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Excess molar enthalpies of the ternary mixtures: ethyl *tert*-butylether + 2,2,4-trimethylpentane + (*n*-heptane or *n*-octane)

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Abstract

Microcalorimetric measurements of excess molar enthalpies, at 298.15 K, are reported for the two ternary mixtures $x_1CH_3CH_2OC(CH_3)_3 + x_2(CH_3)_3CCH_2CH(CH_3)_2 + x_3CH_3(CH_2)_vCH_3$, where $v = 5$ and 6. Smooth representations of the results are presented and used to construct constant excess molar enthalpy contours on Roozeboom diagrams. It is shown that good 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.

Keywords: Ethyl *tert*-butylether; 2,2,4-Trimethylpentane; *n*-Heptane; *n*-Octane; Excess molar enthalpy; Ternary system

1. Introduction

A recent paper [1], from our laboratory, reported excess molar enthalpies for the ternary systems consisting of ethyl *tert*-butylether (ETBE), *n*-hexane (*n*-C6) together with either *n*-heptane (*n*-C7) or *n*-octane (*n*-C8). As an extension of that investigation, similar measurements have been made for the analogous systems, in which 2,2,4-trimethylpentane (TMP) replaced the *n*-C6 used previously.

2. Experimental

The ETBE, used for the present measurements, was obtained from TCI America and had a stated purity of 97+ mol%. The TMP (HPLC Grade, with purity > 99.7 mol%), *n*-C7 and *n*-C8 were obtained from Sigma–Aldrich. The *n*-alkanes had purities of at least 99 mol%. Apart from partial degassing, all of the components were used as received from the manufacturers. Densities, measured at 298.15 K in an Anton–Paar digital densimeter, were 735.39, 687.92, 679.93 and 698.80 kg m^{-3} for ETBE, TMP, *n*-C7 and *n*-C8, respectively. These are in reasonable agreement with values in [2,3].

An LKB flow microcalorimeter (Model 10700-1), maintained at 298.150 ± 0.003 K, was used to measure the excess molar enthalpies H_m^E. Details of the equipment and the operating procedure have been described previously [4,5].

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In studying the ternary systems $x_1CH_3CH_2OC(CH_3)_3 + x_2(CH_3)_3CCH_2CH(CH_3)_2 + x_3CH_3(CH_2)_v CH_3$, for $v = 5$ and 6, the excess molar enthalpy $H^E_{m,1+23}$ was determined for several pseudo-binary systems in which ETBE 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

$$H^E_{m,123} = H^E_{m,1+23} + (1 - x_1)H^E_{m,23} \qquad (1)$$

where $H^E_{m,23}$ is the excess molar enthalpy of the particular binary mixture of TMP and either *n*-C7 or *n*-C8. Over most of the mole fraction range of component 1, the errors of $H^E_{m,1+23}$ are estimated to be <0.5%. Errors in the mole fractions of the final ternary mixtures are estimated to be $<5 \times 10^{-4}$.

3. Results and discussion

Excess molar enthalpies $H^E_{m,ij}$ $(i < j)$, at $T = 298.15$ K, for four of the five constituent binary systems of present interest, have been reported in our previous papers: ETBE(1) + *n*-C7(3) [6], TMP(2) + *n*-C7(3) [7], ETBE(1) + *n*-C8(3) [6] and TMP(2) + *n*-C8(3) [7]. The experimental values of x_1 and $H^E_{m,12}$, measured at 298.15 K for ETBE(1)+ TMP(2) are summarized in Table 1. These results were fitted with the smoothing function

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

by the method of least-squares with all points weighted equally. The values of the coefficients h_k, obtained from the analysis, 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 five other constituent-binaries.

The excess molar enthalpies reported by Mier et al. [8] for ETBE(1) + TMP(2) at 298.15 K are larger than the present results, at all mole fractions. For an equimolar mixture, the difference is more than 60 J mol^{-1}. It is unlikely that this difference can be attributed to differences in the purities of the samples of ETBE used in the two investigations.

