

# Excess Enthalpies of the Ternary Mixtures: Tetrahydrofuran + (Hexane or Cyclohexane) + Decane at 298.15 K

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Excess molar enthalpies, measured at 298.15 K in a flow microcalorimeter, are reported for tetrahydrofuran + hexane + decane and for tetrahydrofuran + cyclohexane + decane. Smooth representations of the results are described and used to construct constant excess molar enthalpy contours on Roozeboom diagrams. The latter are compared with diagrams obtained when the model of Liebermann and Fried is used to estimate the excess molar enthalpies of the ternary mixtures from the physical properties of the components and the excess molar enthalpies of their binary mixtures.

## Introduction

Recent papers from our laboratory have reported excess molar enthalpies of some binary and ternary mixtures formed by mixing an oxygenate with various hydrocarbons. Most recently, excess molar enthalpies of the systems tetrahydrofuran + (2,2,4-trimethylpentane or heptane) + methylcyclohexane were reported.<sup>1</sup> As a continuation of that investigation, similar calorimetric measurements at 298.15 K have been made for the two ternary systems formed by mixing tetrahydrofuran (THF) with binary mixtures of either hexane (nC6) or cyclohexane (cC6) with decane (nC10).

## Experimental Section

All of the components were obtained from Aldrich, and in all cases, the purities stated by the manufacturer were at least 99 mol %. Apart from partial degassing, the components were used without further purification. Densities,  $\rho/\text{kg m}^{-3}$ , measured at 298.15 K in an Anton-Paar digital densimeter, were 882.09, 655.42, 773.92, and 726.37 for THF, nC6, cC6, and nC10, respectively. These results are in reasonable agreement with values in the TRC Tables.<sup>2,3</sup>

An LKB flow microcalorimeter (model 10700-1), thermostated at  $(298.150 \pm 0.003)$  K, was used to measure the excess molar enthalpies  $H_m^E$ . Details of the equipment and its operation have been described previously (Tanaka et al.<sup>4</sup> and Kimura et al.<sup>5</sup>).

In studying the ternary systems, the excess molar enthalpy  $H_{m,1+23}^E$  was determined for several pseudobinary mixtures in which component 1, (THF) was added to binary mixtures of components 2 (either nC6 or cC6) and 3 (nC10). For this purpose, binaries with fixed mole ratios,  $x_2/x_3$ , were prepared by mass. The excess molar enthalpy  $H_{m,123}^E$  of the ternary mixture was then obtained from the relation

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

where  $x_1$  is the mole fraction of THF and  $H_{m,23}^E$  is the excess molar enthalpy of the particular binary mixture. Over most of the mole fraction range, the errors of the

excess molar enthalpies and the mole fractions of the final mixtures are estimated to be less than 0.5% and  $5 \times 10^{-4}$ , respectively.

## Results and Discussion

Excess molar enthalpies  $H_{m,ij}^E$  ( $i < j$ ), measured at 298.15 K, for four of the five constituent-binaries of present interest have been reported previously: THF(1) + cC6(2),<sup>6</sup> THF(1) + nC10(3),<sup>7</sup> nC6(2) + nC10(3),<sup>8</sup> and cC6(2) + nC10(3).<sup>9</sup> In the course of the present investigation, excess molar enthalpies at 298.15 K were determined for the three-constituent binaries involving THF. These results are listed in Table 1. Our new results for THF(1) + cC6(2) and THF(1) + nC10(3) tend to be higher than those reported previously. For equimolar mixtures, the differences are approximately 12 and 15 J mol<sup>-1</sup>, respectively.

The smoothing function

$$H_{m,ij}^E/\text{J mol}^{-1} = x_i(1 - x_j) \sum_{k=1}^n h_k(1 - 2x_j)^{k-1} \quad (i < j) \quad (2)$$

was fitted to the results in Table 1 by a least-squares method, with all points weighted equally. Values of the coefficients  $h_k$  are listed in Table 2, along with the standard deviation  $s$  for the representation. For convenience, the coefficients and standard deviations for the representations of  $H_{m,ij}^E$  for the two other binary mixtures are also included in Table 2.

