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Enthalpic and volumetric studies of the interactions of propionamide in aqueous carboxylic acid solutions at 298.15 K

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Abstract

The enthalpies of solution and the molar volumes have been determined for propionamide in aqueous ethanoic acid and propionic acid solutions using a C-80 microcalorimeter and a DMA60/602 densimeter. The enthalpic and volumetric interaction coefficients of propionamide with the two carboxylic acids have been evaluated. Combining the previous studies for butyramide, the pair interaction coefficients are discussed from the point of view of dipole–dipole and structural interactions and the triplet interaction coefficients are interpreted by using the solvent separated association pattern.

Keywords: Apparent volume; Aqueous carboylic acid solutions; Enthalpy of solution; Interaction coefficients; Propionamide

1. Introduction

As the basis of life, proteins play an important role in the process of life and life phenomena. It is well known that most of the biological activities are realized with proteins. But because proteins have a complicated structure and some complicated effects on its structure, it is very difficult to study the interactions between proteins directly. Consequently, in thermodynamics, the behavior of proteins is often investigated on some simple model interactions between groups associated with proteins. As model compounds of polypeptides [1,2], amides play an important part in understanding the conformational stability of proteins and providing insights into physiochemical phenomena in life [3,4]. As is known, amide + carboxylic acid mixtures have an significant effect on regulating the order ⇔ disorder transformation reaction, which exists inter- and intramolecularly of proteins because they could give much information about solute–solute and long-range intramolecular interactions. Such systems have been investigated by a variety of physicochemical techniques such as infrared, NMR, viscosity, conductance and mass spectra, etc. [5–7]. We have reported the enthalpic and volumetric interaction coefficients of butyramide with carboxylic acids in water [8]. The present paper will further present the enthalpic and volumetric interactions between propionamide and ethanoic acid, propionic acid at 298.15 K.

2. Experimental

Analytical grade propionamide used in the experiments was recrystallized from acetone + water mixtures and dried under vacuum, thereafter, stored over P_2O_5 in a desiccator prior to use. The purification of analytical grade ethanoic acid, propionic acid and water has been described carefully [8].

The enthalpies of solution were measured at 298.15 K using a C-80 calorimeter (Setaram). About 0.05–0.3 g of propionamide was used every time and was weighed on a single-pan balance (Mettler) with a sensitivity of 10 ug. About 5 ml solvent was weighed on an analytical balance with a sensitivity of 0.1 mg. The calibration of the calorimeter was performed with an EJ-2 Joule-effect device (Setaram). The total uncertainty in the enthalpy of solution was about ±0.5%.

Solution densities were measured at 298.15 K with a vibrating-tube digital density meter (Model 60/602, Anton Paar). The precision of the density meter is $1.5 \times 10^{-6}\,\mathrm{g\,cm^{-3}}$. The density meter was calibrated with the known densities of dry air and conductivity water daily. The density of pure water at 298.15 K was taken as

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$0.99705\,\mathrm{g\,cm^{-3}}$ [9]. Reproducibility of the density measurements was better than $3 \times 10^{-6}\,\mathrm{g\,cm^{-3}}$.

3. Theoretical relations

According to the McMillan and Mayer theory [10], all the thermodynamic properties of multi-component solutions can be expressed by using a viral expansion in m, which relates to the non-ideal contributions of any total thermodynamic function to a series of pair, triplet and higher order interaction coefficients. The enthalpies of transfer of propionamide (b) from pure water (w) to carboxylic acid (a) aqueous solutions can be expressed as mentioned in [11]:

$$\Delta_{\mathrm{tr}} H_{\mathrm{b}}(\mathrm{w} \to \mathrm{w}+\mathrm{a}) = 2h_{\mathrm{ab}}m_{\mathrm{a}} + 3h_{\mathrm{abb}}m_{\mathrm{a}}m_{\mathrm{b}} + 3h_{\mathrm{aab}}m_{\mathrm{a}}^{2} + \cdots \quad (1)$$

