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Excess molar enthalpies of the ternary mixtures: (diisopropyl ether or tetrahydrofuran) + 3-methylpentane +*n*-dodecane at 298.15 K

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Abstract

Microcalorimetric measurements of excess molar enthalpies, at 298.15 K, are reported for the two ternary systems formed by mixing either diisopropyl ether or tetrahydrofuran with binary mixtures of 3-methylpentane and *n*-dodecane. Smooth representations of the results are presented and used to construct constant excess molar enthalpy contours on Roozeboom diagrams. It is shown that useful estimates of the ternary enthalpies can be obtained from the Liebermann and Fried model, using only the physical properties of the components and their binary mixtures.

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1. Introduction

Recent papers [1,2], from our laboratory, reported excess molar enthalpies at 298.15 K for the ternary systems formed by mixing either diisopropyl ether (DIPE) or tetrahydrofuran (THF) with binary mixtures of 3-methylpentane (3MP) and either *n*-octane (*n*C8) or *n*-decane (*n*C10). As a continuation of those investigations, similar measurements have been made for the analogous systems, in which the third component was replaced by *n*-dodecane (*n*C12).

2. Experimental

The components used for the present measurements, were obtained from the Aldrich Chemical Co. The DIPE, 3MP and *n*C12, all had stated purities exceeding 99.0 mol%. The THF had a stated purity >99.5 mol% and was inhibitor free. Apart from partial degassing, the components were used as received from the manufacturer. Densities, measured at 298.15 K in an Anton-Paar digital densimeter, were 718.70, 882.09, 659.94 and 745.32 kg m^{-3} for DIPE, THF, 3MP and *n*C12, respectively. These are in reasonable agreement with values in the literature [3–5].

An LKB flow microcalorimeter (Model 10700-1), maintained at 298.150 ± 0.003 K, was used to measure the excess molar enthalpies $H_{\mathrm{m}}^{\mathrm{E}}$. Details of the equipment and the operating procedure have been described previously [6,7].

For the ternary systems $x_1\{(CH_3)_2CH\}_2O + x_2CH_3CH_2CH(CH_3)CH_2CH_3 + x_3CH_3(CH_2)_{10}CH_3$ and $x_1C_4H_8O + x_2CH_3CH_2CH(CH_3)CH_2CH_3 + x_3CH_3(CH_2)_{10}CH_3$, the excess molar enthalpy $H_{\mathrm{m},1+23}^{\mathrm{E}}$ was determined for several pseudo-binary systems in which DIPE or THF was added to a binary mixture of components 2 and 3, having a fixed mole ratio x_2/x_3. For this purpose, binary mixtures with $x_2/x_3 \approx 0.3$, 1.0 and 3.0 were prepared by weighing. The excess molar enthalpy of the ternary system was obtained from the relation:

$$H_{\mathrm{m},123}^{\mathrm{E}} = H_{\mathrm{m},1+23}^{\mathrm{E}} + (1 - x_1)H_{\mathrm{m},23}^{\mathrm{E}} \tag{1}$$

where $H_{\mathrm{m},23}^{\mathrm{E}}$ is the excess molar enthalpy of the particular binary mixture of 3MP and *n*C12. Over most of the mole fraction range of component 1, the errors of $H_{\mathrm{m},1+23}^{\mathrm{E}}$ are estimated to be less than 0.5%. Errors in the mole fractions of the final ternary mixtures are estimated to be less than 5×10^{-4}.

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3. Results and discussion

Excess molar enthalpies $H^{\mathrm{E}}_{\mathrm{m},ij}$ ($i < j$), at $T = 298.15\,\mathrm{K}$, for four of the constituent-binary systems of present interest, have been reported previously: DIPE(1) + 3MP(2) [1], DIPE(1) + nC12(3) [8], 3MP(2) + nC12(3) [9], and THF(1) + 3MP(2) [2]. The experimental values of x_1 and $H^{\mathrm{E}}_{\mathrm{m},12}$, measured for THF(1)+$n$C12(3) are listed in Table 1. Coefficients h_k for the representations of those results by the smoothing function:

$$H^{\mathrm{E}}_{\mathrm{m},ij}\,(\mathrm{J\,mol^{-1}}) = x_i(1-x_i)\sum_{k=1}^{m} h_k(1-2x_i)^{k-1}, \quad i < j \quad (2)$$

