

Excess enthalpies of 1,1,2,2-tetrachloroethane + n-alkanes. Measurement and comparison with the DISQUAC model

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Enthalpy-of-mixing measurements are reported at 298.15 K for 1,1,2,2-tetrachloroethane + n-alkanes (pentane through heptadecane). All the mixtures are endothermic with molar excess enthalpies increasing regularly with the chain length of the n-alkane. The experimental data are in good agreement with the values predicted by the DISQUAC group contribution model for n-alkanes containing less than eight C atoms. The discrepancies increase with the chain length of the n-alkane up to 10 % (in the central range of composition) for heptadecane.

1. INTRODUCTION

In continuation of our systematic experimental studies on the excess molar enthalpies H^E of mixtures containing chloroalkanes (ORTJ0915), we have determined H^E of 1,1,2,2-tetrachloroethane + n-alkanes (pentane through heptadecane). H^E measurements have been published previously only for 1,1,2,2-tetrachloroethane + n-hexane (PARJ04870). These data, along with data on vapor-liquid equilibria (FARJ0950) and excess volumes (LINJ0950), will be used to test the applicability of various group-contribution models.

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

C₂H₂Cl₄, 1,1,2,2-Tetrachloroethane 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. $n(D, 298.15 \text{ K}) = 1.4918$; $\rho_i(298.15 \text{ K})/\text{kg m}^{-3} = 1588.37$.

C₅H₁₂, Pentane. Fluka AG (Buchs, Switzerland) 'puriss' grade material of stated GLC purity > 99.5 mole %, purified as above; $n(D, 298.15 \text{ K}) = 1.3547$; $\rho_i(298.15 \text{ K})/\text{kg m}^{-3} = 621.31$.

C₆H₁₄, Hexane. Fluka AG (Buchs, Switzerland) 'puriss' grade material of stated GLC purity > 99.5 mole %, purified as above; $n(D, 298.15 \text{ K}) = 1.3723$; $\rho_i(298.15 \text{ K})/\text{kg m}^{-3} = 654.71$.

C₇H₁₆, Heptane. Fluka AG (Buchs, Switzerland) 'puriss' grade material of stated GLC purity > 99.5 mole %, purified as above; $n(D, 298.15 \text{ K}) = 1.3851$; $\rho_i(298.15 \text{ K})/\text{kg m}^{-3} = 679.46$.

C₈H₁₈, Octane. Fluka AG (Buchs, Switzerland) 'puriss' grade material of stated GLC purity > 99.5 mole %, purified as above; $n(D, 298.15 \text{ K}) = 1.3951$; $\rho_i(298.15 \text{ K})/\text{kg m}^{-3} = 698.29$.

C₉H₂₀, Nonane. Fluka AG (Buchs, Switzerland) 'purum' grade material of stated GLC purity > 99.0 mole %, purified as above; $n(D, 298.15 \text{ K}) = 1.4033$; $\rho_i(298.15 \text{ K})/\text{kg m}^{-3} = 713.85$.

C₁₀H₂₂, Decane. Fluka AG (Buchs, Switzerland) 'purum' grade material of stated GLC purity > 99.0 mole %, purified as above; $n(D, 298.15 \text{ K}) = 1.4098$; $\rho_i(298.15 \text{ K})/\text{kg m}^{-3} = 726.13$.

C₁₁H₂₄, Undecane. Fluka AG (Buchs, Switzerland) 'purum' grade material of stated GLC purity > 98.0 mole %, purified as above; $n(D, 298.15 \text{ K}) = 1.4154$; $\rho_i(298.15 \text{ K})/\text{kg m}^{-3} = 736.80$.

C₁₂H₂₆, Dodecane. Fluka AG (Buchs, Switzerland) 'purum' grade material of stated GLC purity > 98.0 mole %, purified as above; $n(D, 298.15 \text{ K}) = 1.4196$; $\rho_i(298.15 \text{ K})/\text{kg m}^{-3} = 745.19$.

C₁₃H₂₈, Tridecane. Fluka AG (Buchs, Switzerland) 'puriss' grade material of stated GLC purity > 99.5 mole

%, purified as above; $n(D, 298.15 \text{ K}) = 1.4238$; $\rho_i(298.15 \text{ K})/\text{kg m}^{-3} = 752.79$.

C₁₄H₃₀, Tetradecane. Fluka AG (Buchs, Switzerland) 'puriss' grade material of stated GLC purity > 99.5 mole %, purified as above; $n(D, 298.15 \text{ K}) = 1.4271$; $\rho_i(298.15 \text{ K})/\text{kg m}^{-3} = 759.36$.

C₁₅H₃₂, Pentadecane. Fluka AG (Buchs, Switzerland) 'purum' grade material of stated GLC purity > 98 mole %, purified as above; $n(D, 298.15 \text{ K}) = 1.4298$; $\rho_i(298.15 \text{ K})/\text{kg m}^{-3} = 764.80$.

C₁₆H₃₄, Hexadecane. Fluka AG (Buchs, Switzerland) 'purum' grade material of stated GLC purity > 98 mole %, purified as above; $n(D, 298.15 \text{ K}) = 1.4325$; $\rho_i(298.15 \text{ K})/\text{kg m}^{-3} = 770.10$.

