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# Surface Tensions and Relative Adsorptions in Hydrogen-Bonded Systems

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Liquid–air interfacial tensions of representative alkanol + alkane and alkanol + amine systems measured by the differential capillary rise technique at 298.15 K are reported. The binary systems studied are hexane + ethanol and + 1-propanol, heptane + ethanol and + 1-propanol, and *n*-butylamine + methanol, + ethanol, and + 1-butanol. The excess surface tensions in the alkanol + alkane systems are negative, while in the alkanol + amine systems they are positive. The surface tensions themselves for the alkanol + amine systems, when plotted against mole fraction, exhibit a maximum. The two classes of systems exhibit characteristic patterns in their relative adsorptions. The results are discussed in relation to the bulk properties of these hydrogen-bonded systems.

## Introduction

Over the last few years we have conducted a systematic study of the thermophysical properties of hydrogen-bonded systems. With the exception of a recent work (*1*), our focus so far has been on bulk properties. In the present work we extend this systematic study to the surface tension of two representative classes of hydrogen-bonded systems: 1-alkanol + *n*-alkane and 1-alkanol + *n*-alkylamine systems. Alkanols, which are the common components in these systems, self-associate strongly in both the pure state and in a mixture. Alkanes are inert "solvents" or "diluents", reducing the degree of hydrogen bonding in the mixture. On the other hand, *n*-alkylamines self-associate weakly but cross-associate strongly with 1-alkanols (*2*).

The properties of alkanol + alkane and alkanol + amine mixtures deviate remarkably from ideal solution behavior due to the strong specific intermolecular interactions. The OH–NH interaction, in particular, is one of the strongest hydrogen-bonding interactions, resulting in relatively large negative enthalpies of mixing in alkanol + amine mixtures (*2*). In contrast, the enthalpies of mixing in alkanol + alkane mixtures are positive. It is expected that the interfacial properties of these systems will be strongly influenced by their propensity to form hydrogen-bonding association complexes.

In this work we report surface tensions of seven binary mixtures from these two classes of systems. In parallel we have studied a number of excess quantities of these mixtures including their enthalpies and volumes of mixing (*3*). Thus, we are able to estimate their nonideal behavior and convert the surface tension results to relative adsorptions.

## Materials and Methods

All pure liquids used in this work were pro-analysis grade from Merck. The reported purities, verified by GLC, are the following: methanol, >99.8%; ethanol, >99.8%; 1-propanol, >99.5%; 1-butanol, >99%; *n*-hexane, >99.5%; *n*-heptane, >99%; *n*-propylamine, >99%; *n*-butylamine, >99%. No further purification was attempted.

Pure component properties are reported in Table 1. The mixtures were prepared by mass from the pure components with a precision of ±0.0001 g. Precautions were taken to minimize evaporation losses during the preparation of the mixtures and the subsequent determination of densities and surface tensions.

**Table 1. Density $\rho$ and Surface Tension $\sigma$ of the Pure Components**

| | | $\rho$/(kg·m$^{-3}$) | | $\sigma$/(mN·m$^{-1}$) | |
|---|---|---|---|---|---|
| liquid | $t$/°C | measd | lit. | measd | lit. |
| methanol | 25 | 786.66 | 786.64 (*8*) | 22.12 | 22.10 (*9*) |
| ethanol | 25 | 785.19 | 785.12 (*10*) | 21.76 | 21.68 (*6*) |
| propanol | 25 | 799.63 | 799.69 (*10*) | 23.15 | 23.18 (*6*) |
| butanol | 25 | 805.92 | 805.86 (*10*) | 24.20 | 24.18 (*9*) |
| hexane | 25 | 654.90 | 654.80 (*11*) | 18.42$^a$ | 18.41$^a$ (*6*) |
| heptane | 25 | 679.42 | 679.57 (*10*) | 19.60 | 19.70 (*12*) |
| butylamine | 25 | 732.20 | 733.10 (*13*) | 23.12 | 19.7$^b$ (*14*) |

$^a$ At 20 °C. $^b$ At 41 °C.

The densities, $\rho$, were measured with a vibrating tube densitometer, model DMA 60/602 of Anton Paar. Bidistilled water and air were used as calibrating substances. The temperature in the measuring cell was regulated to 25.00 ± 0.01 °C through a Haake ultrathermostat and measured by a precision digital thermometer, model S1220 of Systemteknik. The estimated error in the density is ±5 × 10$^{-6}$ g·cm$^{-3}$.

The surface tensions, $\sigma$, were measured by the differential capillary rise technique (*4–6*) as described previously (*7*). The precision glass capillaries used had internal radii of $r_1$ = 0.1475 mm and $r_2$ = 0.3851 mm. The differential rise, $\Delta h$, in the capillaries, was measured by a PTI cathetometer with an accuracy of ±0.01 mm. The estimated error in surface tension is ±0.05 mN·m$^{-1}$ in absolute values.

