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Physico-Chemical Studies of DL-Alanine in Aqueous Sodium Nitrate Solution[1]

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Abstract—In this study solubility of DL-alanine is measured in aqueous solutions with various $NaNO_3$ concentrations using the "formol titrimetry" method in the temperature range from 288.15 to 308.15 K. The standard transfer Gibbs energy and entropy have been evaluated for alanine. Chemical effects of the transfer Gibbs energy [$\Delta G^0_{i,\mathrm{ch}}(i)$], were obtained by subtracting theoretically computed $\Delta G^0_{\mathrm{t,cav}}(i)$ and $\Delta G^0_{\mathrm{t},d-d}(i)$ from the total transfer free energy, $\Delta G^0_{\mathrm{t}}(i)$. $T\Delta S^0_{i,\mathrm{ch}}(i)$ have been evaluated after elimination of cavity and dipole-dipole interaction effects from total transfer [$T\Delta S^0_{\mathrm{t}}(i)$] entropy. Various solvents and thermodynamic parameters are presented in this manuscript. The data accumulated demonstrated that solubility was affected by the electrolyte.

Keywords: DL-alanine, solubility, sodium nitrate, transfer energetics, hydrophobic interaction

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INTRODUCTION

Solubility data of amino acids in different solvents systems, such as aquo-organic, non-aqueous and aqueous electrolyte solutions may support the studies of solubility of other biomolecules.

Salts can modify the structure of proteins [1] by affecting their solubility, denaturation, folding and unfolding processes [2, 3], and activity of enzymes. For these reasons solubility and thermodynamic studies were carried out in different aqueous-organic [4–15] and non-aqueous solvent systems [9, 16]. In this regard Tanford, Nozaki and other authors [10, 11] reported solubilities, transfer free energies and entropies of some amino acids from water to urea, water-sodium sulphate [7], water to sodium chloride [25], water–glycerol [26], water–DMSO [27], and water–DMF [28] solvent systems.

The objective of this study was accumulation of new experimental data for solubility at various temperatures and thermodynamic parameters of DL-alanine in aqueous solution of sodium nitrate.

[1] The text was submitted by the authors in English.

RESULTS AND DISCUSSION

Calculation of total transfer Gibbs energy and entropy of solution from solubility. The important parameters of DL-alanine and aqueous solution of $NaNO_3$ are presented in Table 1. The solubilities of DL-alanine are listed in Table 2.

In the earlier studies [12, 17–19] of various types of biomolecules the standard Gibbs energies of solutions (ΔG^0_{S}) on molal scale were calculated for each solvent using Eq. (1). In the current study we also measured Gibbs energies of solutions by using the same equation as given below and presented in Table 3 [20].

$$\Delta G^0_{\mathrm{S}}(i) = -RT\ln C_\gamma = -RT\ln m. \quad (1)$$

The free energies, ΔG^0_{S} at different temperatures are fitted by the method of least squares in the following equation [11]:

$$\Delta G^0_{\mathrm{S}} = a + bT + cT\ln T. \quad (2)$$

The coefficients *a, b, c* (Table 4) reproduce the experimental data within ±0.04. Transfer Gibbs energies ΔG^0_{t} and entropies ΔS^0_{t} of amino acids from water to aqueous $NaNO_3$ mixtures were calculated at 298.15 K on mole fraction scale by using the Eqs. (3) and (4):

$$\Delta G^0_{\mathrm{t}}(i) = {}_{\mathrm{S}}\Delta G^0_{\mathrm{S}}(i) - {}_{\mathrm{R}}\Delta G^0_{\mathrm{S}}(i),$$

Table 1. Values of solvent parameters [hard sphere diameter of co-solvent σ_S, ($NaNO_3 + H_2O$), and $\sigma_{S-x} = 1/2(\sigma_S + \sigma_x)$], dipole moment of co-solvent (μ_S), and isobaric thermal expansibility constant α of the $H_2O + NaNO_3$ system at 298.15 K

Molality of $NaNO_3$	Mole fraction of $NaNO_3$ (z_S)	Mole fraction of water (z_R)	Molar mass M_S, kg/mol	Density $d_S \times 10^3$, kg/m^3	Molar volume V_S, dm^3/mol	σ_S, nm	σ_{S-x}, nm	Dipole moment μ_S	$\alpha \times 10^{-3}$
0.00	0.000	1.000	18.015	0.9970[a]	18.0692	0.274	0.445	1.831[a]	0.257[a]
0.25	0.009	0.991	18.618	1.0081	18.4684	0.275	0.446	1.831	0.257
0.50	0.018	0.982	19.219	1.0197	18.8477	0.276	0.446	1.831	0.257
1.00	0.036	0.964	20.426	1.0424	19.5952	0.278	0.447	1.831	0.257
2.00	0.072	0.928	22.837	1.0877	20.9957	0.282	0.449	1.831	0.257
3.00	0.107	0.893	25.181	1.1318	22.2486	0.287	0.452	1.831	0.257
5.00	0.179	0.821	30.004	1.2225	24.5432	0.296	0.456	1.831	0.257

[a] [22].