C₁₇H₃₆, Heptadecane. Fluka AG (Buchs, Switzerland) 'purum' grade material of stated GLC purity > 99 mole %, purified as above; $n(D, 298.15 \text{ K}) = 1.4347$; $\rho_i(298.15 \text{ K})/\text{kg m}^{-3} = 774.33$.

3. RESULTS

The direct experimental HE values are tabulated and graphed in the Appendix and saved on disk as Standard ELDATA Files ORTJ0953.001 through ORTJ0953.013.

The data were fitted to Eq. (1):

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

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

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

where N is the number of experimental values, are less than 10 J mol^{-1} (ca. 0.4 % at $x_1 = 0.5$).

4. DISCUSSION AND CONCLUSIONS

All the mixtures are endothermic with HE increasing regularly with the chain length of the n-alkane. Our results for 1,1,2,2-tetrachloroethane + n-hexane are in excellent agreement (ca 1 % at $x_1 = 0.5$) with the literature data (PARJ04870). The experimental

equimolar HE values for short-chain n-alkanes agree within better than 4 % with DISQUAC predictions, using previously published parameters (KEHH0880). It is interesting to note that these parameters were estimated from structure-property correlations taking into account the intramolecular proximity effect of the $-\text{CHCl}_2$ groups. Characteristic discrepancies (up to 10 %) are noted in mixtures with long-chain n-alkanes. These are usually attributed to the perturbation of the conformational order of long-chain n-alkanes when mixed with globular molecules such as 1,1,2,2-tetrachloroethane (PATD0760).

REFERENCES

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Placido, Jose [PLAJ0]

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Property Code: [HMSD0001] HEAT OF MIXING AND SOLUTION

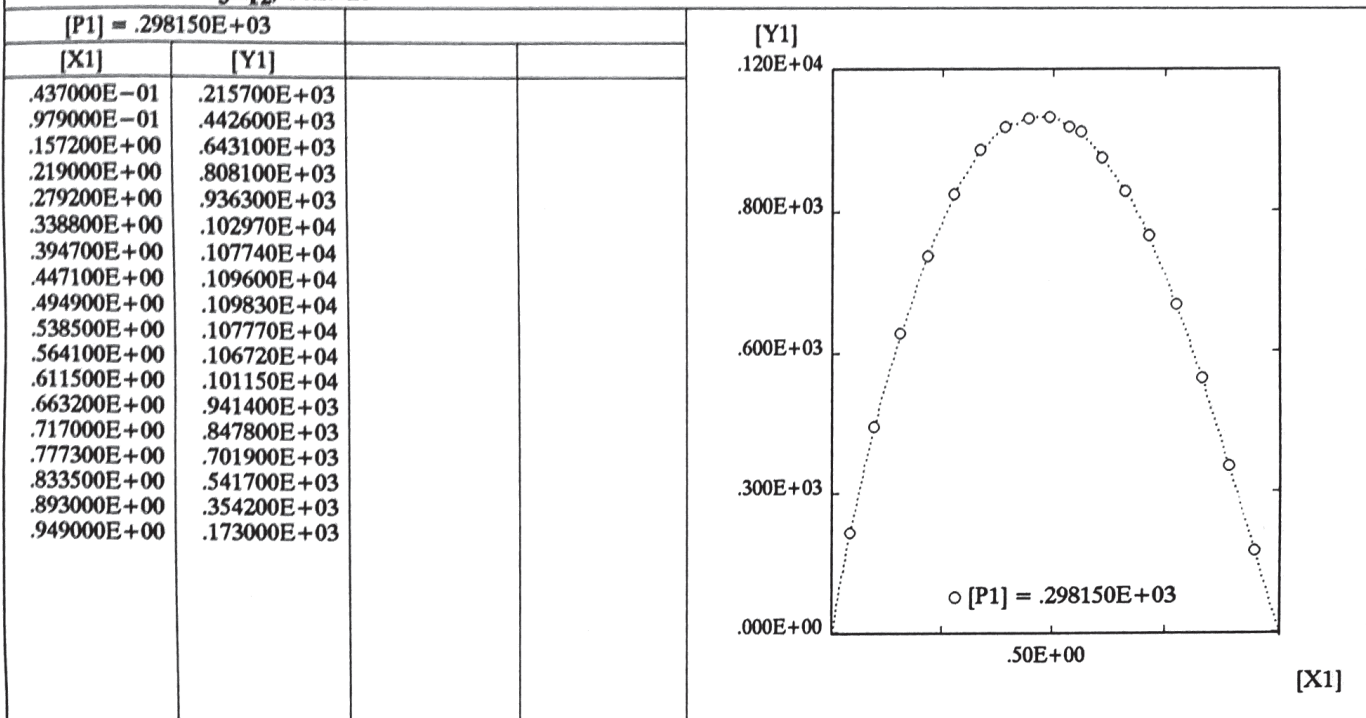
ORTJ0953.001

State: Two-component system, single-phase liquid

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 /Jmol⁻¹, Molar excess enthalpyMethod: Direct low-pressure calorimetric measurement of H^E at variable x_1 and constant T Components: 1. C₂H₂Cl₄, 1,1,2,2-Tetrachloroethane2. C₅H₁₂, Pentane

