Excess Enthalpies of the Ternary Mixtures Diisopropyl Ether + 3-Methylpentane + (Octane or Decane) 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 + 3-methylpentane + octane and for diisopropyl ether + 3-methylpentane + 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 constituent 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 (THF) + 3-methylpentane (3MP) + {octane (nC8) or decane (nC10)} were reported.¹ As a continuation of that investigation, similar calorimetric measurements at 298.15 K have been made for the two ternary systems formed when the THF was replaced by diisopropyl ether (DIPE).

Experimental Section

The DIPE, nC8, and nC10 used in the present work were obtained from the Aldrich Chemical Co. The 3MP was Research Grade material from the Phillips Chemical Co. In all cases the purities stated by the manufacturers were at least 99 mol %. Apart from partial degassing, the components were used without further purification. Densities, $\rho/\text{kg·m}^{-3}$, measured with a precision of 0.03 kg·m⁻³ at 298.15 K in an Anton-Paar digital densimeter, were 718.70, 659.94, 698.72, and 726.30, for DIPE, 3MP, nC8, and nC10, respectively. These results are in reasonable agreement with values in the literature.^{2,3}

An LKB flow microcalorimeter (model 10700-1), thermostated at (298.150 \pm 0.003) K, was used to measure the excess molar enthalpies $H^{\rm E}_{\rm m}$. Details of the equipment and its operation have been described previously.^{4,5}

In studying the ternary systems, the excess molar enthalpy $H_{m,1+23}^E$ was determined for several pseudo binary mixtures in which component 1 (DIPE) was added to binary mixtures of component 2 (3MP) and 3 (either nC8 or 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

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

where x_1 is the mole fraction of DIPE 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

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mixtures are estimated to be less than 0.5% and 5 \times 10 $^{-4},$ respectively.

Results and Discussion

Excess molar enthalpies $H_{m,jj}^{E}$ (i < j), measured at 298.15 K, for four of the five constituent binaries of present interest have been reported previously: DIPE (1) + nC8 (3),⁶ 3MP (2) + nC8 (3),⁷ DIPE (1) + nC10 (3),⁶ and 3MP (2) + nC10 (3).⁸ In the course of the present investigation, excess molar enthalpies at 298.15 K were determined for the fifth constituent binary, DIPE (1) + 3MP (2). These results are listed in Table 1.

The smoothing function

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

was fitted to the results 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 four 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 nC8 and nC10, 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 DIPE (1) + 3MP (2) given in Table 1 and the results for DIPE (1) + nC8 (3) and DIPE (1) + nC10 (3) reported by Zhu et al.⁶ In both figures, it can be seen that the results for the three ternary mixtures fall between those for the two constituent binaries.

The values of $H_{m,1+23}^{E}$ were represented as a sum of binary terms⁹ with an added ternary contribution:

$$H_{m,1+23}^{E} = [x_{2}/(1-x_{1})]H_{m,12}^{E} + [x_{3}/(1-x_{1})]H_{m,13}^{E} + H_{m,T}^{E}$$
(3)

Table 1. Experimental Mole Fractions, x_1 , and Excess Molar Enthalpies, $H_{m,j}^E$ for the Mixture DIPE (1) + 3MP (2) at 298.15 K

	$H_{\mathrm{m},ij}^{\mathrm{E}}$		$H_{\mathrm{m},ij}^{\mathrm{E}}$		$H_{\mathrm{m},ij}^{\mathrm{E}}$		$H_{\mathrm{m},ij}^{\mathrm{E}}$		$H_{\mathrm{m},ij}^{\mathrm{E}}$
<i>X</i> 1	J•mol ^{−1}	<i>X</i> 1	J•mol ^{−1}	<i>X</i> 1	J•mol ^{−1}	x ₁	J•mol ^{−1}	X 1	J•mol ^{−1}
0.0500	42.23	0.2503	155.55	0.4500	197.43	0.6500	176.75	0.8500	93.71
0.1000	78.58	0.2998	171.60	0.5003	197.63	0.7002	162.13	0.9000	65.94
0.1499	109.67	0.3501	183.83	0.5502	193.77	0.7501	143.57	0.9500	35.79
0.2000	136.10	0.3997	192.31	0.5998	187.30	0.8003	121.54		