Table 2. Solubility, mol/kg, of DL-alanine in aqueous mixtures of $NaNO_3$ at different temperature

	Temperature, K				
Molality of $NaNO_3$	288.15	293.15	298.15	303.15	308.15
0.0 [Water]	1.680 (1.652) [17] (1.634) [15]	1.808 (1.715) [17] (1.720) [15]	1.921 (1.800) [17] (1.800) [15]	2.105 (2.100) [17] (2.300) [15]	2.405 (2.350) [17] (2.400) [15]
0.25	1.836 (±0.001)[a]	1.950 (±0.002)[a]	2.045 (±0.001)[a]	2.543 (±0.002)[a]	2.745 (±0.001)[a]
0.50	2.257 (±0.001)[a]	2.397 (±0.001)[a]	2.564 (±0.002)[a]	2.849 (±0.001)[a]	3.018 (±0.001)[a]
1.00	2.422 (±0.003)[a]	2.708 (±0.001)[a]	3.102 (± 0.002)[a]	3.360 (±0.002)[a]	3.624 (±0.001)[a]
2.00	2.846 (±0.001)[a]	3.204 (±0.002)[a]	3.928 (±0.001)[a]	4.102 (±0.003)[a]	4.204 (±0.002)[a]
3.00	3.684 (±0.001)[a]	3.820 (±0.003)[a]	4.360 (±0.002)[a]	4.460 (±0.001)[a]	4.743 (±0.003)[a]
5.00	4.122 (±0.003)[a]	4.732 (±0.001)[a]	5.044 (±0.001)[a]	5.222 (±0.003)[a]	5.389 (±0.001)[a]

[a] Standard deviation.

Table 3. Standard Gibbs energies of solutions (ΔG_S^0) on molal scale in their respective solubilities of DL-alanine in aqueous mixtures of $NaNO_3$ at different temperature

Temperature, K									
288.15		293.15		298.15		303.15		303.18	
S, mol/kg	ΔG_S^0, kJ/mol	S, mol/kg	ΔG_S^0, kJ/mol	S, mol/kg	ΔG_S^0, kJ/mol	S, mol/kg	ΔG_S^0, kJ/mol	S, mol/kg	ΔG_S^0, kJ/mol
1.680	–1.2428	1.808	–1.4433	1.921	–1.6182	2.105	–1.8759	2.405	–2.2482
1.836	–1.4555	1.950	–1.6276	2.045	–1.7733	2.543	–2.3523	2.745	–2.5870
2.257	–1.9501	2.397	–2.1306	2.564	–2.3339	2.849	–2.6387	3.018	–2.8299
2.422	–2.1192	2.708	–2.4280	3.102	–2.8061	3.360	–3.0545	3.624	–3.2987
2.846	–2.5056	3.204	–2.8379	3.928	–3.3913	4.102	–3.5574	4.204	–3.6790
3.684	–3.1239	3.820	–3.2665	4.360	–3.6499	4.460	–3.7683	4.743	–3.9881
4.122	–3.3930	4.732	–3.7883	5.044	–4.0112	5.222	–4.1659	5.389	–4.3152

Table 4. Thermodynamic parameters of transfer of DL-alanine on mole fraction scale from H_2O to aqueous mixtures of $NaNO_3$ at 298.15 K

Molality of $NaNO_3$	a, kJ/mol	b, kJ/mol	a, kJ mol^{-1} K^{-1}	$\Delta G_t^0(i)$, kJ/mol	$T\Delta S_t^0(i)$, kJ/mol
0.0 [Water]	–203.65	4.8166	–0.72646	0.0000	0.000
0.25	–267.76	6.3131	–0.95152	–0.3280	3.319
0.50	–51.26	1.3570	–0.21155	–0.8830	3.343
1.00	146.53	–3.0139	0.44109	–1.1330	3.357
2.00	425.76	–9.2892	1.37779	–2.2520	4.003
3.00	65.90	–1.3066	0.18843	–2.7530	–0.713
5.00	293.79	–6.4367	0.95442	–3.6204	–0.582

$$\Delta G_t^0(i) = (a_S - a_R) + (b_S - b_R)T + (c_S - c_R)T\ln T - RT\ln (M_S/M_R), \quad (3)$$

$$\Delta S_t^0(i) = (b_R - b_S) + (c_R - c_S)(1 + \ln T) + R\ln (M_S/M_R). \quad (4)$$