Table 1
Experimental excess molar enthalpies $H^E_{m,12}$ for $x_1CH_3CH_2OC(CH_3)_3 + x_2(CH_3)_3CCH_2CH(CH_3)_2$ at the temperature 298.15 K

x_1	$H^E_{m,12}$ (J mol^{-1})	x_1	$H^E_{m,12}$ (J mol^{-1})	x_1	$H^E_{m,12}$ (J mol^{-})	x_1	$H^E_{m,12}$ (J mol^{-1})
0.0499	45.27	0.3002	199.04	0.5501	234.50	0.7999	151.23
0.1001	85.65	0.3501	214.76	0.6001	227.62	0.8501	122.06
0.1501	121.84	0.3998	226.84	0.6504	216.23	0.9002	86.59
0.1999	154.59	0.4496	232.90	0.6999	199.80	0.9499	45.60
0.2501	180.22	0.5006	236.45	0.7503	179.05		

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

Component		h_1	h_2	h_3	h_4	s (J mol^{-1})
i	j					
ETBE	TMP	945.80	0.54	25.09		0.95
ETBE	*n*-C7[a]	1124.24	12.58	61.80	70.75	0.68
TMP	*n*-C7[b]	44.03	−0.80			0.10
ETBE	*n*-C8[a]	1242.87	−20.94	56.10	59.69	1.18
TMP	*n*-C8[b]	104.49	−0.032	1.88		0.10

[a] From Peng et al. [6].

[b] From Peng et al. [7].

The experimental results for $H^{\mathrm{E}}_{\mathrm{m},1+23}$ and the corresponding values of $H^{\mathrm{E}}_{\mathrm{m},123}$ are summarized in Tables 3 and 4 for $x_1\mathrm{CH_3CH_2OC(CH_3)_3} + x_2\mathrm{(CH_3)_3CCH_2CH(CH_3)_2} + x_3\mathrm{CH_3(CH_2)_{\mathit{v}}CH_3}$, with $v = 5$ and 6, respectively. The results for $H^{\mathrm{E}}_{\mathrm{m},1+23}$ in Table 3 are plotted in Fig. 1, and those in Table 4 are plotted in Fig. 2. Also plotted in these figures are the results from Table 1 for ETBE(1) + TMP(2), which corresponds to the case $x_3 = 0$, and curves for $x_2 = 0$, calculated from Eq. (2) with the coefficients given in Table 2.

For both systems, the maximum values of $H^{\mathrm{E}}_{\mathrm{m},1+23}$ and $H^{\mathrm{E}}_{\mathrm{m},123}$ occur near $x_1 = 0.5$. The values of $H^{\mathrm{E}}_{\mathrm{m},1+23}$ fall between the results for the two constituent binaries and at constant x_1, $H^{\mathrm{E}}_{\mathrm{m},1+23}$ decreases monotonically as x_2/x_3 increases.

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

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

which consists of a sum of binary contributions [9], and an added ternary term $H^{\mathrm{E}}_{\mathrm{m},T}$. The form

$$H^{\mathrm{E}}_{\mathrm{m},T} = x_1x_2x_3(c_0 + c_1x_1 + c_2x_2 + c_3x_1^2 + c_4x_1x_2 + c_2x_2^2 + \cdots) \qquad (4)$$

which was adopted for the latter is similar to the form used by Morris et al. [10]. 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 $H^{\mathrm{E}}_{\mathrm{m},1+23}$ in Tables 3 and 4. In doing this, the values of

