Liquid Molar Volumes of CH_2FCF_3, CH_3CClF_2, and CH_3CHF_2 and the Mixtures $CHF_2Cl + CH_3CClF_2$ and $CHF_2Cl + CH_3CHF_2$

Krister H. U. Ström* and Urban B. Grén

Department of Chemical Engineering Design, Chalmers University of Technology, Gothenburg, Sweden

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The liquid molar volumes of CH_2FCF_3, CH_3CClF_2, and CH_3CHF_2 and the binary mixtures of $CHClF_2 + CH_3CClF_2$ and $CHClF_2 + CH_3CHF_2$ have been measured in the temperature range 260–320 K. Characteristic volumes for CH_2FCF_3, CH_3CClF_2, and CH_3CHF_2 required for the Thomson–Brobst–Hankinson correlation have been determined. Interaction parameters for the equations of state have been determined from experimental data. In a comparative study, the deviation between experimental data and calculated data from three correlation methods is presented. The three methods are the liquid volume correlation according to Thomson–Brobst–Hankinson and the two equations of state according to Carnahan–Starling–de Santis and Lee–Kesler–Plöcker. The Thomson–Brobst–Hankinson correlation gives the least deviation of the three methods studied, for both pure components and mixtures. Of the two equations of state, the Carnahan–Starling–de Santis equation gives the best agreement with experimental data.

Introduction

Binary refrigerant mixtures are considered promising as working fluids for heat pump systems and other industrial applications. Recently they have also been suggested as the substitutes for completely halogenated refrigerants which have the strongest potential of ozone depletion. However, in order to obtain a satisfactory design implementing these new fluid mixtures, relevant physicochemical data need to be determined.

Liquid volumes have been determined experimentally for three hydrochlorofluorocarbons, CH_2FCF_3, CH_3CClF_2, and CH_3CHF_2, and for the latter two compounds in mixtures with $CHClF_2$. A comparative study between the experimental data and the correlated data using the Thomson–Brobst–Hankinson liquid volume correlation and the two equations of state according to Carnahan–Starling–de Santis and Lee–Kesler–Plöcker is also presented in this paper.

Correlations of Liquid Volume Studied

Thomson and Hankinson (*1*) proposed a correlation for liquid volume at saturated conditions. This correlation has been extended to subcooled conditions by Thomson, Brobst, and Hankinson (*2*).

For pure components, the critical temperature, the acentric factor, and a characteristic volume, which is similar to the critical volume, are required. The Carnahan–Starling–de Santis (*3*) equation combines the hard sphere repulsive compressibility term of Carnahan and Starling (*4*) and the attraction term of Redlich and Kwong (*5*). The equation of state parameters have been determined by Morrison and McLinden (*6*). The Lee–Kesler (*7*) equation of state with mixing rules according to Plöcker et al. (*8*) is a modification of the Benedict–Webb–Rubin (*9*) equation with generalized parameters.

Experimental Method

The density determination was based on measurements of the period of oscillation of a vibrating U-shaped sample tube, which was completely filled with a sample. The following relation exists between the period of time, θ, and the density, ρ:

$$\rho = A^{-1}(\theta^2 - B) \qquad (1)$$

A and B are instrument constants which are determined by calibrations with fluids of known density. The densitometer was calibrated using water and gaseous nitrogen as reference substances. The density data for water and nitrogen were taken from VDI Wasserdampftafeln (*10*) and Jacobsen (*11*).

The vibrating U-tube was kept at a constant temperature by circulating thermostated water around the tube. Temperature control was provided by a thermostatic bath maintained to ±0.001 °C, and the temperature was measured inside the measuring device with a platinum resistance thermometer with a sensitivity of ±0.01 °C. The pressure was measured by a pressure transducer with a precision of ±0.05%.

Density measurements were made with an Anton Paar DMA 602 TP densitometer giving an estimated error of less than ±0.02% in pressure. The refrigerants $CHClF_2$ and CH_2FCF_3 had purities of 99.8 mass %, and CH_3CClF_2 and CH_3CHF_2 had purities corresponding to the quality most commonly available, i.e., technical grade better than 98 mass %.

