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Thermodynamic Parameters of Solvation of Nonelectrolytes in Aqueous Solutions with Hydrogen Bond Networks

A. M. Zaichikov[a] **and M. A. Krest'yaninov**[b]

[a] *Ivanovo State University of Chemical Technology,*
pr. Engel'sa 7, Ivanovo, 153000 Russia

[b] *Institute of Solution Chemistry, Russian Academy of Sciences, Ivanovo, Russia*

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Abstract—The contributions from interaction and reorganization to the enthaplies of solvation of polar and nonpolar nonelectrolytes in aqueous solutions of formamide, ethanediol, and 1,2-propanediol, forming 3D-associated structures in the entire composition range, were calculated. The enthalpy terms of the solvation of nonelectrolytes in aqueous solutions of methyl- and dimethylformamide were estimated. The data were compared considering the thermodynamic characteristics of these aqueous systems that we determined previously. It was found that the shape of the concentration dependences of the enthalpies of solvation of nonelectrolytes in all the examined solutions is determined by the reorganization term. The fact that the solvation of nonelectrolytes in water is the most exothermic compared to the aqueous–organic systems under consideration is due to the lowest value of the reorganization term in water, despite the fact that nonelectrolytes interact with water more weakly than with the nonaqueous components.

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It is known that both polar and nonpolar nonelectrolytes are solvated in water more exothermally than in all the other solvents [1, 2]. Many researchers attribute this effect to a specific response of the hydrogen bond network of water to introduction of solutes [3–5]. Other researchers attribute, e.g., the exothermicity of the hydration of alkanes and noble gases to direct nonelectrolyte–water interaction [6–8] or to the combined effect of many factors [9, 10]. The diversity of the existing viewpoints on these problems was reflected in [1, 2, 4]. Along with water, there are other solvents with hydrogen bond networks (formamide, ethanediol, etc.), whose physicochemical parameters are similar to those of water [11]. However, the solvation characteristics of nonelectrolytes in these solvents differ essentially from those in aqueous solutions [1, 2]. Organic solvents with H-bond networks, when mixed with water, form solutions with a 3D hydrogen bond network in the entire composition range [12–14].

This work continues our studies on the thermodynamic properties of aqueous electrolyte solutions with H-bond networks [15–17]. Revealing and applying appropriate characteristics of mixtures allowing efficient estimation of features of inter-molecular interactions in such solutions is still a topical problem of solution chemistry [2, 11]. It was found, e.g., that in the water–formamide system the concentration dependence of the enthalpy of solvation of nonelectrolytes only slightly depends on the concentration of the nonaqueous component [15] and does not have a pronounced extremum typical of aqueous solutions of tertiary amides and, to a lesser extent, secondary amides [18]. The main goal of this study was to elucidate causes of such behavior of the enthalpies of solvation of vairous nonelectrolytes in solvents with hydrogen bond networks and in their aqueous solutions. To this end, we estimated the enthalpy terms of the solvation of nonelectrolytes in the entire range of compositions of aqueous–organic mixtures.

It is known [2, 19, 20] that the enthalpy of solvation is determined by the sum of contributions from the solute–solvent interactions (E_{int}) and solvent reorganization induced by the solute (E_{reorg}):

$$\Delta H_{\text{solv}} = E_{\text{int}} + E_{\text{reorg}}. \qquad (1)$$

Whereas previously the main problems in theoretical calculations of ΔH_{solv} were due to errors in calculating E_{reorg}, now it has been reliably established that

