.99	1539.136	



(c)

Figure 1(a-c): Variation in density, viscosity and ultrasonic velocity of L-Valine in 2% aqueous solution of sodium salt of different concentration (0.02 to 0.12) mol dm⁻³ at different temperature range T=(288.15, 293.15, 298.15 and 303.15) K

With rise in temperature, decrease in viscosity is observed. As temperature increases, molecules in the solution acquires more and more thermal energy due to which molecular motion increases at the expense of cohesive forces acting between the molecules. Thus the solution faces lesser resistance to flow which results in decrease in viscosity of the solution [16].

The increase in ultrasonic velocity Figure 2c with rise in concentration at different temperature in the present study confirms molecular association [17,18]. Breaking of number of hydrogen bond increases with rise in temperature of the solution under the study which results in formation of more and more number of monomeric water molecules. These molecules then enter in the cage-like water structure and get trapped to form closed packed structure. This closed-packed structure forms stiff material medium for the propagation of ultrasonic wave due to which ultrasonic velocity increases. Using the experimentally measured basic parameters such as density, viscosity and ultrasonic velocity, various thermodynamic parameters are evaluated and are as given in Table 2.

It is observed from the Figure 2(a) that the density of L-Valine solution in 2% aqueous NaCl increases with increase in concentration of solution; however it falls with the rise in temperature. Increase in density with rise in concentration is due to increase in number of solute particles in the solvent. Figure 2(b) shows the variation in viscosity of L-Valine solution in 2% aqueous electrolyte at different temperatures. The viscosity in the present investigation increases with concentration and decreases with rise in temperature. Increase in viscosity of solution with concentration indicates enhancement in molecular aggregation phenomenon.

The variations in these derived thermodynamic parameters with concentration at different temperatures are as shown in Figure 3 (a-q).

The decrease in free length Figure 3(b) with rise in concentration of L-valine solution in 2% aqueous salt solution suggest that there is a significant interaction between solute and solvent molecules. It also suggests the structure promoting behavior [19,20] as well as the presence of dipole-dipole and acceptor-donor interaction between solute and solvent molecules. With the rise in temperature of solution at a given concentration, increase in spacing between the molecule in the present investigation is observed. This increase in free length might be due to rise in thermal energy of the solution causing increasing molecular motion [21]. The variation of acoustic impedance with concentration at different temperature is as shown in Figure 3(c). The increase in acoustic impedance with the rise in concentration as well as temperature is due to change in elastic and inertial properties of solution. This indicates the greater association of solute and solvent molecules through hydrogen bonding [22].

Table 2: Derived thermodynamic parameters of L-Valine in 2% aqueous NaCl salt for different concentrations at temperatures T=(288.15, 293.15, 298.15, 303.15) K

