

Excess enthalpies of 12 binary liquid mixtures of α,ω -dichloroalkanes (C₂, C₄, C₆) + benzene, toluene, ethylbenzene, or butylbenzene at 298.15 K

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(Received in final form February 3, 1997)

Enthalpy-of-mixing H^E measurements are reported at 298.15 K for 12 binary liquid mixtures containing 1,2-dichloroethane, 1,4-dichlorobutane, or 1,6-dichlorohexane + benzene, + toluene, + ethylbenzene, or + butylbenzene at 298.15 K over the entire range of composition. The H^E curves represented as a function of the mole fraction of dichloroalkane are asymmetrical and may be positive, or negative, or S-shaped. The H^E values are always smaller at low concentrations of the dichloroalkane. They decrease from benzene to toluene and then increase with the number of C-atoms of the n-alkylbenzene. For a given n-alkylbenzene, H^E decreases with the number of C-atoms of the dichloroalkane. H^E is the result of an exothermic contribution, due to dipole-induced dipole ($n\pi$) interactions between the polar Cl-groups and the π -electrons of the aromatic hydrocarbon, and an endothermic contribution mainly due to dipole-dipole interactions between the polar Cl-groups. The more negative H^E with toluene, compared to benzene, is due to the inductive effect of the methyl group in toluene. The endothermic contribution decreases with the number of C-atoms in the dichloroalkane, but increases with the number of C-atoms in the n-alkylbenzene.

1. INTRODUCTION

In continuation of our systematic experimental studies on the excess thermodynamic properties of α,ω -dichloroalkane + n-alkane mixtures [BLAA0930; ORTJ0930] we have determined the excess molar enthalpies, H^E , of 12 systems containing an α,ω -dichloroalkane (1,2-dichloroethane, 1,4-dichlorobutane, or 1,6-dichlorohexane) + an aromatic hydrocarbon (benzene, toluene, ethylbenzene, or butylbenzene) at 298.15 K. Many H^E data have been published in the literature for 1,2-dichloroethane + benzene, at temperatures from 288.15 to 318.15 K, and for 1,2-dichloroethane + toluene, at temperatures from 298.15 to 308.15 K [AMAK0580; BANI0830; BAUE1150; BROI0552; CHEG0520; KIRV2370; KORA3530; MAHB0771; NIGR0801; OTTJ2660; PATH0790;

RUIL0550; SIEL0511; SUNS0760; TSCH0485]. We found only a few data sources for the other systems: 1,2-dichloroethane + n-alkylbenzenes (methyl through octyl) at (295 \pm 1) K [TSCH0485], 1,3-dichloropropane and 1,4-dichlorobutane + benzene at 303.15 K [ROYF1800], and 1,3-dichloropropane, 1,4-dichlorobutane, 1,5-dichloropentane, or 1,6-dichlorohexane + benzene at 298.15 K [GROJ0734].

1-Chloroalkane + benzene mixtures have been carefully examined in terms of the DISQUAC group-contribution model [GARI0890]. Several similar studies on α,ω -dichloroalkane + n-alkane mixtures [HAHG0860; POLC0800; KEHH0880] revealed the occurrence of the Cl-Cl 'proximity effect' in α,ω -dichloroalkanes. The same effect appears in α,ω -dichloroalkane + CCl₄ mixtures [KEHH0881] and we expect to observe it in α,ω -dichloroalkane + aromatic hydrocarbons. A small H^E database of 1-chloroalkane or α,ω -dichloroalkane + aromatic hydrocarbons has been analyzed using the classical quasi-chemical lattice theory in terms of group surface interactions [GROJ0733; GROJ0734]. The H^E database revised and completed in the present investigation, along with data on vapor-liquid equilibria, and data for mixtures containing other α,ω -dihaloalkane and aromatic hydrocarbons, will be used to test the applicability of DISQUAC and other group-contribution models to α,ω -dichloroalkane + aromatic hydrocarbon mixtures.

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 (ITS-90) was maintained constant at (298.15 \pm 0.02) K. 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

C₂H₄Cl₂, 1,2-Dichloroethane. (Ethylene dichloride). Fluka AG (Buchs, Switzerland) 'puriss' grade material of stated GLC purity > 99.5 %, was degassed ultrasonically, dried over molecular sieves Type 3A (reference 69828, from Fluka), and used without further purification. $n(D, 298.15 \text{ K}) = 1.4422$ (1.4421 [RIDJ0860]); $\rho(298.15 \text{ K})/\text{kg m}^{-3} = 1245.38$ (1246.37 [RIDJ0860]).

