Alternative Refrigerants CH₂F₂ and C₂HF₅: Critical Temperature, Refractive Index, Surface Tension, and Estimates of Liquid, Vapor, and Critical Densities

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Refractive index data and capillary rise data are reported for CH_2F_2 and C_2HF_5, which are denoted as R32 and R125 by the refrigeration industry. For each fluid, the data extend from 296 K to the critical point and yield the critical temperature T_c and the temperature-dependent capillary length. the refractive index data were combined with liquid density data at 303 K to determine the Lorentz–Lorenz constant k. This constant and the data were used to estimate the liquid, vapor, and critical densities, and the surface tension σ up to the critical point. For both fluids, the surface tension σ is given by the expression $\sigma = S_0 t^{1.26} k_B T_c (N_A/V_c)^{2/3}$ with $S_0 = 5.5$ for R32 and $S_0 = 6.0$ for R125. Here k_B, N_A, T_c, and V_c are the Boltzmann constant, the Avogadro constant, the critical temperature, and the molar critical volume, respectively, and $t \equiv (T_c - T)/T_c$. The present values for S_0 are close to the average value $S_0 = 5.7$ for seven other refrigerants.

I. Introduction

The Montreal Protocol has mandated phasing out the production of the fully halogenated chlorofluorocarbon compounds because of their role in the destruction of the earth's ozone layer. These compounds have enjoyed wide use in industry as refrigerants, foam blowing agents, and solvents. In two previous publications (*1*, *2*) we reported refractive index and capillary rise measurements for seven fluids under consideration as replacements for the fully halogenated refrigerants R11 and R12. Here, we report results for difluoromethane (CH_2F_2) and pentafluoroethane (C_2HF_5), which the refrigeration industry denotes as R32 and R125, respectively. Both R32 and R125 are under consideration as replacements for chlorodifluoromethane (R22) as components of mixtures. R22 is a widely used refrigerant in domestic and industrial applications. The primary focus of this work is the surface tension of R32 and R125. The surface tension is needed to predict heat transfer when phase changes occur. The surface tension influences the nucleation of bubbles in evaporators and the drainage of condensate from condenser surfaces.

We directly measured the critical temperature, the capillary rise, and the refractive indices of both the liquid and the vapor phases of R32 and R125. The refractive indices were combined with other measurements (*3*, *4*) of the liquid densities at 303 K using the Lorentz–Lorenz relation to estimate the densities of the liquid and the vapor phases of R32 and R125 from 296 K to their critical temperatures. (T_c = 351.35 ± 0.02 and 339.33 ± 0.02 K on the ITS90 scale for R32 and R125, respectively.) Because the Lorentz–Lorenz relation is not exact, the estimated densities are subject to systematic errors which, in previous work, we have estimated to be less than 1% of the density of the liquids near their normal boiling points (approximately 10 kg/m³). In the present case, high-quality density data for R32 are consistent with our estimated densities within this uncertainty. Because we determined the critical temperatures precisely from the refractive index data and from observations of the disappearance and reappearance of the meniscus, the values of T_c are not subject to error from imperfections in the Lorentz–Lorenz relation. We determined the surface tension σ from the densities derived from the Lorentz–Lorenz relation and from our capillary rise measurements.

II. Materials

The R32 was obtained from the E.I. DuPont Co. (*5*) and was used without further purification. The purity was estimated at better than 99.9 mol %. No peaks other than R32 were seen from the gas chromatographic analysis (*6*).

A first sample of R125 was obtained from PCR Corp. (*5*) and used without further purification. From the gas chromatograph (*4*), the purity of this sample, as supplied, was estimated to be 99.975 mol % and the principal impurity appeared to be CO_2. However, the aliquot which was used for the present density measurements may have become enriched in CO_2 as a consequence of unwise handling.

A second sample of R125 was obtained from the E.I. DuPont Co. and was used without further purification (*5*). The manufacturer's analysis indicated the presence of R115 (0.267 mol %), CO_2 (2.0 ppm), and CO (0.4 ppm). This sample was used for the present determination of T_c for R125. (R115 forms a nearly ideal mixture with R125, and the critical temperature of R115 is only 14 K above that of R125.)

