Excess Molar Enthalpies of Binary and Ternary Mixtures for 1-Propanol + 3-Pentanone + 1-Hexanol or + 1-Heptanol at 298.15 K

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Excess molar enthalpies at the temperature 298.15 K were measured for 1-propanol + 3-pentanone + 1-hexanol or + 1-heptanol, for 1-propanol + 1-hexanol or + 1-heptanol, and for 3-pentanone + 1-hexanol or + 1-heptanol, using a Calvet microcalorimeter. The experimental results, except for the two binary alcohol–alcohol mixtures, agreed quite well with those calculated by using the Nitta–Chao model.

Introduction

In previous works (*1*, *2*) we determined the excess molar enthalpies of $CH_3(CH_2)_2OH$ (1) + $CH_3CH_2COCH_2CH_3$ (2) + $CH_3(CH_2)_nCH_3$ (3) (n = 4–6) at the temperature 298.15 K. We report here excess molar enthalpies at the temperature 298.15 K and normal atmospheric pressure of $CH_3(CH_2)_2OH$ (1) + $CH_3CH_2COCH_2CH_3$ (2) + $CH_3(CH_2)_nOH$ (3) (n = 5, 6), and of the corresponding binary mixtures. Enthalpy changes were measured using a Calvet microcalorimeter. A variable polynomial has been fitted to each set of experimental results. The ternary results have been fitted by means of an equation proposed by Nagata and Tamura (*3*). The excess molar enthalpies were used to test the group-contribution model of Nitta–Chao (*4*) which is based on the Carnahan–Starling hard-sphere equation of state (*5*).

Experimental Section

The chemical substances employed were supplied by Fluka, and subjected to no further purification other than being dried with Union Carbide 0.4-nm molecular sieves and degassed. The mole-fraction purities were for 1-propanol puriss >99.5 mass %, for 3-pentanone puriss >99.5% mass %, for 1-hexanol puriss >99.5 mass %, and for 1-heptanol puriss >99.5 mass %.

All experimental measurements were carried out using a Calvet microcalorimeter with a calorimeter-cell volume of approximately 10 cm^3 and equipped with a device allowing operation in the absence of a vapor phase. Calibration was performed electrically using a Setaram EJP 30 stabilized current source. Details of the employed procedure have been published previously (*6*, *7*). The precision of the excess molar enthalpies is estimated as better than $\pm 0.01 H^E_m$.

Three experimental runs were carried out for ternary mixtures formed by adding 1-hexanol or 1-heptanol to a binary mixture of 1-propanol (1) + 3-pentanone (2). A ternary mixture may be considered as a pseudobinary mixture composed of one binary mixture and 1-hexanol or 1-heptanol (3). The ternary excess molar enthalpy at x_1, x_2, and x_3 can be given as

$$H^E_{123,m} = H^E_{m,\psi} + (1 - x_3) H^E_{12,m} \quad (1)$$

where $H^E_{m,\psi}$ is the observed excess molar enthalpy for the pseudobinary mixture and $H^E_{12,m}$ is the binary excess molar enthalpy of the 1-propanol + 3-pentanone. Values of $H^E_{12,m}$ at three mole fractions were interpolated by using a spline-fit method. Equation 1 does not involve any approximation (*8*).