Table 3

Experimental excess molar enthalpies $H^{\mathrm{E}}_{\mathrm{m},1+23}$ at 298.15 K for the addition of ETBE to a binary mixture of TMP and *n*-C7 to form $x_1\mathrm{CH_3CH_2OC(CH_3)_3} + x_2\mathrm{(CH_3)_3CCH_2CH(CH_3)_2} + x_3\mathrm{CH_3(CH_2)_5CH_3}$, 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	$H^{\mathrm{E}}_{\mathrm{m},1+23}$ (J mol^{-1})[a]	$H^{\mathrm{E}}_{\mathrm{m},123}$ (J mol^{-1})	x_1	$H^{\mathrm{E}}_{\mathrm{m},1+23}$ (J mol^{-1})[a]	$H^{\mathrm{E}}_{\mathrm{m},123}$ (J mol^{-1})	x_1	$H^{\mathrm{E}}_{\mathrm{m},1+23}$ (J mol^{-1})[a]	$H^{\mathrm{E}}_{\mathrm{m},123}$ (J mol^{-1})
$x_2/x_3 = 0.3341$, $H^{\mathrm{E}}_{\mathrm{m},23}$ (J mol^{-1}) = 8.19								
0.0499	51.25	59.03	0.4006	260.57	265.48	0.7004	223.57	226.02
0.1002	100.17	107.54	0.4504	267.87	272.37	0.7506	199.79	201.83
0.1498	142.09	149.05	0.5001	268.36	272.45	0.7998	171.56	173.20
0.2010	177.31	183.85	0.5506	265.66	269.34	0.8500	135.32	136.55
0.2496	207.58	213.73	0.6002	256.87	260.14	0.8997	97.00	97.82
0.2992	228.62	234.36	0.6505	241.72	244.58	0.9500	50.69	51.10
0.3505	248.64	253.96						
$x_2/x_3 = 1.0000$, $H^{\mathrm{E}}_{\mathrm{m},23}$ (J mol^{-1}) = 11.01								
0.0501	50.13	60.59	0.4001	248.88	255.48	0.7003	213.80	217.10
0.1001	95.95	105.85	0.4504	254.80	260.85	0.7498	192.93	195.68
0.1499	134.65	144.01	0.5006	256.95	262.45	0.7998	164.49	166.69
0.2001	168.47	177.27	0.5500	254.07	259.02	0.8501	131.13	132.78
0.2502	195.84	204.09	0.6001	246.25	250.65	0.9005	89.81	90.90
0.3008	217.51	225.21	0.6502	233.07	236.92	0.9496	48.06	48.61
0.3497	237.17	244.33						
$x_2/x_3 = 3.0005$, $H^{\mathrm{E}}_{\mathrm{m},23}$ (J mol^{-1}) = 8.33								
0.0501	46.31	54.22	0.4006	236.67	241.66	0.7005	206.82	209.31
0.1001	89.15	96.65	0.4505	245.68	250.26	0.7495	185.40	187.49
0.1501	128.18	135.26	0.5000	246.93	251.09	0.8006	155.09	156.75
0.1998	161.07	167.74	0.5501	243.69	247.44	0.8502	124.77	126.02
0.2501	187.73	193.98	0.5999	236.32	239.65	0.8998	89.22	90.05
0.2996	207.82	213.65	0.6503	223.88	226.79	0.9500	46.80	47.22
0.3502	224.88	230.29						

[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}) $= x_1x_2x_3(-257.58 + 1363.63x_1 - 18.76x_2 - 1844.59x_1^2)$; s (J mol^{-1}) = 1.00.

