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Calorimetric and volumetric studies of the interactions of formamide with alkan-1-ol in water at 298.15 K

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Abstract

Enthalpies of solution and apparent molar volumes have been measured for formamide in aqueous methanol, ethanol and propanol solutions by using a C-80 microcalorimeter and vibrating tube digital densimeter. According to these data, enthalpic and volumetric interaction coefficients were obtained. The pair interaction coefficients are discussed from the perspective of dipole–dipole interaction and structural interaction. At the same time, attempts have been made to interpret the triplet coefficients by using the solvent separated association pattern.

Keywords: Formamide; Alkan-1-ol; Apparent volume; Enthalpy of solution; Interaction coefficients

1. Introduction

It is commonly admitted that protein, as the basis of life, has been under extensive research in solution and biological chemistry. To study the interactions between groups in protein in aqueous solutions is of great importance in exploring the structure of protein and the non-bonding interaction of protein. As model compounds of polypeptides [1,2], amides play an important role in understanding the conformational stability of proteins and providing insights into physiochemical phenomena in life [3,4]. We choose here the system amide + alkan-1-ols as our research object to study their enthalpic and volumetric interactions.

2. Experimental

Formamide (AR) was kept over $CaCO_3$ overnight and then distilled twice at reduced pressure. Methanol (AR), ethanol (AR) and propanol (AR) were distilled, respectively, under atmospheric pressure with middle fraction collected. All of the purified products were stored over P_2O_5 in a desiccator before use. The water used in our experiment was deionized and redistilled.

Enthalpies of solution were measured in a C-80 calorimeter (Setaram) using reversed-mixing vessel. The reversed-mixing vessel has two chambers separated by a tilting lid. About 0.05–0.25 g formamide, which was weighed on a single-pan balance (Mettler) with a sensitivity of 10 μg, was introduced into the vessel and then the lid was put in place. In order to obtain a complete separation of the chambers, 0.5 g of mercury was put into the vessel to seal the gapping between the vessel and the lid. About 5 ml solvent was weighed on an analytical balance with a sensitivity

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of 0.1 mg and introduced into the vessel. The calibration of the calorimeter was performed with an EJ2 Joule-effect device (Setaram). The total uncertainty in the measurement was about ±0.5%.

Solution densities were measured with a vibrating-tube digital density meter (model 60/602, Anton Paar), which has been described carefully in [5]. The accuracy of the density meter is estimated to be $\pm 1.5 \times 10^{-6}\,\mathrm{g\,cm^{-3}}$. The temperature around the density meter cell was controlled by circulating water from a constant-temperature bath (Scott, Germany). A CT14500 temperature controller and a CK-100 ultracryostat were employed to maintain the bath temperature at 298.15 ± 0.005 K. The density meter was calibrated daily with dry air and deionized water. The density of pure water at 298.15 K was taken as $0.99705\,\mathrm{g\,cm^{-3}}$ [6]. Reproductivity of the density measurements was better than $\pm 3 \times 10^{-6}\,\mathrm{g\,cm^{-3}}$.

3. Theoretical relations

According to McMillan and Mayer approach [7], 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 formamide (b) from pure water (w) to alkan-1-ol solutions (a) can be expressed as mentioned in [8]

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

where m_a and m_b are the molalities defined per kilogram of pure water, h_{ab} is the heteroactic enthalpic pair interaction coefficients, and h_{abb}, h_{aab} are the enthalpic triplet interaction coefficients. For the system studied here, we used the truncated form of the equation as mentioned in [9]

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

Apparent molar volumes of formamide in pure water were calculated by using

$$\Phi_{\mathrm{Vb}} = \frac{M_{\mathrm{b}}}{\rho} - 10^{3} \times \frac{\rho - \rho_0}{m_{\mathrm{b}}\rho\rho_0} \tag{3}$$

where M_b and m_b are, respectively, the molecular weight and the molality of formamide and ρ, ρ_0 are, respectively, the densities of solution and water. In ternary systems formamide + alkan-1-ol + water, the apparent molar volumes of formamide Φ_{Vb} can be calculated as

$$\Phi_{\mathrm{Vb}} = \frac{1000 + m_{\mathrm{a}}M_{\mathrm{a}} + m_{\mathrm{b}}M_{\mathrm{b}}}{m_{\mathrm{b}}\rho} - \frac{1000 + m_{\mathrm{a}}M_{\mathrm{a}}}{m_{\mathrm{b}}\rho_0} \tag{4}$$

where m_a and m_b are the molalities defined per kilogram of pure water, ρ is the densities of the ternary aqueous solution and ρ_0 is the density of alkan-1-ol + water binary solution. Just as the transfer enthalpy in Eq. (1), the standard transfer volumes of formamide from pure water to aqueous alkan-1-ol solutions can be expressed as follows:

$$\Delta_{\mathrm{tr}} \Phi_{\mathrm{Vb}}(\mathrm{w} \rightarrow \mathrm{w}+\mathrm{a}) = 2V_{\mathrm{ab}}m_{\mathrm{a}} + 3V_{\mathrm{abb}}m_{\mathrm{a}}m_{\mathrm{b}} + 3V_{\mathrm{aab}}m_{\mathrm{a}}^{2} + \cdots \tag{5}$$