C₄H₈Cl₂, 1,4-Dichlorobutane. Aldrich Chem. Co., Inc. (Milwaukee, WI, USA) material of stated purity > 99 %, was degassed ultrasonically, dried over molecular sieves Type 3A (reference 69828, from Fluka), and used without further purification. $n(D, 298.15 \text{ K}) = 1.4522$ (1.4522 [TRC00640]); $\rho(298.15 \text{ K})/\text{kg m}^{-3} = 1133.06$ (1135.3 [TRC00640]).

C₆H₆, Benzene. Aldrich Chem. Co., Inc. (Milwaukee, WI, USA) material of stated purity > 99.8 %, was degassed ultrasonically, dried over molecular sieves Type 3A (reference 69828, from Fluka), and used without further purification. $n(D, 298.15 \text{ K}) = 1.4980$ (1.49792 [RIDJ0860]); $\rho(298.15 \text{ K})/\text{kg m}^{-3} = 873.51$ (873.60 [RIDJ0860]).

C₆H₁₂Cl₂, 1,6-Dichlorohexane. Aldrich Chem. Co., Inc. (Milwaukee, WI, USA) material of stated purity > 99 %, was degassed ultrasonically, dried over molecular sieves Type 3A (reference 69828, from Fluka), and used without further purification. $n(D, 298.15 \text{ K}) = 1.4545$; $\rho(298.15 \text{ K})/\text{kg m}^{-3} = 1067.59$.

C₇H₈, Toluene. Aldrich Chem. Co., Inc. (Milwaukee, WI, USA) material of stated purity > 99.8 %, was degassed ultrasonically, dried over molecular sieves Type 3A (reference 69828, from Fluka), and used without further purification. $n(D, 298.15 \text{ K}) = 1.4940$ (1.49413 [RIDJ0860]); $\rho(298.15 \text{ K})/\text{kg m}^{-3} = 862.15$ (862.19 [RIDJ0860]).

C₈H₁₀, Ethylbenzene. Aldrich Chem. Co., Inc. (Milwaukee, WI, USA) material of stated purity > 99.8 %, was degassed ultrasonically, dried over molecular sieves Type 3A (reference 69828, from Fluka), and used without further purification. $n(D, 298.15 \text{ K}) = 1.4928$ (1.49320 [RIDJ0860]); $\rho(298.15 \text{ K})/\text{kg m}^{-3} = 862.60$ (862.53 [RIDJ0860]).

C₁₀H₁₄, Butylbenzene. Aldrich Chem. Co., Inc. (Milwaukee, WI, USA) material of stated purity > 99 %, was degassed ultrasonically, dried over molecular sieves Type 3A (reference 69828, from Fluka), and used without further purification. $n(D, 298.15 \text{ K}) = 1.4875$ (1.48742 [RIDJ0860]); $\rho(298.15 \text{ K})/\text{kg m}^{-3} = 856.17$ (856.07 [RIDJ0860]).

3. RESULTS

The direct experimental H^E values are tabulated and graphed in the Appendix and saved on disk as Standard ELDATA Files **ORTJ0962.001** through **ORTJ0962.012**.

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 8 J mol^{-1} .

4. DISCUSSION AND CONCLUSIONS

The H^E values at 298.15 K reported in the literature for 1,2-dichloroethane + benzene are in good agreement with our measurements. However, considerable discrepancies exist between our measurements and the data obtained by other authors in the case of 1,2-dichloroethane + toluene and other α,ω -dichloroalkane + n-alkylbenzene mixtures. The largest difference, 300 J mol^{-1} at equimolar composition and 298.15 K, was found in the case of 1,6-dichlorohexane + benzene [GROJ0734].

Our H^E results change regularly with the molecular structure of the components. When represented as a function of the mole fraction of the dichloroalkane, the H^E curves are asymmetrical and may be positive, or negative, or S-shaped. The H^E values are always smaller at low concentrations of the dichloroalkane. They decrease from benzene to toluene and then increase with the number of C-atoms of the n-alkylbenzene. For a given n-alkylbenzene, H^E decreases with the number of C-atoms of the dichloroalkane [ORTJ0963].

