

Excess enthalpies of 14 binary liquid mixtures of trichloromethane + methyl n-alkanoates (C₃ - C₁₆) at 298.15 K

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Enthalpy-of-mixing measurements are reported at 298.15 K for trichloromethane (chloroform) + methyl n-alkanoates (ethanoate through pentadecanoate). All the mixtures are strongly exothermic suggesting the existence of strong specific interactions due to hydrogen bonding between the H atoms of trichloromethane and the O atoms of the n-alkanoates.

1. INTRODUCTION

In continuation of our experimental studies on the excess molar enthalpies H^E of 1,1,2,2-tetrachloroethane + methyl-, ethyl-, or propyl-n-alkanoates [ORTJ0915; ORTJ0951], we have determined H^E of trichloromethane + methyl n-alkanoates (ethanoate through pentadecanoate) at 298.15 K. These data, along with our previous data, will be used to test the applicability of various group-contribution models to mixtures involving complex formation. As far as we know, no H^E measurements have been published previously on this class of mixtures except for trichloromethane + methyl ethanoate at 308.15 K [OHTT0800] and for equimolar mixtures of trichloromethane + methyl ethanoate or + methyl propanoate [SEAS1531] at 298.15 K.

2. EXPERIMENTAL SECTION

2.1. Apparatus and Procedure

The experimental data were taken at atmospheric pressure by means of a Calvet type microcalorimeter, model MS-80D (SETARAM, Lyon, France) with a stainless steel batch mixing cell (volume *ca.* 8 cm³) and with negligible vapor phase. The temperature T was maintained constant to within 0.02 K at (298.15 ± 0.02) K. All temperatures are on ITS-90. The microcalorimeter was calibrated electrically after each measurement (see [ORTJ0881]). Check measurements on cyclohexane + hexane and benzene + heptadecane are in agreement to within 1 % (over central range of concentration) with the data reported in [MCGM0690] and [DIAM0742]. The estimated uncertainties in the

mole fraction composition x_i and H^E are, respectively, $\sigma(x_i) = 0.0005$ and $\sigma(H^E) = 0.02 |H^E|$ (over central range of concentration).

2.2. Materials

CHCl₃, Trichloromethane (Chloroform). Aldrich Chem. Co., Inc. (Milwaukee, WI, USA) material of stated purity > 99 mole %, was degassed ultrasonically, dried over molecular sieves Type 3A (reference 69828, from Fluka), and used without further purification. $n(D, 298.15 \text{ K}) = 1.4429$ (1.44293 [RIDJ0860]); $\rho(298.15 \text{ K})/\text{kg m}^{-3} = 1480.03$ (1479.70 [RIDJ0860]).

C₃H₆O₂, Methyl ethanoate (Methyl acetate). Fluka AG (Buchs, Switzerland) 'puriss' grade material of stated GLC purity > 99.0 mole %, purified as above; $n(D, 298.15 \text{ K}) = 1.3589$ (1.3589 [RIDJ0860]); $\rho(298.15 \text{ K})/\text{kg m}^{-3} = 927.03$ (927.9 [RIDJ0860]).

C₄H₈O₂, Methyl propanoate (Methyl propionate). Fluka AG (Buchs, Switzerland) 'purum' grade material of stated GLC purity > 99.0 mole %, purified as above; $n(D, 298.15 \text{ K}) = 1.3745$; $\rho(298.15 \text{ K})/\text{kg m}^{-3} = 909.30$.

C₅H₁₀O₂, Methyl butanoate (Methyl butyrate). Fluka AG (Buchs, Switzerland) 'purum' grade material of stated GLC purity > 99.0 mole %, purified as above; $n(D, 298.15 \text{ K}) = 1.3849$; $\rho(298.15 \text{ K})/\text{kg m}^{-3} = 892.31$.

C₆H₁₂O₂, Methyl pentanoate (Methyl valerate). Fluka AG (Buchs, Switzerland) 'purum' grade material of stated GLC purity > 99.0 mole %, purified as above; $n(D, 298.15 \text{ K}) = 1.3947$; $\rho(298.15 \text{ K})/\text{kg m}^{-3} = 884.52$.

C₇H₁₄O₂, Methyl hexanoate (Methyl caproate). Fluka AG (Buchs, Switzerland) 'puriss' grade material of stated GLC purity > 99.0 mole %, purified as above; $n(D, 298.15 \text{ K}) = 1.4035$; $\rho(298.15 \text{ K})/\text{kg m}^{-3} = 879.52$.

C₈H₁₆O₂, Methyl heptanoate (Methyl enanthate). Fluka AG (Buchs, Switzerland) 'puriss' grade material of stated GLC purity > 99.0 mole %, purified as above; $n(D, 298.15 \text{ K}) = 1.4095$; $\rho(298.15 \text{ K})/\text{kg m}^{-3} = 875.40$.

C₉H₁₈O₂, Methyl octanoate (Methyl caprylate). Fluka AG (Buchs, Switzerland) 'puriss' grade material of stated GLC purity > 99.0 mole %, purified as above; $n(D, 298.15 \text{ K}) = 1.4148$; $\rho(298.15 \text{ K})/\text{kg m}^{-3} = 872.38$.

C₁₀H₂₀O₂, Methyl nonanoate (Methyl pelargonate). Fluka AG (Buchs, Switzerland) 'puriss' grade material of

stated GLC purity > 99.0 mole %, purified as above; $n(D, 298.15 \text{ K}) = 1.4208$; $\rho(298.15 \text{ K})/\text{kg m}^{-3} = 870.11$.

$\text{C}_{11}\text{H}_{22}\text{O}_2$, **Methyl decanoate** (Methyl caprate). Fluka AG (Buchs, Switzerland) 'puriss' grade material of stated GLC purity > 99.0 mole %, purified as above; $n(D, 298.15 \text{ K}) = 1.4235$; $\rho(298.15 \text{ K})/\text{kg m}^{-3} = 868.09$.

