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Volumetric and acoustic studies of glycine in aqueous solutions of sulphathiazole drug at T=(288.15 to 308.15) K

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The densities (ρ) and speeds of sound (u) for glycine in aqueous solutions of (0.01, 0.02, 0.03, and 0.04) mol·kg^{-1} sulphathiazole drug have been measured at T = (288.15, 293.15, 298.15, 303.15 and 308.15) K, using vibrating tube digital densimeter and sound analyser Anton–Paar Model DSA - 5000. The apparent molar properties like apparent molar volume (V_φ), apparent molar adiabatic compressibility ($K_{\varphi,s}$) and the apparent molar volume ($V_\varphi{}^0$) and adiabatic compressibility at infinite dilution ($K_{\varphi,s}{}^0$) of glycine have been determined in water as well as in ternary mixtures containing sulphathiazole drug at different concentrations and at different temperatures from experimental data of densities and speeds of sound under atmospheric pressure. These data were also used to calculate the transfer parameters. Transfer parameters have been explained from the point of view of concentration dependence of solute-solute and solute-solvent interactions. The limiting apparent molar expansibility (E_φ°) values for glycine in aqueous solutions of drug have been calculated. The calculated values of thermal expansion coefficient (α_2) have small and positive values. These results are explained on the basis of drug-amino acid - water interactions and hydrophobic –hydrophilic interactions.

Keywords: Glycine, Sulphathiazole, Apparent molar volume, Apparent molar adiabatic compressibility, Solute-solute and solute-solvent interactions

Various biological processes involve volume changes and hydration of molecules, and their complete understanding needs a proper idea of the state and the behaviour of the molecules in the medium[1,2]. Further, proteins are essential components of living organisms. Proteins stimulate various biological reactions. However, the direct study of interactions of protein in solution is too difficult because of their complex structure. Generally, the functioning of protein is monitored by studying their component molecules, amino acids[3,4]. The behaviour of biomolecules in mixtures is affected by many factors such as chemical structure, pH, surface charge distribution, and type of electrolyte present and its concentration. The effect of the presence of electrolytes in solutions of biochemicals is of interest in a number of separation processes, such as the reverse micellar extraction of amino acids and proteins which may not occur without the presence of an electrolyte[5,6]. Furthermore, electrolytes are known to influence the stability of biologically important molecules such as proteins[7,8]. In addition, amino acids are the building blocks of other biomolecules such as peptides and proteins. Thus, it is important to study their behaviour in aqueous systems containing electrolytes. The interactions of water with the functional groups of proteins play important factor in determining the conformational stability of proteins[9,10]. Non-bonding interactions such as hydrogen bonding, electrostatic and hydrophobic interactions are also important in the stabilization of native conformations of biological macromolecules. The study of the solvent effect on the properties of model compounds such as amino acids is quite helpful in understanding water-protein interactions in solutions[11].

Although viscosity and partial molar volume properties of amino acids in aqueous electrolyte solutions have been extensively studied by many research groups[12], Physico-chemical properties of drugs are of interest to know during action at the molecular level. The action of a drug must be regarded as the vital outcome of physicochemical interactions between the drug and functionally important molecules in the living organism. Most drugs are organic molecules with both solvophylic

and solvophobic groups due to which these molecules show specific as well as electrostatic interactions. Hence, knowledge of the physicochemical properties of the drugs plays an important role in understanding their physicochemical actions which are highly dependent on the solution behaviour. Volumetric data of drug can provide clues to the interaction occurring in cellular fluids. Amino acid exists as zwitterionic species in aqueous solutions, thus their thermodynamic properties in a variety of media can provide valuable information about the stability and denaturation of proteins[13].

The partial molar volume and the related volumetric parameters of drug compounds in dilute aqueous solutions at different temperatures have been investigated by several authors[2,14-17]. The drug–water molecular interaction and their temperature dependence are useful in the understanding of drug action. It is generally accepted that proteins stabilize because of hydrophobic effect[18], although there was a dispute reported by Makhatadze and Privalov[19] who predicts that binding model describes well but then Franks[20] strongly argued in the article "Protein stability" regarding involvement of hydrophobicity rather than binding which is responsible for solubilizing and denaturing effects. Although no definite principle has been laid down in predicting the effect of solvent on the structure and reactivity of solutes, but much progress has been achieved[21,22].

Sulphathiazole is an organosulfur compound that has been used as a short-acting sulfa drug. It once was a common oral and topical antimicrobial until less toxic alternatives were discovered. It is still occasionally used, sometimes in combination with sulfabenzamide and sulfacetamide, and in aquariums. Recently, the study of small peptides, the building block of proteins having significant biological and industrial importance, has attracted wide interest as their peptides contain more complex structure and more components of proteins than amino acids. Further, these peptides are widely used in many applications mainly, in the pharmaceutical, food and chemical industries. There have been some source investigators in aqueous saccharide solutions[23-26] but very few in aqueous drug solutions[27-31] probably due to complex nature of their interactions. Therefore, the systematic study of peptides can provide valuable information about their behavior in solutions and insight into the hydration of biological systems in presence of drug solutions. Thus, from practical and academic point of view, it is necessary to investigate the solvation behaviour with small peptides in drug solutions at different temperatures. Furthermore, such kind of studies can enlighten the interaction behaviour of drug in biomolecular mixed systems. For this purpose, in the present paper we report the density and speed of sound measurements of glycine in aqueous drug solutions at T= (288.15 to 308.15) K and at atmospheric pressure from which the values for infinite dilution of apparent molar volumes are calculated with the help of least-square method[32]. The limiting apparent molar volumes and limiting apparent molar adiabatic compressibilities have been calculated in order to discuss the types of interactions occurring in the present systems.

