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Excess Molar Enthalpies of an Alkanol + a Branched Chain Ether at the Temperature 298.15 K

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Excess molar enthalpies H_m^E measured at 298.15 K in a flow calorimeter are reported for 12 mixtures of methanol or ethanol or propan-1-ol or propan-2-ol + bis(1-methylethyl) ether (IPE) or 1,1-dimethylethyl methyl ether (TBME) or 1,1-dimethylpropyl methyl ether (TAME). For all of the systems investigated, except the methanol mixtures, H_m^E is positive over the whole mole fraction range and increases in the order methanol < ethanol < propan-1-ol < propan-2-ol. Small exothermic H_m^E values are obtained for methanol + bis(1-methylethyl) ether or 1,1-dimethylethyl methyl ether or 1,1-dimethylpropyl methyl ether at high methanol mole fractions. The results are explained in terms of the strong self-association of the alkanol and the cross-association of the ether oxygen and the hydroxyl alcohol group.

Introduction

The present work is an extension of our previous work on the thermodynamic properties of binary mixtures containing at least one highly polar component (Letcher and Domańska, 1994; Letcher et al., 1994a,b). Customarily, polar organic substances are divided into two classes: those which can act as proton donors in a hydrogen bond, such as the protic alcohols, the primary, or the secondary amines, and those which cannot, such as the aprotic ethers, ketones, nitriles, and nitro compounds (Grolier et al., 1994). In this work we have considered the mixing of a component from each of these groups. As the protic component we have chosen short chain primary alcohols which show strong self-association, and as the aprotic component, branched chain ethers which exhibit little self-association. In addition, the ethers were chosen with increased branching of the alkyl groups (CH_2 and CH_3). The thermodynamic properties of (1,1-dimethylethyl methyl ether or 1,1-dimethylpropyl methyl ether) have been the subject of considerable interest recently because of their use as "blending agents" in the petrochemical industry (Wang et al., 1992b; Tusel-Langer et al., 1991; Zhu et al., 1994a,b; Antosik and Sandler, 1994).

We have determined H_m^E for methanol or ethanol or propanol-1-ol or propan-2-ol + bis(1-methylethyl) ether (IPE) or 1,1-dimethylethyl methyl ether (TBME) or 1,1-dimethylpropyl methyl ether (TAME). The excess properties are used to obtain information on the strength of the O···HO hydrogen bond interaction.

To our knowledge, of the 12 systems investigated here, only H_m^E measurements at 298.15 K of methanol + TBME (Tusel-Langer et al., 1991), ethanol + TBME (Zhu et al., 1994), propan-1-ol + TBME (Zhu et al., 1994), and propan-2-ol + IPE (Blanks and Prausnitz, 1963) have been reported.

Experimental Section

The alkanols were purified and dried by refluxing with magnesium and iodine followed by distillation (Riddick et al., 1986) and stored in a drybox prior to use. Bis(1-methylethyl) ether was supplied by Riedel-deHaen, 1,1-dimethylethyl methyl ether by Janssen Chimica, and 1,1-dimethylpropyl methyl ether by Aldrich Chemicals. Each of the branched ethers were distilled, dried, and degassed before use as previously described (Letcher and Domańska, 1994). The mole fraction of water in each of the liquids was determined by a Karl Fischer titration to be <0.01 mol %. The liquids were each analyzed by GLC and were found to be 99.8 mol % pure in the worst case (1,1-dimethylpropyl methyl ether.)

A ThermoMetric flow microcalorimeter 2277 was used to determine the molar enthalpies of mixing. All the measurements were made at constant temperature (298.15 ± 0.01 K). The calibration and experimental determinations have been described elsewhere (Letcher and Scoones, 1982; Letcher et al., 1992b). The performance of the calorimeter was checked by measuring H_m^E of the test mixture (benzene + cyclohexane). Agreement with literature results (Battler et al., 1985) was always within 1%.

Results and Discussion

The H_m^E results are given in Table 1, together with the deviations δH_m^E calculated from the smoothing equation

$$\delta H_m^E\ (\text{J}\cdot\text{mol}^{-1}) = H_m^E\ (\text{J}\cdot\text{mol}^{-1}) - x(1-x)\sum_{r=0}^{k} A_r(1-2x)^r \quad (1)$$

The values of the parameters A_r were determined by least squares and are given in Table 2.

