UNIQUE NONANALYTIC EQUATION OF STATE OF THE REFRIGERANT R218

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A unique equation of state of the refrigerant R218 is derived which adequately describes a thermodynamic surface in the region of small pressure densities and in the vicinity of the critical point.

Presently, both analytic and nonanalytic equations of state (ES) are widely used to describe the thermodynamic properties of fluids.

Deriving unique analytic equations of state involves the complicated problem of describing a set of thermodynamic properties with the accuracy of initial experimental data for densities ranging from that of an ideal-gas state to that of a liquid at the triple point and for temperatures ranging from the triple point to the Boyle temperature and higher. The analytic ES mainly successfully solve this problem: within a large region of the thermodynamic surface, referred to as regular, they define the thermodynamic functions of the gas and liquid to an accuracy corresponding to that of state-of-the-art experimental studies [1, 2]. However, there is a drawback inherent in the equations of state indicated, i.e., they are incapable of qualitatively and correctly describing the abnormal behavior of a substance in the critical region, especially in the immediate vicinity of the critical point. In this connection, as a line of the liquid-vapor phase equilibrium in a wide neighborhood of the critical point is being approached, an error of the analytic equations increases sharply, going far beyond the allowable limits. Specially performed investigations [3] succeeded in solving the problem of the qualitative estimation of an inoperative region of the unique analytic ES.

An inability of the analytic equations to describe specific features of the critical region prompted the development of methods for constructing nonanalytic ES. Nonanalytic equations include those in which, in order to take into account the characteristics of the near-critical region, use is made of the terms with nonintegral indices which provide a singularity of the relevant thermodynamic functions.

The problem of devising wide-range methods for describing the thermodynamic properties of gases and fluids which satisfy relations of the scaling theory (ST) of critical phenomena was treated in [4-17]. Those studies worked out effective methods of selecting structural forms of the internal energy and of Helmholtz free energy (in the density-temperature coordinates) as well as of the chemical potential (in the pressure-temperature coordinates) which reproduce the ST power laws and, simultaneously, identify singularities of the thermodynamic surface in the rarefied gas region.

Among the above-noted approaches [1-14], one of the most promising is that proposed in [7-9]. Within the framework of this approach, a unique nonanalytic equation of state is constructed which describes, in accordance with the requirements of the modern theory of critical phenomena [13], a wide vicinity of the critical point of the liquid-gas system and, moreover, ensures that the requirement of equality of the chemical potentials is fulfilled

$$\mu'(T) = \mu''(T), \qquad (1)$$

and that the values of the density, pressure, and temperature on the discrete set of points of the coexistence curve are in agreement. Study [10] revealed that the problem in question is solved most successfully based on a single-structural form of the Helmholtz free energy given by

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$$\frac{F(\rho,\ T)}{RT} = \ln\rho + \frac{F_0(T)}{RT} + \omega \sum_{i=0}^{m_1} \sum_{j=0}^{j_1(i)} C_{ij}\tau_1^j \Delta\rho^i + + f(\omega) \sum_{i=0}^{m_0} u_i\, |\Delta\rho|^{\delta+1+\frac{\Delta_i}{\beta}} a_i(x), \tag{2}$$

where $F_0(T)$ is the ideal-gas component of the free energy, $\Delta_0 = 0$, and $\Delta_1 = \Delta$.

The scale functions $a_i(x)$ that enter into expression (2) can be chosen both in the parametric form of the ST representation and in the physical density-temperature variables. In the latter case, the functions $a_i(x)$ have the form [14]

$$\begin{gathered}
a_0(x) = A_1\left[(x+x_1)^{2-\alpha} - \frac{x_1}{x_2}(x+x_2)^{2-\alpha}\right] + B_1(x+x_3)^{\gamma} + C_1, \\
a_1(x) = A_2\left[(x+x_4)^{2-\alpha+\Delta} - \frac{x_4}{x_5}(x+x_5)^{2-\alpha+\Delta}\right] + B_2(x+x_6)^{\gamma+\Delta} + C_2, \\
a_2(x) = A_3\left[(x+x_7)^{2-\alpha+\Delta_2} - \frac{x_7}{x_8}(x+x_8)^{2-\alpha+\Delta_2}\right] + C_3, \\
a_3(x) = A_4(x+x_9)^{\gamma+\Delta_3} + C_4, \\
a_4(x) = A_5[(x+x_{10})^{\gamma+\Delta_4} - (x+x_{11})^{\gamma+\Delta_4}] + C_5,
\end{gathered} \tag{3}$$

