Enthalpies of mixing of water with *N,N*-disubstituted amides of aliphatic carboxylic acids

A. M. Zaichikov[a]* *and M. V. Kulikov*[b]

[a]*Ivanovo State Academy of Chemistry and Technology,*
7 prosp. F. Engel'sa, 153460 Ivanovo, Russian Federation.
Fax: 007 (093 2) 32 9502
[b]*Institute of the Chemistry of Non-Aqueous Solutions, Russian Academy of Sciences,*
1 ul. Akademicheskaya, 153045 Ivanovo, Russian Federation.
Fax: 007 (093 2) 37 8507

The enthalpies of mixing of *N,N*-dialkylpropionamides with water were measured at 298.15 K. A comparative analysis of enthalpy effects (H^E) of mixing of water with *N,N*-disubstituted amides of formic, acetic, and propionic acids was performed. It was established that the H^E values depend on the length of the *N,N*-alkyl substituents and the size of acidic radicals. The size of the nonpolar group and the electron-donor ability of amide molecules primarily affect the enthalpy of mixing. The relative electron-donor abilities of the amides were estimated by the calorimetric method. The results obtained were discussed by invoking thermochemical data for aqueous solutions of hexamethylphosphoric triamide.

Key words: enthalpies of mixing, enthalpies of solution; *N,N*-disubstituted amides, hexamethylphosphoric triamide, aqueous solutions; electron-donor ability.

N,N-Disubstituted (tertiary) amides of aliphatic carboxylic acids and their aqueous solutions have attracted considerable interest[1,2] because of their high dissolving ability. Despite their wide practical use, the physicochemical properties of both individual and mixed solvents based on them have been little studied.

In this work, the enthalpies of mixing of water with *N,N*-dimethylpropionamide (DMPA), *N,N*-diethylpropionamide (DEPA), and *N,N*-dipropylpropionamide (DPPA) were determined at 298.15 K. Previously,[3–6] we have measured the excess thermodynamic functions (H^E, G^E, and TS^E) of aqueous solutions of hexamethylphosphoric triamide (HMPA), *N,N*-dimethylformamide (DMF), *N,N*-dimethylacetamide (DMA), and *N*-methylpyrrolidone (NMP) at various temperatures. The entropies and enthalpies of formation of the heterocomponent water—amide hydrogen bond[6] have been calculated on the basis of a model using data on G^E and H^E. The results previously obtained[4,5] in studying the peculiarities of interparticle interactions of several *N,N*-disubstituted formamides and acetamides with water[7] were supplemented with the measurements of the enthalpies of mixing of water with *N,N*-diethylformamide (DEF), *N,N*-dipropylformamide (DPF), and *N,N*-diethylacetamide (DEA). The coeffcients of the binary amide—amide interactions in dilute solutions have been reported in the literature.[8]

To discuss the properties of the water—*N,N*-dialkylamide systems, we used thermochemical data for aqueous solutions of HMPA,[3] which has an extremely high solvation ability of the phosphoryl group and a number of other interesting physicochemical properties.[9] In addition, the enthalpies of mixing of amides with hexane and chloroform were measured.

Experimental

The enthalpies of mixing were measured on a hermetic isothermic-shell swinging calorimeter described previously.[10] The relative errors of the measurements did not exceed 1%. Bidistilled water and *N,N*-disubstituted amides of "pure" grade, which were vacuum distilled, dried over molecular sieves (4 Å), and repeatedly vacuum distilled, were used in the experiments. Hexamethylphosphoric triamide of "pure" grade was purified following the procedure described previously.[3] The water content in these organic solvents determined by the Fischer method did not exceed 0.03 wt.%. To remove the ethanol admixture, chloroform of "pure" grade was washed with bidistilled water and then purified following the known procedure.[9] Hexane of "pure" grade was used without further purification.