H^E is the result of an exothermic contribution, due to dipole-induced dipole ($n-\pi$) interactions between the polar Cl-groups and the π -electrons of the aromatic hydrocarbon, and an endothermic contribution mainly due to dipole-dipole interactions between the polar Cl-groups. The more negative H^E with toluene, compared to benzene, is due to the inductive effect of the methyl group in toluene. The endothermic contribution decreases with the number of C-atoms in the dichloroalkane, but increases with the number of C-atoms in the n-alkylbenzene.

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ORTJ0962.001

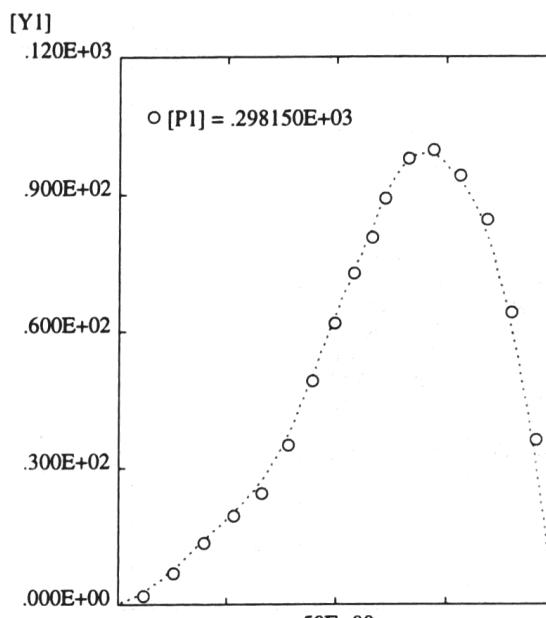
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Pure component 1, liquid
Pure component 2, liquid**Parameters:** [P1] T/K, Temperature
Variables: [X1] x_1 , Mole fraction of component 1[Y1] $H^E/J \text{ mol}^{-1}$, Molar excess enthalpy**Method:** Direct low-pressure calorimetric measurement of H^E at variable x_1 and constant T **Components:** 1. $\text{C}_2\text{H}_4\text{Cl}_2$, 1,2-Dichloroethane
2. C_6H_6 , Benzene

[P1] = .298150E+03

[X1]

[Y1]

.609000E-01	.193200E+01
.128800E+00	.676200E+01
.199100E+00	.133630E+02
.267700E+00	.194810E+02
.332400E+00	.243110E+02
.392500E+00	.350980E+02
.448300E+00	.491050E+02
.498400E+00	.616630E+02
.541500E+00	.726110E+02
.581500E+00	.805000E+02
.611800E+00	.890330E+02
.664800E+00	.977270E+02
.721400E+00	.994980E+02
.781500E+00	.940240E+02
.842400E+00	.842030E+02
.899200E+00	.637560E+02
.954800E+00	.359030E+02



[X1]

ORTJ0962.002

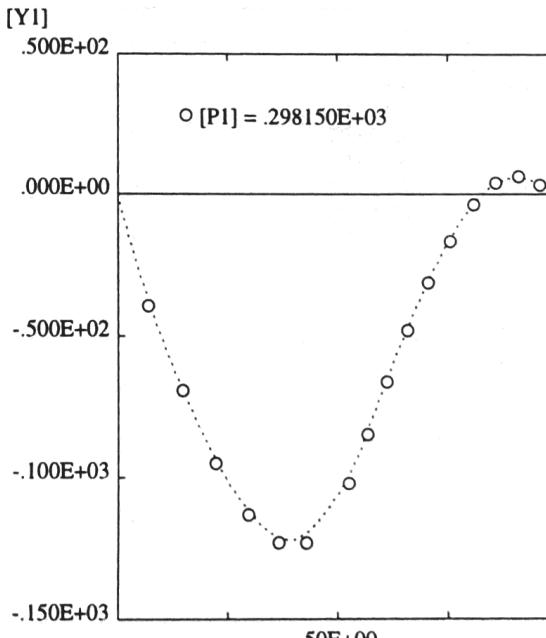
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Pure component 1, liquid
Pure component 2, liquid**Parameters:** [P1] T/K, Temperature**Variables:** [X1] x_1 , Mole fraction of component 1
[Y1] $H^E/J \text{ mol}^{-1}$, Molar excess enthalpy**Method:** Direct low-pressure calorimetric measurement of H^E at variable x_1 and constant T **Components:** 1. $\text{C}_2\text{H}_4\text{Cl}_2$, 1,2-Dichloroethane
2. C_7H_8 , Toluene

[P1] = .298150E+03

[X1]

[Y1]

