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Excess molar enthalpies of the ternary systems pentan-3-one + *n*-hexane + *n*-decane and *n*-tetradecane at 298.15 K

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Abstract

Excess molar enthalpies at a temperature of 298.15 K were measured for (pentan-3-one + *n*-hexane + *n*-decane and *n*-tetradecane) and also for (pentan-3-one + *n*-decane and *n*-tetradecane) and (*n*-hexane + *n*-decane) using a Calvet microcalorimeter. The experimental results agreed quite well with those calculated by using the Nitta–Chao model.

INTRODUCTION

In previous work [1, 2] we determined the excess molar enthalpies of ternary systems at 298.15 K. We report here the excess molar enthalpies at 298.15 K and normal atmospheric pressure of (pentan-3-one + *n*-hexane + *n*-decane and *n*-tetradecane) 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 system was fitted by means of an equation proposed by Nagata and Tamura [3]. The obtained 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

The chemical substances employed were supplied by Fluka, and were subjected to no further purification other than being dried with Union

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Carbide 0.4 nm molecular sieves and degasified. The mole-fraction purities were all >99.5% for pentan-3-one, *n*-hexane, *n*-decane and *n*-tetradecane.

All experimental measurements were carried out using a Calvet microcalorimeter equipped with a device allowing operation in the absence of vapour phase. Calibration was performed electrically using a Setaram EJP 30 stabilized current source and tested further with the hexane + cyclohexane mixture [6, 7]. Details of the procedure employed have been published previously [8, 9]. The precision of the excess molar enthalpies is estimated as better than $0.01H_m^E$.

Three experimental runs were carried out for ternary mixtures formed by adding *n*-decane or *n*-tetradecane to a binary mixture of pentan-3-one (x_1') + *n*-hexane (x_2'). A ternary mixture may be considered as a pseudo-binary mixture composed of one binary mixture and *n*-alkane x_3. The ternary excess molar enthalpy at x_1, x_2 and x_3 can be expressed as

$$H^E_{123,m} = H^E_{m,\psi} = (x_1 + x_2)H^E_{12,m} \tag{1}$$

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 pentan-3-one + *n*-hexane. 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 [10].

RESULTS AND DISCUSSION

Excess molar enthalpies of the binary mixtures are listed in Table 1. A variable degree polynomial of the form

$$H^E_{ij,m}\,(\mathrm{J\ mol^{-1}}) = x_i x_j \sum_{m=1}^{n} A_m (x_i - x_j)^m \tag{2}$$

was fitted. The least squares method was employed in both cases. The number of parameters was determined in each case using an *F*-test [11]. The parameters A_m and the standard deviations of the fits are listed in Table 2.

Figure 1 show the experimental excess molar enthalpies of binary systems plotted against x together with the fitted curves and the predictions of the Nitta–Chao model.

The differences between the experimental values in the literature [12, 13] for the results for binary mixtures fitted by eqn. (2) were about 2.3% for (pentan-3-one + *n*-decane) and 5.6% for (*n*-hexane + *n*-decane). Tables 3 and 4 list the ternary excess molar enthalpies $H^E_{123,m}$ and $H^E_{m,\psi}$ at 298.15 K. The values of $H^E_{123,m}$ were adequately correlated by

$$H^E_{123,m} = H^E_{12,m} + H^E_{13,m} + H^E_{23,m} + x_1 x_2 x_3 \Delta_{123} \tag{3}$$

