Isothermal Vapor−Liquid Equilibria, Excess Molar Enthalpies, and Excess Molar Volumes of Trichloromethane + 1,2-Epoxybutane at (288.15, 298.15, and 313.15) K

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Isothermal vapor−liquid equilibria, VLE, excess Gibbs energies, G_m^E, excess molar enthalpies, H_m^E, and excess molar volumes, V_m^E, were determined for the binary mixture trichloromethane + 1,2-epoxybutane at (288.15, 298.15, and 313.15) K and at atmospheric pressure. All excess properties are negative. The vapor−liquid equilibrium data are thermodynamically consistent and have been fitted to the Redlich−Kister equation. The influence of temperature has been discussed, and the relative magnitudes and signs of the excess properties have been outlined in terms of molecular interactions. Smooth representations of the results are presented.

Introduction

The present investigation is one experimental step of a systematic testing of a thermodynamic model (Francesconi et al., 1991) designed to account for the behavior of binary mixtures whose components can interact so strongly they produce new chemical species.

To exploit the possibility of clarifying macroscopic behavior by probing its microscopic causes, we are considering mixtures of very simple, rigid hydrogen donors and cyclic saturated monoethers, henceforth referred to as “ethers”, saturated and monofunctional to avoid π-interactions and multiple interactions, respectively, and cyclic to reduce the number of possible molecular configurations. Thus, the dominant interaction should be hydrogen bonding.

As to the cycle, we plan to examine ethers of variable ring size, to see whether any effect may be correlated with the number of low-frequency deformations (puckerings) which characterize the ring. In the present case, such deformations are absent, because the ring is a three-membered one.

In this paper we report isothermal vapor−liquid equilibria (VLE), excess molar Gibbs energies, G_m^E, excess molar enthalpies, H_m^E, and excess molar volumes, V_m^E, for the binary mixture trichloromethane + 1,2-epoxybutane at (288.15, 298.15, and 313.15) K.

Becker and Rhensius (1986) have determined excess molar enthalpies, H_m^E, for the same mixture at 298.15 K. We have repeated these measurements using a flow microcalorimeter, and our results agree with theirs in the central range of composition (for example, at the minimum, for $x_1 = 0.5200$, $H_m^E = -2033$ J·mol^{-1} (lit. H_m^E −2039.1 J·mol^{-1}).

To the best of our knowledge, we are not aware of any other measurements on the excess properties of this mixture.

Experimental Section

Chemicals. Both components used in this study were purchased from Aldrich Chemical Co. Trichloromethane (99.9+ mol %, ACS HPLC grade) was used without further purification. 1,2-Epoxybutane (99+ mol %) was distilled over sodium at atmospheric pressure in a Vigreux 20-plate column, and only the middle fraction distilling at constant temperature was collected (after purification GCL purity >99.8 mol %). Before use, both components were stored in dark bottles over molecular sieves (Union Carbide, type 4A, 1/16-in. pellets). The experimental densities, ρ, refractive indices, n(D), and boiling points, T_b, of the pure components are listed in Table 1 and compared with literature values.

Apparatus and Procedure. An all-glass recirculation still (Fritz Gmbh, Normag, Hofheim, Germany), the same as that used in our previous work, was used in the vapor−liquid measurements. The details of the still and its operating procedure are described by Gmehling et al. (1980).

Temperature and pressure were measured by digital electronic instruments (Otto Fritz Gmbh, Normag, Germany) with accuracies of ±0.05 K and ±0.05 kPa, respectively.

The compositions of the liquid and condensed vapor were determined by refractometry (Abbe refractometer, Carl-Zeiss-Jena, precision ±0.0001 (Castellari et al., 1988)).

The refractive indices of solutions of known mole fraction, prepared by mass (Mettler balance, model AE 160, accuracy ±0.0001 g), are given in Table 2. The mole fraction composition derived from these measurements is estimated to be precise to within ±0.0005.

