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METHOD OF CONSTRUCTING A SINGLE EQUATION OF STATE SATISFYING THE REQUIREMENTS OF THE SCALING HYPOTHESIS

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We present an equation of state which quantitatively and qualitatively describes correctly the regular part of the thermodynamic surface and also the region near the critical point.

By a single equation of state, we mean a single structural form which within some given small error describes the experimental thermal and caloric data in the gas and liquid phases, and also on the liquid—vapor coexistence curve for temperatures ranging from T_3 up to T_c, and which describes correctly all of the features of the behavior of the material over this temperature range [1].

Among the important features are the following [2]:

1) The limit $\rho \to 0$ and $p \to 0$ (the equation of state of an ideal gas)

$$p(\rho \to 0,\ T) = R\rho T, \tag{1}$$

2) the equality of the chemical potentials on both branches of the liquid—vapor coexistence curve

$$\mu'' - \mu' = 0, \tag{2}$$

3) the Planck—Gibbs rule

$$\left(\frac{dp_n}{dT}\right)_{T=T_c} = \left(\frac{\partial p}{\partial T}\right)_{v=v_c,\, T=T_c}, \tag{3}$$

4) the critical condition

$$\left(\frac{\partial p}{\partial v}\right)_{v=v_c,\, T=T_c} = \left(\frac{\partial^2 p}{\partial v^2}\right)_{v=v_c,\, T=T_c} = 0. \tag{4}$$

In addition to these characteristics it is important to note the singularities of the thermodynamic surface near the critical point. The modern view, based on accurate experimental research and the scaling theory of critical phenomena, holds that the behavior of the

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thermodynamic functions near the critical point (for pure materials) is singular in nature and can be described by power laws [3]:

on the critical isotherm

$$\Delta p = D(\Delta\rho)\,|\Delta\rho|^{\delta-1}, \tag{5}$$

on the coexistence curve

$$\Delta\rho = \pm B|\tau|^{\beta}, \tag{6}$$

$$C_v = A_0^-\,|\Delta\rho|^{-\alpha/\beta},\ C_p \sim K_T = \Gamma_0^-|\Delta\rho|^{-\gamma/\beta}, \tag{7}$$

on the critical isochore and elasticity line

$$\frac{d^2 p_{\mathrm{ci,\,n}}}{dT^2} = P^{+}\,|\tau|^{-\alpha}, \tag{8}$$

$$C_v = A^{\pm}\,|\tau|^{-\alpha},\ C_p \sim K_T = \Gamma^{\pm}\,|\tau|^{-\gamma}. \tag{9}$$

In the above relations α, β, δ, and γ are the critical exponents which satisfy the Griffiths equalities [3]: $\alpha + 2\beta + \gamma = 2$, $\alpha + \beta(\delta + 1) = 2$.

It is well known that of the above relations (1)-(9), the analytic single equations of state that are normally used give a correct qualitative description only of (1)-(4) [4]. In principle, relations (5)-(9) cannot be satisfied with an analytic description of the thermodynamic surface [5].

In [6-12] the problem of working out a single equation of state free from the insufficiencies present in the analytic equations was considered. The basic approach of [6, 7] is the use of relations coupling the internal energy of the material $u(\rho, T)$ and the thermal parameters:

$$\left(\frac{\partial u}{\partial v}\right)_T = T\left(\frac{\partial p}{\partial T}\right)_v - p. \tag{10}$$

This will guarantee that the equation of state [7] gives the ideal gas limit (1), but it does not correctly reproduce the temperature dependence of the second virial coefficient.

An attempt to obtain a single equation of state using the coordinates p, T and satisfying (5)-(19) was made in [8, 9]. But it is then necessary to "match" the derivatives of the enthalpy $i(p, T)$ on the critical isochore [9], and this is a complicated mathematical problem in the coordinates p, T.

In [10-12] the derivatives $(\partial p/\partial v)_T$ and d^2p/dT^2 are analytic functions on the critical isochore and this does not satisfy (9) for K_T and C_p; furthermore, it contradicts the scaling theory which predicts that d^2p/dT^2 is nonanalytic on the critical isochore (8).