We can also use the truncated form as

$$\frac{\Delta_{\mathrm{tr}} \Phi_{\mathrm{Vb}}(\mathrm{w} \rightarrow \mathrm{w}+\mathrm{a})}{m_{\mathrm{a}}} = 2V_{\mathrm{ab}} + 3V_{\mathrm{abb}}m_{\mathrm{b}} + 3V_{\mathrm{aab}}m_{\mathrm{a}} \tag{6}$$

where V_{ab} is the volumetric pair interaction coefficients and V_{abb}, V_{aab} are the volumetric triplet interaction coefficients.

4. Results and discussion

The measured enthalpies of solution of formamide in water and in aqueous methanol, ethanol and propanol solutions are given, respectively, in Tables 1–4. And the densities of solution of formamide

Table 1
The enthalpies, ΔH_m ($\mathrm{J\,mol^{-1}}$), of solution of formamide in pure water at 298.15 K

m_b ($\mathrm{mol\,kg^{-1}}$)	ΔH_m	m_b ($\mathrm{mol\,kg^{-1}}$)	ΔH_m
0.2272	2065	0.7987	1983
0.4118	2022	0.9449	1952
0.5880	2003	1.1344	1940

Table 2
The enthalpies, ΔH_m (J mol^{-1}), of solution of formamide in aqueous methanol solutions at 298.15 K

m_b (mol kg^{-1})	ΔH_m	m_b (mol kg^{-1})	ΔH_m	m_b (mol kg^{-1})	ΔH_m	m_b (mol kg^{-1})	ΔH_m
m_a = 0.4997 (mol kg^{-1})		m_a = 1.0000 (mol kg^{-1})		m_a = 2.0000 (mol kg^{-1})		m_a = 3.0010 (mol kg^{-1})	
0.2244	2358	0.2390	2626	0.2449	3032	0.2644	3307
0.4069	2331	0.4240	2576	0.4530	2981	0.4458	3271
0.5929	2295	0.6011	2543	0.6232	2935	0.6502	3230
0.7757	2266	0.7954	2512	0.8252	2895	0.8449	3207
0.9467	2234	0.9795	2467	1.0036	2857	1.0408	3168
1.1608	2201	1.1570	2445	1.2077	2808	1.2204	3128

Table 3
The enthalpies, ΔH_m (J mol^{-1}), of solution of aqueous formamide in ethanol solutions at 298.15 K

m_b (mol kg^{-1})	ΔH_m	m_b (mol kg^{-1})	ΔH_m	m_b (mol kg^{-1})	ΔH_m	m_b (mol kg^{-1})	ΔH_m
m_a = 0.5000 (mol kg^{-1})		m_a = 1.0000 (mol kg^{-1})		m_a = 2.0000 (mol kg^{-1})		m_a = 3.0010 (mol kg^{-1})	
0.2301	2551	0.2315	3004	0.2673	3882	0.2676	4531
0.4380	2516	0.4313	2948	0.4396	3840	0.4632	4461
0.5907	2484	0.6053	2915	0.6478	3767	0.6627	4375
0.7727	2448	0.8261	2858	0.8295	3689	0.8732	4296
0.9514	2417	0.9763	2836	1.0303	3603	1.0898	4222
1.1537	2372	1.1662	2794	1.2563	3560	1.2762	4166