Table 1
Experimental mole fractions x_1 and excess molar enthalpies $H^{\mathrm{E}}_{\mathrm{m},13}$ for $x_1\mathrm{C_4H_8O} + (1-x_1)\mathrm{CH_3(CH_2)_{10}CH_3}$ binary mixtures at 298.15 K

x_1	$H^{\mathrm{E}}_{\mathrm{m},13}$	x_1	$H^{\mathrm{E}}_{\mathrm{m},13}$	x_1	$H^{\mathrm{E}}_{\mathrm{m},13}$	x_1	$H^{\mathrm{E}}_{\mathrm{m},13}$
0.0500	164.53	0.3001	780.11	0.5505	1013.83	0.7998	764.33
0.1000	322.55	0.3503	860.20	0.6005	1007.48	0.8501	633.27
0.1500	457.79	0.4001	930.49	0.6498	982.14	0.9002	468.66
0.2000	571.53	0.4500	977.74	0.7002	930.03	0.9500	262.85
0.2501	685.57	0.4997	996.68	0.7501	860.83		

Table 2
Coefficients h_k and standard deviations s for the representations of the excess molar enthalpies $H^{\mathrm{E}}_{\mathrm{m},ij}$ of the constituent-binary mixtures at 298.15 K by Eq. (2)

i	j	h_1	h_2	h_3	h_4	h_5	s (J mol^{-1})
DIPE	3MP[a]	790.32	51.08	26.96	58.27		0.73
DIPE	nC12[b]	1698.42	−198.39	140.61			1.20
3MP	nC12[c]	298.54	−5.92	−9.38			0.10
THF	3MP[d]	2887.50	23.24	261.10			2.21
THF	nC12[e]	4012.92	−187.49	344.17	−324.07	369.10	3.51

[a] From Wang et al. [1].
[b] From Zhu et al. [8].
[c] From Hamam et al. [9].
[d] From Wang et al. [2].
[e] Present work.

Table 3
Experimental excess molar enthalpies $H^{\mathrm{E}}_{\mathrm{m},1+23}$ at 298.15 K for the addition of DIPE to a binary mixture of 3MP and nC12 to form $x_1\{\mathrm{(CH_3)_2CH}\}_2\mathrm{O} + x_2\mathrm{CH_3CH_2CH(CH_3)CH_2CH_3} + x_3\mathrm{CH_3(CH_2)_{10}CH_3}$ and values of $H^{\mathrm{E}}_{\mathrm{m},123}$ calculated from Eq. (1) using $H^{\mathrm{E}}_{\mathrm{m},23}$ obtained from Eq. (2) with coefficients from Table 2

x_1	$H^{\mathrm{E}}_{\mathrm{m},1+23}$[a] (J mol^{-1})	$H^{\mathrm{E}}_{\mathrm{m},123}$ (J mol^{-1})	x_1	$H^{\mathrm{E}}_{\mathrm{m},1+23}$[a] (J mol^{-1})	$H^{\mathrm{E}}_{\mathrm{m},123}$ (J mol^{-1})	x_1	$H^{\mathrm{E}}_{\mathrm{m},1+23}$[a] (J mol^{-1})	$H^{\mathrm{E}}_{\mathrm{m},123}$ (J mol^{-1})
$x_2/x_3 = 0.3335$, $H^{\mathrm{E}}_{\mathrm{m},23}$ (J mol^{-1}) = 54.99								
0.0499	59.92	112.17	0.4008	325.27	358.22	0.6998	303.41	319.92
0.1001	116.09	165.58	0.4498	337.48	367.73	0.7503	274.03	287.76
0.1500	168.49	215.23	0.4996	344.39	371.91	0.8003	237.54	248.52
0.1999	212.29	256.29	0.5495	344.99	369.77	0.8501	192.60	200.84
0.2499	249.84	291.09	0.5999	338.29	360.29	0.8999	137.93	143.43
0.3001	284.88	323.37	0.6498	324.43	343.69	0.9500	74.47	77.22
0.3506	305.85	341.56						
$x_2/x_3 = 1.0005$, $H^{\mathrm{E}}_{\mathrm{m},23}$ (J mol^{-1}) = 74.64								
0.0500	52.27	123.17	0.4001	274.32	319.10	0.7002	249.36	271.74
0.1000	99.11	166.28	0.4499	284.38	325.44	0.7496	225.55	244.24
0.1500	142.50	205.94	0.5001	288.75	326.06	0.8003	194.00	208.91
0.2001	180.32	240.02	0.5496	290.02	323.64	0.8498	155.88	167.09
0.2501	214.46	270.43	0.5997	281.87	311.75	0.9000	109.72	117.19
0.3016	238.44	290.56	0.6497	269.07	295.22	0.9500	59.09	62.82
0.3499	258.53	307.05						
$x_2/x_3 = 3.0008$, $H^{\mathrm{E}}_{\mathrm{m},23}$ (J mol^{-1}) = 56.08								
0.0500	43.29	96.57	0.3999	227.21	260.86	0.7003	201.27	218.08
0.1000	84.99	135.47	0.4497	234.80	265.67	0.7501	180.39	194.40
0.1500	119.66	167.33	0.5001	237.72	265.75	0.7999	154.45	165.67
0.2000	151.93	196.79	0.5490	234.75	260.05	0.8500	123.73	132.14
0.2501	179.34	221.40	0.6005	228.19	250.59	0.9000	87.14	92.75
0.3001	198.17	237.42	0.6491	218.15	237.83	0.9500	45.70	48.50
0.3497	214.67	251.14						

[a] Ternary term for representing $H^{\mathrm{E}}_{\mathrm{m},1+23}$ by Eqs. (3) and (4): $H^{\mathrm{E}}_{\mathrm{m},T}$ (J mol^{-1}) $= [x_1x_2x_3/(1-x_1+x_2)](-1816.28 + 4177.73x_1 + 3686.09x_2 - 5577.29x_1^2 - 3028.96x_1x_2 - 4392.61x_2^2 + 2572.48x_1^3)$; s (J mol^{-1}) = 1.19.