Thus, the problem of constructing a single nonanalytic equation of state satisfying the requirements (5)-(9) of the scaling theory is still of current interest. In the present paper, we solve the problem using a single structural form of the free energy for the liquid and gas phases:

$$F(\rho,\ T) = \int p_{\mathrm{n}}(T_s)\frac{d\rho}{\bar\rho^2} + F_0(\rho,\ T) - \int\left(\frac{\partial F_{\mathrm{n}}(\rho,\ T)}{\partial\rho}\right)_{T=T_s} d\rho - \int\left(\frac{\partial F_{\mathrm{r}}(\rho,\ T)}{\partial\rho}\right)_T \frac{T_s(\rho)}{T}\,d\rho, \tag{11}$$

where $p_n(T_s)$ and $T_s(\rho)$ are the equations of the elasticity line and coexistence curve, respectively; $F_n(\rho, T)$ and $F_r(\rho, T)$ are the nonanalytic and analytic parts of the function $F_0(\rho, T) = F_n(\rho, T) + F_r(\rho, T)$.

We first study how the requirement that the chemical potentials be equal on the coexistence curves $T_s(\rho)$ [relation (2)] can be satisfied in our approach.

We substitute (11) into the thermodynamic relation $\mu(\rho, T) = F(\rho, T) + \rho(\partial F(\rho, T)/\partial\rho)_T$, and then equate the resulting values of $\mu(\rho, T)$ on the right and left branches of the coexistence curve. We then obtain

$$F(\rho', T_s) - F(\rho'', T_s) = p_{\mathrm{n}}(T_s)(v'' - v'). \tag{12}$$

But (12) is identical to (2) [1].

Therefore, the form of the free energy (11) guarantees in principle the equality of the chemical potentials (2) on the elasticity line $p_n(T_s)$ and also the internal consistency of v', v'', and $p_n(T_s)$ at each point on the coexistence curve.

The functions $p_n(T_s)$ and $T_s(\rho)$ are given in the form of [7, 13]:

$$\ln p_{\mathrm{n}}(T_s) = P_{-1}/T_s + P_0 + P_1 T_s + P_2 (T_s - T_{\mathrm{c}})^{2-\alpha} + P_3 T_s^3, \tag{13}$$

$$T_s(\rho) = T_{\mathrm{c}}(1 - D_0 |\Delta\rho|^{1/\beta} + D_1^{\pm} \Delta\rho^{i_1^{\pm}} + D_2^{\pm} \Delta\rho^{i_2^{\pm}} + D_3^{\pm} \Delta\rho^{i_3^{\pm}}), \tag{14}$$

where $1 - D_0 + (-1)^{i_1^+} D_1^+ + (-1)^{i_2^+} D_2^+ + (-1)^{i_3^+} D_3^+ = 0.$

The above forms for $p_n(T_s)$ and $T_s(\rho)$ describe the liquid—vapor phase equilibrium line in correspondence with the requirements of the scaling theory (6), (8), and also have the following physically reasonable forms in the limit of small densities and pressures [7]: $\ln p_n(T_s) = P_{-1}/T_s$, $T_s(\rho \to 0) \sim \rho$.

Using now the method of looking for the singular terms in the thermodynamic functions described above, we find a structural form of the function $F_0(\rho, T)$ entering (11), which guarantees that the scaling relations (5)-(9) and also (3) and (4) will be satisfied, and also qualitatively reproduces the correct behavior at small densities and pressures.