Conc.	Temp. K	0	0.02	0.04	0.06	0.08	0.1	0.12
(moia m)								
$\beta a 10^{-10} (m^2 N^{-1})$	288.15 K	4.4078	4.3915	4.3753	4.3541	4.3185	4.3002	4.284
	293.15 K	4.3614	4.358	4.3423	4.3213	4.2846	4.2655	4.2475
10 (11 11)	298.15 K	4.2802	4.3059	4.2952	4.2694	4.243	4.2198	4.1966
	303.15 K	4.2291	4.2539	4.24	4.2143	4.1889	4.1/12	4.1565
T	288.15 K	4.0568	4.0493	4.0418	4.032	4.0155	4.007	3.9994
10^{-11} (m)	293.15 K	4.0703	4.0687	4.0614	4.0515	4.0343	4.0253	4.0168
	298.15 K	4.069	4.0812	4.0762	4.0639	4.0513	4.0402	4.0291
	303.15 K	4.0803	4.0922	4.0855	4.0731	4.0608	4.0522	4.0451
7	288.15 K	1.5175	1.5196	1.5232	1.5278	1.5355	1.5392	1.5429
$10^{6} (\text{Kg}^2 \text{s}^{-1})$	293.15 K	1.5241	1.5242	1.5276	1.5322	1.5403	1.5444	1.5488
10 (119 5)	298.15 K	1.5375	1.5320	1.555	1.5407	1.540/	1.5510	1.5567
	303.15 K	1.5456	1.5401	1.5438	1.5496	1.5554	1.5598	1.5631
_;	288.15 K	3.1	3.0729	3.0944	3.1158	3.1237	3.1161	3.1238
$10^9 (\text{Nm}^{-2})$	293.15 K	2.8836	2.8601	2.8684	2.877	2.8865	2.9071	2.9169
10 (1111)	298.15 K	2.7523	2.7199	2.7431	2.7384	2.7482	2.7555	2.7634
	303.15 K	2.643	2.6363	2.6482	2.6581	2.6679	2.6646	2.6734
	288.15 K	7.4437	7.345	7.4546	7.5566	7.5726	7.5448	7.5879
$\tau_{10^{-13}}$ (s)	293.15 K	6.2117	6.1432	6.1858	6.2223	6.2411	6.3353	6.3762
10 (5)	298.15 K	5.4313	5.3488	5.4514	5.4263	5.457	5.4871	5.5176
	303.15 K	4.8292	4.8516	4.8965	4.9274	4.9579	4.9441	4.9834
	288.15 K	5.9608	5.9077	5.9666	6.0207	6.0291	6.0145	6.0372
ΔG 10 ⁻²¹	293.15 K	5.4014	5.3566	5.3845	5.4083	5.4205	5.4812	5.5073
$(kJmol^{-1})$	298.15 K	5.0104	4.9474	5.0256	5.0066	5.0298	5.0525	5.0753
· · · ·	303.15 K	4.6722	4.6915	4.73	4.7564	4.7823	4.7706	4.8037
	288.15 K	1.1347	1.1585	1.1324	1.1077	1.099	1.1042	1.0941
$V_{\rm f}$ 10 ⁻⁸ (m ³ mol ⁻¹)	293.15 K	1.4788	1.5078	1.4916	1.4761	1.4619	1.4279	1.4122
10 (1111101)	298.15 K	1.785	1.8407	1.7899	1.7974	1.777	1.7591	1.741
	303.15 K	2.1124	2.1151	2.0847	2.0594	2.0349	2.0405	2.0158
	288.15 K	2.6958	2.7165	2.7286	2.742	2.7595	2.7747	2.7869
b 10 ⁻⁵ (m ³ mol ⁻¹)	293.15 K	2.6912	2.7049	2.7174	2.731	2.7478	2.7626	2.7736
10 (11 1101)	298.15 K	2.7007	2.6998	2.7109	2.7257	2.74	2.7565	2.7724
	303.15 K	2.6986	2.7016	2.7107	2.7254	2.7403	2.7512	2.7642
	288.15 K	1.8129	1.8174	1.8197	1.8217	1.823	1.8258	1.828
$R_{10^{-4}}$ (m ³ mol ⁻¹)	293.15 K	1.8151	1.8194	1.8219	1.824	1.8249	1.8275	1.8292
10 (11 1101)	298.15 K	1.8166	1.8206	1.8234	1.8251	1.8267	1.8292	1.8314
	303.15 K	1.8185	1.8235	1.8252	1.827	1.8287	1.8305	1.833
	288.15 K	3.9152	3.9281	3.9339	3.9395	3.9445	3.9521	3.9577
$W = 10^{-4} (m^3 mol^{-1})$	293.15 K	3.9285	3.939	3.9453	3.951	3.9552	3.9622	3.9664
10 (11 1101)	298.15 K	3.9438	3.9498	3.9563	3.9616	3.9664	3.9738	3.9805
	303.15 K	3.9567	3.9659	3.9697	3.975	3.9804	3.9846	3.9913
	288.15 K	1.3878	1.3913	1.3934	1.3957	1.3986	1.4012	1.4032
10^{-10} (m)	293.15 K	1.387	1.3893	1.3915	1.3938	1.3966	1.3991	1.401
io (iii)	298.15 K	1.3886	1.3885	1.3904	1.3929	1.3953	1.3981	1.4008
	303.15 K	1.3883	1.3888	1.3903	1.3928	1.3954	1.3972	1.3994
- 12	288.15 K	9.4835	9.8179	9.6656	9.7965	9.9125	9.9017	9.8473
$\frac{\alpha}{10^{-15}}$	293.15 K	8.1243	8.1421	8.0466	8.0911	8.1239	8.1221	8.2295
10	298.15 K	7.0137	7.0483	6.9604	7.0874	7.0386	7.0621	7.0846
	303.15 K	6.1606	6.2246	6.2679	6.32	6.3453	6.3698	6.3432
-	288.15 K	0.1585	0.1269	0.1229	0.1205	0.1173	0.1116	0.1083
I I	293.15 K	0.1432	0.1159	0.1147	0.1122	0.1089	0.1032	0.0998
	298.15 K	0.1265	0.0981	0.103	0.1014	0.0972	0.093	0.0888
	303.15 K	0.1138	0.0857	0.09	0.0883	0.084	0.0798	0.0772
V_a 10 ⁻⁶ (m ³ mol ⁻¹)	288.15 K	1.4887	1.183	1.1478	1.1257	1.0955	1.0411	1.011
	293.15 K	1.3412	1.0797	1.0703	1.0479	1.0172	0.9615	0.9307
,,	270.13 K 303 15 V	1.179	0.91	0.9397	0.9431	0.9038	0.0002	0.0200
ļ	303.13 K	1.0399	0.7943	0.0309	0.0213	0.7014	0.7414	0.7175
	288.15 K	0.00725	0.00711	0.00702	0.00705	0.00706	0.00702	0.00698