Table I. Experimental Excess Molar Enthalpies H^E_m at the Temperature 298.15 K

x	H^E_m/(J·mol^{-1})	x	H^E_m/(J·mol^{-1})	x	H^E_m/(J·mol^{-1})
	x $CH_3(CH_2)_2OH$ + (1 − x) $CH_3(CH)_5OH$				
0.1149	23.0	0.4751	63.0	0.7301	56.3
0.1950	36.8	0.5013	63.8	0.8167	45.3
0.2690	47.0	0.5375	64.2	0.8702	35.2
0.3386	54.6	0.5542	64.3	0.9211	23.8
0.3750	57.6	0.5948	64.0		
0.4281	61.0	0.6580	61.6		
	x $CH_3(CH_2)_2OH$ + (1 − x) $CH_3(CH)_6OH$				
0.0818	27.5	0.4419	100.4	0.6497	97.9
0.1568	49.4	0.4650	101.9	0.7049	91.2
0.2220	66.2	0.5103	103.3	0.7316	86.9
0.2480	71.9	0.5416	103.8	0.7924	73.4
0.3284	87.4	0.5654	103.2	0.8472	58.9
0.3923	96.1	0.6050	101.3	0.9138	36.2
	x $CH_3CH_2COCH_2CH_3$ + (1 − x) $CH_3(CH)_5OH$				
0.0821	427.3	0.4809	1428.2	0.8261	986.6
0.1356	660.4	0.5248	1449.1	0.8442	914.5
0.1881	847.2	0.5440	1450.4	0.8905	707.1
0.2426	1012.9	0.5781	1444.8	0.9238	524.2
0.2859	1122.0	0.6253	1419.9		
0.3303	1217.5	0.6652	1377.7		
0.3594	1271.8	0.7192	1289.1		
0.4135	1360.6	0.7734	1157.0		
	x $CH_3CH_2COCH_2CH_3$ + (1 − x) $CH_3(CH)_6OH$				
0.0838	452.6	0.4783	1498.8	0.7946	1186.9
0.1355	688.1	0.5080	1516.7	0.8770	851.4
0.1660	810.7	0.5473	1529.1	0.9185	619.2
0.2244	1004.8	0.5704	1531.0	0.9536	382.1
0.2712	1142.9	0.6024	1520.7		
0.3300	1282.2	0.6516	1484.1		
0.3886	1390.2	0.7075	1398.3		
0.4212	1432.4	0.7665	1267.9		

Results and Discussion

Excess molar enthalpies of the binary mixtures are listed in Table I. A variable-degree polynomial of the form

$$H^E_{ij,m}/(\text{J·mol}^{-1}) = x_i x_j \sum_{m=0}^{n} A_m (x_i - x_j)^m \quad (2)$$

was fitted to the binary systems using a least-squares method.

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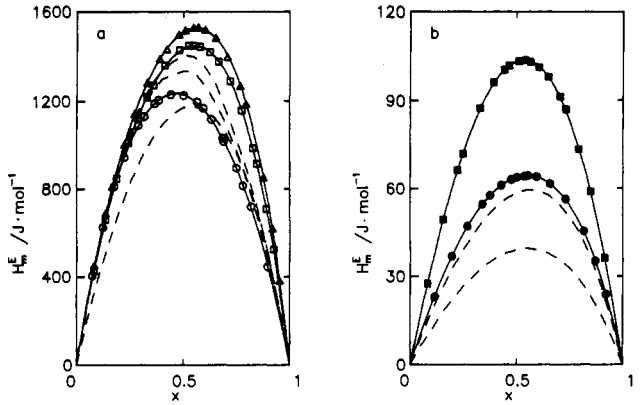

Figure 1. Excess molar enthalpies for (○) x $CH_3(CH_2)_2OH$ + $(1-x)$ $CH_3CH_2COCH_2CH_3$ (*1*), (□) x $CH_3CH_2COCH_2CH_3$ + $(1-x)$ $CH_3(CH_2)_5OH$, (△) x $CH_3CH_2COCH_2CH_3$ + $(1-x)$ $CH_3(CH_2)_6OH$, (●) x $CH_3(CH_2)_2OH$ + $(1-x)$ $CH_3(CH_2)_5OH$, and (■) x $CH_3(CH_2)_2OH$ + $(1-x)$ $CH_3(CH_2)_6OH$ at the temperature 298.15 K. Continuous curves were calculated from eq 2. Broken lines were calculated by means of the Nitta–Chao model.

