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## Ultrasonic and Thermodynamic Interaction Studies of Glycine in Aqueous Maltose and Raffinose Solution at Constant Temperature



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### ABSTRACT

Density and Ultrasonic velocity of Glycine in aqueous D (+) maltose and D (+) raffinose (2.0, 4.0 and 6.0) mass % of saccharides solutions have been determined experimentally at 308 K. The results obtained from density and ultrasonic velocity measurement have been used to calculate the adiabatic compressibility  $K_s$ , Acoustic impedance  $Z$ , apparent molal volume ( $\phi_v$ ), apparent molal adiabatic compressibility, ( $\phi K_s$ ), and partial molal volume,  $\phi_0 v$  partial molal adiabatic compressibility, ( $\phi_0 K_s$ ) at infinite dilution and experimental slopes. The results are interpreted in terms of solute-co-solute and solute-solvent interactions in these systems. It has been observed that there exist strong solute-solvent and solute-solute interactions and complex formation between in these ternary systems. The properties of the glycine aqueous maltose and aqueous raffinose systems are discussed in terms of the structure making and hydrogen bonding effect.

## 1. INTRODUCTION

In continuation of our earlier work (1) on the study of interaction between amino acids and electrolytes in aqueous medium, we present in this paper, the study of interaction between glycine in aqueous maltose and raffinose 308 K. The hydration behavior of saccharides has been found to be related to number or configuration of hydroxyl group. Saccharides and their derivatives are very important chemicals in life processes, while amino acids are basic components of protein molecules. There has been an increased interest in the physicochemical properties of amino acids and carbohydrates in aqueous as well as aqueous electrolyte media to understand the role played by the biological molecules in living organism. Therefore studies on carbohydrates protein interactions are very important in immunology, biosynthesis, pharmacology and medicine (2-6). Amino acids have zwitterion and are the constituents of the most important class of biopolymers, i.e. Proteins. Disarrangement water and electrolyte balance in living systems cause a wide variety of health problems. Therefore, a knowledge of water-amino acid interaction the effects on several biological processes occurring in living organism. In recent years, a number of workers have utilized density and viscosity data to deduce the thermodynamic properties (relative viscosity, Jones-Dole coefficient and free energy of activation of viscous flow) for a number of mixtures solutions (7-9). Very recently, we have made systematic effort to investigate the ultrasonic and volumetric properties of glycerin and dextrose in  $\text{Na}_2\text{CO}_3$  and  $\text{NaHCO}_3$  buffer solutions and amino acids in concentrated electrolytic solution (10-12). The thermodynamic parameters were utilized to study of various interactions taking place in the solutions of these ternary systems were measured at 308 and 313K. It was found that solute-solvent interaction and complex formation between in these ternary systems. It was found that  $\text{NaCl}$  and  $\text{MgCl}_2$  increase the apparent molar volume and decrease the adiabatic compressibility of amino acids. This increase could be attributed to the interactions of the ions of the  $\text{NaCl}$  and  $\text{MgCl}_2$  electrolytes and zwitterion head group of amino acids, causing the transfer of hydrated water molecule to the bulk state. Structural interactions of ionic solutes with electrolytes solvent are important in many fields of chemistry and biochemistry.

In the present paper, we report densities,  $\rho$  and ultrasonic velocities,  $u$  of Glycine (0.05, 0.07, 0.10, 0.13, 0.16 and 0.19) m in aqueous D (+) maltose and D(+) raffinose (2.0,4.0 and 6.0 %) solutions have been determined experimentally at 308K. From these experimental data, a number of thermodynamic parameters namely, adiabatic compressibility  $K_s$ , Acoustic

impedance  $Z$ , apparent molal volume  $\phi_v$ , partial molal adiabatic compressibility,  $\phi K_s$ , and partial molal volume  $\phi_v^0$ , partial molal adiabatic compressibility,  $\phi K_s^0$  at infinite dilution and experimental slopes  $S_v$  and  $S_{K_s}$  respectively have been calculated. These parameters were utilized to study various interactions taking place in the solutions of glycine in aqueous maltose and raffinose at 308 K.