The enthalpies of mixing $H^E$ were measured with a Hart Scientific flow calorimeter, model 7501, with a procedure fully described in ref 3. A detailed presentation of the experimental $H^E$ results for all studied systems will be given in a forthcoming publication.

## Results

The results for the surface tension of the binary liquid mixtures over the whole concentration range are reported in Tables 2 and 3. The reported values are the average of at least three independent measurements. For alkanol + amines the surface tension at intermediate compositions exceeds the surface tension of both pure components. The excess surface

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**Table 2. Surface Tensions $\sigma$ for Alkanes + Alkanols**

| $x_1$ | $\sigma$/(mN·m$^{-1}$) | $x_1$ | $\sigma$/(mN·m$^{-1}$) | $x_1$ | $\sigma$/(mN·m$^{-1}$) | $x_1$ | $\sigma$/(mN·m$^{-1}$) |
|---|---|---|---|---|---|---|---|
| Hexane (1) + Ethanol (2) | | Hexane (1) + Propanol (2) | | Heptane (1) + Ethanol (2) | | Heptane (1) + Propanol (2) | |
| 0.0000 | 21.76 | 0.0000 | 23.15 | 0.0000 | 21.76 | 0.0000 | 23.15 |
| 0.0491 | 20.98 | 0.0235 | 22.83 | 0.0251 | 21.47 | 0.0497 | 22.47 |
| 0.0996 | 20.37 | 0.0501 | 22.40 | 0.0503 | 21.13 | 0.0976 | 21.92 |
| 0.1114 | 20.24 | 0.0966 | 21.82 | 0.0761 | 20.89 | 0.1964 | 21.16 |
| 0.1991 | 19.43 | 0.1955 | 20.68 | 0.1006 | 20.68 | 0.2981 | 20.59 |
| 0.2489 | 19.08 | 0.2964 | 19.76 | 0.1270 | 20.52 | 0.3974 | 20.27 |
| 0.2893 | 18.79 | 0.3446 | 19.40 | 0.1499 | 20.40 | 0.4952 | 20.06 |
| 0.3566 | 18.53 | 0.4093 | 19.06 | 0.1757 | 20.25 | 0.5955 | 19.83 |
| 0.4004 | 18.34 | 0.4471 | 18.87 | 0.2023 | 20.18 | 0.6971 | 19.75 |
| 0.4486 | 18.23 | 0.4971 | 18.61 | 0.2979 | 19.96 | 0.7973 | 19.66 |
| 0.5379 | 18.08 | 0.5942 | 18.33 | 0.3517 | 19.83 | 0.8969 | 19.59 |
| 0.6057 | 18.03 | 0.6522 | 18.14 | 0.4030 | 19.77 | 0.9503 | 19.58 |
| 0.6324 | 17.96 | 0.6951 | 18.11 | 0.5551 | 19.66 | 0.9750 | 19.59 |
| 0.6939 | 17.92 | 0.7490 | 18.00 | 0.5998 | 19.63 | 1.0000 | 19.60 |
| 0.8007 | 17.87 | 0.7990 | 17.94 | 0.6297 | 19.62 | | |
| 0.8513 | 17.83 | 0.8481 | 17.89 | 0.6515 | 19.60 | | |
| 0.9000 | 17.83 | 0.9008 | 17.83 | 0.6978 | 19.60 | | |
| 0.9242 | 17.80 | 0.9491 | 17.80 | 0.7509 | 19.58 | | |
| 0.9502 | 17.79 | 0.9846 | 17.80 | 0.8008 | 19.59 | | |
| 1.0000 | 17.81 | 1.0000 | 17.81 | 0.8501 | 19.59 | | |
| | | 0.9412 | | 0.9412 | 19.57 | | |
| | | | | 1.0000 | 19.60 | | |

**Table 3. Surface Tensions $\sigma$ for 1-Alkanols + *n*-Alkylamines**

| $x_1$ | $\sigma$/(mN·m$^{-1}$) | $x_1$ | $\sigma$/(mN·m$^{-1}$) | $x_1$ | $\sigma$/(mN·m$^{-1}$) |
|---|---|---|---|---|---|
| Methanol (1) + Butylamine (2) | | Ethanol (1) + Butylamine (2) | | Butanol (1) + Butylamine (2) | |
| 0.0000 | 23.12 | 0.0000 | 23.12 | 0.0000 | 23.12 |
| 0.0915 | 23.25 | 0.1007 | 23.20 | 0.0492 | 23.27 |
| 0.2007 | 23.41 | 0.1980 | 23.27 | 0.1004 | 23.42 |
| 0.3010 | 23.52 | 0.2492 | 23.30 | 0.2001 | 23.66 |
| 0.4000 | 23.71 | 0.3994 | 23.33 | 0.2978 | 23.87 |
| 0.4997 | 23.79 | 0.5003 | 23.32 | 0.3960 | 24.02 |
| 0.5999 | 23.85 | 0.6027 | 23.12 | 0.4988 | 24.19 |
| 0.6504 | 23.80 | 0.7111 | 22.86 | 0.5821 | 24.25 |
| 0.7018 | 23.71 | 0.8488 | 22.41 | 0.6999 | 24.31 |
| 0.7773 | 23.38 | 0.9006 | 22.23 | 0.7977 | 24.30 |
| 0.8523 | 23.01 | 1.0000 | 21.80 | 0.8976 | 24.26 |
| 0.9010 | 22.70 | | | 0.9472 | 24.24 |
| 0.9512 | 22.38 | | | 1.0000 | 24.20 |
| 1.0000 | 22.12 | | | | |

