# Excess Enthalpies of the Ternary Mixtures: (Diisopropyl Ether or 2-Methyltetrahydrofuran) + Hexane + Hexadecane at 298.15 K

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Excess molar enthalpies, measured at 298.15 K in a flow microcalorimeter, are reported for diisopropyl ether + hexane + hexadecane and for 2-methyltetrahydrofuran + hexane + hexadecane. 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

Previous papers from our laboratory have reported excess molar enthalpies of some binary and ternary mixtures comprised of either diisopropyl ether (DIPE)[1−3] or 2-methyltetrahydrofuran (MTHF)[4−6] and various *n*-alkanes. As a continuation of those investigations, similar calorimetric measurements at 298.15 K have been made for the two ternary systems formed from either DIPE or MTHF with mixtures of hexane (nC6) and hexadecane (nC16).

## 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, $\rho$/kg·m$^{-3}$, measured at 298.15 K in an Anton-Paar digital densimeter, were 718.69, 848.10, 655.43, and 770.13, for DIPE, MTHF, nC6, and nC16, respectively. The result for DIPE is within 0.03% of that reported by Obama et al.;[7] that for MTHF is within 0.15% of the value reported recently by De Lorenzi et al.[8] The results for the alkanes are in reasonable agreement with values in the TRC Tables.[9]

An LKB flow microcalorimeter (Model 10700-1), thermostated at (298.150 ± 0.003) K, was used to measure the excess molar enthalpies $H^{E}_{m}$. Details of the equipment and its operation have been described previously (Tanaka et al.[10] and Kimura et al.[11]).

In studying the ternary systems, the excess molar enthalpy $H^{E}_{m,1+23}$ was determined for several pseudobinary mixtures in which component 1 (DIPE or MTHF) was added to binary mixtures of components 2 (nC6) and 3 (nC16). For this purpose, binaries with fixed mole ratios, $x_2/x_3$, were prepared by mass. The excess molar enthalpy $H^{E}_{m,123}$ of the ternary mixture was then obtained from the relation

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

where $x_1$ is the mole fraction of either DIPE or MTHF and $H^{E}_{m,23}$ 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 $H^{E}_{m,ij}$ ($i < j$), at 298.15 K, for four of the five constituent binaries of present interest have been reported previously: DIPE (1) + nC6 (2),[1] DIPE (1) + nC16 (3),[1] nC6 (2) + nC16 (3),[12] and MTHF (1) + nC6 (2).[4] The experimental results for the excess molar enthalpies $H^{E}_{m,13}$ of MTHF (1) + nC16 (3) at 298.15 K are listed in Table 1. The smoothing function

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

was fitted to the results 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 $H^{E}_{m,ij}$ for the four other binary mixtures are also included in Table 2.

The experimental results for $H^{E}_{m,1+23}$ and the corresponding values of $H^{E}_{m,123}$ calculated from eq 1 are listed in Tables 3 and 4 against the mole fraction $x_1$ for the ternary systems containing DIPE and MTHF, respectively. For a selected value of $x_2/x_3$ in these tables, the maximum values of $H^{E}_{m,1+23}$ and $H^{E}_{m,123}$ 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 $H^{E}_{m,1+23}$ from Tables 3 and 4 are plotted in Figures 1 and 2, respectively. Also shown in Figure 2 are the results for MTHF (1) + nC16 (3) given in Table 1.