Results and Discussion

Experimental values of the excess enthalpy of mixing ($H^E{}_{\text{exp}}$) in the binary systems water—dialkylamide of propionic acid were approximated by the Redlich—Kister polynomials using the least squares method

$$H^E{}_{\text{calc}} = X_1X_2\sum_{n=1}^{p} c_n(X_1 - X_2)^{n-1}, \qquad (1)$$

where X_1 and X_2 are the mole fractions of the components of the systems, and p is the number of coefficients in the polynomial. Hereafter subscript 1 is referred to

Translated from *Izvestiya Akademii Nauk. Seriya Khimicheskaya*, No. 10, pp. 1805—1810, October, 1997.

Table 1. Excess experimental enthalpies of mixing (H^E_{exp}/J mol^{-1}) in H_2O—*N,N*-disubstituted propionamide systems at 298.15 K

H_2O—DMPA		H_2O—DEPA		H_2O—DPPA	
X_2	$-H^E_{exp}$	X_2	$-H^E_{exp}$	X_2	$-H^E_{exp}$
0.0274	377.8	0.0253	365.7	0.0013	30.58
0.0771	803.9	0.0784	665.8	0.0024	54.71
0.1093	971.0	0.1101	749.0	0.0034	77.27
0.1587	1144.0	0.1606	817.4	0.0045	97.57
0.2173	1261.0	0.2103	837.8	...	...
0.2865	1320.0	0.2510	834.4	...	...
0.3524	1326.0	0.3026	812.6	0.6547	−138.70
0.4571	1263.0	0.3658	771.0	0.7217	−126.30
0.5602	1137.0	0.4432	708.0	0.7715	−103.10
0.6509	967.1	0.4924	666.1	0.8267	−76.04
0.7001	860.6	0.5369	631.8	0.8820	−44.18
0.7501	742.9	0.6347	550.1	0.9323	−16.79
0.8501	474.4	0.7412	432.3	0.9656	−6.34
0.9351	212.4	0.8401	297.6		
0.9601	134.9	0.9255	150.8		
0.9780	78.02	0.9507	102.0		
		0.9702	62.34		

Note. X_2 is the mole fraction of amide. For the H_2O—DMPA system, $c_1 = -4868.21$, $c_2 = -2522.05$, $c_3 = -1997.94$, $c_4 = -972.074$, $c_5 = -444.006$, $c_6 = -2664.78$, $c_7 = -2461.83$, and $\sigma = 3.7$; here c_i/J mol^{-1} are the coefficients of Eq. (1) and σ/J mol^{-1} is the standard error of approximation.

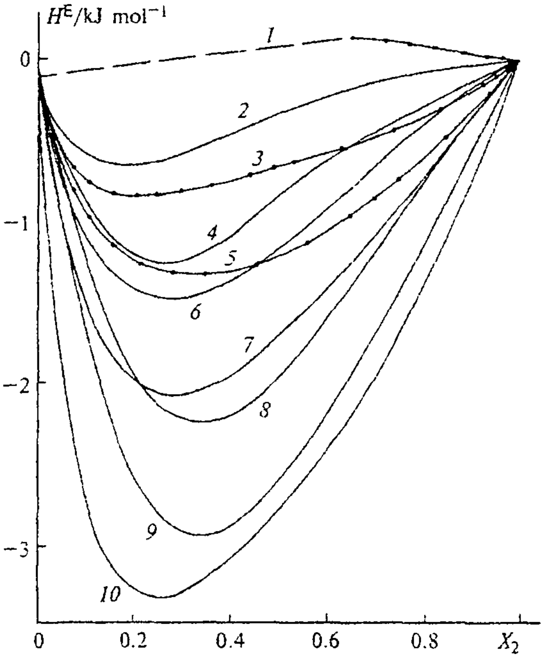

Fig. 1. Dependences of enthalpies of mixing (H^E) of water with *N,N*-dusubstituted amides of carboxylic acids and HMPA at 298.15 K: DPPA (*1*), DPF[7] (*2*), DEPA (*3*), DPA[11] (*4*), DMPA (*5*), DEF[7] (*6*), DEA[7] (*7*), DMF[4] (*8*), DMA[5] (*9*), and HMPA[3] (*10*).

water, hexane, and chloroform, while subscript 2 is referred to amides. The H^E_{exp} values, the coefficients of the Redlich—Kister equation (c_i), and the standard deviation of the approximation are given in Table 1. We failed to adequately describe the experimental data for the H_2O—DEPA system using a reasonable number of the c_i coefficients in polynomial (1) because of the rather complicated shape of the $H^E(X_2)$ curve. Stratification is observed in the H_2O—DPPA system.