Materials and Methods

Glycine was procured from Hi Media, India minimum assay 99.0% and used after drying over silica gel in a vacuum desiccator at room temperature. Sulphathiazole from Sigma Chemicals Co. were dried for 24 h in a vacuum desiccator before use. The structure of this drug is given in Fig. 1.

All solutions were prepared using deionized doubly glass-distilled water (having specific conductance less than 10^{-6} S·cm^{-1} that have been freshly degassed by vacuum pump). Solutions of glycine in the concentration range of 0.05 to 0.30 mol·kg^{-1} were made by mass on the molality concentration scale with an accuracy of $\pm 1 \times 10^{-5}$. The weighing were done on an Afcoset electronic balance (India, Model ER-182A) with a precision of ± 0.01 mg. The uncertainties in the solution molalities were in the range $\pm 2 \times 10^{-5}$ mol·kg^{-1}. Densities and speeds of sound, of glycine in aqueous sulphathiazole solutions at different temperatures were measured simultaneously and automatically, using an Anton Paar (model DSA 5000) vibrating-tube densimeter. Both the speed of sound and the density are extremely sensitive to temperature and hence, it was controlled to $\pm 1 \times 10^{-2}$ K by a built-in solid state thermostat. The apparatus was also tested with the density of a known molality of aqueous NaCl using the data given by Pitzer et al.[33].The reproducibility of

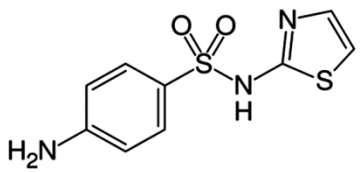

Fig. 1 — Structure of sulphathiazole drug

the instrument corresponded to a precision in density and speed of sound measurements of ± 1 × 10^{-3} kg·m^{-3} and 1 × 10^{-2} m·s^{-1}. The uncertainty of the density and speed of sound estimates was found to be less than ± 5 × 10^{-3} kg·m^{-3} and ± 5 × 10^{-2} m·s^{-1}. Before each series of measurements, the instrument was pre-calibrated with doubly distilled, deionized, degassed water, and dry air for the temperature range investigated. The absorption spectra of samples were recorded on Double beam UV-visible spectrophotometer.

Results and Discussion

Density, speed of sound and partial molar properties

The values of the density, and speed of sound, for glycine in aqueous solutions of sulphathiazole at T = (288.15, 293.15, 298.15, 303.15 and 308.15) K are reported in Table 1. The apparent molar

Table 1 — Densities (ρ) and speeds of sound (u) of glycine in aqueous solutions of sulphathiazole drug at different temperatures