Results and Discussion

Compressed liquid volumes were measured from 260 to 320 K and at pressures from 0.5 to 1.5 MPa for CH_2FCF_3, CH_3CClF_2, and CH_3CHF_2 and for the two nonazeotropic mixtures $CHClF_2 + CH_3CClF_2$ and $CHClF_2 + CH_3CHF_2$. All data are given in Tables I and II.

In Figure 1 our results for pure refrigerants are plotted together with literature values. It can be concluded from both Figure 1 and Table I that the agreement for CH_2FCF_3 and CH_3CHF_2 is generally better than 0.5%. For CH_3CClF_2 the data of Valtz (*14*) agrees within 0.5–1%. The data for CH_3CClF_2 by Tschernejewa (*15*) show a little higher liquid volume at temperatures over 280 K.

In Figure 2 our results for the mixture x $CHClF_2$ + $(1 - x)$ CH_3CClF_2 are plotted with literature values. There is good agreement with the results by Valtz (*14*). The results presented by Valtz (*14*) at 372 K show that the liquid volume is higher for the mixture with $x = 0.75$ in comparison with the mixture with $x = 0.25$. This is not in accordance with the

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* To whom correspondence should be addressed.

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Table I. Molar Volumes of Liquid Refrigerants 1,1,1,2-Tetrafluoroethane, 1,1-Difluoro-1-chloroethane, and 1,1-Difluoroethane at Different Pressures *P* and Temperatures *T*

CH_2FCF_3			CH_3CF_2Cl			CH_3CHF_2		
P/MPa	T/K	v/(cm^3/mol)	P/MPa	T/K	v/(cm^3/mol)	P/MPa	T/K	v/(cm^3/mol)
0.942	263.50	76.82	0.500	263.88	83.97	0.668	260.51	66.05
1.105	263.50	76.80	1.005	263.88	83.90	1.006	260.54	66.02
1.596	263.50	76.79	1.502	263.88	83.83	1.499	260.54	65.97
0.940	272.91	78.56	0.504	272.87	85.29	0.470	261.85	66.30
1.137	272.91	78.52	0.997	272.87	85.36	1.004	261.85	66.26
1.587	272.91	78.44	1.497	272.87	85.29	1.508	261.85	67.57
0.942	282.58	80.56	1.005	282.84	87.06	0.490	263.59	66.47
1.139	282.58	80.50	1.497	282.84	87.15	1.086	263.58	66.40
1.645	282.58	80.39	1.497	282.84	86.97	1.593	263.57	66.34
0.909	292.70	82.86	0.506	293.09	89.07	0.490	273.03	68.09
1.136	292.70	82.78	1.004	293.09	88.96	1.087	273.03	68.00
1.601	292.70	82.65	1.499	293.09	88.85	1.593	273.03	67.93
1.096	302.45	85.41	0.498	302.99	91.14	0.490	282.78	69.86
1.580	302.45	85.22	1.003	302.99	91.02	1.087	282.78	69.75
1.114	312.89	88.52	1.498	302.99	90.89	1.593	282.78	69.66
1.580	312.89	88.24	1.010	313.23	93.29	0.490	288.12	70.92
1.592	323.09	91.83	1.506	313.23	93.11	1.087	303.02	74.10
			1.008	323.43	95.80	1.593	303.02	73.96
			1.503	323.43	95.58	1.087	312.65	76.55
						1.593	312.65	76.37
						1.593	322.89	79.33

Table II. Molar Volumes of the Mixtures Difluorochloromethane + 1,1-Difluoro-1-chloroethane and Difluorochloromethane + 1,1-Difluoroethane at Different Pressures *P* and Temperatures *T*