The $H^E(X_2)$ dependences in the systems water—*N,N*-disubstituted amide of carboxylic acid and water—HMPA calculated at 298.15 K and supplemented with the literature data[11] on the excess enthalpies of aqueous solutions of *N,N*-dipropylacetamide (DPA) are presented in Fig. 1. A dependence of the absolute values of H^E on the size of the alkyl substituents at the N atom is observed in the region of medium concentrations. For tertiary formamides and acetamides, the exothermic effect of mixing rapidly decreases in the series DMF > DEF > DPF and DMA > DEA > DPA. The same is also observed for mixtures of water with *N,N*-dialkylpropionamides; additionally, stratification occurs and the $H^E(X_2)$ dependence becomes S-shaped in the region of endoeffects in the H_2O—DPPA system.

Previously,[3—6] it has been shown that the large negative values of H^E in aqueous solutions of HMPA, DMF, DMA, and NMP are due to the fact that the exoeffects of the formation of stable heterocomponent hydrogen bonds, which are prevailing intermolecular interactions in these solutions, substantially exceed the endoeffects of destruction of self-associated structures of the components. Variations in the H^E values in these systems should mainly be associated with different strengths of the H-bonds formed between these amides and water (Table 2). The donor numbers (*DN*) of DEF and DEA are known to be higher than those of their dimethyl-substituted analogs (see Table 2). Taking into account the correlation between the energies of the HOH...OY hydrogen bonds and the *DN* of OY substance[12] one can assume that the diethylamides of carboxylic acids form stronger hydrogen bonds with water. Correspondingly, one could also expect larger negative H^E values in their aqueous solutions as compared to those of dimethylamides. However, it can be seen in Fig. 1 that for all diethylsubstituted amides the endoeffects of their mixing with water are larger than those for the corresponding dimethylamides.

Table 2. Enthalpies of hydrogen bond water—amide (ΔH/kJ mol^{-1}) at 298.15 K and the donor numbers of amides (*DN*/kcal mol^{-1})

Amide	$-\Delta H$ *	*DN* **	Amide	$-\Delta H$ *	*DN* **
DMF	17.6	26.6	NMP	18.3	27.3
DEF	—	31.0	HMPA	20.1	38.8
DMA	18.7	27.8	DMCA	—	23.0
DEA	—	32.1			

* According to Ref. 6. ** According to Ref. 9.

Table 3. Excess enthalpies of mixtures of chloroform with amides (H^E) at a 2 : 1 ratio of the components and the enthalpies of solution at infinite dilution of amides in hexane ($\Delta_{sol}H^\circ$) at 298.15 K

Amide	$-H^E$	$\Delta_{sol}H^\circ$	Amide	$-H^E$	$\Delta_{sol}H^\circ$
	kJ mol^{-1}			kJ mol^{-1}	
DMF	3.20	13.76±0.11	DMPA	3.16	10.52±0.05
DEF	3.57	12.39±0.19	DEPA	3.42	9.93±0.16
DPF	3.72	11.68±0.20	DPPA	3.96	9.69±0.15
DMA	3.91	13.11±0.15	HMPA	6.71	11.66±0.12
DEA	4.39	11.32±0.04			

A comparison of the enthalpy effects of mixing of water with dimethylamides shows a rapid decrease in the absolute values of H^E in the series DMA > DMF > DMPA. It is obvious that the larger exoeffect of mixing of water with DMA as compared to that of mixing with DMF is due to strengthening the interaction of this amide with water owing to the inductive effect of the additional methyl group on the oxygen atom of the carbonyl group. On the contrary, a further increase in the size of the acidic radical in the DMPA molecule to the size of the ethyl group results in appreciably smaller absolute values of H^E in the H_2O—DMPA system as compared to the expected values.