where $\Delta_2 = \gamma - \alpha$, $\Delta_3 = \Delta_4 = \gamma+\beta - 1$, and $A_3 = A_4 = A_5 = 1$, and the coefficients A_1 and A_2 are defined in terms of the constants of the parametric equations of state of the ST:

$$\begin{gathered}
A_1 = -\frac{k\gamma(\gamma-1)}{2\alpha b^2(2-\alpha)(1-\alpha)(1-x_1/x_2)}, \\
A_2 = -\frac{k(\gamma+\Delta)}{2(2-\alpha+\Delta)(1-\alpha+\Delta)\, b^2(1-x_4/x_5)}, \quad B_1 = B_2 = 1/2k.
\end{gathered} \tag{4}$$

The constants C_i entering into the scaling functions (3) are determined from the equality $(2 - \alpha+\Delta_i)a_i(-x_0) + x_0 a_i'(-x_0) = 0$, where $-x_0$ is the value of the variable x on the coexistence curve, $k = [(b^2 - 1)]/x_0]^{\beta}$ and $b^2 = (\gamma - 2\beta)/[\gamma - 2\beta)]$. The functions $a_0(x)$ and $a_1(x)$ are "responsible" for the representation of asymptotic and nonasymptotic terms of the thermodynamic function in accordance with the ST requirements. The functions $a_2(x)$, $a_3(x)$, and $a_4(x)$ allow one to account for the difference of the real liquid-vapor system from a lattice-gas model.

In view of the foregoing, when constructing the unique ES of the refrigerant R218 which meets the requirements of the fluctuation theory of critical phenomena, expression (2) with the scaling functions (3) is used as the initial thermodynamic function.

The refrigerant R218 is chosen as the object of the study for the following reasons. First, the equation of state of R218 describing its equilibrium properties in the near-critical region is lacking until now. At the same time, there is a sufficient collection of precise experimental data on r^*, p-v-T, ρ', ρ'', C_p, and C_v obtained in a wide range of the state parameters, including the vicinity of the critical point [18-28]. Second, the known analytic ES of R218 [19, 20, 22, 29-31] were constructed disregarding the experimental data on r^* and C_v in a wide range of the state parameters.

Study [3] showed that the analytic equations of state neither qualitatively nor quantitatively describe the thermodynamic properties of pure materials in the near-critical region.

Third, a reliable bulk of the experimental data pertaining to the various properties of the refrigerant R218 in the near-critical region and the accurate thermodynamic tables describing the regular part of the thermodynamic surface, including both the rarefied gas and the dense liquid regions $0.01 \leq \omega \leq 3.1$, allow a dependable evaluation of the range of applicability of the unique nonanalytic ES set up on the basis of expression (2).

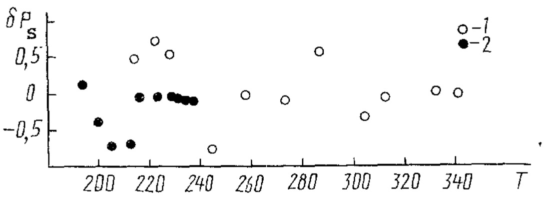

Fig. 1. Deviations of values of the saturation pressure P_s, calculated from the equation of state of the present study from experimental data: 1) [19]; 2) [20]. δP_s, %; T, K.

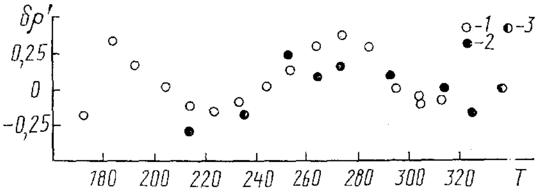

Fig. 2. Deviations of values of the saturated liquid density ρ' calculated from the equation of state of the present study from experimental data: 1) [20]; 2) [18]; 3) [22]. $\delta\rho'$, %.