The experimental results for  $H_{m,1+23}^E$  and the corresponding values of  $H_{m,123}^E$  calculated from eq 1 are listed in Tables 3 and 4 against the mole fraction  $x_1$  for the ternary systems containing nC6 and cC6, respectively. For a selected value of  $x_2/x_3$  in these tables, the maximum values of  $H_{m,1+23}^E$  and  $H_{m,123}^E$  occur near  $x_1 = 0.5$ . For both systems at constant  $x_1$ , the enthalpies decrease as  $x_2/x_3$  increases.

The experimental values of  $H_{m,1+23}^E$  from Tables 3 and 4 are plotted in Figures 1 and 2, respectively. Also shown in these figures are the results for THF(1) + nC6(2), THF(1) + nC10(3), and THF(1) + cC6(2), reported in Table 1. In Figure 1, it can be seen that the results for the three ternary mixtures containing nC6 fall between those for the

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**Table 1. Experimental Mole Fractions,  $x_1$ , and Excess Molar Enthalpies,  $H_{m,ij}^E$  for the Mixtures THF(1) + nC6(2), THF(1) + cC6(2), and THF(1) + nC10(3) at 298.15 K**

$x_1$	$\frac{H_{m,ij}^E}{\text{J mol}^{-1}}$	$x_1$	$\frac{H_{m,ij}^E}{\text{J mol}^{-1}}$	$x_1$	$\frac{H_{m,ij}^E}{\text{J mol}^{-1}}$	$x_1$	$\frac{H_{m,ij}^E}{\text{J mol}^{-1}}$	$x_1$	$\frac{H_{m,ij}^E}{\text{J mol}^{-1}}$
THF(1) + nC6(2)									
0.0500	151.57	0.2500	573.75	0.4504	748.03	0.6503	695.86	0.8501	396.08
0.1000	283.77	0.2998	643.55	0.5005	750.72	0.7002	646.10	0.8998	289.83
0.1499	394.02	0.3503	688.14	0.5508	750.14	0.7500	581.02	0.9499	152.77
0.2001	492.96	0.4003	724.95	0.6003	729.39	0.8015	498.97		
THF(1) + cC6(2)									
0.0500	161.26	0.2499	585.32	0.4504	728.05	0.6500	656.38	0.8505	358.88
0.1000	296.70	0.2997	639.68	0.4999	729.98	0.7003	603.99	0.9003	250.46
0.1500	413.31	0.3500	684.33	0.5498	720.56	0.7498	528.92	0.9500	129.36
0.2001	506.35	0.4003	714.05	0.6000	695.74	0.8000	451.11		
THF(1) + nC10(3)									
0.0500	155.22	0.2503	645.62	0.4509	895.78	0.6501	892.50	0.8502	560.57
0.0999	303.09	0.3000	731.21	0.4999	920.84	0.7003	843.66	0.9011	410.24
0.1500	434.95	0.3498	804.37	0.5499	929.53	0.7502	773.95	0.9500	228.08
0.2000	547.09	0.4002	864.30	0.6002	921.92	0.8003	678.28		

**Table 2. Coefficients,  $h_k$ , and Standard Deviations,  $s$ , for the Representation of  $H_{m,ij}^E$  ( $i < j$ ) by Eq 2 for the Constituent–Binary Mixtures at 298.15 K**

component		$s$				
$i$	$j$	$h_1$	$h_2$	$h_3$	$h_4$	$\text{J mol}^{-1}$
THF	nC6 <sup>a</sup>	3021.50	-33.63	224.97		2.57
THF	cC6 <sup>a</sup>	2926.64	202.83	191.77	209.78	2.30
THF	nC10 <sup>a</sup>	3688.15	-622.13	427.41	-219.91	2.58
nC6	nC10 <sup>b</sup>	58.61	9.87	-4.77		0.04
cC6	nC10 <sup>c</sup>	1204.96	-439.85	212.59	-14.82	1.2

<sup>a</sup> Present work. <sup>b</sup> Hamam and Benson.<sup>8</sup> <sup>c</sup> Arenosa et al.<sup>9</sup>**Table 3. Experimental Excess Molar Enthalpies,  $H_{m,1+23}^E$ , Measured at 298.15 K, for the Addition of THF to nC6 + nC10 Mixtures to Form THF(1) + nC6(2) + nC10(3), and Values of  $H_{m,123}^E$  Calculated from Eq 1**