Table 4
The enthalpies, ΔH_m (J mol^{-1}), of solution of formamide in aqueous propanol solutions at 298.15 K

m_b (mol kg^{-1})	ΔH_m	m_b (mol kg^{-1})	ΔH_m	m_b (mol kg^{-1})	ΔH_m	m_b (mol kg^{-1})	ΔH_m
m_a = 0.5001 (mol kg^{-1})		m_a = 1.0000 (mol kg^{-1})		m_a = 2.0000 (mol kg^{-1})		m_a = 3.0000 (mol kg^{-1})	
0.2518	2676	0.2348	3228	0.2528	4123	0.2684	4529
0.4204	2638	0.4277	3177	0.4524	4064	0.5088	4425
0.6096	2603	0.6552	3122	0.6662	3972	0.6894	4362
0.7993	2561	0.8087	3083	0.8785	3924	0.9064	4280
0.9924	2525	0.9887	3038	1.0467	3858	1.1277	4207
1.1486	2497	1.2306	2982	1.2639	3819	1.3504	4148

in pure water and in aqueous alkan-1-ol solutions at 298.15 K are given, respectively, in Tables 5–8. The molar enthalpies of solution and apparent molar volumes of formamide in pure water can be expressed by the following equations:

$$\Delta H_m(\mathrm{J\,mol^{-1}}) = 2086 - 134m_b \quad (\mathrm{S.D.} = 8) \tag{7}$$

$$\Phi_{Vb}(\mathrm{cm^3\,mol^{-1}}) = 38.483 + 0.104m_b \quad (\mathrm{S.D.} = 0.007) \tag{8}$$

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

m_b (mol kg^{-1})	ρ (g cm^{-3})	Φ_{Vb} (cm^3 mol^{-1})	m_b (mol kg^{-1})	ρ (g cm^{-3})	Φ_{Vb} (cm^3 mol^{-1})
0.20293	0.998386	38.500	0.78123	1.002025	38.576
0.30114	0.999022	38.511	1.00251	1.003368	38.590
0.40537	0.999688	38.526	1.16929	1.004369	38.594
0.62638	1.001078	38.549	1.21188	1.004608	38.608

Table 6
Densities and apparent molar volumes of formamide in aqueous methanol solutions at 298.15 K

m_b (mol kg^{-1})	ρ (g cm^{-3})	Φ_{Vb} (cm^3 mol^{-1})	m_b (mol kg^{-1})	ρ (g cm^{-3})	Φ_{Vb} (cm^3 mol^{-1})
m_a = 0.49320 (mol kg^{-1})			m_a = 0.97682 (mol kg^{-1})		
0.00000	0.994164		0.00000	0.991517	
0.11044	0.994894	38.484	0.06582	0.991952	38.476
0.21476	0.995574	38.503	0.13657	0.992413	38.509
0.28010	0.995996	38.513	0.17084	0.992635	38.518
0.33516	0.996349	38.520	0.27942	0.993334	38.534
0.41395	0.996848	38.537	0.33129	0.993663	38.547
0.48732	0.997310	38.548	0.39488	0.994061	38.569
0.52827	0.997566	38.555	0.46264	0.994483	38.586
0.56596	0.997797	38.567	0.57673	0.995192	38.599
m_a = 1.49118 (mol kg^{-1})			m_a = 1.97944 (mol kg^{-1})		
0.00000	0.988874		0.00000	0.986556	
0.16431	0.989947	38.509	0.16065	0.987600	38.513
0.20632	0.990218	38.514	0.23128	0.988051	38.534
0.25044	0.990498	38.536	0.29412	0.988445	38.563
0.31130	0.990886	38.544	0.34931	0.988788	38.586
0.36821	0.991244	38.559	0.39302	0.989058	38.601
0.43716	0.991670	38.586	0.46686	0.989522	38.597
0.50313	0.992074	38.608	0.54736	0.990007	38.631
0.58199	0.992557	38.623	0.63680	0.990541	38.661

As is shown above, the homoactic enthalpic interaction coefficient (h_{bb}) is −134 J kg mol^{-1} which agrees very well with −115 J kg mol^{-1} in [10], the standard partial molar volume of formamide is 38.483 cm^3 mol^{-1} which agrees very well with 38.5 cm^3 mol^{-1} as reported in [11–13]. The transfer enthalpies and standard transfer volumes of formamide from pure water to aqueous alkan-1-ols solutions have been calculated

Table 7
Densities and apparent molar volumes of formamide in aqueous ethanol solutions at 298.15 K