$\text{C}_{12}\text{H}_{24}\text{O}_2$, **Methyl undecanoate**. Fluka AG (Buchs, Switzerland) 'puriss' grade material of stated GLC purity > 98.0 mole %, purified as above; $n(D, 298.15 \text{ K}) = 1.4270$; $\rho(298.15 \text{ K})/\text{kg m}^{-3} = 866.75$.

$\text{C}_{13}\text{H}_{26}\text{O}_2$, **Methyl dodecanoate** (Methyl laurate). Fluka AG (Buchs, Switzerland) 'puriss' grade material of stated GLC purity > 99.5 mole %, purified as above; $n(D, 298.15 \text{ K}) = 1.4298$; $\rho(298.15 \text{ K})/\text{kg m}^{-3} = 865.12$.

$\text{C}_{14}\text{H}_{28}\text{O}_2$, **Methyl tridecanoate**. Fluka AG (Buchs, Switzerland) 'puriss' grade material of stated GLC purity > 98.0 mole %, purified as above; $n(D, 298.15 \text{ K}) = 1.4329$; $\rho(298.15 \text{ K})/\text{kg m}^{-3} = 863.94$.

$\text{C}_{15}\text{H}_{30}\text{O}_2$, **Methyl tetradecanoate** (Methyl myristate). Fluka AG (Buchs, Switzerland) 'puriss' grade material of stated GLC purity > 99.5 mole %, purified as above; $n(D, 298.15 \text{ K}) = 1.4345$; $\rho(298.15 \text{ K})/\text{kg m}^{-3} = 863.27$.

$\text{C}_{16}\text{H}_{32}\text{O}_2$, **Methyl pentadecanoate**. Fluka AG (Buchs, Switzerland) 'puriss' grade material of stated GLC purity > 99.0 mole %, purified as above; $n(D, 298.15 \text{ K}) = 1.4370$; $\rho(298.15 \text{ K})/\text{kg m}^{-3} = 862.35$.

3. RESULTS

The direct experimental H^E values are tabulated and graphed in the Appendix and saved on disk as Standard ELDATA Files **ORTJ0961.001** through **ORTJ0961.014**.

The data were fitted to Eq. (1):

$$H^E_{\text{calc}}/\text{J mol}^{-1} = x_1 x_2 \sum A_i [x_1/(x_1 + kx_2)]^{i-1} \quad (1)$$

all points weighted equally. With an adjusted coefficient k and $n = 4$ coefficients A_i , the standard deviations $\sigma(H^E)$, defined by Eq.(2):

$$\sigma(H^E) = [\sum (H^E_{\text{calc}} - H^E)^2 / (N-n)]^{1/2} \quad (2)$$

where N is the number of experimental values, are less than 10 J mol^{-1} (*ca.* 0.5 % at $x_1 = 0.5$).

4. DISCUSSION AND CONCLUSIONS

Comparison with literature values is possible only for trichloromethane + methyl ethanoate or + methyl propanoate. The equimolar H^E values at 298.15 K reported by [SEAS1531] are by 200-300 J mol^{-1} less negative than our measurements. However, the same discrepancies exist between the measurements of [SEAS1531] and other authors. For example, the data of [SEAS1531] are also by 200-300 J mol^{-1} less negative than the data reported by [BEAL0690] and [HALF0720] for trichloromethane + diethyl ether or the data reported by [HANY0750] and [HALF0720] for trichloromethane + propan-2-one.

The excess enthalpies of all the systems studied are strongly negative, between -1700 and -2300 J mol^{-1} at the minimum situated at a mole fraction of trichloromethane of *ca.* $x_1 = 0.55$. This suggests that in trichloromethane + methyl n-alkanoate mixtures there exist specific interactions between the unlike molecules (mainly due to hydrogen bonding) with formation of 1:1 and 2:1 complexes. The ideal association model of [MCGM0580], based on the assumption of this type of complex formation, has already been used to describe the properties of trichloromethane + ethyl methanoate, + methyl ethanoate, or + ethyl ethanoate [OHTT0800].

As expected, because of the decrease of the association constant with temperature, the H^E value of trichloromethane + methyl ethanoate at 308.15 K, obtained by [OHTT0800], is less negative than we obtained at 298.15 K.

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Property Code: [HMSD1000] HEAT OF MIXING AND SOLUTION			ORTJ0961.001
State: Two-component system, single-phase liquid or two-phase liquid-liquid (LL)			
Pure component 1, liquid			
Pure component 2, liquid			
Parameters: [P1] T/K, Temperature			
Variables: [X1] x_1 /-, Mole fraction of component 1			
[Y1] H^E /J mol ⁻¹ , Molar excess enthalpy			
Method: Direct low-pressure calorimetric measurement of H^E at variable x_1 and constant T			
Components: 1. CHCl ₃ , Trichloromethane 2. C ₃ H ₆ O ₂ , Methyl ethanoate			
[P1] = .298150E+03			
[X1]	[Y1]		
.409000E-01	-.185300E+03		
.806000E-01	-.349200E+03		
.132300E+00	-.588100E+03		
.187400E+00	-.837600E+03		
.247300E+00	-.108970E+04		
.305500E+00	-.130310E+04		
.360900E+00	-.150440E+04		
.413200E+00	-.166090E+04		
.458700E+00	-.178100E+04		
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.938700E+00	-.456500E+03		
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			-.100E+04
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			-.200E+04
			.50E+00
			[X1]