where m_{a} and m_{b} are the molalities defined per kilogram of pure water, h_{ab} is the enthalpic pair interaction coefficient. h_{abb} and h_{aab} are the enthalpic triplet interaction coefficients. For the system studied, we used the truncated form of the equation as [8]:

$$\frac{\Delta_{\mathrm{tr}} H_{\mathrm{b}}(\mathrm{w} \to \mathrm{w}+\mathrm{a})}{m_{\mathrm{a}}} = 2h_{\mathrm{ab}} + 3h_{\mathrm{abb}}m_{\mathrm{b}} + 3h_{\mathrm{aab}}m_{\mathrm{a}} \quad (2)$$

The density data are used to calculate the apparent molar volumes of propionamide. In propionamide + water binary systems, the apparent molar volumes of propionamide can be expressed as:

$$\Phi v_{\mathrm{b}} = \frac{M_{\mathrm{b}}}{\rho} - \frac{10^{3}(\rho-\rho_{0})}{m_{\mathrm{b}}\rho\rho_{0}} \quad (3)$$

where M_{b} and m_{b} are, respectively, the molecular weight and the molality of propionamide. ρ and ρ_0 are, respectively, the densities of the solutions and water. In propionamide + carboxylic acid + water ternary systems, the apparent molar volumes of propionamide are calculated using

$$\Phi v_{\mathrm{b}} = \frac{1000 + m_{\mathrm{a}}M_{\mathrm{a}} + m_{\mathrm{b}}M_{\mathrm{b}}}{m_{\mathrm{b}}\rho} - \frac{1000 + m_{\mathrm{a}}M_{\mathrm{a}}}{m_{\mathrm{b}}\rho_{0}} \quad (4)$$

where m_{a} and m_{b} are, respectively, the molalities defined per kilogram of pure water and solvent. ρ is the densities of the ternary aqueous solution. ρ_0 is the density of carboxylic acid + water binary solution. According to the McMillan and Mayer approach, just the same as the transfer enthalpy in Eq. (1), the standard transfer volumes of propionamide from pure water to aqueous carboxylic acid solutions, can be expressed as follows:

$$\Delta_{\mathrm{tr}} \Phi v_{\mathrm{b}}(\mathrm{w} \to \mathrm{w}+\mathrm{a}) = 2v_{\mathrm{ab}}m_{\mathrm{a}} + 3v_{\mathrm{abb}}m_{\mathrm{a}}m_{\mathrm{b}} + 3v_{\mathrm{abb}}m_{a}^{2} + \cdots \quad (5)$$

where v_{ab} is the volumetric pair interaction coefficient, v_{abb} and v_{aab} are the volumetric triplet interaction coefficients. We also use the truncated formed as [8]:

$$\frac{\Delta_{\mathrm{tr}} \Phi v_{\mathrm{b}}(\mathrm{w} \to \mathrm{w}+\mathrm{a})}{m_{\mathrm{a}}} = 2v_{\mathrm{ab}} + 3v_{\mathrm{abb}}m_{\mathrm{b}} + 3v_{\mathrm{aab}}m_{\mathrm{a}} \quad (6)$$

4. Results and discussion

The measured enthalpies of solutions of propionamide in water and in various carboxylic acid are, respectively, given in Tables 1–3. The molar solution enthalpies of propionamide in pure water can be expressed by the following equation:

$$\Delta_{\mathrm{sol}} H_{\mathrm{m}}\ (\mathrm{J\,mol^{-1}}) = 6926 + 210m_{\mathrm{b}} \quad (7)$$

The transfer enthalpies of propionamide from pure water to the aqueous solutions of carboxylic acids have been calculated and fitted to Eq. (2) using a least squares method. The enthalpic interaction coefficients thus obtained, are given in Table 4, and the enthalpic interaction coefficients between butyramide and two carboxylic acids are also presented in the same table.

