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Excess molar enthalpies of methyl *tert*-butyl ether + *n*-hexane + (*n*-decane or *n*-dodecane) ternary mixtures at 298.15 K

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Abstract

Excess molar enthalpies, measured at 298.15 K in a flow microcalorimeter, are reported for the ternary mixtures $x_1C_5H_{12}O + x_2C_6H_{14} + x_3C_vH_{2v+2}$, with $v = 10$ and 12. Smooth representations of the results are described and used to construct contours of constant enthalpy on a Roozeboom diagram. An analysis of the results in terms of the Flory theory of mixtures is also presented.

INTRODUCTION

Recent papers [1, 2] from our laboratory have described measurements of the excess molar enthalpies of some ternary systems containing an alcohol (ethanol or *n*-propanol) and two *n*-alkanes (C_6, C_{10} or C_{12}). The results enable estimates of the temperature variation of the vapor–liquid equilibria in such mixtures to be made, and are useful in considering the alcohols as gasoline blending agents.

Methyl *tert*-butyl ether (MTBE) is also of interest as a relatively cheap blending agent with high octane number. Extending our earlier work, we have now measured excess molar enthalpies for the two ternary systems (MTBE + *n*-hexane + *n*-decane) and (MTBE + *n*-hexane + *n*-dodecane), both at 298.15 K.

EXPERIMENTAL

Materials

MTBE (HPLC grade), with stated purity exceeding 99 mol.%, was obtained from the Aldrich Chemical Co. Its density, measured at 298.15 K

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in an Anton-Paar digital densimeter, was 735.67 kg m^{-3}. The n-hexane, n-decane, and n-dodecane were the same as used previously [1, 2]. The compositions of the mixtures are stated in terms of the mole fractions, x_i of their components; the subscripts 1, 2, and 3 are used to designate MTBE, n-hexane, and either n-decane or n-dodecane, respectively.

Equipment and technique

Excess molar enthalpies H_m^E were measured in an LKB flow microcalorimeter (Model 10700-1) thermostated at 298.150 ± 0.002 K. This equipment and its modifications have been described previously [3, 4].

In studying the ternary systems $x_1C_5H_{12}O + x_2C_6H_{14} + x_3C_vH_{2v+2}$ for $v = 10$ or 12, the excess molar enthalpy $H_{m,1+23}^E$ was determined for several pseudo-binary systems in which MTBE was added to binary mixtures of components 2 and 3 having a fixed mole ratio x_2/x_3. These binaries were prepared by weighing. The excess molar enthalpy $H_{m,123}^E$ of the ternary system was then obtained from the relation

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

where $H_{m,23}^E$ is the excess molar enthalpy of the particular binary n-alkane mixture. Values of $H_{m,23}^E$ for n-hexane + n-decane and n-hexane + n-dodecane were taken from the literature [5, 6]. The errors of $H_{m,1+23}^E$ are estimated to be less than 0.5% over most of the MTBE mole fraction range. Errors in the mole fractions of the final ternary mixtures are estimated to be less than 5×10^{-4}.

RESULTS AND DISCUSSION

Binary mixtures

Table 1 summarizes the excess molar enthalpies $H_{m,1j}^E$ measured for the three binary systems formed by mixing MTBE with each of the n-alkanes. Representations of the results in the form

$$H_{m,1j}^E = x_1(1 - x_1)\sum_{k=1}^{n} h_k z^{k-1} \qquad z = 1 - 2x_1 \qquad j > 1 \tag{2}$$

and the standard deviations s of these representations are given in the footnotes to the table. The experimental data and their representations by eqn. (2) are plotted in Fig. 1.

We are not aware of any directly comparable previous results for these systems. However, the system MTBE + n-heptane has been investigated by Tusel-Langer et al. [7]. The dotted curve shown in Fig. 1 was calculated from the smoothing equation reported by those authors. It lies somewhat closer to our curve for n-hexane than might be expected from the results for the other n-alkanes.

TABLE 1

Experimental results for the excess molar enthalpies $H^{\mathrm{E}}_{\mathrm{m},1j}$ of $x_1\mathrm{C_5H_{12}O} + x_j\mathrm{C}_v\mathrm{H}_{2v+2}$ mixtures at 298.15 K and their representations by eqn. (2)