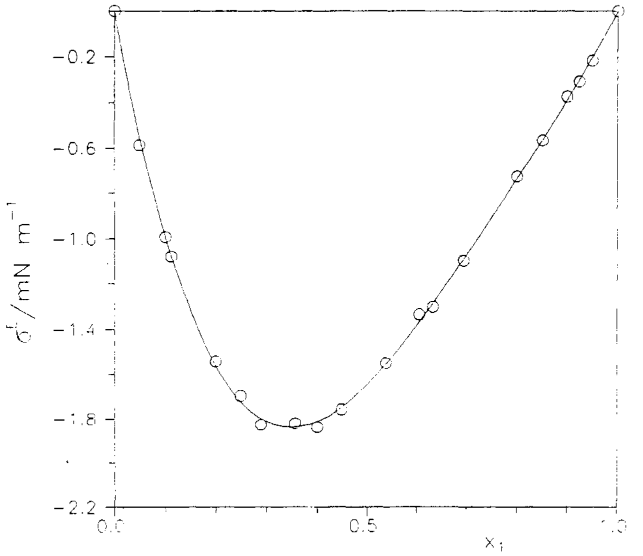

**Figure 1.** Experimental excess surface tension for the system hexane (1) + ethanol (2) at 298.15 K.

**Table 4. Coefficients of Eq 2 and Percent Absolute Average Deviation (% AAD)**

| | hexane (1) + ethanol (2) | hexane (1) + propanol (2) | heptane (1) + ethanol (2) | heptane (1) + propanol (2) |
|---|---|---|---|---|
| $b_0$ | -6.5913 | -5.4116 | -3.9111 | -5.4116 |
| $b_1$ | 4.4769 | 3.1813 | 2.8148 | 3.1813 |
| $b_2$ | -1.7188 | -1.8672 | -3.1122 | -1.8672 |
| $b_3$ | -0.4089 | 0.7133 | 2.3006 | 0.7133 |
| $b_5$ | | -0.8064 | | |
| % AAD | 1.7053 | 1.2656 | 2.8378 | 1.2656 |

tension, $\sigma^E$, of the mixture is defined as

$$\sigma^E = \sigma - x_1\sigma_1 - x_2\sigma_2 \qquad (1)$$

where $x_i$ and $\sigma_i$ are the mole fraction and the surface tension of pure liquid $i$ while $\sigma$ is the surface tension of the mixture of composition $x_i$. Although there is no justification for the ideal surface tension being equal to $x_1\sigma_1 + x_2\sigma_2$, the surface tension deviation $\sigma^E$ of eq 1 is customarily referred to as excess surface tension. In Figures 1–4 are shown the excess surface tensions for the alkanol + alkane mixtures while in Figures 5–7 are shown the corresponding quantities for the alkanol + amine mixtures. These excess quantities are negative over the mole fraction range for the first class of systems but positive for the second class. The curves in these figures were calculated with a Redlich–Kister–Scatchard-type equation:

$$\sigma^E/(\mathrm{mN{\cdot}m^{-1}}) = x_1x_2\sum_{j=0}^{n} b_j(1 - 2x_1)^j \qquad (2)$$

The coefficients $b_j$ along with the average absolute percent deviation are reported in Tables 4 and 5.

Also shown are the enthalpies of mixing of two of the systems. Figure 8 gives $H^E$ for ethanol (1) + heptane (2)