Since close to the critical point the scaling singularities in the function $F(\rho, T)$ are given [according to (5)-(9)] by power laws with nonintegral exponents, we take as a starting point the following functional for the nonregular part of $F_0(\rho, T)$:

$$\frac{F_{\mathrm{n}}(\rho, T)}{RT} = \omega^n \left| A \left(\sum_{i=0}^{n_1} A_i \tau^{\varphi_i} \Delta\rho^{\eta_i} \right)^{\xi_1} + B \left(\sum_{j=0}^{n_2} B_j \tau^{\psi_j} \Delta\rho^{\varepsilon_j} \right)^{\xi_2} \right|, \tag{15}$$

where R is the universal gas constant, $\omega = \rho/\rho_c$; $\varphi_0 < \varphi_1 < \ldots$. $\psi_0 < \psi_1 < \ldots$; $\eta_i = (1 + \delta)/\xi_1 - \varphi_i/\beta$; $\varepsilon_j = (1 + \delta)/\xi_2 - \psi_j/\beta$.

On the other hand, in the region of small densities and pressures, a physically correct expression for the free energy has the form

$$\frac{F_{\mathrm{r}}(\rho, T)}{RT} = \ln \rho + \sum_{i=0}^{n_3} \left(\sum_{j=0}^{n_4(i)} C_{ij} \left(\frac{T_{\mathrm{c}}}{T} - 1 \right)^j \right) \Delta\rho^i, \tag{16}$$

where P_i, $D_i^{\pm}$, A_i, B_j, and C_{ij} are constants.

We find relations between the exponents $\varphi_0, \varphi_1, \ldots, \psi_0, \psi_1, \ldots, \xi_1$ and ξ_2 and the critical exponents α, β, δ, and γ and also conditions relating the coefficients of the expressions (15) and (16) such that the equation of state will satisfy relations (5)-(9).

We substitute (15) and (16) into (11) and express the thermodynamic functions $K_T(\rho, T)$, $C_V(\rho, T)$, and $p(\rho, T)$ in terms of derivatives of the free energy $F(\rho, T)$ [2]. Then asymptotically close to the critical point ($\Delta\rho \to 0$, $\tau \to 0$) we represent C_V, K_T, and p on the critical isotherm ($\tau = 0$), the coexistence curve [$T = T_s(\rho)$], and on the critical isochore ($\Delta\rho = 0$) as power series in $\Delta\rho$ and τ. Substituting these series into the corresponding scaling relations (5)-(9) and using the Griffiths relations, we equate the exponents of the leading terms on the right and left sides of the resulting relations. We then find the required relations for $\varphi_0, \psi_0, \psi_1, \varphi_1, \varepsilon_0, \varepsilon_1, \eta_0, \xi_1, \xi_2$ and α, β and also for the coefficients C_{ij} of (16):

$$\begin{cases} \varphi_0 = \psi_0 = 0, \ \eta_0\xi_1 = 1 + \delta, \ \varphi_1\xi_1 = 2 - \alpha, \ \varphi_1 = 1 \text{ or } 2, \\ \varepsilon_0\xi_2 = 1 + \delta, \ \varepsilon_1\xi_2 = 2, \ \psi_1\xi_2 = \gamma, \ \psi_1 = 1 \text{ or } 2, \end{cases} \tag{17}$$

$$2C_{10} + 2C_{20} + 1 = 0, \ C_{10} + 1 + Z_c f = 0, \tag{18}$$

where Z_c is the critical compressibility and $f = P_{-1}/T_c - P_1 T_c - 3P_3 T^3_c$.

It follows from (15)-(18) that the required expression for the function F(ρ, T) has the form

$$\frac{F_0(\rho,\ T)}{RT} = \omega^n |\Delta\rho|^{\delta+1} [A(A_1 x^{\varphi_1} + A_0)^{\xi_1} + B(B_1 x^{\psi_1} + B_0)^{\xi_2}] + \ln\rho + \sum_{i=0}^{n_3} \left(\sum_{j=0}^{n_4(i)} C_{ij} \left(\frac{T_K}{T} - 1 \right)^j \right) \Delta\rho^i, \quad (19)$$

where the coefficients C_{ij} and the exponents ψ_1, ξ_1, ξ_2 satisfy the relations (17) and (18) and where $x = \tau/|\Delta\rho|^{1/\beta}$ is the scaling variable and A_0, A_1, B_0, $B_1 > 0$.