$x\ CHF_2Cl + (1-x)\ CH_3CF_2Cl$				$x\ CHF_2Cl + (1-x)\ CH_3CF_2Cl$				$x\ CHF_2Cl + (1-x)\ CH_3CHF_2$				$x\ CHF_2Cl + (1-x)\ CH_3CHF_2$			
x	P/MPa	T/K	v/ (cm^3/mol)	x	P/MPa	T/K	v/ (cm^3/mol)	x	P/MPa	T/K	v/ (cm^3/mol)	x	P/MPa	T/K	v/ (cm^3/mol)
0.2654	0.496	264.00	79.19	0.5583	1.005	282.87	77.03	0.1679	0.662	262.31	66.12	0.5522	1.232	263.64	65.72
	1.011	264.00	79.10		1.518	282.87	76.94		1.014	262.31	66.09		0.806	273.17	67.45
	1.504	264.00	79.05		1.006	292.89	79.00		1.513	262.31	66.04		1.242	273.17	67.74
	0.499	273.19	80.70		1.493	292.89	78.88		0.650	273.84	68.03		0.800	283.10	69.38
	1.006	273.19	80.59		1.008	293.15	79.08		1.010	273.84	67.99		1.270	283.10	69.28
	1.497	273.19	80.52		1.293	293.15	78.96		1.500	273.84	67.93		1.253	293.10	71.41
	0.499	282.92	82.44		1.006	302.98	81.17		0.791	283.06	69.87		1.269	303.03	73.80
	1.004	282.92	82.33		1.509	302.90	81.01		0.988	283.06	69.84		1.249	312.68	76.44
	1.496	282.92	82.22		1.493	313.12	83.49		1.521	283.06	69.75	0.6826	0.792	261.51	65.18
	0.497	293.05	84.39		1.494	323.19	86.31		0.983	293.09	71.95		0.999	261.51	65.09
	0.998	293.05	84.20	0.7669	0.505	263.96	70.06		1.503	293.09	71.84		1.504	261.51	65.07
	1.500	293.05	84.08		1.010	263.96	69.96		1.020	303.16	74.27		0.782	273.24	67.14
	1.000	293.34	84.65		1.516	263.95	69.89		1.500	303.09	74.14		1.025	273.24	67.11
	1.496	293.34	84.54		1.017	273.22	71.49	0.2400	0.557	262.65	66.09		1.493	273.24	67.06
	1.001	303.04	86.63		1.526	273.22	71.39		1.013	262.65	65.95		0.781	283.09	69.13
	1.500	303.04	86.46		1.009	282.89	73.24		1.479	262.65	65.91		1.004	283.09	69.10
	1.005	313.30	89.05		1.510	282.89	73.12		0.808	273.03	67.83		1.512	283.09	69.02
	1.499	313.30	88.86		1.015	292.91	75.25		1.007	273.03	67.74		1.014	293.20	71.29
	1.491	323.36	91.49		1.525	292.91	75.10		1.503	273.28	67.69		1.486	293.20	71.18
0.5583	0.509	263.98	73.86		1.005	293.16	72.35		1.014	282.50	69.68		0.992	303.07	73.73
	1.099	263.95	73.78		1.524	293.16	75.19	0.3773	0.786	263.37	65.94		1.516	303.07	73.57
	1.503	263.94	73.71		1.501	302.89	77.37		1.242	263.28	65.89		1.498	313.10	76.40
	1.009	273.24	75.28		1.527	323.22	83.10		0.805	273.13	67.67	0.7764	0.792	263.59	65.46
	1.509	273.24	75.21		1.527	313.14	80.02		1.242	273.13	67.59		1.232	263.59	65.39
									0.809	283.07	69.58		0.820	273.39	67.22
									1.264	283.07	69.48		1.229	273.39	67.15
									0.791	293.08	71.72		0.819	282.90	69.11
									1.263	293.08	71.58		1.290	282.90	69.02
									0.802	303.05	74.09		0.806	292.91	71.39
								0.5522	1.263	303.00	73.93		1.245	292.97	71.24
									1.258	312.91	76.52		1.253	303.07	73.83
									0.791	263.64	65.77		1.245	312.87	76.68

other data obtained by Valtz (*14*) at other temperatures. Our results for the mixture $CHClF_2 + CH_3CHF_2$ are presented in Table II, but there are no literature data available for comparison.

On the basis of the experimental data presented, characteristic volumes have been determined. The result is presented in Table IV. In the table are also included the critical properties and acentric factors. The equation of state parameters in the Carnahan–Starling–de Santis equation for CH_2FCF_3 have been determined from the experimental data by Wilson (*17*), which are presented in the Appendix.