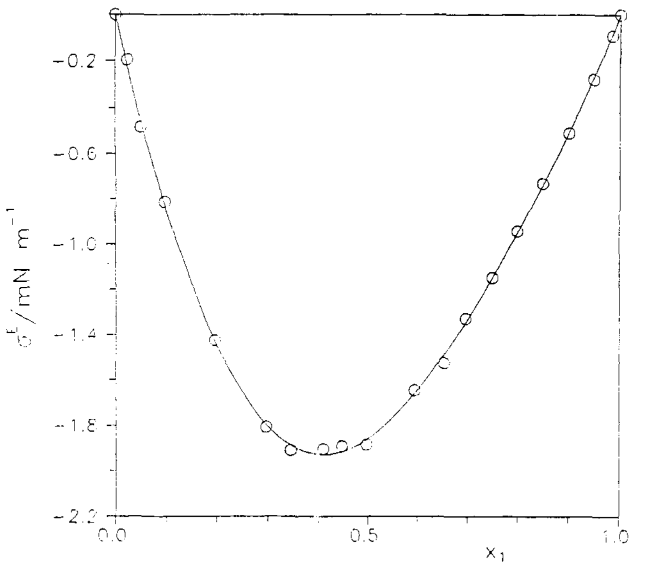

**Figure 2.** Experimental excess surface tension for the system hexane (1) + propanol (2) at 298.15 K.

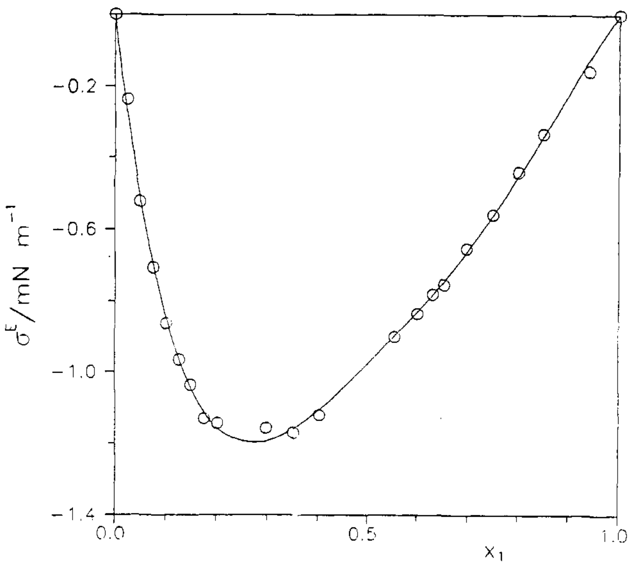

**Figure 3.** Experimental excess surface tension for the system heptane (1) + ethanol (2) at 298.15 K.

**Table 5. Coefficients of Eq 2 and Percent Absolute Average Deviation (% AAD)**

| | methanol (1) + butylamine (2) | ethanol (1) + butylamine (2) | butanol (1) + butylamine (2) |
|---|---|---|---|
| $b_0$ | 4.7707 | 3.3624 | 2.0659 |
| $b_1$ | 3.8708 | 0.5594 | 0.1515 |
| $b_2$ | 0.1353 | -1.9311 | -0.0702 |
| $b_3$ | -3.4425 | 0.0320 | -0.4658 |
| $b_4$ | -1.9250 | 1.8093 | 0.0505 |
| % AAD | 1.4520 | 1.4840 | 1.3050 |