$v = 6$		$v = 10$		$v = 12$	
x_1	$H^{\mathrm{E}}_{\mathrm{m},12}$ [a] (J mol^{-1})	x_1	$H^{\mathrm{E}}_{\mathrm{m},13}$ [b] (J mol^{-1})	x_1	$H^{\mathrm{E}}_{\mathrm{m},13}$ [c] (J mol^{-1})
0.0501	69.4	0.0500	81.5	0.0500	85.2
0.0501	68.9	0.1002	156.3	0.0500	88.5
0.1000	131.5	0.1501	226.3	0.0999	172.9
0.1000	135.2	0.2001	289.8	0.1500	252.3
0.1500	190.3	0.2500	347.9	0.1998	328.6
0.2000	238.9	0.3474	434.9	0.2498	394.8
0.2501	281.2	0.4003	465.9	0.3000	446.3
0.2998	311.5	0.4506	488.5	0.3501	494.9
0.3501	337.1	0.5002	500.0	0.4009	530.8
0.3998	355.1	0.5496	501.4	0.4501	554.2
0.4499	365.3	0.5999	496.6	0.4998	569.8
0.5002	371.8	0.6501	479.5	0.5494	573.3
0.5503	366.3	0.7003	451.2	0.5999	569.6
0.6498	335.5	0.7495	411.4	0.6503	554.0
0.6997	309.1	0.7997	358.5	0.7501	481.2
0.7506	274.7	0.8500	291.8	0.7997	421.7
0.8001	232.5	0.8999	211.9	0.8502	343.3
0.8499	182.8	0.9500	111.5	0.9000	252.1
0.9001	125.9			0.9500	135.6
0.9500	59.7			0.9500	137.5

[a] $H^{\mathrm{E}}_{\mathrm{m},12}$ (J mol^{-1}) $= x_1(1 - x_1)(1477.80 + 0.68z + 64.01z^2 + 95.22z^3 - 213.66z^4)$; $s = 1.5$.
[b] $H^{\mathrm{E}}_{\mathrm{m},13}$ (J mol^{-1}) $= x_1(1 - x_1)(2000.50 - 313.32z + 110.94z^2 - 100.36z^3 - 87.36z^4)$; $s = 1.2$.
[c] $H^{\mathrm{E}}_{\mathrm{m},13}$ (J mol^{-1}) $= x_1(1 - x_1)(2279.30 - 411.74z + 255.00z^2 - 208.78z^3 - 215.53z^4)$; $s = 1.5$.

Ternary mixtures

Experimental results for $H^{\mathrm{E}}_{\mathrm{m},1+23}$ and the corresponding values of $H^{\mathrm{E}}_{\mathrm{m},123}$ are listed in Tables 2 and 3 for the two ternary systems. The values of $H^{\mathrm{E}}_{\mathrm{m},1+23}$ are plotted in Figs. 2 and 3, along with curves calculated for the constituent binaries from the representations given in the footnotes of Table 1. In all cases, the maximum values of $H^{\mathrm{E}}_{\mathrm{m},1+23}$ and $H^{\mathrm{E}}_{\mathrm{m},123}$ occur near $x_1 = 0.5$, and for comparable x_2/x_3, the maximum is larger for the mixture containing *n*-dodecane. For both systems at constant x_1, the enthalpies increase as x_2/x_3 decreases.

As in our previous work [1, 2], the values of $H^{\mathrm{E}}_{\mathrm{m},1+23}$ were represented as a sum of binary terms [8] with an added ternary contribution