m_b (mol kg^{-1})	ρ (g cm^{-3})	Φ_{Vb} (cm^3 mol^{-1})	m_b (mol kg^{-1})	ρ (g cm^{-3})	Φ_{Vb} (cm^3 mol^{-1})
m_a = 0.49687 (mol kg^{-1})			m_a = 0.99313 (mol kg^{-1})		
0.00000	0.992888		0.00000	0.989200	
0.14757	0.993859	38.498	0.17260	0.990322	38.541
0.20057	0.994203	38.510	0.23803	0.990737	38.571
0.25194	0.994535	38.516	0.27411	0.990965	38.582
0.31969	0.994968	38.532	0.32344	0.991274	38.599
0.37940	0.995347	38.543	0.38320	0.991649	38.607
0.42433	0.995628	38.557	0.43534	0.991971	38.622
0.49935	0.996099	38.567	0.48009	0.992250	38.624
0.55658	0.996454	38.577	0.55521	0.992709	38.641
m_a = 1.48481 (mol kg^{-1})			m_a = 1.99738 (mol kg^{-1})		
0.00000	0.985934		0.00000	0.98278	
0.11328	0.986662	38.592	0.15413	0.983748	38.691
0.20091	0.987215	38.626	0.19650	0.984007	38.722
0.24536	0.987491	38.648	0.25805	0.984382	38.748
0.31499	0.987924	38.662	0.33000	0.984815	38.778
0.41000	0.988509	38.680	0.38700	0.985167	38.762
0.44061	0.988700	38.675	0.44299	0.985491	38.804
0.49968	0.989061	38.683	0.49441	0.985798	38.810
0.55791	0.989401	38.717	0.58830	0.986343	38.843

Table 8
Densities and apparent molar volumes of formamide in aqueous propanol solutions at 298.15 K

m_b (mol kg^{-1})	ρ (g cm^{-3})	Φ_{Vb} (cm^3 mol^{-1})	m_b (mol kg^{-1})	ρ (g cm^{-3})	Φ_{Vb} (cm^3 mol^{-1})
m_a = 0.50025 (mol kg^{-1})			m_a = 0.99361 (mol kg^{-1})		
0.00000	0.992249		0.00000	0.988190	
0.14816	0.993218	38.512	0.10943	0.988893	38.580
0.19506	0.993518	38.537	0.15038	0.989152	38.599
0.24304	0.993826	38.543	0.19049	0.989401	38.633
0.30642	0.994227	38.563	0.31905	0.990197	38.674
0.37828	0.994680	38.575	0.36588	0.990481	38.694
0.42745	0.994984	38.592	0.39971	0.990684	38.710
0.50504	0.995466	38.604	0.44161	0.990938	38.719
0.59165	0.995992	38.628	0.49535	0.991258	38.738
m_a = 1.48546 (mol kg^{-1})			m_a = 2.00331 (mol kg^{-1})		
0.00000	0.984589		0.00000	0.981122	
0.08958	0.985147	38.724	0.10761	0.981765	38.927
0.12751	0.985375	38.789	0.18443	0.982210	38.999
0.18011	0.985693	38.815	0.26868	0.982689	39.057
0.26240	0.986185	38.849	0.30170	0.982867	39.106
0.30615	0.986443	38.868	0.37360	0.983275	39.114
0.36171	0.986766	38.897	0.42231	0.983549	39.112
0.44469	0.987258	38.897	0.50820	0.984026	39.141
0.51321	0.987649	38.925	0.56090	0.984305	39.176

and fitted, respectively, to Eqs. (2) and (6) by using a least-squares method. The enthalpic interaction coefficients are given in Table 9 and volumetric interaction coefficients in Table 10.

4.1. Pair interaction

The interaction in between polar non-electrolyte molecules includes [14]: (1) dipole–dipole interaction between polar groups, (2) hydrophobic–hydrophobic interaction between alkyl groups, (3) “thermochemical repulsion” between the solvated polar groups and solvated apolar alkyl groups. For the system we studied here, formamide has no alkyl groups. Therefore, when discussing the pair interactions between formamide and alkan-1-ols, we can only take dipole–dipole interaction and “thermochemical repulsion” into account.

The pair enthalpic interaction coefficient, h_{bb}, between two formamide molecules, is negative as mentioned above, which is just a result of dipole–dipole interaction. Between formamide and alkan-1-ols, apart from dipole–dipole interaction, there still exits “thermochemical repulsion,” which is related to hydrophobic hydration. The “iceberg formation” model proposed by Frank and Evans [15] has stated that the structure of water around hydrophobic solutes

Table 9
The enthalpic interaction coefficients of formamide with methanol, ethanol and propanol in aqueous solutions at 298.15 K

Alkan-1-ol	h_{ab} (J kg mol^{-2})	h_{abb} (J kg^2 mol^{-3})	h_{aab} (J kg^2 mol^{-3})
Methanol	317.3 ± 3	−15.9 ± 2	−21.5 ± 1
Ethanol	517.6 ± 4	−32.2 ± 3	−18.5 ± 1
Propanol	670.8 ± 6	−33 ± 4	−51.2 ± 2

Table 10
The volumetric interaction coefficients of formamide in aqueous alkan-1-ol solutions at 298.15 K

