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# Equations of State for the Ozone-Safe Refrigerants R32 and R125[1]

**A. A. Vasserman[2, 3] and D. V. Fominsky[2]**

Equations of state for gaseous and liquid difluoromethane (R32) and pentafluoroethane (R125) were developed. The coefficients of the equations were determined using experimental density data and heat capacities $c_v$ and $c_s$. The equations satisfy Maxwell's rule. The equations describe the thermodynamic properties of R32 and R125 at temperatures from 140 to 433 K and from 178 to 480 K, respectively, and at pressures up to 70 MPa within the experimental uncertainties. In particular, the root-mean-square deviations of the calculated values of density from the most reliable experimental data are equal to 0.10% for R32 and 0.12% for R125.

**KEY WORDS:** density; difluoromethane; equation of state; heat capacity; pentafluoroethane; R32; R125.

## 1. INTRODUCTION

The necessity of replacing the traditional refrigerants that deplete atmospheric ozone with alternative refrigerants caused an intensive study of the properties of such substances. Difluoromethane (R32) and pentafluoroethane (R125) are alternative ozone-safe refrigerants. The data on their thermodynamic properties are required for designing refrigeration plants using difluoromethane, pentafluoroethane, or their mixtures with other substances as working media.

Recently equations of state for R32 and R125 [1, 2] were published. However, the newest $p$, $\rho$, $T$-data [3] were not used. Comparisons showed that, in general, these equations of state describe data [3] satisfactorily;

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[2] Odessa State Maritime University, Mechnikova Str. 34, Odessa 65029, Ukraine.

[3] To whom correspondence should be addressed. E-mail: avas@writeme.com

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however, the errors of their analytical description for each substance are still higher than the errors of the experiments. It should be noted that the data [3] cover a wide range of temperature and pressure and the number of experimental points is more than the quantity obtained in all previous research. We decided to develop new equations of state for R32 and R125 taking into account these data [3].

## 2. EQUATIONS OF STATE

The equations of state developed in this work have the form,

$$\frac{F(\omega, \tau)}{RT} = \alpha_0(\omega, \tau) + \alpha(\omega, \tau) \tag{1}$$

where $F(\omega, \tau)$ is the Helmholtz free energy, $\omega = \rho/\rho_{\mathrm{cr}}$ is the reduced density, and $\tau = T/T_{\mathrm{cr}}$ is the reduced temperature. The first term of Eq. (1) is an ideal-gas part that is determined using ideal gas heat capacity data $c_{p0}$. The function $\alpha(\omega, \tau)$ is the real part of the equation of state and can be written as

$$\alpha(\omega, \tau) = \sum_{i=1}^{m} \sum_{j=0}^{r_i} a_{ij} \frac{\omega^i}{\tau^j} + \exp(-\omega^2) \sum_{i=1}^{n} \sum_{j=1}^{s_i} b_{ij} \frac{\omega^i}{\tau^j} \tag{2}$$

Coefficients $a_{ij}$ and $b_{ij}$ are determined on the basis of the experimental data for the thermodynamic properties of compressed gas and liquid.

The equation of state in the form of Eq. (1) was effectively used for an analytical description of the data on thermodynamic properties of a number of substances, including difluoromethane [2, 4, 5].

The equations of state for R32 and R125 used the $p$, $\rho$, $T$-data for gaseous and liquid phases listed in Tables I and II. In addition to the experimental data used in the single-phase region, calculated values of saturated vapor and liquid densities and saturated vapor pressure were also used. The equations for calculating these thermodynamic properties were taken from Ref. 1 for both refrigerants. Use of the calculated values of the properties for the saturation line was required because of the small amount of experimental data for the density of the saturated vapor and because of the necessity to have values of $\rho'$, $\rho''$, and $p_s$ at the same temperatures in order to ensure satisfaction of Maxwell's rule.