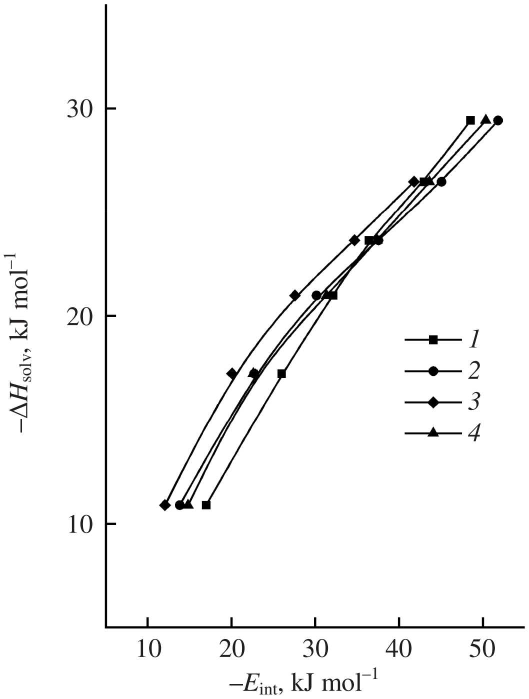

Fig. 1. Correlation between the enthalpies of hydration of alkanes and the energies of alkane–water interaction at 298.15 K: (*1*) our calculation, (*2*) data of [20], (*3*) data of [6], and (*4*) data of [8].

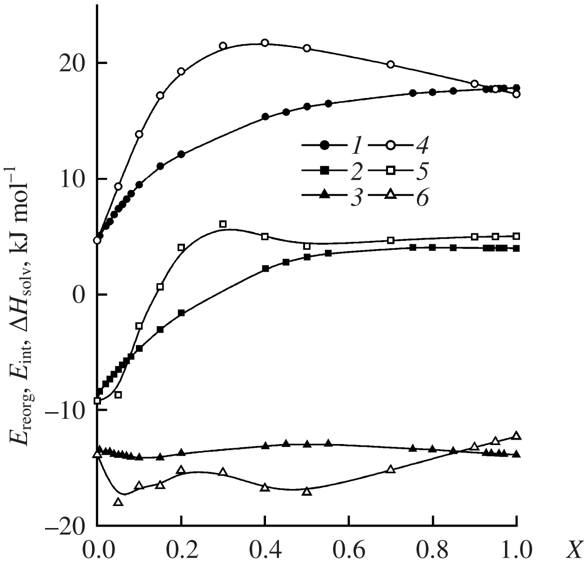

Fig. 2. Contributions from (*1, 4*) reorganization and (*3, 6*) interaction to the (*2, 5*) enthalpies of solvation of argon in aqueous solutions of (*1–3*) formamide and (*4–6*) DMF at 298.15 K.

for aqueous solutions of polar and nonpolar substances $E_{reorg} > 0$ [20, 21]. The same conclusion was confirmed in recent theoretical studies [22, 23] in which it was shown that the term E_{reorg} in solvation of alkanes and noble gases in both aqueous and nonaqueous solutions can be estimated from Eq. (2):

$$E_{reorg} = T(\alpha/\beta)V_s, \tag{2}$$

where V_s is the partial molar volume of the solute, and in the factor $T(\alpha/\beta)$ representing the internal pressure [1, 2] α and β are the coefficients of volume expansion and isothermal compressibility of the solvent, respectively. Calculation of the quantity E_{reorg} including the properties of a solvent [$T(\alpha/\beta)$] and dissolved nonelectrolyte (V_s) would allow, knowing ΔH_{solv}, estimation of E_{int} which cannot be determined experimentally. Figure 1 shows the correlation of the experimental enthalpies of solvation of alkanes in water with the qunantities E_{int} calculated from Eqs. (1) and (2). The enthalpies of solvation of alkanes in water [24] are given, as in [22], in the Ben-Naim standard [25], and the parameters of the individual components, required for the calculation, are taken from [24, 26]. Good agreement of the values obtained with the results of computer simulation [6, 22] and theoretical calculations [8] demonstrates the efficiency of this method for estimating both energy terms of solvation of nonpolar electrolytes in water.