A thermal ES computed in terms of the free energy (2) from the formula $P = \rho^2(\partial F/\partial\rho)_T$ has the form

$$\begin{gathered} Z = 1 + \omega \sum_{i=0}^{m_1} \Bigl(\sum_{j=0}^{j_1(i)} C_{ij}\tau_1^j \Delta\rho^i (i\omega + \Delta\rho) \Bigr) + \\ + \omega \sum_{i=0}^{4} u_i |\Delta\rho|^{\delta + \frac{\Delta_i}{\beta}} \left[\frac{f'(\omega)}{\beta} \operatorname{sign}(\Delta\rho) h_i(x) + f'(\omega) a_i(x) \right], \end{gathered} \tag{5}$$

where $Z = P/(\rho RT)$; $h_i(x) = (2 - \alpha + \Delta_i)a_i(x) - x a_i'(x)$; $f'(\omega) = df/d\omega$; $f(\omega) = [(1 - \omega)^2 - 2]^{n0}$.

In order for the equation of state, Eq. (5), to satisfy the ST power laws, the critical conditions must be fulfilled

$$\left(\frac{\partial^n P}{\partial \rho^n} \right)_T \bigg|_{T=T_c,\ v=v_c} = 0, \qquad \left(\frac{\partial P}{\partial \rho} \right)_T \bigg|_{T\to T_c,\ v\to v_c} \sim |\tau|^\gamma,$$

where n = 1-4.

Taking Eq. (6) into account, let us transform the regular component of the ES (5):

$$\begin{gathered} Z = 1 + \omega y_1 + \omega^2 y_2 + (y_3 + \omega y_4) C_{01}\omega + (y_5 + \omega y_0) C_{20}\omega + \\ + \sum_{i=6}^{m_1} C_{i0}\Delta\rho^{i-1}(i\omega + \Delta\rho) + \omega\tau_1(2\omega - 3)C_{11} + \omega^2\tau_1(3\omega - 4)C_{21} + \\ + \omega\tau_1 \sum_{i=3}^{m_1} C_{i1}\Delta\rho^{i-1}(i\omega + \Delta\rho) + \omega \sum_{i=0}^{m_1} \sum_{j=2}^{j_1(i)} C_{ij}\tau_1^j \Delta\rho^{i-1}(i\omega + \Delta\rho) + \omega \sum_{i=0}^{4} u_i |\Delta\rho|^{\delta + \frac{\Delta_i}{\beta}} \left[\frac{f(\omega)}{\beta} \operatorname{sign}(\Delta\rho) h_i(x) + f'(\omega) a_i(x) \right], \end{gathered} \tag{7}$$

where $y_1 = dy_2/dw$, $y_3 = dy_4/dw$, and $y_5 = dy_6/dw$, whereas the functions y_2, y_4, and y_6 are defined by the relations

$$y_1 = -\frac{15{,}4}{12} + \frac{5{,}8}{12}\Delta\rho - \frac{1{,}1}{6}\Delta\rho^2 + 0{,}05\Delta\rho^3,$$