For the qualitative representation of caloric properties of difluoromethane, 73 experimental values of the isochoric heat capacity $c_v$ in the temperature range from 153 to 341 K at pressures from 5.2 to 32 MPa and 101 values of the saturated liquid heat capacity $c_s$ in the temperature range 141

**Table I.** List of $p, \rho, T$ Data Used in the Development of the Equation of State for R32 and the Root-Mean-Square Deviations $\delta\rho_{\mathrm{m}}$ of the Experimental Values of Density from the Calculated Results

| Year | Authors | Range of parameters | | Number of points | $\delta\rho$ (%) | $\delta\rho_{\mathrm{m}}$ (%) |
|---|---|---|---|---|---|---|
| | | $T$ (K) | $p$ (MPa) | | | |
| 1993 | Holste [6] | 150–375 | 1.5–72 | 126 | 0.1 | 0.14 |
| 1994 | Defibaugh et al. [7] | 243–373 | 0.3–9.8 | 379 | 0.1 | 0.12 |
| 1996 | Magee [8] | 142–396 | 3.8–35 | 135 | 0.03 | 0.03 |
| 1996 | Zhang et al. [9] | 290–370 | 0.1–6.5 | 81 | 0.1 | 0.10 |
| 1997 | de Vries [3] | 224–433 | 0.02–20.6 | 1143 | 0.05 | 0.08 |
| | | 349–352 | 5.5–5.9 | 6 | 0.5 | 0.09[a] |
| 1995 | Outcalt and McLinden, $\rho''$ [1] | 140–340 | $8\times10^{-5}$–4.6 | 41 | 0.1 | 0.17 |
| 1995 | Outcalt and McLinden, $\rho'$ [1] | 140–340 | $8\times10^{-5}$–4.6 | 41 | 0.05 | 0.03 |

[a] The value of $\delta p_{\mathrm{m}}$ is used for 6 points in the critical region.

to 342 K [15] were included in the set of used data. Satisfaction of Maxwell's rule was ensured by the use of the values of $\rho'$, $\rho''$, and $p_s$ from the set of $p, \rho, T$-data for the saturation line given at 41 temperatures in the temperature range 140 to 340 K.

The equation of state for R125 used 99 experimental values of heat capacity $c_v$ in the temperature range from 200 to 342 K at pressure from

**Table II.** List of $p, \rho, T$ Data used in the Development of the Equation of State for R125 and the Root-Mean-Square Deviations $\delta\rho_{\mathrm{m}}$ of the Experimental Values of Density from the Calculated Results

| Year | Authors | Range of parameters | | Number of points | $\delta\rho$ (%) | $\delta\rho_{\mathrm{m}}$ (%) |
|---|---|---|---|---|---|---|
| | | $T$ (K) | $p$ (MPa) | | | |
| 1991 | Zausaev and Kletsky [10] | 273–443 | 0.6–6.1 | 43 | 0.1 | 0.23 |
| 1992 | Defibaugh et al. [11] | 275–369 | 1.6–6.3 | 150 | 0.1 | 0.16 |
| 1993 | Holste [12] | 180–480 | 0.2–68 | 211 | 0.1 | 0.17 |
| 1995 | Ye et al. [13] | 290–390 | 0.1–3.6 | 93 | 0.1 | 0.14 |
| 1995 | Boyes and Weber [14] | 273–363 | 0.3–4.6 | 80 | 0.05 | 0.05 |
| 1996 | Magee [8] | 178–398 | 3.6–35 | 77 | 0.03 | 0.02 |
| 1997 | de Vries [3] | 243–413 | 0.02–19.8 | 952 | 0.05 | 0.08 |
| | | 339–341 | 3.6–3.8 | 9 | 0.5 | 0.13[a] |
| 1995 | Outcalt and McLinden, $\rho''$ [1] | 180–330 | $6\times10^{-3}$–3.0 | 31 | 0.1 | 0.34 |
| 1995 | Outcalt and McLinden, $\rho'$ [1] | 180–330 | $6\times10^{-3}$–3.0 | 31 | 0.05 | 0.07 |

[a] The value of $\delta p_{\mathrm{m}}$ is used for 9 points in the critical region.

3.8 to 33 MPa and 93 values of heat capacity $c_s$ in the temperature range 176–278 K [15]. To ensure satisfaction of Maxwell's rule, 31 values of $\rho'$, $\rho''$, and $p_s$ in the temperature range from 180 to 330 K were used.