Table 4
Experimental excess molar enthalpies $H^{E}_{m,1+23}$ at 298.15 K for the addition of THF to a binary mixture of 3MP and *n*C12 to form $x_1C_4H_8O + x_2CH_3CH_2CH(CH_3)CH_2CH_3 + x_3CH_3(CH_2)_{10}CH_3$, and values of $H^{E}_{m,123}$ calculated from Eq. (1) using $H^{E}_{m,23}$ obtained from Eq. (2) with coefficients from Table 2

x_1	$H^{E}_{m,1+23}$ [a] (J mol^{-1})	$H^{E}_{m,123}$ (J mol^{-1})	x_1	$H^{E}_{m,1+23}$ [a] (J mol^{-1})	$H^{E}_{m,123}$ (J mol^{-1})	x_1	$H^{E}_{m,1+23}$ [a] (J mol^{-1})	$H^{E}_{m,123}$ (J mol^{-1})
$x_2/x_3 = 0.3335$, $H^{E}_{m,23}$ (J mol^{-1}) = 54.99								
0.0500	152.52	204.76	0.4001	873.86	906.85	0.7001	856.29	872.78
0.1000	303.86	353.35	0.4503	899.58	929.81	0.7502	787.01	800.75
0.1500	422.39	469.14	0.5003	926.58	954.06	0.8000	693.87	704.87
0.2000	549.53	593.52	0.5494	939.96	964.74	0.8499	573.92	582.18
0.2501	649.33	690.57	0.6000	932.00	953.99	0.9003	421.57	427.05
0.2999	736.34	774.84	0.6536	901.68	920.73	0.9500	233.33	236.08
0.3501	808.64	844.38						
$x_2/x_3 = 1.0005$, $H^{E}_{m,23}$ (J mol^{-1}) = 74.64								
0.0500	153.14	224.05	0.4001	815.46	860.24	0.7006	772.61	794.95
0.1000	292.01	359.18	0.4498	837.69	878.75	0.7502	704.11	722.75
0.1499	415.71	479.16	0.5003	856.88	894.17	0.8000	618.88	633.81
0.1999	520.57	580.28	0.5500	862.00	895.59	0.8499	508.69	519.89
0.2501	616.62	672.59	0.6004	849.74	879.56	0.9000	368.98	376.45
0.2999	691.73	743.98	0.6498	820.74	846.88	0.9500	206.34	210.07
0.3498	758.87	807.39						
$x_2/x_3 = 3.0008$, $H^{E}_{m,23}$ (J mol^{-1}) = 56.08								
0.0499	143.91	197.20	0.4022	749.08	782.60	0.6996	692.03	708.88
0.1001	274.39	324.86	0.4502	773.10	803.94	0.7499	628.26	642.29
0.1500	386.86	434.53	0.4997	787.12	815.18	0.7998	544.81	556.04
0.2000	493.58	538.45	0.5499	785.28	810.52	0.8503	442.39	450.79
0.2501	577.11	619.17	0.6006	769.17	791.57	0.8999	322.11	327.73
0.3000	649.27	688.53	0.6499	740.51	760.14	0.9500	172.31	175.11
0.3500	712.71	749.16						

[a] Ternary term for representing $H^{E}_{m,1+23}$ by Eqs. (3) and (4): $H^{E}_{m,T}$ (J mol^{-1}) $= x_1x_2x_3(-1805.20 + 7330.17x_1 + 4436.42x_2 - 11995.21x_1^2 - 5286.26x_1x_2 - 4855.84x_2^2 + 7463.30x_1^3)$; s (J mol^{-1}) = 3.38.

are listed in Table 2, along with the standard deviation s of the representation. Also included in Table 2 are the representations of $H^{E}_{m,ij}$ for the other constituent-binaries [1,2,8,9].