The literature data on the electron-donor properties of tertiary propionamides are rather contradictory. Thus, the enthalpy of the hydrogen bond between water and DMPA was found to have a larger negative value than that of the hydrogen bond between water and DMA.[13] It follows from Ref. 12 that the donor number of DEPA is larger than that of DEA. This is inconsistent with conclusions drawn in Ref. 14, in which the lower electron-donor ability of dialkylpropionamides as compared to dialkylacetamides was found on the basis of the IR spectra. The published data on the degree of association of pure *N,N*-disubstituted amides[4,5] are also rather contradictory. All this generated a need for additional study of the association and donor properties of the molecules of dialkylamides.

Previously,[15] a correlation between the *DN* and enthalpies of mixing of organic solvents with chloroform in the 1 : 2 ratio has been established. The excess enthalpies of mixtures of the studied amides with chloroform at $X_2 \approx 0.33$ are listed in Table 3. Thermochemical data indicate that the electron-donor abilities of amides of formic, acetic, and propionic acids increase gradually as the size of the alkyl substituent at the N atom successively increases. This tendency is in agreement with the available values of the *DN* for dimethyl- and diethyl-substituted formamides and acetamides (see Table 2). However, a comparison of the H^E values of mixtures of chloroform with amides containing the same substituents at the N atom and a lengthened acidic radical shows that the relative electron-donor abilities of DMA and DEA are higher than those of DMPA and DEPA, respectively. These results are unexpected from the viewpoint of concepts of the stronger electron-donor properties of the ethyl radical as compared to those of the methyl radical.[16] The decrease in the electron-donor properties of propionamides as compared to those of acetamides is explained by the decrease in the conjugation between the p-electron pair of the nitrogen atom and the π-bond of the carbonyl group.[14] In this case the acidic radical acts as an electron-withdrawing substituent. Dimethylchloroacetamide (DMCA) can serve as an example of a substituted amide with the known *DN* (see Table 2). Noteworthy is the abormally high value of the enthalpy of mixing of chloroform with HMPA in a 2 : 1 ratio, which nearly coincides with the data obtained previously.[15] This value exceeds more than twice the absolute value of H^E in a mixture of chloroform with DMF, which corresponds to the latest calorimetric determination[17] of the *DN* of HMPA, which is equal to 50.3 kcal mol^{-1}.

Previously,[18] it has been proposed to estimate the enthalpy of association of pure organic solvents from their enthalpies of solution at an infinite dilution in hexane. For this purpose, we carried out four measurements of the enthalpies of mixing of hexane with each amide in the region of its low concentration ($0.004 < X_2 < 0.035$). The values of the enthalpies of solution of amides ($\Delta_{sol}H = H^E/X_2$) were extrapolated to zero concentration using the relation

$$H^E/X_2 = A + BX_2, \qquad (2)$$

which was successfully tested on more than 50 binary organic systems.[19] Doubled standard deviations of this approximation are given as the errors of the values presented in Table 3. The enthalpies of solution ($\Delta_{sol}H^\circ$) of tertiary amides in hexane, which exhibit a tendency to decrease as both the acidic radical and the substituent at the N atom increase in size (see Table 3), form a sequence DMF > DMA > DEF > DPF > DEA > DMPA > DEPA > DPPA. The data obtained are in agreement with the results reported in Ref. 20, where the association equilibria in disubstituted formamides and acetamides were studied by NMR spectroscopy. It is likely that the decrease in the $\Delta_{sol}H^\circ$ values reflects a decrease in the content of the dipole-dipole dimers because of increasing steric hindrances upon their formation owing to the increasing size of the alkyl substituents.