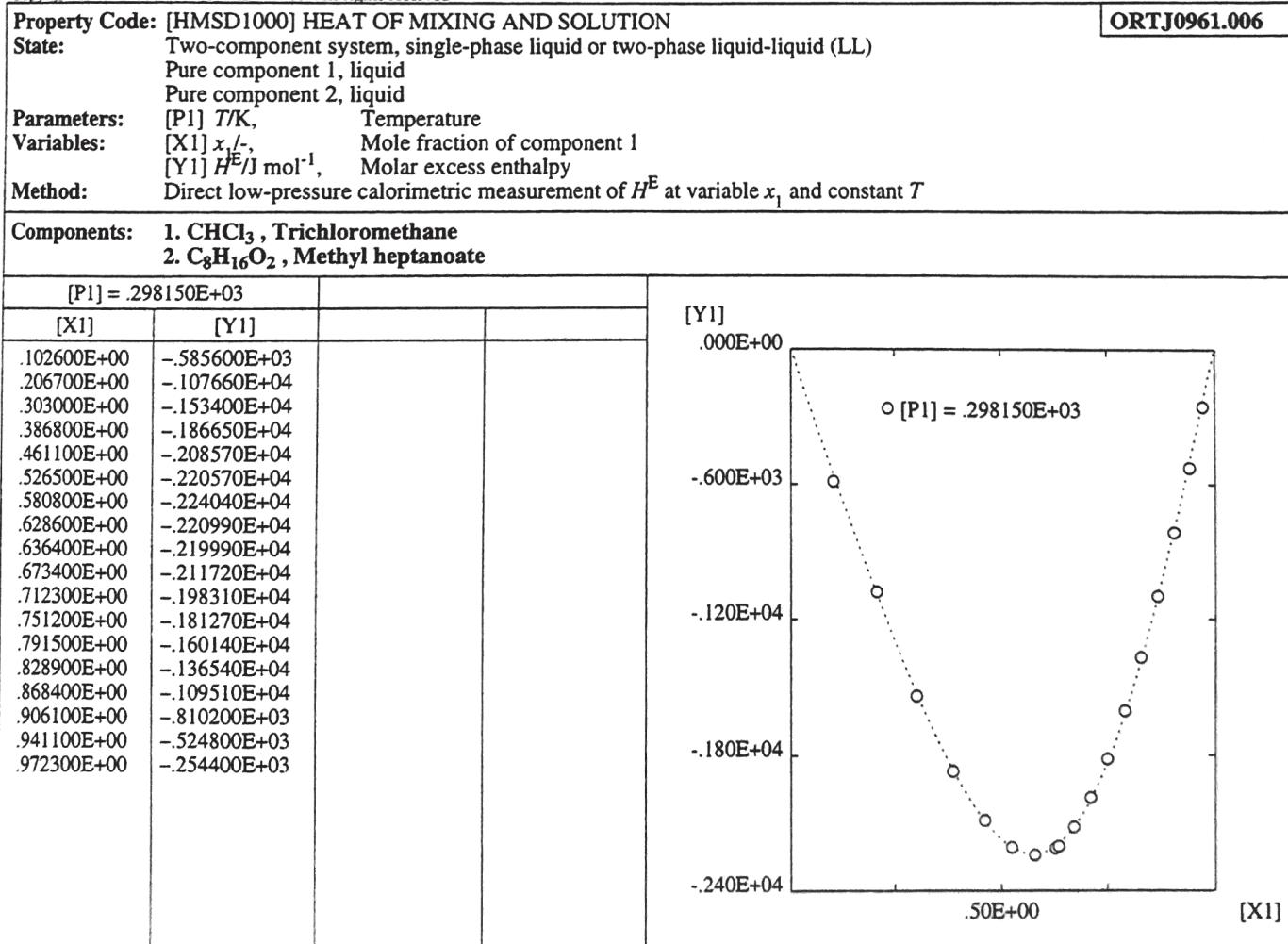
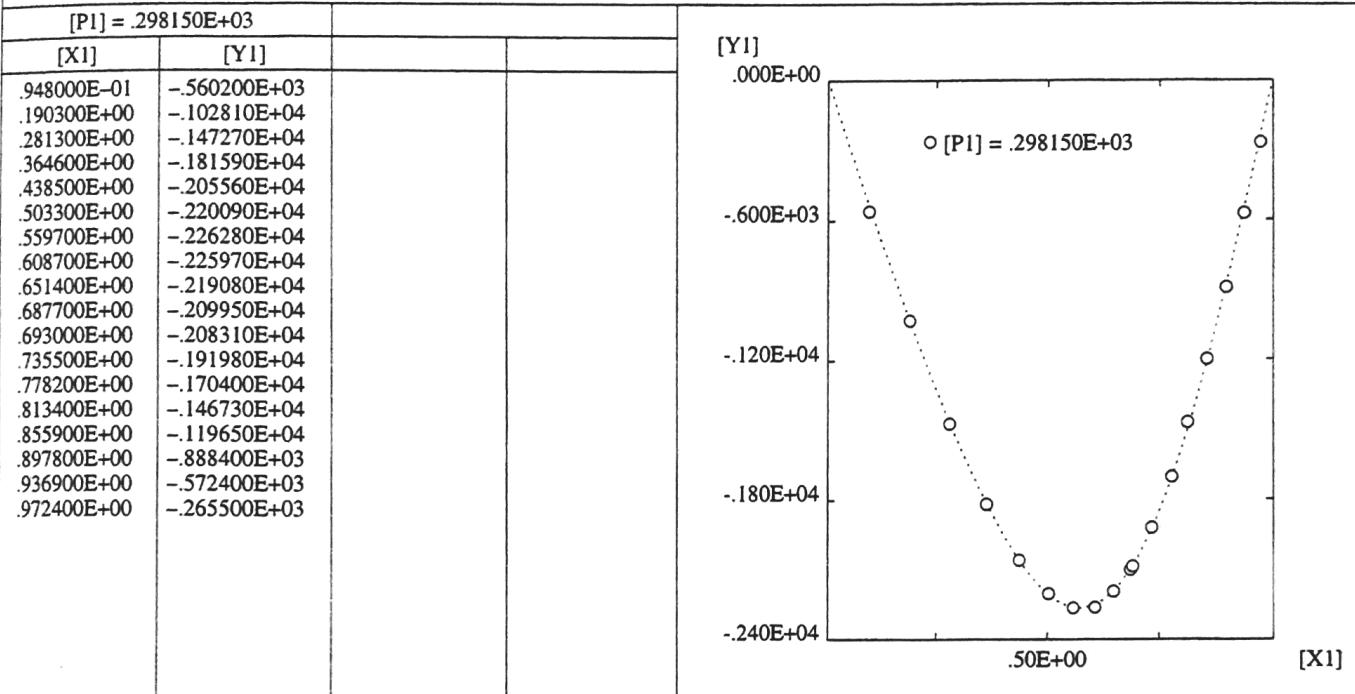
Property Code: [HMSD1000] HEAT OF MIXING AND SOLUTION			ORTJ0961.002
State: Two-component system, single-phase liquid or two-phase liquid-liquid (LL)			
Pure component 1, liquid			
Pure component 2, liquid			
Parameters: [P1] T/K, Temperature			
Variables: [X1] x_1 /-, Mole fraction of component 1			
[Y1] H^E /J mol ⁻¹ , Molar excess enthalpy			
Method: Direct low-pressure calorimetric measurement of H^E at variable x_1 and constant T			
Components: 1. CHCl ₃ , Trichloromethane 2. C ₄ H ₈ O ₂ , Methyl propanoate			
[P1] = .298150E+03			
[X1]	[Y1]		
.626000E-01	-.317500E+03		
.129200E+00	-.651000E+03		
.194500E+00	-.989700E+03		
.258700E+00	-.129680E+04		
.319800E+00	-.156090E+04		
.379400E+00	-.178120E+04		
.431600E+00	-.194080E+04		
.477400E+00	-.204660E+04		
.510300E+00	-.210520E+04		
.584400E+00	-.213440E+04		
.621900E+00	-.210690E+04		
.659200E+00	-.203630E+04		
.694100E+00	-.192430E+04		
.729900E+00	-.179500E+04		
.763300E+00	-.165010E+04		
.799700E+00	-.146640E+04		
.836500E+00	-.125340E+04		
.873800E+00	-.100940E+04		
.909600E+00	-.729400E+03		
.955100E+00	-.390200E+03		
		[Y1]	.000E+00
			-.600E+03
			-.120E+04
			-.180E+04
			-.240E+04
			.50E+00
			[X1]