Following Tsao and Smith,[13] the values of $H^{E}_{m,1+23}$ were represented as a sum of binary terms with an added ternary contribution

$$H^{E}_{m,1+23} = [x_2/(1 - x_1)]H^{E}_{m,12} + [x_3/(1 - x_1)]H^{E}_{m,13} + H^{E}_{m,T} \qquad (3)$$

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**Table 1. Experimental Mole Fractions, $x_1$, and Excess Molar Enthalpies, $H^E_{m,13}$, for MTHF (1) + nC16 (3) Mixtures at 298.15 K**

| $x_1$ | $H^E_{m,13}$/J·mol$^{-1}$ | $x_1$ | $H^E_{m,13}$/J·mol$^{-1}$ | $x_1$ | $H^E_{m,13}$/J·mol$^{-1}$ | $x_1$ | $H^E_{m,13}$/J·mol$^{-1}$ | $x_1$ | $H^E_{m,13}$/J·mol$^{-1}$ |
|---|---|---|---|---|---|---|---|---|---|
| 0.0500 | 138.89 | 0.2499 | 617.51 | 0.4503 | 901.14 | 0.6501 | 922.04 | 0.8500 | 605.77 |
| 0.1000 | 277.38 | 0.3002 | 711.99 | 0.5003 | 921.69 | 0.7000 | 879.85 | 0.9002 | 450.36 |
| 0.1499 | 413.89 | 0.3496 | 787.03 | 0.5502 | 938.68 | 0.7499 | 816.48 | 0.9500 | 252.68 |
| 0.2002 | 514.55 | 0.4000 | 853.69 | 0.5998 | 941.29 | 0.7998 | 726.74 | | |

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

| component | | | | | | | | $s$ |
|---|---|---|---|---|---|---|---|---|
| $i$ | $j$ | $h_1$ | $h_2$ | $h_3$ | $h_4$ | $h_5$ | $h_6$ | J·mol$^{-1}$ |
| DIPE | nC6[a] | 901.54 | 48.97 | 44.94 | 70.11 | −136.66 | | 1.0 |
| DIPE | nC16[a] | 2241.24 | −417.28 | 351.93 | 219.29 | −491.54 | −598.53 | 2.8 |
| nC6 | nC16[b] | 453.2 | 10.2 | −57.3 | | | | 0.6 |
| MTHF | nC6[c] | 2145.77 | 42.35 | 54.73 | | | | 1.73 |
| MTHF | nC16[d] | 3706.59 | −928.87 | 526.97 | −423.85 | | | 4.14 |

[a] Zhu et al.[1] [b] Miller and Williamson.[12] [c] Wang et al.[4] [d] Present work.