TABLE 1

Experimental excess molar enthalpies H_m^E at 298.15 K

x	H_m^E ($J\,mol^{-1}$)	x	H_m^E ($J\,mol^{-1}$)	x	H_m^E ($J\,mol^{-1}$)	x	H_m^E ($J\,mol^{-1}$)
$xC_2H_5COC_2H_5 + (1-x)C_{10}H_{22}$							
0.1293	654.3	0.3665	1230.5	0.5718	1267.1	0.7703	957.8
0.1807	840.6	0.4013	1256.2	0.6173	1223.2	0.8044	861.5
0.2402	1012.4	0.4612	1290.5	0.6411	1196.5	0.8500	712.3
0.2851	1110.9	0.5033	1290.0	0.6935	1120.4	0.8997	520.2
0.3267	1175.0	0.5358	1283.2	0.7421	1025.6	0.9423	316.1
$xCO_2H_5COC_2H_5 + (1-x)C_{14}H_{30}$							
0.0551	360.0	0.2802	1239.7	0.5557	1490.4	0.8245	922.0
0.0762	484.2	0.3315	1342.8	0.6105	1440.0	0.8653	755.7
0.1207	696.7	0.3675	1401.9	0.6754	1336.4	0.9122	533.9
0.1605	861.5	0.3952	1443.7	0.7023	1280.1	0.9372	402.0
0.2130	1047.1	0.4421	1490.0	0.7530	1160.2		
0.2442	1140.6	0.5013	1505.2	0.8051	997.6		
$xC_6H_{14} + (1-x)C_{10}H_{22}$							
0.0698	4.7	0.3233	14.0	0.5276	14.9	0.7401	10.6
0.1241	7.2	0.3820	14.9	0.5651	14.5	0.7968	8.8
0.1802	9.6	0.4241	15.2	0.6274	13.6	0.8498	6.8
0.2362	11.8	0.4501	15.2	0.6634	12.9	0.8921	4.8
0.2663	12.7	0.4826	15.2	0.6986	12.0		

TABLE 2

Parameters A_i, B_i for eqns. (2) and (4) and standard deviations s

$xC_2H_5COC_2H_5 + (1-x)C_6H_{14}$ [1]
$A_0 = 4094$, $A_1 = -779.1$, $A_2 = 635.3$, $A_3 = -283.2$ $s = 4.6$

$xC_2H_5COC_2H_5 + (1-x)C_{10}H_{22}$
$A_0 = 5163$, $A_1 = -179.5$, $A_2 = 1180$, $A_3 = 125.1$, $A_4 = -162.2$ $s = 2.3$

$xC_6H_{14} + (1-x)C_{10}H_{22}$
$A_0 = 60.61$, $A_1 = -9.187$, $A_2 = -1.180$, $A_3 = -1.620$ $s = 0.10$

$xC_2H_5COC_2H_5 + (1-x)C_{14}H_{30}$
$A_0 = 6022$, $A_1 = 67.40$, $A_2 = 473.8$, $A_3 = -137.1$, $A_4 = 808.7$ $s = 3.3$

$xC_6H_{14} + (1-x)C_{14}H_{30}$ [2]
$A_0 = 275.3$, $A_1 = -6.821$, $A_2 = -147.9$ $s = 1.1$

$xC_2H_5COC_2H_5 + x_2C_6H_{14} + (1-x_1-x_2)C_{10}H_{22}$
$B_0 = -1.365$, $B_1 = -21.20$, $B_2 = 1.863$, $B_3 = 56.48$, $B_4 = -5.546$, $B_5 = 1.785$, $B_6 = -43.20$, $B_7 = 4.769$ $s = 3.9$

$x_1C_2H_5COC_2H_5 + x_2C_6H_{14} + (1-x_1-x_2)C_{14}H_{30}$
$B_0 = -2.534$, $B_1 = -29.92$, $B_2 = 0.0289$, $B_3 = 79.87$, $B_4 = 4.448$, $B_5 = -2.336$, $B_6 = -64.54$, $B_7 = -3.759$ $s = 4.1$