Property Code:	[HMSD1000] HEAT OF MIXING AND SOLUTION	ORTJ0961.003
State:	Two-component system, single-phase liquid or two-phase liquid-liquid (LL)	
Pure component 1, liquid		
Pure component 2, liquid		
Parameters:	[P1] T/K, Temperature	
Variables:	[X1] $x_1/-$, Mole fraction of component 1	
	[Y1] $H^E/J\ mol^{-1}$, Molar excess enthalpy	
Method:	Direct low-pressure calorimetric measurement of H^E at variable x_1 and constant T	
Components:	1. CHCl ₃ , Trichloromethane 2. C ₅ H ₁₀ O ₂ , Methyl butanoate	
[P1] = .298150E+03		
[X1]	[Y1]	
.679000E-01 .145900E+00 .218400E+00 .293500E+00 .362300E+00 .425900E+00 .480800E+00 .532200E+00 .577200E+00 .607300E+00 .651900E+00 .699800E+00 .749900E+00 .798600E+00 .848900E+00 .889600E+00 .931300E+00 .970000E+00	-.408300E-03 -.811700E+03 -.119130E+04 -.153620E+04 -.181130E+04 -.201410E+04 -.214070E+04 -.221400E+04 -.223390E+04 -.222040E+04 -.215180E+04 -.203130E+04 -.185010E+04 -.162180E+04 -.133590E+04 -.102570E+04 -.669300E+03 -.322000E+03	
		[Y1] .000E+00 -.600E+03 -.120E+04 -.180E+04 -.240E+04
		[X1] .50E+00

Property Code:	[HMSD1000] HEAT OF MIXING AND SOLUTION	ORTJ0961.004
State:	Two-component system, single-phase liquid or two-phase liquid-liquid (LL)	
Pure component 1, liquid		
Pure component 2, liquid		
Parameters:	[P1] T/K, Temperature	
Variables:	[X1] $x_1/-$, Mole fraction of component 1	
	[Y1] $H^E/J\ mol^{-1}$, Molar excess enthalpy	
Method:	Direct low-pressure calorimetric measurement of H^E at variable x_1 and constant T	
Components:	1. CHCl ₃ , Trichloromethane 2. C ₆ H ₁₂ O ₂ , Methyl pentanoate	
[P1] = .298150E+03		
[X1]	[Y1]	
.860000E-01 .173900E+00 .259800E+00 .340200E+00 .413100E+00 .476600E+00 .532100E+00 .581800E+00 .593200E+00 .624100E+00 .665600E+00 .709600E+00 .754300E+00 .799000E+00 .842300E+00 .887300E+00 .929600E+00 .966000E+00	-.514500E+03 -.957300E+03 -.138750E+04 -.173840E+04 -.199650E+04 -.216020E+04 -.224630E+04 -.227340E+04 -.226490E+04 -.223820E+04 -.214940E+04 -.199930E+04 -.180300E+04 -.156140E+04 -.127740E+04 -.954600E+03 -.618100E+03 -.311300E+03	
		[Y1] .000E+00 -.600E+03 -.120E+04 -.180E+04 -.240E+04
		[X1] .50E+00

Property Code:	[HMSD1000] HEAT OF MIXING AND SOLUTION	ORTJ0961.005
State:	Two-component system, single-phase liquid or two-phase liquid-liquid (LL)	
Pure component 1, liquid		
Pure component 2, liquid		
Parameters: [P1] T/K, Temperature		
Variables: [X1] $x_1/-$, Mole fraction of component 1		
[Y1] $H^E/J\ mol^{-1}$, Molar excess enthalpy		
Method: Direct low-pressure calorimetric measurement of H^E at variable x_1 and constant T		