Studies of gas solubilities are used for a long time for comparing the properties of aqueous nonelectrolyte solutions. The papers dealing with the thermodynamics of solvation of argon and ethers [18, 27, 28] show that the shapes of the $\Delta H_{solv}(X)$ dependences for these nonelectrolytes in aqueous solutions of formamide and dimethylformamide (DMF) (here and hereinafter, X is the mole fraction of the organic component) differ essentially. It is interesting to elucidate the causes of these differences, the more so as the previously obtained thermodynamic characteristics of these systems also differ essentially [12]. The contributions E_{reorg} and E_{int}, calculated from Eqs. (1) and (2), to the enthalpies of solvation of argon [27], given in the Ben-Naim standard [25], in aqueous solutions of formamide in DMF are plotted in Fig. 2. The quantities ΔV_s for argon in aqueous solutions of formamide and DMF were calculated using the scale particle model [29], and the required parameters of the components were taken from [2, 30, 31]. Data on the internal pressure in aqueous solutions of these amides were obtained by us previously [12]. The terms E_{reorg} and E_{int} in each system are close in the order of magnitude but opposite in sign. The solvation of argon in solutions with a low content of the nonaqueous component is exothermic, because $|E_{\mathrm{reorg}}| < |E_{\mathrm{int}}|$. This fact agrees with the conclusions made in [6–8] that the exothermicity of solvation of nonpolar nonelectrolytes in water is due to solute–water van der Waals interactions, rather than to enhancement of the interaction of water molecules with each other. At $X > 0.3$ in formamide solutions and $X > 0.17$ in DMF solutions, the argon solvation is already endothermic because $|E_{\mathrm{reorg}}| > |E_{\mathrm{int}}|$. It can be readily seen (Fig. 2) that the shape of the dependences $\Delta H_{\mathrm{solv}}(X)$ is determined by the shape of the dependences $E_{\mathrm{reorg}}(X)$, because the terms E_{int} are approximately constant. In turn, the shape of the dependences $E_{\mathrm{reorg}}(X)$ is mainly determined by the shape of the dependences $T\alpha/\beta(X)$, because V_s with increasing X varies insignificantly. Therefore, apparently, the extremum in the dependence $\Delta H_{\mathrm{solv}}(X)$ for the water–DMF system is due to the maximum in the concentration dependence of $T\alpha/\beta$ at $X = 0.3$ [12], in contrast to the mixtures with formamide where these quantities vary monoton-ically because of greater similarity of the component properties. We believe that the same factors are responsible for different shapes of the dependences $\Delta H_{\mathrm{solv}}(X)$ for the solvation of the $–CH_2–$ fragment of nonelectrolyte molecules in these mixtures [32].

The contributions E_{reorg} and E_{int}, calculated from Eqs. (1) and (2), to the enthalpies of solvation of xenon [33, 34] in aqueous solutions of ethanediol and 1,2-propanediol at 298.15 K are plotted in Fig. 3. The values of V_s for xenon in these solutions were estimated within the framework of the scale particle theory [29], and the component parameters required for the calculation were taken from [22, 35]. Data on the internal pressure in the water–ethanediol system were obtained by us previously [36], and those for aqueous solutions of 1,2-propanediol were taken from [37]. The enthalpies of solvation of xenon in both mixtures are given in the Ben-Naim standard [25]. The exothermicity of solvation of this noble gas in the entire range of compositions of both systems is due to the fact that $|E_{\mathrm{reorg}}| < |E_{\mathrm{int}}|$. Figure 3 shows that the shape of the dependences $\Delta H_{\mathrm{solv}}(X)$, similarly to aqueous solutions of amides, is determined by the shape of the dependence $E_{\mathrm{reorg}}(X)$, because the terms E_{int} vary insignificantly, passing through a shallow minimum. Introduction of an additional alkyl radical into the ethanediol molecule leads to a decrease in the absolute values of E_{int} and in the corresponding enthalpies of solvation of xenon in the water–1,2-propanediol system, because the terms $E_{\mathrm{reorg}}(X)$ in both systems are close. The functions $E_{\mathrm{reorg}}(X)$ and $\Delta H_{\mathrm{solv}}(X)$ in aqueous solutions of 1,2-propanediol pass through a weakly pronounced maximum. For the water–ethanediol system, the maximum of $E_{\mathrm{reorg}}(X)$ is still less pronounced, and the function $\Delta H_{\mathrm{solv}}(X)$ is monotonically ascending. It can be seen from Fig. 3 that xenon interacts with diols more exothermally than with water.