III. Apparatus and Procedures

The horizontal cross-section of the experimental cell is shown schematically in Figure 1. The cell's body was a horizontal stainless steel cylinder 2.54 cm long and 3.5 cm in diameter. A volume of approximately 6 cm³ was milled out of the cell's body to contain the sample. This volume had a nearly rectangular cross-section. A 1.59-mm (1/16-in.)-outside-diameter stainless steel tube was welded to the side of the cell's body and led to the valve that was used to seal the sample within the cell. Sapphire windows 3.07 cm in diameter and 0.63 cm thick were sealed to the ends of the cylindrical cell with gold wire O-rings. The cell contained four Pyrex (*5*) capillary tubes and a fused silica pentagonal prism that had been cut from an optical-quality right triangular prism. To facilitate a compact and rigid assembly, flats were ground on the outside of the capillary tubes and on the prism. A stainless steel spring clip retained the tubes and the prism in place.

We determined the capillary length a by using a differential capillary rise technique that avoids the difficult measurement of the height h_0 that a meniscus would have in a very wide cell. Instead, at each temperature, we measured the difference in the height Δh between the menisci in two different capillary tubes with larger and smaller radii r_l and r_s, respectively. The
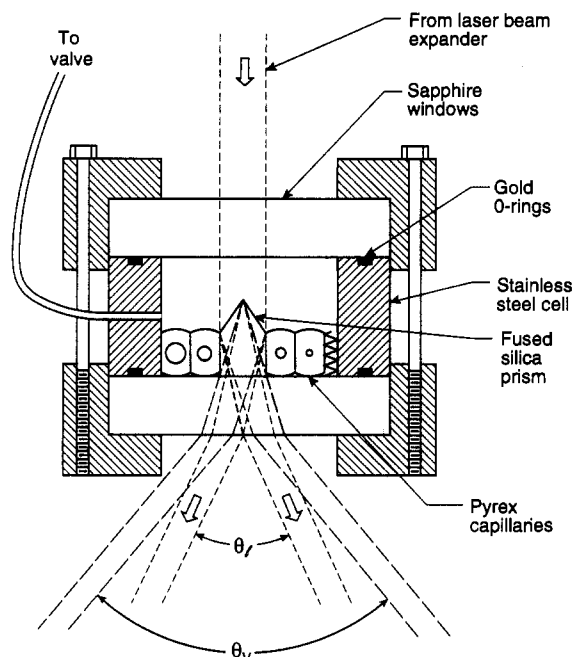
Figure 1. A horizontal cross-section of the stainless steel cell containing four precision bore capillary tubes and a fused silica prism used for refractive index measurements. The windows are sapphire and clamped against the cell and sealed with gold O-rings. Two light beams emerged from the cell from each phase; only one pair is shown here.

meniscus heights were measured with a precision of 0.01 mm with a cathetometer. The capillary length was determined from Δh by an iterative procedure that Lane (7) claimed is more accurate than the use of Sugden's (8) tables. Lane provided polynomial functions $\varphi(r/a)$ which were combined with an initial estimate of a^2 to calculate the capillary rise h_l in the larger of two tubes using the relation

$$h_l = (a_i^2/r_l) \; \varphi(r_l/a_i) \qquad (1)$$

(The subscripts i, l, and s denote the ith iteration, the larger of two tubes, and the smaller of two tubes, respectively.) h_l was then used to obtain a better estimate of a^2 through

$$a_{i+1}^2 = r_s(\Delta h + h_l)/\varphi(r_s/a_i) \qquad (2)$$

Lane's procedure was iterated until the solution converged. Lane's procedure assumes that the liquid–glass contact angle is zero. It appeared to be so.

The surface tension σ is related to the capillary length through

$$\sigma = a^2(\rho_l - \rho_v)g/2 \qquad (3)$$

where ρ_l and ρ_v are the density of the liquid and vapor, respectively, and g is the acceleration due to gravity, 9.80 m/s².

The sample cell contained four capillaries with the following bore radii: r_1 = 0.4961 ± 0.0015 mm, r_2 = 0.2560 ± 0.0010 mm, r_3 = 0.1304 ± 0.0002 mm, and r_4 = 0.0611 ± 0.0001 mm. Those with the larger radii were used far from T_c; those with the smaller radii were used closer to T_c. The radii had been determined prior to the assembly of the cell by partially filling the capillaries with mercury plugs, measuring the lengths of the plugs with a traveling microscope, and then weighing the mercury.