.698000E-01	-.394000E+02
.146900E+00	-.692000E+02
.223000E+00	-.949000E+02
.297300E+00	-.113200E+03
.366400E+00	-.122900E+03
.428400E+00	-.123200E+03
.526000E+00	-.102100E+03
.567700E+00	-.848000E+02
.611700E+00	-.664000E+02
.659300E+00	-.481000E+02
.706000E+00	-.315000E+02
.755400E+00	-.166000E+02
.809100E+00	-.360000E+01
.858300E+00	.420000E+01
.910800E+00	.630000E+01
.958500E+00	.330000E+01



[X1]

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Property Code:	[HMSD1000] HEAT OF MIXING AND SOLUTION	ORTJ0962.003
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	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. C ₂ H ₄ Cl ₂ , 1,2-Dichloroethane 2. C ₈ H ₁₀ , Ethylbenzene	
[P1] = .298150E+03		
[X1]	[Y1]	
.844000E-01	-.117000E+02	
.170700E+00	-.143000E+02	
.256400E+00	-.184000E+02	
.337600E+00	-.103000E+02	
.412100E+00	.490000E+01	
.474700E+00	.205000E+02	
.532700E+00	.347000E+02	
.583000E+00	.461000E+02	
.603200E+00	.538000E+02	
.642300E+00	.633000E+02	
.687200E+00	.721000E+02	
.734200E+00	.764000E+02	
.783200E+00	.767000E+02	
.832000E+00	.725000E+02	
.877100E+00	.636000E+02	
.922000E+00	.487000E+02	
.964100E+00	.256000E+02	

The graph plots Molar excess enthalpy [Y1] on the y-axis against mole fraction [X1] on the x-axis. The y-axis ranges from -300E+02 to 900E+02. The x-axis has a tick mark at 50E+00. Data points are represented by open circles connected by a dotted line. A solid horizontal line is drawn at Y1 = 0.000E+00.

[X1]	[Y1]
0.000000E+00	0.000E+00
0.100000E+00	-0.117E+02
0.200000E+00	-0.143E+02
0.300000E+00	-0.184E+02
0.400000E+00	0.490E+01
0.450000E+00	0.205E+02
0.500000E+00	0.347E+02
0.550000E+00	0.461E+02
0.600000E+00	0.538E+02
0.650000E+00	0.633E+02
0.700000E+00	0.721E+02
0.750000E+00	0.764E+02
0.800000E+00	0.767E+02
0.850000E+00	0.725E+02
0.900000E+00	0.636E+02
0.950000E+00	0.487E+02
1.000000E+00	0.256E+02

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Property Code: [HMSD1000] HEAT OF MIXING AND SOLUTION**ORTJ0962.005****State:** Two-component system, single-phase liquid or two-phase liquid-liquid (LL)

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Pure component 2, liquid

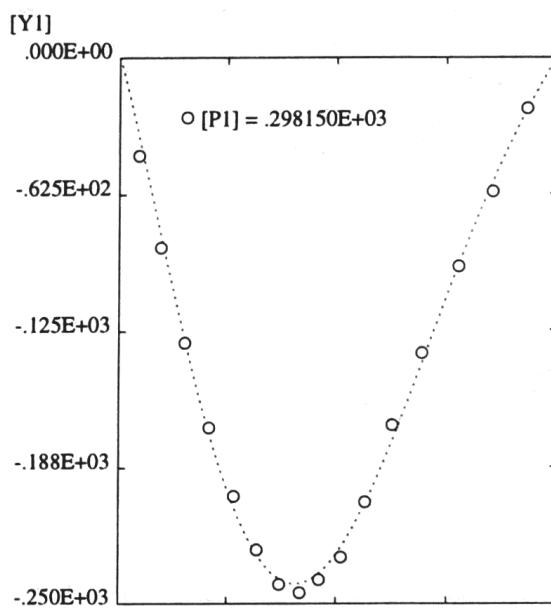
Parameters: [P1] T/K, Temperature**Variables:** [X1] x_1 , Mole fraction of component 1[Y1] $H^E/J \text{ mol}^{-1}$, Molar excess enthalpy**Method:** Direct low-pressure calorimetric measurement of H^E at variable x_1 and constant T **Components:** 1. $\text{C}_4\text{H}_8\text{Cl}_2$, 1,4-Dichlorobutane
2. C_6H_6 , Benzene

[P1] = .298150E+03

[X1]

[Y1]