$x_1$	$\frac{H_{m,1+23}^E}{\text{J mol}^{-1}}$	$\frac{H_{m,123}^E}{\text{J mol}^{-1}}$ <sup>a</sup>	$x_1$	$\frac{H_{m,1+23}^E}{\text{J mol}^{-1}}$	$\frac{H_{m,123}^E}{\text{J mol}^{-1}}$ <sup>a</sup>	$x_1$	$\frac{H_{m,1+23}^E}{\text{J mol}^{-1}}$	$\frac{H_{m,123}^E}{\text{J mol}^{-1}}$ <sup>a</sup>
$x_2/x_3 = 0.3334$ , $H_{m,23}^E$ (J mol <sup>-1</sup> ) = 11.69								
0.0501	169.23	180.34	0.3999	831.33	838.35	0.7001	792.72	796.23
0.1000	295.50	306.02	0.4503	857.27	863.70	0.7502	723.48	726.40
0.1500	425.54	435.48	0.4995	875.51	881.36	0.8000	622.77	625.11
0.1999	534.45	543.80	0.5501	886.74	892.00	0.8501	509.76	511.51
0.2499	629.00	637.77	0.5999	875.01	879.69	0.8997	370.20	371.37
0.3000	710.13	718.31	0.6499	841.65	845.74	0.9499	194.41	195.00
0.3502	780.37	787.97						
$x_2/x_3 = 0.9999$ , $H_{m,23}^E$ (J mol <sup>-1</sup> ) = 14.65								
0.0501	158.95	172.87	0.4002	791.79	800.58	0.7000	746.66	751.06
0.0999	288.13	301.32	0.4500	823.99	832.05	0.7499	677.78	681.44
0.1500	414.61	427.06	0.5001	834.60	841.92	0.8002	587.83	590.76
0.2002	518.23	529.95	0.5500	843.49	850.08	0.8498	479.57	481.77
0.2502	610.04	621.03	0.5999	829.64	835.50	0.8999	347.45	348.92
0.3002	689.57	699.82	0.6515	794.77	799.88	0.9500	185.46	186.19
0.3500	751.43	760.95						
$x_2/x_3 = 3.0014$ , $H_{m,23}^E$ (J mol <sup>-1</sup> ) = 9.84								
0.0499	150.16	159.51	0.4005	756.57	762.47	0.7000	696.95	699.90
0.1001	287.25	296.10	0.4502	783.94	789.35	0.7494	628.69	631.16
0.1500	399.16	407.52	0.5002	794.46	799.38	0.8007	545.01	546.97
0.1999	507.92	515.79	0.5503	795.14	799.56	0.8504	442.59	444.06
0.2501	590.15	597.53	0.6001	780.37	784.30	0.8995	322.44	323.43
0.2998	664.93	671.82	0.6502	745.90	749.34	0.9500	169.79	170.28
0.3502	722.01	728.40						

<sup>a</sup> Ternary term for representation of  $H_{m,123}^E$  by eqs 3 and 4:  $H_{m,T}^E/\text{J mol}^{-1} = [x_1x_2x_3/(1-x_1+x_2)](-76.08-778.61x_1+4327.53x_2-146.25x_1^2-8173.97x_1x_2-5429.03x_2^2)$ ,  $s = 4.25 \text{ J mol}^{-1}$ .

two constituent-binaries; this is not the case for the cC6 system in Figure 2.

Representation of  $H_{m,123}^E$  was based on the relation

$$H_{m,123}^E = \sum_{i=1}^2 \sum_{j=i+1}^3 (x_i + x_j)^2 H_{m,ij}^E \left( \frac{x_i}{x_i + x_j}, \frac{x_j}{x_i + x_j} \right) + H_{m,T}^E \quad (3)$$

**Table 4. Experimental Excess Molar Enthalpies,  $H_{m,1+23}^E$ , Measured at 298.15 K, for the Addition of THF to cC6 + nC10 Mixtures to Form THF(1) + cC6(2) + nC10(3), and Values of  $H_{m,123}^E$  Calculated from Eq 1**