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

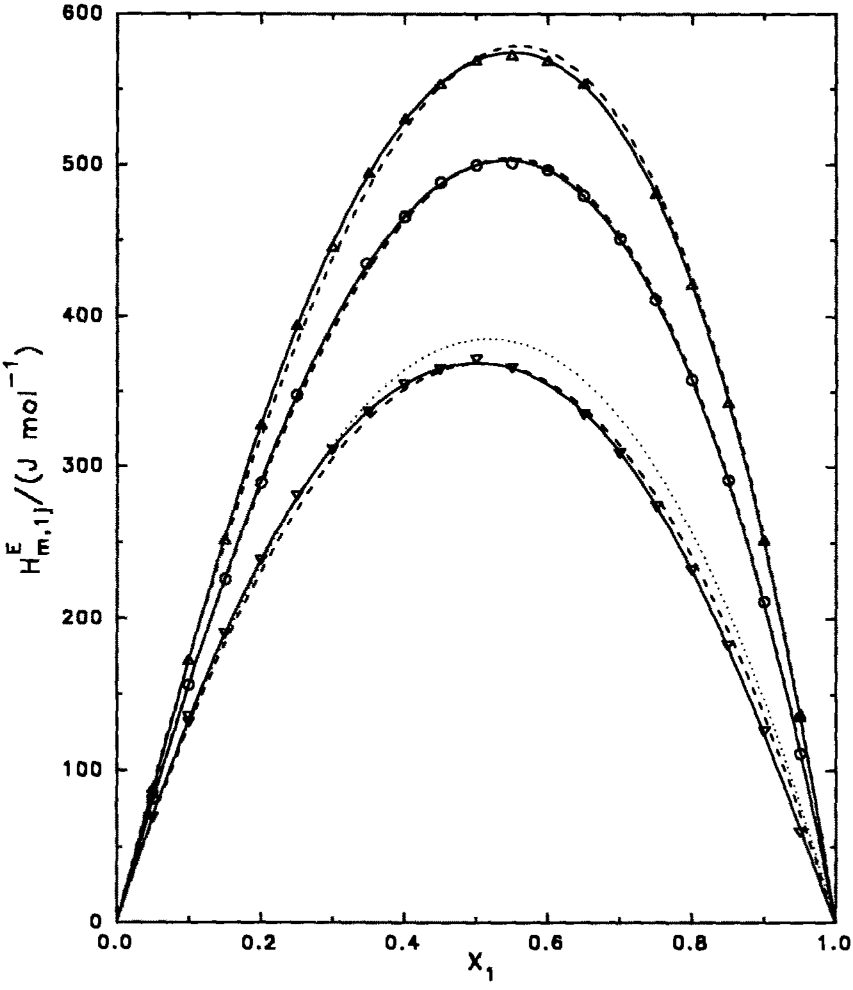

Fig. 1. Excess molar enthalpies $H^E_{m,1j}$ for $x_1C_5H_{12}O + x_jC_vH_{2v+2}$ mixtures at 298.15 K. Experimental results: ∇, $v = 6$; ○, $v = 10$; △, $v = 12$. Curves: · · · ·, $v = 7$, Tusel-Langer et al. [7]; ——, calculated from eqn. (2) using the representations in the footnotes of Table 1; – – – , calculated from the Flory theory.

and following Morris et al. [9], the form

$$H^E_{m,T} = x_1x_2x_3(c_0 + c_1x_1 + c_2x_2 + c_3x_1^2 + c_4x_1x_2 + c_5x_2^2 + \ldots) \tag{4}$$

was adopted for the ternary term. Least-squares analyses in which eqns. (3) and (4) were fitted to the values of $H^E_{m,1+23}$ indicated that adequate representations of the present results were obtained with 4 or 5 adjustable coefficients, c_i. These representations are given in the footnotes of Tables 2 and 3, along with their standard deviations s. Curves calculated from these representations are shown in Figs. 2 and 3.

TABLE 2

Experimental excess molar enthalpies $H^{E}_{m,1+23}$ at 298.15 K for the addition of methyl *tert*-butyl ether to a binary mixture of *n*-hexane and *n*-decane to form $x_1C_5H_{12}O + x_2C_6H_{14} + x_3C_{10}H_{22}$, and values of $H^{E}_{m,123}$ calculated from eqn. (1) using $H^{E}_{m,23}$ from ref. 5

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})
$x_2/x_3 = 0.3477$, $H^{E}_{m,23}$ (J mol^{-1}) = 11.8								
0.0500	83.4	94.6	0.4004	441.3	448.3	0.6997	417.7	421.2
0.0500	82.3	93.5	0.4500	459.2	465.6	0.7500	377.9	380.8
0.1000	155.2	165.8	0.5000	469.6	475.4	0.8002	328.4	330.7
0.1499	223.6	233.6	0.5502	471.0	476.3	0.8501	266.5	268.3
0.1999	284.6	294.0	0.6007	463.0	467.7	0.9007	191.6	192.8
0.2999	380.6	388.9	0.6502	446.7	450.8	0.9500	102.2	102.8
0.3515	415.1	422.7						
$x_2/x_3 = 0.9984$, $H^{E}_{m,23}$ (J mol^{-1}) = 14.4								
0.0397	61.6	75.3	0.3438	388.1	397.5	0.6467	417.3	422.4
0.0398	64.8	78.6	0.3933	413.1	421.8	0.7029	385.2	389.5
0.1218	178.7	191.3	0.4402	427.7	435.7	0.7587	342.8	346.3
0.1642	231.7	243.7	0.4901	439.3	446.7	0.8167	284.2	286.8
0.2073	278.4	289.8	0.5412	442.2	448.8	0.8761	209.4	211.2
0.2519	322.5	333.2	0.5932	433.8	439.6	0.9372	114.9	115.8
0.2975	357.2	367.3	0.6465	416.1	421.1			
$x_2/x_3 = 3.1408$, $H^{E}_{m,23}$ (J mol^{-1}) = 9.3								
0.0397	59.9	68.8	0.3915	385.6	391.2	0.7027	349.8	352.6
0.0804	119.0	127.5	0.4402	393.7	398.9	0.7588	305.4	307.6
0.1217	173.8	181.9	0.4901	407.1	411.8	0.8168	252.8	254.5
0.1644	221.9	229.6	0.5408	400.7	405.0	0.8762	184.4	185.5
0.2520	306.8	313.7	0.5934	396.7	400.4	0.9373	100.6	101.2
0.2987	332.5	339.0	0.6472	374.5	377.8	0.9374	100.1	100.7
0.3440	356.6	362.7						

[a] Ternary term for representation of $H^{E}_{m,1+23}$ by eqns. (3) and (4): $H^{E}_{m,T}$ (J mol^{-1}) $= x_1x_2x_3(651.9 - 2783.5x_1 - 24.9x_2 + 3607.8x_1^2)$; $s = 2.6$.