The experimental results for $H^{E}_{m,1+23}$ and the corresponding values of $H^{E}_{m,123}$ are summarized in Tables 3 and 4 for $x_1\{(CH_3)_2CH\}_2O + x_2CH_3CH_2CH(CH_3)CH_2CH_3 + x_3CH_3(CH_2)_{10}CH_3$ and $x_1C_4H_8O + x_2CH_3CH_2CH(CH_3)CH_2CH_3 + x_3CH_3(CH_2)_{10}CH_3$, respectively. The results for $H^{E}_{m,1+23}$ in Table 3 are plotted in Fig. 1 along with the results reported previously for the constituent-binaries DIPE(1)+3MP(2) [1], and DIPE(1)+*n*C12(3) [8]. The values of $H^{E}_{m,1+23}$ in Table 4 are plotted in Fig. 2 along with the values of $H^{E}_{m,13}$ given in Table 1 for the constituent-binary THF(1) + *n*C12(3) and the results reported previously for THF(1) + 3MP(2) [2]. For both systems, the maximum values of $H^{E}_{m,1+23}$ and $H^{E}_{m,123}$ occur near $x_1 = 0.5$. In the case of the THF system there is a tendency for the maximum values of $H^{E}_{m,1+23}$ to occur at lower values of x_1 as x_2/x_3 increases.

Representation of the values of $H^{E}_{m,1+23}$ was based on the relation:

$$H^{E}_{m,1+23} = \left(\frac{x_2}{1-x_1}\right)H^{E}_{m,12} + \left(\frac{x_3}{1-x_1}\right)H^{E}_{m,13} + H^{E}_{m,T} \quad (3)$$

which consists of a sum of binary contributions [10], and an added ternary term $H^{E}_{m,T}$. The form

$$H^{E}_{m,T}\ (\mathrm{J\,mol^{-1}}) = \left[\frac{x_1x_2x_3}{1+b\{x_2-x_1\}}\right] \times (c_0 + c_1x_1 + c_2x_2 + c_3x_1^2 + c_4x_1x_2 + c_5x_2^2 + c_6x_1^3 + c_7x_1^2x_2 + c_8x_1x_2^2 + c_9x_2^3) \quad (4)$$

which was adopted for the latter is similar to the form used by Morris et al. [11] with the possibility of having an extra skewing factor $(1 + b\{x_2 - x_1\})^{-1}$ for non-zero values of b. The values of the coefficients c_j were adjusted by least-squares analyses in which Eqs. (3) and (4) were fitted to the values of $H^{E}_{m,1+23}$ in Tables 3 and 4, with $b = 1$ for the DIPE system and $b = 0$ for the THF system. In doing this, the values of $H^{E}_{m,ij}$ for the binary contributions were calculated from Eq. (2) using the appropriate coefficients from Table 2. The resulting forms for $H^{E}_{m,T}$ are given in the footnotes of Tables 3 and 4, along with the standard deviations s for the representation of the values of $H^{E}_{m,1+23}$.

The solid curves in Figs. 1 and 2 were calculated from Eq. (3) using values of $H^{E}_{m,T}$ given by the formulae in the footnotes of Tables 3 and 4. It is evident from these, that the representation of the experimental results is quite good.