Property Code: [HMSD0001] HEAT OF MIXING AND SOLUTION

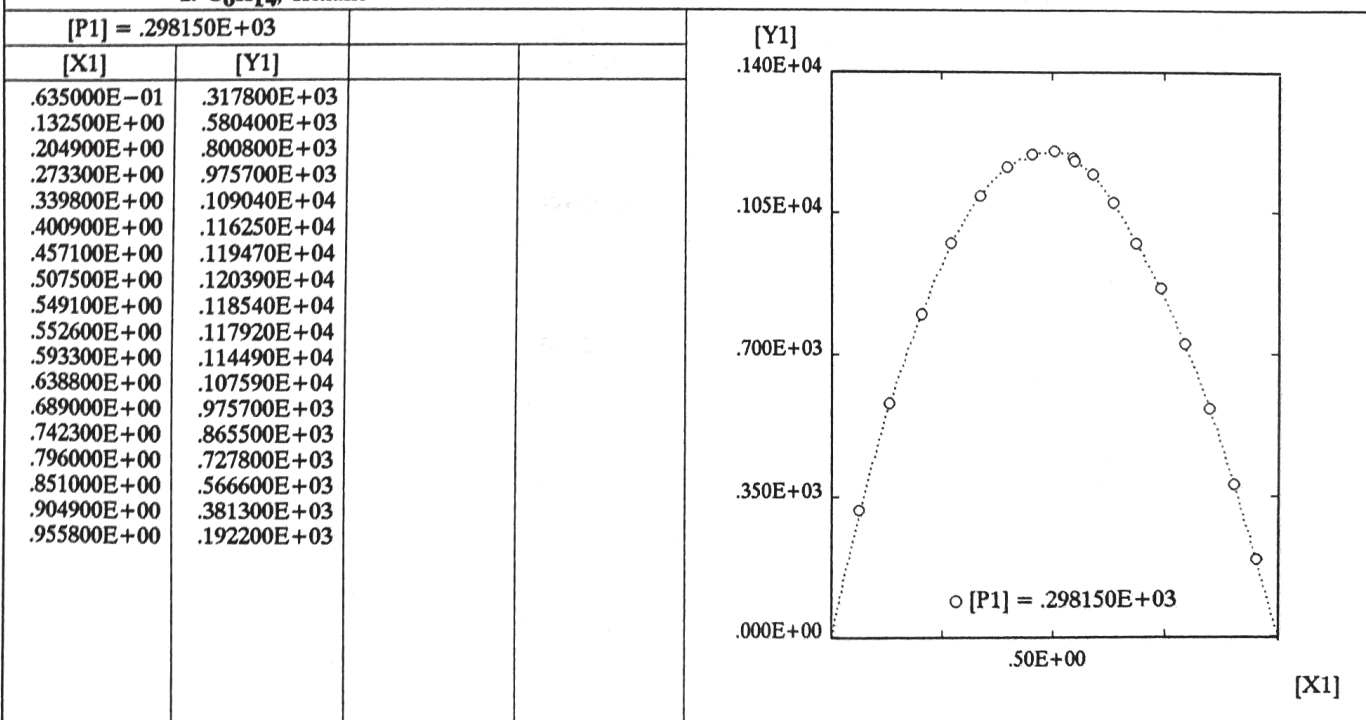
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State: Two-component system, single-phase liquid