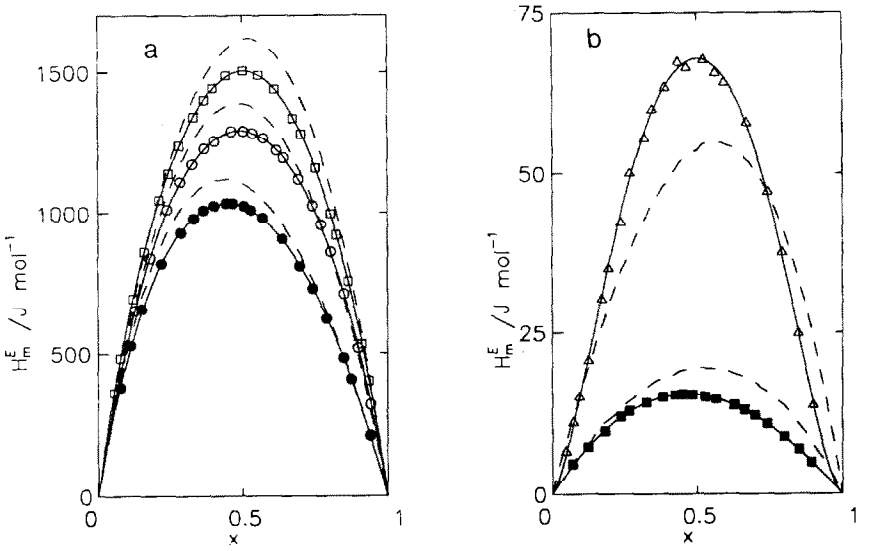

Fig. 1. Excess molar enthalpies at 298.15 K for $\{xC_2H_5COC_2H_5 + (1-x)CH_3(CH_2)_nCH_3\}$ and $\{xCH_3(CH_2)_mCH_3 + (1-x)CH_3(CH_2)_lCH_3\}$: (a) (●), $n = 4$ [1]; (○), $n = 8$; (□), $n = 12$; (b) (■), $m = 4$, $l = 8$; (△), $m = 4$, $l = 12$ [2]. Continuous curves were calculated from eqn. (2). Broken lines were calculated by means of the Nitta–Chao model.

TABLE 3

Experimental excess molar enthalpies for $x_1C_2H_5COC_2H_5 + x_2C_6H_{14} + x_3C_{10}H_{22}$ at 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})
$x_1' = 0.2505$	$H^E_{12,m} = 875.8\ J\ mol^{-1}$				
0.2454	0.7339	6.6	864.4	−4.2	4.5
0.2323	0.6945	34.3	846.0	−3.6	16.1
0.2186	0.6538	61.7	825.8	−1.8	26.5
0.2131	0.6372	72.1	816.8	−0.7	29.9
0.1996	0.5968	95.7	793.1	3.1	35.9
0.1877	0.5613	110.1	766.1	3.9	38.0
0.1795	0.5368	117.4	744.7	3.8	37.8
0.1682	0.5029	121.0	708.7	−0.2	35.6
0.1635	0,4889	123.5	694.9	−0.0	34.1
0.1508	0.4510	127.9	655.0	0.3	28.5
0.1361	0.4068	128.3	603.8	−0.7	20.2
0.1227	0.3669	124.2	553.0	−3.4	12.1
0.1177	0.3519	120.3	531.6	−6.3	9.1
0.0993	0.2970	118.1	465.2	−2.6	−0.7
0.0891	0.2663	113.3	424.5	−2.8	−4.9
0.0733	0.2192	103.5	359.6	−3.7	−9.0
0.0511	0.1528	86.1	264.6	−3.4	−9.4
0.0333	0.0994	63.6	179.8	−4.8	−6.1

TABLE 3 (continued)