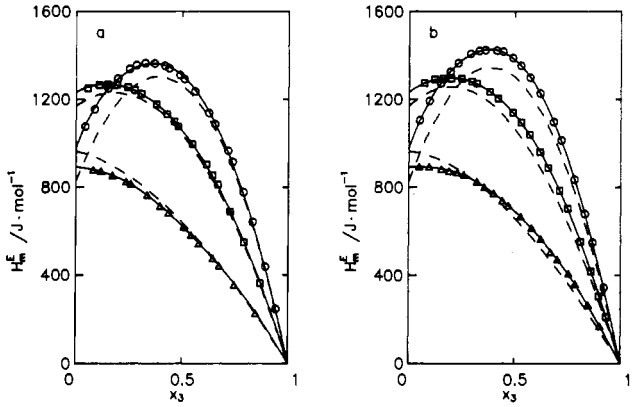

Figure 2. Excess molar enthalpies $H^E_{123,m}$ at the temperatue 298.15 K of the pseudobinaries (○) 0.2495 $CH_3(CH_2)_2OH$ + 0.7503 $CH_3CH_2COCH_2CH_3$ + $CH_3(CH_2)_nOH$ (3), (□) 0.5000 $CH_3(CH_2)_2OH$ + 0.5000 $CH_3CH_2COCH_2CH_3$ + $CH_3(CH_2)_nOH$ (3), and (△) 0.7505 $CH_3(CH_2)_2OH$ + 0.2495 $CH_3CH_2COCH_2CH_3$ + $CH_3(CH_2)_nOH$ (3): (a) $n = 5$, (b) $n = 6$. Continuous curves were calculated form eq 3, and broken lines were calculated by using the Nitta–Chao model.

The number of parameters was determined in each case using the *F*-test (*9*). The parameters A_m and the standard deviations of the fits are listed in Table II.

Table II. Parameters A_i and B_i for Equations 2–4 and Standard Deviations s

x $CH_3(CH_2)_2OH$ + $(1-x)$ $CH_3CH_2COCH_2CH_3$ (*1*)					
$A_0 = 4928$	$A_1 = 450.6$	$A_2 = 65.51$	$A_3 = 14.17$	$A_4 = 683.2$	$s = 3.6$
x $CH_3(CH_2)_2OH$ + $(1-x)$ $CH_3(CH_2)_5OH$					
$A_0 = 255.3$	$A_1 = 45.15$	$A_2 = 30.00$	$A_3 = 21.97$		$s = 0.16$
x $CH_3(CH_2)_2OH$ + $(1-x)$ $CH_3(CH_2)_6OH$					
$A_0 = 413.5$	$A_1 = 54.15$	$A_2 = 2.58$	$A_3 = 7.29$		$s = 0.27$
x $CH_3CH_2COCH_2CH_3$ + $(1-x)$ $CH_3(CH_2)_5OH$					
$A_0 = 5740$	$A_1 = 1011$	$A_2 = 1111$			$s = 3.5$
x $CH_3CH_2COCH_2CH_3$ + $(1-x)$ $CH_3(CH_2)_6OH$					
$A_0 = 6041$	$A_1 = 1139$	$A_2 = 1454$	$A_3 = 3491$		$s = 3.9$
$CH_3(CH_2)_2OH$ (1) + $CH_3CH_2COCH_2CH_3$ (2) + $CH_3(CH_2)_5OH$ (3)					
$B_0 = 1.104$	$B_1 = 10.66$	$B_2 = -10.00$	$B_3 = -19.95$	$B_4 = 42.56$	$s = 5.5$
$B_5 = -7.970$	$B_6 = 15.26$	$B_7 = -33.47$			
$CH_3(CH_2)_2OH$ (1) + $CH_3CH_2COCH_2CH_3$ (2) + $CH_3(CH_2)_6OH$ (3)					
$B_0 = 2.526$	$B_1 = 12.32$	$B_2 = -4.086$	$B_3 = -20.10$	$B_4 = 30.41$	$s = 2.5$
$B_5 = -9.622$	$B_6 = 14.65$	$B_7 = 22.02$			

Figure 1 shows the experimental excess molar enthalpies plotted against x together with the curves calculated using eq 2 and the predictions of the Nitta–Chao model.

The differences between the experimental values of the literature (*10*) in the binary mixtures and our results fitted with eq 2 were about 3.6% for $CH_3(CH_2)_2OH$ + $CH_3(CH_2)_5OH$.

Tables III and IV list the ternary excess molar enthalpies $H^E_{123,m}$ and H^E_m at the temperature 298.15 K. The values of $H^E_{123,m}$ were adequately correlated by

$$H^E_{123,m} = H^E_{12,m} + H^E_{13,m} + H^E_{23,m} + x_1x_2x_3\Delta_{123} \quad (3)$$

where

$$\Delta_{123}/RT = B_0 - B_1x_1 - B_2x_2 - B_3x_1^2 - B_4x_2^2 - B_5x_1x_2 - B_6x_1^3 - B_7x_2^3 \quad (4)$$

Equation 3 was used by Morris et al. (*11*) in correlating their ternary excess molar enthalpies. Equation 4 was suggested by Nagata and Tamura (*3*). The B_i parameters were calculated by the unweighted least-squares method using a nonlinear optimization algorithm due to Marquardt (*12*). Table II presents the values of the parameters and the corresponding standard deviations.