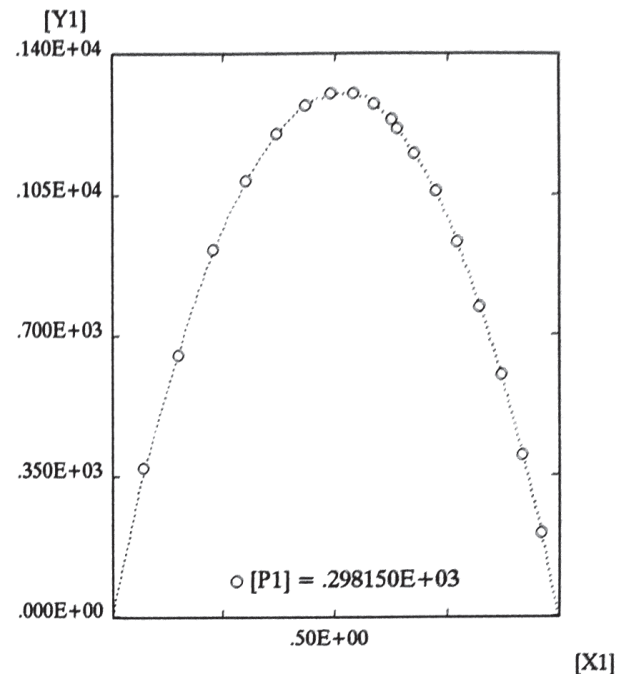
Pure component 1, liquid

Pure component 2, liquid

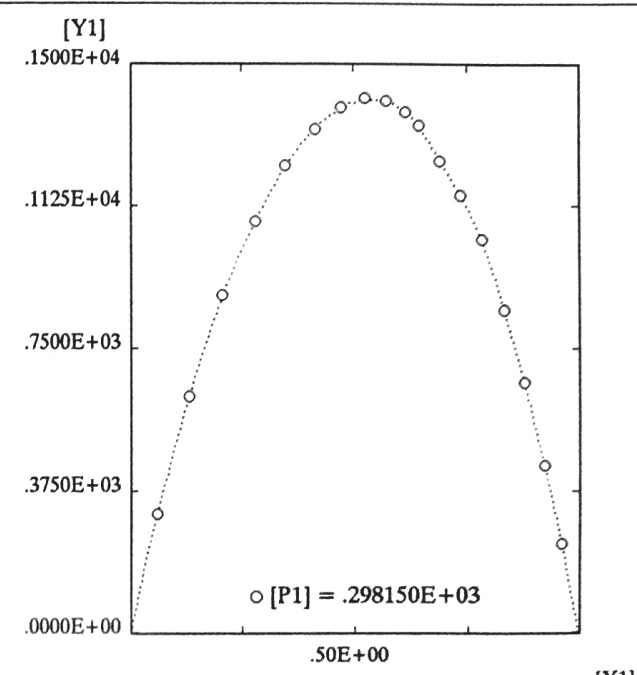
Parameters: [P1] T/K, Temperature

Variables: [X1] x_1 /%, Mole fraction of component 1[Y1] H^E /Jmol⁻¹, Molar excess enthalpyMethod: Direct low-pressure calorimetric measurement of H^E at variable x_1 and constant T Components: 1. C₂H₂Cl₄, 1,1,2,2-Tetrachloroethane2. C₆H₁₄, Hexane

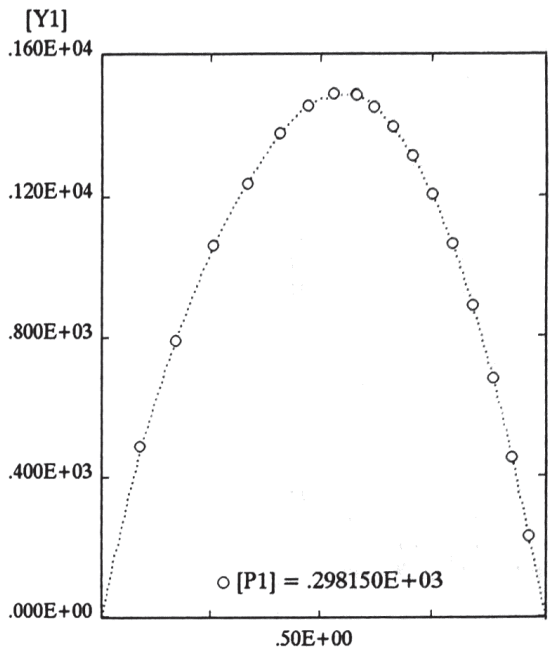
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State: Two-component system, single-phase liquid			
Pure component 1, liquid			
Pure component 2, liquid			
Parameters:	[P1] T/K, Temperature		
Variables:	[X1] x_1 /-, Mole fraction of component 1		
	[Y1] H^E /Jmol ⁻¹ , Molar excess enthalpy		
Method: Direct low-pressure calorimetric measurement of H^E at variable x_1 and constant T			
Components: 1. C ₂ H ₂ Cl ₄ , 1,1,2,2-Tetrachloroethane 2. C ₇ H ₁₆ , Heptane			
[P1] = .298150E+03			
[X1]	[Y1]		
.732000E-01	.370200E+03		
.152100E+00	.652700E+03		
.230600E+00	.913800E+03		
.304300E+00	.108630E+04		
.372800E+00	.120080E+04		
.436300E+00	.127100E+04		
.493400E+00	.130100E+04		
.543200E+00	.130070E+04		
.587800E+00	.127430E+04		
.627000E+00	.123650E+04		
.640200E+00	.121390E+04		
.678100E+00	.115210E+04		
.725800E+00	.106020E+04		
.772700E+00	.932000E+03		
.822300E+00	.768700E+03		
.872000E+00	.600400E+03		
.919200E+00	.400000E+03		
.963100E+00	.205900E+03		



Property Code: [HMSD0001] HEAT OF MIXING AND SOLUTION		ORTJ0953.004	
State: Two-component system, single-phase liquid			
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 /Jmol ⁻¹ , Molar excess enthalpy		
Method: Direct low-pressure calorimetric measurement of H^E at variable x_1 and constant T			
Components: 1. C ₂ H ₂ Cl ₄ , 1,1,2,2-Tetrachloroethane 2. C ₈ H ₁₈ , Octane			
[P1] = .298150E+03			
[X1]	[Y1]		
.618000E-01	.313300E+03		
.134200E+00	.625700E+03		
.208800E+00	.889300E+03		
.283200E+00	.108520E+04		
.351300E+00	.123120E+04		
.417400E+00	.132670E+04		
.476100E+00	.138510E+04		
.529300E+00	.140930E+04		
.576100E+00	.140160E+04		
.617500E+00	.137160E+04		
.648100E+00	.133660E+04		
.692300E+00	.124270E+04		
.738900E+00	.115250E+04		
.785800E+00	.103500E+04		
.833000E+00	.849800E+03		
.879400E+00	.661100E+03		
.923700E+00	.442900E+03		
.963200E+00	.233600E+03		