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$\alpha(K^{-1})$	293.15 K	0.00664	0.00644	0.00638	0.00637	0.00636	0.00633	0.00634
	298.15 K	0.00611	0.00593	0.00589	0.00593	0.00588	0.00587	0.00585
	303.15 K	0.00569	0.00553	0.00555	0.00556	0.00554	0.00553	0.0055
V	288.15 K	1.0901	1.0702	1.0678	1.0663	1.0644	1.061	1.059
$(m^3 mol^{-1})$	293.15 K	1.0804	1.0635	1.0628	1.0613	1.0594	1.0559	1.054
	298.15 K	1.0699	1.053	1.0559	1.0549	1.0525	1.05	1.0476
	303.15 K	1.0623	1.0458	1.0483	1.0473	1.0449	1.0425	1.041
	288.15 K	2.9983	3.0542	3.0301	3.0484	3.0657	3.068	3.0587
ΔH 106	293.15 K	2.8482	2.8464	2.8251	2.8308	2.836	2.8395	2.8575
$(k \text{Imol}^{-1})$	298.15 K	2.7176	2.7201	2.6893	2.7105	2.7019	2.7074	2.7123
(303.15 K	2.6074	2.6161	2.6127	2.6207	2.6268	2.6327	2.6256

4.10E-011

4.09E-011











Figure 3(e)



Figure 3(b)







Figure 3(f)









Figure 3(d) shows the variation of internal pressure of L-Valine in 2% aqueous salt with concentration at different temperature. It is observed that internal pressure of the solution under the study increases with rise in concentration at a constant temperature and decreases with increase in temperature. The decrease in internal pressure with increase in temperature is due to the thermal agitation of molecules which reduces the interaction between the molecules in the system. Increase in internal pressure confirms the structure making tendency of solute and solvent and also the increase in solute solvent interactions through hydrogen bonding [23].

The intermolecular interactions can also be detected from physical parameter relaxation time. It is observed from Figure 3(e) that relaxation time increases slowly with increase in concentration but decreases with increase in temperature. Decrease in internal pressure is due to weakening of hydrogen bonds due to thermal motion [24].

The increase in Gibbs' free energy Figure 3(f) with concentration suggests greater association among components of the solution through intermolecular hydrogen bonding. Decrease in Gibbs' free energy with increase in temperature indicates that less time is required for rearrangement of molecules in the solution [25]. Free volume is defined as the average volume in which the center of the molecules can moves in uniform potential field inside the hypothetical cell due to repulsion of surrounding molecules. The decrease in free volume with rise in concentration and temperature Figure 3(g) suggests decrease in spacing between solute and solvent molecules through intermolecular hydrogen bonding.

Figure 3(h) and (i) shows the variations of Van der Waal's constant and molecular radius of solutions under the study with concentration and temperature. Van der Waal's constant and molecular radius depends on the size of associated complex formed between solute and solvent molecules. Greater size indicates more association. The increase in Van der Waal's constant and molecular radius values indicates greater association among the components of the mixture and enhanced solute solvent interactions.

As temperature increases, the Van der Waal's forces become weak. This makes rupture of water molecules of the last hydration shell easy and thus size of the complex gets reduced which results decrease in values of Van der Waal's constant and molecular radius with increase in temperature.

Rao constant and Wada constant are useful to study physiochemical behavior of liquid and liquid mixtures. The increasing trend of both constants Figure 3(j) and (k) with rise in concentration as well as with temperature suggests that in the present system more number of components are available in a given volume of the medium and the presence of the solute-solvent interactions [26-28]. This indicates greater molecular association and close packing of the medium.

Absorption coefficient is the characteristic of the liquid medium and depends upon the composition, temperature, pressure, and frequency. The increase in absorption coefficient (Figure 3(1)) observed in the present study with rise in concentration clearly indicates the intermolecular association through dipole-dipole interaction between solute and solvent. Absorption coefficient is found to decrease with increase in temperature. As temperature increases, the thermal motion of the particles increases. Hence less sound energy is needed to break such weak intermolecular forces which lead to less sound absorption. Relaxation strength directly correlates adiabatic compressibility. The decrease in values of relaxation strength with concentration at constant temperature is due to the formation of complex by solvent molecules around solute molecules which is clear indication of the greater association (Figure 3 (m)). Figure 3(n) and (p) shows the variation of available and reduced volume of the solution under study with concentration at constant temperature. These variation curves are supporting the facts indicated by the earlier physical parameters under the study. The decreasing trend of Volume expansivity (Figure 3(0)) with concentration shows the greater molecular association due to strong hydration of solute with solvent molecules. The variation in enthalpy of L-valine (Figure 3(q)) also shows the supporting nature.

CONCLUSIONS

Basic physical parameters such as ultrasonic velocity, density and viscosity of L-Valine solution in 2% aqueous salt of different concentration at different temperature were measured initially. Using these basic parameters various acoustical parameters were evaluated. The temperature effect on these parameters was discussed appropriately. These parameters were interpreted in connection with the molecular interactions. The addition of solute in solvent has structure making tendency through intermolecular hydrogen bonding.

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