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

onent						S
j	h_1	h_2	h_3	h_4	h_5	$\overline{\mathbf{J}\cdot\mathbf{mol}^{-1}}$
$3MP^{a}$	790.32	51.08	26.96	58.27		0.73
nC8 ^b	1102.00	-49.71	53.24	2.16	-121.37	1.29
nC8 ^c	65.33	-2.71				0.046
nC10 ^b	1357.07	-109.31	162.86	-58.25	-202.62	0.88
$nC10^d$	162.42	-1.51	-1.63			0.07
	<u>j</u> 3MP ^a nC8 ^b nC8 ^c nC10 ^b nC10 ^d	$\begin{tabular}{ c c c c c }\hline \hline j & h_1 \\ \hline \hline j & h_1 \\ \hline $3MP^a$ & 790.32 \\ $nC8^b$ & 1102.00 \\ $nC8^c$ & 65.33 \\ $nC10^b$ & 1357.07 \\ $nC10^d$ & 162.42 \\ \hline \end{tabular}$	$\begin{tabular}{ c c c c c c c } \hline & & & & & & & & \\ \hline \hline j & h_1 & h_2 \\ \hline \hline 3MP^a & 790.32 & 51.08 \\ nC8^b & 1102.00 & -49.71 \\ nC8^c & 65.33 & -2.71 \\ nC10^b & 1357.07 & -109.31 \\ nC10^d & 162.42 & -1.51 \\ \hline \end{tabular}$	$\begin{tabular}{ c c c c c c c c c c c c c c c c c c c$	$\begin{tabular}{ c c c c c c c c c c c c c c c c c c c$	$ \begin{array}{c c c c c c c c c c c c c c c c c c c $

^a Present work. ^b Zhu et al.⁶ ^c Hamam et al.⁷ ^d Hamam and Benson.⁸



Figure 1. Excess molar enthalpies, $H_{m,1+23}^{E}$, for DIPE (1) + 3MP (2) + nC8 (3) at 298.15 K. Experimental results: \triangle , $x_2/x_3 = 0.3332$; \bigcirc , $x_2/x_3 = 1.0002$; \bigtriangledown , $x_2/x_3 = 2.9995$; \diamondsuit , $x_2 = 0$, Zhu et al.;⁶ \square , $x_3 = 0$. Curves: \neg , 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.

where the values of $H^{\rm E}_{{\rm m}, ij}$ were calculated from the appropriate smoothing functions.

The form

$$H_{m,T}^{E}/J \cdot mol^{-1} = [x_{1}x_{2}x_{3}/(1 + b\{x_{2} - x_{1}\})](c_{0} + c_{1}x_{1} + c_{2}x_{2} + c_{3}x_{1}^{2} + c_{4}x_{1}x_{2} + c_{5}x_{2}^{2} + c_{6}x_{1}^{3} + c_{7}x_{1}^{2}x_{2} + c_{8}x_{1}x_{2}^{2} + c_{3}x_{2}^{3})$$
(4)

which was adopted for the latter contribution is similar to the form used by Morris et al.¹⁰ with the possibility of having an extra skewing factor $\{1 + b(x_2 - x_1)\}^{-1}$ for a nonzero value of *b*. Values of the parameters c_i were



Figure 2. Excess molar enthalpies, $H_{m,1+23}^{E}$, for DIPE (1) + 3MP (2) + nC10 (3) at 298.15 K. Experimental results: \triangle , $x_2/x_3 = 0.3333; \bigcirc$, $x_2/x_3 = 0.9994; \bigtriangledown$, $x_2/x_3 = 3.0013; \diamondsuit$, $x_2 = 0$, Zhu et al.;⁶ \Box , $x_3 = 0$. 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.

obtained from least-squares analyses in which eqs 3 and 4 were fitted to the values of $H_{m,1+23}^E$ in Tables 3 and 4, with b = 0 for the nC8 system and b = 1 for the nC10 system. 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 2 were calculated from eq 3, using these representations.

Equations 1-4 were also used to calculate the constant $H^{\rm E}_{m,123}$ contours plotted on the Roozeboom diagrams in Figures 3a and 4a. The general characteristics of these are similar. In both figures, all of the contours extend to the

Table 3.	Experimental	l Excess Molar	Enthalpies, $H_{m,14}^{E}$	-23, Measured	at 298.15 K,	for the Add	ition of DIPE to 3	3MP + nC8
Mixtures	s To Form DIP	PE (1) + 3MP (2	2) + nC8 (3), and V	alues of $H_{m,12}^{E}$	a Calculated	from Eq 1		