Property Code: [HMSD0001] HEAT OF MIXING AND SOLUTION		ORTJ0953.005	
State: Two-component system, single-phase liquid 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 /Jmol ⁻¹ , Molar excess enthalpy			
Method: Direct low-pressure calorimetric measurement of H^E at variable x_1 and constant T			
Components: 1. C ₂ H ₂ Cl ₄ , 1,1,2,2-Tetrachloroethane 2. C ₉ H ₂₀ , Nonane			
[P1] = .298150E+03			
[X1]	[Y1]		
.893000E-01	.488200E+03		
.172100E+00	.790700E+03		
.257900E+00	.106130E+04		
.336900E+00	.123620E+04		
.409800E+00	.137860E+04		
.473900E+00	.145580E+04		
.531600E+00	.148870E+04		
.581300E+00	.148640E+04		
.583500E+00	.148580E+04		
.623000E+00	.145190E+04		
.664200E+00	.139650E+04		
.707900E+00	.131510E+04		
.752300E+00	.120490E+04		
.797100E+00	.106350E+04		
.842900E+00	.886100E+03		
.887800E+00	.679800E+03		
.929500E+00	.453300E+03		
.967100E+00	.228100E+03		



[Y1]

.160E+04

.120E+04

.800E+03

.400E+03

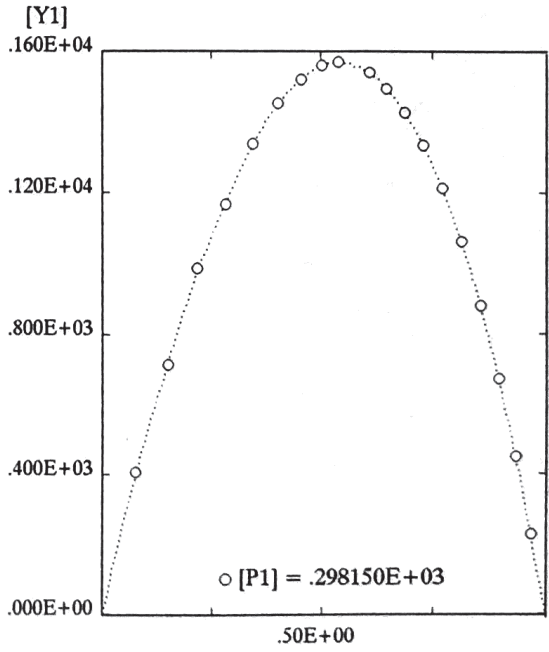
.000E+00

.50E+00

[X1]

○ [P1] = .298150E+03

Property Code: [HMSD0001] HEAT OF MIXING AND SOLUTION		ORTJ0953.006	
State: Two-component system, single-phase liquid 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 /Jmol ⁻¹ , Molar excess enthalpy			
Method: Direct low-pressure calorimetric measurement of H^E at variable x_1 and constant T			
Components: 1. C ₂ H ₂ Cl ₄ , 1,1,2,2-Tetrachloroethane 2. C ₁₀ H ₂₂ , Decane			
[P1] = .298150E+03			
[X1]	[Y1]		
.780000E-01	.407700E+03		
.152000E+00	.715600E+03		
.220200E+00	.985100E+03		
.285900E+00	.116740E+04		
.348100E+00	.133850E+04		
.405400E+00	.145230E+04		
.458200E+00	.152110E+04		
.505700E+00	.156000E+04		
.542700E+00	.157110E+04		
.613000E+00	.154120E+04		
.650900E+00	.149500E+04		
.690900E+00	.142770E+04		
.732300E+00	.133520E+04		
.774600E+00	.121290E+04		
.817000E+00	.106160E+04		
.859000E+00	.879900E+03		
.899200E+00	.673100E+03		
.937200E+00	.448800E+03		
.970600E+00	.227600E+03		



[Y1]

.160E+04

.120E+04

.800E+03

.400E+03

.000E+00

.50E+00

[X1]

○ [P1] = .298150E+03

Property Code: [HMSD0001] HEAT OF MIXING AND SOLUTION

ORTJ0953.007

State: Two-component system, single-phase liquid

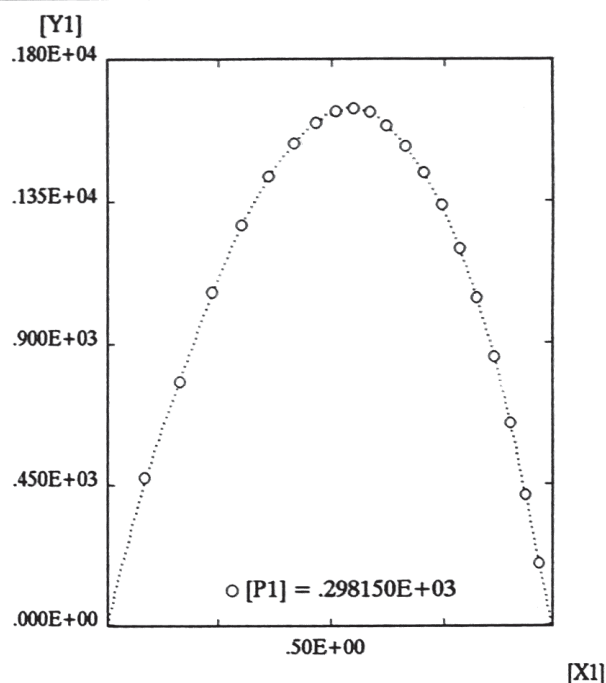
Pure component 1, liquid

Pure component 2, liquid

Parameters: [P1] T/K, Temperature

Variables: [X1] x_1 /-, Mole fraction of component 1[Y1] H^E /Jmol⁻¹, Molar excess enthalpyMethod: Direct low-pressure calorimetric measurement of H^E at variable x_1 and constant T Components: 1. C₂H₂Cl₄, 1,1,2,2-Tetrachloroethane
2. C₁₁H₂₄, Undecane