The refractive indices of the refrigerants were measured using an expanded and collimated beam of laser light (λ = 0.633 μm) that was directed horizontally through the cell as indicated in Figure 1. The prism within the cell divided the

Table 1. Capillary Length Data and Refractive Index Data

$(T_c - T)/T_c$	a^2/mm^2	n_v	n_l
	R32		
0.15583	1.563	1.0073	1.1946
0.12371	1.311	1.0104	1.1861
0.09399	1.024	1.0155	1.1744
0.06545	0.728	1.0186	1.1614
0.03747	0.446	1.0303	1.1472
0.02046	0.253	1.0385	1.1331
0.01435	0.168	1.0437	1.1266
0.00874	0.109	1.0490	1.1185
0.00577	0.070	1.0539	1.1131
0.00315	0.035	1.0582	1.1074
0.00153		1.0623	1.1009
0.00049		1.0694	1.0953
0.00036		1.0712	1.0942
0.00005		1.0769	1.0896
	R125		
0.12260	0.734	1.0125	1.1847
0.08911	0.552	1.0186	1.1708
0.05823	0.371	1.0262	1.1582
0.03862	0.250	1.0303	1.1473
0.02238	0.148	1.0398	1.1353
0.01450	0.102	1.0463	1.1276
0.00963	0.063	1.0498	1.1222
0.00378	0.032	1.0596	1.1109
0.00198	0.024	1.0643	1.1054
0.00133		1.0657	1.1030
0.00050		1.0714	1.0987
0.00022		1.0745	1.0947

beam into two beams that diverged at an angle θ that depended upon the refractive index of the fluid in contact with the prism through the relation

$$n = -\sin(\theta/2)\,\tan(\theta_p/2) + [n_p^2 - \sin^2(\theta/2)]^{1/2} \qquad (4)$$

Here n_p = 1.4571 is the refractive index of the fused silica prism and θ_p = 90.056° is the angle of the prism that divided the light beam. The expanded laser beam illuminated both the liquid and the vapor phases of the refrigerant. Thus, four beams exited the cell; the lower two diverged from the liquid at the angle $\theta = \theta_l$, and the upper two diverged from the vapor at the angle θ_v. The angles θ_l and θ_v were measured using a mirror that was fixed to a calibrated rotational mount. The mount was rotated until, successively, each diverging beam was reflected into a stationary telescope focused at infinity. When the beam was coincident with the cross hairs in the telescope, the orientation of the rotational mount was read. The uncertainty in measuring θ_l and θ_v was approximately ±0.001 rad.

Prior to the final filling, the cell was flushed with the refrigerant under study and then evacuated. The cell was partially filled with liquid by condensing the refrigerant into the cell at approximately 273 K.

The cell was placed in a thermostated oil bath with flat transparent windows. The bath's temperature was monitored with a calibrated platinum resistance thermometer that showed temperature fluctuations of ±0.02 K. In most cases 1 h was allowed to elapse at each temperature before measurements of the refractive index, capillary rise, and temperature were made. The data are listed in Table 1.

For each sample, the critical temperature was bracketed by repeatedly observing the disappearance and reappearance of the meniscus. The values of T_c determined in this fashion were always consistent with those deduced from the refractive index and capillary rise data.

IV. Analysis

The refractive index differences $(n_l - n_v)$ were fitted by the equation

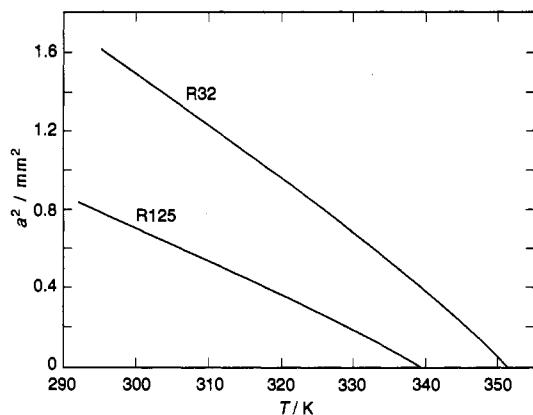$$n_l - n_v = 2\Delta n_0 t^{0.325}(1 + n_1 t^{0.5} + n_2 t) \qquad (5)$$

Figure 2. Square of the capillary rise parameter a^2 vs temperature. The plotted values are from eq 8, with the parameters a_0 and a_1 listed in Table 2.

where the reduced temperature is defined by

$$t = (T_c - T)/T_c \qquad (6)$$

The fit determined T_c, the refractive index amplitude Δn_0, and two additional parameters, n_1 and n_2. The exponents 0.325 and 0.5 were obtained from theories (9) appropriate in the critical region. The refractive index averages $(n_l + n_v)/2$ were fitted by the following two-parameter equation:

$$n_l + n_v = 2n_c(1 + n_d t) \qquad (7)$$

Here n_c is the refractive index at the critical point and n_d is the slope of the "rectilinear diameter" of the refractive index as a function of the temperature.