$$y_3 = 5 - 4\Delta\rho + 3\Delta\rho^2 - 2\Delta\rho^3 + \Delta\rho^4,$$

$$y_5 = 4 - 3\Delta\rho + 2\Delta\rho^2 - \Delta\rho^3 + \Delta\rho^5.$$

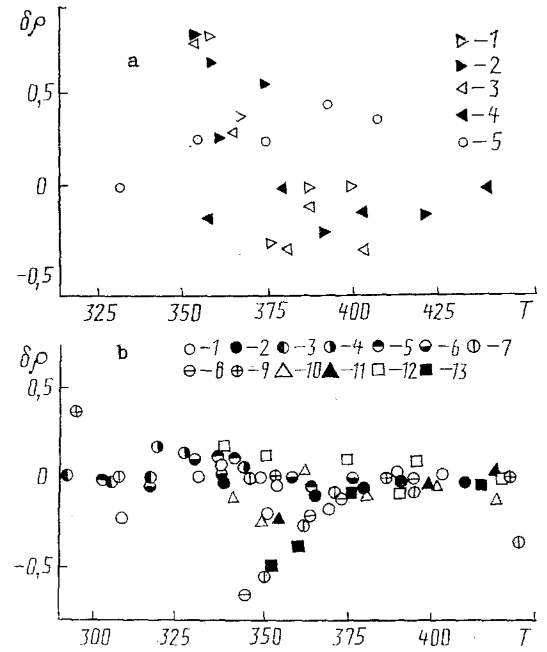

Fig. 3. Deviations of values of the density ρ in a single-phase state calculated from the equation of state of the present study from experimental data [19] (a) and [25] (b): a – 1) ρ = 618.8 kg/m^3; 2) 479.9; 3) 564.6; 4) 375.3; 5) 187.8 kg/m^3; b – 1) ρ = 1136 kg/m^3; 2) 1104; 3) 1377; 4) 1325; 5) 1280; 6) 1220; 7) 171.0; 8) 296.5; 9) 64.7; 10) 927.5; 11) 883; 12) 1003; 13) 706 kg/m^3. $\delta\rho$, %.

When $n_0 \geq 2$, expression Eq. (7) in the region $\rho \to 0$ and $P \to 0$ goes over into the ES of an ideal gas

$$Z|_{\rho\to 0} = 1. \tag{8}$$

For the case $\Delta\rho \to 0$ and $\tau \to 0$, Eq. (7) goes over into a scaling ES in physical variables [14] which reproduce the power laws that follow from the fluctuation theory of critical phenomena.

In constructing the unique ES satisfying the ST requirements, along with the p-v-T data, the experimental data on C_v [4] should be included in the initial body of experimental data from which the coefficients of the equation of state are determined. The expression for the specific heat at constant volume computed from Eq. (2) by the formula $C_v = -T(\partial^2F/\partial T^2)_V$ has the form

$$\begin{aligned} C_v = &-R\omega \sum_{i=0}^{m_1} \Big(\sum_{j=2}^{j_1(i)} C_{ij} j (j-1) t^{-2} \tau_1^{j-2} \Delta\rho^i \Big) - \\ &- RTf(\omega) \sum_{i=0}^{4} u_i a_i''(x) |\Delta\rho|^{\frac{-\alpha+\Delta_i}{\beta}} + C_v^0(T), \end{aligned} \tag{9}$$

where $C_v^0(T)$ is the ideal-gas component of the specific heat at constant volume.

The coefficients of the equation of state (7) are found from the condition of the minimum of the functional

$$\Phi = \Phi_p + \Phi_{C_v} + \Phi_{p_s}. \tag{10}$$

Here, Φ_p, Φ_{C_V}, and Φ_{p_s} are the terms of the functional being minimized which account for the thermal characteristics and the specific heat at constant volume in a single-phase region as well as the pressure and density on the saturation line, respectively.

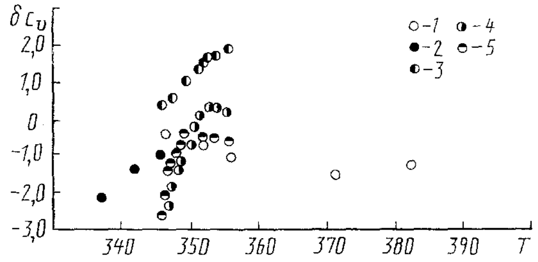

Fig. 4. Deviations of values of C_v calculated from the equation of state of the present study from experimental data [23] [1) ρ = 377.1 kg/m^3; 2) 995.0] and [27] [3) 806.5 kg/m^3; 4) 685.7; 5) 503.8]. δC_v, %.