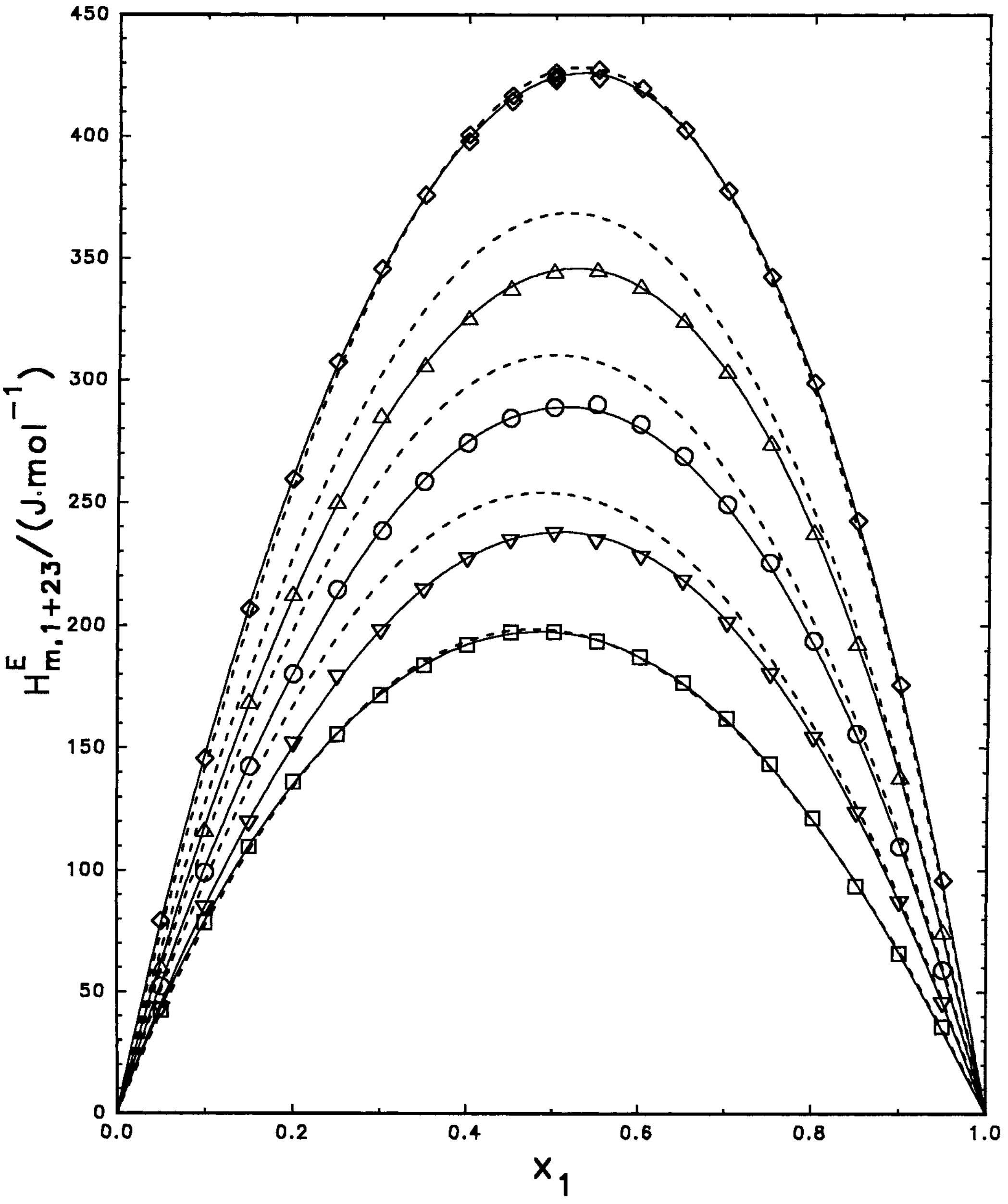

Fig. 1. Excess molar enthalpies, $H^{\mathrm{E}}_{\mathrm{m},1+23}$, for $x_1\{(CH_3)_2CH\}_2O + x_2CH_3CH_2CH(CH_3)CH_2CH_3 + x_3CH_3(CH_2)_{10}CH_3$ mixtures at 298.15 K. Experimental results: (△) $x_2/x_3 = 0.3335$; (○) $x_2/x_3 = 1.0005$; (▽) $x_2/x_3 = 3.0008$; (◇) $x_2 = 0$, Zhu et al. [8]; (□) $x_3 = 0$, Wang et al. [1]; (—) calculated from Eq. (3) with $H^{\mathrm{E}}_{\mathrm{m},T}$ from the footnote of Table 3; (- - -) estimated by the Liebermann–Fried model.

Some constant $H^{\mathrm{E}}_{\mathrm{m},123}$ contours, calculated from Eqs. (1)–(4), are plotted on the Roozeboom diagrams in part (a) of Figs. 3 and 4. The general characteristics of these are similar. In both figures, all of the contours extend to the edges of the triangle, and there is no indication of an internal extremum.

In our earlier work [1,2] on the enthalpies of the analogous systems containing *n*C8 or *n*C10 in place of *n*C12, it was found that the Liebermann–Fried model [12,13] could provide useful estimates of the ternary enthalpies using only the properties of the pure components and interaction parameters derived from analyses of the excess enthalpies of their constituent-binaries. This approach was investigated for the present systems. Reference can be made to the work of Wang et al. [14] for the equations used in this application.

The values of the Liebermann–Fried interaction parameters A_{ij} and A_{ji} for the constituent-binaries are given in Table 5. These were obtained by fitting the Liebermann–Fried formula for $H^{\mathrm{E}}_{\mathrm{m},ij}$ to the primary experimental data for the excess molar enthalpies, as given in Table 1 and Refs. [1,2,8,9]. Also included in the table are