m_A / (mol kg^{-1})	T (K)									
	288.15		293.15		298.15		303.15		308.15	
	ρ·10^{-3}/ (kg m^{-3})	u/ (m s^{-1})	ρ·10^{-3}/ (kg m^{-3})	u/ (m s^{-1})	ρ·10^{-3}/ (kg m^{-3})	u/ (m s^{-1})	ρ·10^{-3}/ (kg m^{-3})	u/ (m s^{-1})	ρ·10^{-3}/ (kg m^{-3})	u/ (m s^{-1})
	0.01 m_B sulphathiazole + glycine									
0.00000	1.000325	1469.92	0.999405	1486.13	0.998225	1499.61	0.996810	1511.66	0.995183	1521.47
0.05051	1.001954	1471.82	1.000999	1487.96	0.999801	1501.32	0.998370	1513.34	0.996725	1523.12
0.10054	1.003564	1473.93	1.002577	1489.99	1.001362	1503.39	0.999915	1515.36	0.998254	1525.08
0.15023	1.005160	1476.26	1.004144	1492.36	1.002911	1505.74	1.001450	1517.75	0.999774	1527.37
0.19879	1.006716	1478.71	1.005674	1494.98	1.004425	1508.44	1.002950	1520.39	1.001260	1529.96
0.25038	1.008366	1481.64	1.007298	1498.17	1.006032	1511.56	1.004543	1523.76	1.002841	1533.09
0.29747	1.009870	1484.55	1.008779	1501.36	1.007497	1514.70	1.005997	1526.89	1.004284	1536.31
	0.02 m_B sulphathiazole + glycine									
0.00000	1.001475	1470.95	1.000544	1486.78	0.999354	1500.61	0.997930	1512.41	0.996291	1523.07
0.05029	1.003078	1472.96	1.002112	1488.77	1.000898	1502.51	0.999454	1514.27	0.997784	1524.89
0.09996	1.004661	1475.15	1.003661	1491.02	1.002425	1504.65	1.000963	1516.39	0.999263	1526.96
0.14933	1.006233	1477.59	1.005202	1493.66	1.003945	1507.09	1.002468	1518.79	1.000739	1529.36
0.19938	1.007826	1480.23	1.006765	1496.75	1.005487	1509.83	1.003997	1521.54	1.002243	1532.09
0.25102	1.009468	1483.28	1.008378	1500.43	1.007081	1512.87	1.005581	1524.72	1.003798	1535.23
0.29572	1.010890	1486.09	1.009775	1503.85	1.008461	1515.93	1.006954	1528.02	1.005149	1538.29
	0.03 m_B sulphathiazole + glycine									
0.00000	1.003074	1471.32	1.002033	1487.38	1.000707	1501.37	0.999146	1513.51	0.997492	1523.96
0.04983	1.004575	1473.59	1.003500	1489.56	1.002156	1503.52	1.000571	1515.66	0.998905	1526.08
0.09992	1.006080	1476.07	1.004980	1491.94	1.003621	1505.87	1.002018	1517.99	1.000343	1528.39
0.14983	1.007574	1478.74	1.006458	1494.53	1.005089	1508.42	1.003476	1520.57	1.001793	1530.85
0.20001	1.009073	1481.53	1.007949	1497.36	1.006571	1511.28	1.004956	1523.39	1.003269	1533.54
0.24928	1.010540	1484.49	1.009415	1500.29	1.008034	1514.31	1.006427	1526.36	1.004736	1536.36
0.29986	1.012042	1487.77	1.010929	1503.53	1.009545	1517.59	1.007944	1529.79	1.006264	1539.67
	0.04 m_B sulphathiazole + glycine									
0.00000	1.004068	1472.75	1.003103	1488.63	1.001801	1502.49	1.000261	1514.55	0.998595	1524.89
0.05055	1.005538	1475.27	1.004537	1491.07	1.003199	1504.92	1.001646	1516.96	0.999965	1527.19
0.10055	1.006994	1477.99	1.005971	1493.68	1.004611	1507.45	1.003049	1519.61	1.001358	1529.66
0.14987	1.008435	1480.88	1.007399	1496.46	1.006029	1510.21	1.004461	1522.36	1.00277	1532.23
0.20139	1.009943	1484.09	1.008909	1499.61	1.007544	1513.2	1.005973	1525.39	1.004279	1534.98
0.24766	1.011301	1487.23	1.010283	1502.58	1.008931	1515.97	1.007355	1528.31	1.005668	1537.61
0.29883	1.012811	1490.76	1.011818	1505.99	1.010491	1519.09	1.008923	1531.55	1.007236	1540.76

volumes (V_φ) and the apparent molar adiabatic compressibilities ($K_{\varphi,s}$) of glycine in aqueous sulphathiazole solution were calculated from the experimentally measured densities and speeds of sound using the following equations and are given in Table 2.

$$V_\varphi = \left(\frac{M}{\rho}\right) - \{1000(\rho - \rho_o)m_A\rho\rho_o\} \quad \ldots (1)$$

$$K_{\varphi,s} = (M\beta_s/\rho) - \{1000\,(\beta_{s,o}\rho - \beta_s\rho_o)/m_A\rho\rho_o\} \quad \ldots (2)$$

where, M is the molar mass of glycine, and ρ, ρ_o, β_S and $\beta_{S,0}$ are the densities and coefficient of adiabatic compressibilities of solution and the solvent (drug+water), respectively and m_A is the molality of solute, that is, glycine in aqueous sulphathiazole. The coefficients of adiabatic compressibilities were calculated using the equation

$$\beta_s = (1/u^2\rho) \quad \ldots (3)$$

For the dilute solutions used in the present study, the variation of V_φ and $K_{\varphi,s}$ with molality can be represented by the following equation,

$$Y_\varphi = Y_\varphi^o + S_Q m_A \quad \ldots (4)$$

where Y_φ^0 (denotes V_φ^0 or $K_{\varphi,s}^0$) is the limiting value of partial molar property (equal to the infinite dilution

Table 2 — Apparent molar volume (V_φ) and apparent molar adiabatic compressibility ($K_{\varphi,s}$) of glycine in aqueous solutions of sulphathiazole drug at different temperatures