					•	-		
	$H^{\!\mathrm{E}}_{\mathrm{m},1+23}$ a	$H_{\mathrm{m},123}^{\mathrm{E}}$		$H^{\!\mathrm{E}}_{\mathrm{m},1+23}$ a	$H_{\mathrm{m},123}^{\mathrm{E}}$		$H^{\!\mathrm{E}}_{\mathrm{m},1+23}$ a	$H_{\mathrm{m,123}}^{\mathrm{E}}$
<i>X</i> ₁	$J \cdot mol^{-1}$	J•mol ^{−1}	<i>X</i> 1	J•mol ^{−1}	J•mol ^{−1}	<i>X</i> 1	$J \cdot mol^{-1}$	$J \cdot mol^{-1}$
			$x_2/x_3 = 0.3$	332; H_{m23}^{E} (J·mol	l^{-1}) = 11.99			
0.0501	48.81	60.20	0.3999	245.49	252.69	0.6998	217.29	220.89
0.1000	90.79	101.58	0.4504	253.55	260.14	0.7498	194.63	197.63
0.1500	131.45	141.64	0.5000	256.21	262.21	0.8004	166.06	168.45
0.1999	163.70	173.30	0.5501	254.59	259.99	0.8508	132.53	134.32
0.2501	193.86	202.85	0.6000	247.22	252.02	0.8999	93.75	94.95
0.3017	211.88	220.26	0.6502	234.32	238.52	0.9499	50.22	50.82
0.3498	232.96	240.76						
			$x_2/x_3 = 1.0$	002; $H_{\rm m}^{\rm E}_{23}$ (J·mol	l^{-1}) = 16.33			
0.0499	44.78	60.30	0.4001	226.62	236.42	0.7000	197.57	202.47
0.1000	85.76	100.46	0.4500	233.63	242.61	0.7504	176.34	180.42
0.1500	122.77	136.65	0.5001	234.18	242.34	0.7999	150.93	154.20
0.2002	154.18	167.24	0.5503	233.20	240.54	0.8500	120.83	123.28
0.2503	178.31	190.55	0.6003	224.62	231.15	0.8999	84.98	86.61
0.3007	198.49	209.91	0.6500	213.74	219.46	0.9499	45.44	46.26
0.3502	215.29	225.90						
			$x_2/x_3 = 2.9$	995; H_{m}^{E} 23(J·mol	l^{-1}) = 12.50			
0.0500	42.58	54.46	0.3995	208.36	215.87	0.7000	178.87	182.62
0.1000	81.33	92.58	0.4501	214.40	221.28	0.7500	159.38	162.51
0.1500	115.05	125.68	0.4994	215.21	221.47	0.7992	135.53	138.04
0.1999	143.71	153.72	0.5503	212.50	218.12	0.8517	105.98	107.83
0.2508	165.46	174.83	0.5999	205.39	210.39	0.8998	75.97	77.22
0.3004	186.18	194.93	0.6497	194.19	198.57	0.9499	40.42	41.05
0.3498	197.97	206.10						

^{*a*} Ternary term for representation of $H_{m,1+23}^{E}$ by eqs 3 and 4: $H_{m,T}^{E}/J \cdot mol^{-1} = x_1 x_2 x_3 (141.72 - 250.27 x_1 - 254.17 x_2)$, $s = 1.03 J \cdot mol^{-1}$.

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

	$H_{\rm m,1+23}^{\rm E}$ a	$H_{ m m,123}^{ m E}$		$H_{\mathrm{m,1+23}}^{\mathrm{E}}$	$H_{ m m,123}^{ m E}$		$H_{\rm m,1+23}^{ m E}$ a	$H_{\mathrm{m},123}^{\mathrm{E}}$
<i>X</i> 1	J•mol ^{−1}	$\overline{\mathbf{J}\cdot\mathbf{mol}^{-1}}$	<i>X</i> 1	J•mol ^{−1}	$\overline{\mathbf{J}\cdot\mathbf{mol}^{-1}}$	<i>X</i> 1	$J \cdot mol^{-1}$	$\overline{\mathbf{J}\cdot\mathbf{mol}^{-1}}$
			$x_2/x_3 = 0.3$	333; H ^E _{m 23} (J·mol	$ ^{-1}) = 30.23$			
0.0496	42.86	71.60	0.3973	276.61	294.83	0.6977	254.04	263.18
0.0990	92.27	119.51	0.4470	287.24	303.96	0.7478	228.81	236.44
0.1485	139.74	165.48	0.4963	291.20	306.43	0.7984	196.39	202.48
0.1983	179.35	203.59	0.5467	293.70	307.41	0.8489	156.81	161.38
0.2478	213.64	236.38	0.5979	286.65	298.81	0.8989	110.24	113.30
0.2977	238.45	259.69	0.6472	273.58	284.25	0.9494	56.19	57.72
0.3477	260.44	280.16						
			$x_2/x_3 = 0.9$	994; H ^E 22(J·mol	$ ^{-1}) = 40.60$			
0.0495	48.78	87.37	0.3971	249.27	273.75	0.6972	224.00	236.29
0.0990	93.66	130.25	0.4503	256.79	279.11	0.7477	201.24	211.49
0.1485	132.45	167.03	0.4971	260.91	281.33	0.7982	172.65	180.85
0.1982	165.70	198.26	0.5461	260.30	278.73	0.8481	138.12	144.29
0.2477	195.71	226.26	0.5975	253.86	270.20	0.8990	97.68	101.78
0.2976	216.77	245.29	0.6475	241.42	255.73	0.9494	51.35	53.41
0.3469	235.12	261.64						
			$x_2/x_3 = 3.0$	013; $H_{\rm m}^{\rm E}_{23}$ (J·mol	$ ^{-1}) = 30.51$			
0.0499	45.11	74.10	0.3999	220.16	238.47	0.7007	190.95	200.08
0.0998	85.53	113.00	0.4501	226.66	243.44	0.7497	170.86	178.50
0.1500	119.47	145.40	0.4966	228.45	243.81	0.8001	144.66	150.76
0.2000	150.24	174.65	0.5500	225.57	239.30	0.8503	114.95	119.52
0.2498	176.08	198.97	0.6001	218.46	230.66	0.8998	81.61	84.67
0.2996	193.52	214.89	0.6497	207.05	217.74	0.9499	43.20	44.73
0.3502	209.35	229.18						