The values of the isobaric heat capacities for the ideal gas which were necessary for the use of the heat capacities $c_v$ and $c_s$ while developing the equation of state were taken as average values between the data [16, 17] for R32 and [1, 18] for R125. These values were approximated by Eq. (3) for R32 and Eq. (4) for R125.

$$c_{p0}/R = 4.3914 - 2.5143\tau + 5.3885\tau^2 - 1.6057\tau^3 \tag{3}$$

$$c_{p0}/R = 2.9072 + 11.5586\tau - 2.1135\tau^2 \tag{4}$$

The weights for the experimental data were assumed to be inversely proportional to the accuracy of the data and calculated according to the method given in Ref. 19. The values of the relative deviations $\delta\rho$ used for the calculation of the weights of $p$, $\rho$, $T$-data are shown in the next-to-last column of Table I for R32 and in Table II for R125. The values of the errors of the data on the heat capacities $c_v$ and $c_s$ and the data used for the satisfaction of Maxwell's rule were assumed to be equal to $\delta c_v = 0.5\%$, $\delta c_s = 0.7\%$, and $\delta p_s = 0.1\%$ for both substances. The accuracy and the mutual agreement of the data and also the amount of information for each property influenced the choice of these values.

A step-wise regression analysis method [20] was used for determining the coefficients of Eq. (2) for both R32 and R125. The method is based on the minimization of the square functional formed on the basis of the approximated data. In this method the number of coefficients used in the equation increases from an initial minimum to optimum by means of adding the statistically most significant terms chosen from the empirical bank of terms. These terms are determined at each stage of the calculations in accordance with the significance level of the Student *t*-criterion. The Fisher criterion is used to determine if the current equation significantly improved the quality of the approximation of the data in comparison with the previous one. If the improvement is significant, a new term is added to the equation. Otherwise the equation is taken as final.

The bank of terms used for both equations of state contained 70 terms in the polynomial part ($m = 10$, $r = 6$) and 50 terms in the exponential part ($n = 10$, $s = 5$). A reliable approximation of the data over a wide range of parameters was ensured by assigning the value 0.05 to the significance level of the Student *t*-criterion and the value 0.25 to the level of the Fisher criterion.

As discussed above, different data are included to determine the coefficients. Therefore, the square functional was complicated by gradually

**Table III.** Coefficients $a_{ij}$ and $b_{ij}$ of the Equation of State, Eq. (2), for R32

| $i$ | $j$ | $a_{ij}$ | $i$ | $j$ | $b_{ij}$ |
|---|---|---|---|---|---|
| 1 | 0 | $1.183486$ | 1 | 4 | $-6.546357 \times 10^{-2}$ |
| 1 | 1 | $-2.430934$ | 1 | 5 | $-2.784785 \times 10^{-1}$ |
| 1 | 4 | $-1.472179 \times 10^{-2}$ | 2 | 1 | $1.113400$ |
| 2 | 0 | $-4.506743 \times 10^{-1}$ | 2 | 2 | $-2.954417$ |
| 2 | 1 | $1.721527$ | 2 | 4 | $4.898234$ |
| 2 | 2 | $-1.349166$ | 2 | 5 | $-2.354906$ |
| 3 | 1 | $-6.052212 \times 10^{-1}$ | 3 | 5 | $-7.709682 \times 10^{-1}$ |
| 3 | 2 | $9.265910 \times 10^{-1}$ | 4 | 5 | $6.502963 \times 10^{-1}$ |
| 4 | 0 | $8.081905 \times 10^{-2}$ | 5 | 3 | $2.168338 \times 10^{-1}$ |
| 4 | 2 | $-1.999587 \times 10^{-1}$ | 5 | 4 | $-5.499117 \times 10^{-1}$ |
| 4 | 3 | $3.655934 \times 10^{-3}$ | 6 | 3 | $1.978099 \times 10^{-2}$ |
| 5 | 2 | $8.217181 \times 10^{-3}$ | 6 | 5 | $9.535163 \times 10^{-2}$ |
| 6 | 0 | $-3.230880 \times 10^{-3}$ | 8 | 4 | $-1.425744 \times 10^{-2}$ |
| 6 | 1 | $5.778584 \times 10^{-3}$ | 9 | 2 | $3.921874 \times 10^{-3}$ |
| 10 | 0 | $-2.536027 \times 10^{-6}$ | | | |

including $p, \rho, T$-data for the single-phase region and for the saturation line, data ensuring satisfaction of Maxwell's rule and data on heat capacities $c_v$ and $c_s$. This method allows one to estimate mutual agreement between different data and to find better assigned values of the relative errors used for calculating weights.