.456000E-01	-.444000E+02
.976000E-01	-.867000E+02
.154400E+00	-.130300E+03
.210900E+00	-.168900E+03
.267200E+00	-.200400E+03
.320800E+00	-.225200E+03
.372500E+00	-.241200E+03
.419000E+00	-.244900E+03
.463000E+00	-.238800E+03
.512600E+00	-.228600E+03
.568700E+00	-.203400E+03
.629200E+00	-.168000E+03
.695700E+00	-.135100E+03
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.855900E+00	-.613000E+02
.933400E+00	-.234000E+02



[X1]

Property Code: [HMSD1000] HEAT OF MIXING AND SOLUTION**ORTJ0962.006****State:** Two-component system, single-phase liquid or two-phase liquid-liquid (LL)

Pure component 1, liquid

Pure component 2, liquid

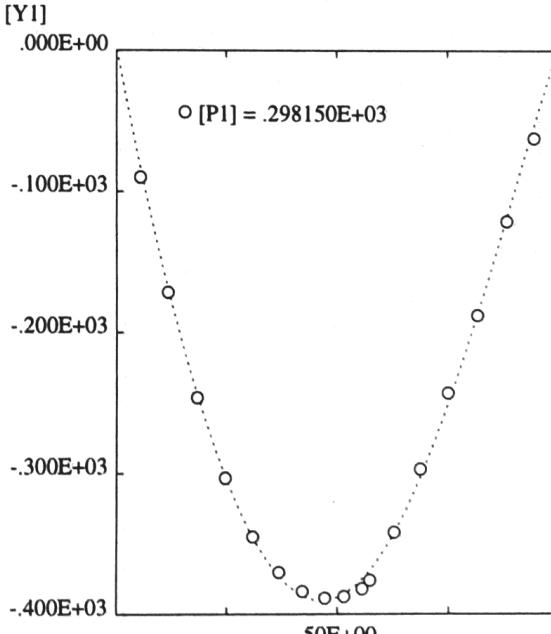
Parameters: [P1] T/K, Temperature**Variables:** [X1] x_1 , Mole fraction of component 1[Y1] $H^E/J \text{ mol}^{-1}$, Molar excess enthalpy**Method:** Direct low-pressure calorimetric measurement of H^E at variable x_1 and constant T **Components:** 1. $\text{C}_4\text{H}_8\text{Cl}_2$, 1,4-Dichlorobutane
2. C_7H_8 , Toluene

[P1] = .298150E+03

[X1]

[Y1]

.557000E-01	-.899000E+02
.118000E+00	-.171100E+03
.184100E+00	-.246400E+03
.247700E+00	-.303400E+03
.310600E+00	-.344600E+03
.369000E+00	-.370300E+03
.422500E+00	-.383600E+03
.471900E+00	-.388500E+03
.517100E+00	-.387300E+03
.556900E+00	-.381700E+03
.574900E+00	-.375700E+03
.628800E+00	-.341700E+03
.687500E+00	-.297900E+03
.750800E+00	-.243700E+03
.816400E+00	-.188400E+03
.881600E+00	-.121300E+03
.944200E+00	-.624000E+02