Table 4
Experimental excess molar enthalpies $H^{\mathrm{E}}_{\mathrm{m},1+23}$ at 298.15 K for the addition of ETBE to a binary mixture of TMP and *n*-C8 to form $x_1\mathrm{CH_3CH_2OC(CH_3)_3} + x_2\mathrm{(CH_3)_3CCH_2CH(CH_3)_2} + x_3\mathrm{CH_3(CH_2)_6CH_3}$, 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	$H^{\mathrm{E}}_{\mathrm{m},1+23}$ (J mol^{-1})[a]	$H^{\mathrm{E}}_{\mathrm{m},123}$ (J mol^{-1})	x_1	$H^{\mathrm{E}}_{\mathrm{m},1+23}$ (J mol^{-1})[a]	$H^{\mathrm{E}}_{\mathrm{m},123}$ (J mol^{-1})	x_1	$H^{\mathrm{E}}_{\mathrm{m},1+23}$ (J mol^{-1})[a]	$H^{\mathrm{E}}_{\mathrm{m},123}$ (J mol^{-1})
x_2/x_3 = 0.3342, $H^{\mathrm{E}}_{\mathrm{m},23}$ (J mol^{-1}) = 19.70								
0.0500	54.04	72.76	0.3998	273.81	285.63	0.6497	262.99	269.89
0.1002	104.58	122.31	0.4004	275.10	286.91	0.7002	243.71	249.62
0.1501	147.60	164.34	0.4498	283.58	294.42	0.7499	218.71	223.64
0.2000	188.93	204.69	0.4995	287.31	297.17	0.8003	187.17	191.10
0.2501	218.38	233.15	0.4999	287.06	296.91	0.8500	149.90	152.85
0.3000	242.68	256.47	0.5502	283.56	292.42	0.9000	106.90	108.87
0.3001	241.11	254.90	0.5999	276.55	284.43	0.9500	56.75	57.74
0.3502	263.64	276.44						
x_2/x_3 = 1.0004, $H^{\mathrm{E}}_{\mathrm{m},23}$ (J mol^{-1}) = 26.12								
0.0500	50.22	75.04	0.3998	258.47	274.15	0.7001	226.86	234.69
0.1001	98.47	121.98	0.4502	265.42	279.78	0.7502	203.40	209.93
0.1500	139.38	161.58	0.4999	268.29	281.35	0.8000	174.13	179.36
0.1999	175.15	196.05	0.5499	265.90	277.66	0.8502	138.00	141.91
0.2497	205.04	224.64	0.5999	258.13	268.58	0.9000	99.01	101.62
0.3002	225.90	244.18	0.6499	244.48	253.62	0.9500	51.87	53.18
0.3499	244.94	261.92						
x_2/x_3 = 3.0003, $H^{\mathrm{E}}_{\mathrm{m},23}$ (J mol^{-1}) = 19.68								
0.0500	47.41	66.11	0.3983	240.42	252.26	0.6995	212.51	218.42
0.0999	91.64	109.36	0.4500	248.01	258.83	0.7496	190.14	195.07
0.1499	129.96	146.69	0.4993	251.27	261.12	0.7998	162.28	166.22
0.1998	163.41	179.16	0.5500	248.97	257.83	0.8499	130.30	133.25
0.2499	190.93	205.69	0.5998	241.63	249.51	0.8999	92.17	94.14
0.3003	211.06	224.83	0.6501	229.02	235.91	0.9499	49.88	50.87
0.3499	228.18	240.98						