^{*a*} Ternary term for representation of $H_{m,1+23}^{E}$ by eqs 3 and 4: $H_{m,T}^{E}/J \cdot mol^{-1} = [x_1x_2x_3/(1-x_1+x_2)](-2299.60+7663.10x_1+6132.61x_2-10725.92x_1^2-12730.74x_1x_2-4033.34x_2^2+4867.66x_1^3+10774.47x_1^2x_2), s = 1.52 J \cdot mol^{-1}.$

edges of the triangle, and there is no indication of an internal maximum.

the Liebermann-Fried model can represent the enthalpies of the present ternary systems.

Recent work^{11,12} indicates that an extension of the model of Liebermann and Fried^{13,14} 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 equations used in this application have been outlined by Wang et al.¹² 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



Figure 3. Contours for constant values of $H_{m,123}^{E}/J \cdot mol^{-1}$ for DIPE (1) + 3MP (2) + nC8 (3) at 298.15 K: a, calculated from the representation of the experimental results by eqs 1–4 with $H_{m,T}^{E}$ from the footnote of Table 3; b, estimated by means of the Liebermann–Fried model.

for the other relevant binaries by Zhu et al.,⁶ Hamam et al.,⁷ and Hamam and Benson.⁸ Also included in Table 5 are values of the standard deviations *s* achieved in the fitting process and values^{2,15,16} of the isobaric thermal expansivities α_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 the fits of the data for the constituent binaries are relatively good. In the case of the mixtures with nC8, the Liebermann–Fried model predicts the ternary enthalpies quite well. In the case of the mixtures with nC10, the model tends to overestimate the ternary enthalpies. The mean absolute relative deviations between the 57 experimental and estimated values of $H_{m,1+23}^{E}$ in Tables 3 and 4 amount to 0.8% and 3.6%, 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



Figure 4. Contours for constant values of $H^{\rm E}_{m,123}/J \cdot {\rm mol}^{-1}$ for DIPE (1) + 3MP (2) + nC10 (3) at 298.15 K: a, calculated from the representation of the experimental results by eqs 1–4 with $H^{\rm E}_{m,T}$ from the footnote of Table 4; b, 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, α_{p} , at 298.15 K for Liebermann–Fried Model Calculations

component				S	α _p (k	K ⁻¹)
i	j	A_{ij}	A_{ji}	$\overline{\mathbf{J}\cdot\mathbf{mol}^{-1}}$	i	j
DIPE	3MP	0.8486	1.0156	1.27	1.455 ^a	1.396 ^b
DIPE	nC8	0.9429	0.8618	1.60	1.455^{a}	1.164 ^c
3MP	nC8	1.0588	0.9251	0.05	1.396^{b}	1.164 ^c
DIPE	nC10	0.9689	0.7941	2.07	1.455^{a}	1.051 ^c
3MP	nC10	1.0597	0.8958	0.07	1.396^{b}	1.051 ^c

^a Obama et al.² ^b Riddick et al.¹⁵ See page 96. ^c Benson et al.¹⁶

parts a, which were derived from the representations of the experimental data for the ternary mixtures. It is evident from these comparisons that the Liebermann– Fried model provides useful predictions of the ternary enthalpy behavior without requiring the direct investigation of the excess enthalpies of any ternary mixtures.

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