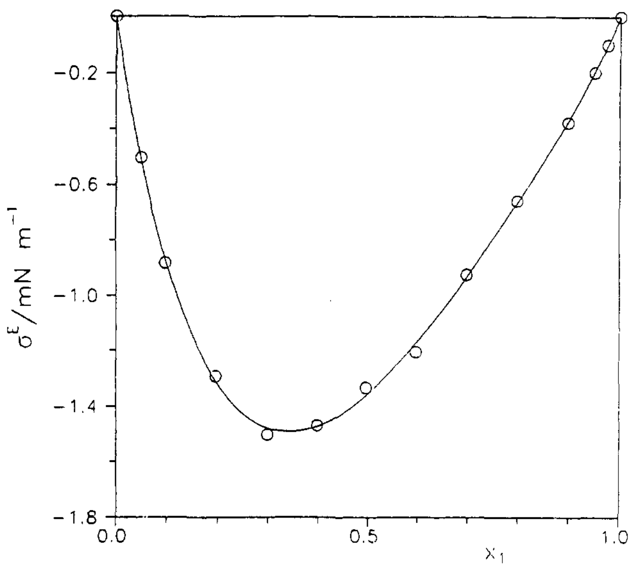

**Figure 4.** Experimental excess surface tension for the system heptane (1) + propanol (2) at 298.15 K.

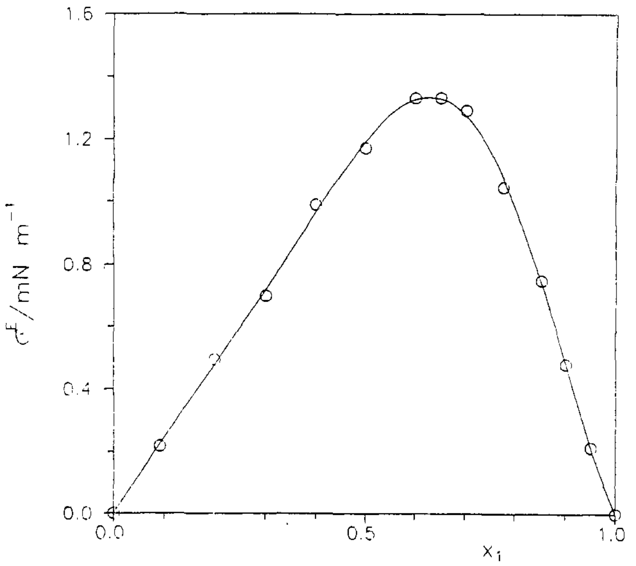

**Figure 5.** Experimental excess surface tension for the system methanol (1) + butylamine (2) at 298.15 K.

**Table 6. Coefficients of Eq 4 and Percent Absolute Average Deviation (% AAD)**

| | hexane (1) + ethanol (2) | hexane (1) + propanol (2) | heptane (1) + ethanol (2) | heptane (1) + propanol (2) |
|---|---|---|---|---|
| $v_0$ | 1.6059 | 0.6006 | 1.8512 | 1.1631 |
| $v_1$ | 0.2272 | 0.7181 | 0.0675 | 0.8172 |
| $v_2$ | 0.3547 | 0.7392 | 0.6939 | 0.2595 |
| $v_3$ | 1.6345 | 0.7413 | 0.5978 | -0.8868 |
| $v_4$ | 1.9471 | | 0.0361 | -0.1951 |
| $v_5$ | -4.6705 | | -0.5687 | 1.7692 |
| % AAD | 1.2926 | 2.1696 | 1.1635 | 3.0166 |

while Figure 9 gives $H^E$ for ethanol (1) + butylamine (2) at 298.15 K. The two classes of systems have $H^E$ values differing not only in their absolute values but also in their sign.

Similar patterns are observed for the volumes of mixing, $V^E$. The latter quantity is defined by

$$V^E = V - x_1V_1 - x_2V_2 = x_1M_1(1/\rho - 1/\rho_1) + x_2M_2(1/\rho - 1/\rho_2) \quad (3)$$

where $V_j$ and $M_j$ are the molar volume and molar mass, respectively, of component $j$. $V^E$ values calculated from the density results have been fitted with a Redlich–Kister–Scatchard-type equation analogous to eq 2, namely,

$$V^E/(\text{cm}^3\cdot\text{mol}^{-1}) = x_1x_2\sum_{j=0}^{n} v_j(1 - 2x_1)^j \quad (4)$$

Coefficients $v_j$ along with the standard deviation of fit are reported in Tables 6 and 7. Contrary to the case of alkanol + alkane systems, the excess volumes for alkanol + amine systems are relatively large and negative.