The experimental excess molar enthalpies of ternary mixtures plotted against x together with the curves fitted and the predictions of the Nitta–Chao model are shown in Figure 2. The Nitta–Chao model displays a $0.10H^E_m$ deviation for $CH_3(CH_2)_2OH$ + $CH_3(CH_2)_5CH_3$, $0.40H^E_m$ for $CH_3(CH_2)_2$-OH + $CH_3(CH_2)_6OH$, $0.42H^E_m$ for $CH_3(CH_2)_2OH$ + $CH_3(CH_2)_6OH$, $0.11H^E_m$ for $CH_3CH_2COCH_2CH_3$ + $CH_3(CH_2)_5$-

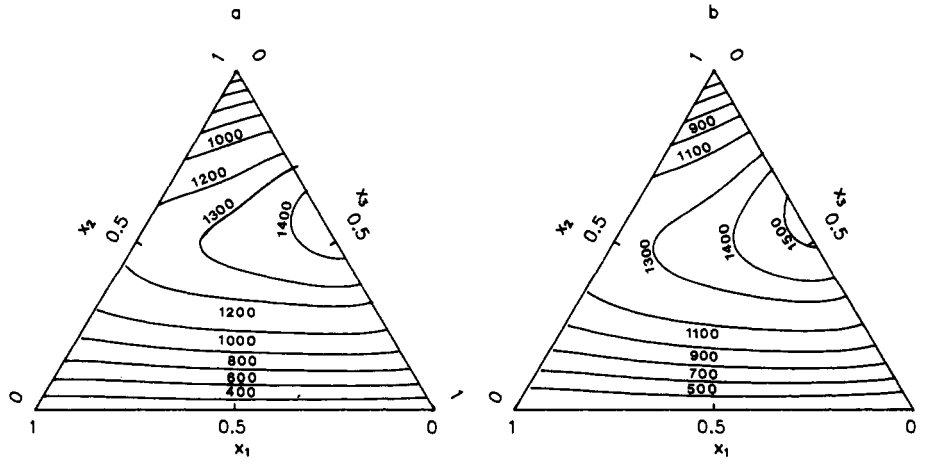

Figure 3. Curves of constant $H^E_{123,m}$/(J·mol^{-1}) for (a) $CH_3(CH_2)_2OH$ (1) + $CH_3CH_2COCH_2CH_3$ (2) + $CH_3(CH_2)_5OH$ (3) and (b) $CH_3(CH_2)_2OH$ (1) + $CH_3CH_2COCH_2CH_3$ (2) + $CH_3(CH_2)_6OH$ (3) calculated from eq 3 at the temperature 298.15 K.

Table III. Experimental Excess Molar Enthalpies for $CH_3(CH_2)_2OH$ (1) + $CH_3CH_2COCH_2CH_3$ (2) + $CH_3(CH_2)_5OH$ (3) at the Temperature 298.15 K