A flow microcalorimeter (LKB Produkter, model 2107, Bromma, Sweden) was used to determine the excess molar enthalpies, H_m^E. Details of the equipment and its operation procedure have been described by Monk and Wadso (1968) and Francesconi and Comelli (1986).

The temperature was measured with a precision of ±0.01 K by means of an electronic thermometer. Values of the mole fraction x_1 of component 1 (trichloromethane) of the mixtures obtained from the calibrated flow rates are estimated to be precise to 3×10^{-4}.

The performance of the calorimeter was checked by measuring H_m^E of the test mixture hexane + cyclohexane

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Table 1. Densities, ρ, Refractive Indices, n(D), and Boiling Points, T_b, of Pure Components

	ρ(298.15 K)/(g·cm^{-3})		n(D, 298.15 K)		T_b(101.3 kPa)/K	
component	exptl	lit.[a]	exptl	lit.[a]	exptl	lit.[a]
trichloromethane	1.47919	1.47970	1.4431	1.44293	334.35	334.328
1,2-epoxybutane	0.82463	0.824	1.3815	1.381	336.65	336.574

[a] Riddick et al. (1986).

Table 2. Refractive Indices n(D) for the Binary Mixture Trichlormethane (1) + 1,2-Epoxybutane as a Function of Mole Fraction x_1

x_1	n(D, 298.15 K)	x_1	n(D, 298.15 K)
0.0104	1.3822	0.5197	1.4134
0.0970	1.3877	0.5499	1.4152
0.1455	1.3906	0.5779	1.4169
0.1682	1.3919	0.6020	1.4184
0.2182	1.3948	0.6413	1.4208
0.2860	1.3990	0.7361	1.4268
0.3467	1.4027	0.7940	1.4304
0.4166	1.4071	0.8863	1.4360
0.4499	1.4090	0.9452	1.4398
0.4503	1.4091		

Table 3. Clausius Clapeyron Coefficients A and B, Correlation Coefficients R, and Standard Deviations $\sigma(P°_i)$ for tha Vapor Pressure $P°_i$ of Pure Components

component	temp range/K	A	B	R	$\sigma(P°_i)$/kPa
trichloromethane	280−331	6.9134	−1639.82	−0.99996	0.003
1,2-epoxybutane	288−323	7.0486	−1693.34	−0.99998	0.002

[a] $\log(P°_i/\mathrm{kPa}) = A + B/T(\mathrm{K})$.

at 298.15 K, for which literature values (Benson, 1974) are known. The agreement with the literature was better than 0.5% over the central range of concentration.

The solutions for the measurements of densities, ρ, necessary to determine V^E_m values, were prepared by mass and kept in closed bottles (capacity ~15 cm^3). The possible error in determining the mole fraction is estimated to be ±0.0003.

Densities, ρ, were measured using a vibrating densitymeter (Anton Paar, model DMA 60/602, Graz, Austria) previously described by Fermeglia and Lapasin (1988), and we followed the same procedure also in preparing solutions.

The temperature was determined by digital thermometers (Anton Paar DT 100-10; 100-25; 100-40). A Hetotherm bath circulator (Heto, type 01 DBT 623, Birkerød, Denmark) maintained the temperature constant to within ±0.005 K. Freshly bidistilled and degassed water (Wagenbreth and Blanke, 1968) and dry air (Kohlrausch, 1968) were used as standards of calibration.

The excess molar volumes, V^E_m, were determined from the equation

$$V^E_m = (x_1M_1 + x_2M_2)/\varrho - (x_1M_1/\varrho_1 + x_2M_2/\varrho_2) \quad (1)$$

x_1, x_2; M_1, M_2; and ρ_1, ρ_2 being the mole fractions, the molecular weights, and the densities of component 1 and component 2, whereas ρ is the density of the mixture.

Corrections for buoyancy and evaporation of the components were carried out, but only a negligible contribution to the mole fraction was detected.

Prior to each set of measurements, the apparatus was calibrated with the test mixture benzene + cyclohexane (Wilhelm, 1985), and the results agreed with literature data (discrepancy ±0.5% in the central range of composition).