m_A / (mol kg^{-1})	T(K)									
	$V_\varphi \cdot 10^6$/ (m^3 mol^{-1})					$K_{\varphi,s} \cdot 10^6$/ (m^3 mol^{-1}GPa^{-1})				
	288.15	293.15	298.15	303.15	308.15	288.15	293.15	298.15	303.15	308.15
0.01 m_B sulphathiazole + glycine										
0.05051	42.74	43.45	43.82	44.16	44.54	-19.0	-16.8	-14.4	-13.2	-12.0
0.10054	42.71	43.39	43.75	44.09	44.45	-20.6	-18.2	-16.8	-15.4	-14.0
0.15023	42.67	43.32	43.69	44.02	44.37	-22.2	-20.3	-18.8	-17.7	-16.1
0.19879	42.64	43.27	43.63	43.95	44.29	-23.6	-22.3	-21.1	-19.8	-18.2
0.25038	42.61	43.21	43.57	43.88	44.21	-25.4	-25.4	-23.2	-22.4	-20.4
0.29747	42.57	43.16	43.52	43.82	44.14	-27.0	-27.0	-25.1	-24.3	-22.5
0.02 m_B sulphathiazole + glycine										
0.05029	43.11	43.81	44.31	44.72	45.37	-20.3	-18.5	-16.4	-14.7	-13.4
0.09996	43.04	43.74	44.22	44.62	45.26	-21.7	-20.4	-18.1	-16.6	-15.1
0.14933	42.98	43.66	44.13	44.51	45.14	-23.4	-22.8	-20.0	-18.5	-17.1
0.19938	42.92	43.59	44.05	44.40	45.00	-24.9	-25.4	-21.9	-20.5	-19.1
0.25102	42.87	43.51	43.95	44.28	44.88	-26.7	-28.2	-23.6	-22.6	-21.1
0.29572	42.81	43.44	43.88	44.18	44.77	-28.1	-30.4	-25.5	-25.0	-22.9
0.03 m_B sulphathiazole + glycine										
0.04983	44.83	45.53	45.91	46.42	46.69	-22.4	-19.7	-18.3	-17.1	-15.9
0.09992	44.81	45.41	45.76	46.21	46.45	-23.8	-21.1	-19.6	-18.4	-17.3
0.14983	44.79	45.30	45.61	45.98	46.21	-25.1	-22.5	-21.0	-20.0	-18.5
0.20001	44.76	45.19	45.47	45.77	45.96	-26.2	-24.1	-22.7	-21.6	-19.8
0.24928	44.74	45.09	45.33	45.54	45.72	-27.4	-25.4	-24.3	-23.1	-21.2
0.29986	44.71	44.97	45.18	45.34	45.46	-28.8	-26.8	-25.8	-24.9	-23.0
0.04 m_B sulphathiazole + glycine										
0.04983	45.85	46.58	47.31	47.60	47.93	-24.2	-21.6	-20.0	-18.8	-16.6
0.09992	45.77	46.36	46.95	47.20	47.49	-25.8	-23.1	-21.2	-20.7	-18.2
0.14983	45.66	46.15	46.63	46.84	47.04	-27.3	-24.6	-22.8	-22.2	-19.5
0.20001	45.56	45.92	46.25	46.43	46.60	-28.8	-26.3	-24.2	-23.6	-20.6
0.24928	45.47	45.70	45.92	46.09	46.21	-30.3	-27.7	-25.3	-25.0	-21.8
0.29986	45.35	45.46	45.56	45.68	45.78	-31.6	-29.0	-26.4	-26.2	-23.2

partial molar property) and S_Q (denotes Sv or S_K) is the experimental or limiting slope.

The V_φ and $K_{\varphi,s}$ data have been used to see the effect of temperature and drug concentration on solute-solvent interactions occurring in the ternary mixture of the present study. Hence, V_φ^0 and $K_{\varphi,s}^0$ at infinite dilution obtained by the least square fitting of V_φ and $K_{\varphi,s}$data using Eqn 4 are summarized in Table 3. At infinite dilution, the solute-solute interactions (that is, from the experimental slopes Sv or S_K) are negligible; therefore, the standard partial molar volume with temperature dependence gives valuable information of the solute-solvent interactions[34].

Table 3 reveals that glycine studied here has large positive V_φ^0 values and negative $K_{\varphi,s}^0$ values in aqueous sulphathiazole solutions at all investigated temperatures, which indicates the presence of strong amino acid-drug-water[24] interactions. The V_φ^0 values increase with increase in temperature and also with increase in the concentration of sulphathiazole. It indicates that solute-solvent interactions are increasing both with an increase in the concentration of sulphathiazole and temperature. Moreover, the $K_{\varphi,s}^0$ values are negative for glycine in aqueous sulphathiazole solutions. The negative $K_{\varphi,s}^0$ values indicate that water molecules around the solute are less compressible than water present in the bulk. Usually, partial molar volumes V_φ^0 and compressibilities $K_{\varphi,s}^0$ at infinite dilution are correlated in such way that the compressibility increases as the volume increases[25]. Further, it can be seen from Table 3, the values of $K_{\varphi,s}^0$ become more negative with increase in concentration of sulphathiazole and less negative with increase in temperature indicating the release of more water molecules from the secondary solvation layer of sulphathiazole into the bulk. This feature is similar to that observed for glycine in aqueous solutions of saccharides[24,32]. The experimental S_V values in Table 3 for glycine in sulphathiazole are found to be negative, suggesting that solute-solute interactions are weaker than solute-solvent interactions in the system under study.

Table 3 — Partial molar properties, V_φ^0 and $K_{\varphi,s}^\circ$ and their corresponding slopes, S_v and S_K of glycine in aqueous solutions of sulphathiazole drug at different temperatures