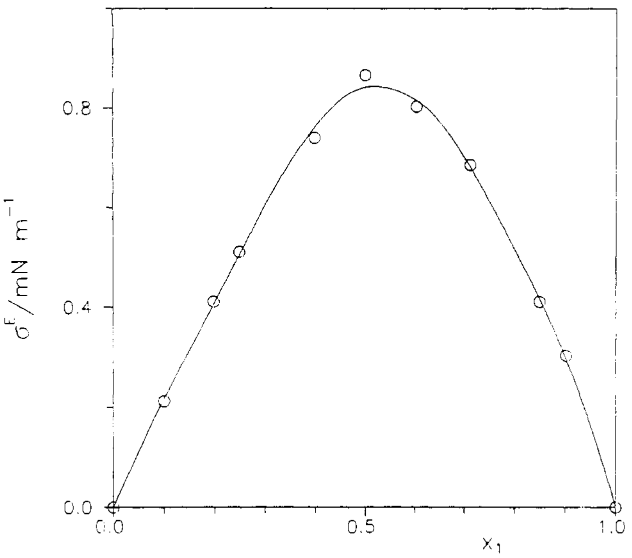

**Figure 6.** Experimental excess surface tension for the system ethanol (1) + butylamine (2) at 298.15 K.

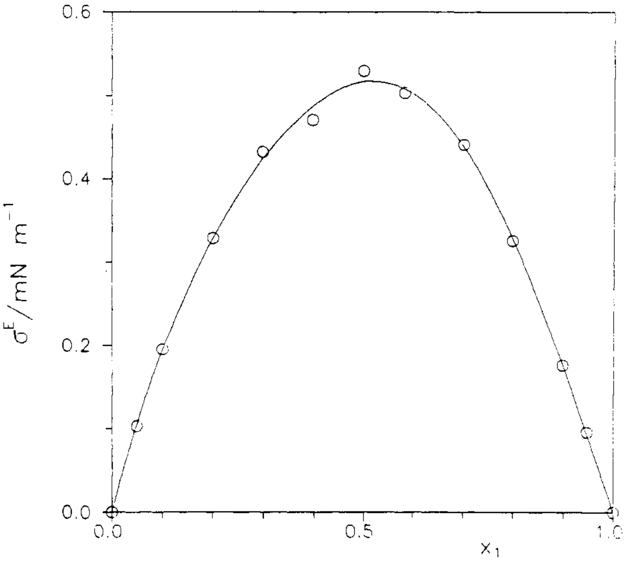

**Figure 7.** Experimental excess surface tension for the system butanol (1) + butylamine (2) at 298.15 K.

**Table 7. Coefficients of Eq 4 and Percent Absolute Average Deviation (% AAD)**

| | methanol (1) + butylamine (2) | ethanol (1) + butylamine (2) | butanol (1) + butylamine (2) |
|---|---|---|---|
| $v_0$ | −6.7038 | −5.1413 | −4.7850 |
| $v_1$ | −0.8393 | −0.9076 | −0.5174 |
| $v_2$ | 1.2197 | −0.2181 | 0.2316 |
| $v_3$ | 0.4995 | 1.8230 | 1.9760 |
| $v_4$ | −0.3936 | −0.2553 | −0.3937 |
| % AAD | 0.3599 | 1.3338 | 1.9478 |

In a binary mixture, the relative adsorption of component 2 at the liquid–air interface is defined by

$$\Gamma_{2,1} = -[\mathrm{d}\sigma/\mathrm{d}\mu_2]_T = -[(\mathrm{d}\sigma/\mathrm{d}x_2)(\mathrm{d}x_2/\mathrm{d}\mu_2)]_T \qquad (5)$$

where $\mu_2$ is the chemical potential of component 2 in the mixture. From eq 5, the calculation of the relative adsorptions requires a knowledge of the chemical potential in the mixture as a function of composition. In refs 2 and 3 we have presented an equation-of-state theory of hydrogen-bonded fluids and correlated the thermodynamic quantities of mixing for systems belonging to the two classes of mixtures studied here. This theory provides the required expression for the chemical potential for the mixtures of our interest (*3*). The pure component parameters are given in ref 2. The hydrogen-bonding parameters for the OH–NH interaction are $E° = -39.3$ kJ·mol$^{-1}$, $S° = -62$ J·K$^{-1}$·mol$^{-1}$, and $V° = -7.0$ cm$^3$·mol$^{-1}$. The physical interaction parameters of the theory are reported in Table 8.