Inasmuch as, for R218, the collection of data on the density ρ'' on the vapor branch of the coexistence curve is limited [22], along with the experimental values use is also made of the calculated density values obtained based on experimental data for "apparent" evaporation heat [23] from the equation

$$\frac{1}{\rho''} = \frac{r^*}{P_S' T}, \tag{11}$$

where $P_s' = dP_s/dT$, and r^* is defined by the expression

$$r^* = \frac{P_c}{\rho_c}(d_1 + d_2|\tau|^{\beta} + d_3|\tau|^{\beta+\Delta} + d_4|\tau|^{1-\alpha} + d_5\tau^2). \tag{12}$$

The elasticity line $P_s(T)$, whose expression enters in Eq. (11), is described by the equation

$$P_S = P_c \exp(-f_0/t\tau^2)(1 + f_1\tau + f_2|\tau|^{2-\alpha} + f_3|\tau|^{2-\alpha+\Delta} + f_4\tau^2). \tag{13}$$

The constants entering into Eq. (12) were determined from the experimental data regarding the density ρ'' [22] and the evaporation heat r^* [26]. The coefficients of Eqs. (11)-(13) were found by the method of least squares (MLS) and, in the case of Eq. (11), from experimental data [19-23,26]. As result, the following values are obtained: $f_1 = d_1 = 7.61651$, $f_2 = 32.6890$, $f_3 = -41.2878$, $f_4 = -15.7713$, $d_2 = 14.5618$, $d_3 = 108.065$, $d_4 = -93.3097$, $d_5 = 0.138159$, $\beta = 0.321$, $\gamma = 1.24$, $\Delta = 0.5$, $T_c = 0.138159$, $T_c = 345.05$ K, $P_c = 2.707$ MPa, and $\rho_c = 628$ kg/m^3.The mean-square deviations of the values computed from Eqs. (11)-(13) from the experimental data [19-23, 26] in the temperature range 190-344 K constituted $\delta r^* = 0.34\%$ for the "apparent" evaporation heat and $\delta P_s = 0.23\%$ for the pressure on the saturation line.

The proposed approach permits describing the vapor branch of the coexistence curve and ensuring the agreement of the values of ρ'', r^*, P_s', and P for temperatures ranging from $T_{tr.p}$ to T_c (Figs. 1, 2).

The ideal-gas components of the thermodynamic functions of R218 are calculated from the equation [32] for the ideal-gas component of the specific heat at constant pressure

$$C_p^0 = 0{,}1170 - 2{,}908 \cdot 10^{-3} T - 2{,}226 \cdot 10^{-6} T^2, \quad \text{kJ/(kg·K)}. \tag{14}$$

To determine the coefficients of the unique nonanalytic ES (7) by the MLS based on the experimental data [19-28], use was made of a singular expansion procedure which enables one to efficiently take account of the rounding errors and of the initial information [33]. Here, the parameters of Eq. (7) take on the following values: R = 44.221 J/(kg·K),

$$u_0 = 6{,}31816330291, \quad u_1 = 0{,}148495725730, \quad u_2 = -0{,}450306666721,$$
$$u_3 = 3{,}50136562801, \quad u_4 = 14{,}2098681944, \quad C_{10} = 0{,}0224627237173,$$
$$C_{20} = 0{,}0562490433606, \quad C_{60} = -0{,}161194693556, \quad C_{70} = 0{,}0549098357091,$$

$$C_{80} = 0{,}534145902917, \quad C_{90} = -0{,}487142212748, \quad C_{10,0} = -0{,}267986696661,$$

$$C_{11,0} = 0{,}395206010446, \quad C_{12,0} = -0{,}059050995026, \quad C_{11} = 0{,}630049612964,$$

$$C_{21} = 1{,}22674700462, \quad C_{31} = -1{,}86258073185, \quad C_{41} = -0{,}920140426292,$$

$$C_{51} = 2{,}07911122684, \quad C_{61} = -2{,}48067715977, \quad C_{71} = 1{,}10431045890,$$

$$C_{81} = 2{,}42241102233, \quad C_{91} = -2{,}55266247167, \quad C_{10,1} = 0{,}589351124415,$$