Components: 1. CHCl₃, Trichloromethane
2. C₇H₁₄O₂, Methyl hexanoate



Property Code: [HMSD1000] HEAT OF MIXING AND SOLUTION

State: Two-component system, single-phase liquid or two-phase liquid-liquid (LL)

ORTJ0961.007

Pure component 1, liquid

Pure component 1, liquid
Pure component 2, liquid

Parameters: P_{tot} component 2, liquid [P1] T/K Temp

Parameters: [P1] T/R; Temperature
Variables: [X1] x₁/z₁; Mole fraction

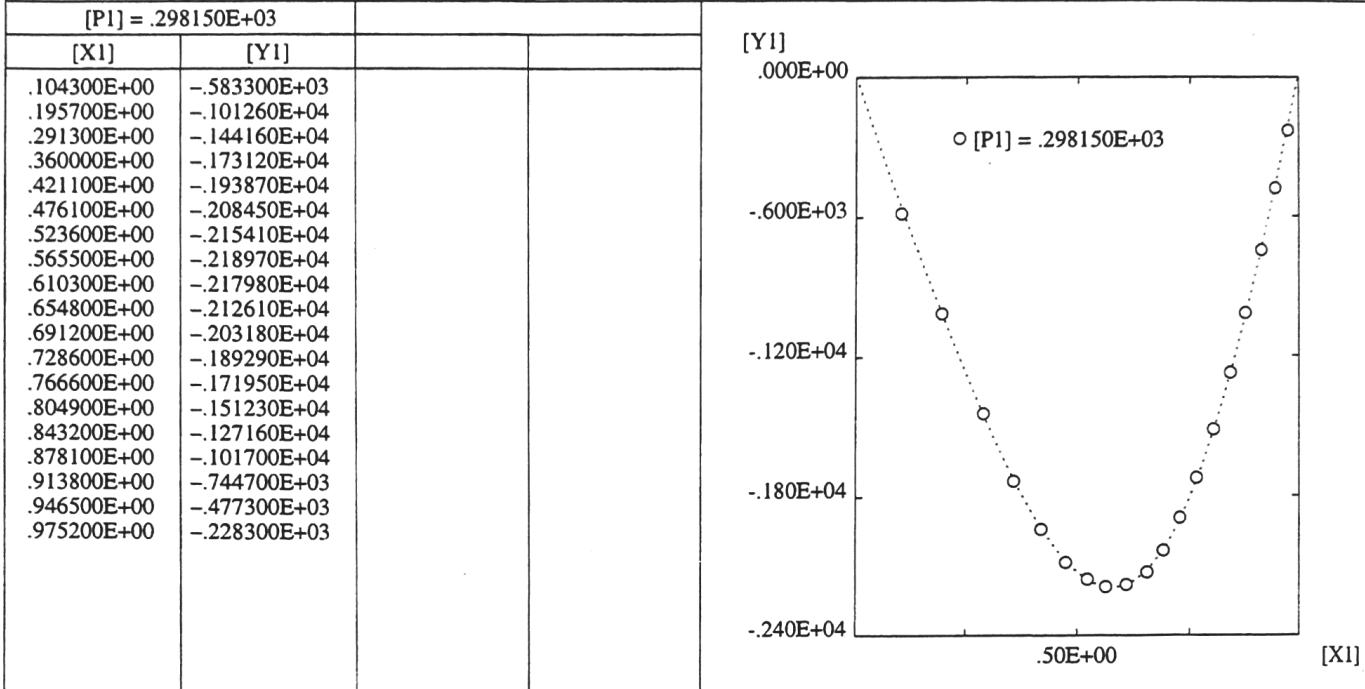
$[X_1] x_1^E$, Mole fraction of component 1
 $[Y_1] H_E^E/J \text{ mol}^{-1}$, Molar excess enthalpy

Method: Direct low-pressure calorimetric measurement

Method: Direct low-pressure calorimetric measurement of H°_f at variable x_1 and constant T

Components: 1. CHCl_3 , Trichloromethane
2. $\text{C}_2\text{H}_5\text{O}_2$, Methyl acetate

2. C₉H₁₈O₂, Methyl octanoate



Property Code: [HMSD1000] HEAT OF MIXING AND SOLUTION

State: Two-component system, single-phase liquid or two-phase liquid-liquid (LL)