Equations (2)–(4) were used to calculate the constant $H^{E}_{m,123}$ contours plotted in Figs. 4 and 5. The general characteristics of these are very similar, and neither shows any indication of a maximum lying within the triangle, as found for the *n*-propanol + *n*-hexane + *n*-dodecane system [2].

Estimation from the Flory theory

In the past, we have found the Flory theory [10, 11] to be useful in correlating the excess molar enthalpies of a number of ether + *n*-alkane binary mixtures (see Wang et al. [12], where references to earlier work may be found). Accordingly, a similar treatment of the present systems seemed worthwhile. In this regard, the possibility of predicting the

TABLE 3

Experimental excess molar enthalpies $H^E_{m,1+23}$ at 298.15 K for the addition of methyl *tert*-butyl ether to a binary mixture of *n*-hexane and *n*-dodecane to form $x_1C_5H_{12}O + x_2C_6H_{14} + x_3C_{12}H_{26}$, and values of $H^E_{m,123}$ calculated from eqn. (1) using $H^E_{m,23}$ from ref. 6

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})
$x_2/x_3 = 0.3738$, $H^E_{m,23}$ (J mol^{-1}) = 32.3								
0.0500	89.1	119.8	0.3502	457.1	478.1	0.7003	472.5	482.2
0.0500	88.2	118.9	0.4015	485.2	504.5	0.7499	432.0	440.1
0.1002	177.0	206.0	0.4503	501.5	519.3	0.8002	378.2	384.7
0.1501	243.7	271.1	0.4999	515.8	531.9	0.8500	306.6	311.4
0.2000	308.9	334.8	0.5501	520.8	535.4	0.9000	224.0	227.3
0.2500	368.6	392.9	0.5999	515.5	528.4	0.9499	118.3	120.0
0.2998	418.8	441.4	0.6505	499.3	510.6			
$x_2/x_3 = 0.9845$, $H^E_{m,23}$ (J mol^{-1}) = 39.5								
0.0397	69.6	107.5	0.2982	381.0	408.7	0.6483	452.7	466.6
0.0805	131.7	168.0	0.3460	412.6	438.4	0.7028	421.0	432.7
0.1221	188.6	223.3	0.3923	439.5	463.5	0.7597	375.3	384.8
0.1646	242.8	275.8	0.4405	463.4	485.5	0.8173	314.4	321.6
0.2079	293.5	324.8	0.5414	475.2	493.3	0.8765	231.2	236.1
0.2524	343.2	372.7	0.5680	474.1	491.2	0.9374	128.8	131.2
$x_2/x_3 = 2.8956$, $H^E_{m,23}$ (J mol^{-1}) = 28.1								
0.0500	74.3	101.0	0.3505	386.3	404.6	0.6999	373.6	382.1
0.0500	76.5	103.1	0.3999	409.7	426.5	0.7505	335.1	342.1
0.1000	146.2	171.5	0.4501	419.9	435.3	0.8000	290.1	295.7
0.1501	206.2	230.1	0.4995	427.3	441.4	0.8500	233.7	238.0
0.2002	256.6	279.1	0.5499	423.4	436.1	0.9007	167.8	170.5
0.2499	307.0	328.1	0.6001	414.5	425.7	0.9502	87.7	89.0
0.2972	349.7	369.5	0.6497	398.8	408.6	0.9502	88.0	89.4

[a] Ternary term for representation of $H^E_{m,1+23}$ by eqns. (3) and (4): $H^E_{m,T}$ (J mol^{-1}) $= x_1x_2x_3(1522.2 - 6710.2x_1 - 1893.3x_2 + 7079.7x_1^2 + 5467.8x_1x_2)$; $s = 3.4$.

behavior of the ternary system from analyses of its constituent binaries was particularly attractive.