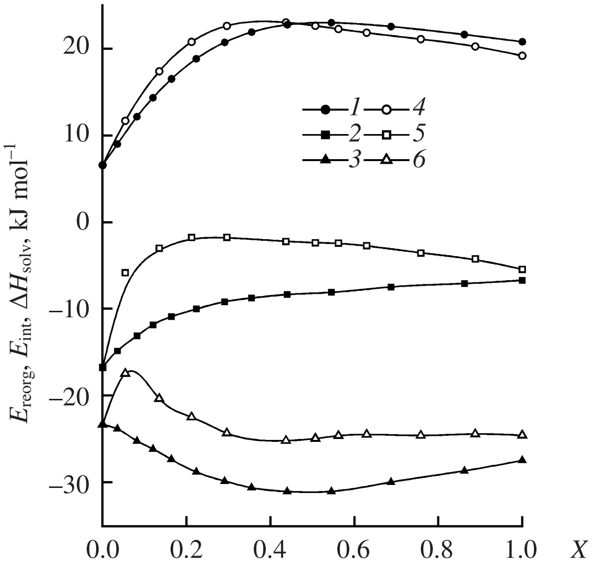

Fig. 3. Contributions from (*1, 4*) reorganization and (*3, 6*) interaction to the (*2, 5*) enthalpies of solvation of xenon in aqueous solutions of (*1–3*) ethanediol and (*4–6*) 1,2-propanediol at 298.15 K.

It was already shown in [8] that alkanes interact with formamide more strongly than with water. It is known that the parameter $T(\alpha/\beta)V_s$ characterizes a decrease in the cohesion energy of a solvent due to solvation of a nonelectrolyte [22]. It was found previously that the parameter $T(\alpha/\beta)$ is mainly determined by short-range intermolecular van der Waals interactions in unit volume of a solvent, which are the most sensitive to changes in the nearest intermolecular surrounding [38]. Dack [38, 39] showed that the contribution of specific interactions can be estimated by taking the cohesion energy density as the full measure of intermolecular interactions and assuming that the internal pressure reflects nonspecific van der Waals

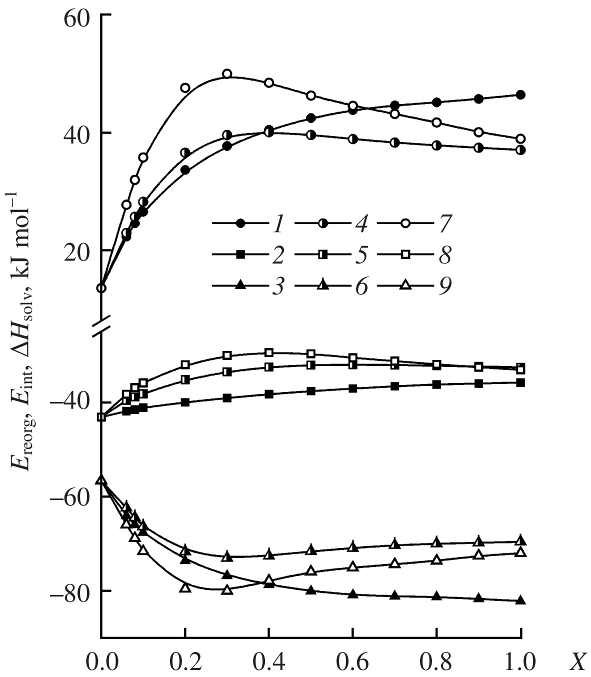

Fig. 4. Contributions from (*1, 4, 7*) reorganization and (*3, 6, 9*) interaction to the (2, 5, 8) enthalpies of solvation of dioxane in aqueous solutions of (*1–3*) formamide, (*4–6*) methylformamide, and (*7–9*) DMF at 298.15 K.