ORTJ0961.008

Pure component 1, liquid

Pure component 2, liquid

Parameters: [P1] T/K , Temp

Variables: $[X_1] x_1$, Mole fraction of component 1

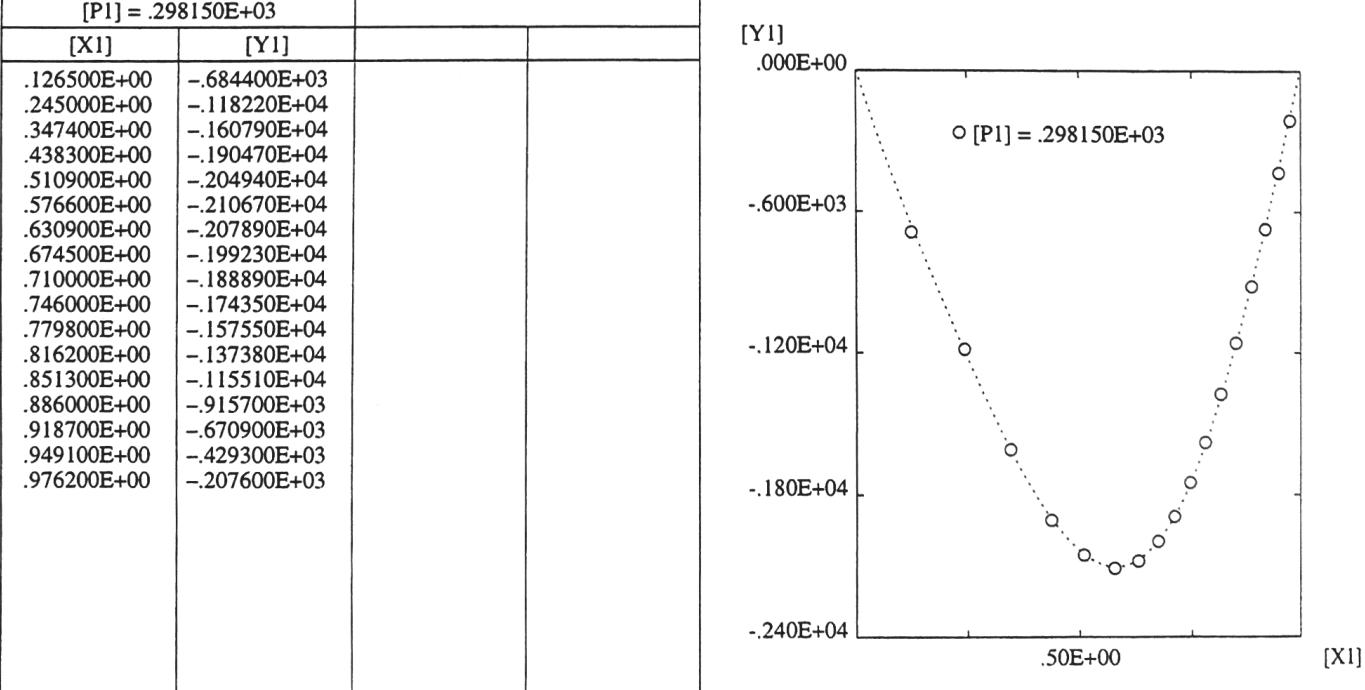
[Y1] $H^E/J \text{ mol}^{-1}$, Molar excess enthalpy

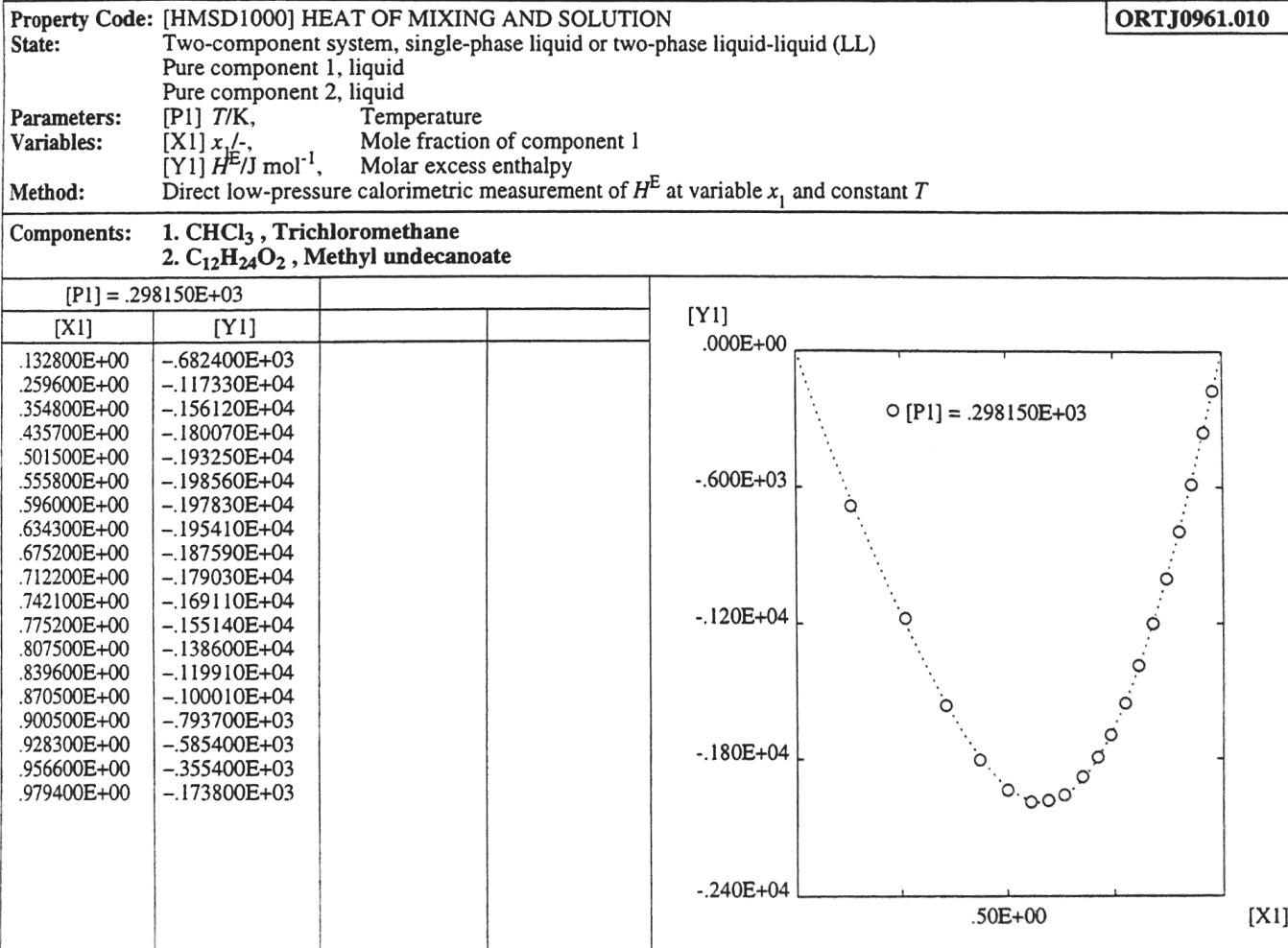
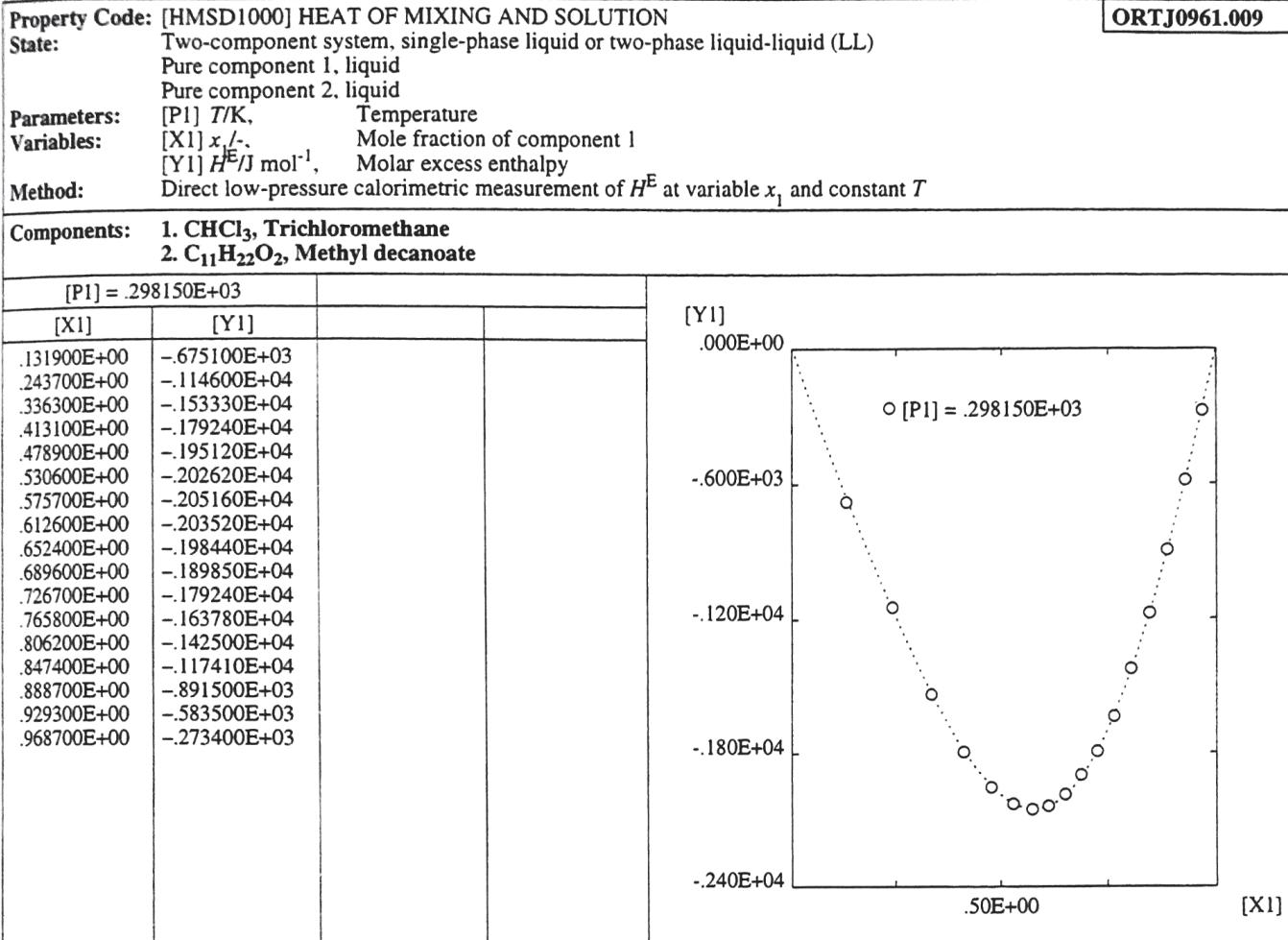
Method: Direct low-pressure calorimetric measurement