Table 4. Pressure P, Liquid and Vapor Phase Mole Fractions x_1 and y_1, and Activity Coefficients γ_i (Calculated from a Five-Parameter Redlich−Kister Equation) for Trichloromethane (1) + 1,2-Epoxybutane (2) at (288.15, 298.15, and 313.15) K

P/kPa	x_1	y_1	γ_1	γ_2
		T = 288.15 K		
14.80 (14.81)[a]	0.000	0.000	0.22[c]	1.00
14.30	0.067	0.028	0.34	0.99
13.95	0.091	0.046	0.37	0.99
13.65	0.120	0.063	0.40	0.97
13.25	0.168	0.089	0.43	0.96
12.75	0.211	0.125	0.45	0.95
12.35	0.260	0.168	0.46	0.94
11.75	0.325	0.238	0.49	0.92
11.35	0.399	0.340	0.54	0.87
11.10	0.514	0.537	0.65	0.74
11.80	0.633	0.732	0.78	0.59
12.65	0.711	0.831	0.84	0.55
13.35	0.768	0.889	0.88	0.44
14.40	0.841	0.937	0.92	0.36
15.50	0.916	0.980	0.97	0.25
15.65	0.921	0.983	0.97	0.24
16.75 (16.80)[b]	1.000	1.000	1.00	0.11[c]
		T = 298.15 K		
23.40 (23.45)[a]	0.000	0.000	0.23[c]	1.00
22.30	0.094	0.045	0.37	0.98
21.75	0.129	0.065	0.41	0.97
20.95	0.179	0.101	0.45	0.95
20.25	0.228	0.145	0.48	0.94
19.50	0.293	0.208	0.52	0.91
18.75	0.353	0.285	0.56	0.88
18.30	0.431	0.400	0.63	0.82
18.15	0.511	0.535	0.70	0.73
18.95	0.617	0.710	0.81	0.62
20.25	0.700	0.805	0.87	0.53
21.15	0.752	0.864	0.90	0.48
22.50	0.810	0.915	0.93	0.43
23.80	0.874	0.952	0.96	0.37
25.00	0.930	0.981	0.98	0.29
26.30 (26.20)[b]	1.000	1.000	1.00	0.19[c]
		T = 313.15 K		
43.85 (43.85)[a]	0.000	0.000	0.20[c]	1.00
42.50	0.065	0.025	0.33	0.98
41.45	0.090	0.040	0.37	0.97
40.40	0.143	0.080	0.44	0.95
38.85	0.200	0.127	0.50	0.93
37.15	0.277	0.202	0.55	0.90
36.35	0.347	0.285	0.63	0.87
35.20	0.438	0.412	0.65	0.82
35.30	0.526	0.553	0.72	0.74
36.65	0.633	0.718	0.82	0.62
38.50	0.710	0.810	0.88	0.54
39.85	0.749	0.853	0.91	0.50
42.10	0.823	0.920	0.95	0.42
43.80	0.860	0.950	0.96	0.38
45.50	0.911	0.968	0.98	0.33
47.10	0.960	0.990	0.99	0.27
48.10 (48.10)[b]	1.000	1.000	1.00	0.23[c]

[a] Jónasson et al. (1994). [b] Smith and Srivastava (1986). [c] Calculated from a five-parameter Redlich−Kister equation at $x_1 \to 0$ or $x_1 \to 1$.

Results and Discussion

The values of the vapor pressure $P°_i$ of pure components were fitted to the Clausius−Clapeyron equation. The parameters A and B are listed in Table 3 with their

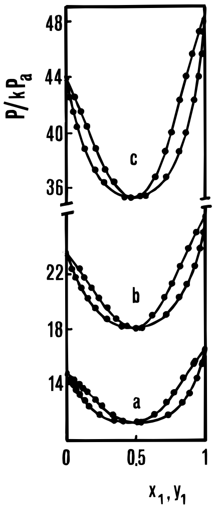

Figure 1. VLE data for the binary mixture trichloromethane + 1,2-epoxybutane at (288.15, 298.15, and 313.15) K: (a), (b), (c) T = 288.15, 298.15, 313.15 K, respectively; (●), experimental points; solid curves, Redlich−Kister equation.

correlation coefficients R and standard deviations $\sigma(P°_i)$; our values agree with those of the literature (Smith and Srivastava, 1986; Jónasson et al., 1994) within the limits of the experimental errors.