Thus, despite the increase in the electron-donor properties and decrease in the enthalpies of association (see Table 1—3), the endothermic effects of mixing of diethylamides of carboxylic acids with water are much larger than those of mixing of the corresponding dimethylamides (see Fig. 1). A comparison of the H^E values in mixtures of water with diethylamides with those in mixtures of water with dipropylamides shows that this tendency is retained. An increase in the size of the alkyl groups in the amide molecules is a reasonable explanation of this phenomenon, characteristic of aqueous solutions. As is known, the structure of water is defined by a random network of hydrogen bonds.[1,2] An increase in size or number of the hydrocarbon radicals results in a higher degree of destruction of the network.[2] Additionally, the probability of formation of hydrogen bonds between water and the oxygen atom of the molecule of the organic component decreases as the latter increases in size.[21] The decrease in the electron-donor ability of propionamides in combination with maximum sizes of their hydrocarbon groups favors the minimum exothermic effect of mixing of water with amides. The steric factor is likely the main reason for the close H^E values observed in aqueous solutions of HMPA and DMA (see Fig. 1) despite the lesser electron-donor ability of DMA. However, it follows from a comparison of the data reported previously in Refs. 4, 5, and 7 that the exothermic effects of mixing of DEA and DEF with water in the narrow range of compositions ($0 < X_2 < 0.05$) at 298.15 K are larger than those of mixing of DMA and DMF with water, respectively. The same is also observed for mixtures of water with DMPA and DEPA, but at lesser concentrations of the amides. The formation of solutions with such a high water content occurs without breaking of the network of H-bonds in water.[1] Combination of the effects of hydrophilic and hydrophobic hydration is the reason for the larger exothermic effects in the interaction between amides with larger alkyl radicals and water.

The strong electron-donor properties of the amide molecules and their much larger volumes as compared to those of the water molecule lead to a situation where a fraction of the molecules of the organic component does not form hydrogen bonds with water even in solutions with high water content.[22] In aqueous solutions of HMPA whose molecule has six Me groups, these molecules were found at $X_2 = 0.02$.[22] We calculated the excess partial molar enthalpies of water on the basis of the obtained data on the excess enthalpies of water—amide mixtures:

$$\bar{H}_1^E = H^E_{calc} - X_2(\partial H^E_{calc}/\partial X_2)_{T,p}. \qquad (3)$$

Their values are indirect evidence for distinctions in the hydrate shells of the dissolved amides (Fig. 2). The partial molar excess enthalpies of water are negative if the HMPA content in solutions is less than 1 mol.% and are close to zero in other systems at the above concentrations. The most probable cause for this phenomenon is that the added water is incorporated into the hydrate shells of HMPA, since it is known that the effect of HMPA on the water structure is extended to a distance equal to several diameters of the water molecules in this range of compositions.[1] The decrease in the absolute values of $\bar{H}_1^E$ in other aqueous solutions of amides (see Fig. 2) is due to the smaller surface areas of their alkyl radicals. The partial molar excess enthalpies of water successively take more negative values as the hydrocarbon groups of dialkylamides increase in size and are minimum in the H_2O—DEA system.

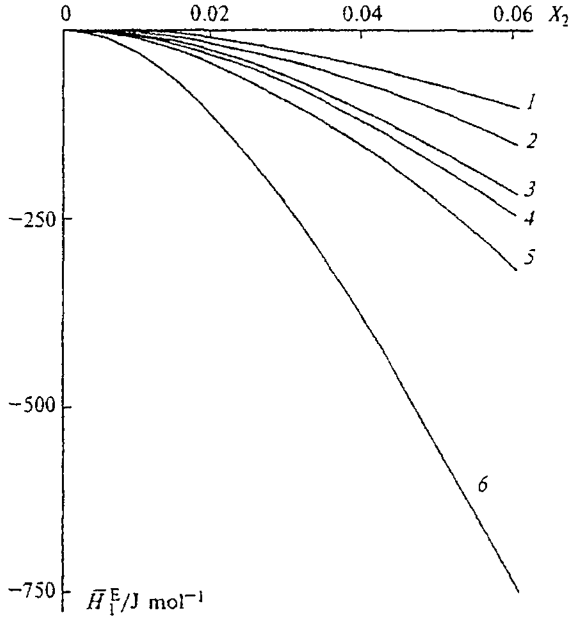

Fig. 2. Excess partial molar enthalpies ($\bar{H}_1^E$) of water at low concentrations of amides (T = 298.15 K) in the systems: H_2O—DMF (*1*), H_2O—DMA (*2*), H_2O—DMPA (*3*), H_2O—DEF (*4*), H_2O—DEA (*5*), and H_2O—HMPA (*6*).