We discuss the analysis of (19) in more detail. If we put $\psi_1 = \varphi_1 = 1$ in (19) then [12, 14] the equation $A_1 x + A_0 = 0$ is the equation of a pseudospinodal curve which is a set of singular points of C_V on the thermodynamic surface, and the isotherms are discontinuous in the two-phase region on this curve.

It follows from (19) that in the case $\psi_1 = \varphi_1 = 2$ there is only one singular point on the thermodynamic surface, the critical point, and the isotherms in the two-phase region have the van der Waals form.

We substitute (19) into (11) and using the thermodynamic relation $p = \rho^2 (\partial F/\partial\rho)_T$ obtain a single equation of state for the gas and liquid phases, which satisfies all of the requirements (1)-(9):

$$p(\rho,\ T) = p_n(T_s) + RT\omega^n |\Delta\rho|^\delta [F_1(\rho,\ T) - F_1(\rho,\ T_s)] + RT\rho \left(1 - \frac{T_s}{T} \right) \left[1 + \rho \sum_{i=1}^{n_3} \left(\sum_{j=0}^{n_4(i)} C_{ij} \tau_1^j \right) \frac{i}{\rho_c} \Delta\rho^{i-1} \right]; \quad (20)$$

here

$$F_1(\rho,\ T) = F_2(\rho,\ T) + F_3(\rho,\ T);$$

$$F_2(\rho,\ T) = n(\Delta\rho)\rho [A_0^* (x^{\varphi_1} + B_0^*)^{\xi_1} + A_1^* (x^{\psi_1} + B_1^*)^{\xi_2}];$$

$$F_3(\rho,\ T) = \rho\omega\, \mathrm{sign}(\Delta\rho) [A_1^* (\tau^{\psi_1} |\Delta\rho|^{\varepsilon_1 - 1 + \delta} \varepsilon_1 + B_1^* \varepsilon_0)\, \xi_2 (x^{\psi_1} + B_1^*)^{\xi_2 - 1} + A_0^* B_0^* \eta_0 \xi_1 (x^{\varphi_1} + B_0^*)^{\xi_1 - 1}];$$

$$A_0^* = AA_1^{\xi_1}, \quad B_0^* = A_0/A_1, \quad A_1^* = BB_1^{\xi_2}, \quad B_1^* = B_0/B_1.$$

We discuss how completely the equation of state (20) reproduces the regular part of the thermodynamic surface for small densities. Since $p_n(T_s)|_{\rho\to 0} \sim \exp(P^*_{-1}/\rho)$, where $P^*_{-1} < 0$, we obtain from (20) in the limit $\rho \to 0$

$$Z = 1 + \sum_{i=1}^{n-1} H^{(i)}(T)\,\omega^i + O(\rho^n), \quad (21)$$

where

$$H^{(1)}(T) = -\frac{R_1 T_c}{T} + \sum_{i=1}^{n_3} \left(\sum_{j=0}^{n_4(i)} C_{ij} \tau_1^j \right) i;$$

$$H^{(2)}(T) = \frac{R_2 T}{2T} - \sum_{i=1}^{n_3} \left(\sum_{j=0}^{n_4(i)} C_{ij} \tau_1^j \right) i \left(1 - i + \frac{R_1}{T} \right);$$

$$R_1 = D_0 \frac{1}{\beta} + i_1^+ D_1^+ + i_2^+ D_2^+ + i_3 D_3^+; \quad \tau_1 = \frac{T_c}{T} - 1;$$

$$R_2 = \frac{1}{\beta} \left(\frac{1}{\beta} - 1 \right) D_0 - i_1^+ (i_1^+ - 1) D_1^+ - i_2^+ (i_2^+ - 1) D_2^+ - i_3 (i_3^+ - 1) D_3^+.$$

It follows from (21) that if $n \geq 3$, our approach not only gives the correct limit to the equation of state of an ideal gas, but also gives the qualitatively correct temperature dependence of the second and third virial coefficients.

Quantitative analysis was carried out on the equation of state (20) using the most reliable p—v—T and C_V data on Ar [15-17]. Experimental data on C_V were included together with the thermal data because the specific heat at constant volume is the most sensitive test of the accuracy of a thermal equation of state [18].