$$C_{11,1} = 0{,}0745160471754, \quad C_{12,1} = -0{,}026628238323, \quad C_{02} = 3{,}93560539238,$$

$$C_{03} = -11{,}8536218855, \quad C_{12} = -3{,}6394955802, \quad C_{13} = 10{,}5007725892,$$

$$C_{14} = -13{,}1781973408, \quad C_{22} = -16{,}7133147460, \quad C_{23} = 20{,}9569739053,$$

$$C_{24} = 12{,}3104811269, \quad C_{32} = 17{,}0771440228, \quad C_{33} = -17{,}2833118701,$$

$$C_{34} = -2{,}85177881914, \quad C_{35} = 12{,}2593198229, \quad C_{43} = -20{,}3045785030,$$

$$C_{52} = -12{,}1967276713, \quad C_{53} = 4{,}29285829479, \quad C_{62} = -2{,}99230926065,$$

$$C_{63} = 37{,}5925438758, \quad C_{72} = -0{,}932654043439, \quad C_{73} = -18{,}1544644111,$$

$$C_{82} = 2{,}18289603260, \quad C_{83} = -23{,}4733713450, \quad C_{92} = 2{,}79166635271,$$

$$C_{93} = 27{,}7752156754, \quad C_{10,2} = -2{,}68708075850, \quad C_{10,3} = -11{,}9176513857,$$

$$C_{11,2} = 0{,}847048226592, \quad C_{11,3} = 2{,}40680158132,$$

$$C_{12,2} = -0{,}0829789676672, \quad C_{12,3} = -0{,}192256177589.$$

The root-mean-square deviations of the values predicted by the equation of state (7) from the experimental data [21-28] were $\delta\rho = 0.25\%$ for the density in the single-phase region (Fig. 3), $\delta C_V = 0.6\%$ for the specific heat at constant volume (Fig. 4), $\delta\rho' = 0.34\%$ for the density on the saturation line, and $\delta P_s = 0.27\%$ for the pressure on the saturation line.

The maximum deviations were $\delta\rho^{\max} = 1.1\%$ for the density and $\delta C_V{}^{\max} = 2.8\%$ for the specific heat at constant volume.

The root-mean-square deviation of $\delta\mu'(T)$ from $\mu''(T)$ in the temperature range $190 \le T < T_c$ was equal to 0.09%.

A comparison of the values of s, h, w, C_p, C_v, K_T, and ρ with the corresponding data tabulated in [19, 22, 24] revealed a satisfactory agreement in the range of the state parameters $0.1 \le P \le 50$ MPa and $190 \le T \le 440$ K (here, the density varied within $0.01 \le \omega < 3.1$) in the regular part of the thermodynamic surface. At the same time, in a wide region about the critical point, the maximum deviations of the thermodynamic functions computed from the ES (7) from the tabulated values [24] comprised $\delta\rho = 9\%$ for the density in a one-phase state, $\delta\rho'' = 6\%$ for the density on the saturation line, $\delta C_V = 11\%$ for the specific heat at constant volume, and $\delta C_p = 16\%$ for the specific heat at constant pressure.

NOTATION

T, absolute temperature; P, pressure; ρ, density; $\omega = \rho/\rho_c$; $t = T/T_c$; $\tau = 1 - t$; $\tau_1 = 1 - t^{-1}$; P_c, ρ_c, T_c, critical parameters; R, gas constant; μ, chemical potential; ρ'', ρ', densities of the saturated vapor and liquid, respectively; P_s, pressure on the saturation line; C_v, C_p, specific heat at constant volume and at constant pressure, respectively; F_0, Helmholtz free energy; $F_0(T)$, $C_v{}^0(T)$, $C_p{}^0(T)$, ideal-gas functions; $a_i(x)$, $h_i(x)$, $f_i(x)$, scaling functions of free energy, chemical potential, and specific heat at constant volume, respectively; s, entropy, K_T, isothermal compressibility; w, speed of sound; r^*, "apparent" evaporation heat; α, β, γ, δ, Δ, critical exponents; $v_c = 1/\rho_c$; $\Delta\rho = (\rho - \rho_c)/\rho_c$.

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