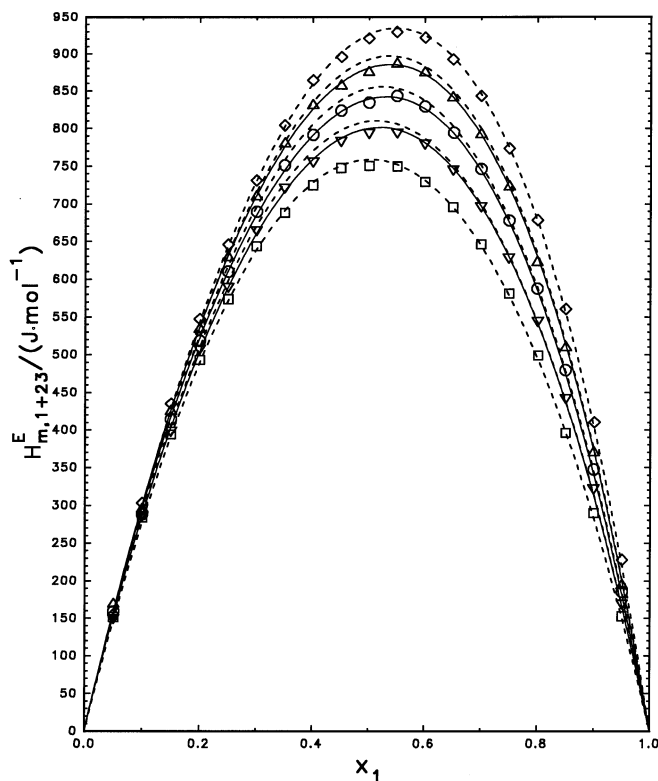
$x_1$	$\frac{H_{m,1+23}^E}{\text{J mol}^{-1}}$	$\frac{H_{m,123}^E}{\text{J mol}^{-1}}$ <sup>a</sup>	$x_1$	$\frac{H_{m,1+23}^E}{\text{J mol}^{-1}}$	$\frac{H_{m,123}^E}{\text{J mol}^{-1}}$ <sup>a</sup>	$x_1$	$\frac{H_{m,1+23}^E}{\text{J mol}^{-1}}$	$\frac{H_{m,123}^E}{\text{J mol}^{-1}}$ <sup>a</sup>
$x_2/x_3 = 0.3413$ , $H_{m,23}^E$ (J mol <sup>-1</sup> ) = 197.00								
0.0501	160.48	347.61	0.4000	788.90	907.09	0.7001	749.43	808.52
0.1000	284.67	461.98	0.4511	814.57	922.71	0.7498	683.82	733.11
0.1500	396.24	563.69	0.5006	832.55	930.92	0.7996	596.95	636.43
0.2000	506.53	664.13	0.5501	838.73	927.37	0.8503	488.82	518.30
0.2500	594.33	742.08	0.5996	826.16	905.05	0.8999	357.10	376.82
0.3001	674.35	812.23	0.6498	797.26	866.26	0.9499	193.88	203.74
0.3502	740.68	868.69						
$x_2/x_3 = 0.9997$ , $H_{m,23}^E$ (J mol <sup>-1</sup> ) = 301.22								
0.0500	146.12	432.28	0.3000	632.63	843.48	0.7003	668.21	758.48
0.0514	146.47	432.21	0.3502	686.16	881.91	0.7497	603.83	679.23
0.1000	270.32	541.43	0.4001	719.91	900.61	0.7999	526.24	586.52
0.1501	375.67	631.68	0.4507	748.45	913.91	0.8498	426.60	471.85
0.1999	475.87	716.87	0.5502	760.86	896.34	0.8997	307.04	337.24
0.2001	476.62	717.58	0.5997	748.24	868.84	0.9495	166.60	181.82
0.2503	560.28	786.12	0.6504	718.40	823.70			
$x_2/x_3 = 2.9976$ , $H_{m,23}^E$ (J mol <sup>-1</sup> ) = 277.55								
0.0500	144.59	408.25	0.3999	678.46	845.01	0.7003	599.56	682.73
0.0999	262.48	512.30	0.4501	697.78	850.39	0.7501	537.28	606.63
0.1501	371.79	607.67	0.4997	701.98	840.84	0.7999	460.59	516.14
0.2000	460.71	682.76	0.5498	702.15	827.10	0.8501	366.48	408.08
0.2502	537.76	745.86	0.6001	684.14	795.12	0.9001	241.70	269.44
0.3001	603.45	797.70	0.6498	645.35	742.55	0.9500	118.26	132.12
0.3501	644.21	824.59						

<sup>a</sup> Ternary term for representation of  $H_{m,123}^E$  by eqs 3 and 4:  $H_{m,T}^E/\text{J mol}^{-1} = [x_1x_2x_3/(1-x_1+x_2)](2238.80-16713.51x_1-6660.68x_2+30596.20x_1^2+62961.82x_1x_2+3330.48x_2^2-16490.90x_1^3-72266.73x_1^2x_2-53181.13x_1x_2^2)$ ,  $s = 5.40 \text{ J mol}^{-1}$ .