## 2 EXPERIMENTAL

### 2.1 Chemical and Preparation

Glycine (99.5% purity), D(+) maltose and D(+) raffinose (99.8% purity) were procured from Merck and S d Fine Ltd. They were used as such without further purification, after drying over calcium chloride in desiccators for more than 48 hours. The ultrasonic velocities and densities of glycine in aqueous saccharides solution at various concentrations as well as in double distilled de-ionized water were measured experimentally. Aqueous solutions of maltose and raffinose (2.0, 4.0 and 6.0 %) were prepared and these were used as solvents to prepare glycine solutions on mass basis covering the whole composition range. All the solutions were prepared by mass in dry box and were stored in special air-tight bottles and kept in dark to avoid photochemical degradation. The weighing was done on an Afcoset ER-120A electronic balance with an accuracy  $\pm 0.1$  mg.

### 2.2. Measurement of density and ultrasonic velocity

The densities were measured with a single capillary pycnometer (made of Borosil glass) of bulb capacity of  $8 \times 10^{-6}$  m<sup>3</sup>. The marks of the stems were calibrated using double distilled water at 308 K. The pycnometer was kept for about 30 minutes in a thermostatic water bath so that the thermal fluctuation in density was minimized. The ultrasonic velocities in solutions were measured using a single crystal variable path interferometer. The temperature of the test solutions was maintained at  $308 \pm 1 \times 10^{-2}$  K in an electronically controlled thermostatic water bath. The velocity and density data were found to be accurate within  $\pm 0.10\%$  and  $\pm 0.01\%$  respectively.

## 3. RESULTS AND DISCUSSION

The densities,  $\rho$  and ultrasonic velocities,  $u$  of Glycine (0.05, 0.07, 0.10, 0.13, 0.16 and 0.19) m in aqueous D(+) maltose and D(+) raffinose (2.0,4.0 and 6.0 %) solutions have been

determined experimentally at 308K are presented in Table 1. It is observed from Table 1 that densities and ultrasonic velocities for all the ternary systems increase with an increase in molalities of glycine. The values of  $\rho$  and  $u$  increase with an increase in concentration of amino acid in all the ternary systems under investigation, which appear to be due to hydrophobic properties of solutes i.e. H-bond forming. This may be attributed to the formation of clusters by the amino acids and strong intermolecular forces in the solute. The changes in structure of solvent or solution as a result of H- bond formation lead to decrease in intermolecular free length (13). Solute may occupy the interstitial spaces in solvent or get solvated forming new weaker bonds. It was suggested that what is experimentally observed for any system, reflects the compromise between the tendency for the ion and the peptide to interact with each other and inclination of the solutes to associate with the solvent (14-16). The variation of ultrasonic velocity with the concentration of glycine,  $(du/dc)$  can be shown to depend upon the concentration derivations of the density and adiabatic compressibility of the system investigated.

Thus in the relation:

$$du/dc = -u/2 \left[ \frac{1}{\rho} \left( \frac{d\rho}{dc} \right) + \left( \frac{1}{K_s} \right) \left( \frac{dK_s}{dc} \right) \right] \quad (1)$$

The quantity  $(1/\rho)(d\rho/dc)$  is negative while  $(1/K_s)(dK_s/dc)$  is positive and the net value is negative. This makes  $du/dc$  positive, showing that  $u$  increases with increase the concentration of the ternary systems.

### 3.1 Adiabatic Compressibility

The adiabatic compressibility of the glycine + water + D(+) maltose and glycine + water + D(+) raffinose was determined at 308K from the density and velocity data. The adiabatic compressibility was calculated by this relation

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

The per usual of Table 1 exhibits the values of the adiabatic compressibility,  $K_s$  are found to be decrease with increase the concentration of solute glycine in all the ternary systems. Such a decrease in adiabatic compressibility observed in the ternary systems (glycine + water + D(+) maltose and D(+) raffinose ) clearly confirms the conclusion drawn from the ultrasonic velocity and density data. It is well known fact that when a solute dissolves in a solvent.