Table III. Characteristic Volumes v^*, Critical Temperatures T_c, Critical Pressures P_c, and Acentric Factors ω of the Pure Components

component	v^*/(cm^3/mol)	T_c/K	P_c/MPa	ω_{SRK}	ω_{LKP}
CHF_2Cl	163.7	369.15	4.979	0.2215	0.2210
CH_2FCF_3	197.6	374.25	4.067	0.3269	0.3175
CH_3CF_2Cl	222.8	410.20	4.123	0.2386	0.2269
CH_3CHF_2	173.0	386.60	4.519	0.2614	0.2529

In Table II the experimental liquid volume data are given for $CHClF_2 + CH_3CClF_2$ and $CHClF_2 + CH_3CHF_2$. The binary interaction parameters for the mixtures, according to the mixing rules described elsewhere (*12*), have been determined. To adjust the interaction parameter, the objective function, eq 2, has been used. The criterion for an optimal interaction parameter is the minimal value of the objective

Table IV. Relative Standard Deviations σ for Various Correlations

component	reference	N_p	$\sigma_{LKP}/\%$	$\sigma_{CSD}/\%$	$\sigma_{TBH}/\%$
CH_2FCF_3	this work[a]	17	3.78	0.63	0.10
	13[b]	25	3.13	0.78	0.37
	13[b]	10	3.42	1.02	0.19
CH_3CF_2Cl	this work[a]	19	2.39	0.30	0.04
	15[b]	15	1.94	1.29	1.30
	14[b]	4	1.50	1.63	0.63
CH_3CHF_2	this work[a]	21	13.2	0.80	0.11
	16[b]	18	11.6	0.49	0.82

[a] Subcooled conditions. [b] Saturated conditions.

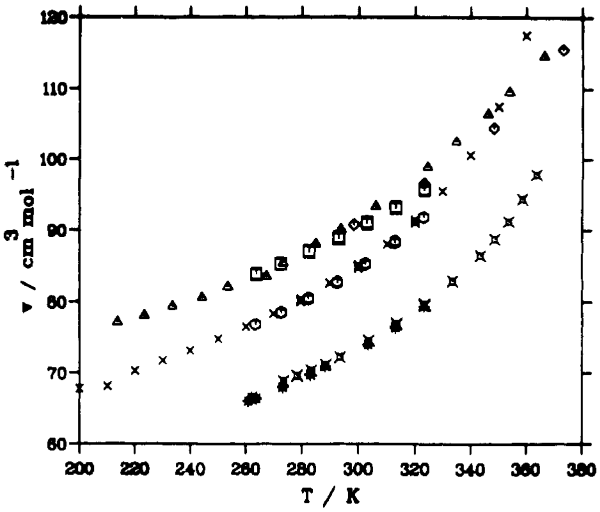

Figure 1. Comparison with literature values: ×, CH_2FCF_3 (*13*); ⊕, CH_2FCF_3 (this work); ◇, CH_3CClF_2 (*14*); □, CH_3CClF_2 (this work); ▲, CH_3CClF_2 (*15*); ✖, CH_3CHF_2 (*16*); *, CH_3CHF_2 (this work).

$$Q = \sum_i \left(\frac{v_i^{\text{exptl}} - v_i^{\text{calcd}}}{v_i^{\text{exptl}}} \right)^2 \qquad (2)$$

function, Q. The binary interaction parameters and the relative standard deviations are given in Table V.

A comparative study has been performed where the relative standard deviation between the calculated data from the three methods and the experimental data including $CHClF_2$, for which the liquid volume of the pure component has been presented earlier (*12*), has been calculated. It was found that the relative deviation of the experimental liquid volume from the Lee–Kesler equation was negative and from 2.0% to 2.5% for CH_2FCF_3; see Figure 3. The results of Maezawa (*13*) at the saturation pressure show the lowest relative standard deviation, 3.1%, compared to compressed conditions. A negative deviation was also found for CH_3CClF_2 in the volume calculations with the Lee–Kesler equation of about 1.5–2.5%. The deviation for the liquid volume data from this work and Valtz (*14*) is nearly constant in the temperature range studied; however, according to Tschernejewa (*15*), the deviation will increase at higher temperatures. For CH_3CHF_2 the relative standard deviation is over 10% for both the data by Sato (*16*) and the data from this work. A plausible reason for these large deviations may be attributed to the critical properties used in the calculations.