Data on the status of the water molecules in solutions with a high amide content are of great importance for elucidating the nature of intermolecular interactions in the whole range of compositions of the water—amide systems. Since analogous studies are few and nonsystematic, we attempted to reveal a qualitative relation between the enthalpies of solution and solvation of water and the electron-donor properties and chemical structure of amides. The enthalpies of solvation of water ($\Delta_s H^\circ$) were determined from the values of the enthalpies of solution of water at infinite dilution in amides (ΔH_1°); for the systems approximated by the Redlich—Kister polynomial, the ΔH_1° values were calculated from the relation

$$\Delta H_1^\circ = \lim_{X_1 \to 0} (\partial H^E/\partial X_1) = \sum_{n=1}^{p} c_n(-1)^{n-1}. \qquad (4)$$

The calculated values of $\Delta_s H^\circ$ of water in DMF and DMA are in good agreement with the known data.[23] The ΔH_1° values for mixtures of water with DPF, DPA, DEPA, and DPPA were found from the H^E values at $X_2 > 0.9$ using Eq. (2). The largest absolute values of the enthalpies of solution and solvation of water are observed in HMPA (Table 4). However, they only slightly differ from other values when substantial differences between the electron-donor properties of HMPA and tertiary amides are considered. Steric hindrances arising in the formation of the $H_2O \cdot 2HMPA$ complex result in

Table 4. Enthalpies of solution of water in amides at infinite dilution ($\Delta H_1{}^\circ$) and the enthalpies of solvation of water in amides ($\Delta_s H^\circ$) at 298.15 K

Amide	$\Delta H_1{}^\circ$	$\Delta_s H^\circ$	Amide	$\Delta H_1{}^\circ$	$\Delta_s H^\circ$
	kJ mol^{-1}			kJ mol^{-1}	
DMF	−3.7	47.7	DPA	−2.5	46.5
DEF	−1.8	45.8	DMPA	−3.6	47.6
DPF	−0.1	44.1	DEPA	−2.1	46.1
DMA	−5.5	49.6	DPPA	0.1	43.9
DEA	−4.1	48.1	HMPA	−7.7	51.7

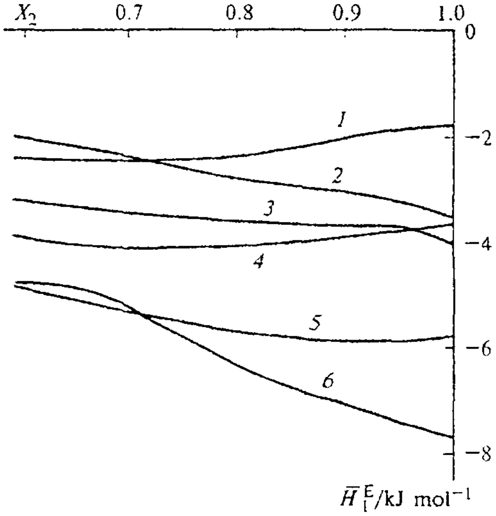

Fig. 3. Excess partial molar enthalpies ($\bar{H}_1{}^E$) of water (T = 298.15 K) in the systems: H_2O—DEF (*1*), H_2O—DMPA (*2*), H_2O—DEA (*3*), H_2O—DMF (*4*), H_2O—DMA (*5*), and H_2O—HMPA (*6*).

a situation where (according to IR spectroscopy data[21,23]) the fraction of free OH groups in the range of highly diluted solutions of water in this solvent amounts to 20%. As has been established recently,[23] the non-specific solvation of water depends slightly on the nature of the solvent, while the effect of the environment is mainly due to distinctions in the energies of heteromolecular H-bonds. It has been shown previously[2,24] that the $\Delta H_1{}^\circ$ values are mainly determined by differences between the energy of the H-bonds in water and those in its complexes with electron donors. Taking into account the substantial difference between the enthalpies of H-bonds in the H_2O—HMPA (see Table 1) and H_2O—H_2O [25] systems, the absolute values of $\Delta H_1{}^\circ$ and $\Delta_s H^\circ$ decrease appreciably due to the endoeffect caused by inability of forming the second H-bond with HMPA by a fraction of water molecules.