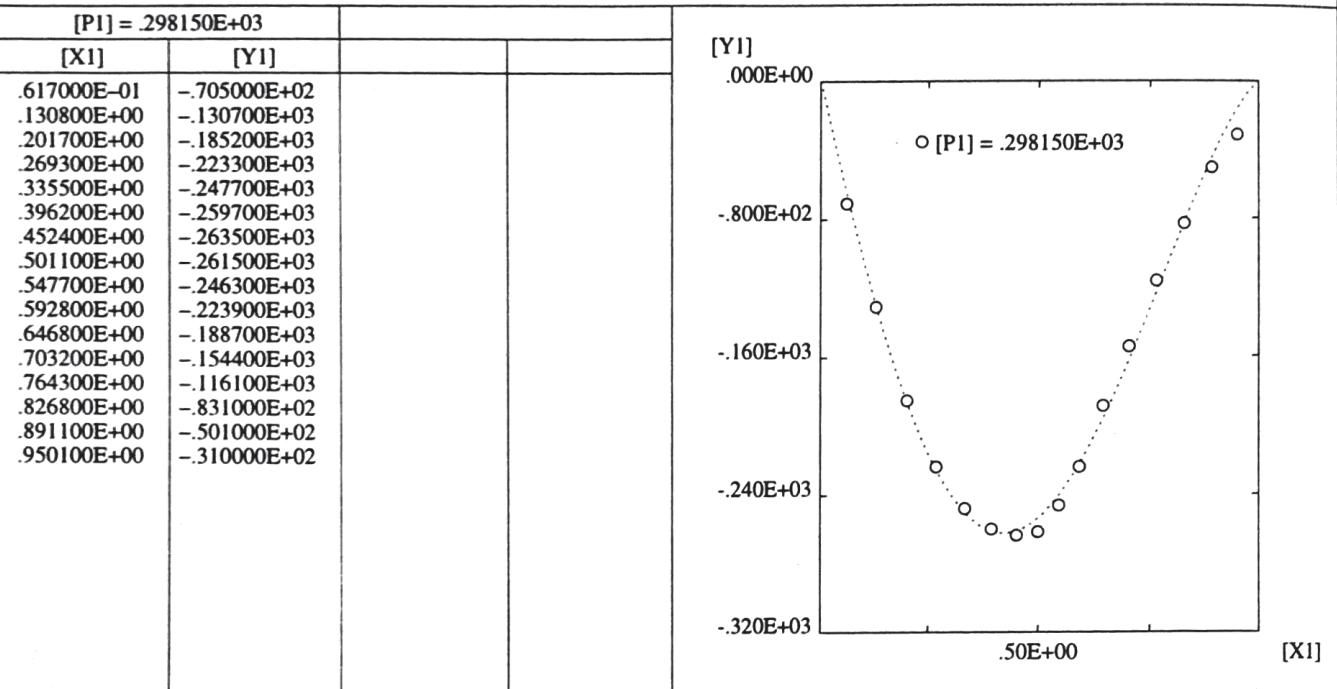


[X1]

ORTJ0962.007

Property Code:	[HMSD1000] HEAT OF MIXING AND SOLUTION
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 enthalpy
Method:	Direct low-pressure calorimetric measurement of H^E at variable x_1 and constant T

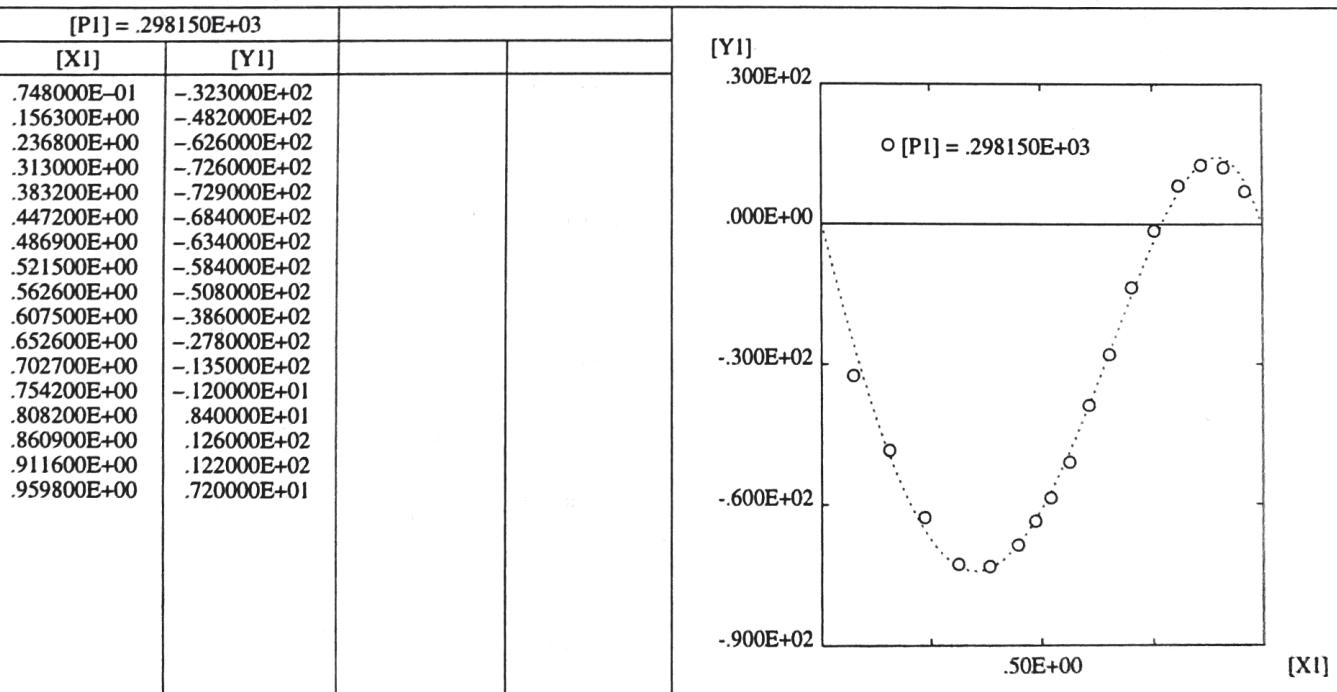
Components: 1. $\text{C}_4\text{H}_8\text{Cl}_2$, 1,4-Dichlorobutane
2. C_8H_{10} , Ethylbenzene