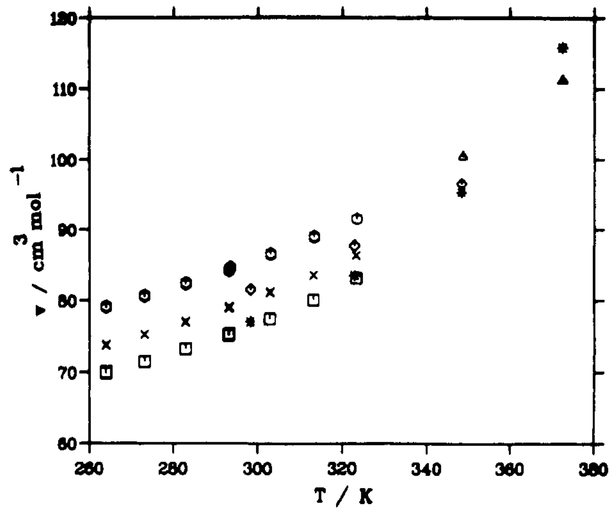

Figure 2. Comparison with literature values for the mixture $CHClF_2 + CH_3CClF_2$: ▲, $x = 0.25$ (*14*); ◇, $x = 0.50$ (*14*); *, $x = 0.75$ (*14*); ⊕, $x = 0.2654$ (this work); ×, $x = 0.5583$ (this work); □, $x = 0.7669$ (this work).

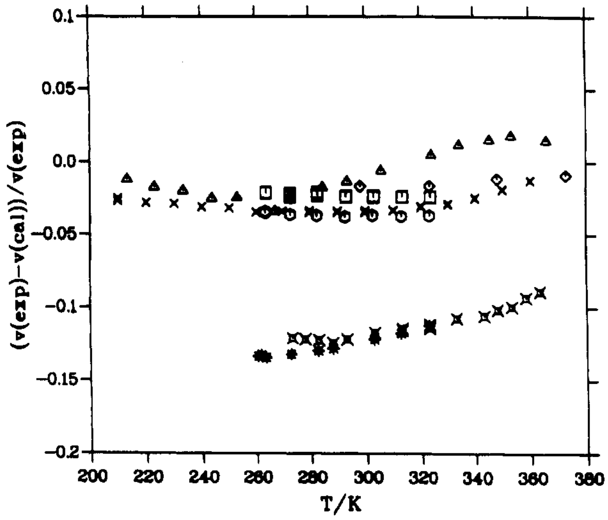

Figure 3. Comparison of literature and calculated values with the Lee–Kesler equation: ×, CH_2FCF_3 (*13*); ⊕, CH_2FCF_3 (this work); ◇, CH_3CClF_2 (*14*); □, CH_3CClF_2 (this work); ▲, CH_3CClF_2 (*15*); ✖, CH_3CHF_2 (*16*); *, CH_3CHF_2 (this work).

With the equation of state according to Carnahan–Starling–de Santis, the experimental data, from both this work and the literature, give deviations of ±1%, see Figure 4, except for the data by Tschernejewa (*15*) which show higher deviations in the temperature range 300–360 K. At higher temperatures, that is, near the critical temperature, the deviation increases. The influence of the pressure may be observed in the liquid volume calculations with the Carnahan–Starling–de Santis equation, Figure 4, in that the relative deviation will increase at higher pressures. This may depend on the fact that the equation of state parameters are determined from data at saturated conditions (*6*).

The Thomson–Brobst–Hankinson liquid volume correlation gives relative deviations around ±1%, Figure 5. For CH_2FCF_3 the data of Maezawa (*13*) give a relative deviation,

Table V. Relative Standard Deviations σ and Interaction Parameters k_{ij} for the Mixtures

			LKP		CSD		TBH	
mixture	reference	N_p	k_{ij}	$\sigma/\%$	k_{ij}	$\sigma/\%$	k_{ij}	$\sigma/\%$
$CHF_2Cl + CH_3CF_2Cl$	this work	48	1.093	0.63	1.010	0.30	0.989	0.02
	14	10	1.046	2.30	1.005	1.66	1.009	0.51
$CHF_2Cl + CH_3CHF_2$	this work	64	1.322	3.49	1.006	0.42	1.001	0.10