x_1	x_2	$H^E_{m,\psi}$/ (J·mol^{-1})	$H^E_{123,m}$/ (J·mol^{-1})	$\delta H^E_{123,m}$/ (J·mol^{-1})	$x_1x_2x_3\Delta E_{123}$/ (J·mol^{-1})
		$x_1' = 0.2495$, $H^E_{12,m} = 976.0$ J·mol^{-1}			
0.2385	0.7172	143.2	1075.9	−6.8	−28.5
0.2280	0.6856	261.7	1153.3	−7.7	−53.1
0.2120	0.6376	420.2	1249.5	1.1	−79.3
0.2009	0.6042	508.3	1294.1	1.8	−87.9
0.1872	0.5630	605.4	1337.5	6.3	−87.3
0.1791	0.5386	654.5	1354.9	7.9	−81.8
0.1688	0.5075	704.2	1364.3	5.0	−70.6
0.1577	0.4744	745.6	1362.6	−0.0	−54.9
0.1456	0.4380	781.8	1351.4	−2.5	−35.2
0.1406	0.4227	790.0	1339.8	−6.4	−26.9
0.1278	0.3845	809.2	1309.2	−6.1	−6.9
0.1230	0.3698	810.9	1291.9	−7.2	−0.0
0.1097	0.3299	806.6	1235.7	−5.9	15.7
0.0935	0.2813	771.3	1137.1	−7.3	27.3
0.0859	0.2583	750.5	1086.5	−1.7	29.7
0.0709	0.2131	689.1	966.3	8.7	29.1
0.0655	0.1970	658.5	914.7	9.9	27.5
0.0529	0.1591	570.3	777.2	9.2	21.5
0.0419	0.1261	478.0	642.0	7.3	15.1
0.0268	0.0807	334.2	439.2	9.1	6.7
0.0143	0.0430	191.1	247.1	6.7	1.9
		$x_1' = 0.5000$, $H^E_{12,m} = 1232.1$ J·mol^{-1}			
0.4724	0.4724	84.7	1248.8	−7.1	−16.9
0.4459	0.4460	165.1	1264.0	−2.8	−25.9
0.4258	0.4259	217.5	1266.9	−1.3	−28.3
0.4048	0.4048	267.5	1264.9	1.1	−27.3
0.3789	0.3790	320.2	1254.0	3.2	−22.2
0.3626	0.3626	347.5	1241.0	2.7	−17.2
0.3469	0.3469	369.5	1224.4	1.2	−11.5
0.3114	0.3115	409.6	1177.1	−0.2	2.6
0.2860	0.2860	426.3	1131.1	−2.9	12.5
0.2687	0.2687	436.7	1098.9	−0.6	18.5
0.2592	0.2592	437.2	1075.9	−2.7	21.4
0.2260	0.2260	440.1	996.9	1.4	29.0
0.1924	0.1924	428.6	902.8	7.6	31.9
0.1780	0.1781	415.6	854.3	7.4	31.6
0.1663	0.1663	402.3	812.0	6.8	30.8
0.1374	0.1374	351.1	689.8	−4.4	26.6
0.1048	0.1048	291.5	549.8	−4.2	19.3
0.0663	0.0663	199.1	362.4	−6.2	9.5
		$x_1' = 0.7505$, $H^E_{12,m} = 892.1$ J·mol^{-1}			
0.6886	0.2289	58.2	876.8	−1.9	−1.4
0.6609	0.2197	86.5	872.1	2.0	0.9
0.6209	0.2064	114.7	852.7	−0.3	4.9
0.5731	0.1905	143.3	824.6	0.1	8.7
0.5566	0.1850	152.8	814.4	1.9	9.6
0.4997	0.1661	170.5	764.4	0.8	10.7
0.4541	0.1510	175.2	715.0	−2.1	10.0
0.4243	0.1410	178.4	682.7	−0.8	9.1
0.3698	0.1229	180.2	619.7	2.3	7.3
0.3435	0.1142	174.5	582.8	−0.7	6.7
0.3166	0.1053	167.8	544.2	−3.5	6.2
0.2663	0.0885	159.2	475.6	−1.4	5.8
0.2442	0.0812	151.2	441.4	−3.0	5.8
0.1916	0.0637	127.5	355.2	−7.7	5.8
0.1155	0.0384	88.7	226.0	−6.1	4.4

OH, and $0.12H^E_m$ for $CH_3CH_2COCH_2CH_3$ + $CH_3(CH_2)_6OH$ whereas the ternary mixtures show $0.04H^E_m$ for $CH_3(CH_2)_2$-OH + $CH_3CH_2COCH_2CH_3$ + $CH_3(CH_2)_5OH$, and $0.06H^E_m$ for $CH_3(CH_2)_2OH$ + $CH_3CH_2COCH_2CH_3$ + $CH_3(CH_2)_6OH$.

The lines of constant ternary excess molar enthalpy calculated by use of eqs 3 and 4 are shown in Figure 3. Figure 4 shows lines of constant $H^E_{123,m}$ calculated by use of eq 4. The so-called "ternary contribution" represents the difference between the experimental value and that predicted from the