interactions. Hydrogen bonds, being localized in space, act as a significant restricting factor for short-range van der Waals forces [1, 40]. Calculations [38, 39] show that in water the contribution of nonspecific interactions, reaching ~10% of the total intermolecular interaction energy, is abnormally low even in comparison with organic nonelectrolytes with hydrogen bond networks. According to our data, this contribution at 298.15 K is minimal in formamide (~40%) and reaches ~45% in ethanediol and 1,2-propanediol [41]. Therefore, it seems apparent that the higher exothermicity of solvation of noble gases (Figs. 2, 3) in water, compared to organic solvents with hydrogen bond networks, is mainly due to lower values of E_{reorg} in water, which, in turn, is associated with the less perfect structure of hydrogen bond networks in nonaqueous solvents [11]. These reasonings, on the whole, well correlate with the principle of minimum possible change in the openwork structure of water determined by the network of hydrogen bonds [42] under the action of solute species. The same conclusion follows from a direct neutron scattering experiment, which shows that the water structure does not become more openwork around dissolved methane molecules and a weak opposite effect is observed [43].

It can be seen that the first additions of the organic solvents with H-bond networks to water cause the contributions E_{reorg} to increase considerably, with the most pronounced increase observed in the water–DMF system (Fig. 2). One of the causes of this effect is that, on adding DMF to water, the H-bond network of water starts to degrade and becomes fully destroyed at the DMF mole fraction X ~0.3 [30]. Furthermore, the molecules of DMF added to water have a high dipole moment. As a result, as we showed in [41], the van der Waals interactions in solution are sharply enhanced and the specific interactions are weakened, which is not compensated by the formation of water–amide heteroassociates. Addition to water of organic solvents with H-bond networks also leads to destruction of the intrinsic hydrogen bond network in water, but at considerably higher concentrations of the nonaqueous component [44]; furthermore, these water–nonelectrolyte systems are 3D-associated in the entire composition range [45]. This fact, together with the larger similarity of component properties, results in monotonic enhancement of van der Waals interactions in such mixtures with increasing concentration of the nonaqueous component. As a result, the dependences $E_{\mathrm{reorg}}(X)$ are also smooth, without pronounced extrema, especially for the water–formamide system. An increase in the size of the hydrocarbon core in 1,2-propanediol relative to ethanediol results in more pronounced break of the water structure and enhancement of van der Waals interactions. As a result, a smooth maximum appears in the $E_{\mathrm{reorg}}(X)$ dependence for the water–1,2-propanediol system, whereas for the water–ethanediol system this maximum is ill-defined (Fig. 3).

It was shown recently [46, 47] that relationship (2) [22] is applicable to estimating the reorganization contribuiton for not only nonpolar but also polar nonelectrolytes in both aqueous and nonaqueous polar solvents. The same follows from the previously found linear correlations between $\Delta H_{\mathrm{solv}}(X)$ and $E_{\mathrm{int}}(X)$, and also between $E_{\mathrm{int}}(X)$ and $E_{\mathrm{reorg}}(X)$ for various nonelectrolytes in water [48]. The contributions E_{reorg} and E_{int} to the enthalpies of solvation of 1,4-dioxane in aqueous solutions of formamide, methylformamide, and DMF at 298.15 K [28] are plotted in Fig. 4. The required quantities for the calculation performed similarly to the previous calculations were taken from [27, 30], and the internal pressure in the water–methylformamide system was estimated by us in [49]. As in Fig. 2, the dependences $E_{\mathrm{reorg}}(X)$ and $\Delta H_{\mathrm{solv}}(X)$ for