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})
$x_1' = 0.5020$	$H^E_{12,m} = 1024\,J\,mol^{-1}$				
0.4768	0.4730	78.2	1051.1	−2.3	1.1
0.4545	0.4508	149.3	1076.5	1.5	7.0
0.4225	0.4191	235.8	1097.8	−0.0	20.5
0.4011	0.3979	287.2	1105.5	−1.5	31.1
0.3792	0.3761	340.8	1114.3	3.8	41.9
0.3679	0.3650	355.8	1106.4	−3.5	47.0
0.3407	0.3380	401.2	1096.3	−4.4	57.3
0.3173	0.3147	436.1	1083.4	−0.2	63.0
0.2857	0.2835	463.4	1046.4	−0.1	65.1
0.2792	0.2770	466.4	1036.1	−0.7	64.8
0.2508	0.2488	477.5	989.3	3.1	60.0
0.2322	0.2304	481.1	954.9	9.1	54.5
0.1982	0.1966	455.5	859.9	2.1	40.7
0.1864	0.1849	445.0	825.3	2.2	35.3
0.1580	0.1567	412.6	734.9	3.7	22.1
0.1392	0.1381	385.2	669.2	4.9	14.1
0.1061	0.1053	321.8	538.3	3.7	3.0
0.0686	0.0680	233.3	373.2	4.5	−2.7
0.0361	0.0358	136.5	210.1	3.5	−2.2
$x_1' = 0.7554$	$H^E_{12,m} = 704.2\,J\,mol^{-1}$				
0.7290	0.2359	120.2	799.7	−5.2	4.7
0.7027	0.2274	228.6	883.6	−3.7	4.4
0.6759	0.2187	325.6	955.6	−1.7	1.9
0.6584	0.2131	378.2	991.9	−4.7	0.0
0.6268	0.2028	478.1	1062.3	4.4	−2.3
0.6089	0.1970	524.6	1092.1	4.7	−2.6
0.5761	0.1864	598.5	1135.5	2.0	−0.8
0.5434	0.1759	664.0	1170.5	0.5	3.9
0.5072	0.1642	730.9	1203.7	3.8	11.5
0.4826	0.1562	767.2	1217.0	3.2	17.5
0.4524	0.1464	801.3	1222.9	−0.3	25.0
0.3834	0.1241	846.5	1203.8	−6.3	38.1
0.3588	0.1161	853.7	1188.1	−4.5	40.3
0.3535	0.1144	854.9	1184.4	−3.5	40.5
0.3170	0.1026	845.3	1140.8	−5.0	40.2
0.2612	0.0845	801.0	1044.4	−3.4	33.2
0.2490	0.0806	783.7	1015.8	−5.1	30.9
0.2191	0.0709	744.0	948.3	2.0	24.3
0.1703	0.0551	643.6	802.4	4.8	12.9
0.1125	0.0364	477.3	582.1	4.6	2.6
0.0669	0.0216	311.6	373.9	4.8	−0.7

TABLE 4

Experimental excess molar enthalpies for $x_1C_2H_5COC_2H_5 + x_2C_6H_{14} + x_3C_{14}H_{30}$ at 298.15 K

x_1	x_2	$H^E_{m,\psi}$ ($J\,mol^{-1}$)	$H^E_{123,m}$ ($J\,mol^{-1}$)	$\delta H^E_{123,m}$ ($J\,mol^{-1}$)	$x_1x_2x_3\Delta^E_{123}$ ($J\,mol^{-1}$)
$x'_1 = 0.2505$	$H^E_{12,m} = 875.8\,J\,mol^{-1}$				
0.2443	0.7306	15.6	869.5	−4.9	4.9
0.2293	0.6857	62.3	863.7	−2.3	12.4
0.2242	0.6704	77.9	861.4	−0.1	13.9
0.2136	0.6388	101.2	847.7	−1.7	15.4
0.2000	0.5979	130.2	829.0	0.3	14.8
0.1901	0.5684	148.2	812.4	2.4	12.9
0.1768	0.5286	164.3	782.1	1.9	8.9
0.1662	0.4969	175.8	756.6	3.7	4.8
0.1535	0.4589	180.7	717.0	0.9	−0.9
0.1477	0.4417	182.9	699.1	0.9	−3.6
0.1331	0.3980	183.3	648.4	−0.8	−10.5
0.1282	0.3834	181.6	629.6	−2.3	−12.7
0.1177	0.3519	180.6	591.8	−1.2	−17.1
0.1054	0.3153	171.4	539.9	−5.7	−21.3
0.0851	0.2545	159.2	456.6	−5.0	−25.1
0.0609	0.1821	134.8	347.7	−5.6	−22.8
0.0393	0.1174	102.2	239.4	−6.5	−14.7
$x'_1 = 0.5021$	$H^E_{12,m} = 1024.2\,J\,mol^{-1}$				
0.4904	0.4862	58.7	1058.9	1.9	6.4
0.4695	0.4656	153.6	1111.3	3.4	17.6
0.4419	0.4382	264.3	1165.7	3.1	32.4
0.4238	0.4202	329.8	1194.3	3.5	42.1
0.3981	0.3947	410.6	1222.6	2.2	55.3
0.3803	0.3771	452.7	1228.4	−4.9	63.5
0.3663	0.3632	485.0	1232.2	−6.6	69.2
0.3480	0.3451	528.0	1237.9	−1.8	75.1
0.3251	0.3223	565.6	1228.6	−1.4	79.6
0.3000	0.2974	590.2	1202.0	−3.5	80.4
0.2761	0.2737	605.3	1168.4	−0.3	76.8
0.2650	0.2627	605.8	1146.3	−0.8	73.7
0.2408	0.2388	604.5	1095.7	4.8	64.3
0.2133	0.2115	583.8	1018.9	6.3	50.0
0.2021	0.2004	570.2	982.5	5.7	43.5
0.1809	0.1794	540.7	909.7	6.5	30.7
0.1512	0.1499	485.8	794.2	4.8	13.7
0.1371	0.1359	452.8	732.3	1.1	6.7
0.1072	0.1063	386.3	605.0	3.9	−4.2
0.0696	0.0690	280.2	422.2	1.2	−8.5