Since the solvent molecules are oriented in ionic field i.e. electrostatic field of ions of glycine ( $\text{NH}_3^+$ ,  $\text{COO}^-$ ). The solvent molecules are more compactly packed in the primary solvation shells. This is the reason, why 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 phenomenon called 'Electrostriction'. Since the solvent molecules are compressed, they do not respond to a further application of pressure. So the solutions become harder to compress. This will lead to the decrease in compressibility values. This may further ascribed that such a decrease in adiabatic compressibility,  $K_s$  is attributed to the influence of the electrostatic field of the ions on the accompanying molecules. The increasing electrostrictive compression of solvent around the molecules results in a large decrease in compressibility of solutions. It may be inferred that the weakening of hydrogen bond strength formed by the solute and solvent molecules and maximum complex formation may also be the reason for decrease in compressibility. This is in accordance with the view proposed by Jones and Falkenhafen et al. (17 & 18).

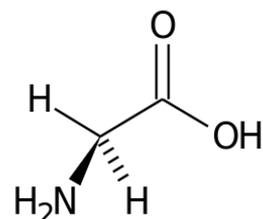
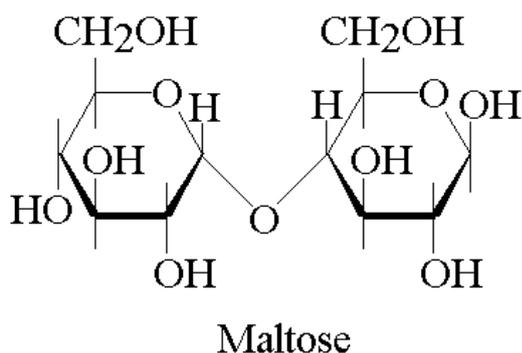
### 3.2 Acoustic impedance

The values of acoustic impedance,  $Z$  of these ternary systems (glycine + aqueous + maltose and glycine + aqueous +raffinose ) were calculated by the following relation and are reported in Table 1.

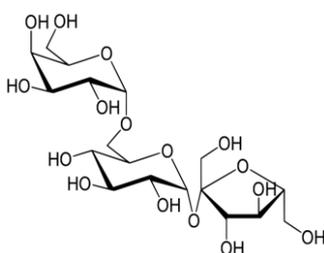
$$Z = u \cdot \rho \quad (3)$$

The values of  $Z$  increase with increase in molalities of glycine which indicates solute-solvent interaction are present and they behave as structure makers are shown in Table 1. The specific acoustic impedance of the medium for the particular type of wave being propagated and through it. The transmission of acoustic wave from one medium to another is important for transmission phenomenon in fluids. It may also correlate with ionic size  $\text{NH}_3^+$ ,  $\text{COO}^-$  and  $\text{OH}^-$  of ions. The behavior of acoustic impedance can be explained on the basis of lyophobic interaction between solute (glycine) and solvent (aqueous maltose and raffinose) molecules, which maximum complex ion formation and become responsible for the propagation of ultrasonic wave.

## Structures



Glycine



Raffinose  
HUMAN

### 3.3 Apparent molal volume

The apparent molal volume,  $\phi_v$  were calculated from measured density data of glycine in aqueous maltose and raffinose solution at different molalities at 308 K using the following equation:

$$\phi_v = [1000 (\rho^0 - \rho) / m \rho^0] + M / \rho \quad (4)$$

Where M is the molecular mass of the solutes,  $\rho^0$  and  $\rho$  density of solution. In these cases where molality dependence of  $\phi_v$ , having definite trend points, The  $\phi_v$  values increase due to reduction in the electrostriction effect at terminals, whereas it decreases due to disruption of side group hydration by that of the charged end.

The partial molal volume at infinite dilution  $\phi_v^0$  was calculated by taking an average data points. The linear variation is obtained by least square fitting to the following equation.