The extension of Flory's theory to multicomponent systems has been outlined by Brostow and Sochanski [13]. Here, only the equations needed for application to the present ternary systems are summarized; the notation is essentially the same as in ref. 13.

$$H^E_{m,123} = \left(\sum x_i V_i^*\right)\left[\sum \phi_i P_i^*(\tilde{v}_i^{-1} - \tilde{v}^{-1}) + \tilde{v}^{-1}\psi\right] \tag{5}$$

$$T^* = \left(\sum \phi_i P_i^* - \psi\right) \Big/ \left(\sum \phi_i P_i^*/T_i^*\right) \tag{6}$$

where

$$\psi = \phi_1\theta_2X_{12} + \phi_2\theta_3X_{23} + \phi_3\theta_1X_{13}(s_3/s_1) \tag{7}$$

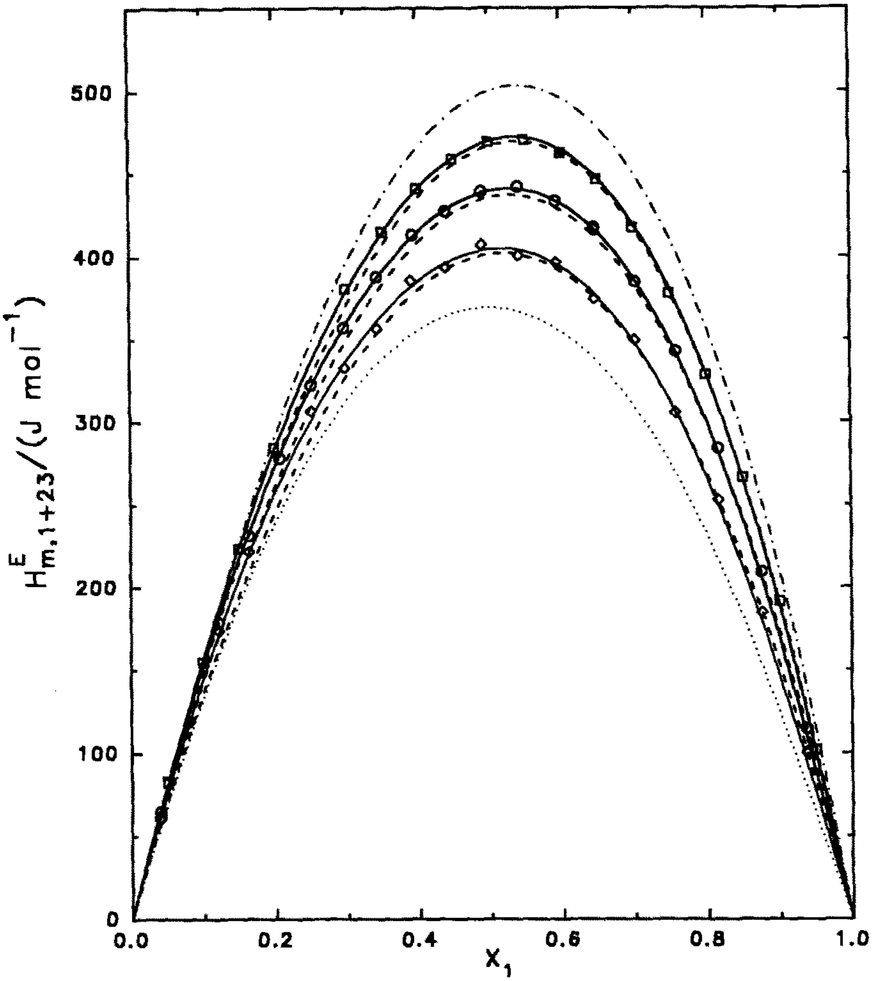

Fig. 2. Excess molar enthalpies $H^E_{m,1+23}$ for $x_1C_5H_{12}O + x_2C_6H_{14} + x_3C_{10}H_{22}$ mixtures at 298.15 K. Experimental results: □, $x_2/x_3 = 0.3477$; ○, $x_2/x_3 = 0.9984$; ◇, $x_2/x_3 = 3.1408$. Curves: – · –, $x_2 = 0$, calculated from eqn. (2) using the representation in the footnotes of Table 1; · · ·, $x_1 + x_2 = 1$, calculated from eqn. (2) using the representation in the footnotes of Table 1; ——, calculated from eqns. (2)–(4) using the representations in the footnotes of Tables 1 and 2; – – –, calculated from the Flory theory.