Here the subscripts "S" and "R" refer to the aqueous $NaNO_3$ mixtures and reference solvent (H_2O) respectively and M_R and M_S are the molar masses of the pure and mixed solvent respectively. $\Delta G_t^0(i)$ and $T\Delta S_t^0(i)$ values of α-amino acid were thus obtained (Table 4). The involved uncertainties in $\Delta G_t^0(i)$ and $\Delta S_t^0(i)$ are about ±0.05 kJ/mol and 2 J mol^{-1} K^{-1}, respectively.

Computation of cavity, transfer dipole–dipole interactions, enthalpy due to cavity formation, chemical parts of transfer Gibbs energy, and entropy**.** The term $\Delta P_t^0(i)$ [Eq. (5), where $P = G$ or S) may be ascribed as the sum of the following terms (assuming dipole induced dipole term to be negligibly small) [17].

$$\Delta P_t^0(i) = \Delta P_{t,cav}^0(i) + \Delta P_{t,d-d}^0(i) + \Delta P_{t,ch}^0(i). \quad (5)$$

Here, $\Delta P_{t,cav}^0(i)$ indicates the transfer energy contribution of the cavity effect which is involved due to creation of cavities for the species, DL-alanine in H_2O and aqueous $NaNO_3$ mixtures, and $\Delta P_{t,d-d}^0(i)$ stands for the dipole–dipole interaction effect involving interaction between dipolar-zwitter-ionic amino acid and the solvent molecules.

On the other hand, $\Delta P_{t,ch}^0(i)$ includes all other effects such as those arising from acid-base or short-range dispersion interaction, hydrophilic or hydrophobic hydration and structural effects, etc. $\Delta G_{t,cav}^0(i)$ can be computed using Scaled Particle Theory (SPT) [11, 17, 19].

The involved equations are given as follow:

$$\Delta G_{cav}^0(i) = G_C + RT\ln (RT/V_S), \quad (6)$$

$$G_C = RT[-\ln (1-z) + \{3x/(1-z)\}\sigma_x + \{3y/(1-z)\}\sigma^2{}_x + \{9x^2/4(1-z)^2\}\sigma^2{}_x],$$

$$z = \pi N_A/6V_S(z_R\sigma_R^3 + z_S\sigma_S^3),$$

$$x = \pi N_A/6V_S(z_R\sigma_R^2 + z_S\sigma_S^2),$$

$$y = \pi N_A/6V_S(z_R\sigma_R + z_S\sigma_S),$$

$$V_S = M_S/d_S.$$

In this expression N_A is Avogadro's number, z_R and z_S are the mole fraction of reference solvent water and co-solvent respectively. σ_x, σ_R, and σ_S are the hard sphere diameters of amino acid, water and aqueous sodium nitrate mixtures respectively. Where the terms M_S, d_S represent for molar mass and molar density of the solvent.

Therefore, the required $\Delta G_{t,cav}^0(i)$ represents the difference:

$$\Delta G_{t,cav}^0(i) = {}_S\Delta G_{cav}^0(i) - {}_R\Delta G_{cav}^0(i) = {}_SG_C - {}_RG_C + RT\ln (V_R/V_S), \quad (7)$$

$$\Delta G_{t,d-d}^0(i) = [{}_S\Delta G_{d-d}^0(i) - {}_R\Delta G_{d-d}^0(i)] \quad (8)$$

and $\Delta S_{t,d-d}^0(i) = [{}_S\Delta S_{d-d}^0(i) - {}_R\Delta S_{d-d}^0(i)]$ are calculated by means of the Keesom-orientation expression, [14, 21] for ${}_S\Delta G_{d-d}^0(i)$ in a solvent S, as given below:

$${}_S\Delta G_{d-d}^0(i) = -(8\Pi/9)N_A^2\mu_S^2\mu_x^2\sigma_{S-x}^{-3}(kT)^{-1}\nu_S^{-1} = A/TV_S, \quad (9)$$

$$A = -(8\Pi/9)N_A^2\mu_S^2\mu_x^2\sigma_{S-x}^{-3}(k)^{-1};\ V_S = M_S/d_S,$$

$${}_S\Delta S_{d-d}^0(i) = -\{\delta_S\Delta G_{d-d}^0(i)/\delta T\}_p, \quad (10)$$

$$T_S\Delta S_{d-d}^0(i) = {}_S\Delta G_{d-d}^0(i)[1 + T\alpha],$$

where N_A stands for Avogadro number, μ_S, μ_x are the dipole moment of aqueous $NaNO_3$ mixtures and amino