	T(K)				
	288.15	293.15	298.15	303.15	308.15
0.01 m_B sulphathiazole + glycine					
$V_\varphi^0 \cdot 10^6$ (m^3 mol^{-1})	42.77(±0.003)	43.50(±0.005)	43.87(±0.004)	44.35(±0.02)	44.61(±0.004)
$S_v \cdot 10^6$ (m^3 $kg^{1/2}mol^{-3/2}$)	-0.68(±0.02)	-1.17((±0.02)	-1.21(±0.02)	-1.38(±0.012)	-1.61(±0.02)
$K_{\varphi,s}^\circ \cdot 10^6$ (m^3 $mol^{-1}GPa^{-1}$)	-17.28(±0.10)	-14.14(±0.41)	-12.34(±0.12)	-10.88(±0.09)	-9.76(±0.06)
$S_K \cdot 10^6$ (kg m^3 $mol^{-2}GPa^{-1}$)	-32.31(±0.55)	-43.06(±2.15)	-43.28(±0.63)	-45.36(±0.51)	-42.58(±0.33)
0.02 m_B sulphathiazole + glycine					
$V_\varphi^0 \cdot 10^6$ (m^3 mol^{-1})	43.16(±0.003)	44.00(±0.004)	44.53(±0.004)	45.15(±0.02)	45.50(±0.006)
$S_v \cdot 10^6$ (m^3 $kg^{1/2}mol^{-3/2}$)	-1.19(±0.02)	-1.97((±0.02)	-1.75(±0.02)	-3.22(±0.08)	-2.47(±0.03)
$K_{\varphi,s}^\circ \cdot 10^6$ (m^3 $mol^{-1}GPa^{-1}$)	-18.59(±0.08)	-15.65(±0.25)	-14.48(±0.09)	-12.47(±0.21)	-11.31(±0.08)
$S_K \cdot 10^6$ (kg m^3 $mol^{-2}GPa^{-1}$)	-32.07(±0.42)	-34.32(±0.56)	-36.91(±0.51)	-41.53(±1.11)	-39.00(±0.43)
0.03 m_B sulphathiazole + glycine					
$V_\varphi^0 \cdot 10^6$ (m^3 mol^{-1})	44.85(±0.004)	45.63(±0.004)	46.06(±0.005)	46.63(±0.008)	46.94(±0.013)
$S_v \cdot 10^6$ (m^3 $kg^{1/2}mol^{-3/2}$)	-1.46(±0.02)	-2.21((±0.02)	-2.92(±0.02)	-4.35(±0.042)	-4.95(±0.07)
$K_{\varphi,s}^\circ \cdot 10^6$ (m^3 $mol^{-1}GPa^{-1}$)	-21.22(±0.08)	-19.28(±0.10)	-17.64(±0.08)	-14.96(±0.08)	-12.61(±0.08)
$S_K \cdot 10^6$ (kg m^3 $mol^{-2}GPa^{-1}$)	-25.10(±0.44)	-24.99(±0.53)	-27.22(±0.44)	-32.38(±0.43)	-34.71(±0.40)
0.04 m_B sulphathiazole + glycine					
$V_\varphi^0 \cdot 10^6$ (m^3 mol^{-1})	45.96(±0.009)	46.81(±0.009)	47.32(±0.009)	47.98(±0.009)	48.36(±0.01)
$S_v \cdot 10^6$ (m^3 $kg^{1/2}mol^{-3/2}$)	-2.02(±0.046)	-4.50((±0.05)	-7.04(±0.05)	-7.69(±0.04)	-8.66(±0.06)
$K_{\varphi,s}^\circ \cdot 10^6$ (m^3 $mol^{-1}GPa^{-1}$)	-22.75(±0.08)	-20.09(±0.10)	-18.70(±.0.16)	-17.58(±0.2)	-15.45(±0.13)
$S_K \cdot 10^6$ (kg m^3 $mol^{-2}GPa^{-1}$)	-29.98(±0.45)	-30.27(±0.52)	-26.35(±0.83)	-29.58(±1.05)	-25.89(±0.69)

V_φ^0 and $K_{\varphi,s}^\circ$ for glycine in water at the temperatures of (288.15, 293.15, 298.15, 303.15, and 308.15) K are taken from the references 28, 31 and 32

Partial molar properties of transfer

The transfer partial molar volumes ($\Delta_{tr}V_{\varphi}^{\circ}$) and transfer partial molar adiabatic compressibilities ($\Delta_{tr}K_{\varphi,s}^{\circ}$) at infinite dilution of glycine from water to aqueous solutions of sulphathiazole drug have been determined as

$$\Delta_{tr}Y_{\phi}^{\circ} = Y_{\phi}^{0} \text{ (in aqueous sulphathiazole solutions)} - Y_{\phi}^{0} \text{ (in water)} \quad \ldots (5)$$

The experimental values V_{φ}^{0} and $K_{\varphi,s}^{0}$ for glycine in water at T= (288.15, 293.15, 298.15, 303.15, and 308.15) K have been taken from literatures[32,35,36]. The calculated results are given in Table 4 and illustrated in Figs 2 and 3. The values of $\Delta_{tr}V_{\varphi}^{\circ}$ and ($\Delta_{tr}K_{\varphi,s}^{\circ}$) are by definition free from solute- solute interactions[37] and thereby provide qualitative and quantitative information regarding the interactions of a co-solvent and a solute as at infinite dilution the interactions between solute molecules become negligible. Table 4 and Figs 2 and 3 show that $\Delta_{tr}V_{\varphi}^{\circ}$ and $\Delta_{tr}K_{\varphi,s}^{\circ}$ are positive. From the data, it has been remarked that the $\Delta_{tr}V_{\varphi}^{\circ}$ values increase with elevation in temperature for all concentrations of drug. The positive $\Delta_{tr}V_{\varphi}^{\circ}$ value stands for the expansion in volume of glycine in moving from water to aqueous drug solutions [38,39].