**Table 3. Experimental Excess Molar Enthalpies, $H^E_{m,1+23}$, Measured at 298.15 K, for the Addition of DIPE to nC6 + nC16 Mixtures to Form DIPE (1) + nC6 (2) + nC16 (3), and Values of $H^E_{m,123}$ Calculated from Eq 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$^{-1}$ | $x_1$ | $H^E_{m,1+23}$[a]/J·mol$^{-1}$ | $H^E_{m,123}$/J·mol$^{-1}$ |
|---|---|---|---|---|---|---|---|---|
| | | | $x_2/x_3 = 0.3335$, $H^E_{m,23}$ (J·mol$^{-1}$) = 83.27 | | | | | |
| 0.0501 | 80.40 | 159.50 | 0.4002 | 426.96 | 476.91 | 0.7002 | 412.28 | 437.25 |
| 0.1001 | 147.94 | 222.88 | 0.4502 | 443.89 | 489.67 | 0.7497 | 375.70 | 396.55 |
| 0.1500 | 215.65 | 286.43 | 0.4999 | 460.12 | 501.76 | 0.8001 | 327.95 | 344.60 |
| 0.2001 | 274.63 | 341.24 | 0.5501 | 461.80 | 499.26 | 0.8506 | 267.58 | 280.02 |
| 0.2502 | 328.29 | 390.73 | 0.6003 | 454.97 | 488.25 | 0.9000 | 195.25 | 203.58 |
| 0.3001 | 369.48 | 427.77 | 0.6503 | 438.53 | 467.65 | 0.9500 | 107.39 | 111.55 |
| 0.3496 | 398.28 | 452.44 | | | | | | |
| | | | $x_2/x_3 = 1.0004$, $H^E_{m,23}$ (J·mol$^{-1}$) = 113.30 | | | | | |
| 0.0502 | 67.21 | 174.83 | 0.3498 | 330.02 | 403.68 | 0.6499 | 352.07 | 391.74 |
| 0.1000 | 124.65 | 226.62 | 0.4001 | 350.51 | 418.48 | 0.7000 | 329.46 | 363.45 |
| 0.1501 | 177.92 | 274.21 | 0.4509 | 365.45 | 427.66 | 0.7500 | 298.83 | 327.16 |
| 0.2000 | 229.46 | 320.10 | 0.5003 | 372.38 | 428.99 | 0.7999 | 259.72 | 282.39 |
| 0.2501 | 270.59 | 355.55 | 0.5508 | 372.11 | 423.01 | 0.8500 | 210.70 | 227.69 |
| 0.3002 | 305.21 | 384.50 | 0.6002 | 366.73 | 412.03 | 0.9500 | 81.55 | 87.22 |
| | | | $x_2/x_3 = 2.9865$, $H^E_{m,23}$ (J·mol$^{-1}$) = 81.34 | | | | | |
| 0.0494 | 52.24 | 129.56 | 0.4008 | 279.88 | 328.62 | 0.6999 | 254.43 | 278.84 |
| 0.1000 | 101.62 | 174.82 | 0.4536 | 291.81 | 336.25 | 0.7503 | 230.36 | 250.67 |
| 0.1500 | 146.71 | 215.85 | 0.5004 | 294.94 | 335.57 | 0.8002 | 197.71 | 213.96 |
| 0.2002 | 185.72 | 250.77 | 0.5509 | 292.86 | 329.39 | 0.8501 | 159.26 | 171.45 |
| 0.2503 | 218.96 | 279.94 | 0.6005 | 285.50 | 317.99 | 0.9001 | 116.75 | 124.88 |
| 0.2972 | 242.81 | 299.97 | 0.6499 | 273.21 | 301.68 | 0.9500 | 63.11 | 67.18 |
| 0.3487 | 263.37 | 316.35 | | | | | | |

[a] Ternary term for representation of $H^E_{m,1+23}$ by eqs 3 and 4: $H^E_{m,T}$ (J·mol$^{-1}$) = $x_1x_2x_3(765.19 - 8562.98x_1 + 293.01x_2 + 15474.78x_1^2 + 3158.09x_1x_2 - 1582.53x_2^2 - 11329.16x_1^3)$, $s$ = 2.32 J·mol$^{-1}$.

where values of the $H^E_{m,ij}$ were calculated from the appropriate smoothing functions. The form

$$H^E_{m,T}/\text{J·mol}^{-1} = x_1x_2x_3(c_0 + c_1x_1 + c_2x_2 + c_3x_1^2 + c_4x_1x_2 + c_5x_2^2 + ...) \quad (4)$$

which was adopted for the ternary contribution, is similar to that used by Morris et al.[14] Values of the parameters $c_i$ were obtained from least-squares analyses in which eqs 3 and 4 were fitted to the experimental values in Tables 3 and 4. The resulting representations of $H^E_{m,T}$ are given in the footnotes of the tables, along with the standard deviations $s$ of the fits. The solid curves for $H^E_{m,1+23}$ in Figures 1 and 2 were calculated from eqs 3 and 4, using these representations.

Equations 1−4 were also used to calculate the constant $H^E_{m,123}$ contours plotted on the Roozeboom diagrams[15] in Figures 3a and 4a. The general characteristics of these are similar and there is no indication of an internal extremum.