The VLE experimental data are listed in Table 4, and the $P-x-y$ plots are shown in Figure 1.

The liquid-phase "experimental" activity coefficients of component i were evaluated by the equation

$$\gamma_i = Py_1/P_ix_i \tag{2}$$

where P_i is the "corrected vapor pressure" (Prausnitz, 1969) obtained from the virial coefficients following the procedure described by Francesconi and Cojutti (1971).

The area test (Prausnitz, 1969)

$$I = \int_0^1 [\ln \gamma_1/\ln \gamma_2 + (V^E_m/RT)\, dP/dx_1]\, dx_1 \tag{3}$$

was used to check the thermodynamic consistency of the isothermal VLE data. It must be remarked that the contribution of the V^E_m/RT dP/d$x_1$term to the integral I is negligible since dP/dx_1 is small and has opposite signs for $x_1 < 0.5$ and $x_1 > 0.5$.

This integral should be zero, and thus only a comparison of I with the uncertainty δI may decide the consistency of the data. Thermodynamic consistency of VLE data is assumed when $|I|$ and δI are of the same order or

$$\delta I \geq |I| \tag{4}$$

In our case, for the three temperatures, $\delta I \approx |I|$, and thus the criterion is verified. The excess molar Gibbs energy, G^E_m, was calculated from

$$G^E_m = RT(x_1 \ln \gamma_1 + x_2 \ln \gamma_2) \tag{5}$$

with γ_i calculated from a five-parameter Redlich−Kister equation to which the experimental γ_i values of eq 2 were fitted, following the least-squares procedure described by Castellari et al. (1988). These adjustable parameters are reported in Table 7, whereas Table 4 reports calculated γ_i values.

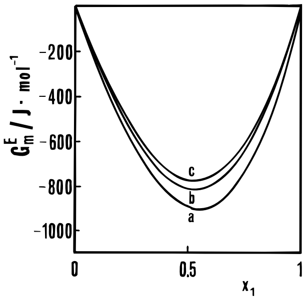

Figure 2. Excess molar Gibbs energies, G^E_m, for the binary mixture trichloromethane + 1,2-epoxybutane: (a), (b), (c) T = 288.15, 298.15, 313.15 K, respectively.

Table 5. Excess Molar Enthalpies, H^E_m, as a Function of the Mole Fraction x_1 for the Mixture Trichloromethane (1) + 1,2-Epoxybutane at (288.15, 298.15, and 313.15) K

x_1	H^E_m/(J·mol^{-1})	x_1	H^E_m/(J·mol^{-1})
	T = 288.15 K		
0.0432	−47	0.6190	−1994
0.0828	−192	0.6842	−1804
0.1192	−378	0.7647	−1404
0.1529	−570	0.8125	−1097
0.2131	−943	0.8667	−718
0.2653	−1253	0.8966	−508
0.3513	−1656	0.9286	−301
0.4193	−1913	0.9630	−117
0.5199	−2078		
	T = 298.15 K		
0.0432	−131	0.6192	−1936
0.0828	−269	0.6842	−1747
0.1193	−417	0.7647	−1385
0.1530	−586	0.8125	−1102
0.2131	−917	0.8667	−735
0.2653	−1218	0.8966	−518
0.3514	−1653	0.9286	−309
0.4194	−1887	0.9630	−118
0.5200	−2033		
	T = 313.15 K		
0.0432	−63	0.6193	−1853
0.0829	−181	0.6844	−1680
0.1194	−339	0.7649	−1326
0.1530	−518	0.8126	−1057
0.2132	−865	0.8668	−707
0.2655	−1181	0.8966	−513
0.3516	−1616	0.9286	−314
0.4195	−1845	0.9630	−125
0.5202	−1971		

Mean deviations between experimental activities given by eq 2 and the ones calculated by the Redlich−Kister equation with the parameters of Table 7 are on the order of 3 × 10^{-2}.