The capillary length data for each refrigerant were fitted by the equation

$$a^2 = a_0^2 t^{0.935}(1 + a_1 t) \qquad (8)$$

where a_0^2 and a_1 were the fitted parameters and the exponent 0.935 was taken from theory and is believed to be asymptotically correct (10) near T_c. T_c was taken from the fit of eq 5 to the refractive index data. Equation 8 for a^2 is plotted in Figure 2, and the deviations of the data from eq 8 are plotted in Figure 3. The parameters that resulted from the fitting procedure are listed in Table 2, together with t_{max}, the maximum value of t used in the fit.

The refractive index data and liquid density data ρ^* at reference states (3, 4) for each system were combined to obtain the Lorentz–Lorenz "constant" k that appears in the approximate relation

$$\frac{n^2 - 1}{n^2 + 2} = k\rho^* \qquad (9)$$

The reference states ρ^* are listed in Table 3. Using eq 9 and the value of k obtained from the reference point, densities were calculated at each temperature for which refractive indices were measured. These densities were fitted by the following equations that closely parallel eqs 5 and 7 and are algebraically simpler than combining eqs 5, 7, and 9:

$$\rho_l - \rho_v = 2\Delta\rho_0 t^{0.325}(1 + \rho_1 t^{0.5} + \rho_2 t) \qquad (10)$$

$$\rho_l + \rho_v = 2\rho_c(1 + \rho_d t) \qquad (11)$$

In eq 10, $\Delta\rho_0$, ρ_1, ρ_2, and, implicitly, T_c are the fitted parameters. In eq 11 the critical density ρ_c and the rectilinear diameter ρ_d are the fitted parameters. The fitted equations for the density are plotted in Figure 4, and the fitted parameters are listed in Table 4. The deviations from eqs 10 and 11 are plotted in Figure 5.
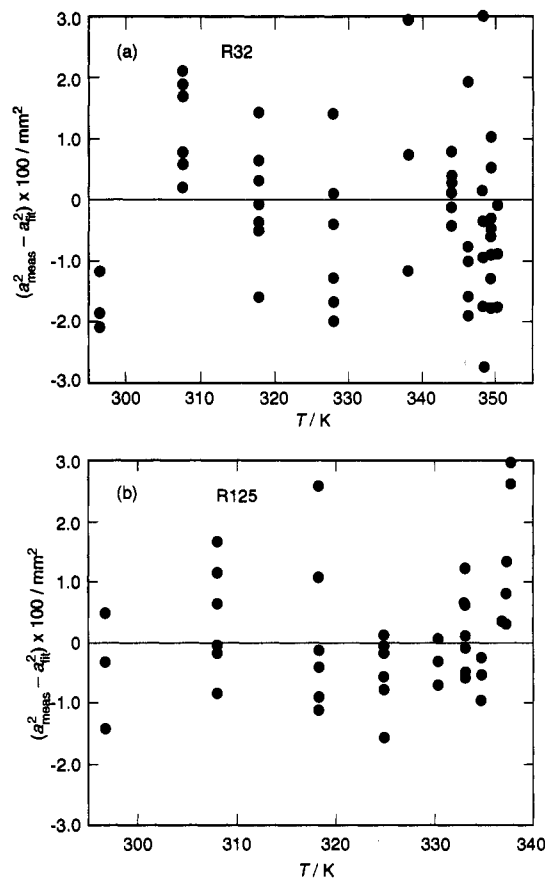

Figure 3. Deviations of the capillary rise data from the fitted equations shown in Figure 2.

Equation 1 was used to calculate the surface tension from the fitted parameters for the capillary lengths and the density differences. The resulting representations of the surface tension have five numerical parameters in addition to T_c and the exponents. To obtain a simpler two-parameter representation, the surface tensions were also fitted with the function

$$\sigma = \sigma_0 t^{1.26}(1 + \sigma_1 t) \qquad (12)$$

This two-parameter function is within 0.02 mN/m of the full five-parameter function that it replaces. The parameters σ_0 and σ_1 are listed in Table 4.