[P1] = .298150E+03	
[X1]	[Y1]
.870000E-01	.472800E+03
.166700E+00	.779800E+03
.239000E+00	.106310E+04
.304300E+00	.127550E+04
.363800E+00	.142980E+04
.418800E+00	.153610E+04
.467300E+00	.160160E+04
.511800E+00	.163740E+04
.550900E+00	.164510E+04
.586100E+00	.163560E+04
.622600E+00	.159120E+04
.665100E+00	.152680E+04
.705100E+00	.144170E+04
.745400E+00	.133730E+04
.786100E+00	.119890E+04
.825900E+00	.104220E+04
.865700E+00	.856700E+03
.904100E+00	.645300E+03
.939900E+00	.415700E+03
.972000E+00	.199400E+03



Property Code: [HMSD0001] HEAT OF MIXING AND SOLUTION

ORTJ0953.008

State: Two-component system, single-phase liquid

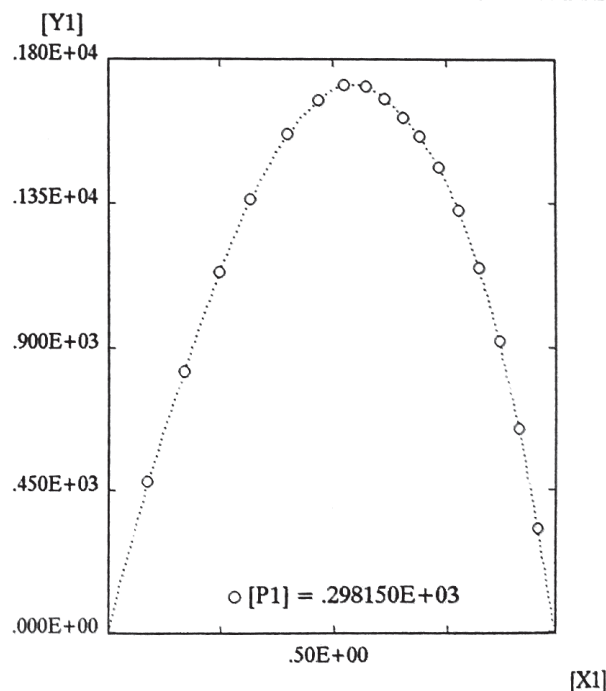
Pure component 1, liquid

Pure component 2, liquid

Parameters: [P1] T/K, Temperature

Variables: [X1] x_1 /-, Mole fraction of component 1[Y1] H^E /Jmol⁻¹, Molar excess enthalpyMethod: Direct low-pressure calorimetric measurement of H^E at variable x_1 and constant T Components: 1. C₂H₂Cl₄, 1,1,2,2-Tetrachloroethane
2. C₁₂H₂₆, Dodecane

[P1] = .298150E+03	
[X1]	[Y1]
.900000E-01	.478900E+03
.173000E+00	.827500E+03
.250300E+00	.113510E+04
.319200E+00	.136310E+04
.402000E+00	.156690E+04
.470500E+00	.167310E+04
.527700E+00	.172120E+04
.576200E+00	.171580E+04
.617100E+00	.167720E+04
.656800E+00	.161890E+04
.692600E+00	.156110E+04
.734500E+00	.146360E+04
.779100E+00	.132770E+04
.824800E+00	.114950E+04
.871200E+00	.922200E+03
.916800E+00	.645600E+03
.962400E+00	.328400E+03



Property Code: [HMSD0001] HEAT OF MIXING AND SOLUTION

ORTJ0953.009

State: Two-component system, single-phase liquid
 Pure component 1, liquid
 Pure component 2, liquid

Parameters: [P1] T/K, Temperature

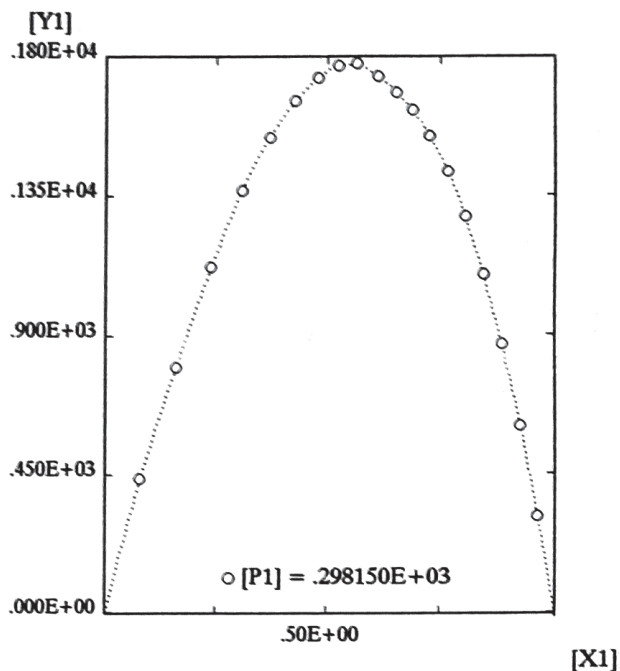
Variables: [X1] x_1 /-, Mole fraction of component 1