Components: 1. CHCl₃ Trichloromethane

Components: 1. CHCl_3 , Trichloromethane
2. $\text{C}_{12}\text{H}_{22}\text{O}_2$ - Methyl nonanoate

2. C₁₀H₂₀O₂, Methyl nonanoate





Property Code:	[HMSD1000] HEAT OF MIXING AND SOLUTION	ORTJ0961.011
State:	Two-component system, single-phase liquid or two-phase liquid-liquid (LL)	
Pure component 1, liquid		
Pure component 2, liquid		
Parameters:	[P1] T/K, Temperature	
Variables:	[X1] $x_1/-$, Mole fraction of component 1	
[Y1] $H^E/J\ mol^{-1}$, Molar excess enthalpy		
Method:	Direct low-pressure calorimetric measurement of H^E at variable x_1 and constant T	
Components:	1. <chem>CHCl3</chem> , Trichloromethane 2. <chem>C13H26O2</chem> , Methyl dodecanoate	
[P1] = .298150E+03		
[X1]	[Y1]	
.147000E+00 .268500E+00 .366400E+00 .441200E+00 .508300E+00 .566900E+00 .617100E+00 .658000E+00 .692700E+00 .736100E+00 .771800E+00 .808700E+00 .846500E+00 .878800E+00 .912100E+00 .944100E+00 .973800E+00	-.715100E+03 -.114490E+04 -.150260E+04 -.173130E+04 -.186370E+04 -.190120E+04 -.189320E+04 -.183810E+04 -.175930E+04 -.165320E+04 -.150770E+04 -.133190E+04 -.111520E+04 -.899700E+03 -.670200E+03 -.432000E+03 -.209400E+03	
		[Y1] .000E+00 -.500E+03 -.100E+04 -.150E+04 -.200E+04
		[X1] .50E+00