Table IV. Experimental Excess Molar Enthalpies for $CH_3(CH_2)_2OH$ (1) + $CH_3CH_2COCH_2CH_3$ (2) + $CH_3(CH_2)_6OH$ (3) at the Temperature 298.15 K

x_1	x_2	$H^E_{m,\psi}$/ (J·mol^{-1})	$H^E_{123,m}$/ (J·mol^{-1})	$\delta H^E_{123,m}$/ (J·mol^{-1})	$x_1x_2x_3\Delta E_{123}$/ (J·mol^{-1})
		$x_1' = 0.2495$, $H^E_{12,m} = 976.0$ J·mol^{-1}			
0.2368	0.7120	179.3	1105.3	1.9	−51.3
0.2255	0.6781	311.2	1193.1	1.0	−83.1
0.2124	0.6388	441.2	1271.9	−0.9	−104.0
0.2010	0.6045	538.7	1324.8	−3.0	−109.4
0.1918	0.5767	613.3	1363.3	0.1	−106.1
0.1768	0.5319	715.5	1407.2	3.2	−89.4
0.1669	0.5018	768.0	1420.7	0.5	−72.8
0.1596	0.4801	799.7	1424.1	−2.1	−59.0
0.1461	0.4394	849.8	1421.3	−2.2	−31.7
0.1385	0.4164	875.3	1416.9	3.3	−16.6
0.1272	0.3826	891.1	1388.6	1.3	4.0
0.1176	0.3536	892.5	1352.4	−0.7	18.9
0.1068	0.3213	880.7	1298.5	−2.9	31.9
0.0946	0.2844	856.1	1226.0	1.3	41.2
0.0787	0.2368	789.6	1097.6	0.7	44.5
0.0698	0.2100	740.3	1013.4	2.9	42.4
0.0543	0.1634	622.2	834.7	−1.6	33.6
0.0423	0.1272	512.8	678.3	−2.1	24.0
0.0330	0.0992	420.5	549.5	1.7	16.2
0.0197	0.0591	266.5	343.4	2.5	6.5
		$x_1' = 0.5000$, $H^E_{12,m} = 1232.1$ J·mol^{-1}			
0.4639	0.4640	130.2	1273.5	0.0	−27.2
0.4398	0.4398	207.4	1291.1	2.1	−34.7
0.4197	0.4197	260.2	1294.4	−1.1	−35.5
0.4011	0.4011	305.2	1293.5	−3.2	−32.5
0.3761	0.3761	365.3	1292.0	0.8	−24.0
0.3522	0.3523	412.6	1280.6	2.2	−12.5
0.3399	0.3399	434.1	1271.7	3.0	−5.8
0.3089	0.3089	472.4	1233.5	−1.4	12.1
0.2894	0.2894	489.4	1202.6	−3.9	23.1
0.2544	0.2544	512.6	1139.5	−0.4	40.0
0.2344	0.2344	517.9	1095.4	3.1	47.2
0.2144	0.2144	510.6	1039.0	1.2	52.2
0.1899	0.1899	491.4	959.3	−1.2	54.8
0.1706	0.1706	467.9	888.2	−3.5	54.1
0.1439	0.1439	430.7	785.2	0.5	49.3
0.1249	0.1249	395.6	703.3	2.9	43.5
0.0949	0.0949	318.2	552.0	−1.8	31.5
0.0699	0.0699	244.7	416.9	−2.9	20.3
0.0506	0.0506	179.8	304.6	−5.6	12.0
0.0339	0.0339	126.6	210.1	−0.6	6.0
		$x_1' = 0.7505$, $H^E_{12,m} = 892.1$ J·mol^{-1}			
0.7133	0.2371	45.4	893.3	0.6	−3.5
0.6782	0.2255	87.2	893.4	2.1	−2.0
0.6345	0.2109	127.6	881.8	−2.6	3.2
0.6068	0.2017	151.0	872.3	−4.1	7.2
0.5777	0.1920	179.7	866.4	1.8	11.4
0.5551	0.1845	196.9	856.7	3.7	14.4
0.5094	0.1693	220.0	825.5	2.1	19.7
0.4830	0.1606	226.6	800.7	−1.9	22.2
0.4494	0.1494	238.1	772.3	−0.2	24.7
0.4189	0.1392	241.5	739.4	−2.2	26.6
0.3976	0.1322	244.7	717.3	−1.0	27.6
0.3536	0.1176	246.9	667.3	1.8	28.9
0.3141	0.1044	241.8	615.2	2.6	29.4
0.2818	0.0937	229.6	564.6	−0.9	29.0
0.2469	0.0821	212.6	506.1	−4.5	27.9
0.2233	0.0742	205.9	471.3	0.4	26.5
0.1845	0.0613	187.0	406.3	5.0	23.2
0.1617	0.0537	166.7	358.9	1.2	20.5
0.1151	0.0383	128.5	265.3	2.9	13.9
0.0733	0.0244	81.8	168.9	−1.6	7.3

binary mixtures by employing eq 2. This contribution shows a maximum of 34 J·mol^{-1}, $x_1 = 0.13$, $x_2 = 0.21$, and a minimum of −89 J·mol^{-1}, $x_1 = 0.21$, $x_2 = 0.58$, for $CH_3(CH_2)_2OH$ + CH_3-