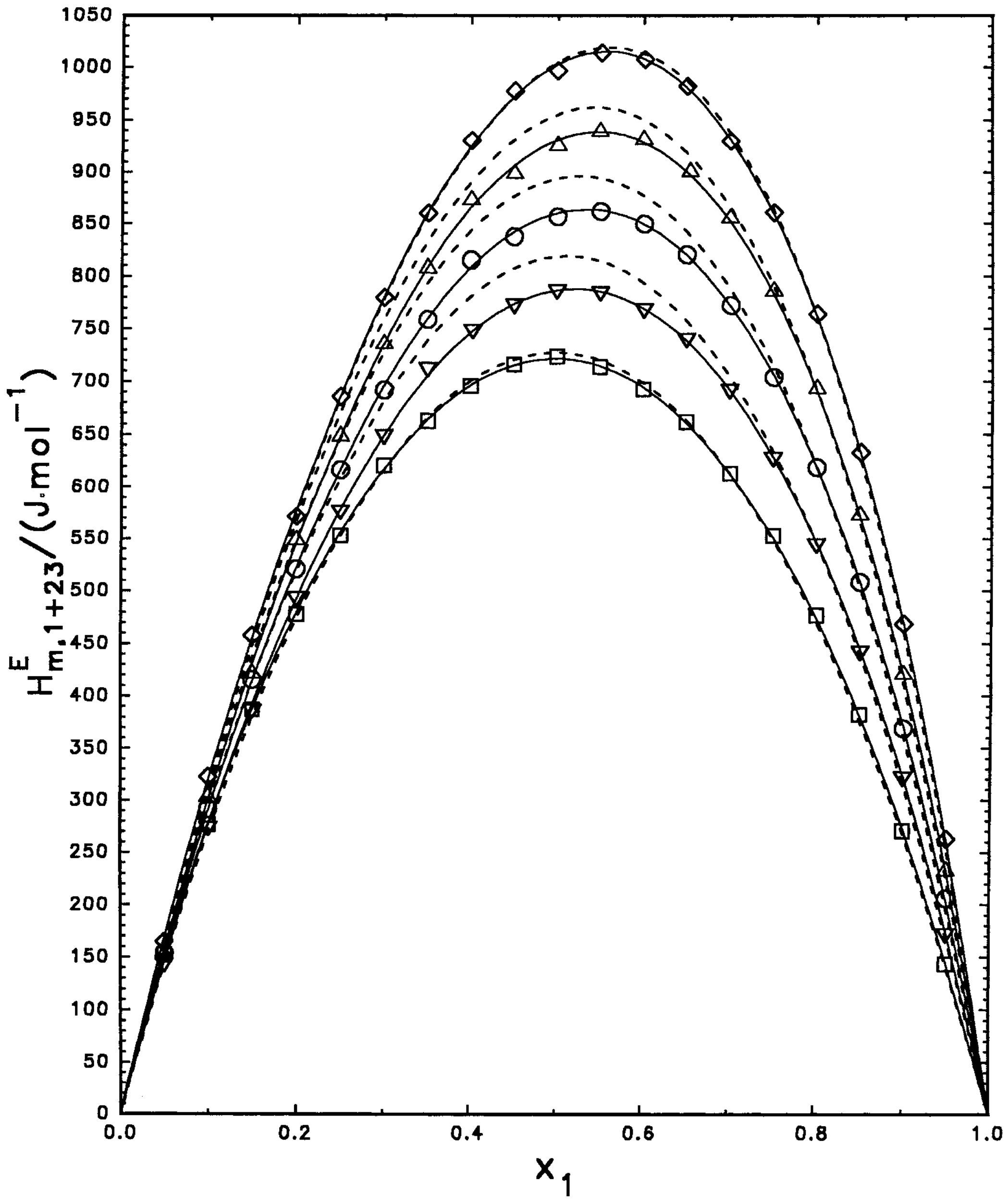

Fig. 2. Excess molar enthalpies, $H^{\mathrm{E}}_{\mathrm{m},1+23}$, for $x_1\mathrm{C_4H_8O} + x_2\mathrm{CH_3CH_2CH(CH_3)CH_2CH_3} + x_3\mathrm{CH_3(CH_2)_{10}CH_3}$ mixtures at 298.15 K. Experimental results: (△) $x_2/x_3 = 0.3335$; (○) $x_2/x_3 = 1.0005$; (▽) $x_2/x_3 = 3.0008$; (◇) $x_2 = 0$; (□) $x_3 = 0$, Wang et al. [2]; (—) calculated from Eq. (3) with $H^{\mathrm{E}}_{\mathrm{m},T}$ from the footnote of Table 4; (- - -) estimated by the Liebermann–Fried model.

Table 5

Values of the interaction parameters A_{ij} and A_{ji}, standard deviation s and the isobaric thermal expansivity α_{p} at 298.15 K, for Liebermann and Fried model calculations

i	*j*	A_{ij}	A_{ji}	s (J mol^{-1})	α_{p} (kK^{-1})	
					i	*j*
DIPE	3MP	0.8486	1.0156	1.27	1.455[a]	1.396[b]
DIPE	*n*C12	0.9665	0.7448	3.13	1.455[a]	0.960[c]
3MP	*n*C12	1.7027	0.8505	0.14	1.396[b]	0.960[c]
THF	3MP	0.7791	0.8045	5.46	1.138[b]	1.396[b]
THF	*n*C12	0.9284	0.5599	5.84	1.138[b]	0.960[c]

[a] From Obama et al. [3].

[b] From Riddick et al. [15].

[c] From Benson et al. [16].

values of the standard deviation s achieved in the fitting process, and values of the isobaric expansivity α_{p} [3,15,16], used in evaluating the contributions due to different sizes of the molecules.