and the sums are taken over the three components i. The asterisk is used to indicate characteristic values of the pressure P, volume V, and temperature T; the tilde indicates their reduced values obtained by dividing by the corresponding characteristic values. The segment fractions ϕ_i, and site fractions θ_j are defined by the relations

$$\phi_j = x_j V_j^* / \sum x_i V_i^* \tag{8}$$

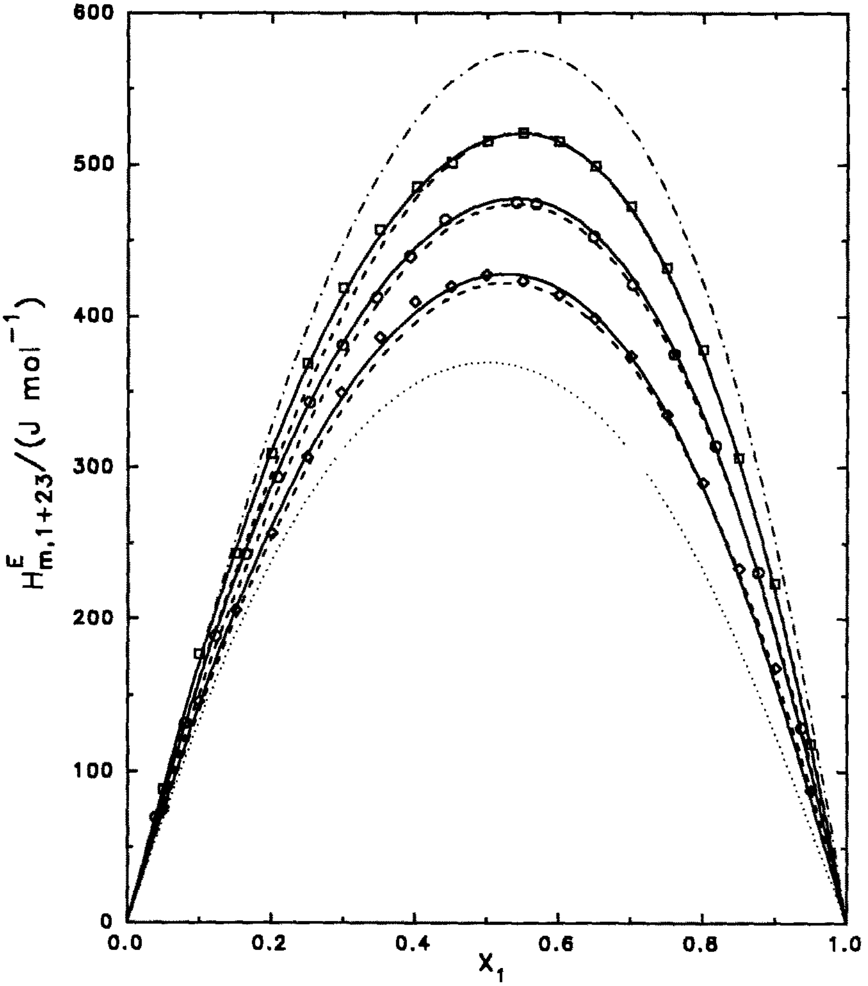

Fig. 3. Excess molar enthalpies $H^E_{m,1+23}$ for $x_1C_5H_{12}O + x_2C_6H_{14} + x_3C_{12}H_{26}$ mixtures at 298.15 K. Experimental results: □, $x_2/x_3 = 0.3738$; ○, $x_2/x_3 = 0.9845$; ◇, $x_2/x_3 = 2.8956$. Curves: – · –, $x_2 = 0$, calculated from eqn. (2) using the representation in the footnotes of Table 1; · · ·, $x_1 + x_2 = 1$, calculated from eqn. (2) using the representation in the footnotes of Table 1; ——, calculated from eqns. (2)–(4) using the representations in the footnotes of Tables 1 and 3; – – –, calculated from the Flory theory.

and

$$\theta_j = \phi_j / \sum \phi_i (s_i/s_j) \tag{9}$$

where s_i is the number of contact sites per segment of a molecule of species i. The quantities X_{ij} in eqn. (7) denote the mixed-pair interchange interaction energies between sites on species i and j. The factor s_3/s_1 in the last term of eqn. (7) has been introduced to allow for the asymmetry in Flory's definition of X_{ij} because, in the present usage, the convention $i < j$