Densities, at 298.15 K, of solutions and the apparent molar volumes of propionamide in water and in the two carboxylic acid aqueous solutions, are given in Tables 5–7. The apparent molar volumes of propionamide in pure water is found to be a very linear function of the amide molality over the molality range studied and can be expressed as:

$$\Phi v_{\mathrm{b}}\ (\mathrm{cm^{3}\,mol^{-1}}) = 71.469 - 0.349m_{\mathrm{b}} \quad (8)$$

The standard partial molar volume of propionamide in Eq. (8) agrees very well with the literature [12]. The standard transfer volumes of propionamide, are thus obtained, and fitted to Eq. (6) using a least squares method. The volumetric interaction coefficients obtained are given in Table 8, together with the volumetric interaction coefficients between butyramide and the two corresponding carboxylic acid molecules.

4.1. Pair interaction

Seen from Table 4, h_{ab} have positive values, and increase with increasing size of the apolar side chain of carboxylic acid molecules. Also shown in Table 8, v_{ab} are negative varying from small to large values with the hydrocarbon chain extended. These are consistent with the interactions between butyramide and ethanoic acid or propionic acid.

Table 1
The enthalpies of solution of propionamide in pure water at 298.15 K

m_{b} (mol kg^{-1})	$\Delta_{\mathrm{sol}}H_{\mathrm{m}}$ (J mol^{-1})	m_{b} (mol kg^{-1})	$\Delta_{\mathrm{sol}}H_{\mathrm{m}}$ (J mol^{-1})
0.1397	6957	0.5487	7040
0.2992	6981	0.6310	7051
0.3857	7015	0.7060	7079
0.4789	7018		

Table 2
The enthalpies of solution of propionamide in acetic acid solutions at 298.15 K ($\mathrm{J\,mol^{-1}}$)

m_b ($\mathrm{mol\,kg^{-1}}$)	$\Delta_{sol}H_m$ ($m_a = 0.5000$)	m_b ($\mathrm{mol\,kg^{-1}}$)	$\Delta_{sol}H_m$ ($m_a = 0.9999$)	m_b ($\mathrm{mol\,kg^{-1}}$)	$\Delta_{sol}H_m$ ($m_a = 2.0001$)	m_b ($\mathrm{mol\,kg^{-1}}$)	$\Delta_{sol}H_m$ ($m_a = 3.0000$)
0.1455	7331	0.1462	7723	0.1564	8298	0.1684	8705
0.2612	7374	0.2667	7751	0.2730	8318	0.2952	8708
0.3773	7403	0.3864	7784	0.4020	8339	0.4132	8750
0.4966	7439	0.4952	7806	0.5281	8415	0.5495	8800
0.5969	7473	0.6224	7840	0.6445	8431	0.6886	8842
0.7190	7505	0.7277	7876	0.7800	8464	0.8002	8882

Table 3
The enthalpies of solution of propionamide in propionic acid solutions at 298.15 K ($\mathrm{J\,mol^{-1}}$)

m_b ($\mathrm{mol\,kg^{-1}}$)	$\Delta_{sol}H_m$ ($m_a = 0.5000$)	m_b ($\mathrm{mol\,kg^{-1}}$)	$\Delta_{sol}H_m$ ($m_a = 1.0000$)	m_b ($\mathrm{mol\,kg^{-1}}$)	$\Delta_{sol}H_m$ ($m_a = 1.9998$)	m_b ($\mathrm{mol\,kg^{-1}}$)	$\Delta_{sol}H_m$ ($m_a = 2.9997$)
0.1445	7465	0.1509	7925	0.1624	8712	0.1788	9283
0.2629	7501	0.2718	7957	0.2853	8736	0.3028	9300
0.3785	7543	0.3870	7990	0.4094	8767	0.4454	9319
0.4896	7569	0.5080	8013	0.5337	8805	0.5636	9335
0.6103	7628	0.6246	8061	0.6695	8810	0.7038	9352
0.7234	7635	0.7526	8108	0.7998	8822	0.8447	9360

Table 4
The enthalpic interaction coefficients of propionamide (pa) and butyramide (ba) with acetic acid and propionic acid in aqueous solutions at 298.15 K

Carboxylic acid	h_{ab} ($\mathrm{J\,kg\,mol^{-2}}$)		h_{abb} ($\mathrm{J\,kg^2\,mol^{-3}}$)		h_{aab} ($\mathrm{J\,kg^2\,mol^{-3}}$)	
	pa	ba[a]	pa	ba[a]	pa	ba[a]
Acetic acid	416 ± 5	660 ± 4	23 ± 5	−21 ± 5	−31 ± 1	−36 ± 1
Propionic acid	552 ± 7	847 ± 7	17 ± 8	−18 ± 9	−41 ± 2	−42 ± 2

[a] Ref. [8].