[Y1] H^E /Jmol⁻¹, Molar excess enthalpy

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

Components: 1. C₂H₂Cl₄, 1,1,2,2-Tetrachloroethane
 2. C₁₃H₂₈, Tridecane

[P1] = .298150E+03	
[X1]	[Y1]
.804000E-01	.436600E+03
.164000E+00	.797000E+03
.242000E+00	.112260E+04
.311700E+00	.136620E+04
.374000E+00	.153910E+04
.430600E+00	.165860E+04
.481800E+00	.173350E+04
.527200E+00	.177090E+04
.568000E+00	.177850E+04
.615000E+00	.173900E+04
.655100E+00	.168640E+04
.691400E+00	.162950E+04
.729600E+00	.154460E+04
.769400E+00	.142720E+04
.809300E+00	.128060E+04
.849500E+00	.109510E+04
.889900E+00	.868500E+03
.929600E+00	.603300E+03
.967000E+00	.314700E+03



Property Code: [HMSD0001] HEAT OF MIXING AND SOLUTION

ORTJ0953.010

State: Two-component system, single-phase liquid
 Pure component 1, liquid
 Pure component 2, liquid

Parameters: [P1] T/K, Temperature

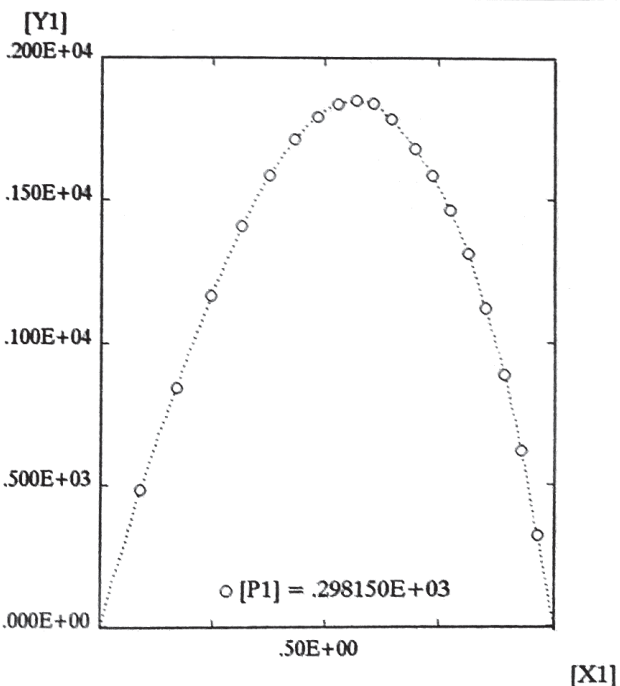
Variables: [X1] x_1 /-, Mole fraction of component 1

[Y1] H^E /Jmol⁻¹, Molar excess enthalpy

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

Components: 1. C₂H₂Cl₄, 1,1,2,2-Tetrachloroethane
 2. C₁₄H₃₀, Tetradecane

[P1] = .298150E+03	
[X1]	[Y1]
.899000E-01	.484400E+03
.170800E+00	.844900E+03
.247300E+00	.116560E+04
.316300E+00	.140910E+04
.378600E+00	.158690E+04
.435500E+00	.171380E+04
.486200E+00	.179480E+04
.532200E+00	.183820E+04
.572900E+00	.185020E+04
.609800E+00	.184010E+04
.649700E+00	.178520E+04
.701100E+00	.168240E+04
.738400E+00	.158540E+04
.776900E+00	.146430E+04
.816000E+00	.131050E+04
.854500E+00	.112240E+04
.894000E+00	.888900E+03
.931800E+00	.618300E+03
.967900E+00	.321400E+03



Property Code: [HM5D0001] HEAT OF MIXING AND SOLUTION

ORTJ0953.011

State: Two-component system, single-phase liquid
 Pure component 1, liquid
 Pure component 2, liquid

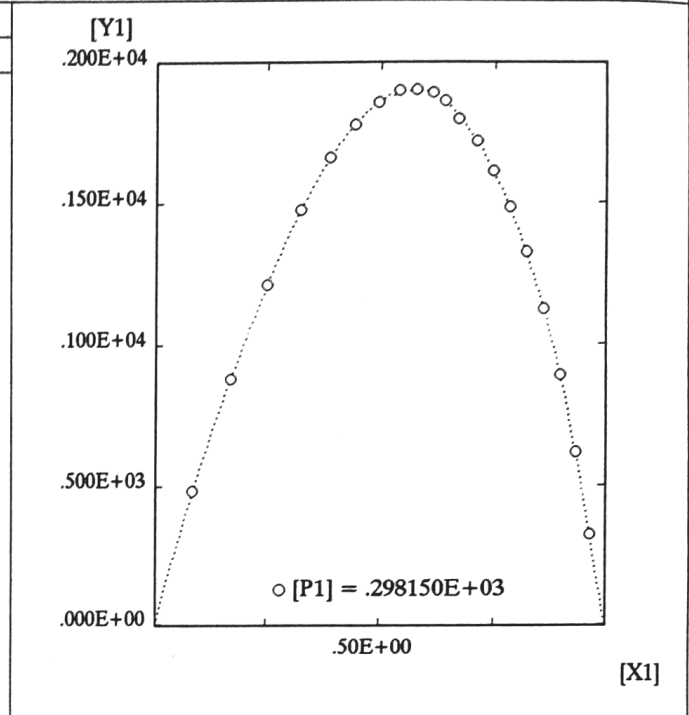
Parameters: [P1] T/K, Temperature

Variables: [X1] x_1 /, Mole fraction of component 1
 [Y1] H^E /Jmol⁻¹, Molar excess enthalpy