Estimates of $H^{\mathrm{E}}_{\mathrm{m},1+23}$, derived from the Liebermann–Fried model, are shown as dashed curves in Figs. 1 and 2. It can be seen that, although the fits of the constituent-binaries are reasonable, as indicated by the standard deviations in Table 5, the model tends to over-estimates $H^{\mathrm{E}}_{\mathrm{m},1+23}$ for the ternary mixtures. For the 57 points in each of Tables 3 and 4, the root mean square deviations are 15.3 and 22.0 J mol^{-1}, respectively.

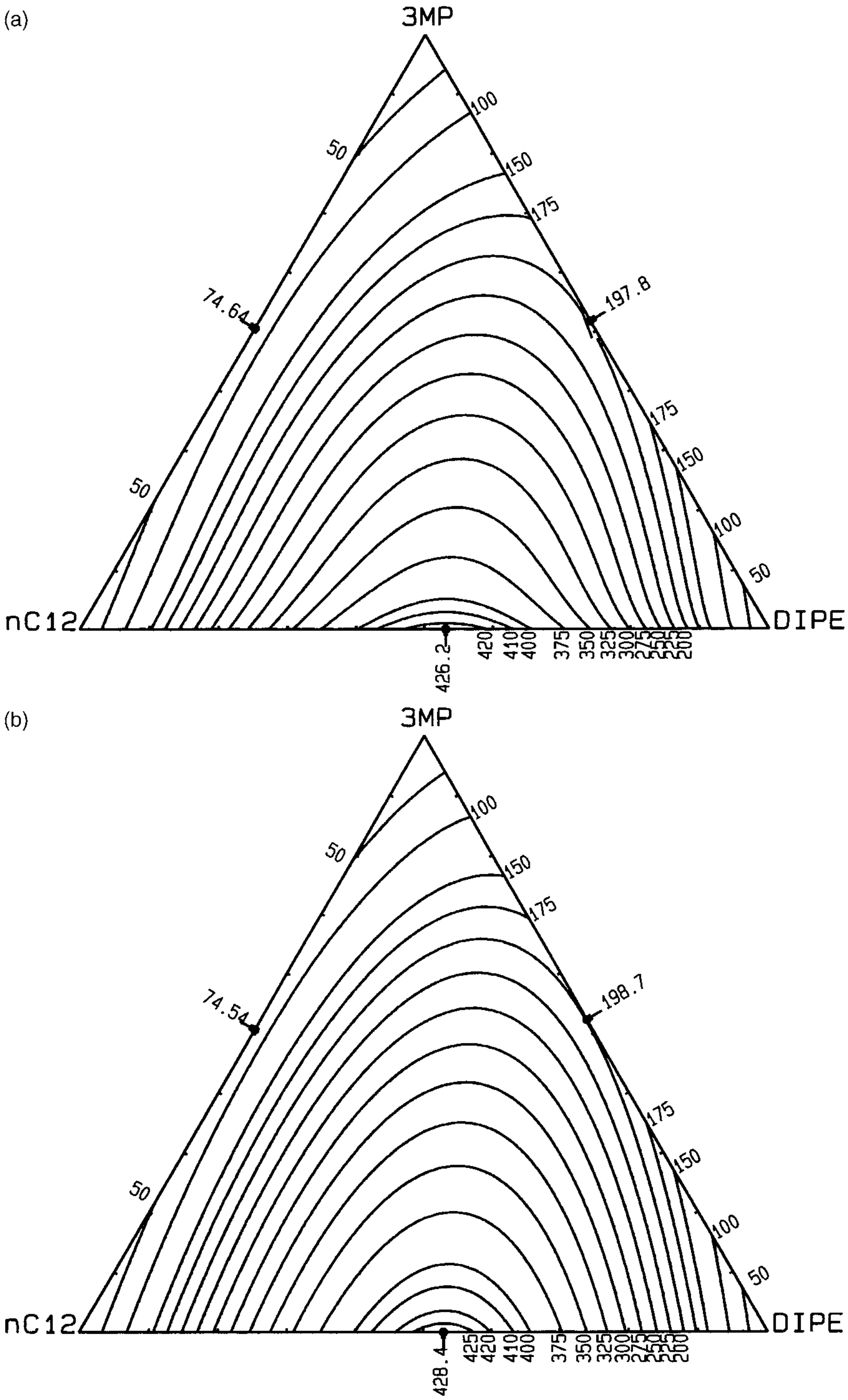

Fig. 3. Contours for constant values of $H^{E}_{m,123}$ ($J\,mol^{-1}$) for $x_1\{(CH_3)_2CH\}_2O + x_2CH_3CH_2CH(CH_3)CH_2CH_3 + x_3CH_3(CH_2)_{10}CH_3$ mixtures at 298.15 K: (a) calculated from the representation of the experimental results by Eqs. (1)–(4) with $H^{E}_{m,T}$ from the footnote of Table 3; (b) estimated by the Liebermann–Fried model.