Recent work[16,17] indicates that an extension of the model of Liebermann and Fried[18,19] can be useful in representing the excess enthalpies of binary mixtures and also has the potential for estimating the enthalpies of ternary mixtures 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.[17] 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 $H^E_{m,ij}$ to the primary experimental results in Table 1 and those reported

**Table 4. Experimental Excess Molar Enthalpies, $H^{E}_{m,1+23}$, Measured at 298.15 K, for the Addition of MTHF to nC6 + nC16 Mixtures To Form MTHF (1) + nC6 (2) + nC16(3), and Values of $H^{E}_{m,123}$ Calculated from Eq 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$^{-1}$ | $x_1$ | $H^{E}_{m,1+23}$ [a] J·mol$^{-1}$ | $H^{E}_{m,123}$ J·mol$^{-1}$ |
|---|---|---|---|---|---|---|---|---|
| | | | $x_2/x_3 = 0.3336$, $H^{E}_{m,23}$ (J·mol$^{-1}$) = 83.28 | | | | | |
| 0.0500 | 125.21 | 204.32 | 0.4001 | 753.17 | 803.13 | 0.7001 | 760.71 | 785.68 |
| 0.1000 | 253.87 | 328.82 | 0.4500 | 786.48 | 832.28 | 0.7500 | 701.25 | 722.07 |
| 0.1500 | 363.71 | 434.50 | 0.5002 | 814.10 | 855.72 | 0.7999 | 620.46 | 637.13 |
| 0.2000 | 464.46 | 531.08 | 0.5500 | 823.96 | 861.43 | 0.8499 | 514.63 | 527.13 |
| 0.2499 | 558.67 | 621.13 | 0.5996 | 820.33 | 853.68 | 0.9000 | 378.95 | 387.28 |
| 0.3001 | 632.81 | 691.10 | 0.6503 | 801.69 | 830.82 | 0.9500 | 210.26 | 214.42 |
| 0.3500 | 700.90 | 755.03 | | | | | | |
| | | | $x_2/x_3 = 1.0002$, $H^{E}_{m,23}$ (J·mol$^{-1}$) = 113.30 | | | | | |
| 0.0500 | 124.91 | 232.54 | 0.3996 | 680.70 | 748.73 | 0.7005 | 667.73 | 701.67 |
| 0.1000 | 235.12 | 337.10 | 0.4507 | 712.98 | 775.22 | 0.7498 | 611.51 | 639.85 |
| 0.1500 | 338.26 | 434.57 | 0.4998 | 732.98 | 789.66 | 0.8000 | 536.61 | 559.27 |
| 0.2000 | 429.64 | 520.28 | 0.5498 | 737.45 | 788.46 | 0.8500 | 439.81 | 456.80 |
| 0.2503 | 516.34 | 601.29 | 0.6000 | 730.29 | 775.61 | 0.9004 | 319.94 | 331.22 |
| 0.3000 | 586.50 | 665.81 | 0.6506 | 706.21 | 745.80 | 0.9500 | 173.72 | 179.38 |
| 0.3497 | 642.31 | 715.98 | | | | | | |
| | | | $x_2/x_3 = 3.0010$, $H^{E}_{m,23}$ (J·mol$^{-1}$) = 81.32 | | | | | |
| 0.0500 | 117.71 | 194.96 | 0.4000 | 591.21 | 640.00 | 0.6499 | 590.62 | 619.09 |
| 0.0999 | 214.42 | 287.61 | 0.4499 | 609.29 | 654.02 | 0.6998 | 555.03 | 579.44 |
| 0.1502 | 302.41 | 371.52 | 0.5000 | 624.87 | 665.53 | 0.7502 | 501.28 | 521.59 |
| 0.2002 | 385.56 | 450.60 | 0.5501 | 627.21 | 663.79 | 0.8000 | 434.51 | 450.78 |
| 0.2501 | 455.48 | 516.46 | 0.5963 | 619.89 | 652.72 | 0.8500 | 355.20 | 367.40 |
| 0.3000 | 514.24 | 571.17 | 0.5997 | 618.48 | 651.03 | 0.9500 | 139.59 | 143.65 |
| 0.3498 | 553.51 | 606.38 | | | | | | |

[a] Ternary term for representation of $H^{E}_{m,1+23}$ by eqs 3 and 4: $H^{E}_{m,T}$ (J·mol$^{-1}$) = $x_1x_2x_3(-2838.33 + 3995.75x_1 + 12127.97x_2 - 1550.43x_1^2 - 9294.34x_1x_2 - 12397.64x_2^2)$, $s$ = 4.68 J·mol$^{-1}$.