We used the following values of the parameters in the equation of state (20): $\varphi_1 = 1$, $\psi_1 = 1$, $\xi_1 = 2 - \alpha$, $n_0 = 1/\beta$, $\xi_2 = \gamma$, $\varepsilon_1 = 2/\gamma$, $\varepsilon_0 = 1/\beta$, $\alpha = 0.116$, $\beta = 0.34$. The functions $T_S(\rho)$ and $p_n(T_S)$ were chosen in correspondence with [7].

Because $\tau + B^*_0|\Delta\rho|^{1/\beta} = 0$ and $\tau + B^*_1|\Delta\rho|^{1/\beta} = 0$ are the equations of pseudospinodal curves, the nonlinear parameters B^*_0 and B^*_1 were determined from the relations $B^*_0 = y_1^{1/\beta}D_0$, $B^*_1 = y_2^{1/\beta}D_0$, where $D_0 = -0.6349$, $y_1 = 1.41$, $y_2 = 1.07$ [11].

The mean-square deviations between the calculations using the equation of state (20) and the experimental data of [15-17] in the region $0.78 \leq T/T_c \leq 2.7$, $0.2 \leq \omega \leq 2.1$ were as follows: the density in the single-phase region $\delta\rho_{sp} = 0.21\%$, the specific heat at constant volume $\delta C_V = 3.7\%$, the density on the coexistence curve $\delta\rho = 0.34\%$, and the pressure on the elasticity line $\delta p_n = 0.12\%$.

Therefore, our equation of state (20) satisfies all of the requirements (1)-(9) and gives the correct temperature dependence of the leading virial coefficients; in this respect, it compares favorably to the equations of state in [6, 7]. Also, the presence of the derivative $T_n'(\rho)$ in the equation of state of [6, 7] makes it difficult to control the growth of errors in the region of small densities. Equation (20) is free from this problem. To a large extent, this explains its better accuracy in the single-phase region of the thermodynamic surface in comparison with [6, 7].

NOTATION

μ, chemical potential; ρ, density; p, pressure; T, absolute temperature; R, universal gas constant; T_3, triple point; v_c, p_c, T_c, ρ_c, critical parameters; v, specific volume; $p_n(T_S)$, elasticity line; T_S, coexistence curve; $\Delta p = (p - p_c)/p_c$, $\Delta\rho = (\rho - \rho_c)/\rho_c$, $\tau = (T - T_c)/T_c$; μ' and μ'', chemical potentials on the coexistence curve; C_V, heat capacity at constant volume; C_p, heat capacity at constant pressure; K_T, isothermal compressibility; $p_{ci}(T)$, critical isochore; F, free energy; α, β, δ, γ, critical exponents; Z_c, critical compressibility; Z, compressibility; ρ' and ρ'', values of the density on the vapor and liquid branches of the coexistence curve.

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DENSITY OF ADSORBED WATER IN POWDER SYSTEMS. 3. γ-Al_2O_3

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The choice of dispersion medium is considered. Estimates are made of the density of water adsorbed on γ-Al_2O_3.

The measurement methods and techniques have been given in [1, 2]. The dispersion media were nitrobenzene and toluene. Figure 1 shows the results on the density of γ-Al_2O_3 as a function of water content.

There is a clear-cut effect on the density from the adsorbed water concentration in the dependence of the adsorbent mass or m/V relation (m is the mass of dehydrated adsorbent and V is the volume of the dispersed system).

The experiments showed that nitrobenzene is a liquid of zero effect in relation to γ-Al_2O_3, since the density of the dehydrated solid phase measured with this medium is independent of m/V. However, Fig. 1b shows that the density of the adsorbent in the same state as measured in toluene decreases as m/V increases, which indicates that the concentration of the liquid molecules in the surface layers of the dehydrated solid phase is less than in the bulk.

We now examine the changes in the relative densities of the dispersion media as affected by the water contents. The quantities are calculated from

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