Alkan-1-ols	V_{ab} (cm^3 kg mol^{-2})	V_{abb} (cm^3 kg^2 mol^{-3})	V_{aab} (cm^3 kg^2 mol^{-3})
Methanol	−0.013 ± 0.009	0.002 ± 0.001	0.039 ± 0.003
Ethanol	−0.016 ± 0.003	0.019 ± 0.001	0.044 ± 0.004
Propanol	−0.021 ± 0.005	0.041 ± 0.002	0.079 ± 0.006

is strengthened, which has also been examined and confirmed both experimentally and theoretically [16]. Investigations indicated that the structure strengthening is associated with the local density decreasing around them [16,17]. The dipole–dipole interaction between formamide and alkan-1-ols will do negative contribution to h_{ab}. The partial desolvation accompanied, between polar groups, according to destructive overlap model [18,19], will make the previously electrostricted water molecules return to their normal structure, causing a positive contribution to V_{ab}. On the contrary, the "thermochemical repulsion" between formamide and alkyl groups of alkan-1-ols will break the hydrophobic hydration structure, causing a positive contribution to h_{ab} and a negative contribution to V_{ab}. Seen from Tables 9 and 10, h_{ab} all are positive and V_{ab} all are negative, which indicate that "thermochemical repulsion" effect is prevailing. For the system studied here, the absolute values of h_{ab} and V_{ab} increase as the alkyl chain of alkan-1-ols increases. This is because "thermochemical repulsion" effect will increase as the hydrophobic group increases. This is consistent with what Lilley and co-workers have determined [20].

4.2. Triplet interaction

According to McMillan and Mayer approach [7], there are two sets of triplet interaction coefficients. One is a–a–b type and the other is a–b–b type. Just as Lilley and co-workers have proposed, as for polar molecules with longer alkyl chains, two molecules associate in side-by-side manner and the enthalpic pair interaction coefficient is generally positive. Whereas, as for polar molecules with shorter or no alkyl chains, two molecules associate in head-on fashion and the enthalpic pair interaction coefficient is negative [21]. For the a–b–b type triplet interaction among two formamide molecules and one alkan-1-ol molecule, two formamide molecules with strong polarity will first interact in head-on fashion and form a cyclic dimer, which takes on electric neutrality. The cyclic dimer of formamide molecules is similar to an apolar molecule. When an alkan-1-ol molecule with long alkyl chain interacts with the cyclic dimer, the dominating interaction will be hydrophobic–hydrophobic interaction. According to the cosphere overlap model [21], this interaction will break the hydrophobic hydration structure and do positive contribution to h_{abb}. Therefore, the negative value of h_{abb} is smaller than that of h_{bb}. The negative value of h_{abb} is still dipole–dipole interaction. In this process, the previous electrostatic water molecules are squeezed out and will return to their normal structure, which will do positive contribution to volumetric coefficients. Therefore, V_{abb} are all positive.

In the system studied here, the a–a–b type triplet interaction is related to the interaction between one formamide molecule and two alkan-1-ol molecules. As is shown in Tables 9 and 10, h_{aab} are all negative and V_{aab} are all positive. Due to the strong polarity, the formamide molecule will take place interaction with one alkan-1-ol molecule and do positive contribution to h_{aab} (see the values of h_{ab} in Table 9). For methanol, ethanol and propanol molecules, their homoactic pair enthalpic interaction coefficients are all positive [22,23], which is characteristic of the interaction in side-by-side manner. If another alkan-1-ol molecule interacts with the associated formamide–alkan-1-ol molecule pair in side-by-side manner, h_{aab} should have more positive value. However, the experimental data obtained are all negative. It suggests that other pattern interaction exists.

Franks [24] has proposed that there exist solvent separated association between hydrophobic solutes. In a Monte Carlo simulation study [25], some evidences have been found and have proved the coexistence of solvent separated association and direct contact configurations. We think that no dipole–dipole interaction occurs in triplet interaction between the second alkan-1-ol molecule and the formamide–alkan-1-ol molecule pair. Therefore, their alkyl groups will not take place direct contact, but take place solvent separated association. Direct contact will break the hydrophobic hydration structure of alkyl groups and do positive contributions to enthalpic functions and negative to volumetric coefficients. Solvent separated association will not only break the hydrophobic hydration structure, but also can produce a mutual shielding effect with alkyl groups closing to each other [26]. As a result, this effect will strengthen hydrophobic hydration structure and produce enthalpic and volumetric effect that is contrary to what direct contact has produced, causing h_{aab} to be negative and V_{aab} positive.

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