literature values but on the assumption that the infinite dilution activity coefficient (γ) of the reference and test compound is equal. The latter assumption can lead to problems when GC is used to determine P°_L for polar substances. P°_{GC} overestimates P°_L by up to factors of 5–10 for some organophosphate pesticides and phthalate esters. Caution should be exercised when applying the GC method to these and other polar substances.

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Registry No. γ-HCH, 58-89-9; *p,p'*-DDE, 72-55-9; *o,p'*–DDT, 789-02-6; *p,p'*-DDD, 72-54-8; *p,p'*-DDT, 50-29-3; α-HCH, 319-84-6; naphthalene, 91-20-3; 1-methylnaphthalene, 90-12-0; 1,2,3,4-tetrachlorobenzene, 634-66-2; biphenyl, 92-52-4; 2-chlorobiphenyl, 2051-60-7; 4-chlorobiphenyl, 2051-62-9; fluorene, 86-73-7; hexachlorobenzene, 118-74-1; phenanthrene, 85-01-8; anthracene, 120-12-7; aldrin, 309-00-2; 2,4,6-trichlorobiphenyl, 35693-92-6; pyrene, 129-00-0; dieldrin, 60-57-1; 2,2',5,5'-tetrachlorobiphenyl, 35693-99-3; fluoranthene, 206-44-0; 2,2',4,5,5'-pentachlorobiphenyl, 37680-73-2; eicosane, 112-95-8; benz[*a*]anthracene, 56-55-3; 2,2',3,3',5,5',6,6'-octachlorobiphenyl, 2136-99-4; decachlorobiphenyl, 2051-24-3; benzo[*e*]pyrene, 192-97-2; benzo[*a*]pyrene, 50-32-8; mirex, 2385-85-5; endosulfan I, 959-98-8; endosulfan II, 33213-65-9; endosulfan sulfate, 1031-07-8; *trans*-chlordane, 5103-74-2; *cis*-chlordane, 5103-71-9; cypermethrin, 52315-07-8; fenvalerate, 51630-58-1; heptachlor, 76-44-8; α-chlordene, 56534-02-2; γ-chlordene, 56641-38-4; *trans*-nonachlor, 39765-80-5; *cis*-nonachlor, 5103-73-1; endosulfan ether, 3369-52-6; endosulfan lactone, 3868-61-9; phorate, 298-02-2; diazinon, 333-41-5; methyl parathion, 298-00-0; ethyl parathion, 56-38-2; malathion, 121-75-5; fenitrothion, 122-14-5; chlorpyrifos, 2921-88-2; tris(butoxyethyl) phosphate, 78-51-3; tris(2-ethylhexyl) phosphate, 78-42-2; tricresyl phosphate, 1330-78-5; dimethyl phthalate, 131-11-3; diethyl phthalate, 84-66-2; di-*n*-butyl phthalate, 84-74-2; bis(2-ethylhexyl) phthalate, 117-81-7; *cis*-permethrin, 61949-76-6; *trans*-permethrin, 61949-77-7.

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Vapor Pressures and Gas-Phase *PVT* Data for 1,1-Dichloro-2,2,2-trifluoroethane

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We present new data for the vapor pressure and gas-phase *PVT* surface of 1,1-dichloro-2,2,2-trifluoroethane (refrigerant 123) in the temperature range 338–453 K at densities up to 0.67 mol/L. The data have been represented analytically to demonstrate the precision and to facilitate calculation of thermodynamic properties.

Introduction

1,1-Dichloro-2,2,2-trifluoroethane (CCl_2HCF_3 or R 123) has been suggested as a possible substitute for CCl_3F (R11) for use in the blowing of polyurethane and phenolic foams. Since large quantities of these blowing agents are released directly into the atmosphere, it is desirable that they have a short atmospheric lifetime.

This work is part of an experimental program to determine the thermophysical properties of environmentally acceptable chlorofluorocarbons. In this paper we present measurements of the vapor pressure and gas-phase *PVT* surface for R 123. The data were measured in an existing apparatus, which has been proven capable of making measurements of high precision and accuracy. In the next section we give a brief description of the apparatus and measurement techniques and, following that, a discussion of the results. Comparisons are made where possible with the very few data available for this fluid.