The $\Delta_{tr}V_{\varphi}^{\circ}$ value can further be explained on the basis of co-sphere overlap model[40,41]. According to this model, it is suggested that, in the ternary solutions sulpha drug –glycine-water, the ionic-hydrophilic and hydrophilic-hydrophilic group interactions contribute positively, whereas the hydrophilic-hydrophobic group interactions contribute negatively to the $\Delta_{tr}V_{\varphi}^{\circ}$ values. It can also be seen in Table 4 and Fig. 2 that the transfer volume increases with increasing concentrations of drug. It may be concluded that in the ternary solutions, the increased concentrations of drug lead to greater ionic-hydrophilic and

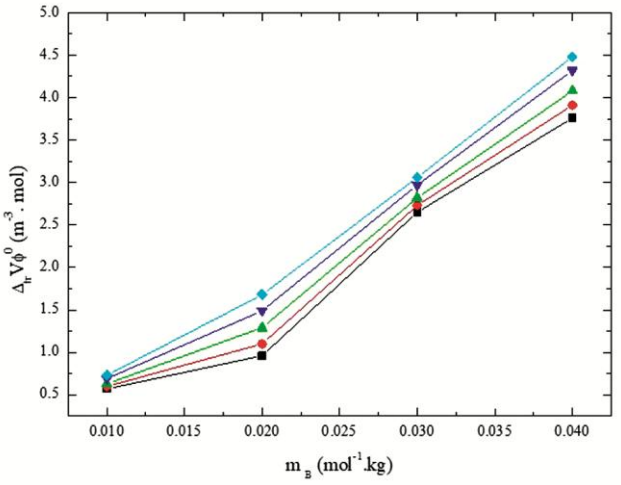

Fig. 2 — Plots of transfer partial molar volume, $\Delta_{tr}V_{\varphi}^{o}$ at infinite dilution versus molality of sulphathiazole drug at T= (■, 288.15 K; ●, 293.15 K; ▲, 298.15 K; ▼, 303.15 K; ○, 308.15 K)

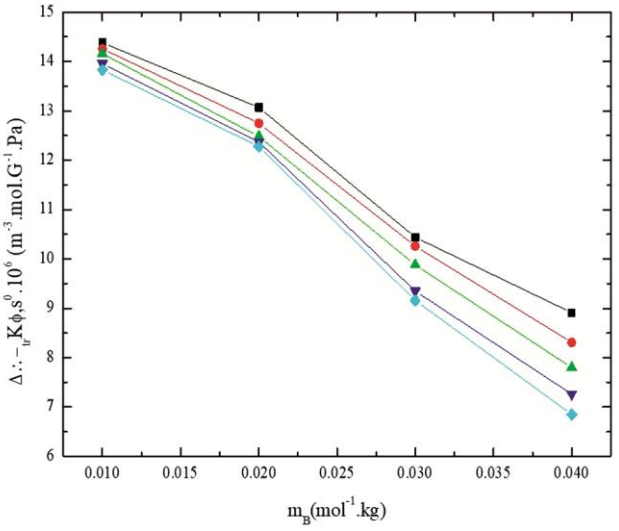

Fig. 3 — Plots of transfer partial molar adiabatic compressibility, $\Delta_{tr}K_{\varphi,s}^{o}$ at infinite dilution versus molality of sulphathiazole drug at T= (■, 288.15 K; ●, 293.15 K; ▲, 298.15 K; ▼, 303.15 K; ○, 308.15 K)

Table 4 — Transfer partial molar volumes, $\Delta_{tr}V_{\varphi}^{\circ}$ and transfer partial molar adiabatic compressibilities, $\Delta_{tr}K_{\varphi,s}^{\circ}$ of glycine in aqueous solutions of sulphathiazole drug at different temperatures

m_B (mol kg^{-1})	T(K)									
	$\Delta_{tr}V_{\varphi}^{\circ}\cdot10^{6}$ (m^{3} mol^{-1})					$\Delta_{tr}K_{\varphi,s}^{\circ}\cdot10^{6}$ (m^{3} mol^{-1}GPa^{-1})				
	288.15	293.15	298.15	303.15	308.15	288.15	293.15	298.15	303.15	308.15
0.01	0.57	0.60	0.63	0.69	0.73	14.38	14.26	14.16	13.96	13.83
0.02	0.96	1.10	1.29	1.49	1.68	13.07	12.75	12.48	12.37	12.28
0.03	2.65	2.73	2.82	2.97	3.06	10.44	10.26	9.88	9.35	9.16
0.04	3.76	3.91	4.08	4.32	4.48	8.91	8.31	7.80	7.26	6.85

m_B stands for the molality of solvent (i.e. aqueous sulphathiazole solution)

hydrophilic-hydrophilic interactions between -NH_3^+ / COO^- group of glycine and sulfuric( ) group of sulpha drug that are not influenced by the hydrophilic-hydrophobic interactions.

The $\Delta_{tr}K^{\circ}_{\varphi,s}$ values decrease both with increase in concentration of sulphathiazole drug and temperature. The observed increase in $\Delta_{tr}V^{\circ}_{\varphi}$ and decrease in $\Delta_{tr}K^{\circ}_{\varphi,s}$ values for glycine in aqueous drug solutions with an increase of temperature may be attributed to the corresponding decrease in the number of electrostricted water molecules and, thereby structure making tendency of the ions increases. That is, the release of water molecules to solvent bulk occurs due to disruption of hydration sphere of the charged end centers of glycine and sulphathiazole drug. As a result, it also leads to larger decrease in the compressibility with increase in sulphathiazole concentration. Thus, $K_{\phi,s}{}^{0}$ values are negative and $\Delta_{tr}K^{\circ}_{\varphi,s}$ values are positive.