ORTJ0962.008

Property Code:	[HMSD1000] HEAT OF MIXING AND SOLUTION
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 enthalpy
Method:	Direct low-pressure calorimetric measurement of H^E at variable x_1 and constant T

Components: 1. $\text{C}_4\text{H}_8\text{Cl}_2$, 1,4-Dichlorobutane
2. $\text{C}_{10}\text{H}_{14}$, Butylbenzene

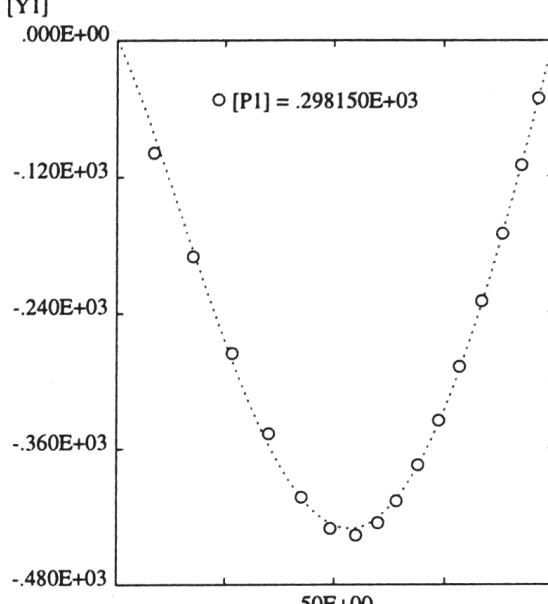


Property Code: [HMSD1000] HEAT OF MIXING AND SOLUTION**State:** Two-component system, single-phase liquid or two-phase liquid-liquid (LL)
Pure component 1, liquid
Pure component 2, liquid**ORTJ0962.009****Parameters:** [P1] T/K, Temperature**Variables:** [X1] x_1 /-, Mole fraction of component 1
[Y1] 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. C₆H₆, Benzene
2. C₆H₁₂Cl₂, 1,6-Dichlorohexane

[P1] = .298150E+03

[X1]

[X1]	[Y1]
.860000E-01	-988000E+02
.177000E+00	-190000E+03
.268600E+00	-275300E+03
.351600E+00	-346600E+03
.427000E+00	-402900E+03
.493400E+00	-430300E+03
.551200E+00	-436500E+03
.600200E+00	-425700E+03
.643300E+00	-406700E+03
.691300E+00	-374700E+03
.739000E+00	-335000E+03
.785400E+00	-287700E+03
.835100E+00	-229800E+03
.882600E+00	-170800E+03
.926100E+00	-109800E+03
.965200E+00	-515000E+02



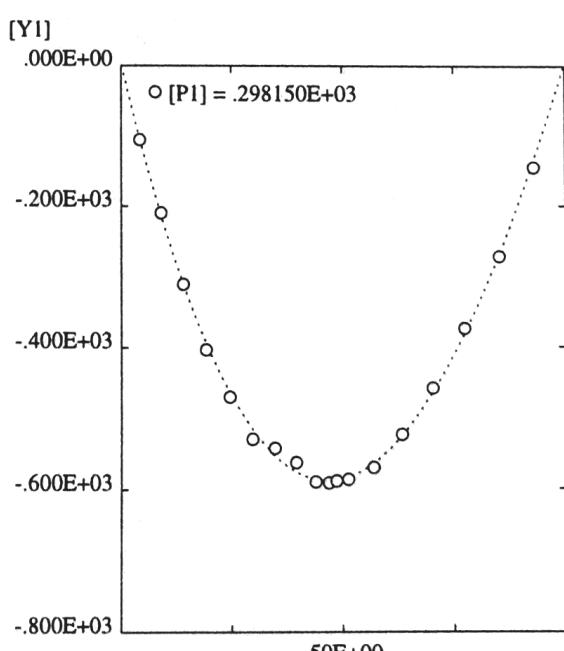
[X1]