The surface tensions of other refrigerants (R123, R123a, R134, R134a, R141b, R142b, and R152a) were found (1, 2) to scale according to the principle of corresponding states. Within 7%, the reduced surface tension S follows the law

$$S = \frac{\sigma}{k_B T_c}\left(\frac{M}{N_A \rho_c}\right)^{2/3} = S_0 t^{1.26} \qquad (13)$$

where M is the molecular weight, k_B is Boltzmann's constant, and N_A is Avogadro's constant. For the seven refrigerants listed above, $S_0 = 5.7 \pm 0.2$. We find the present pair of refrigerants R32 and R125 are best represented with $S_0 = 5.5$ and 6.0, respectively (see Figure 6). Thus, the value $S_0 = 5.7$ is a reasonable estimate for halogenated ethanes and methanes, and we recommend it for cases in which there are no data.

V. Systematic and Random Errors

Random errors in the measurement of the refractive index resulted from imperfections in the rotational mount used for the angle measurements and from small displacements of the cell during the measurements. The rotational mount could be read to approximately 1 arc min or, equivalently, 0.0003

Table 2. Summary of Measurements

refrigerant	T_c/K	Δn_0	n_1	n_2	n_c	n_d	a_0^2/mm²	a_1	t_{max}
R32	351.36	0.1517	0.890	−1.423	1.0828	0.1126	9.55	−0.38	0.16
R125	339.33[a]	0.1565	0.376	−0.394	1.0848	0.1048	5.26	0.00	0.14

[a] Second sample of R125. See section VI in the text.

Table 3. Reference Data for Estimating Densities

refrigerant	T/K	ρ^*/(kg/m³)	P/kPa	M/(g/mol)	k/(cm³/g)	B/(cm³/mol)	C/[(dm³/mol)²]
R32	303.35[a]	938[a]	1939[a]	52.023	0.1295	−297[a]	28.7[a]
R125	303.31[b]	1158[b]	1578[b]	120	0.0984	−359[c]	30.0[c]

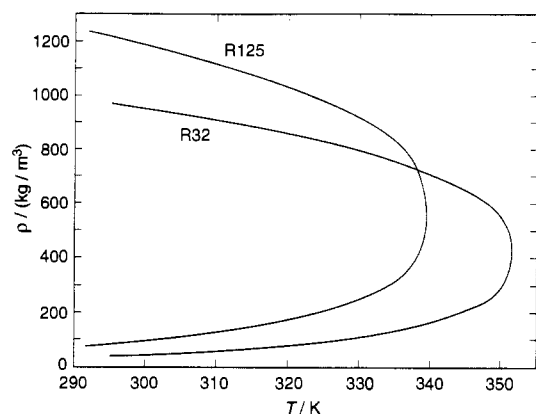[a] Defibaugh, Morrison, and Weber (*3*). [b] Defibaugh and Morrison (*4*). [c] Weber (*13*).

Figure 4. Density as a function of temperature from eqs 10 and 11 and the parameters listed in Table 4.

Table 4. Derived and Estimated Properties

refrigerant	$\Delta\rho_0/\rho_c$	ρ_1	ρ_2	ρ_c/ (kg/m³)	ρ_d	σ_0/ (mN/m)	σ_1
R32	1.806	0.878	−1.468	419	1.289	80.8	−0.49
R125	1.816	0.372	−0.449	565	1.159	55.5	0.19

rad. Two readings were needed for each measurement of θ_l or θ_v, and the resulting errors in the refractive index are expected to be approximately 0.0006. The standard deviation of the refractive index data with respect to eqs 5–7 was 0.001 for R32 and 0.0007 for R125.

A bracket held the cell in the oil bath. Any undetected rotation of this bracket would result in errors in θ_l and θ_v of second order in the rotation. If a rotation of 1.5° had occurred in either direction, it would have decrease θ_l and θ_v by 1.5 arcmin and the refractive indices deduced from the data would have been decreased by 0.000 44. The prism angle θ_p was measured to within 3 arc s. An error of 3 arc s in θ_p would lead to a negligible error (1.4×10^{-5}) in the refractive index.