As follows from the data listed in Table 4, the increase in the proton-withdrawing properties of tertiary amides, which is observed as the substituents at the N atom increase in size, results in a successive decrease in the absolute values of the enthalpies of solution and solvation of water. It has been shown[21] that the fraction of water molecules bound to the organic solvent by H-bonds through both protons decreases as the number of methyl groups in the organic solvent increases. Since the conditions of the formation of H-bonds deteriorate as the alkyl radicals increase in size, the endoeffect of breaking the H-bonds in water is progressively less compensated for by the exoeffect of the formation of hetero bonds. This is most likely the main reason for the nearly zero enthalpy effect of dissolution of water in DPF and DPPA, which are electron donors of medium strength. On the contrary, water dissolves in less branched amides with a much higher exothermic effect. The decrease in steric hindrances in the case of formation of hydrogen bonds between water and these amides is also confirmed by a nearly full absence of free OH-groups in highly concentrated solutions of compact aprotic solvents (DMA, and DMSO).[21] This is also evidenced by the data on the excess partial molar enthalpies of water (Fig. 3). Appreciable negative values of $\bar{H}_1^E$ in aqueous solutions of DMA at $X_2 > 0.8$ indicate that 1 mol of water added interacts with the solution in this range of compositions in the same way as with pure DMA, so that even the presence of the $H_2O \cdot 2$DMA complexes[23] does not impede the formation of new heteroassociates.

Thus, the increase in the size of *N,N*-alkyl radicals, resulting in the increase in the electron-donor properties of dialkylamides, is accompanied by a decrease in the absolute values of the enthalpy effects of mixing over nearly the whole range of compositions of the water—amide systems. Therefore, distinctions in the endoeffects due to destruction of self-associated structures of tertiary amides are vague. The smallest absolute values of H^E observed in mixtures of water with dialkylpropionamides are due to a decrease in the electron-donor ability and to the maximum sizes of their hydrocarbon groups. The deterioration of conditions of heteroassociation because of the increasing size of the alkyl radicals leads to stratification in the H_2O—DPPA system and to the endoeffect of dissolution of water in this amide.

The authors express their gratitude to N. A. Potkin and O. E. Golubinskii for their help in purifying the reagents and measuring the enthalpies of mixing of amides with hexane.

This work was partially supported by the Russian Foundation for Basic Research (Project No. 96-03-33642a).

References

1. Yu. M. Kessler and A. L. Zaitsev, *Sol'vofobnye effekty. Teoriya, eksperiment, praktika* [*Solvophobic Effects. Theory, Experiment, Practice*], Khimiya, Leningrad, 1989, 312 pp. (in Russian).