Table 5. Gibbs energies, enthalpy and entropy of transfer of DL-alanine from H_2O to aqueous mixtures of $NaNO_3$ at 298.15 K

Molality of $NaNO_3$	$\Delta G_t^0(i)$, kJ/mol	$\Delta G_{t,cav}^0(i)$, kJ/mol	$\Delta G_{t,d\text{-}d}^0(i)$, kJ/mol	$\Delta G_{t,ch}^0(i)$, kJ/mol	$T\Delta S_t^0(i)$, kJ/mol	$\Delta H_{t,cav}^0(i)$, kJ/mol	$T\Delta S_{t,cav}^0(i)$, kJ/mol	$T\Delta S_{t,d\text{-}d}^0(i)$, kJ/mol	$T\Delta S_{t,ch}^0(i)$, kJ/mol
0.0 [Water]	0.0000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.0000
0.25	–0.3280	–0.218	0.033	–0.143	3.319	–0.265	–0.047	0.035	3.331
0.50	–0.8830	–0.417	0.110	–0.576	3.343	–0.492	–0.075	0.119	3.299
1.0	–1.1330	–0.785	0.408	–0.756	3.357	–0.886	–0.101	0.439	3.019
2.0	–2.2520	–1.400	1.400	–2.252	4.003	–1.480	–0.080	1.510	2.573
3.0	–2.7530	–1.890	2.750	–3.613	–0.713	–1.880	0.010	2.960	–3.683
5.0	–3.6204	–2.650	5.880	–6.850	–0.582	–2.390	0.260	6.330	–7.118

acid respectively (Table 1). In this study we consider the dipole moment of aqueous Sodium nitrate solution is similar to the reference solvent (H_2O). σ_{S-x} Is the distance at which the attractive and repulsive interactions between the solvent and solute molecules are equal and can be expressed as $1/2(\sigma_S + \sigma_x)$, where σ_S and σ_x are the hard sphere diameters of cosolvent and solute molecules respectively (Table 1). α Is the isobaric thermal expansibility constant of the solvent:

$$\alpha = (\delta \ln V_S/\delta T)_p = -(\delta \ln d_S/\delta T)_p. \quad (11)$$

The enthalpy change due cavity forming interaction in water to aqueous $NaNO_3$ mixtures is measured by the equations:

$$\Delta H_{t,cav}^0(i) = {}_S\Delta H_{cav}^0(i) - {}_R\Delta H_{cav}^0(i), \quad (12)$$

$$\Delta H_{cav}^0(i) = (A + H + K + E)B, \quad (13)$$

$$A = (\Pi N_A/6V_S)(z_R\sigma_R^3 + z_S\sigma_S^3);\ B = \sigma_S RT^2/(1 - A);$$

$$H = \sigma_x 3y/(1 - A);\ K = \sigma_x 3x/(1 - A);\ E = 9\sigma_x^2 x^2/(1 - A)^2;$$

$$x = (\Pi N_A/6V_S)(z_R\sigma_R^2 + z_S\sigma_S^2);\ y = (\Pi N_A/6V_S)(z_R\sigma_R + z_S\sigma_S).$$

Following Marcus [22] and Kim et al. [21] in order to get the $\Delta P_{t,d\text{–}d}^0(i)$ term on mole fraction scale the quantity was again multiplied by the term x_{S1}.

$$x_{S1} = x_S(\mu_S/\sigma_S^3)/(\mu_R/\sigma_R^3). \quad (14)$$

This is the real mole fraction contribution due to dipole-dipole interaction [22]. Subtraction of $\Delta P_{t,cav}^0(i)$ and $\Delta P_{t,d\text{–}d}^0(i)$ from the total gives $\Delta P_{t,ch}^0(i)$ of the solute amino acid (Table 5).

Solubility of DL-alanine increases with increasing temperature in a particular composition of water-$NaNO_3$ mixed solvent system and with the increased concentration of $NaNO_3$. Solubility of the same also increases at higher temperature. The gradual increment of solubility of DL-alanine in presence of $NaNO_3$ may be due to the "salting in effect." The amino acid zwitterions can form associated species with cation and anion of the electrolyte.