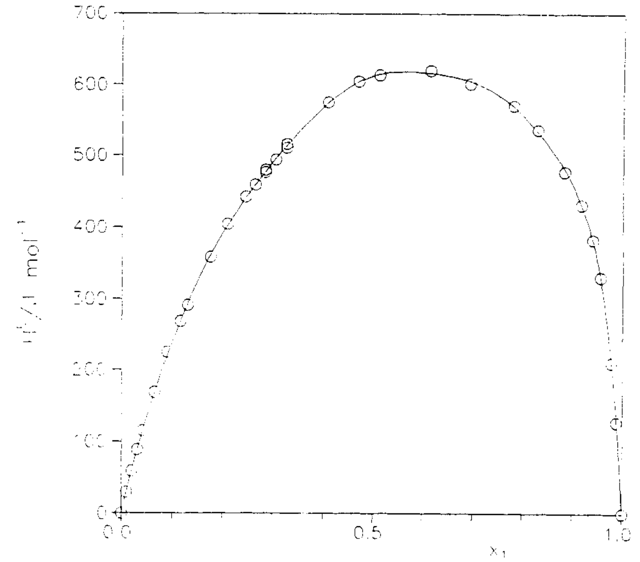

**Figure 8.** Experimental heats of mixing for the system heptane (1) + ethanol (2) at 298.15 K.

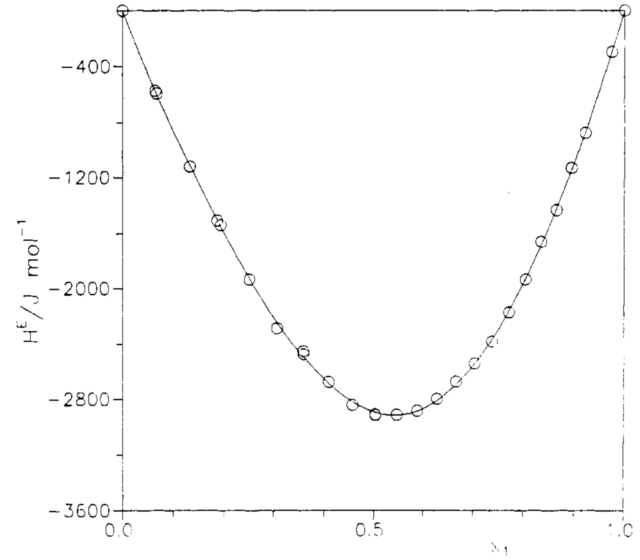

**Figure 9.** Experimental heats of mixing for the system ethanol (1) + butylamine (2) at 298.15 K.

**Table 8. Physical Interaction Parameters of the Lattice Fluid Associated Solutions (LFAS) Model (*2*)**

| system | $\zeta_{12}$ | $\xi_{12}$ | $q_{12}$ |
|---|---|---|---|
| methanol (1) + butylamine (2) | 1.040 | 0.998 | 0.250 |
| ethanol (1) + butylamine (2) | 0.954 | 0.992 | 0.250 |
| butanol (1) + butylamine (2) | 0.936 | 0.992 | 0.250 |
| hexane (1) + ethanol (2) | 1.018 | 0.998 | −0.120 |
| hexane (1) + propanol (2) | 1.020 | 0.992 | −0.145 |
| heptane (1) + ethanol (2) | 1.013 | 0.993 | −0.145 |
| heptane (1) + propanol (2) | 1.016 | 0.994 | −0.120 |

In Figures 10–12 are shown the relative adsorptions at the liquid–air interface of our mixtures. For completeness we have included in Figure 12 the curve for the mixture of 1-propanol with the amine which has been reported previously (*1*). As observed in Figures 10–12 the variation of the relative adsorptions with composition follows distinctively different

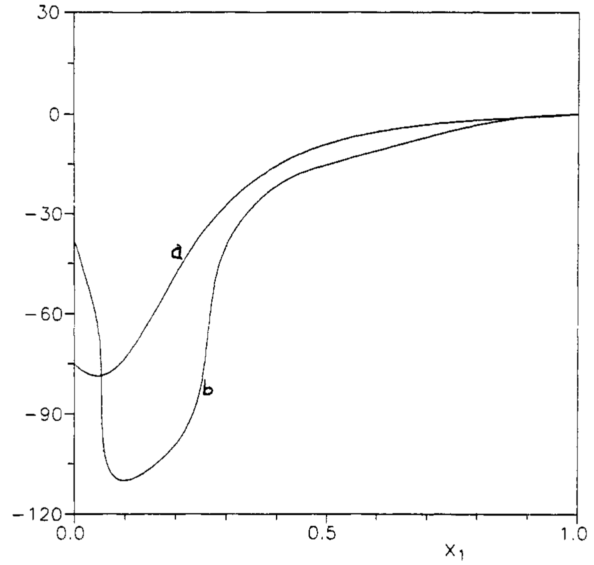

**Figure 10.** Relative adsorption of alkanol at the liquid–air interface in the systems (a) hexane (1) + ethanol (2) and (b) hexane (1) + propanol (2) at 298.15 K.

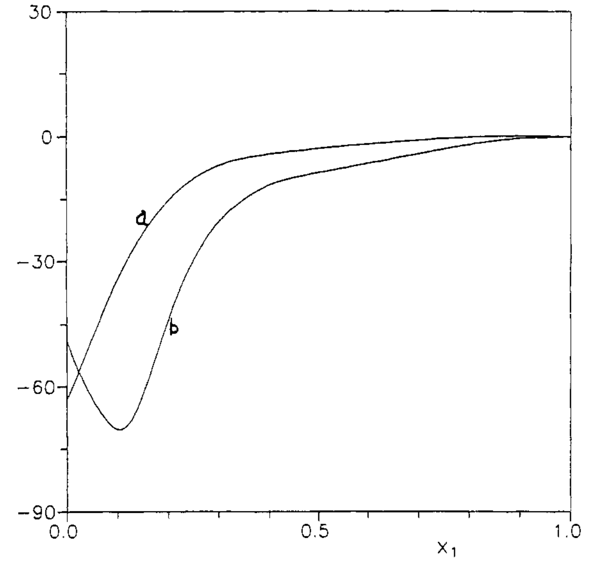

**Figure 11.** Relative adsorption of alkanol at the liquid–air interface in the systems (a) heptane (1) + ethanol (2) and (b) heptane (1) + propanol (2) at 298.15 K.

patterns in the two classes of systems. In alkanol + alkane mixtures the relative adsorption of alkanol is negative with a pronounced minimum in the alkanol-rich region. In the alkanol + amine systems the relative adsorption of alkanol is both positive and negative with a pronounced maximum in the alkanol-rich region and a shallow minimum in the amine-rich region.

## Discussion

The alkanol + alkane mixtures have in general the following properties in common: positive enthalpies and volumes of mixing, positive deviations from Raoult's law, and negative excess surface tensions. The alkanol + amine mixtures have in common the following properties: relatively large negative enthalpies and volumes of mixing, negative deviations from Raoult's law, and positive excess surface tensions. In Table 9 are shown the extreme values for the excess enthalpy, $H^E$, the excess volume, $V^E$, and the excess surface tension, $\sigma^E$, in our systems at 298.15 K. For completeness in the table are included results for the mixtures of propanol with propylamine and butylamine which have been studied earlier (*1*). The distinct differences in these results as well as in the calculated relative adsorptions shown in Figures 10–12 are the outcomes of the distinctively different types of intermolecular interactions in the two classes of systems.