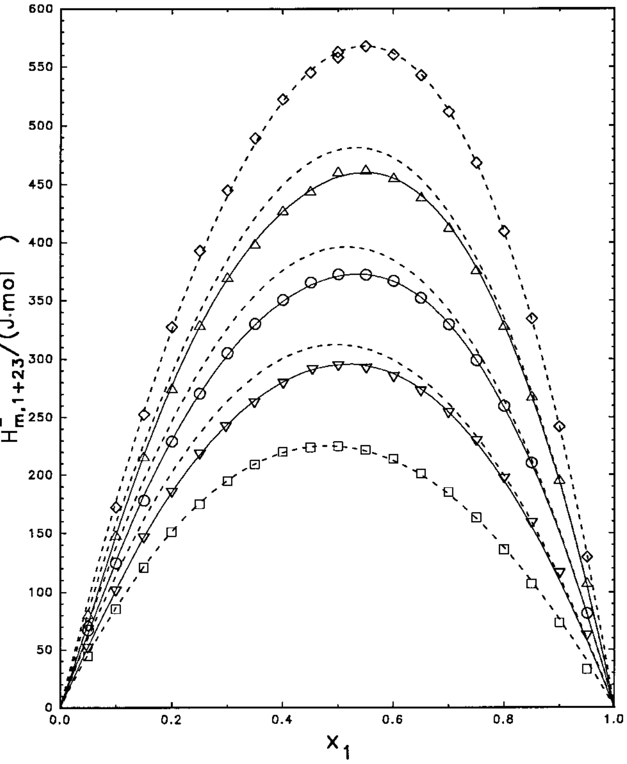

**Figure 1.** Excess molar enthalpies $H^{E}_{m,1+23}$ for DIPE (1) + nC6 (2) + nC16 (3) at 298.15 K. Experimental results: (△) $x_2/x_3$ = 0.3335; (○) $x_2/x_3$ = 1.0004; (▽) $x_2/x_3$ = 2.9865; (◇) $x_2$ = 0, Zhu et al.[1]; (□) $x_3$ = 0, Zhu et al.[1] Curves: (—) calculated from the representation of the results by eq 3, using the ternary term $H^{E}_{m,T}$ given in the footnote of Table 3; (- - -) estimated by means of the Liebermann−Fried model.