[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}) $= x_1x_2x_3(-274.40 + 704.89x_1 + 138.39x_2 - 1165.62x_1^2)$; s (J mol^{-1}) = 1.07.

$H^{\mathrm{E}}_{\mathrm{m},ij}$ for the binary contributions were calculated from Eq. (2) using the appropriate coefficients from Table 2. The resulting forms for $H^{\mathrm{E}}_{\mathrm{m},T}$ are given in the footnotes of Tables 3 and 4, along with the standard deviations s for the representation of the values of $H^{\mathrm{E}}_{\mathrm{m},1+23}$.

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

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. In both cases, there is no indication of an internal extremum, and the maximum value of $H^{\mathrm{E}}_{\mathrm{m},123}$ is located in the edge of the plot for the constituent binary comprising ETBE and the *n*-alkane.

Recently [1,6,11,12], we have found that the Liebermann–Fried model [13,14] can be extended to provide estimates of the thermodynamics properties of multicomponent systems, 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. [1] 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 in [6,7]. Also included in the table are values of the standard deviation s achieved in the

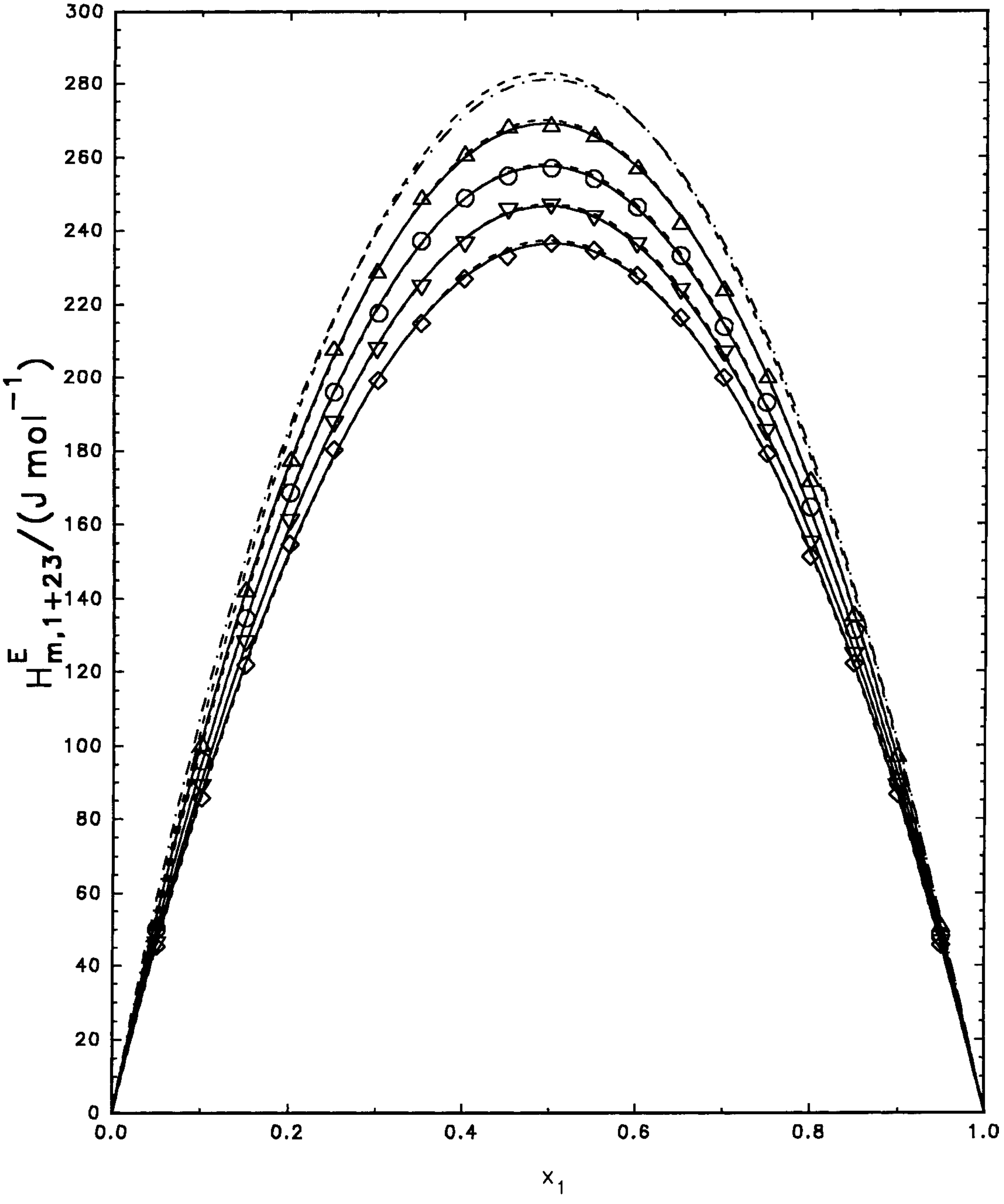

Fig. 1. Excess molar enthalpies, $H^E_{m,1+23}$, for $x_1CH_3CH_2OC(CH_3)_3 + x_2(CH_3)_3CCH_2CH(CH_3)_2 + x_3CH_3(CH_2)_5CH_3$ mixtures at 298.15 K. Experimental results: (◇), $x_3 = 0$; (△), $x_2/x_3 = 0.3341$; (○), $x_2/x_3 = 1.0000$; (▽), $x_2/x_3 = 3.0005$. Curves: (—) calculated from Eq. (3) with $H^E_{m,T}$ from the footnote of Table 3; (—·—·—), $x_2 = 0$, [6]; (- - -), estimated by the Liebermann–Fried model.