2. V. P. Belousov and M. Yu. Panov, *Termodinamika vodnykh rastvorov neelektrolitov* [*Thermodynamics of Aqueous Solutions of Nonelectrolytes*], Khimiya, Leningrad, 1983, 159 pp. (in Russian).
3. A. L. Zaitsev, E. A. Nogovitsyn, A. M. Zaichikov, N. I. Zheleznyak, and G. A. Krestov, *Zh. Fiz. Khim.*, 1991, **65**, 906 [*Russ. J. Phys. Chem.*, 1991, **65** (Engl. Transl.)].
4. A. M. Zaichikov and G. A. Krestov, *Zh. Fiz. Khim.*, 1995, **69**, 389 [*Russ. J. Phys. Chem.*, 1995, **69**, 353 (Engl. Transl.)].
5. A. M. Zaichikov and Yu. G. Bushuev, *Zh. Fiz. Khim.*, 1995, **69**, 1942 [*Russ. J. Phys. Chem.*, 1995, **69**, 1766 (Engl. Transl.)].
6. A. M. Zaichikov, Yu. G. Bushuev, and G. A. Krestov, *J. Therm. Anal.*, 1995, **45**, 687.
7. A. M. Zaichikov and O. E. Golubinskii, *Zh. Fiz. Khim.*, 1996, **70**, 2127 [*Russ. J. Phys. Chem.*, 1996, **70** (Engl. Transl.)].
8. M. V. Kulikov and A. M. Zaichikov, *Izv. Akad. Nauk, Ser. Khim.*, 1997, 1587 [*Russ. Chem. Bull.*, 1997, **46**, 1513 (Engl. Transl.)].
9. C. Reichardt, *Solvents and Solvent Effects in Organic Chemistry*, VCH, Weinheim, 1988.
10. A. M. Zaichikov, E. A. Nogovitsyn, N. I. Zheleznyak, and G. A. Krestov, *Zh. Fiz. Khim.*, 1988, **62**, 3118 [*Russ. J. Phys. Chem.*, 1988, **62** (Engl. Transl.)].
11. P. Assarsson and F. R. Eirich, *Adv. Chem. Ser.*, 1968, **81**, 1.
12. Yu. M. Kessler, Yu. M. Bobrinev, N. A. Borovaya, Yu. P. Boroda, R. Kh. Bratishko, N. V. Vlasova, V. D. Gruba, M. S. Dobrolyubova, R. B. Lyalina, A. I. Mishustin, A. I. Podkovyrin, V. F. Stolypin, and P. S. Yastremskii, in *Problemy sol'vatatsii i kompleksoobrazovaniya* [*Problems of Solvation and Complexation*], IKhTI, Ivanovo, 1978, 31 (in Russian).
13. D. B. Henson and C. A. Swenson, *J. Phys. Chem.*, 1973, **77**, 2401.
14. A. N. Egorochkin and S. E. Skobeleva, *Usp. Khim.*, 1978, **48**, 2216 [*Russ. Chem. Rev.*, 1978, **48** (Engl. Transl.)].
15. V. G. Tsvetkov, B. I. Kozyrkin, L. Ya. Tsvetkova, and I. B. Rabinovich, *Zh. Obshch. Khim.*, 1978, **48**, 2162 [*Russ. J. Gen. Chem.*, 1978, **48** (Engl. Transl.)].
16. A. S. Dneprovskii and T. I. Temnikova, *Teoreticheskie osnovy organicheskoi khimii* [*Theoretical Foundations of Organic Chemistry*], Khimiya, Leningrad, 1991 (in Russian).
17. J.-C. Bollinger, G. Yvernault, and T. Yvernault, *Thermochim. Acta*, 1983, **60**, 137.
18. Yu. A. Aleksandrov, V. G. Tsvetkov, and E. I. Solov'eva, *Zh. Fiz. Khim.*, 1977, **51**, 2137 [*Russ. J. Phys. Chem.*, 1977, **51** (Engl. Transl.)].
19. D. M. Trampe and C. A. Eckert, *J. Chem. Eng. Data*, 1991, **36**, 112.
20. H.-J. Bittrich and D. Kirsch, *Z. Phys. Chem.*, 1975, **256**, 808.
21. A. Burneau, *J. Mol. Liq.*, 1990, **46**, 99.
22. V. I. Korsunskii and Yu. I. Naberukhin, *Zh. Struktur. Khim.*, 1977, **18**, 587 [*J. Struct. Chem. (USSR)*, 1977, **18** (Engl. Transl.)].
23. M. D. Borisover, A. A. Stolov, A. R. Cherkasov, S. V. Izosimova, and B. N. Solomonov, *Zh. Fiz. Khim.*, 1994, **68**, 56 [*Russ. J. Phys. Chem.*, 1994, **68** (Engl. Transl.)].
24. H. Kleeberg, W. A. P. Luck, and H.-Y. Zheng, *Fluid Phase Equil.*, 1985, **20**, 119.
25. G. E. Walrafen, M. R. Fisher, M. S. Hokmabadi, and W.-H. Yang, *J. Chem. Phys.*, 1986, **85**, 6970.

Received January 9, 1997