H_m^E for the mixtures of ethanol or propan-1-ol or propan-2-ol + IPE or TBME or TAME are positive over the whole alkanol mole fraction range with H_m^E(maximum) decreasing with increased symmetry of the ether side chain in the order TAME > TBME > IPE. For methanol + IPE or TBME or TAME, H_m^E is positive over most of the mole fraction range with the H_m^E(maximum) decreasing in the same order as those for the other alkanols. In the methanol-rich region, a small exothermic effect is found.

For all of the mixtures reported here, H_m^E(maximum) lies between 0.28 and 0.4 mole fraction of the alkanol. The skewness reflects the strong self-association of the alkanol.

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Table 1. Experimental Excess Molar Enthalpies, H_m^E, for an Alkanol + a Branched Chain Ether and the Deviations, δH_m^E, Calculated from Eq 1 and Table 2 at the Temperature 298.15 K[a]

x	$H_m^E/(J \cdot mol^{-1})$	$\delta H_m^E/(J \cdot mol^{-1})$	x	$H_m^E/(J \cdot mol^{-1})$	$\delta H_m^E/(J \cdot mol^{-1})$	x	$H_m^E/(J \cdot mol^{-1})$	$\delta H_m^E/(J \cdot mol^{-1})$
$xCH_3OH + (1-x)[CH_3CH(CH_3)]_2O$								
0.088	150.4	−2.2	0.605	103.3	−0.1	0.846	−25.5	2.2
0.132	189.6	0.2	0.606	101.4	−1.6	0.874	−34.8	2.0
0.227	218.6	2.0	0.702	53.2	3.0	0.928	−37.9	2.7
0.276	215.6	1.6	0.708	43.3	−3.4	0.963	−35.5	−6.5
0.340	202.0	−0.0	0.716	39.2	−3.0	0.976	−26.1	−5.0
0.404	179.4	−4.9	0.756	20.0	1.5	0.986	−12.7	1.0
0.532	142.6	5.2	0.775	6.4	−0.9	0.993	−6.1	0.9
$xCH_3OH + (1-x)CH_3C(CH_3)_2OCH_3$								
0.095	205.9	9.4	0.377	310.7	2.1	0.884	16.1	−3.8
0.132	249.6	9.4	0.493	282.3	2.2	0.910	9.4	3.1
0.141	242.2	−6.8	0.494	280.0	0.1	0.921	0.4	−1.2
0.187	271.2	−10.1	0.592	225.2	−9.9	0.922	0.7	−0.7
0.202	280.2	−9.2	0.745	133.1	5.5	0.936	−5.6	−2.4
0.265	313.5	5.3	0.820	68.8	2.5	0.936	−6.1	1.0
0.347	316.9	5.4						
$xCH_3OH + (1-x)CH_3CH_2C(CH_3)_2OCH_3$								
0.185	300.5	−9.9	0.386	382.0	−2.9	0.856	87.7	4.6
0.236	359.4	10.4	0.422	374.7	−5.1	0.929	31.4	0.4
0.350	390.1	4.8	0.644	261.6	−1.7	0.978	−6.2	−12.7
0.372	386.9	−1.3	0.772	156.7	1.6	0.987	−4.3	−7.9
$xC_2H_5OH + (1-x)[CH_3CH(CH_3)]_2O$								
0.080	240.4	14.5	0.459	414.0	3.7	0.871	74.1	2.3
0.106	295.9	10.2	0.564	348.5	−0.2	0.882	59.1	−3.1
0.215	402.8	−10.6	0.641	289.9	1.7	0.934	28.7	4.0
0.299	442.6	−0.6	0.694	233.2	−7.9	0.935	25.7	1.9
0.366	448.2	7.8	0.789	150.0	0.7	0.985	3.4	0.8
$xC_2H_5OH + (1-x)CH_3C(CH_3)_2OCH_3$								
0.077	214.9	−0.6	0.526	470.1	−5.5	0.766	266.1	1.5
0.120	300.8	−5.8	0.581	448.7	2.6	0.825	198.9	2.6
0.222	460.1	7.9	0.591	440.6	2.4	0.830	183.0	−7.2
0.359	525.6	−1.0	0.678	361.0	1.0	0.881	135.5	4.8
0.451	515.8	−2.0	0.686	350.4	−1.5	0.923	79.4	−2.1
0.525	478.1	−5.5	0.732	304.0	2.2	0.982	14.0	−4.4
$xC_2H_5OH + (1-x)CH_3CH_2C(CH_3)_2OCH_3$								
0.075	263.0	3.3	0.551	492.7	−4.6	0.871	160.2	−4.2
0.190	465.6	−9.3	0.643	428.5	1.9	0.923	107.6	−2.1
0.253	536.7	6.4	0.680	380.0	−11.2	0.934	77.2	−3.6
0.341	561.6	0.6	0.778	300.5	17.0	0.984	17.8	−0.8
$xC_3H_7OH + (1-x)[CH_3CH(CH_3)]_2O$								
0.059	182.6	9.9	0.291	453.8	−2.7	0.635	297.7	−7.2
0.081	225.6	2.5	0.334	460.6	−1.5	0.699	246.1	−2.0
0.116	295.7	3.3	0.376	468.4	10.1	0.767	188.2	1.7
0.139	320.9	−10.2	0.461	428.0	−0.4	0.856	107.2	1.4
0.165	369.7	2.7	0.540	386.9	6.8	0.915	60.6	3.4
0.207	408.4	−1.9	0.590	340.2	−2.0	0.979	6.9	−5.6
0.252	435.6	−5.6						
$xC_3H_7OH + (1-x)CH_3C(CH_3)_2OCH_3$								
0.098	322.9	7.7	0.464	562.6	1.9	0.765	314.2	0.3
0.176	460.3	2.2	0.566	518.2	10.2	0.797	270.4	−3.2
0.248	523.6	−9.3	0.612	464.9	−8.1	0.869	172.7	−3.3
0.315	559.8	−7.5	0.639	455.9	6.0	0.925	110.2	12.5
0.386	583.4	7.4	0.696	385.1	−8.8	0.992	7.0	−3.3
$xC_3H_7OH + (1-x)CH_3CH_2C(CH_3)_2OCH_3$								
0.073	220.8	−5.1	0.361	592.2	−2.0	0.787	306.1	−0.3
0.110	320.5	1.2	0.462	577.8	0.5	0.828	261.7	3.2
0.136	380.9	6.1	0.523	552.7	6.1	0.890	179.1	1.0
0.193	473.9	2.6	0.620	472.0	−2.1	0.937	112.8	4.1
0.235	515.9	−7.8	0.677	420.9	−0.6	0.978	30.1	−11.1
0.282	566.2	2.2	0.735	359.1	−3.9			
$xCH_3C(OH)CH_3 + (1-x)[CH_3CH(CH_3)]_2O$								
0.087	379.4	12.4	0.348	688.3	0.5	0.748	386.3	−9.9
0.131	479.0	−3.1	0.452	672.1	2.7	0.787	326.4	−11.6
0.149	520.2	−0.4	0.540	626.2	5.4	0.837	257.6	0.3
0.232	628.7	−7.5	0.626	547.6	1.1	0.879	192.4	4.6
0.287	670.7	−2.3	0.709	456.3	6.9	0.926	120.6	10.2
$xCH_3C(OH)CH_3 + (1-x)CH_3C(CH_3)_2OCH_3$								
0.063	298.6	5.3	0.393	742.3	−6.4	0.631	682.8	−3.3
0.126	471.8	−3.6	0.417	747.4	−4.8	0.645	679.4	−6.2
0.168	547.6	−7.8	0.445	747.0	−0.6	0.684	625.7	−2.2
0.227	640.5	7.2	0.515	739.9	−6.9	0.866	270.2	−5.7
0.343	712.8	−3.5	0.614	692.1	−9.0	0.871	259.5	−4.2