$$\Phi_v = \phi_v^0 + S_v m^{1/2} \quad (5)$$

The intercept which is the limiting apparent molal volume at infinite dilution  $\phi_v^0$  and the experimental slope,  $S_v$  which is considered to be volumetric pairwise interactions. The derived values  $\phi_v^0$  of along with  $S_v$  are summarized in Table 2. Table 2 shows that the values of  $\phi_v^0$  are positive of these ternary systems which indicate ion-solvent interactions are strong. The positive value of  $\phi_v^0$  with saccharides concentration of water molecules as a result of shielding of polar terminal groups ( $\text{NH}_3^+$  and  $\text{COO}^-$ ) of glycine molecules is due to increased interaction between the OH groups of saccharides. They have estimated the contraction of water around the oppositely charge group is caused by electrostatic ion-solvent interaction and is called electrostriction. A mutual comparison of these saccharides shows the values of  $\phi_v^0$  are larger in case of maltose than raffinose. Due to the interaction of  $\text{OH}^-$  ions with glycine of  $\text{NH}_3^+$  are strong and hydration of maltose (Solute - solvent interaction) will be much more than that of raffinose which is in good agreement with the results reported in Glycine– saccharides- water mixture (19 & 20). The  $S_v$  values (Table -2) of maltose and raffinose ternary systems are found to be positive. The positive values indicates ion–ion interactions are strong but in case of maltose systems, ion-ion interactions are strong than the raffinose system. Due to the  $S_v$  values (Table -2) of raffinose systems are found to be lesser than the maltose system.



The types of interactions occurring between the charge centre of glycine and saccharides can be classified as follows:

- i. Hydrophilic ionic interactions between the  $-\text{OH}$  group of saccharides and zwitterions of glycine  $-\text{NH}_3^+$  and  $\text{COO}^-$ .
- ii. The terminal groups of zwitterions of amino acid  $-\text{NH}_3^+$  and  $\text{COO}^-$  are hydrated in electrostatic manner.
- iii. The electrostriction of  $\text{NH}_3^+$  group is greater than the  $\text{COO}^-$  by a factor of 10.
- iv. The overlap of hydration co spheres of terminal groups ( $\text{NH}_3^+$  and  $\text{COO}^-$ ) and of adjacent groups results in volume change.
- v. The hydrophobic –hydrophilic interactions between the  $-\text{OH}$  group of saccharides molecules and non-polar group of the amino acid.

### 3.4 Apparent molal adiabatic compressibility

The density and adiabatic compressibility values were employed for calculated apparent molal adiabatic compressibility,  $\phi K_s$  of glycine in aqueous maltose and raffinose solution at different molalities at 308 K using the following equation:

$$\phi K_s = [1000 (\rho^0 K_s - \rho K_s^0) / m \rho^0] + M K_s / \rho \quad (6)$$

The less negative values of  $\phi K_s$  over the entire range of molality of the ternary systems which indicates due to loss of structural compressibility of solvent on increase in the population of hydrogen bonded solvent molecule in the vicinity of the solute molecules.

The partial molal adiabatic compressibility,  $\phi^0 K_s$  at infinite dilution are obtained from the plot of  $\phi K_s$  versus  $m^{1/2}$  by least square method. The molality dependence of this parameter has been expressed in terms of the following equation

$$\phi_{K_s} = \phi_{K_s}^0 + S_{K_s} m^{1/2} \quad (7)$$

Where  $\phi_{K_s}^0$  is the partial molar adiabatic compressibility at infinite dilution. It provides information regarding solute-solvent interaction and  $S_{K_s}$  is the experimental slope ion-ion interactions. The values of  $\phi_{K_s}^0$  and  $S_{K_s}$  are also included in Table 2. The magnitude of negative values of  $\phi_{K_s}^0$  is higher in maltose systems suggesting that the presence of strong solute-solvent. The hydration of maltose (solute - solvent interaction) will be much more than that of raffinose. So it indicates that the interaction between the maltose (OH<sup>-</sup>) and charged end group (NH<sub>3</sub><sup>+</sup> and COO<sup>-</sup>) of glycine are much stronger than that of raffinose ternary systems. The values of  $S_{K_s}$  glycine + water+ maltose are positive which is indicating the presence of ion-pair interactions is strong. The positive and higher values of  $S_{K_s}$  in case of maltose systems indicates ion-ion interactions are strong than the raffinose systems. Due to the  $S_{K_s}$  values (Table -2) of raffinose systems are found to be lesser than the maltose systems. This suggests glycine in aqueous maltose solution is strong structure maker than glycine in aqueous raffinose solution. From this tabulation, one can notice that the values of  $\phi_{K_s}^0$  in mixed ternary system less than the pure system suggesting the weak ion-solvent interactions in the ternary systems (21-23).

## CONCLUSION

The volume and compressibility data have been used to study of solute –solute and solute-solvent interaction in these ternary systems. It can be concluded that the existence of molecular interaction is in the order of maltose > raffinose. This suggests glycine in aqueous maltose solution is strong structure maker than glycine in aqueous raffinose solution. These parameters reflect that the solute-solvent interactions are predominant over the solute-solute interactions for the glycine in aqueous saccharides solutions.