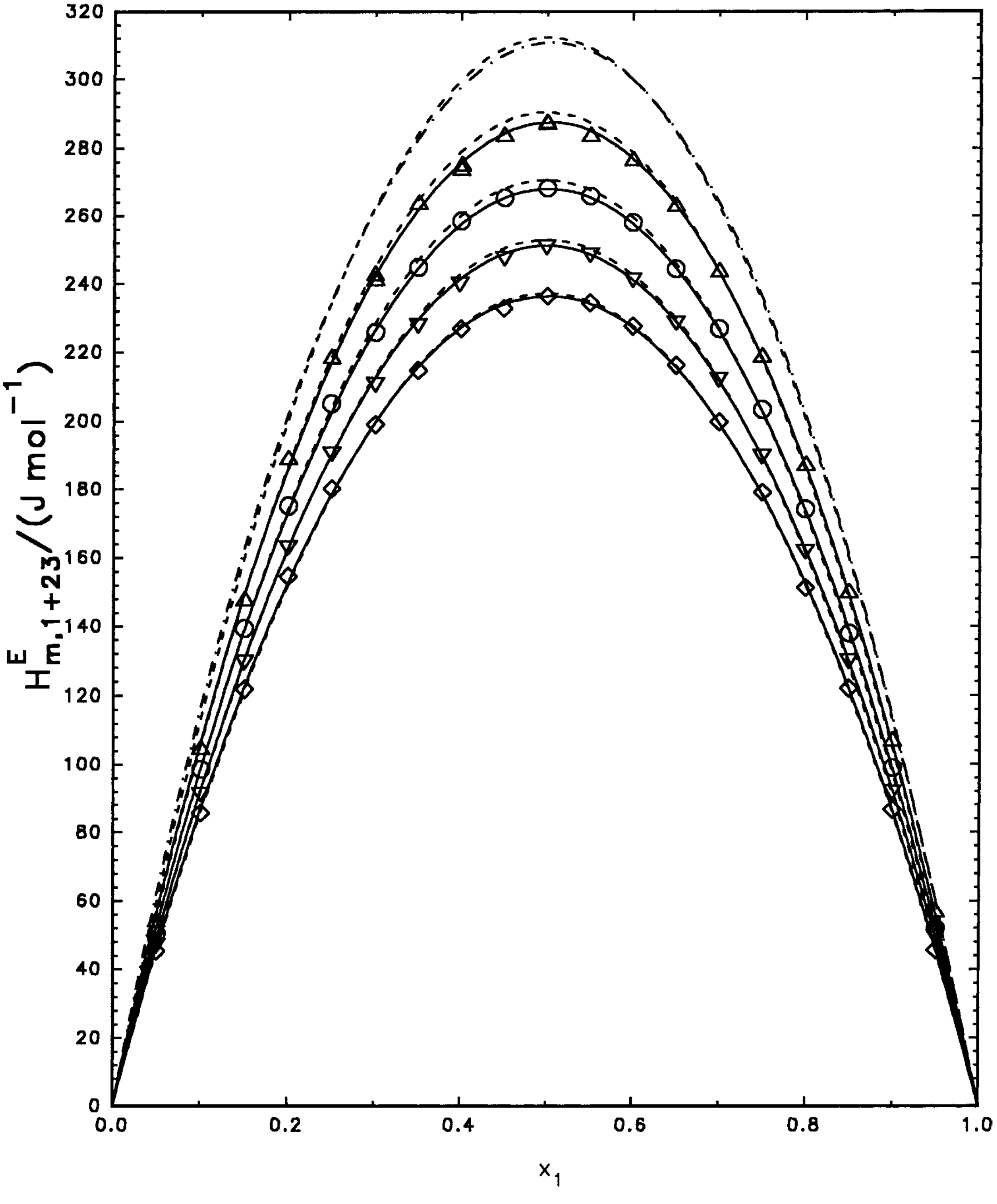

Fig. 2. Excess molar enthalpies, $H^{E}_{m,1+23}$, for $x_1CH_3CH_2OC(CH_3)_3 + x_2(CH_3)_3CCH_2CH(CH_3)_2 + x_3CH_3(CH_2)_6CH_3$ mixtures at 298.15 K. Experimental results: (◇), $x_3 = 0$; (△), $x_2/x_3 = 0.3342$; (○), $x_2/x_3 = 1.0004$; (▽), $x_2/x_3 = 3.0003$. Curves: (—) calculated from Eq. (3) with $H^{E}_{m,T}$ from the footnote of Table 4; (—·—·—), $x_2 = 0$, [6]; (- - -), estimated by the Liebermann–Fried model.