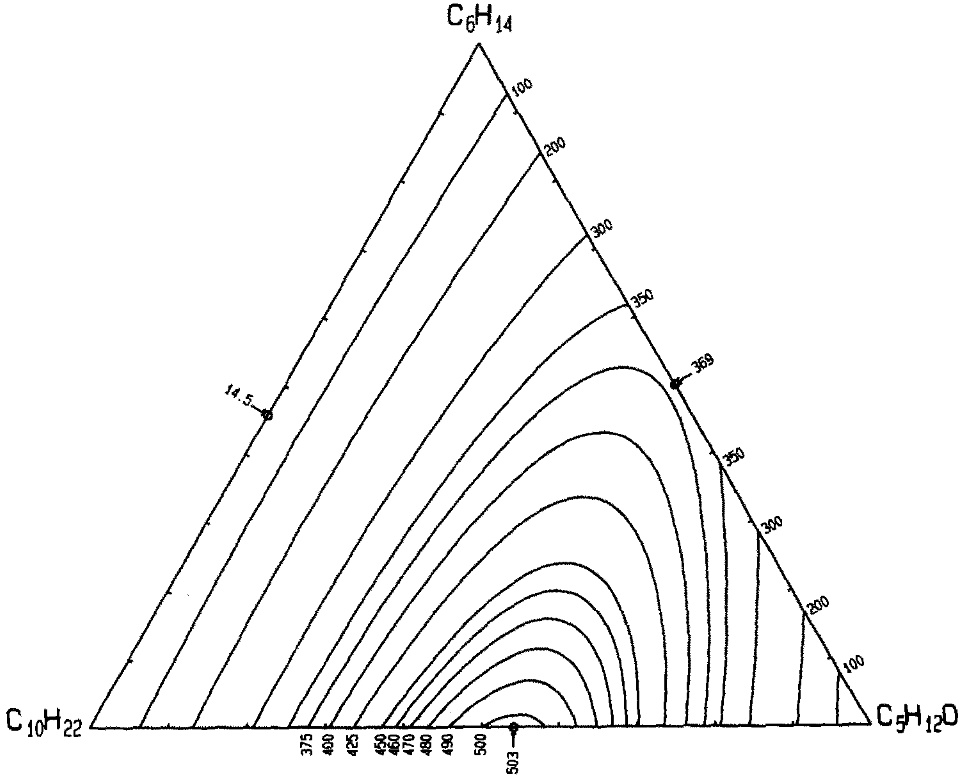

Fig. 4. Contours for constant values of the excess molar enthalpy $H^E_{m,123}$ (J mol^{-1}) of $x_1C_5H_{12}O + x_2C_6H_{14} + x_3C_{10}H_{22}$ at 298.15 K, calculated from eqns. (1)–(4) using the representations given in the footnotes of Tables 1 and 2.

has been adopted. The reduced form of the equation of state is

$$\tilde{p}\tilde{v}/\tilde{T} = \tilde{v}^{1/3}/(\tilde{v}^{1/3} - 1) - 1/\tilde{v}\tilde{T} \qquad (10)$$

which for zero pressure becomes

$$\tilde{v}^{4/3}\tilde{T} - \tilde{v}^{1/3} + 1 = 0 \qquad (11)$$

Values of V_i^* and P_i^* for the pure components are obtained from

$$\tilde{v}^{1/3} = (V_{m,i}/V_i^*)^{1/3} = 1 + \alpha_{p,i}T/3(1 + \alpha_{p,i}T) \qquad (12)$$

and

$$P_i^* = (\alpha_{p,i}/\kappa_{T,i})T\tilde{v}_i^2 \qquad (13)$$

where $V_{m,i}$, $\alpha_{p,i}$, and $\kappa_{T,i}$ are the molar volume, isobaric expansivity, and isothermal compressibility, respectively. The value of T_i^* can then be calculated from eqn. (6).

The component properties and parameters used in the present calculations are given in Table 4. In the case of MTBE, α_p at 298.15 K has been reported by Obama et al. [14], but it appears that a value of κ_T is not