Table 5
Densities and apparent molar volumes of propionamide in pure water at 298.15 K

m_b ($\mathrm{mol\,kg^{-1}}$)	ρ ($\mathrm{g\,cm^{-3}}$)	Φv_b ($\mathrm{cm^3\,mol^{-1}}$)	m_b ($\mathrm{mol\,kg^{-1}}$)	ρ ($\mathrm{g\,cm^{-3}}$)	Φv_b ($\mathrm{cm^3\,mol^{-1}}$)
0.19698	0.997416	71.413	0.60377	0.998231	71.250
0.30214	0.997622	71.361	0.70846	0.998454	71.212
0.40301	0.997822	71.323	0.82658	0.998694	71.188
0.51209	0.998040	71.291	0.89990	0.998851	71.164

Table 6
Densities and apparent molar volumes of propionamide in aqueous acetic acid solutions at 298.15 K

m_b ($\mathrm{mol\,kg^{-1}}$)	ρ ($\mathrm{g\,cm^{-3}}$)	Φv_b ($\mathrm{cm^3\,mol^{-1}}$)	m_b ($\mathrm{mol\,kg^{-1}}$)	ρ ($\mathrm{g\,cm^{-3}}$)	Φv_b ($\mathrm{cm^3\,mol^{-1}}$)
$m_a = 0.50556$			$m_a = 0.99991$		
0.00000	1.001130		0.00000	1.004819	
0.20244	1.001457	71.324	0.20259	1.005089	71.321
0.30354	1.001623	71.303	0.30283	1.005228	71.292
0.39543	1.001773	71.290	0.41214	1.005378	71.276
0.49324	1.001938	71.266	0.49390	1.005488	71.270
0.60244	1.002120	71.248	0.61622	1.005655	71.256
0.71139	1.002312	71.215	0.71208	1.005793	71.234
0.78541	1.002439	71.201	0.83926	1.005979	71.206
0.89989	1.002643	71.171	0.92683	1.006110	71.186
$m_a = 1.49626$			$m_a = 1.99760$		
0.00000	1.008310		0.00000	1.011597	
0.20137	1.008522	71.344	0.19954	1.011753	71.386
0.30530	1.008639	71.309	0.30430	1.011840	71.361
0.40371	1.008739	71.318	0.40027	1.011920	71.346
0.51352	1.008856	71.309	0.48794	1.011988	71.348
0.61254	1.008967	71.291	0.58628	1.012069	71.338
0.70690	1.009080	71.266	0.67512	1.012141	71.332
0.82288	1.009213	71.248	0.82929	1.012258	71.333
0.88644	1.009287	71.237	0.91223	1.012323	71.330

Table 7
Densities and apparent molar volumes of propionamide in aqueous propionic acid solutions at 298.15 K