Property Code: [HMSD1000] HEAT OF MIXING AND SOLUTION**State:** Two-component system, single-phase liquid or two-phase liquid-liquid (LL)
Pure component 1, liquid
Pure component 2, liquid**ORTJ0962.010****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. C₆H₁₂Cl₂, 1,6-Dichlorohexane
2. C₇H₈, Toluene

[P1] = .298150E+03

[X1]

[X1]	[Y1]
.432000E-01	-105500E+03
.910000E-01	-208500E+03
.140800E+00	-310000E+03
.193400E+00	-402700E+03
.246400E+00	-469900E+03
.298100E+00	-528400E+03
.348400E+00	-541100E+03
.396200E+00	-562000E+03
.440400E+00	-588900E+03
.469400E+00	-590100E+03
.487000E+00	-587300E+03
.513300E+00	-585600E+03
.570200E+00	-568200E+03
.633800E+00	-522400E+03
.702600E+00	-457100E+03
.775000E+00	-372700E+03
.851800E+00	-270200E+03
.930100E+00	-144200E+03

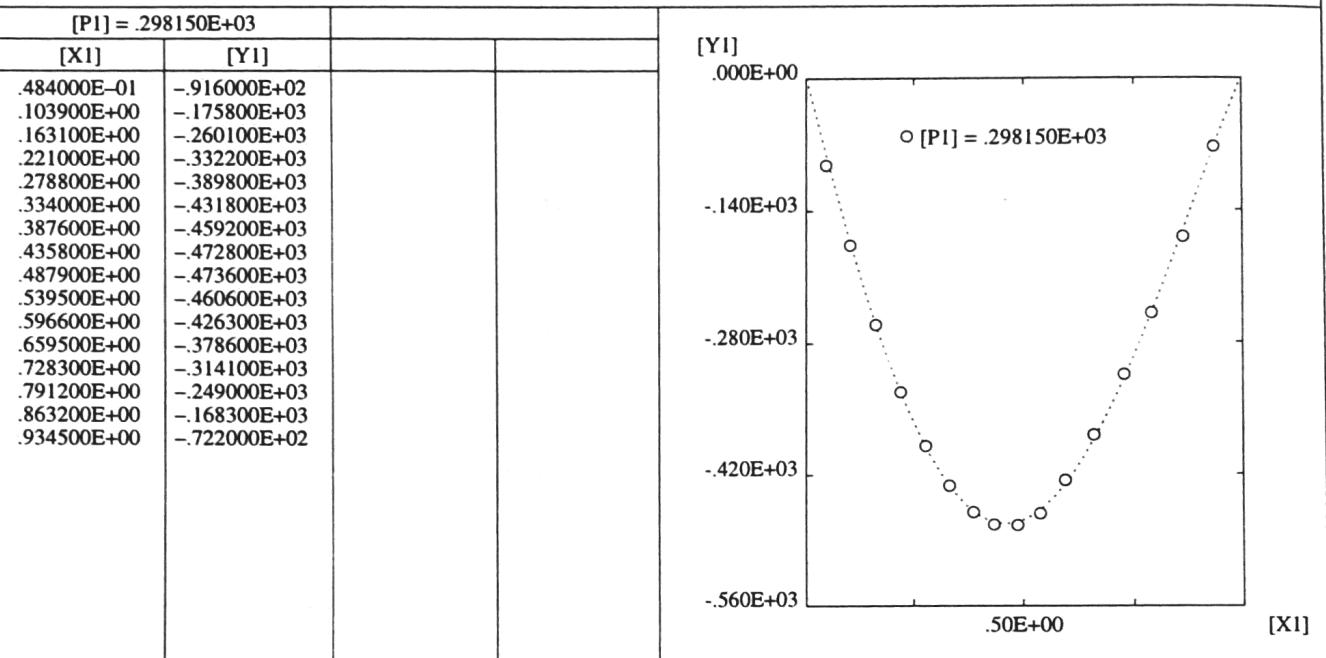


[X1]

ORTJ0962.011

Property Code:	[HMSD1000] HEAT OF MIXING AND SOLUTION
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. $C_6H_{12}Cl_2$, 1,6-Dichlorohexane
2. C_8H_{10} , Ethylbenzene



ORTJ0962.012

Property Code:	[HMSD1000] HEAT OF MIXING AND SOLUTION
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. $C_6H_{12}Cl_2$, 1,6-Dichlorohexane
2. $C_{10}H_{14}$, Butylbenzene