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**Table -1**Densities ( $\rho$ ) and ultrasonic velocity ( $u$ ), acoustic impedance ( $Z$ ) and adiabatic compressibility ( $K_s$ ) of glycine + aqueous D(+) Maltose and glycine + aqueous D(+) Raffinose at 308 K

$m$ (mol.kg <sup>-1</sup> )	$\rho$ (10 <sup>-3</sup> kg m <sup>-3</sup> )	$u$ (m s <sup>-1</sup> )	$Z$ (10 <sup>-3</sup> Kg m <sup>-2</sup> s <sup>-1</sup> )	$K_s$ (10 <sup>-10</sup> m <sup>2</sup> N <sup>-1</sup> )
glycine + 2.00% D(+) Maltose				
0.00	1.001	1524	1525	4.295
0.05	1.002	1527	1530	4.281
0.07	1.003	1528	1532	4.270
0.10	1.004	1530	1536	4.254
0.13	1.006	1532	1538	4.232
0.16	1.007	1533	1541	4.224
0.19	1.008	1534	1546	4.215
glycine + 4.00% D(+) Maltose				
0.00	1.009	1531	1544	4.228
0.05	1.010	1533	1548	4.213
0.07	1.011	1535	1550	4.203
0.10	1.012	1535	1553	4.194
0.13	1.014	1538	1559	4.173
0.16	1.015	1540	1563	4.154
0.19	1.016	1541	1565	4.144
glycine + 6.00% D(+) Maltose				
0.00	1.016	1536	1561	4.172
0.05	1.017	1539	1565	4.151
0.07	1.018	1541	1568	4.136
0.10	1.019	1542	1571	4.127
0.13	1.021	1544	1576	4.108
0.16	1.022	1545	1578	4.099
0.19	1.023	1547	1582	4.085
glycine + 2.00% D(+) Raffinose				
0.00	1.0008	1524	1525	4.302
0.05	1.0021	1527	1530	4.279
0.07	1.0034	1528	1533	4.268
0.10	1.0040	1529	1535	4.260

0.13	1.0050	1531	1538	4.245
0.16	1.0060	1533	1542	4.229
0.19	1.0070	1535	1545	4.216
glycine + 4.00% D(+) Raffinose				
0.00	1.008	1529	1530	4.274
0.05	1.009	1532	1545	4.223
0.07	1.010	1533	1548	4.213
0.10	1.011	1535	1553	4.197
0.13	1.012	1536	1554	4.188
0.16	1.013	1538	1557	4.171
0.19	1.014	1539	1560	4.164
glycine + 6.00% D(+) Raffinose				
0.00	1.016	1534	1558	4.183
0.05	1.018	1538	1565	4.153
0.07	1.019	1539	1568	4.143
0.10	1.020	1540	1570	4.133
0.13	1.021	1541	1574	4.128
0.16	1.022	1543	1575	4.109
0.19	1.023	1545	1580	4.095

**Table -2 Limiting apparent molal volume ( $\Phi_v^0$ ), experimental slope ( $S_v$ ), Limiting apparent molal adiabatic compressibility ( $\Phi_{Ks}^0$ ) and experimental slope ( $S_{Ks}$ ) of glycine + aqueous D(+) maltose and glycine + aqueous D(+) raffinose at 308 K**

Mass %	$\Phi_v^0 \times 10^6$ ( $m^3 mol^{-1}$ )	$S_v \times 10^6$ ( $m^3 l^{1/2} mol^{-3/2}$ )	$-\Phi_{Ks}^0 \times 10^6$ ( $m^3 mol^{-1} GPa^{-1}$ )	$S_{Ks} \times 10^6$ ( $m^3 mol^{-1} GPa^{-1}$ )
glycine + aqueous + D(+) Maltose				
2.00	44.23	1.32	-13.05	36.34
4.00	43.01	13.89	-19.98	9.90
6.00	45.85	1.93	-37.52	97.19
glycine + aqueous + D(+) Raffinose				
2.00	43.81	1.61	-24.04	10.98
4.00	43.35	3.68	-22.87	6.12
6.00	43.64	3.28	-22.34	6.03