The coefficients of the final equations of state were obtained by taking into account all of these properties and are given in Tables III and IV. During the calculations, the following values of the critical parameters and gas constants were assumed

For R32 $T_c = 351.35\ \mathrm{K}$ [1]; $\rho_c = 427.00\ \mathrm{kg \cdot m^{-3}}$ [1]

$\mathrm{R} = 159.821\ \mathrm{J \cdot kg^{-1} \cdot K^{-1}}$

For R125 $T_c = 339.33\ \mathrm{K}$ [1]; $\rho_c = 571.29\ \mathrm{kg \cdot m^{-3}}$ [1]

$\mathrm{R} = 69.275\ \mathrm{J \cdot kg^{-1} \cdot K^{-1}}$

## 3. COMPARISONS WITH EXPERIMENTAL DATA

The results of the comparisons of the initial experimental data for both substances with the values calculated by means of the equations of state are shown in the last column of Table I for R32 and Table II for R125 where the root-mean-square deviations $\delta\rho_{\mathrm{m}}$ for each author are shown. It should be noted that for six points from Ref. 3 for R32 and nine points from the

**Table IV.** Coefficients $a_{ij}$ and $b_{ij}$ of the Equation of State, Eq. (2), for R125

| $i$ | $j$ | $a_{ij}$ | $i$ | $j$ | $b_{ij}$ |
|---|---|---|---|---|---|
| 1 | 0 | $2.825627 \times 10^{-1}$ | 1 | 4 | $1.203782 \times 10^{-1}$ |
| 1 | 1 | $-6.854910 \times 10^{-1}$ | 2 | 1 | $1.687868 \times 10$ |
| 1 | 3 | $-9.975127 \times 10^{-1}$ | 2 | 2 | $-6.643158 \times 10$ |
| 2 | 1 | $6.445149 \times 10^{-1}$ | 2 | 3 | $9.473412 \times 10$ |
| 2 | 3 | $4.024926 \times 10^{-1}$ | 2 | 4 | $-5.988361 \times 10$ |
| 3 | 1 | $-7.060325 \times 10^{-1}$ | 2 | 5 | $1.396914 \times 10$ |
| 4 | 2 | $2.621465 \times 10^{-1}$ | 3 | 2 | $6.765262 \times 10^{-1}$ |
| 5 | 0 | $1.118047 \times 10^{-1}$ | 4 | 1 | $-5.880934 \times 10^{-2}$ |
| 5 | 2 | $-1.217283 \times 10^{-1}$ | 4 | 3 | $-5.648372 \times 10^{-1}$ |
| 6 | 0 | $-3.381068 \times 10^{-2}$ | 5 | 4 | $1.643516 \times 10^{-1}$ |
| 7 | 1 | $7.828134 \times 10^{-3}$ | 6 | 1 | $6.900750 \times 10^{-1}$ |
| 7 | 2 | $2.074012 \times 10^{-3}$ | 6 | 2 | $-1.066543$ |
| 7 | 4 | $-5.912590 \times 10^{-5}$ | 6 | 4 | $3.699827 \times 10^{-1}$ |
| 8 | 3 | $8.017074 \times 10^{-5}$ | 7 | 5 | $-4.472888 \times 10^{-2}$ |
| 10 | 1 | $-5.547434 \times 10^{-5}$ | 10 | 1 | $6.506423 \times 10^{-3}$ |
|  |  |  | 10 | 3 | $-8.564364 \times 10^{-3}$ |

same work for R125 which are in the critical region, where the values of the derivative $\partial\rho/\partial p$ are very large, values of root-mean-square deviations for pressure $\delta p_{\mathrm{m}}$ are given in Tables I and II. For the comparisons with the data for the saturation line, the calculated values of density and of saturated vapor pressure were determined using Maxwell's rule.