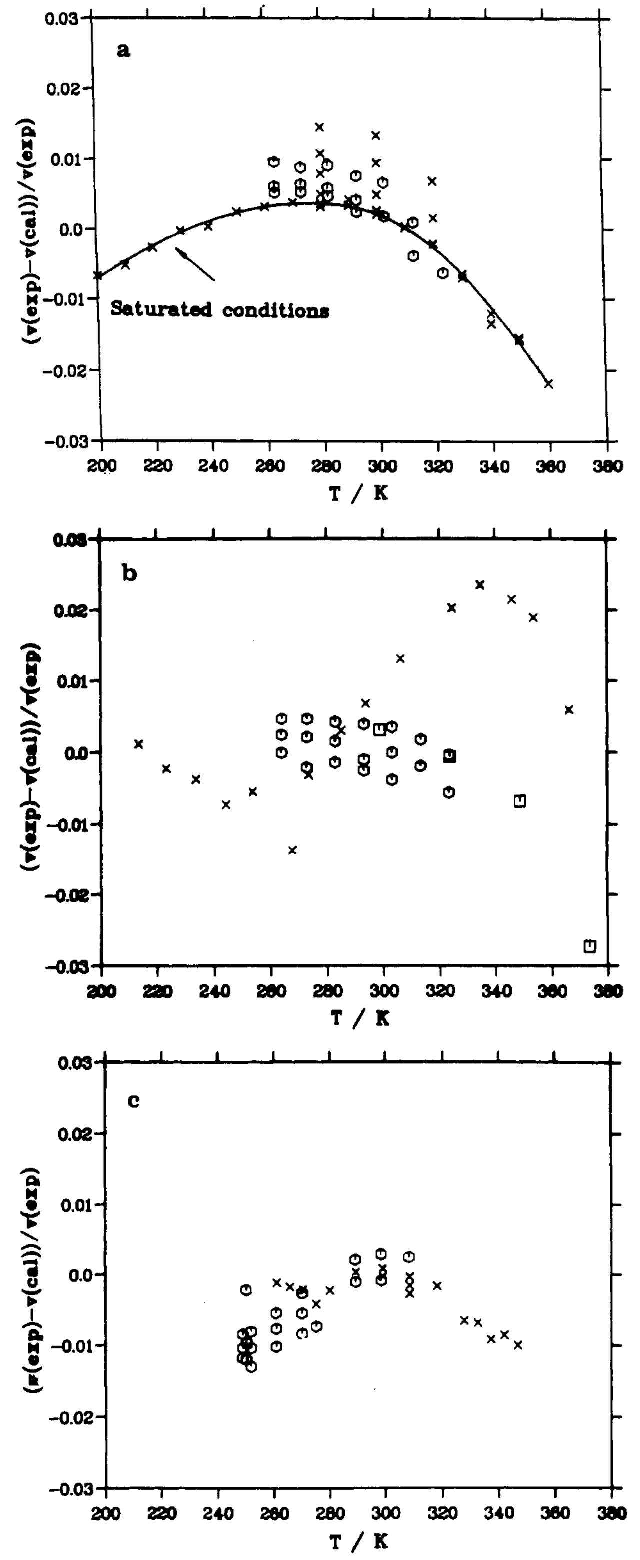

Figure 4. (a) Comparison of literature and calculated values for CH_2FCF_3 with the Caranahan–Starling–de Santis equation: ×, CH_2FCF_3 (*13*); ⊖, CH_2FCF_3 (this work). (b) Comparison of literature and calculated values for CH_3CClF_2 with the Caranahan–Starling–de Santis equation: □, CH_3CClF_2 (*14*); ⊖, CH_3CClF_4 (this work); ×, CH_3CClF_2 (*15*). (c) Comparison of literature and calculated values for CH_3CHF_2 with the Caranahan–Starling–de Santis equation: ×, CH_3CHF_2 (*16*); ⊖, CH_3CHF_2 (this work).