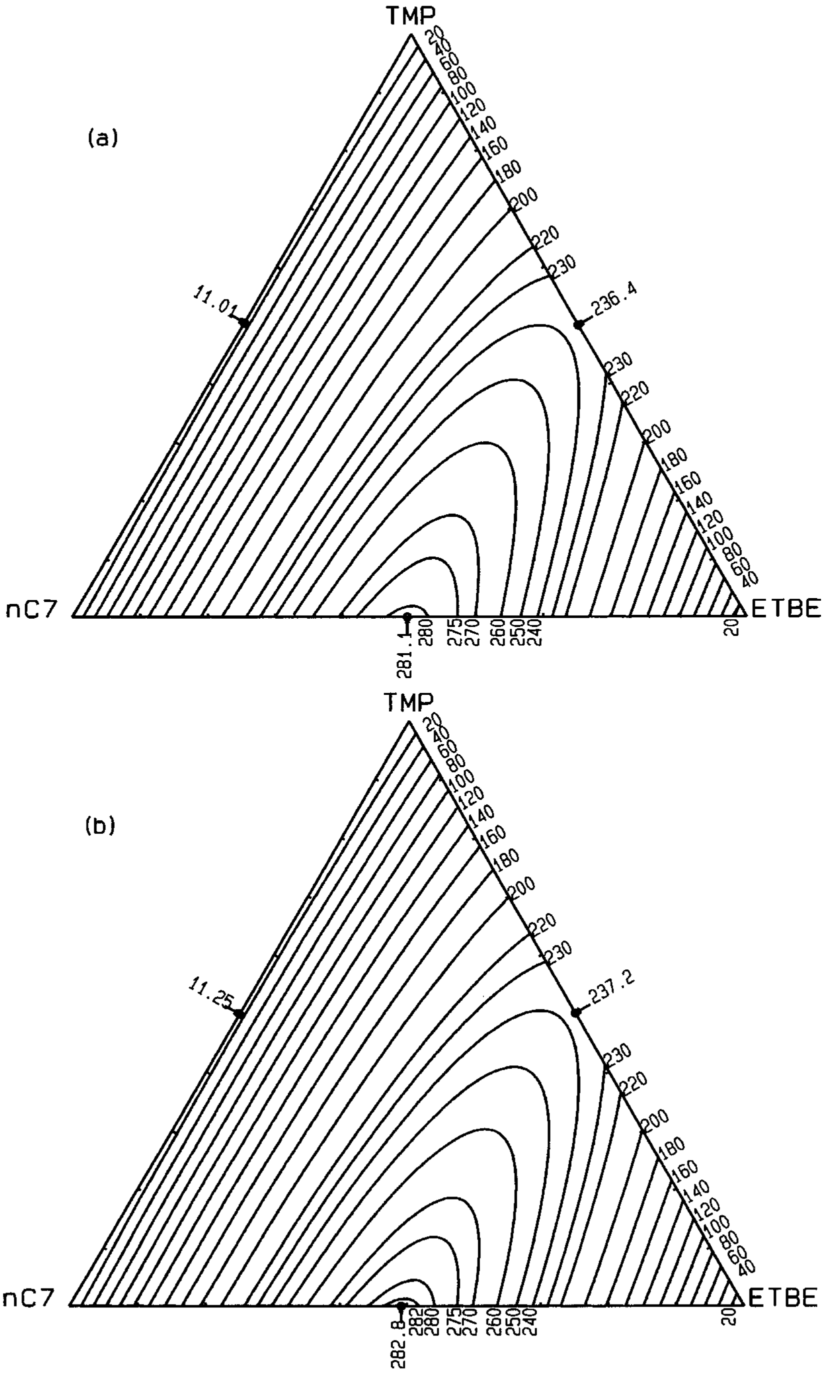

Fig. 3. Contours for constant values of $H^{E}_{m,123}$ (J mol^{-1}) for $x_1CH_3CH_2OC(CH_3)_3 + x_2(CH_3)_3CCH_2CH(CH_3)_2 + x_3CH_3(CH_2)_5CH_3$ mixtures at 298.15 K. (a) Calculated from the representation of the experimental results by Eqs. (1)–(4) with $H^{E}_{m,T}$ from the footnote of Table 3 and (b) estimated by the Liebermann–Fried model.