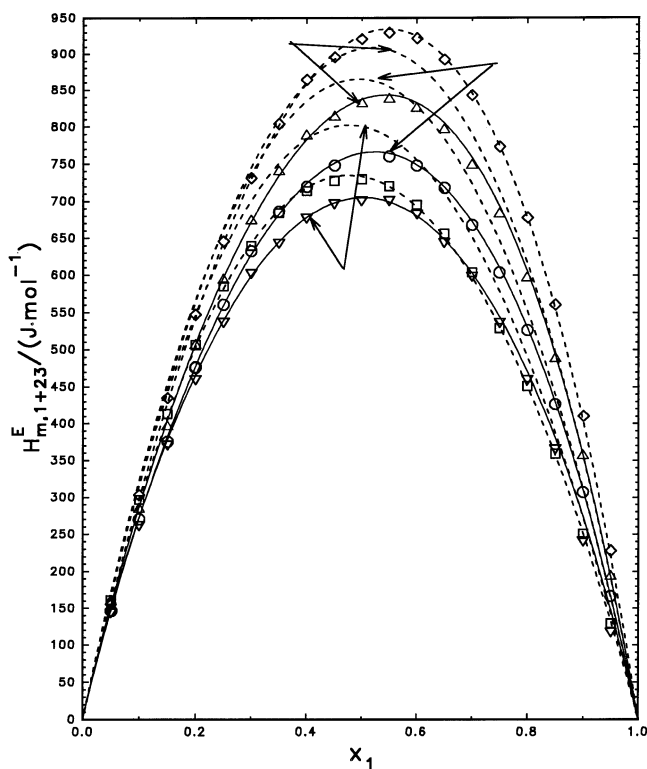
which consists of the sum of binary contributions proposed by Kohler<sup>10,11</sup> and an added ternary term. The form

$$H_{m,T}^E/\text{J mol}^{-1} = [x_1x_2x_3/(1-x_1+x_2)](c_0 + c_1x_1 + c_2x_2 + c_3x_1^2 + c_4x_1x_2 + c_5x_2^2 + c_6x_1^3 + c_7x_1^2x_2 + c_8x_1x_2^2 + c_9x_2^3) \quad (4)$$

which was adopted for the latter contribution is similar to the form used by Morris et al.<sup>12</sup> with an extra skewing factor  $(1-x_1+x_2)^{-1}$  inserted. Values of the parameters  $c_i$  were obtained from least-squares analyses in which eqs 3 and 4 were fitted to the values of  $H_{m,123}^E$  in Tables 3 and 4. The resulting representations of  $H_{m,T}^E$  are given in the footnotes of the tables, along with the standard deviations  $s$  of the fits. The solid curves for  $H_{m,1+23}^E$  in Figures 1 and



**Figure 1.** Excess molar enthalpies  $H_{m,1+23}^E$  for THF(1) + nC6(2) + nC10(3) at 298.15 K. Experimental results:  $\triangle$ ,  $x_2/x_3 = 0.3334$ ;  $\circ$ ,  $x_2/x_3 = 0.9999$ ;  $\nabla$ ,  $x_2/x_3 = 3.0014$ ;  $\diamond$ ,  $x_2 = 0$ ;  $\square$ ,  $x_3 = 0$ . Curves: —, calculated from the representation of the results by eqs 1–4, using the ternary term  $H_{m,T}^E$  given in the footnote of Table 3; ---, estimated by means of the Liebermann–Fried model.