The plot of G^E_m against x_1 for the three temperatures considered is shown in Figure 2.

Tables 5 and 6 list the experimental values and Figures 3 and 4 show the graphical representations of excess molar enthalpies H^E_m and excess molar volumes V^E_m, respectively. Experimental values of excess molar enthalpies and excess molar volumes were fitted to the Redlich−Kister equation

$$Z^E_m/x_1x_2 = \sum_{k\geq 0} a_k(x_1 - x_2)^k \tag{6}$$

(where $Z^E_m = H^E_m$ or V^E_m), by a least-squares analysis procedure, assuming unit statistical weight for each point.

Parameters a_k, based on the lowest values of the standard deviation $\sigma(Z^E_m)$

$$\sigma(Z^E_m) = |\phi/(N - n)|^{0.5} \tag{7}$$

Table 6. Densities, ρ, and Excess Molar Volumes, V_m^E, as a Function of the Mole Fraction x_1 for the Mixture Trichlomethane (1) + 1,2-Epoxybutane (2) at (288.15, 298.15, and 313.15) K

x_1	ρ/(g·cm^{-3})	V_m^E/(cm^3·mol^{-1})	x_1	ρ/(g·cm^{-3})	V_m^E/(cm^3·mol^{-1})
		T = 288.15 K			
0.0103	0.84181	−0.003	0.5195	1.16837	−0.132
0.0967	0.89535	−0.030	0.5498	1.18852	−0.133
0.1454	0.92587	−0.043	0.5777	1.20734	−0.138
0.1681	0.94020	−0.050	0.6018	1.22350	−0.137
0.2180	0.97187	−0.066	0.6412	1.25008	−0.136
0.2858	1.01521	−0.081	0.7359	1.31463	−0.125
0.3465	1.05449	−0.101	0.7939	1.35445	−0.108
0.4164	1.10015	−0.117	0.8862	1.41848	−0.072
0.4497	1.12208	−0.122	0.9451	1.45961	−0.038
0.4501	1.12237	−0.123			
		T = 298.15 K			
0.0103	0.83094	−0.006	0.5195	1.15395	−0.172
0.0969	0.88394	−0.032	0.5498	1.17389	−0.174
0.1454	0.91407	−0.058	0.5777	1.19239	−0.175
0.1681	0.92824	−0.067	0.6018	1.20836	−0.171
0.2180	0.95961	−0.090	0.6412	1.23458	−0.167
0.2858	1.00257	−0.118	0.7359	1.29813	−0.140
0.3465	1.04141	−0.140	0.7939	1.33736	−0.116
0.4164	1.08654	−0.157	0.8862	1.40043	−0.067
0.4497	1.10825	−0.166	0.9451	1.44101	−0.030
0.4502	1.10853	−0.167			
		T = 313.15 K			
0.0103	0.81415	−0.009	0.5195	1.13176	−0.228
0.0969	0.86654	−0.077	0.5498	1.15132	−0.228
0.1454	0.89618	−0.110	0.5777	1.16949	−0.228
0.1681	0.91013	−0.122	0.6018	1.18514	−0.221
0.2180	0.94095	−0.149	0.6412	1.21085	−0.214
0.2858	0.98323	−0.186	0.7359	1.27324	−0.184
0.3465	1.02132	−0.203	0.7939	1.31174	−0.156
0.4164	1.06564	−0.220	0.8862	1.37353	−0.095
0.4497	1.08690	−0.224	0.9451	1.41328	−0.050
0.4502	1.08718	−0.224			

where

$$\phi = \sum_{k=1}^{N} {\eta_k}^2 \qquad (8)$$

with $\eta_k = (H_m^E \text{ or } V_m^E)_{\text{exptl}} - (H_m^E \text{ or } V_m^E)_{\text{calcd}}$, are listed in Table 7.