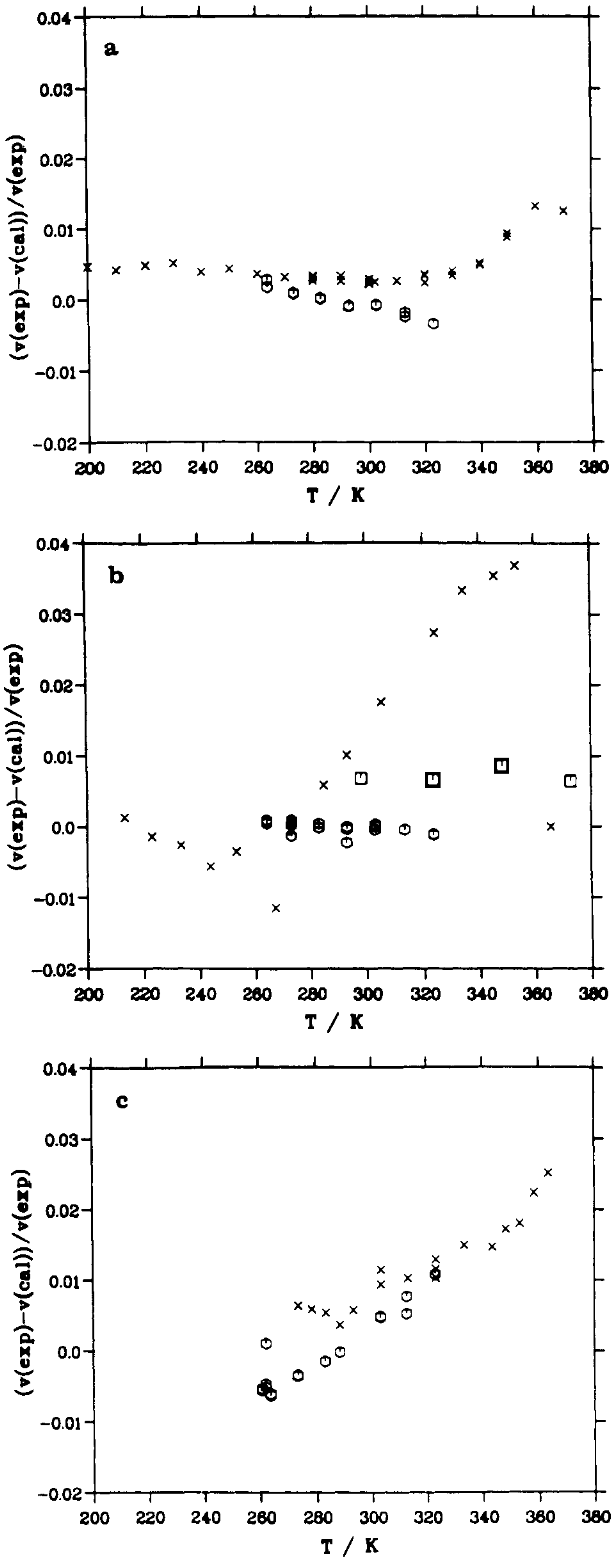

Figure 5. (a) Comparison of literature and calculated values for CH_2FCF_3 with the Thomson–Brobst–Hankinson correlation: ×, CH_2FCF_3 (*13*); ⊖, CH_2FCF_3 (this work). (b) Comparison of literature and calculated values for CH_3CClF_2 with the Thomson–Brobst–Hankinson correlation: □, CH_3CClF_2 (*14*); ⊖, CH_3CClF_2 (this work); ×, CH_3CClF_2 (*15*). (c) Comparison of literature and calculated values for CH_3CHF_2 with the Thomson–Brobst–Hankinson correlation: ×, CH_3CHF_2 (*16*); ⊖, CH_3CHF_2 (this work).

which is constant for temperatures up to 340 K, but then the deviation increases slightly with temperature. Our results show only little influence of the temperature. The liquid volume data by Tschernejewa (*15*) for CH_3CClF_2 show the same trend in the relative deviation, Figure 5b, as with the two equations of state studied although with increasing deviation at higher temperatures. For CH_3CClF_2, the data by Valtz (*14*) give a relative standard deviation of 0.62%, and for this work the deviation is 0.04%. For CH_3CHF_2, the relative deviation increases with increasing temperature (Figure 5c), for the results of both Sato (*16*) and this work.