m_b (mol kg^{-1})	ρ (g cm^{-3})	Φv_b (cm^3 mol^{-1})	m_b (mol kg^{-1})	ρ (g cm^{-3})	Φv_b (cm^3 mol^{-1})
m_a = 0.50298			m_a = 0.99575		
0.00000	1.000120		0.00000	1.002879	
0.20000	1.000473	71.226	0.20818	1.003215	71.134
0.31053	1.000675	71.189	0.30704	1.003376	71.117
0.41766	1.000871	71.163	0.40364	1.003530	71.112
0.50795	1.001039	71.140	0.51618	1.003706	71.093
0.59499	1.001200	71.122	0.58220	1.003832	71.077
0.70046	1.001399	71.097	0.69544	1.004009	71.066
0.81656	1.001621	71.068	0.80015	1.004186	71.044
0.92975	1.001835	71.047	0.90911	1.004364	71.031
m_a = 1.50637			m_a = 2.00230		
0.00000	1.005461		0.00000	1.007726	
0.20387	1.005746	71.136	0.20233	1.007942	71.307
0.30961	1.005895	71.121	0.31633	1.008064	71.297
0.40989	1.006032	71.121	0.41080	1.008164	71.293
0.50764	1.006165	71.118	0.49717	1.008261	71.275
0.61041	1.006316	71.092	0.55197	1.008323	71.264
0.70697	1.006449	71.087	0.70080	1.008484	71.253
0.81494	1.006583	71.086	0.78854	1.008576	71.251
0.90974	1.006726	71.075	0.91467	1.008723	71.227

The physical meaning of the solute–solute interaction coefficients is, generally, related to the variations of thermodynamic properties when two solvated solute particles are brought from an infinite instance to a finite where their hydration shells are perturbed [13]. As for the polar non-electrolyte molecules with alkyl groups, the overall effects among the solutes reflect the following three effects [14]: (1) dipole–dipole interaction between polar groups, (2) hydrophobic–hydrophobic interaction of the alkyl groups, (3) 'thermochemical repulsion' between the solvated apolar alkyl groups and solvated polar groups. The first effect is exothermic and is accompanied by partial desolvation of them, which is an endothermic effect, thus weakening the exothermic effect caused by the dipole–dipole interaction. According to the destructive overlap model [12,15], the partial desolvation of polar groups will make some of the previously electrostricted water molecules return to their normal structure, causing a positive value to v_{ab}. On the other hand, the 'thermochemical repulsion' will make some water molecules in the hydrophobic hydration shell return to the bulk. Because the density of water is lower than that in bulk [16,17], the effect will produce a positive contribution to h_{ab} and a negative contribution to v_{ab}. With alkyl groups, when ethanoic acid or propionic acid interacts with butyramide, they will associate in the side-by-side manner [18]. Apart from dipole–dipole interaction and the 'thermochemical repulsion', there also exist hydrophobic–hydrophobic groups overlapping each other. In this process, more water molecules are squeezed out of the hydrophobic hydration shell to the bulk. Therefore, the effect will cause a larger positive contribution to h_{ab}, and larger negative contribution to v_{ab}. With the alkyl chain extended, the hydrophobic–hydrophobic interaction becomes more and more pronounced. Consequently, for propionamide interacting with carboxylic acid molecules, positive h_{ab} increase from ethanoic acid to propionic acid, and negative v_{ab} decrease in values.

4.2. Triplet interaction

As shown in Table 4, all enthalpic triplet interaction coefficients are much smaller than the pair interaction coefficient, and even present negative values. In addition, we also can see from Table 8 that the triplet volumetric interaction coefficients for all ternary systems are positive and much larger than the corresponding pair interaction coefficients in values. This is just like the interaction of bytyramide with two carboxylic acid molecules. When three polar

Table 8
The volumetric interaction coefficients of propionamide (pa) and butyramide (ba) with acetic acid and propionic acid in aqueous solutions at 298.15 K

Carboxylic acid	v_{ab} (cm^3 kg mol^{-2})		v_{abb} (cm^3 kg^2 mol^{-3})		v_{aab} (cm^3 kg^2 mol^{-3})	
	pa	ba[a]	pa	ba[a]	pa	ba[a]
Acetic acid	−0.088 ± 0.005	−0.047 ± 0.003	0.058 ± 0.004	0.085 ± 0.003	0.019 ± 0.002	0.020 ± 0.001
Propionic acid	−0.239 ± 0.004	−0.245 ± 0.005	0.057 ± 0.003	0.128 ± 0.004	0.062 ± 0.001	0.073 ± 0.002

[a] Ref. [8].