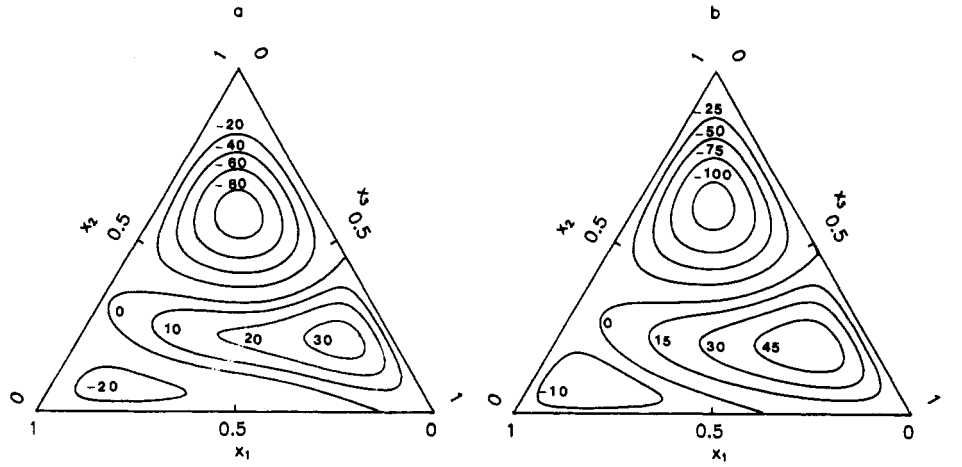

Figure 4. Ternary contributions $x_1x_2x_3\ \Delta_{123}/(J\cdot mol^{-1})$ to excess molar enthalpies of (a) $CH_3(CH_2)_2OH$ (1) + $CH_3CH_2COCH_2CH_3$ (2) + $CH_3(CH_2)_5OH$ (3) and (b) $CH_3(CH_2)_2OH$ (1) + $CH_3CH_2COCH_2CH_3$ (2) + $x_3CH_3(CH_2)_6OH$ (3) at the temperature 298.15 K.

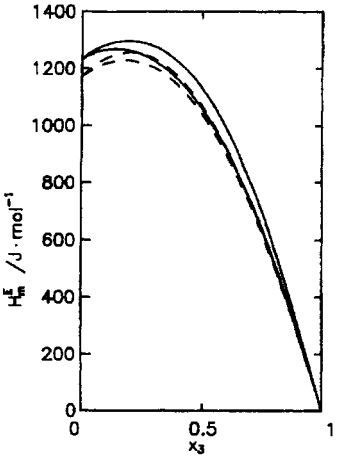

Figure 5. Curves of excess molar enthalpies at the temperature 298.15 K of the pseudobinary system ($0.5CH_3(CH_2)_2OH$ (1) + $0.5CH_3CH_2COCH_2CH_3$ (2)) + $CH_3(CH)_nOH$ (3), n = 5 and n = 6: (- - -) eq 3, (—) Nitta–Chao model.

$CH_2COCH_2CH_3$ + $CH_3(CH_2)_5OH$ and a maximum of 55 $J\cdot mol^{-1}$, $x_1 = 0.16$, $x_2 = 0.20$, and a minimum of $-109\ J\cdot mol^{-1}$, $x_1 = 0.20$, $x_2 = 0.61$, for $CH_3(CH_2)_2OH$ + $CH_3CH_2COCH_2CH_3$ + $CH_3(CH_2)_6OH$.

Figure 5 shows the variation of excess molar enthalpy of ternary systems ($CH_3(CH_2)_2OH$ (1) + $CH_3CH_2COCH_2CH_3$ (2) + $CH_3(CH)_nOH$ (3) (n = 5, 6)) when the 1-alkanol chain is varied; note the rise of the enthalpy with the number of atoms of the 1-alkanol.

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