The histograms of deviations $\delta\rho$ for R32 and R125 were built using the results of the calculations. They are shown in Fig. 1a and b. It can be seen from the figure that the distribution of the deviations is close to normal. While building the histograms, six points for R32 and nine points for R125 lie in the critical region and have deviations $\delta\rho$ larger than 1% (while deviations of $\delta p$ are 0.03 to 0.24%), and these deviations were not taken into account. There are 18 points for R32 and 16 points for R125 that have deviations with absolute values in the range 0.5 to 1% and are not included in the histograms but were taken into account for calculating the values $\delta\rho_{\mathrm{m}} = 0.10\%$ (for 1946 points) and 0.12% (for 1668 points).

For R32, additional comparisons with $p$, $\rho$, $T$-data [21, 22] that were not used in developing the equation of state showed that the corresponding values of the root-mean-square deviations $\delta\rho_{\mathrm{m}}$ are 0.24% and 0.13% (for 123 points in the temperature range 243 to 373 K and pressure range 0.1 to 4.8 MPa and for 95 points at 290 to 370 K and 0.1 to 6.5 MPa, respectively).

The root-mean-square deviations of the initial values of the saturated vapor pressure [1] and the experimental data on the heat capacities $c_v$ and

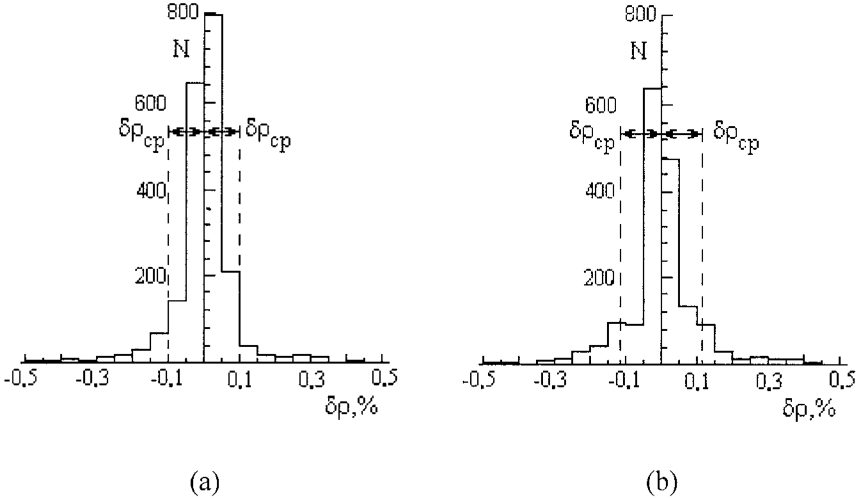

**Fig. 1.** Histograms of the deviations of $\delta\rho$ for R32 (a) and R125 (b).

$c_s$ [15] from the values calculated from the equation of state for R32 are equal to 0.16%, 0.31%, and 0.85%, respectively. Taking into account the errors of these data, the deviations can be considered to be quite satisfactory. It should also be noted that after exclusion of the value −0.72% at the minimum temperature of 140 K where the vapor pressure is small (82 Pa), the value $\delta p_{s\mathrm{m}}$ decreases to 0.12%.

For R32, the experimental data for the saturated vapor pressure [7, 8, 15, 22–28] and the densities of the saturated vapor [7, 24, 28] and liquid [7, 8, 24, 26, 28] are also compared with the values calculated by means of the equation of state. These data cover the temperature range 140 to 351 K for $p_s$, 219 to 345 K for $\rho''$, and 139 to 346 K for $\rho'$ and the numbers of points in the corresponding sets are 327, 55, and 87, respectively. For the majority of the authors, the root-mean-square deviations of data on pressure $p_s$ and density $\rho'$ from the values calculated from the equation of state on the basis of Maxwell's rule are not larger than 0.2%. For the data in Refs. 15 and 23 given in the temperature ranges 140 to 180 K and 149 to 245 K, the values of $\delta p_s$ are 1.36% and 0.92%, respectively. This is caused by big relative errors of the experimental data in the region of low pressure. It should be noted that deviations of the data [15, 23] from the calculations have different signs. The majority of the data for the density of saturated vapor has more significant deviations than data for the density of saturated liquid. The values of $\delta\rho''_{\mathrm{m}}$ lie mainly in the range 0.7 to 1% except for data from Ref. 7 $\delta\rho''_{\mathrm{m}} = 0.13\%$. The uncertainty of the experimental values of $\rho''$ is as a rule larger than of the values of $\rho'$.