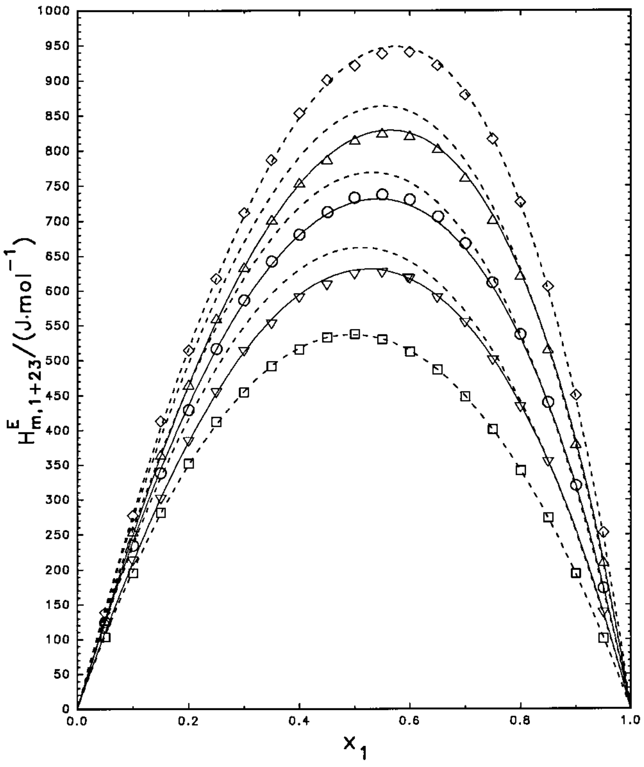

**Figure 2.** Excess molar enthalpies $H^{E}_{m,1+23}$ for MTHF (1) + nC6 (2) + nC16 (3) at 298.15 K. Experimental results: (△) $x_2/x_3$ = 0.3336; (○) $x_2/x_3$ = 1.0002; (▽) $x_2/x_3$ = 3.0010; (◇) $x_2$ = 0; (□) $x_3$ = 0, Wang et al.[4] Curves: (—) calculated from the representation of the results by eq 3, using the ternary term $H^{E}_{m,T}$ given in the footnote of Table 4; (- - -) estimated by means of the Liebermann−Fried model.

for the other relevant binaries by Zhu et al.,[1] Wang et al.,[4] and Miller and Williamson.[12] Also included in Table 5 are values of the standard deviations $s$ achieved in the fitting process and values[7,8,20] of the isobaric thermal expansivities $\alpha_p$ of the components, used in evaluating the contributions due to different molecular sizes.

**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 | | | | *s* | $\alpha_p$ (kK$^{-1}$) | |
|---|---|---|---|---|---|---|
| *i* | *j* | $A_{ij}$ | $A_{ji}$ | J·mol$^{-1}$ | *i* | *j* |
| DIPE | nC6 | 0.8493 | 0.9960 | 2.09 | 1.455[a] | 1.387[b] |
| DIPE | nC16 | 0.9982 | 0.6508 | 5.72 | 1.455[a] | 0.883[b] |
| nC6 | nC16 | 1.0713 | 0.8121 | 0.85 | 1.387[b] | 0.883[b] |
| MTHF | nC6 | 0.8155 | 0.8525 | 1.70 | 1.215[c] | 1.387[b] |
| MTHF | nC16 | 0.9923 | 0.5348 | 5.85 | 1.215[c] | 0.883[b] |

[a] Obama et al.[7] [b] Benson et al.[20] [c] Estimated from the density data of De Lorenzi et al.[8]