Property Code:	[HMSD1000] HEAT OF MIXING AND SOLUTION	ORTJ0961.012
State:	Two-component system, single-phase liquid or two-phase liquid-liquid (LL)	
Pure component 1, liquid		
Pure component 2, liquid		
Parameters:	[P1] T/K, Temperature	
Variables:	[X1] $x_1/-$, Mole fraction of component 1	
[Y1] $H^E/J\ mol^{-1}$, Molar excess enthalpy		
Method:	Direct low-pressure calorimetric measurement of H^E at variable x_1 and constant T	
Components:	1. <chem>CHCl3</chem> , Trichloromethane 2. <chem>C14H28O2</chem> , Methyl tridecanoate	
[P1] = .298150E+03		
[X1]	[Y1]	
.155200E+00 .277700E+00 .376300E+00 .451400E+00 .518100E+00 .573500E+00 .625700E+00 .667500E+00 .699700E+00 .759300E+00 .789400E+00 .824900E+00 .860700E+00 .896100E+00 .930100E+00 .960600E+00 .984600E+00	-.730100E+03 -.115260E+04 -.149970E+04 -.168840E+04 -.179020E+04 -.182790E+04 -.182060E+04 -.176550E+04 -.169060E+04 -.151420E+04 -.139060E+04 -.121180E+04 -.100030E+04 -.769100E+03 -.531000E+03 -.310400E+03 -.133700E+03	
		[Y1] .000E+00 -.500E+03 -.100E+04 -.150E+04 -.200E+04
		[X1] .50E+00

Property Code: [HMSD1000] HEAT OF MIXING AND SOLUTION

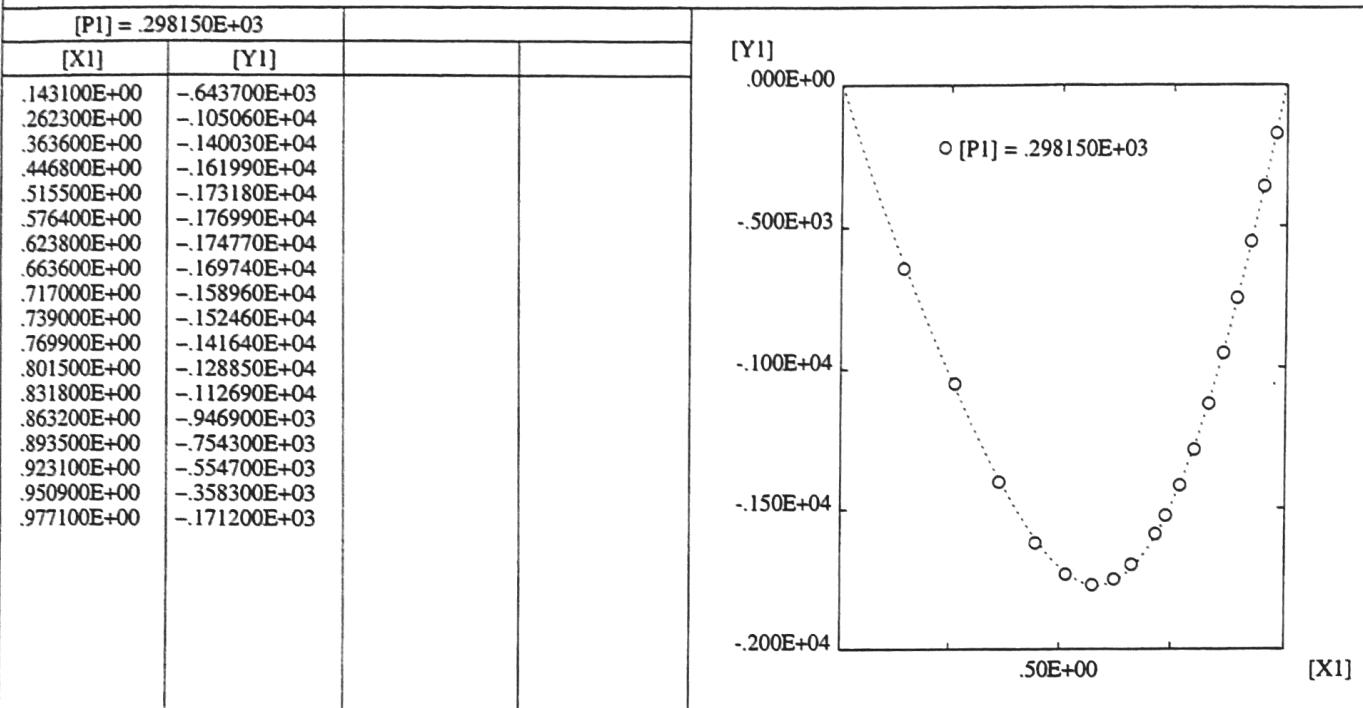
ORTJ0961.013

State: Two-component system, single-phase liquid or two-phase liquid-liquid (LL)

Pure component 1, liquid

Pure component 2, liquid

Parameters: [P1] T/K, Temperature

Variables: [X1] x_1 , Mole fraction of component 1[Y1] $H^E/J\text{ mol}^{-1}$, Molar excess enthalpyMethod: Direct low-pressure calorimetric measurement of H^E at variable x_1 and constant T Components: 1. CHCl_3 , Trichloromethane2. $\text{C}_{15}\text{H}_{30}\text{O}_2$, Methyl tetradecanoate

Property Code: [HMSD1000] HEAT OF MIXING AND SOLUTION

ORTJ0961.014

State: Two-component system, single-phase liquid or two-phase liquid-liquid (LL)

Pure component 1, liquid

Pure component 2, liquid

Parameters: [P1] T/K, Temperature

Variables: [X1] x_1 , Mole fraction of component 1[Y1] $H^E/J\text{ mol}^{-1}$, Molar excess enthalpyMethod: Direct low-pressure calorimetric measurement of H^E at variable x_1 and constant T Components: 1. CHCl_3 , Trichloromethane2. $\text{C}_{16}\text{H}_{32}\text{O}_2$, Methyl pentadecanoate