The present technique uses refractive indeces and the Lorentz–Lorenz relation to obtain densities. Chae et al. (*2*) have reviewed the errors associated with the assumption that k is "constant" in the Lorentz–Lorenz relation. They concluded that the systematic errors in determining the liquid and vapor densities resulting from this assumption do not exceed approximately $0.008\rho^*$, where ρ^* is the density of the liquid at the state used to determine k. This implies that the critical density, which is on the order of 1/2 of the liquid density, might be in error by as much as 1.6% of ρ_c. The analysis of Chae et al. appears to be confirmed by comparing the present data for R32 with high-quality data from the literature (see below).

The random errors in the measurements of θ_l and θ_v lead to random errors in the computed densities on the order of 5 kg/m³. When the density is small (i.e., in the vapor at low temperatures), these random errors greatly exceed all the systematic errors that are proportional to the density.
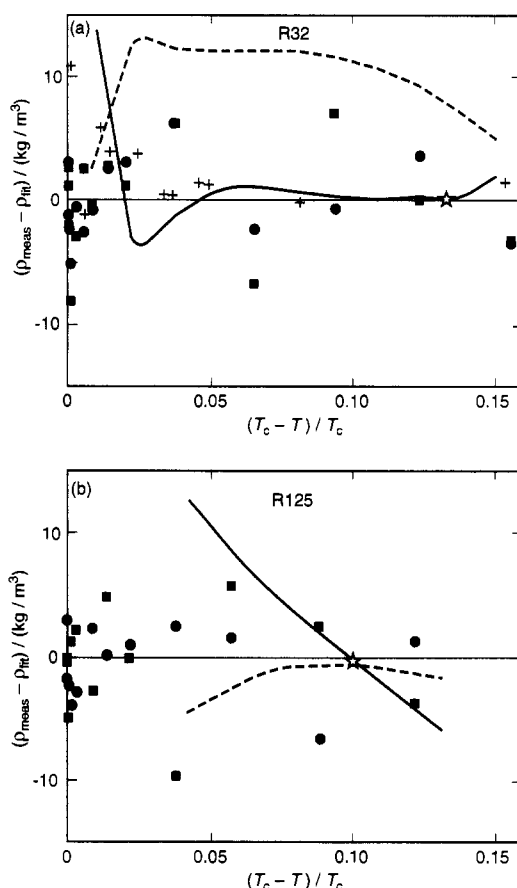

Figure 5. Deviations of densities from eqs 10 and 11 and the parameters listed in Table 4. The solid circles and squares are the liquid and vapor densities, respectively, that were obtained by us from the Lorentz–Lorenz relation. For R32, the results from ref 3 for the liquid and vapor densities are represented by the solid and dashed curves, respectively. For R32, the liquid densities tabulated in ref 11 were obtained with a pycnometer and are represented by plus signs. For R125, the dashed curve for the vapor density was estimated from a correlation of virial coefficients in ref 13 and the solid curve for the liquid density represents extensive measurements using a vibrating tube densimeter in ref 4. For both R32 and R125, the stars indicate the reference states for determining the Lorentz–Lorenz parameters.

The standard deviation of the square of the capillary rise parameter a^2 was approximately 0.02 mm² for both R32 and R125. The random errors in a^2 correspond to random errors in measuring the height of the menisci of ±10 μm. Depending upon the capillary tubes used and upon the temperature, the relative uncertainties in Δh were between 0.2% and 1% far from T_c. This accounts for a relative uncertainty in a^2 of as much as 2%. The systematic errors resulting from the uncertainties in the determination of the capillary radii were
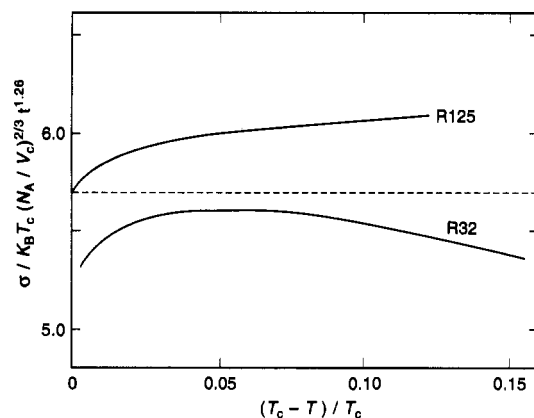
Figure 6. Reduced surface tension $\sigma/k_B T_c (N_A/V_c)^{2/3} t^{1.26}$ as a function of the reduced temperature. The dashed line represents the results obtained from seven refrigerants of refs 1 and 2.

between 0.1% and 0.4%, and they propagate to another systematic error in a^2 as large as 0.8%. The variation of k might lead to systematic errors in the densities and in the surface tension on the order of 1.6%. The sum of these possible systematic errors is 4.4%, and the sum in quadrature is 2.6%, in the worst case. The random errors in surface tension were approximately ±0.05 mN/m.