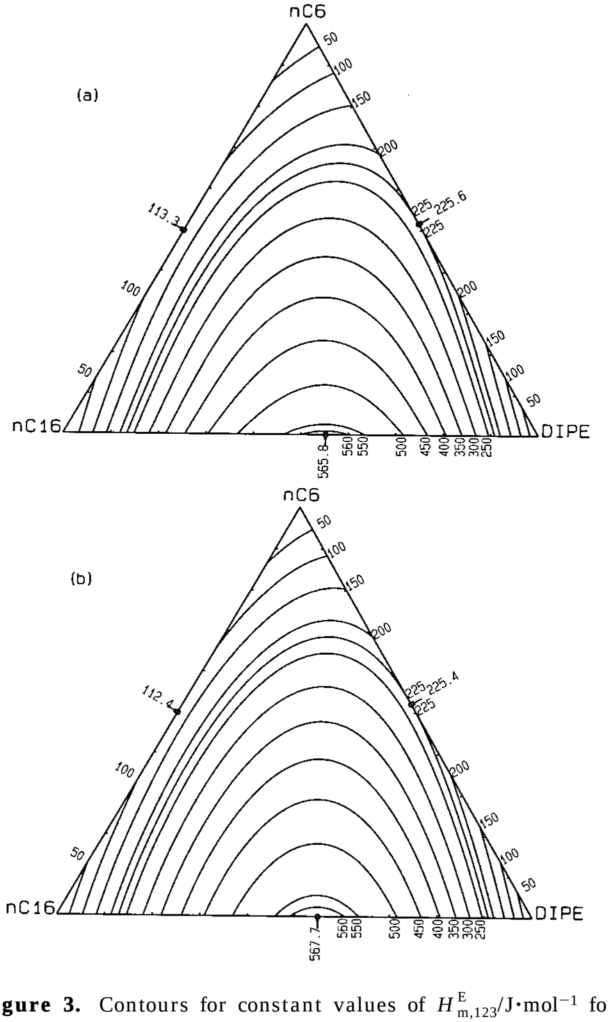

**Figure 3.** Contours for constant values of $H^{E}_{m,123}$/J·mol$^{-1}$ for DIPE (1) + nC6 (2) + nC16 (3) at 298.15 K. Part (a) calculated from the representation of the experimental results by eqs 1 and 3 with $H^{E}_{m,T}$ from the footnote of Table 3; part (b) estimated by means of the Liebermann−Fried model.

Using the values of the parameters given in Table 5, $H^{E}_{m,1+23}$ and $H^{E}_{m,123}$ were calculated for the present mixtures. The resulting values of $H^{E}_{m,1+23}$ for the ternary mixtures and $H^{E}_{m,ij}$ 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 overestimate the ternary enthalpies of both systems. The mean absolute relative deviations between the 56 experimental and estimated values of $H^{E}_{m,1+23}$ in Table 3 and the 57 points in Table 4 amount to 5.3% and 4.8%, respectively.

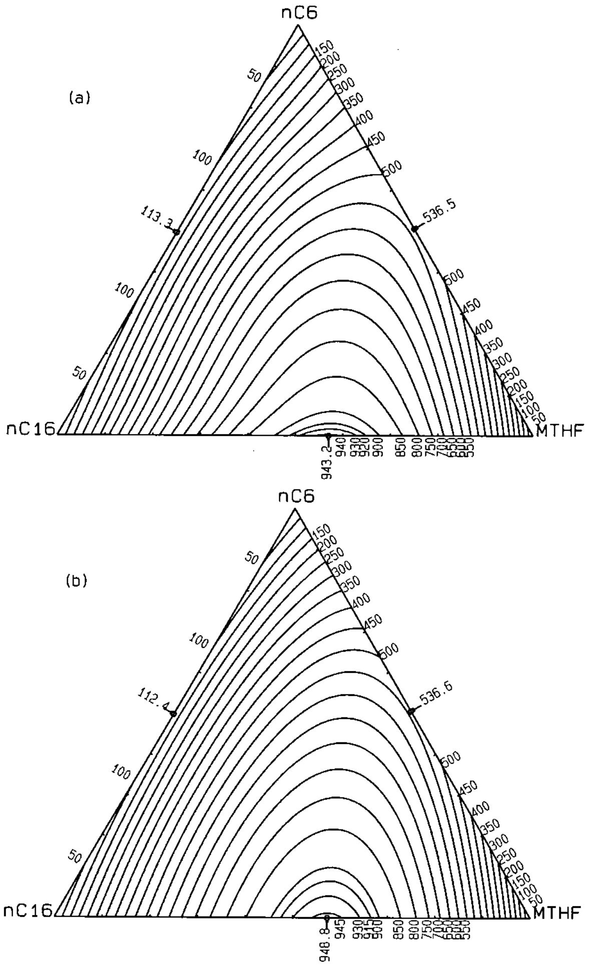

**Figure 4.** Contours for constant values of $H^{E}_{m,123}$/J·mol$^{-1}$ for MTHF (1) + nC6 (2) + nC16 (3) at 298.15 K. Part (a) calculated from the representation of the experimental results by eqs 1 and 3 with $H^{E}_{m,T}$ from the footnote of Table 4; part (b) estimated by means of the Liebermann−Fried model.

Constant $H^{E}_{m,123}$ 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 very similar to those in parts a, which were derived from the representations of the experimental data for the ternary mixtures. This again indicates that the Liebermann−Fried model provides useful estimates of the ternary enthalpy behavior without requiring the direct investigation of the excess enthalpies of any ternary mixtures.

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