The negative increment of $\Delta G_t^0(i)$ values for DL-alanine against the mol % of $NaNO_3$ indicates that DL-alanine would be stabilized with the increased concentration of the electrolyte carrying nitrate anion.

The $\Delta G_{t,cav}^0(i)$ values gradually become negative with $NaNO_3$ concentration (Table 5) which indicates that the amino acid acquire more stability with the increased mole percent of $NaNO_3$ [17, 22]. In the course of introduction of the amino acid into aqueous $NaNO_3$ system there can form strong ion-pair complexes between the zwitterionic amino acid and ions of the electrolyte, $NaNO_3$. This factor is likely be responsible for the stability of DL-alanine during transfer from water to water-$NaNO_3$ solvent system.

The $\Delta G_{t,d\text{–}d}^0(i)$ values (Table 5) depend on hard sphere diameter of the cosolvent and solute molecules, dipole moment of solvent and molar volume of the co-solvent [Eq. (9)].

The variation of $\Delta G_{t,d\text{–}d}^0(i)$ values indicates that amino acid becomes unstable in higher content of sodium nitrate media due to dipole–dipole interactions. The $\Delta G_{t,ch}^0(i)$ values gradually become negative with increase in concentration of $NaNO_3$ in $NaNO_3$-water system. Due to $\Delta G_{t,cav}^0(i)$ the amino acid becomes stabilized and due to $\Delta G_{t,d\text{–}d}^0(i)$ the amino acid becomes unstable. DL-alanine becomes stabilized by

the increased concentration of $NaNO_3$ in water–$NaNO_3$ solution system. The same is true for proteins and biomacromolecules.

Variations of $T\Delta S_t^0(i)$ and $T\Delta S_{t,ch}^0(i)$ are complicated. Actually $T\Delta S_t^0(i)$ is composed of transfer entropy due to cavity, dipole–dipole and chemical interaction effects:

$$T\Delta S_t^0(i) = T\Delta S_{t,cav}^0(i) + T\Delta S_{t,d-d}^0(i) + T\Delta S_{t,ch}^0(i).$$

At the initial concentration of $NaNO_3$ in the solvent system the $T\Delta S_{t,ch}^0(i)$ values exhibit very low negative increment but beyond 7 mol % the $T\Delta S_{t,ch}^0(i)$ values reach sharp negative increment. Increase in concentration of $NaNO_3$ in presence of the amino acid leads to breaking of hydrogen bonds between water molecules. In this region the numbers of free water molecules as well as cations and anions of the electrolyte increase which results in the positive entropy change. But with the increased concentration of $NaNO_3$, ion-pair formation occurs between the cation/anion and zwitterions of the amino acid to a greater extent resulting in lower number of free molecules in the system which is responsible for sharp decrement of $T\Delta S_{t,ch}^0(i)$ value.

EXPERIMENTAL

Chemicals and their purifications. DL-alanine, 99% purity (Merck), was used after drying as described in the previous publication [9]. Sodium nitrate, 99.5% purity (Merck, India) was oven dried for 3–4 days and cooled in a vacuum desiccator for 2 days prior to use. For formol titration standardized NaOH [E Merck] solution and phenolphthalein indicator [LR, BDH] were used. Triple distilled water was used for the preparation of solution and other experimental works.

Preparations of saturated solutions. The aqueous solvent of $NaNO_3$ of the concentrations of 0.0, 0.25, 0.50, 1.0, 2.0, 3.0, and 5.0 M were made. The solvent [H_2O/(H_2O+ $NaNO_3$)] and excess amount of amino acid was placed in stoppered glass tubes. A low-cum-high temperature thermostat was used for all measurements (accuracy ±0.02 K). Solubility of DL-alanine was measured by formol titrimetry method [9, 17]. The measurements were taken at 288.15, 293.15, 298.15, 303.15, and 308.15 K. Four sets of measurements for all co-solvent mixtures were made at all temperatures by equilibrating the solutions from both above and below (±0.02 K) the required temperatures. Solubility data coincided within the range ±(0.2–0.5)%.

CONCLUSIONS

According to our study the electrolyte affects the solubility of the amino acid as well as other biomolecules having hydrophobic side chains. High solubility of DL-alanine and stability of its solution is determined by increased concentration of the electrolyte that contains the nitrate anion. Stability of the amino acid arises mainly due cavity forming interactions, ion-pair formation, and other chemical types of interactions. The amino acid demonstrates higher disorder at lower content of the electrolyte.

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