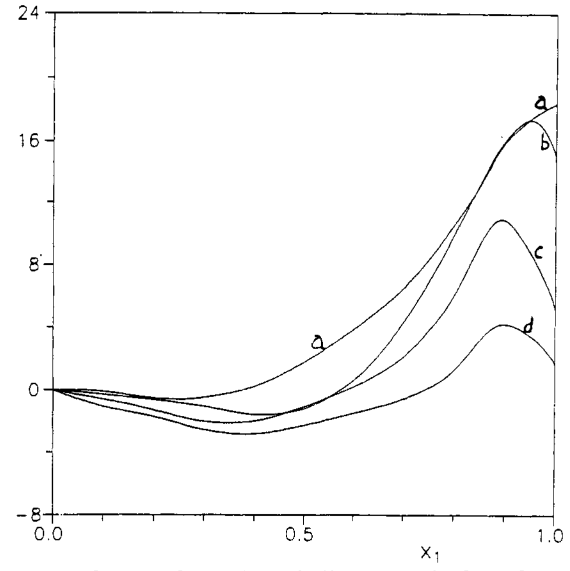

**Figure 12.** Relative adsorption of alkanol at the liquid–air interface in the systems (a) methanol (1), (b) ethanol (1), (c) propanol (1), and (d) butanol (1) with butylamine (2) at 298.15 K.

**Table 9. Extreme Values for the Excess Properties**

| system | $H^E$/ (J·mol$^{-1}$) | $V^E$/ (cm$^3$·mol$^{-1}$) | $\sigma^E$/ (mN·m$^{-1}$) |
|---|---|---|---|
| methanol (1) + butylamine (2) | -3813.9 | -1.676 | 1.33 |
| ethanol (1) + butylamine (2) | -2914.0 | -1.288 | 0.87 |
| butanol (1) + butylamine (2) | -2705.3 | -1.201 | 0.53 |
| hexane (1) + ethanol (2) | 603.7 | 0.406 | -1.84 |
| hexane (1) + propanol (2) | 585.8 | 0.226 | -1.91 |
| heptane (1) + ethanol (2) | 619.5 | 0.463 | -1.17 |
| heptane (1) + propanol (2) | 646.3 | 0.323 | -1.50 |

On the molecular level, both classes of systems interact primarily through hydrogen bonding. The hydrogen-bonding interaction energies, $E°$, for the OH–OH, NH–NH, and OH–NH interactions are −25.1, −13.2, and −39.3 kJ·mol$^{-1}$, respectively (*2*). As a consequence, in the alkanol + alkane mixtures the association complexes formed are "homopolymers" of self-associated alkanols, or [alkanol]$_n$ chains, while in the alkanol + amine mixtures the association complexes are primarily "block copolymers" of the type [alkanol]$_n$–[amine]$_m$ where the amine block is essentially monomeric (*2*). Thus, the alkanol prefers to interact with alkanol in alkanol + alkane systems, but in the case of alkanol + amine systems both the alkanol and the amine prefer to interact with each other rather than with themselves. On the basis of these, one could explain the observed patterns of the relative adsorptions in our systems. Our discussion could be based, equally well, on the surface tension results reported in Tables 2 and 3. The different features of the two classes of systems, however, are more pronounced in Figures 10–12 for the relative adsorptions.

For alkanol + alkane mixtures there are two synergistic factors which force the alkanol to preferentially avoid the interface: the lower surface tension of the alkane and the fact that the hydrogen-bonding interaction can be accomplished more efficiently in the bulk liquid phase rather than in the interface. The role of *n*-alkane in the alkanol-rich region is essentially the breaking of the "structure" of the hydrogen-bonded alkanol chains. This is a highly endothermic process as one can verify from the positive enthalpy of mixing. This explains the tendency to keep the interfacial region rich in alkane rather than in alkanol especially in the alkanol-rich region.

Due to the strength of the OH–NH interaction, in the case of alkanol + amine systems, both compounds tend to prefer the bulk liquid phase rather than the interface where they can accomplish this hydrogen bond more efficiently. Thus, in both diluted regions of the composition, the less abundant component will tend to prefer the bulk, giving rise to s-shaped curves of preferential adsorptions. This is particularly noticeable in the alkanol-rich region (Figure 12). One should contrast Figure 12 with Figures 10 and 11 where the inert alkane in the alkanol-rich region exhibits a strong preference for the interface while the cross-associated amine in the corresponding region in Figure 12 strongly prefers the bulk. This tendency, however, will be moderated by the tendency of the component with the lower surface tension to be preferentially adsorbed at the interface. It is the interplay of these factors which may explain in qualitative terms the behavior observed in Figures 10–12.

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