Experimental Section

The Burnett/isochoric *PVT* apparatus has been thoroughly documented in earlier publications (*1*–*3*), and only a brief description is given here. The sample cell is a heavy-walled nickel vessel with two chambers, gold-plated on the inside. The top chamber communicates via a small tube with a very sensitive diaphragm-type pressure transducer. This apparatus is mounted in a circulated and thermostated oil bath. The transducer separates the sample from an external argon-filled manifold that has several precision pressure gauges and an automated piston-type gas injector, which balances the argon pressure against the sample pressure. Temperature of the sample is measured with a platinum resistance thermometer calibrated on the IPTS-68 temperature scale. A feedback circuit using the thermometer, an AC bridge, a signal conditioner, and a programmable power supply controls the temperature of the oil bath. A microcomputer sets the desired temperature, monitors equilibration, and measures temperature and pressure in the automatic mode, which is used for data acquisition on isochores. For the Burnett expansion data, on isotherms, pressure is measured with a gas-operated deadweight pressure balance, which is not automated.

The Burnett expansion mode is used to establish a base-line isotherm on which density can be calculated as a function of pressure. Thereafter, data are measured as pressure versus temperature on isochores with a pressure measurement on the base-line isotherm being used to determine the density of each isochore.

The range of the apparatus is 273–473 K at pressures from 0–20 MPa. Temperatures are measured to about 1 mK. With the dead-weight gauge, pressures are thought to be accurate to within 2×10^{-5} MPa, and the uncertainty with the automated pressure gauge is about 2×10^{-4} MPa. Gas-phase densities are accurate to within several hundredths of 1%.

The very pure samples used were available from a supplier with a stated purity of 0.9995. They were loaded into a sample cylinder and degassed by pumping while frozen with liquid nitrogen. The freezing and pumping cycle was repeated several times until a final measurement of residual pressure indicated the presence of no more than about 10 ppm air. The water content was determined in two different laboratories (*4*, *5*), and the values 23 and 12 ppm were found. The only other impurity found (*4*) was a small (unmeasurable) amount of R 123a ($CClFHCClF_2$). The cylinder was attached to a sample supply manifold, and both were insulated and heated in order to be able to fill the sample cell at elevated temperatures and pressures.

Results

Vapor Pressure. A set of 44 vapor pressure data were measured in the range 338–453 K with the cell approximately half-full of liquid. These results are shown in Table I. An equation of the form

$$\ln P = a/T_r + b + cT_r + dT_r^3 + e(1 - T_r)^{1.5} \qquad (1)$$

fit the data quite well with a standard deviation of 0.01% in pressure. The reduced temperature, $T_r = T/T_c$, was calculated by using the value 456.87 K for T_c, which we reported earlier (*6*). The best values for the parameters were found to be, $a = -8.42429$, $b = 11.27015$, $c = -1.81144$, $d = 0.26528$, and $e = 1.78725$ with *P* in MPa and *T* in Kelvin. This equation yields a value of 3.668 MPa, with an estimated uncertainty of ±0.004 MPa, for the critical pressure. The experimental data extend from a reduced temperature of 0.74 to 0.992. Thus a short extrapolation of eq 1 to $T_r = 0.7$ allows us to estimate the accentric factor for R 123. We find a value of $\omega = 0.280$.

Table I. Vapor Pressure Data for R 123

T, K	*P*, MPa	*T*, K	*P*, MPa	*T*, K	*P*, MPa
338.143	0.3300	398.171	1.3272	433.081	2.4920
338.251	0.3310	398.264	1.3298	433.119	2.4938
343.107	0.3778	403.127	1.4604	433.260	2.5001
348.122	0.4313	403.247	1.4639	434.163	2.5379
350.821	0.4623	408.114	1.6043	438.133	2.7107
353.164	0.4906	408.258	1.6087	438.152	2.7115
358.141	0.5549	413.118	1.7591	438.241	2.7159
363.179	0.6262	413.212	1.7622	443.100	2.9404
368.172	0.7035	418.147	1.9256	443.154	2.9430
373.153	0.7875	418.239	1.9291	443.217	2.9462
378.169	0.8794	423.160	2.1034	448.109	3.1881
383.115	0.9776	423.240	2.1067	448.213	3.1938
388.134	1.0854	428.146	2.2924	453.126	3.4543
393.124	1.2011	428.210	2.2951	453.135	3.4563
393.295	1.2054	433.080	2.4924		