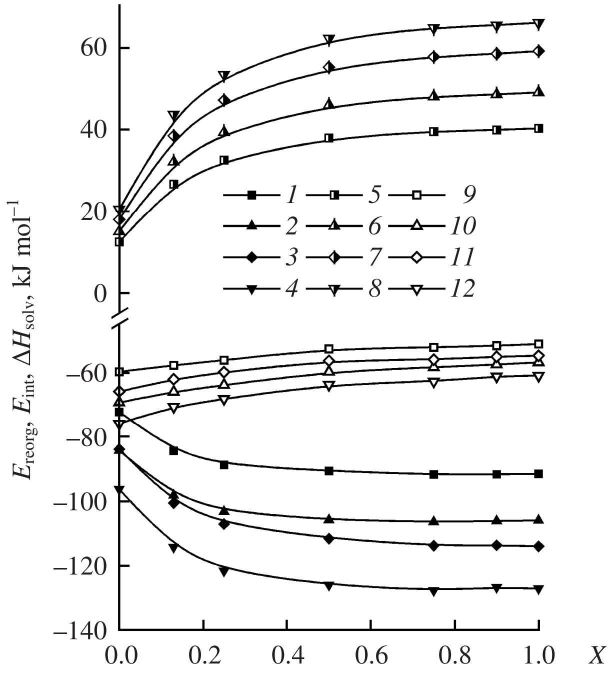

Fig. 5. Contributions from (*5–8*) reorganization and (*1–4*) interaction to the (*9–12*) enthalpies of solvation of dialkylamides in aqueous solutions of formamide at 298.15 K: (*1, 5, 9*) DMF, (*2, 6, 10*) dimethylacetamide, (*3, 7, 11*) diethylformamide, and (*4, 8, 12*) diethylacetamide.

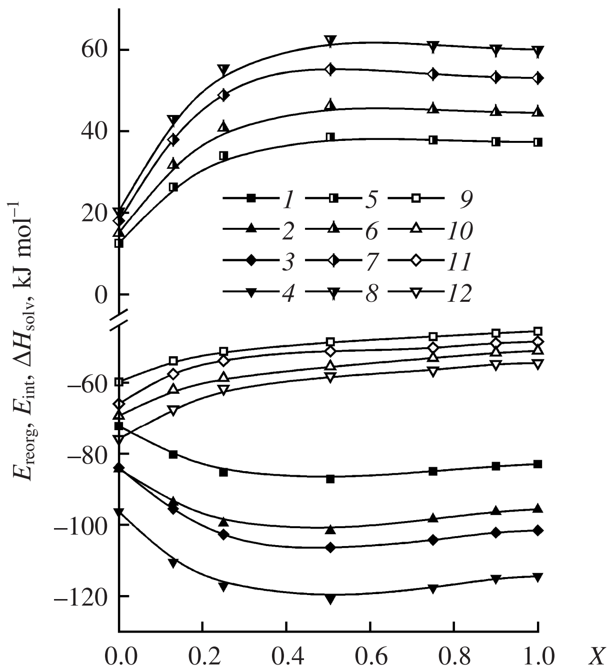

Fig. 6. Contributions from (*5–8*) reorganization and (*1–4*) interaction to the (*9–12*) enthalpies of solvation of dialkylamides in aqueous solutions of ethanediol at 298.15 K: (*1, 5, 9*) DMF, (*2, 6, 10*) dimethylacetamide, (*3, 7, 11*) diethylformamide, and (*4, 8, 12*) diethylacetamide.

aqueous solutions of formamide and DMF show similar trends. In the water–methylformamide system, the hydrogen bond network of water is relatively rapidly broken [49], and the corresponding functions are intermediate between those for the systems with formamide and DMF. This is largely due to the fact that methylformamide, in contrast to formamide which forms network structures, forms only H-bond chains [49]. Figure 4 shows that, despite lower exothermicity of solvation of dioxane in formamide compared to water, this ether interacts with the amide considerably more strongly than with water.