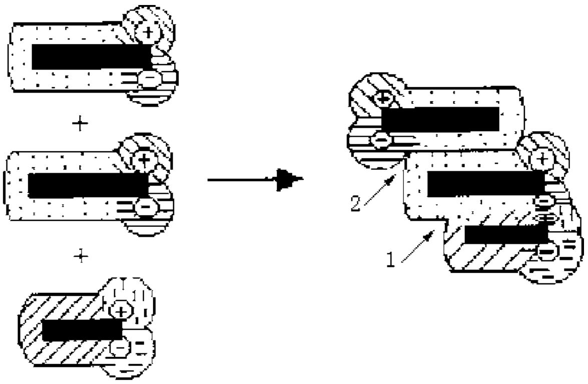

Fig. 1. Schematic representation of the triplet interaction: (1) direct contact; (2) solvent separated association.

non-electrolyte molecules with alkyl groups interact in solutions, two molecules first associate in a side-by-side manner [18,19]. Given another molecule participates in the triplet interaction in similar way to the two overlapping co-region molecules, the triplet coefficients, whether enthalpic or volumetric interaction coefficients would be the same sign with the corresponding pair interaction coefficients and furthermore, present larger values. This is, obviously, inconsistent with the experimental data. These evidences indicate that the triplet interaction and the pair interaction of the interacting molecules are different in the interaction mechanism.

We have given a reasonable explanation to the triplet interaction of butyramide and carboxylic acid molecules [8]. Franks [20] has pointed out that there is solvent separated association among hydrophobic solutes. In a Monte Carlo simulation study [21], some evidences were found for the coexistence of solvent separated and direct contact configurations. We think that no dipole–dipole interaction occurs in the triplet interaction between the third molecule and the pair of two molecules associated in side-by-side manner, and their alkyl groups will take place the solvent separated association (Fig. 1). The direct contact among alkyl groups are hydrophobic–hydrophobic interaction, mentioned often, which will break the hydrophobic hydration structure of the alkyl groups. But the solvent separated association will not have the same effect. The molecular dynamics calculations have obtained that the hydrophobic group has a protecting effect [17] or a blocking effect [16] for the hydration shell water molecules. We have ascribed the two effects to a shielding effect [8]. Hydrophobic group can shield its hydrophobic hydration structure from the shock of other water molecules. For the kind of solvent separated association, aside from the shielding effect of the hydrophobic groups themselves, with no co-region overlap occurring, they will provide a mutual shielding effect among the associated hydrophobic groups. This means that the solvent separated association not only does not break the hydrophobic hydration structure of the alkyl groups but also increase the structure with the mutual shielding effect, which is contrary to the direct contact association. As a result, the solvent separated association will cause a negative value to enthalpic functions and positive to volumetric coefficients. The triplet interaction mechanism could explain why the triplet interaction coefficients are opposite in sign to the corresponding pair coefficients.

As shown in Tables 4 and 8, when butyramide or propionamide interacts with the same carboxylic acid molecule, h_{aab} and v_{aab} are very similar. But h_{abb} for butyramide is smaller than that for propionamide, and v_{abb} is larger for butyramide than for propionamide. These results can also be explained according to the solvate separated association mentioned above. As is well known, carboxylic acid belongs to strong polar molecules. In a–a–b triplet interactions, the two carboxylic acid molecules will associate in side-by-side manner, and amides participate in the triplet interaction by the solvent separated association. Compared to propionamide, the alkyl chain of butyramide is longer, but the length of the alkyl chain which participates in the solvent separated association is the same because both ethanoic acid and propionic acid has a short hydrocarbon chain. Consequently, h_{aab} and v_{aab} are the same. In a–b–b triplet interaction, one carboxylic acid molecule first associates with one amide molecule abiding by the side-by-side manner. The other amide molecule interacts with them according to the solvent separated manner. Compared to propionamide, the two molecules of butyramide will have a larger solvent separated association, thus causing a larger negative contribution to the enthalpic coefficients and a larger positive contribution to the volumetric coefficients. This is why h_{abb} for butyramide is smaller and v_{abb} is larger for butyramide, compared to propionamide.

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