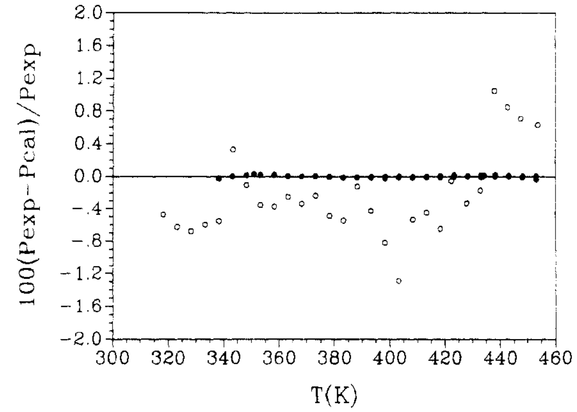

Figure 1. Deviation of the vapor pressure data from eq 1: (●) this work; (○) ref 7.

We found only one other set of documented vapor pressure data for R 123 in the literature, Kubota et al. (*7*). Their data are less precise but extend over a wider range, 273 K to T_c. Figure 1 shows a comparison of the two data sets with eq 1 in the range of our data. The figure shows that our pressures are higher than those of ref 7 over most of the temperature range. In some cases, this could indicate the presence of a volatile impurity in our sample. Because of the care taken in its preparation and handling, we do not feel that such is the case here.

PVT Data. A temperature of 433.074 K was chosen for the Burnett expansion. A higher temperature would have been desirable because it would have allowed us to make *PVT* measurements to higher densities, but operating the expansion valve at high temperatures shortens valve life. Two runs with a total of 9 expansions (11 pressures) were measured, and the results are given in Table II. The data were analyzed by the Burnett method of analysis from ref 8, using an isotherm of the form

$$P = RT\rho(1 + B\rho + C\rho^2) \qquad (2)$$

This truncated virial equation fit the data very well with a

Table II. *PVT* Data on the Burnett Isotherm at 433.074 K

pressure, MPa	density, mol/L	pressure, MPa	density, mol/L
1.808 96	0.672 23	1.381 54	0.468 26
1.158 83	0.377 40	0.847 65	0.262 72
0.698 41	0.211 83	0.499 48	0.147 44
0.407 63	0.118 90	0.288 03	0.082 761
0.233 78	0.066 729	0.164 13	0.046 459
2.081 54	0.834 16		

Table III. Gas-Phase *PVT* Data for R 123

T, K	*P*, MPa	density, mol/L
433.052	1.8088	0.672 2
428.230	1.7715	0.672 3
422.709	1.7281	0.672 5
418.232	1.6923	0.672 6
413.154	1.6512	0.672 8
443.086	1.8857	0.671 9
453.119	1.9611	0.671 6
433.081	1.8093	0.672 2
433.119	1.1590	0.377 4
383.133	0.9614	0.378 2
385.102	0.9701	0.378 2
387.238	0.9790	0.378 1
389.201	0.9870	0.378 1
393.194	1.0032	0.378 0
398.178	1.0233	0.377 9
402.982	1.0425	0.377 9
408.056	1.0625	0.377 8
413.198	1.0826	0.377 7
418.070	1.1015	0.377 6
423.083	1.1208	0.377 5
428.043	1.1398	0.377 5
443.144	1.1968	0.377 2
453.093	1.2340	0.377 1
433.225	1.1596	0.377 4
433.043	0.6984	0.211 83
359.175	0.5483	0.212 49
361.159	0.5527	0.212 47
363.158	0.5569	0.212 45
368.148	0.5675	0.212 41
373.144	0.5780	0.212 36
378.225	0.5885	0.212 32
383.164	0.5987	0.212 27
389.170	0.6110	0.212 22
393.160	0.6191	0.212 19
398.117	0.6291	0.212 14
403.047	0.6391	0.212 10
408.059	0.6490	0.212 05
413.061	0.6590	0.212 01
418.081	0.6690	0.211 96
423.047	0.6787	0.211 92
428.083	0.6887	0.211 88
443.107	0.7179	0.211 74
453.162	0.7374	0.211 65
433.137	0.6986	0.211 83
433.097	0.4077	0.118 91
358.091	0.3277	0.119 28
363.148	0.3332	0.119 26
368.125	0.3387	0.119 23
373.141	0.3441	0.119 21
377.170	0.3494	0.119 18
383.195	0.3548	0.119 16
388.148	0.3601	0.119 13
393.109	0.3654	0.119 11
398.026	0.3706	0.119 09
403.042	0.3760	0.119 06
408.061	0.3813	0.119 04
413.013	0.3865	0.119 01
418.074	0.3918	0.118 99
422.994	0.3969	0.118 96
428.143	0.4024	0.118 94
443.130	0.4180	0.118 86
453.086	0.4284	0.118 81
433.177	0.4078	0.118 91
433.105	0.4077	0.118 91