Table 1 (Continued)

x	H^E_m/(J·mol^{-1})	δH^E_m/(J·mol^{-1})	x	H^E_m/(J·mol^{-1})	δH^E_m/(J·mol^{-1})	x	H^E_m/(J·mol^{-1})	δH^E_m/(J·mol^{-1})
			xCH$_3$C(OH)CH$_3$ + (1 − x)CH$_3$CH$_2$C(CH$_3$)$_2$OCH$_3$					
0.095	400.1	13.1	0.358	738.3	2.2	0.678	577.4	−6.4
0.117	453.1	5.2	0.410	753.4	6.7	0.680	587.2	4.3
0.174	565.5	−2.2	0.460	754.2	−9.4	0.755	458.7	−5.7
0.206	603.4	−13.8	0.566	700.0	0.0	0.819	351.0	9.3
0.274	681.3	−8.2	0.640	625.8	−6.4	0.878	220.7	0.2

[a] The ethers are bis(1-methylethyl) ether, [CH$_3$CH(CH$_3$)$_2$]$_2$O, 1,1-dimethylethyl methyl ether, [CH$_3$C(CH$_3$)$_2$OCH$_3$], and 1,1-dimethylpropyl methyl ether, [CH$_3$CH$_2$C(CH$_3$)$_2$OCH$_3$].