Both $CHClF_2$ and CH_3CClF_2 show negative deviations from the Lee–Kesler equation of state of about 2–3% in the reduced temperature range, 0.6–0.8% for $CHClF_2$ and 2% for CH_3CClF_2 (*12*). This deviation is reflected in the value of the interaction parameter, calculated from the experimental liquid volumes. The relative standard deviation between the Lee–Kesler equation and the liquid volume data from this work for the mixture $CHClF_2$ + CH_3CClF_2 was 0.63%, compared to 2.30% using the data by Valtz.

The importance of a correct parameter is illustrated for the mixture $CHClF_2$ + CH_3CHF_2 in that, by using an optimal interaction parameter of 1.322, a relative standard deviation of 3.5% is obtained, whereas by using one equal to 1.0, a deviation of 8.8% is obtained. The relative standard deviation for the liquid volume calculated from the Carnahan–Starling–de Santis equation, Table V, is low. This equation gives good results in the liquid volume calculations due to the use of a more correct hard sphere attraction term in the equation (*6*).

Tables IV and V show that the Thomson–Brobst–Hankinson correlation has a lower relative standard deviation compared with the two equations of state and thus is the most suitable correlation. This is explained by the fact that it has more specific parameters than the equations of state such as the characteristic volume.

Conclusion

Liquid molar volumes have been measured for the three compounds CH_2FCF_3, CH_3CClF_2, and CH_3CHF_2 and the two binary mixtures $CHClF_2$ + CH_3CClF_2 and $CHClF_2$ + CH_3CHF_2. The characteristic volumes for the Thomson–Brobst–Hankinson correlation have been determined along with the binary interaction parameters for all three methods. The equation of state parameters for the Carnahan–Starling–de Santis equation have been determined for CH_2FCF_3.

In a comparative study the methods most suitable for refrigerants for liquid volume calculations, Lee–Kesler, Carnahan–Starling–de Santis, and Thomson–Brobst–Hankinson, have been compared. The Thomson–Brobst–Hankinson correlation was found to give the best fit, one reason being that the correlation is based on the law of corresponding states and the characteristic volume is an adjustable parameter with approximately the value of the critical volume. However, this correlation cannot be used for phase calculations as is the case with an equation of state.

The Carnahan–Starling–de Santis equation of state gives a better fit than the Lee–Kesler–Plöcker equation of state to the liquid volume data for the components and mixtures studied. A major reason is that the hard sphere term in the Carnahan–Starling–de Santis equation takes the packing problem at high densities into account. However, the deviation in liquid volume calculations still increases near the critical point, as the equation does not account for scaling in the critical region. The Lee–Kesler equation shows negative deviation for CH_3CHF_2. This affects the calculation of the mixture volume since the value of the interaction parameter has a significant influence on the calculated volume.

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Glossary

A,B	parameters in the expression for the calculation of density
k	interaction parameter
N_p	number of points
P	pressure
T	temperature
v	liquid volume
v^*	characteristic volume
x	mole fraction
Q	objective function

Greek Letters

ρ	density
σ	relative standard deviation
θ	period of time
ω	acentric factor

Subscripts

c	critical
i,j	component numbers
LKP	Lee–Kesler–Plöcker
CSD	Carnahan–Starling–de Santis
TBH	Thomson–Brobst–Hankinson
SRK	Soave–Redlich–Kwong
v	volume

Appendix

Equation of state parameters in the Carnahan–Starling–de Santis equation for CH_2FCF_3: $a_0 = 3264.4$ kJ m^3 kmol^{-2}, $a_1 = 2.14761 \times 10^{-3}$ K^{-1}, $a_2 = -2.68483 \times 10^{-6}$ K^{-2}, $b_0 = 0.14352$ m^3 kmol^{-1}, $b_1 = -1.76071 \times 10^{-4}$ m^3 kmol^{-1} K^{-1}, $b_2 = -4.21926 \times 10^{-8}$ m^3 kmol^{-1} K^{-2}, $a/(\text{kJ m}^3\text{ kmol}^{-2}) = a_0 \exp[a_1(T/\text{K}) + a_2(T/\text{K})^2]$, and $b/(\text{m}^3\text{ kmol}^{-1}) = b_0 + b_1(T/\text{K}) + b_2(T/\text{K})^2$.

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