TABLE 4 (continued)

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})
$x'_i = 0.7556$	$H^E_{12,m} = 704.2\,J\,mol^{-1}$				
0.7221	0.2337	210.7	883.8	−3.8	35.5
0.6995	0.2264	327.1	979.1	−6.3	46.1
0.6632	0.2146	488.6	1106.8	−2.6	50.2
0.6257	0.2025	624.6	1207.8	0.7	46.8
0.6082	0.1968	681.0	1247.9	2.9	44.6
0.5729	0.1854	778.5	1312.5	4.2	41.0
0.5339	0.1728	866.9	1364.6	3.4	39.9
0.5106	0.1652	911.7	1387.6	3.1	40.7
0.4826	0.1562	956.7	1406.5	2.5	43.0
0.4635	0.1500	979.4	1411.5	−0.3	44.9
0.4314	0.1396	1010.3	1412.4	−1.7	48.2
0.4094	0.1325	1022.1	1403.7	−3.9	50.0
0.3788	0.1226	1031.2	1384.3	−2.7	51.2
0.3489	0.1129	1022.7	1347.9	−5.4	50.5
0.3340	0.1081	1015.4	1326.7	−4.9	49.3
0.2891	0.0936	979.3	1248.8	2.4	42.4
0.2643	0.0855	939.9	1186.3	−0.5	36.6
0.2172	0.0703	848.5	1051.0	0.3	23.4
0.1709	0.0553	730.3	889.6	0.5	10.4
0.1422	0.0460	638.2	770.7	−4.6	3.9
0.0903	0.0292	448.0	532.2	−8.9	−2.5
0.0494	0.0160	269.9	316.0	−6.8	−2.3

where

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

Equation (3) was used by Morris et al. [14] in correlating their ternary excess molar enthalpy results. Equation (4) was suggested by Nagata and Tamura [3]. The B_i parameters were calculated by the unweighted least squares method using a non-linear optimization algorithm due to Marquardt [15]. Table 2 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 given in Fig. 2. The Nitta–Chao model displays a $0.11H^E_m$ deviation for (pentan-3-one + *n*-hexane); $0.05H^E_m$ for (pentan-3-one + *n*-decane); $0.16H^E_m$ for (pentan-3-one + *n*-tetradecane); $0.31H^E_m$ for (*n*-hexane + *n*-decane) and $0.20H^E_m$ for (*n*-hexane + *n*-tetradecane), whereas the ternary mixtures show $0.10H^E_m$ for (pentan-3-one + *n*-hexane + *n*-decane) and $0.07H^E_m$ for (pentan-3-one + *n*-hexane + *n*-tetradecane).