Table IV. Coefficients for Equation 3 with Pressure in Megapascal and Density in Moles per Liter

$B_0 = 2.473\,62$
$B_1 = -1.802\,84 \times 10^2$
$B_2 = 4.626\,79 \times 10^3$
$B_3 = -4.418\,43 \times 10^4$
$R = 0.008\,314\,41$

$C_0 = 14.0967$
$C_1 = -8.878\,89 \times 10^2$
$C_2 = 1.864\,75 \times 10^4$
$C_3 = -1.301\,48 \times 10^5$

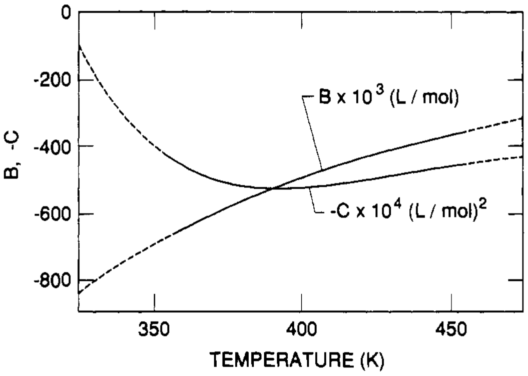

Figure 2. Virial coefficients calculated by eq 3. Dashed portions are extrapolations beyond the range of the data.

standard deviation of 0.13 kPa in pressure. Values of −0.408 43 L/mol and 0.048 45 (L/mol)^2 were found for *B* and *C*, respectively.

A total of 64 *PVT* data were measured at 4 densities with the isochoric method. Temperatures varied from 358 to 453 K, and ensities ranged from 0.119 to 0.67 mol/L. The results are shown in Table III, where the data are listed in chronological order to show that there were no leaks during an isochoric run.

A virial surface, utilizing eq 2 with *B* and *C* being functions of temperature, was fit to the *PVT* data. *B* and *C* were expressed as

$$B(T) = B_0 + B_1\tau + B_2\tau^2 + B_3\tau^3$$

$$C(T) = C_0 + C_1\tau + C_2\tau^2 + C_3\tau^3 \qquad (3)$$

with $\tau = T^{-1/2}$. The coefficients are given in Table IV. This surface fits the data very well, with a standard deviation of 0.17 kPa in pressure, or 0.03% in density (maximum deviation 0.087%). At a temperature of 433.074 K, the calculated values of $B(T)$ and $C(T)$ are −0.4087 L/mol and 0.048 84 (L/mol)^2, respectively, and these agree quite well with the values found above for the Burnett isotherm. Solution of eqs 1–3 allows calculation of the density of the saturated vapor up to a density of 0.67 mol/L. Extrapolation to a density twice this value should cause errors no larger than 0.3% in the calculated density. $B(T)$ and $C(T)$ are plotted as functions of temperature in Figure 2. This surface, along with ideal gas values, allows the calculation of the thermodynamic properties of the gas phase. At the time of this writing, we are not aware of any other published gas-phase *PVT* data for R 123.

Registry No. R123, 306-83-2.

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Isobaric Vapor–Liquid Equilibria in Mixtures of Acetaldehyde and Methyl Iodide

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Vapor–liquid equilibria at 750 Torr were measured for acetaldehyde with methyl iodide. The NRTL equation was used to correlate the experimental results. There was no evidence of a binary azeotrope at high mole fractions of acetaldehyde.

Introduction

The vapor–liquid equilibrium of the acetaldehyde–methyl iodide system is of interest in the production of vinyl acetate. Since a search of the literature failed to find useful information about this equilibrium, the present study was undertaken.