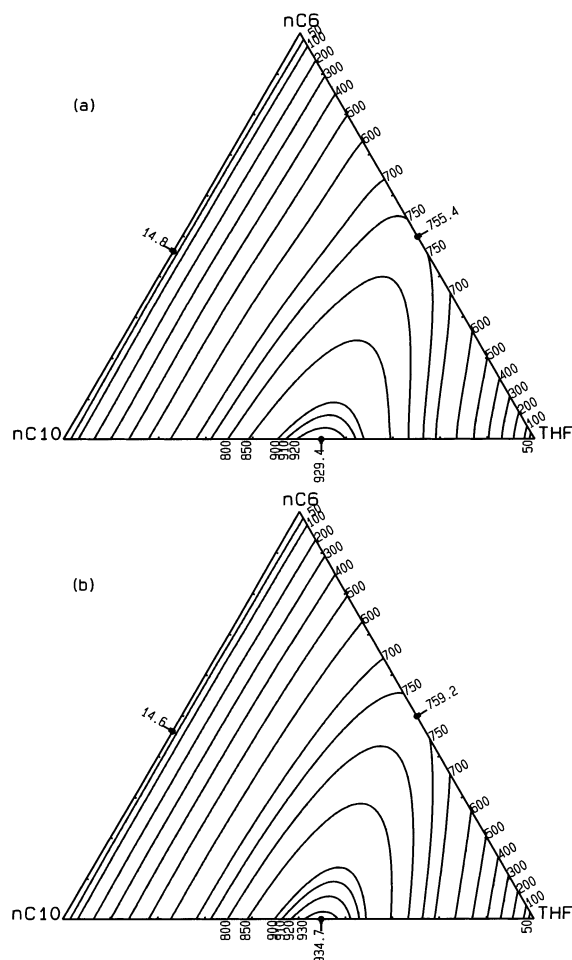


**Figure 2.** Excess molar enthalpies  $H_{m,1+23}^E$  for THF(1) + cC6(2) + nC10(3) at 298.15 K. Experimental results:  $\triangle$ ,  $x_2/x_3 = 0.3413$ ;  $\circ$ ,  $x_2/x_3 = 0.9997$ ;  $\nabla$ ,  $x_2/x_3 = 2.9976$ ;  $\diamond$ ,  $x_2 = 0$ ;  $\square$ ,  $x_3 = 0$ . Curves: —, calculated from the representation of the results by eqs 1–4, using the ternary term  $H_{m,T}^E$  given in the footnote of Table 4; ---, estimated by means of the Liebermann–Fried model.

**Table 5.** Values of the Interaction Parameters,  $A_{ij}$  and  $A_{ji}$ , Standard Deviations,  $s$ , and Isobaric Thermal Expansivities,  $\alpha_p$ , at 298.15 K for Liebermann–Fried Model Calculations

component		$A_{ij}$		$s$	$\alpha_p$ (K <sup>-1</sup> )	
$i$	$j$	$A_{ij}$	$A_{ji}$	J mol <sup>-1</sup>	$i$	$j$
THF	nC6	0.7881	0.7810	4.26	1.138 <sup>a</sup>	1.387 <sup>b</sup>
THF	nC10	0.9014	0.6087	4.87	1.138 <sup>a</sup>	1.051 <sup>b</sup>
nC6	nC10	1.0464	0.9265	0.06	1.387 <sup>b</sup>	1.051 <sup>b</sup>
THF	cC6	0.7139	0.8654	4.06	1.138 <sup>a</sup>	1.220 <sup>c</sup>
cC6	nC10	1.2904	0.6098	2.60	1.220 <sup>c</sup>	1.051 <sup>b</sup>

<sup>a</sup> Riddick et al.<sup>17</sup> see page 309. <sup>b</sup> Benson et al.<sup>18</sup> <sup>c</sup> Riddick et al.<sup>17</sup> see page 90.