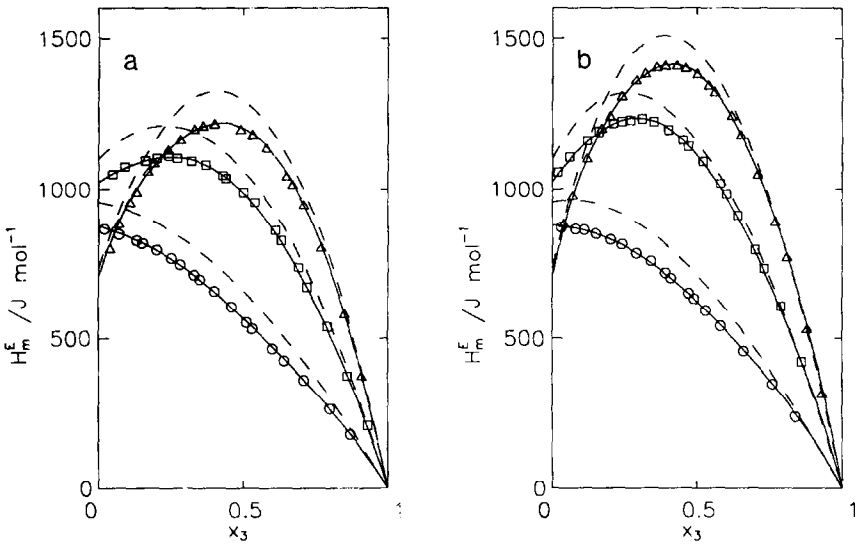

Fig. 2. Excess molar enthalpies $H^E_{123,m}$ at 298.15 K of the pseudobinary: (a) (○) $\{x(0.2505C_2H_5COC_2H_5 + 0.7495C_6H_{14}) + (1-x)C_{10}H_{22})\}$; (□) $\{x(0.5020C_2H_5COC_2H_5 + 0.4980C_6H_{14}) + (1-x)C_{10}H_{22}\}$; (△) $\{x(0.7554C_2H_5COC_2H_5 + 0.2446C_6H_{14}) + (1-x)C_{10}H_{22}\}$; (b) (○) $\{(0.2505C_2H_5COC_2H_5 + 0.7495C_6H_{14}) + (1-x)C_{14}H_{30})\}$; (□) $\{x(0.5021C_2H_5COC_2H_5 + 0.4979C_6H_{14}) + (1-x)C_{14}H_{30})\}$; (△) $\{x(0.7556C_2H_5COC_2H_5 + 0.2444C_6H_{14}) + (1-x)C_{14}H_{30}\}$. Continuous curves were calculated from eqn. (4) and broken lines were calculated by using the Nitta–Chao model.

Figure 3 shows lines of constant ternary excess molar enthalpies (isolines) calculated by using eqn. (3).

The lines of constant ternary excess molar enthalpy calculated by use of eqns. (3) and (4) are shown in Fig. 3. Figure 4 shows lines of constant $H^E_{123,m}$ obtained by use of eqn. (5). The so called "ternary contribution"

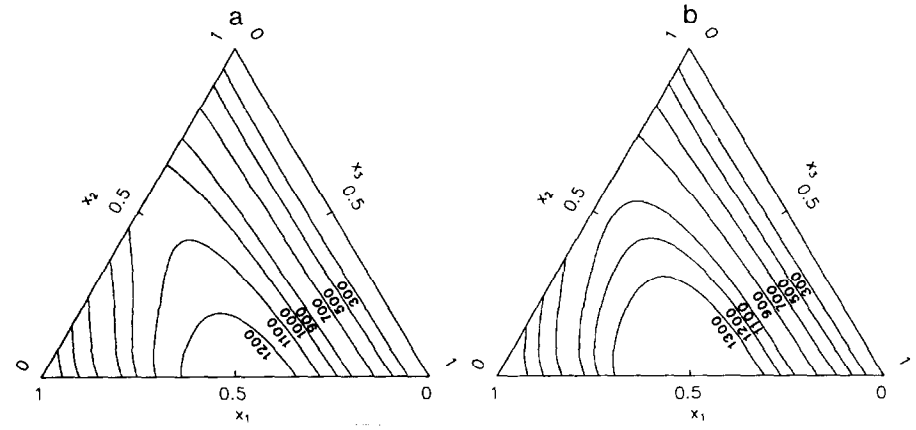

Fig. 3. Curves of constant $H^E_{123,m}$ (J mol^{-1}) for (a) $(x_1C_2H_5COC_2H_5 + x_2C_6H_{14} + x_3C_{10}H_{22})$, (b) $(x_1C_2H_5COC_2H_5 + x_2C_6H_{14} + x_3C_{10}H_{22})$ calculated from eqn. (4) at 298.15 K.