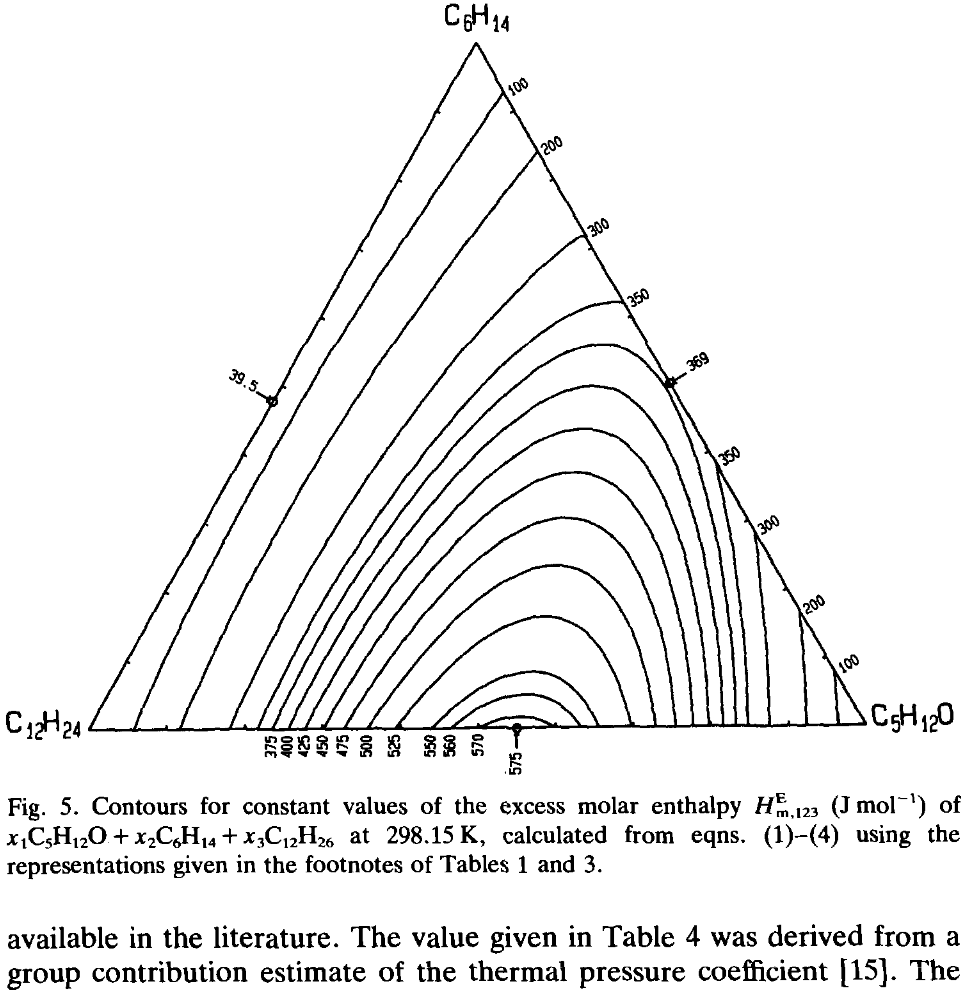

Fig. 5. Contours for constant values of the excess molar enthalpy $H^{E}_{m,123}$ (J mol^{-1}) of $x_1C_5H_{12}O + x_2C_6H_{14} + x_3C_{12}H_{26}$ at 298.15 K, calculated from eqns. (1)–(4) using the representations given in the footnotes of Tables 1 and 3.

available in the literature. The value given in Table 4 was derived from a group contribution estimate of the thermal pressure coefficient [15]. The data for the *n*-alkanes are the same as in our previous work [12].

Values of X_{12} and X_{13} were obtained from least-squares calculations in which eqn. (5) was fitted to the representations of $H^{E}_{m,1j}$ given in the footnotes of Table 1. Values of X_{23} were obtained from similar treatments of $H^{E}_{m,23}$ for *n*-hexane + *n*-decane mixtures [5], and *n*-hexane + *n*-dodecane mixtures [6]. In all of these calculations, it was assumed that the molecules were approximately spherical, and the relation

$$s_i/s_j = (V_j^*/V_i^*)^{1/3} \tag{14}$$

was used to estimate the site ratios.

The dashed curves in Figs. 1–3 were calculated from eqn. (5), using the parameters from Table 4. The agreement between these estimates and the experimental results is surprisingly good, in view of the fact that only data for the pure components and their binary mixtures were used. For the 96 points of the *n*-decane systems, the standard deviation between the

TABLE 4

Component properties and parameters used in Flory calculations for the systems $x_1C_5H_{12}O + x_2C_6H_{14} + x_3C_{10}H_{22}$ [a] and $x_1C_5H_{12}O + x_2C_6H_{14} + x_3C_{12}H_{26}$ [b] at 298.15 K

Component	V_m ($cm^3\ mol^{-1}$)	α_p (kK^{-1})	κ_T (TPa^{-1})	P^* ($J\ cm^{-3}$)	V^* ($cm^3\ mol^{-1}$)	T^* (K)
$C_5H_{12}O$	119.82	1.423	1690.6	442.9	90.20	4385.0
C_6H_{14}	131.57	1.387	1703.9	424.2	99.52	4436.1
$C_{10}H_{22}$	195.94	1.051	1109.6	447.0	155.75	5091.4
$C_{12}H_{26}$	228.55	0.960	987.6	445.2	184.40	5351.4

Interchange interaction energy parameters X_{ij} ($J\ cm^{-3}$):
[a] $X_{12} = 14.7950$; $X_{13} = 18.7764$; $X_{23} = 1.2208$.
[b] $X_{12} = 14.7950$; $X_{13} = 21.1118$; $X_{23} = 2.5204$.

estimated and experimental values of $H^E_{m,123}$ is 5.7 J mol^{-1}, and the mean absolute relative deviation is only 2.0%. For the 99 points of the *n*-dodecane systems, the corresponding deviations are 6.7 J mol^{-1} and 2.1%. Thus the Flory theory provides useful estimates of $H^E_{m,123}$ for systems of the present type. Finally, it should also be noted that X_{1j} is nearly linear in the carbon chain length of the second component, which could be used to provide estimates of $H^E_{m,1j}$ for mixtures of MTBE with other *n*-alkanes.

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