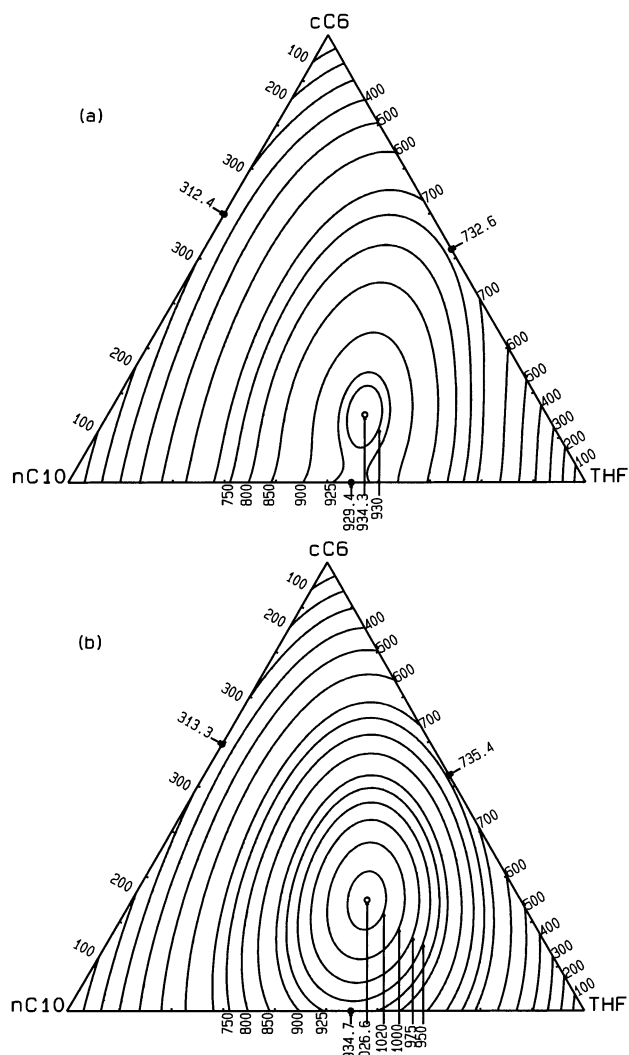


**Figure 3.** Contours for constant values of  $H_{m,123}^E$ /J mol<sup>-1</sup> for THF(1) + nC6(2) + nC10(3) at 298.15 K. Part a calculated from the representation of the experimental results by eqs 1–4 with  $H_{m,T}^E$  from the footnote of Table 3; part b estimated by means of the Liebermann–Fried model.

2 were calculated from eq 1, using these representations of  $H_{m,123}^E$ .

Equations 1–4 were also used to calculate the constant  $H_{m,123}^E$  contours plotted on the Roozeboom diagrams in Figures 3a and 4a. Although the general characteristics of these are similar, they differ through the appearance of a small internal maximum for the cC6 system and no indication of an internal extremum for the nC6 system.

Recent work<sup>13,14</sup> indicates that an extension of the model of Liebermann and Fried<sup>15,16</sup> can be useful in representing the excess enthalpies of binary mixtures, and also has the potential for estimating the enthalpies of ternary mixtures



**Figure 4.** Contours for constant values of  $H_{m,123}^E/\text{J mol}^{-1}$  for THF(1) + cC6(2) + nC10(3) at 298.15 K. Part a calculated from the representation of the experimental results by eqs 1–4 with  $H_{m,T}^E$  from the footnote of Table 4; part b estimated by means of the Liebermann–Fried model.

from data for the pure components and their binary mixtures. It is therefore of interest to examine how well the Liebermann–Fried model can represent the enthalpies of the present ternary systems.

The equations used in this application have been outlined by Wang et al.<sup>14</sup> Values of the Liebermann–Fried interaction parameters,  $A_{ij}$  and  $A_{ji}$ , for each of the binary mixtures are given in Table 5. These were obtained by fitting the Liebermann–Fried formula for  $H_{m,ij}^E$  to the primary experimental results in Table 1 and those reported for the other relevant binaries by Hamam et al.<sup>8</sup> and Arenosa et al.<sup>9</sup> Also included in Table 5 are values of the standard deviations  $s$  achieved in the fitting process and values<sup>17,18</sup> of the isobaric thermal expansivities  $\alpha_p$  of the components, used in evaluating the contributions due to different molecular sizes.

Using the values of the parameters given in Table 5,  $H_{m,1+23}^E$  and  $H_{m,123}^E$  were calculated for the present mixtures. The resulting values of  $H_{m,1+23}^E$  for the ternary mixtures and  $H_{m,ij}^E$  for the constituent-binaries were used to plot the dashed curves in Figures 1 and 2. It can be seen from these that, although the fits of the data for the constituent-binaries are relatively good, the Liebermann–

Fried model tends to slightly over-estimate the ternary enthalpies of the nC6 system, and much more significantly in the case of the cC6 mixtures. The mean absolute relative deviations between the 57 experimental and estimated values of  $H_{m,1+23}^E$  in Table 3 and the 58 points in Table 4 amount to 1.2% and 11.4%, respectively.

Constant  $H_{m,123}^E$  contours were also estimated on the basis of the Liebermann–Fried model and are shown in parts b of Figures 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. In the case of the cC6 system, the Liebermann–Fried treatment predicts the internal maximum, but it is larger than that found from the analysis of the ternary experimental data. Despite this discrepancy, it can be seen that the Liebermann–Fried model provides useful prediction of the ternary enthalpy behavior without requiring the direct investigation of the excess enthalpies of any ternary mixtures.

## Literature Cited

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