Constant $H^{E}_{m,123}$ contours, estimated on the basis of the model, are shown on the Roozeboom diagrams in part (b) of Figs. 3 and 4. Although differing in detail, the overall characteristics of these are similar to those in parts (a), which were derived from the representations of the experimental data for the ternary mixtures. It can be seen from these comparisons, that the Liebermann–Fried model provides useful predictions of the behavior of $H^{E}_{m,123}$ for both of the present systems, without requiring the study of any ternary mixtures.

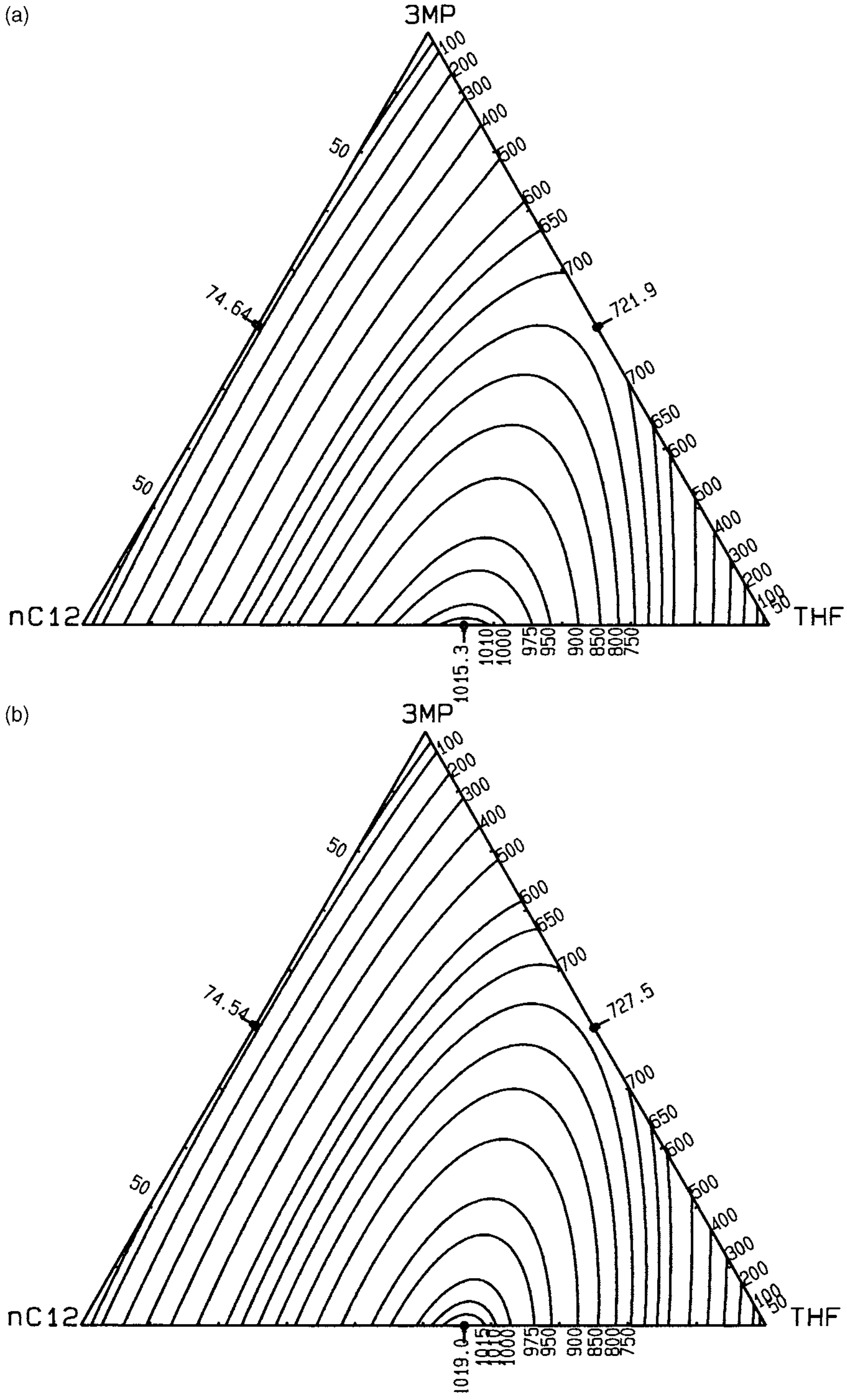

Fig. 4. Contours for constant values of $H^{E}_{m,123}$ ($J\,mol^{-1}$) for $x_1C_4H_8O + x_2CH_3CH_2CH(CH_3)CH_2CH_3 + x_3CH_3(CH_2)_{10}CH_3$ mixtures at 298.15 K: (a) calculated from the representation of the experimental results by Eqs. (1)–(4) with $H^{E}_{m,T}$ from the footnote of Table 4; (b) estimated by the Liebermann–Fried model.

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