Experimental Section

Chemicals. The acetaldehyde was a BDH laboratory reagent with 99.5 mol % minimum purity. In the course of the work, samples of methyl iodide from two different sources were used: analyzed reagent with 99.8 mol % minimum purity from the J. T. Baker Chemical Co. and material with specified boiling point 41–43 °C from Anachemia Chemicals Ltd.

Pure methyl iodide is a colorless liquid but becomes pale yellow or violet on standing due to the liberation of free iodine. The coloration can be removed by immersing a thin strip of copper or a drop of mercury. Apart from this treatment, the component liquids were used as received without further purification.

Measurements. Vapor–liquid equilibria were established at 750 Torr in a modified Dvořák and Boublík circulating still (*1*). The auxiliary equipment for measuring the temperature and pressure was the same as used by Hull and Lu (*2*). Preliminary experimental runs showed that the previous operational procedure (*1*, *3*) needed modification in view of the low boiling point of acetaldehyde, the extremely large difference in the densities of the two components, and the possible reaction between acetaldehyde and free iodine released during operation of the still. Consequently, the insulation of the still was increased around the equilibrium chamber, the liquid and condensate reservoirs, and the mixing vessel. The entire still was covered with aluminum foil to reduce exposure of the contents to light; small windows were provided for occasional observation of the operation. The still was filled with nitrogen prior to introducing the liquid. Vigorous stirring of the liquids in the reservoirs was initiated before starting to heat the mixing vessel. It was crucial to keep the liquid level in the mixing vessel somewhat lower than that maintained in our previous work, in order to avoid any "air lock" occurring in the tube connecting the liquid reservoir with the mixing vessel.

Table I. Isobaric Vapor–Liquid Equilibria for Acetaldehyde (1)–Methyl Iodide (2): Boiling Temperature, *t*, and Liquid- and Vapor-Phase Mole Fractions, x_1 and y_1, at 750 Torr

x_1	y_1	t/°C	y_1(calcd) − y_1(exptl)	[t(calcd) − t(exptl)]/°C
		Set 1		
1.0000	1.0000	19.94		
0.9642	0.9686	19.95	−0.0024	0.01
0.9578	0.9618	19.95	−0.0011	0.02
0.9320	0.9390	19.99	0.0006	0.04
0.8990	0.9120	20.07	0.0029	0.06
0.8464	0.8740	20.29	0.0054	0.06
0.8134	0.8526	20.49	0.0062	0.02
0.6662	0.7678	21.55	0.0072	−0.05
		Set 2		
0.0000	0.0000	41.72		
0.0128	0.0559	40.32	0.0001	0.05
0.0443	0.1692	37.42	−0.0013	0.08
0.1092	0.3112	33.33	0.0094	−0.08
0.1283	0.3412	32.33	0.0120	−0.04
0.1895	0.4188	29.81	0.0180	0.00
0.2518	0.5070	27.80	−0.0050	0.08
0.3220	0.5658	26.18	−0.0037	−0.01
0.4128	0.6282	24.52	−0.0009	−0.07
		Set 3		
0.3730	0.6060	25.06	−0.0059	0.08
0.4081	0.6258	24.49	−0.0016	0.04
0.5170	0.6880	23.02	0.0037	−0.04
0.6026	0.7346	22.06	0.0055	0.00
0.6710	0.7694	21.48	0.0083	−0.02
0.7250	0.8000	21.08	0.0075	−0.02

The temperature of the cooling medium used in the operation of the still was reduced to −4 °C or lower. Vapor formation in the sampling tube was avoided as much as possible by keeping the time of the sampling procedure as short as possible and by maintaining the temperature of the sampling tube and the covered sample container below 4 °C. Immediately after the sampling, the containers were placed in a refrigerator near the still.

A precision digital densimeter (Anton Paar, Model DMA 02C) was used for the analysis of the samples. It was kept at 4 °C by water circulating from a constant-temperature bath cooled by a Neslab bath cooler (Model PBC-4). At least two analyses were carried out for each sample. In the event of disagreement, a new sample was taken from the still.

Results and Discussion

The experimentally determined liquid and vapor compositions, x_1 and y_1, and Celsius temperatures, t, for the isobaric equi-

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