The pair and triplet interaction coefficients estimated from $\Delta_{tr}V^{\circ}_{\varphi}$ and $\Delta_{tr}K^{\circ}_{\varphi,s}$ values as discussed in our previous paper[42] using the following equation

$$\Delta_{tr}Y^{\circ}_{\varphi}(water\ to\ aqueous\ cosolute\ solution) = 2Y_{AB}m_B + 3Y_{ABB}m_B^2 \quad \ldots (6)$$

where the constants Y_{AB} and Y_{ABB} are pairwise and triplet interaction coefficients. Here A denotes glycine, B denotes the co-solute (drug), and m_B is the molality of the co-solute. The $\Delta_{tr}Y^{\circ}_{\varphi}$ values have been fitted to Eqn 6 to obtain Y_{AB} and Y_{ABB}. The corresponding parameters V_{AB} and V_{ABB} for volumes and K_{AB} and K_{ABB} for adiabatic compressibilities, estimated from $\Delta_{tr}V^{\circ}_{\varphi}$ and $\Delta_{tr}K^{\circ}_{\varphi,s}$, respectively, are listed in Table 5.The pair wise interaction coefficients V_{AB} and V_{ABB} are positive for sulpha drug at all temperatures for glycine. Positive values for V_{AB} strengthen our viewpoint that ionic/ hydrophilic-hydrophilic interactions dominate over hydrophobic- ionic interactions between solute and co-solute molecules. The values of V_{AB} for glycine increase with increase in temperature. The pairwise interaction coefficient K_{AB} corresponding to the compressibility is also positive and it decreases with increase in temperature.

Apparent molar expansibilities

The temperature variation of V_{φ}^{0} can be expressed as

$$V_{\varphi}^{0} = a + b(T - T_m) + c(T - T_m)^2 \quad \ldots (7)$$

where T_m represents the midpoint temperature of the range used (T_m =298.15 K). Least- square fitting of Eqn 7 was done to obtain a, b, and c parameters.

Differentiation of Eqn 7 with respect to temperature at constant pressure was done to calculate partial molar isobaric expansions

$$E_2^o = (\partial V_{\varphi}^{0}/\partial T)_P = b + 2c(T - T_m) \quad \ldots (8)$$

It follows from Eqn 8 that the quantity $b+2c(T-T_m)$ is equivalent to E_2^o. The calculated values of partial molar expansion, E_2^o, at different temperatures are included in Table 6. From this table, it has been seen that at each temperature E_2^o value for any solute

Table 5 — Pair, Y_{AB} and triplet, Y_{ABB} interaction coefficients of glycine in aqueous solutions of sulphathiazole at different temperatures

Temperature (K)	$V_{AB}\cdot10^6$ ($m^3\ mol^{-2}\ kg$)	$V_{ABB}\cdot10^8$ ($m^3\ mol^{-3}\ kg^2$)	$K_{AB}\cdot10^8$ ($m^3\ mol^{-2}\ kg\ GPa^{-1}$)	$K_{ABB}\cdot10^9$ ($m^3\ mol^{-3}\ kg^2\ GPa^{-1}$)
288.15	14.35(±11.25)	-5.62(±2.18)	6.23(±1.55)	-8.88(±3.03)
293.15	17.26(± 9.44)	-5.42(±1.83)	6.19((±1.53)	-8.93((±2.97)
298.15	21.25(± 6.81)	-5.08(±1.32)	6.13(±1.53)	-8.93(±2.98)
303.15	25.21(±5.01)	-4.88(±0.97)	6.08(±1.53)	-8.96(±2.97)
308.15	30.18(± 2.46)	-4.36(±0.47)	6.06(±1.50)	-9.01(±2.92)

Table 6 — Partial molar expansions, E_2^0 at infinite dilution and isobaric thermal expansion coefficient, α_2 of glycine in aqueous sulphathiazole solutions at different temperatures

m_B (mol kg^{-1})	$E_2^0\cdot10^6$ ($m^3\ mol^{-1}\ K^{-1}$)				
	288.15 K	293.15 K	298.15 K	303.15 K	308.15 K
0.01	0.0002	0.0030	0.0045	0.0060	0.0075
0.02	0.0012	0.0037	0.0062	0.0087	0.0112
0.03	0.0135	0.0170	0.0205	0.0240	0.0275
0.04	0.0280	0.0320	0.0360	0.0400	0.0440
			α_2 (K)		
0.01	0.00003	0.00007	0.00010	0.00013	0.00017
0.02	0.00003	0.00008	0.00014	0.00019	0.00024
0.03	0.00030	0.00037	0.00044	0.00051	0.00058
0.04	0.00059	0.00067	0.00076	0.00083	0.00090

thought to be sensitive measure of solute-solvent interaction. From Table 6, it has been seen that at each temperature E_2^o value in aqueous drug solution are increasing regularly with rise in temperature, and with the concentration of sulphathiazole drug. It may be noted that E_2^o values are positive favouring the solute-solute interactions.