For the new experimental data for the saturated vapor pressure [3] covering the range $T = 223$ to 351 K (139 points), the value of $\delta p_{sm}$ is equal to 0.04%. Comparison with values of densities of saturated liquid and vapor obtained by extrapolation of the $p$, $v$, $T$-data in Ref. 3 to the saturation line showed that the root-mean-square deviations for 41 points in the temperature range 140 to 340 K are $\delta\rho'_{\mathrm{m}} = 0.04\%$ and $\delta\rho'' = 0.38\%$.

A comparison of calculated values of speed of sound for R32 with experimental data [29, 30] was made. For 120 points [29] covering the temperature range 243 to 373 K and pressure range 0.27 to 33 MPa, the root-mean-square deviation $\delta w_{\mathrm{m}}$ is equal to 0.56%. For 305 points [30] in the temperature range 248 to 343 K and pressure range 1.7 to 65 MPa, the value of $\delta w_{\mathrm{m}}$ is 0.70%.

For R125, additional comparisons with $p$, $\rho$, $T$-data [31, 32] showed that the corresponding values of the root-mean-square deviations $\delta\rho_{\mathrm{m}}$ are 0.26% and 0.43% (for the total amount of 56 points in the temperature range 383 to 424 K and pressure range 0.9 to 11 MPa). Such values of deviations are caused by the disagreement of data [31, 32] with those used in the development of the equation of state.

The root-mean-square deviations of the initial values of the saturated vapor pressure [1] and experimental data on the heat capacities $c_v$ and $c_s$ [15] from the values calculated by means of the equation of state for R125 are equal to 0.07%, 0.45%, and 0.29%, respectively.

For R125, experimental data for the saturated vapor pressure [8, 12–15, 26–28, 31–33] and the densities of the saturated vapor [14] and liquid [8, 11, 26, 28] are also compared with the calculated values. These data cover the temperature range 180 to 338 K for $p_s$, 275 to 320 K for $\rho''$, and 173 to 336 K for $\rho'$ and the numbers of corresponding points are 393, 10, and 54, respectively. For the vast majority of authors, the root-mean-square deviations of data for pressure $p_s$ and density $\rho'$ from the calculated values are not larger than 0.2% and 0.1%, respectively. The value of $\delta\rho''_{\mathrm{m}}$ for data from Ref. 14 is 0.32%.

For the experimental saturated vapor pressure data [3] at temperatures 222 to 339 K (98 points), the value of $\delta p_{sm}$ is 0.08%. Comparison with values of $\rho'$ and $\rho''$ obtained by extrapolation of the $p$, $v$, $T$-data [3] to the saturation line showed that the root-mean-square deviations for 31 points at temperatures from 180 to 330 K are $\delta\rho'_{\mathrm{m}} = 0.02\%$ and $\delta\rho'' = 0.31\%$.

A comparison of calculated values of speed of sound for R125 with experimental data [34, 35] showed that for 72 points [34] in the gaseous phase covering the temperature range 273 to 343 K and pressure range 0.01 to 0.24 MPa, the root-mean-square deviation $\delta w_{\mathrm{m}}$ is equal to 0.07%. For 167 points [35] in the liquid phase at temperatures 241 to 333 K and pressures 0.32 to 32 MPa, the value of $\delta w_{\mathrm{m}}$ is 0.54%.

After compiling the equations of state for R32 and R125, we received the monograph [36] where analogous equations are published. Comparisons showed that the equations of state in Ref. 36 are very similar to our equations, with ours being slightly better. Either equation would be satisfactory for any technical application.

## 4. CONCLUSION

The values of thermal, caloric, and acoustic properties of R32 and R125 calculated with the developed equations of state were compared with the experimental data. The detailed comparisons showed that these equations describe the most reliable experimental data with high precision. Therefore, the new equations of state may be used for calculation of thermodynamic properties of gaseous and liquid difluoromethane and pentafluoroethane in a wide region of temperatures and pressures.

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