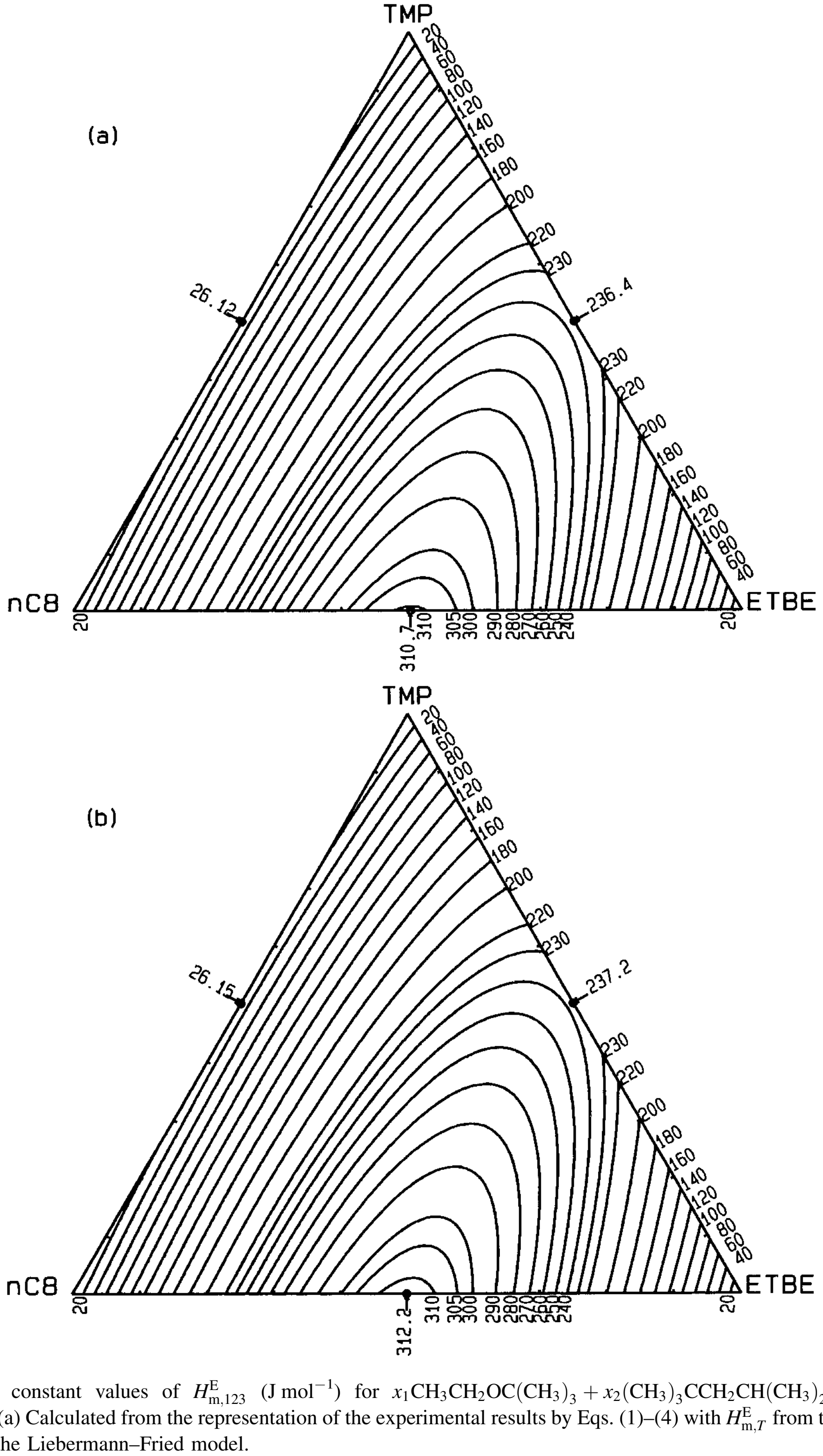

Fig. 4. Contours for constant values of $H^{\mathrm{E}}_{\mathrm{m},123}$ (J mol^{-1}) for $x_1\mathrm{CH_3CH_2OC(CH_3)_3} + x_2\mathrm{(CH_3)_3CCH_2CH(CH_3)_2} + x_3\mathrm{CH_3(CH_2)_6CH_3}$ mixtures at 298.15 K. (a) Calculated from the representation of the experimental results by Eqs. (1)–(4) with $H^{\mathrm{E}}_{\mathrm{m},T}$ from the footnote of Table 4 and (b) estimated by the Liebermann–Fried model.

Table 5
Values of the interaction parameters A_{ij} and A_{ji}, standard deviation s and isobaric thermal expansivity α_p at 298.15 K, for Liebermann–Fried model calculations

Component		A_{ij}	A_{ji}	s (J mol^{-1})	α_p (kK^{-1})	
i	j				i	j
ETBE	TMP	0.9153	0.9118	1.12	1.401[a]	1.197[b]
ETBE	*n*-C7	0.8726	0.9305	2.10	1.401[a]	1.256[c]
TMP	*n*-C7	0.9830	1.0071	0.24	1.197[b]	1.256[c]
ETBE	*n*-C8	0.8930	0.8883	2.03	1.401[a]	1.164[c]
TMP	*n*-C8	0.9901	0.9892	0.12	1.197[b]	1.164[c]

[a] From Obama et al. [2].

[b] From Rajagopal and Subrahmanyam [15].

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

fitting process, and values of the isobaric expansivity α_p [2,15,16], used in evaluating the contributions due to different sizes of the molecules.

Estimates of $H^{E}_{m,1+23}$, derived from the Liebermann–Fried model, are shown as dashed curves in Figs. 1 and 2. It can be seen that there is a tendency for the model to over-estimate $H^{E}_{m,1+23}$. For the 57 points in Table 3 and the 60 points in Table 4, the root mean square deviations are 0.98 and 1.96 J mol^{-1}, respectively.

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. As found previously [1] for the analogous systems containing *n*-C6 in place of TMP, it is clear from a comparison of parts (a) and (b) that the Liebermann–Fried model provides useful estimates of $H^{E}_{m,123}$ for both of the present systems.

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