Table 2. Coefficients A_r for Eq 1 and Standard Deviations σ^a for an Alkanol + a Branched Chain Ether Obtained from the Data in Table 1

branched ether	A_0	A_1	A_2	A_3	σ/(J·mol)
		CH$_3$OH			
[CH$_3$CH(CH$_3$)]$_2$O	602.4	785.4	129.6	1009.7	3.3
CH$_3$C(CH$_3$)$_2$OCH$_3$	1110.4	736.3	129.4	938.0	6.5
CH$_3$C(CH$_3$)$_2$OCH$_3$	1414.8	901.5	6.1	288.3	8.0
		CH$_3$CH$_2$OH			
[CH$_3$CH(CH$_3$)]$_2$O	1556.5	1123.1	374.7	711.6	8.3
CH$_3$C(CH$_3$)$_2$OCH$_3$	1992.4	998.7	137.5	157.5	4.5
C$_2$H$_5$C(CH$_3$)$_2$OCH$_3$	1977.8	998.2	147.4	131.7	12.4
		CH$_3$(CH$_2$)$_2$OH			
[CH$_3$CH(CH$_3$)]$_2$O	1625.4	1225.2	347.2	190.8	5.9
CH$_3$C(CH$_3$)$_2$OCH$_3$	2183.7	935.7	511.5	566.5	8.2
C$_2$H$_5$C(CH$_3$)$_2$OCH$_3$	2240.0	1091.5	490.7	−284.6	5.2
		CH$_3$CH(OH)CH$_3$			
[CH$_3$CH(CH$_3$)]$_2$O	2585.6	1137.6	803.2	951.5	7.8
CH$_3$C(CH$_3$)$_2$OCH$_3$	2995.4	198.5	599.7	2008.0	7.8
C$_2$H$_5$C(CH$_3$)$_2$OCH$_3$	2938.3	706.9	419.1	1339.5	8.7

[a] $\sigma = [\sum(H^E_m(\text{exp}) - H^E_m(\text{calcd}))^2/(n - k)]^{1/2}$, where n is the number of experimental points.

The skewing becomes less prominent in the order methanol > ethanol > propan-1-ol > propan-2-ol for each of the ethers investigated.

Of the 12 mixtures reported here, 4 have been reported elsewhere. Our H^E_m(maximum) results for x(methanol) + (1 − x)TBME of 282.3 J·mol^{-1} at x = 0.493 is 1.3 J·mol^{-1} less than the results reported by Tusel-Langer et al. (1992). Our results of x(ethanol) or x(propan-1-ol) + (1 − x)TBME at x = 0.4 (approximate maximum values) are 520 and 580 J·mol^{-1}, respectively, while the results reported by Zhu et al. (1994a−c) are 514 and 553 J·mol^{-1}, respectively. Our H^E_m(maximum) result of x(propan-2-ol) + (1 − x)IPE at x = 0.35 is 688 J·mol^{-1} while that of Blanks and Prausnitz (1963) is 682 J·mol^{-1} at x = 0.46. To confirm our technique and method, we have repeated H^E_m for a number of reliable x(benzene) + (1 − x)cyclohexane results in the literature (Christiansen et al., 1988), and in all cases our results are within 5 J·mol^{-1} (or 2%) of those reported. We have also repeated the four systems done by other workers at least twice.

The values of H^E_m(maximum) are less positive than those for the corresponding n-alkane mixtures for all of the alkanols investigated in this work (Tusel-Langer et al., 1991; Brown and Ziegler, 1979; Wang et al., 1992a). For example H^E_m(maximum) for x(methanol) + (1 − x) heptane is 454 J·mol^{-1} at x = 0.29, while for x(methanol) + (1 − x) IPE it is 219 J·mol^{-1} at x = 0.23. This suggests that the H^E_m reported here depends on the balance between two opposing contributions: (i) a positive term from the rupture of alkanol−alkanol hydrogen bonds and (ii) a negative term from the formation of −OH··O{CH(CH$_3$)$_2$}$_2$ or −OH··O(CH$_3$)C(CH$_3$)$_3$ or −OH··O(CH$_3$)C(CH$_3$)$_2$C$_2$H$_5$ hydrogen-bonded complexes. These contributions can be semiquantitatively analyzed for H^E_m for each mixture by use of the following expression (Villamanan et al., 1982; Diogo et al., 1993):

$$h^{\text{int}}_{\text{OH-O}} = H^E_{\text{max}} - h_{\text{O}} - h_{\text{OH}} \qquad (2)$$

where h_{OH}, h_{O}, and $h^{\text{int}}_{\text{OH-O}}$ are, respectively, from hydrogen bonds between alcohol molecules, from the enthalpic contributions from ether−ether interactions, and from hydrogen bonds between alcohol and ether molecules.