The contributions E_{reorg} and E_{int} to the enthalpies of solvation of N,N-disubstituted formamides and acetamides in aqueous solutions of formamide, ethanediol, and 1,2-propanediol at 298.15 K are plotted in Figs. 5–7. The required data for the calculation, which was performed similarly to the previous cases, were taken from [50], and the functions $\Delta H_{solv}(X)$ were determined by us previously [15–17]. It should be noted that the shape of the curves $\Delta H_{solv}(X)$ in these solutions (Figs. 5–7) is determined by the shape of the dependences $E_{reorg}(X)$, as it was demonstrated above for the same mixtures with noble gases as solutes. The concentration dependences $E_{reorg}(X)$ and $\Delta H_{solv}(X)$ for the water–1,2-propanediol system show a tendency toward appearance of a maximum, most noticeable with diethylacetamide as solute. In all the three aqueous–organic systems, the reorganization contribution to the enthalpy of solvation of diethylformamide is larger compared to dimethylacetamide, owing to larger molecular size. However, stronger proton-acceptor power of diethylformamide and smaller molecular size of dimethylacetamide result in that dimethylacetamide interacts with solutions more weakly than diethylformamide. As a result of summation of these contributions of opposite signs, dimethylacetamide appears to be solvated in all solutions more exothermally compared to diethylformamide. On the whole, in going from DMF to diethylacetamide in the entire composition range the terms E_{reorg} and the exothermic effect of solvation of the disubstituted amides increase. Despite smaller exothermic effect of the solvation of dialkylamides in organic solvents with hydrogen bond networks compared to water, these amides interact with the nonaqueous components considerably more strongly than with water. The same trend is observed with the enthalpies of solvation of alkanols in water and formamide [8].

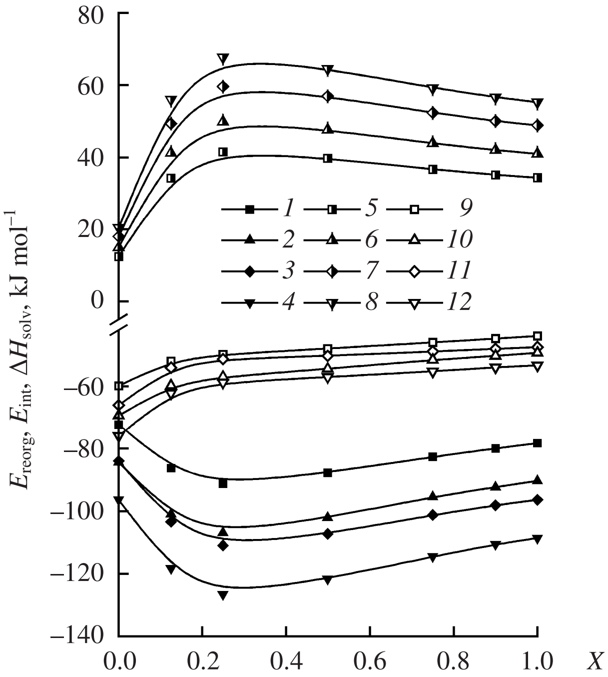

Fig. 7. Contributions from (*5–8*) reorganization and (*1–4*) interaction to the (*9–12*) enthalpies of solvation of dialkylamides in aqueous solutions of 1,2-propanediol at 298.15 K: (*1, 5, 9*) DMF, (*2, 6, 10*) dimethylacetamide, (*3, 7, 11*) diethylformamide, and (*4, 8, 12*) diethylacet-amide.

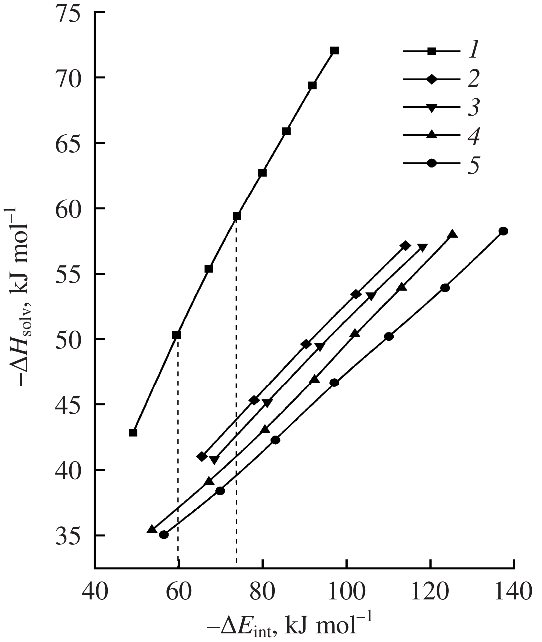

Fig. 8. Correlation between the enthalpies of solvation of alcohols and the energies of alcohol–nonelectrolyte interaction at 298.15 K: (*1*) water, (*2*) dimethylacetamide, (*3*) *N*-methylpyrrolidone, (*4*) ethanediol, and (*5*) formamide.