The effect is that electrostricted water may be released from the loose salvation layer of glycine. Removal of water molecules favours glycine-drug or drug-drug interactions, indicating the value of partial molar expansibility gives information regarding the size of the solute and its hydrophobicity.

The values of V_φ^0 and E_2^o are further used to calculate the isobaric thermal expansion coefficient, α_2 using following relation[43]

$$\alpha_2 = E_2^o / V_\varphi^o \quad \ldots (9)$$

α_2 also plays a vital role in interpretation of solute-solvent interactions[44].

The calculated values of α_2 are included in Table 6. The α_2 values increase with increase in temperature as well as with increase in concentration of sulphathiazole drug indicating that amino acid-drug- water interaction increases as concentration of sulphathiazole drug increases.

Spectral studies

Further as a part of our study, the absorption spectra were recorded for different mixtures to analyze the solute- solvent interactions. The spectra for different concentrations of glycine in different aqueous drug solutions are shown in Fig. 4 at 298.15 K. The values of observed absorption maximum are reported in Table 7. From the spectra, it is observed that absorption maximum increases with increase in

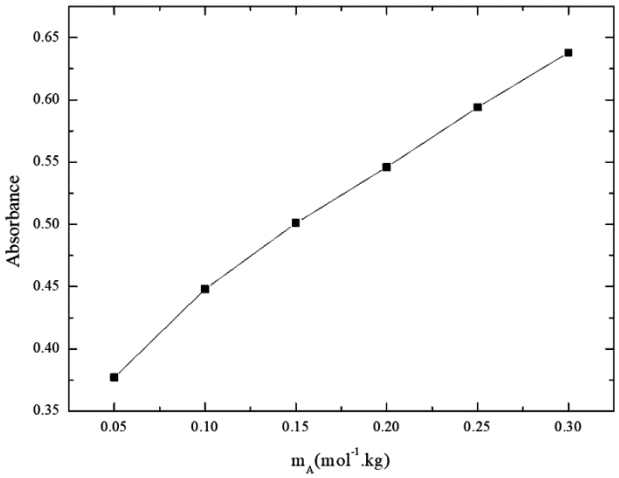

Fig. 4 — Absorption spectra (plot of molality of glycine versus absorbance) for 0.01m sulphathiazole drug solution at 298.15 K

Table 7 — Absorption and emission spectra of glycine with different concentrations of aqueous sulphathiazole drug solutions

m_A(mol kg^{-1})	Absorbance	Wavelength (nm)	$\log_{10}\varepsilon$	Intensity
0.01 $m_B$sulphathiazole + glycine				
0.05	0.377	318	0.8414	303.234
0.10	0.448	318	0.6884	371.399
0.15	0.501	318	0.5289	429.161
0.20	0.546	318	0.4594	445.997
0.25	0.594	318	0.3758	637.549
0.30	0.638	318	0.2996	815.611
0.02 $m_B$sulphathiazole + glycine				
0.05	0.234	322	0.6702	142.887
0.10	0.316	322	0.5263	257.269
0.15	0.383	322	0.4292	301.527
0.20	0.449	322	0.3512	346.218
0.25	0.516	322	0.3147	397.417
0.30	0.561	322	0.2718	438.780
0.03 $m_B$sulphathiazole + glycine				
0.05	0.135	324	0.3222	113.514
0.10	0.227	324	0.4579	201.704
0.15	0.304	324	0.3344	276.351
0.20	0.378	324	0.3096	420.322
0.25	0.457	324	0.2769	593.850
0.30	0.516	324	0.2355	855.510
0.04 $m_B$sulphathiazole + glycine				
0.05	0.070	327	0.1461	65.310
0.10	0.120	327	0.0790	266.640
0.15	0.185	327	0.0660	337.810
0.20	0.257	327	0.0910	546.346
0.25	0.329	327	0.1192	616.147
0.30	0.429	327	0.2209	667.128

concentration of glycine[8] and this feature is similar to that observed for apparent molar volumes. Further, the spectra recorded for a fixed composition of glycine in different aqueous drug solution shows regular decrease in absorption maximum with increase in concentration of sulphathiazole drug solution i.e from 0.01-0.04 mol kg^{-1}. The bathochromic shift observed in case of glycine with aqueous drug solution indicates the coordination of ions of sulphathiazole with glycine by breaking the solvent layers of water that causes extended conjugation resonance[45]. This effect is attributed to the corresponding increase in attraction between glycine and aqueous sulphathaizole solution. This clearly supports and justifies our thermodynamic data.

Conclusions

In this paper, we have presented the volumetric and adiabatic properties of glycine in aqueous sulphathiazole drug solutions at different temperatures. The apparent molar volume values are positive and apparent molar compressibility values are negative in aqueous drug solutions, indicating the presence of strong solute-solvent interactions. The observed positive values of transfer partial molar volumes and transfer partial molar adiabatic compressibilities in ternary solutions sulpha drug – glycine – water suggest that the ion-hydrophilic and hydrophilic – hydrophilic interactions predominate over the hydrophilic- hydrophobic group interactions. From the spectra, it is observed that absorption maximum increases with increase in concentration of glycine. This also supports and justifies our thermodynamic data.

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