VI. Comparison with Other Data

Parts a and b of Figure 5 display the deviation of our density data from eqs 10 and 11. Also displayed in Figure 5a are liquid density data from Malbrunot et al. (*11*) (plotted points) and the densities for R32 deduced by Defibaugh et al. (*3*) (curves) from a multiparameter equation of state.

Defibaugh et al. (*3*) based their multiparameter equation of state on extensive, very accurate vapor pressure measurements and liquid- and vapor-phase *PVT* measurements. Except for a narrow region near the critical point, our results (Figure 5a, plotted points) are consistent with theirs (curves), within the combined random and systematic errors in our data. We recommend using the results from ref 3 for the densities at the saturated vapor pressure, except for the critical region. In the critical region, the form of the equation used in ref 3 is inherently poor at representing the equation of state. Furthermore, the equation of state from ref 3 was deduced without using any density data in the range 200–700 kg/m³ at the saturation conditions. Our R32 density data are also consistent with those obtained by Malbrunot et al. (*11*) using an accurate pycnometer.

In the case of R125 (Figure 5b), our vapor densities are compared with those predicted from a correlation (*13*) of the density virials $B(t)$ and $C(T)$. The agreement is satisfactory. Figure 5b also demonstrates that our liquid densities agree, within their estimated error, with the liquid densities deduced by Defibaugh and Morrison (*4*) from their accurate measurements using a vibrating tube densimeter.

For R32, the critical parameters T_c and ρ_c were determined by Malbrunot et al. (*11*) from liquid and vapor density data. After conversion to ITS90, their results are $T_c = 351.52 \pm 0.20$ K and $\rho_c = 429.6 \pm 1.2$ kg/m³. They also reported the result $T_c = 351.32 \pm 0.10$ K from a sealed tube measurement. The sealed tube measurement is in excellent agreement with the present value $T_c = 351.35 \pm 0.02$ K obtained directly from our observations of the disappearance and reappearance of the meniscus. We also obtained the value $T_c = 351.36 \pm 0.02$ K from fitting eq 5 to the refractive index data. For R32 we obtained the value $\rho_c = 419 \pm 7$ kg/m³. It is 2.5% smaller

than that of Malbrunot et al.'s value and just outside the range spanned by conservatively combining error estimates. In fact, the liquid density data that Malbrunot et al. tabulated agree very well with our own (see Figure 5a). Their single datum closest to T_c is the only one that differs from our correlation by more than 1.5%. However, Malbrunot et al. did not measure the density of the saturated vapor directly. Instead, they fitted equation-of-state data to a Martin–Hou equation of state and extrapolated the Martin–Hou equation to their vapor pressure curve. Perhaps some of the discrepancy between our value for ρ_c and theirs can be explained by problems resulting from using the Martin–Hou equation of state near the critical point.

For R125 the critical parameters have been measured by Wilson et al. (*12*). Their value $T_c = 339.19 \pm 0.20$ K is in very good agreement with the value $T_c = 339.33 \pm 0.02$ K determined by us from observations of the disappearance and reappearance of the meniscus in our second sample. Wilson et al.'s value $\rho_c = 571 \pm 3$ kg/m³ is consistent with the value $\rho_c = 565 \pm 9$ kg/m³ determined for our first sample within the combined errors in the two measurements. (We did not measure ρ_c for our second sample.)

Our first sample of R125 yielded a value for T_c that was 1.22 ± 0.02 K lower than that of the second sample. We believe that the difference in T_c between our two samples may have been due to the presence of more CO_2 in the first sample. CO_2 forms an azeotrope with R125 and is difficult to remove. Such an impurity might lower the estimate of k and raise the estimate of ρ_c by 0.3%.

VII. Summary

We have measured the refractive indices and capillary rise for two proposed substitutes for R22, namely, R32 and R125. The data were combined with reference densities near 303 K to determine the Lorentz–Lorenz constant k from which estimates of the vapor and liquid densities were made up to the critical points. Surface tensions were found to scale according to the expression $\sigma = 5.7 t^{1.26} k_B T_c (N_A/V_c)^{2/3}$ within 7%.

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