The values of H^E_{max} h_{OH}, h_{O}, and $h^{\text{int}}_{\text{OH-O}}$ for the different mixtures are shown in Table 3. For a given alcohol (j = 1, 2, or 3), $h^{\text{int}}_{\text{OH-O}}$ decreases in the sequence TAME < IPE <

Table 3. Excess Molar Enthalpies^a

	H^E_m(maximum)/(J·mol^{-1})						
ether	H^E_m(max)	reference	h_{OH}	reference	h_{O}	reference	($H^E_m - h_{\text{OH}} - h_{\text{O}}$)
				CH$_3$OH			
[CH$_3$CH(CH$_3$)]$_2$O	219	this work	454	Tusel-Langer et al. (1991)	240	Christiansen et al. (1982; 1988)	−475
CH$_3$C(CH$_3$)$_2$OCH$_3$	250	this work	454	Tusel-Langer et al. (1991)	383	Tusel-Langer et al. (1991)	−587
CH$_3$CH$_2$C(CH$_3$)$_2$OCH$_3$	390	this work	454	Tusel-Langer et al. (1991)	268	Zhu et al. (1994b)	−332
				C$_2$H$_5$OH			
[CH$_3$CH(CH$_3$)]$_2$O	448	this work	610	Brown and Ziegler (1979)	240	Christiansen et al. (1982; 1988)	−402
CH$_3$C(CH$_3$)$_2$OCH$_3$	526	this work	610	Brown and Ziegler (1979)	383	Tusel-Langer et al. (1991)	−467
CH$_3$CH$_2$C(CH$_3$)$_2$OCH$_3$	562	this work	610	Brown and Ziegler (1979)	268	Zhu et al. (1994b)	−316
				C$_3$H$_7$OH			
[CH$_3$CH(CH$_3$)]$_2$O	468	this work	580	Wang et al. (1992a)	240	Christiansen et al. (1982; 1988)	−352
CH$_3$C(CH$_3$)$_2$OCH$_3$	583	this work	580	Wang et al. (1992a)	383	Tusel-Langer et al. (1991)	−380
CH$_3$CH$_2$C(CH$_3$)$_2$OCH$_3$	592	this work	580	Wang et al. (1992a)	268	Zhu et al. (1994b)	−256
				CH$_3$CH(OH)CH$_3$			
[CH$_3$CH(CH$_3$)]$_2$O	688	this work	587	Brown and Ziegler (1979)	240	Christiansen et al. (1982; 1988)	−139
CH$_3$C(CH$_3$)$_2$OCH$_3$	747	this work	587	Brown and Ziegler (1979)	383	Tusel-Langer et al. (1991)	−223
CH$_3$CH$_2$C(CH$_3$)$_2$OCH$_3$	754	this work	587	Brown and Ziegler (1979)	268	Zhu et al. (1994b)	−101

[a] $H^E_m(\text{max}) = H^E_m(\text{maximum})\{C_jH_{2j-1}OH + RCOR'\}$; $h_{\text{OH}} = H^E_m(\text{maximum})\{C_jH_{2j-1}OH + C_6H_{14}$ or $C_7H_{16}\}$ for j = 1, 2, or 3 and $h_{\text{O}} = H^E_m(\text{maximum})\{RCOR' + C_6H_{14}\}$.

TBME. The negative value indicates a reasonably strong association between the alkanol and the ether.

The ERAS (extended real associated solution) model of Heintz et al. (1985) will be fitted to the H_m^E data presented here. Details of this treatment will be reported when measurements of V_m^E, α_p (isobaric thermal coefficient), and κ_T (isothermal expansion coefficient) for the same set of mixtures have been determined. The α_p and κ_T physical properties are being measured in Professor Ahluwalia's laboratory in New Delhi by one of the authors (P.U.G.).

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