As can be seen from Figure 1 and Table 4, the mixture trichloromethane + 1,2-epoxybutane forms at T = (288.15, 298.15, and 313.15) K a minimum-boiling azeotrope with negative deviations from Raoult's law (the lowest γ_i values being 0.11, 0.19, and 0.20 for the three temperatures).

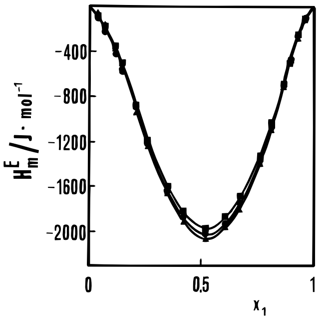

Figure 3. Excess molar enthalpies, H_m^E, for the binary mixture trichloromethane + 1,2-epoxybutane: (▲),(●),(■) experimental points at T = 288.15, 298.15, 313.15 K, respectively; solid curves, Redlich−Kister equation.

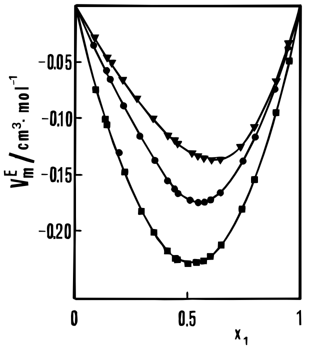

Figure 4. Excess molar volumes, V_m^E, for the binary mixture trichloromethane + 1,2-epoxybutane: (▲),(●),(■) experimental points at T = 288.15, 298.15, 313.15 K, respectively; solid curves, Redlich−Kister equation.

The azeotropic mole fraction is about 0.48, practically unchanged with temperature. The origin of these deviations from ideality is the molecular interactions between the components, a priori substantially the hydrogen bonding between the acid H atom of the chloroalkane (the three electron acceptor Cl atoms being responsible for the positive charge on the H atom) and the negatively charged O atom (with four electron donor CH groups) of the ether.

Evidence of chemical reaction between the components and of consequent complex formation is stressed by Figure 1 and confirmed by the plots of Figure 3, showing strongly hexothermal calorimetric curves by mixing of pure compounds, and also by the volumetric data of Figure 4, where large negative V_m^E values (contraction of the solution with mixing) may be explained by the prevalence of molecular interactions between different compounds over the ones between similar molecules.

Table 7. Least-Squares Parameters a_k and Standard Deviations σ for the Binary Mixture Trichloromethane + 1,2-Epoxybutane at (288.15, 298.15, and 313.15) K

function	a_0	a_1	a_2	a_3	a_4	a_5	σ
			T = 288.15 K				
G_m^E/(J·mol^{-1})[a]	−1.4447	−0.3409	0.2103	0.0254	−0.6329		0.08
H_m^E/(J·mol^{-1})[b]	−8239.6	−1941.0	3686.3	753.8	4061.8		5.1
V_m^E/(cm^3·mol^{-1})[b]	−0.5216	−0.2468					0.01
			T = 298.15 K				
G_m^E/(J·mol^{-1})[a]	−1.3131	−0.1409	0.1319	0.0428	−0.4051		0.04
H_m^E/(J·mol^{-1})[b]	−8110.1	−1338.4	4433.5	−3039.4	1517.7	5255.9	2.8
V_m^E/(cm^3·mol^{-1})[b] −0.6914	−0.1655	0.2473					0.02
			T = 313.15 K				
G_m^E/(J·mol^{-1})[a]	−1.2424	−0.1743	−0.0165	0.2402	−0.2985		0.06
H_m^E/(J·mol^{-1})[b]	−7863.1	−987.2	4344.1	−3657.3	2316.3	4045.6	3.8
V_m^E/(cm^3·mol^{-1})[b]	−0.9160	−0.0524					0.02

[a] Parameters used to calculate G_m^E, eq 5, and obtained from a best fit of the γ_i values of eq 2, through a Redlich−Kister equation. [b] Parameters obtained from a best fit of calorimetric and volumetric data through the polynomial given in eq 6.

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