To elucidate this question, we show in Fig. 8 the correlation of the experimental enthalpies of solvation of alkanols in water, formamide, ethanediol, and amides [32, 51] with the terms E_{int} calculated from Eqs. (1) and (2) at 298.15 K. The required data for the calculation, which was performed similarly to the previous cases, were taken from [24, 50, 52–55]. All the dependences shown in Fig. 8 are well approximated by straight lines, and the parameters of these equations are given in the table together with the correlation coefficients *r* and standard deviations *s*. The correlation between the enthalpies of solvation of alcohols and the energies of alcohol–nonelectrolyte interaction allows prediction of the corresponding parameters if the required experimental or calculated data are lacking. Data for the solvation of alkanols in DMF and methylformamide are not included in Fig. 8 for clarity. The trend that we noted from data on the solvation of polar nonelectrolytes (Figs. 4–7) is manifested here most clearly. Despite the fact that the solvation of alkanols is the most exothermic in water, they interact with water more weakly than with organic components that form hydrogen bond networks and with aprotic amides. Whereas ethanol interacts with four organic solvents more exothermally than with water by only ~10 kJ mol^{-1}, for butanol the difference reaches ~20 kJ mol^{-1} (see dashed lines in Fig. 8). Hence, apparently, the largest exothermic effect of solvation of alkanols in water, compared to nonaqueous components, is due to the smallest value of the reorganization term in water. We believe that the same factor is responsible for the larger exothermicity of solvation of dioxane and dialkylamides in water, which is observed despite the fact that polar nonelectrolytes interact with water more weakly than with the organic solvents under consideration. For nonpolar nonelectrolytes, this may lead even to different signs of the enthalpies of solvation in water and nonaqueous components (Fig. 2).

Thus, within the framework of theoretical concepts [22] we calculated the contributions from interaction and reorganization to the enthalpies of solvation of polar and nonpolar nonelectrolytes in aqueous solutions of formamide, ethanediol, and 1,2-propanediol, 3D-associated in the entire composition range. The

Correlation of the enthalpies of solvation (kJ mol^{-1}) of alkanols in water and organic solvents with the energies (kJ mol^{-1}) of alkanol–nonelectrolyte interaction, $\Delta H_{\mathrm{solv}} = E_{\mathrm{int}}B - A$, at 298.15 K (*r*, correlation coefficient; *s*, standard deviation, kJ mol^{-1})

Solvent	*A*	*B*	*r*	*s*
Water	14.26	0.602	0.998	0.65
Dimethylacetamide	19.36	0.333	0.999	0.17
N-Methylpyrrolidone	18.52	0.328	0.999	0.22
Ethanediol	17.86	0.318	0.999	0.37
Formamide	18.56	0.288	0.999	0.22
DMF	18.84	0.321	0.998	0.37
Methylformamide	17.15	0.348	0.999	0.10

enthalpy terms of solvation of nonelectrolytes in aqueous solutions of methyl- and dimethylformamide were estimated. Their comparison and discussion with the consideration of the thermodynamic characteristics that we obtained previously for these aqueous systems [12, 36, 49, 55] showed that the shape of the concentration dependences of the enthalpies of solvation of nonelectrolytes in all the examined solutions is determined by the reorganization term. It was also found that the largest exothermic effect of solvation of nonelecrolytes in water is due to the smallest value of the reorganization term, despite the fact that nonelectrolytes interact with water more weakly than with nonaqueous components.

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