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

Components: 1. C₂H₂Cl₄, 1,1,2,2-Tetrachloroethane
 2. C₁₅H₃₂, Pentadecane

[P1] = .298150E+03	
[X1]	[Y1]
.874000E-01	.482700E+03
.173300E+00	.879700E+03
.254600E+00	.121470E+04
.327200E+00	.147550E+04
.391700E+00	.166060E+04
.445600E+00	.177790E+04
.497300E+00	.185960E+04
.543100E+00	.190030E+04
.580100E+00	.190360E+04
.615900E+00	.189410E+04
.643100E+00	.186420E+04
.672700E+00	.179980E+04
.713700E+00	.171800E+04
.749900E+00	.161120E+04
.787400E+00	.148430E+04
.824900E+00	.132510E+04
.863500E+00	.112470E+04
.900500E+00	.887900E+03
.936200E+00	.616300E+03
.969700E+00	.323900E+03



Property Code: [HM5D0001] HEAT OF MIXING AND SOLUTION

ORTJ0953.012

State: Two-component system, single-phase liquid
 Pure component 1, liquid
 Pure component 2, liquid

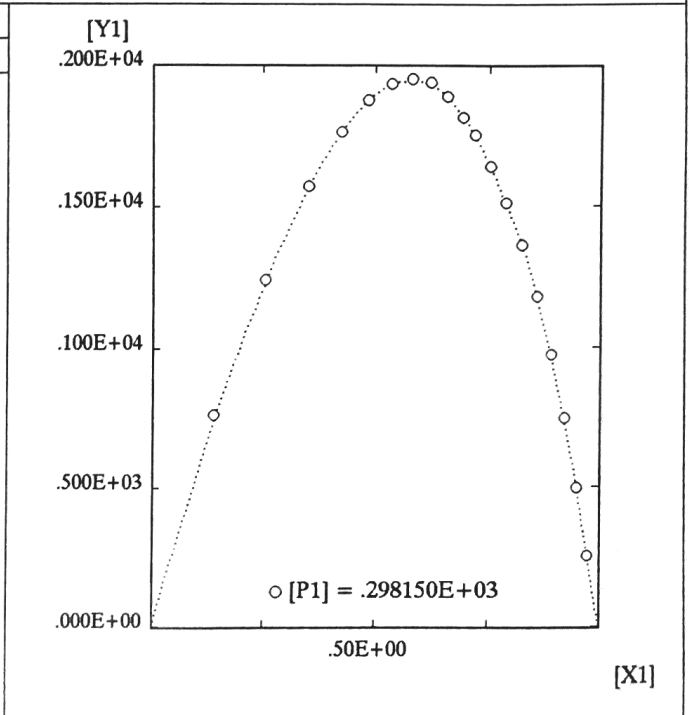
Parameters: [P1] T/K, Temperature

Variables: [X1] x_1 /, Mole fraction of component 1
 [Y1] H^E /Jmol⁻¹, Molar excess enthalpy

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

Components: 1. C₂H₂Cl₄, 1,1,2,2-Tetrachloroethane
 2. C₁₆H₃₄, Hexadecane

[P1] = .298150E+03	
[X1]	[Y1]
.141200E+00	.762300E+03
.257600E+00	.124280E+04
.354000E+00	.157430E+04
.427200E+00	.176670E+04
.486400E+00	.187760E+04
.537700E+00	.193500E+04
.583000E+00	.195470E+04
.623700E+00	.194010E+04
.659400E+00	.189030E+04
.694400E+00	.181510E+04
.720800E+00	.175420E+04
.755500E+00	.164130E+04
.790600E+00	.151150E+04
.825600E+00	.136190E+04
.859900E+00	.118090E+04
.892700E+00	.973800E+03
.923600E+00	.744400E+03
.952500E+00	.496800E+03
.977600E+00	.253200E+03



Property Code: [HMSD0001] HEAT OF MIXING AND SOLUTION

ORTJ0953.013

State: Two-component system, single-phase liquid
 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 /Jmol⁻¹, Molar excess enthalpy

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

Components: 1. C₂H₂Cl₄, 1,1,2,2-Tetrachloroethane
 2. C₁₇H₃₆, Heptadecane

[P1] = .298150E+03

[X1]	[Y1]
.141800E+00	.757600E+03
.251300E+00	.122810E+04
.335600E+00	.154560E+04
.402000E+00	.174590E+04
.461000E+00	.188040E+04
.509500E+00	.195340E+04
.553900E+00	.199360E+04
.593300E+00	.200310E+04
.644800E+00	.197420E+04
.668900E+00	.191470E+04
.699400E+00	.185430E+04
.734600E+00	.175200E+04
.769100E+00	.165870E+04
.804600E+00	.152130E+04
.840100E+00	.135260E+04
.875400E+00	.114630E+04
.909400E+00	.905200E+03
.942200E+00	.625900E+03
.972800E+00	.327700E+03