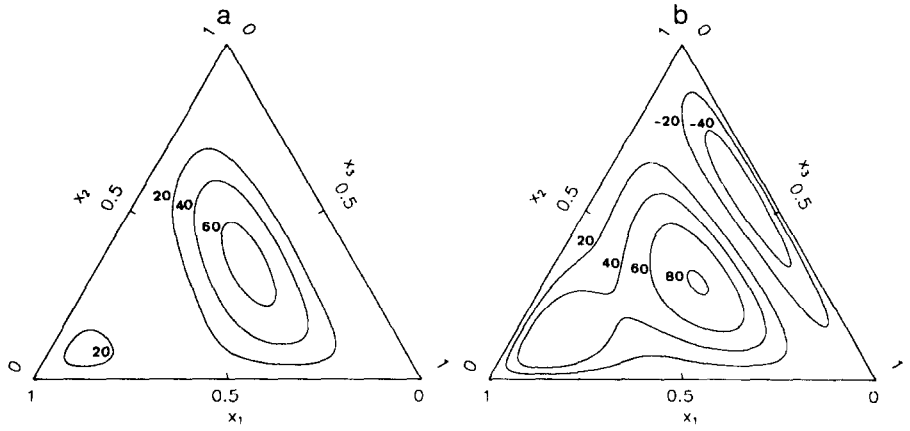

Fig. 4. Ternary contribution isolines $x_1x_2x_3\Delta_{123}$ (J mol^{-1}) to excess molar enthalpy of (a) ($x_1C_2H_5COC_2H_5 + x_2C_6H_{14} + x_3C_{10}H_{22}$), (b) ($x_1C_2H_5COC_2H_5 + x_2C_6H_{14} + x_3C_{10}H_{22}$) calculated from eqn. (4) at 298.15 K.

$x_1x_2x_3\Delta_{123}$ represents the difference between the experimental value and that predicted from the binary mixtures by employing eqns. (2) and (3). This contribution shows a maximum of 66 J mol^{-1} at $x_1 = 0.28$, $x_2 = 0.34$ and a minimum of -18 J mol^{-1} at $x_1 = 0.04$, $x_2 = 0.46$ for (pentan-3-one + *n*-hexane + *n*-decane), and a maximum of 81 J mol^{-1} at $x_1 = 0.32$, $x_2 = 0.28$ and a minimum of -53 J mol^{-1} at $x_1 = 0.06$, $x_2 = 0.56$ for (pentan-3-one + *n*-hexane + *n*-tetradecane).

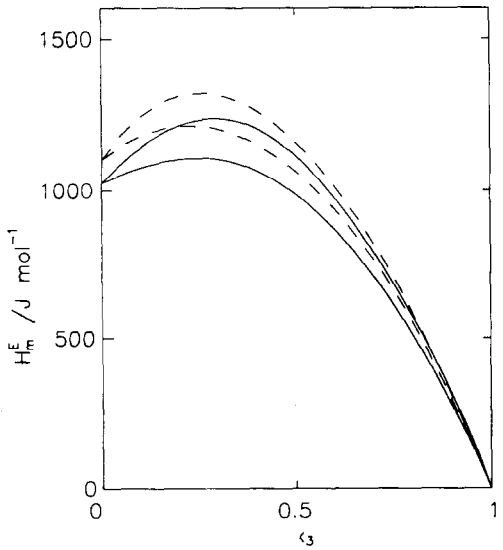

Fig. 5. Curves of excess molar enthalpies $H^E_{m,\psi}$ at 298.15 K of the pseudobinary system $\{x(0.5C_2H_5COC_2H_5 + 0.5C_6H_{14}) + (1-x)C_nH_{2n+2})\}$, $n = 8$ and $n = 12$. – – – – – – –, eqn. (3). ——, Nitta–Chao model.

Figure 5 shows the variation of excess molar enthalpy of ternary systems (pentan-3-one + *n*-hexane + *n*-alkane) when the alkane chain length is varied; note the rise of enthalpy with increasing number of atoms of the alkane.

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