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EQUATION OF STATE TAKING THE FEATURES OF THE INTERNAL ENERGY INTO ACCOUNT AND BASED ON THE SATURATION LINE

E. S. Platunov, V. F. Lysenkov,
N. V. Vas'kova, and A. V. Shustrov

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A method of constructing the equation of state, based on the use of thermodynamic relations between the internal energy of the material and its thermal parameters, is presented.

At present, the construction of a single (for liquid and gas) equation of state is subjected to the condition of describing not only the thermal parameters of the material, but also the caloric data with an accuracy close to that of experiment. However, in most works [1, 2] the question of the qualitative and quantitative account of the thermal and caloric properties in the near-critical regions has been left out of consideration. In [3], on the basis of an analysis of the possibility of using a virial equation of state to describe the thermodynamic properties of argon, it was shown that the structure of the virial equation does not allow the thermal data and the isochoric specific heat in the critical region to be generalized with sufficient accuracy. The boundaries of the region in which the virial equation of state cannot be used were established: $T = T_C \pm 0.1\ T_C$, $\rho = \rho_C \pm 0.4\rho_C$.

If the equation of state is to permit calculations of the thermal and caloric properties over a broad range of parameters of state, including the near-critical region, it is evidently necessary to assign the equation a form that takes account of the features of the material's behavior in the vicinity of the critical point. An attempt to solve this problem was made in [4]. But the equation of state constructed in [4] does not agree at all accurately with the conclusions of scale theory [5], requiring, in particular, divergence of c_V on approaching the critical point along the critical isotherm according to the law $(1-T/T_C)^{-\alpha}$, whereas the value of the critical index is, as shown by subsequent calculations [6], $\alpha \simeq 0.108 \pm 0.010$.

In the present work a method of constructing the equation of state of liquid and gas is proposed, taking account of the features of the behavior of c_V in a broad range of parameters of state, including the near-critical region.

The well-known thermodynamic relation, in differential form, between the thermal parameters of the material and its internal energy u is used:

$$\left(\frac{\partial u}{\partial v}\right)_T = -\rho^2\left(\frac{\partial u}{\partial \rho}\right)_T = -\frac{\partial\left(\frac{p}{T}\right)_V}{\partial\left(\frac{1}{T}\right)}. \qquad (1)$$

Choosing some reference curve $T_{re}(\rho)$ at the thermal surface, Eq. (1) is integrated:

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$$\frac{p(\rho,\ T)}{T} = A_{\mathrm{re}}(\rho) - \rho^2 \int\limits_{T_{\mathrm{re}}(\rho)}^{T} \frac{\left(\frac{\partial u}{\partial \rho}\right)_T}{T^2}\, dT. \tag{2}$$

Here $A_{re}(\rho)$ is a "constant" of integration, equal to the ratio between the pressure and the temperature along the reference curve: $A_{re}(\rho) = [p/T]_{re}$.

The saturation line $T_S(\rho)$ is used as the reference curve. In this case Eq. (2) yields

$$p(\rho,\ T) = \frac{p_S(T_S)}{T_S(\rho)}\, T - \rho^2 T \int\limits_{T_S(\rho)}^{T} \frac{\left(\frac{\partial u}{\partial \rho}\right)_T}{T^2}\, dT. \tag{3}$$

If the equation of state is to allow both the thermal data and the isochoric specific heat to be calculated with acceptable accuracy (even in the near-critical region), the form of the expression for the internal energy $u(\rho,\ T)$ must take account of the features of both the thermal surface and c_V over the whole of the given range of parameters of state, including the near-critical region. The internal energy may be expressed as the sum of three terms:

$$u(\rho,\ T) = u_{\mathrm{IG}}(T) + u_{\mathrm{R}}(\rho,\ T) + u_{\mathrm{I}}(\rho,\ T). \tag{4}$$

As will be made clear by the following, $u_R(\rho,\ T)$ may be expediently written in the form: $u_R(\rho,\ T) = u_{1R}(\rho) + u_{2R}(\rho,\ T)$, where the first term is a function solely of the density.

The constraint imposed on the component of the internal energy $u_I(\rho,\ T)$ is that it must describe the irregular behavior of the isochoric specific heat c_V close to the critical point. Analysis of precision data on the isochoric specific heat of argon [7] leads to the following expression for the irregular component of the internal energy:

$$u_{\mathrm{I}}(\rho,\ T) = f_{\mathrm{I}}(\rho)\left(1 - \frac{T_{\mathrm{I}}(\rho)}{T}\right)^{1-\alpha}. \tag{5}$$

An expression for c_V is found from Eq. (4):

$$c_V(\rho,\ T) = \frac{du_{\mathrm{IG}}(T)}{dT} + \left(\frac{\partial u_{2\mathrm{R}}(\rho,\ T)}{\partial T}\right)_\rho + \left(\frac{\partial u_{\mathrm{I}}(\rho,\ T)}{\partial T}\right)_\rho = \overset{\mathrm{IG}}{c_V}(T) + \overset{\mathrm{R}}{c_V}(\rho,\ T) + \overset{\mathrm{I}}{c_V}(\rho,\ T). \tag{6}$$

According to Eq. (5), the irregular component of the isochoric specific heat is

$$\overset{\mathrm{I}}{c_V}(\rho,\ T) = \left(\frac{\partial u_{\mathrm{I}}(\rho,\ T)}{\partial T}\right)_\rho = \frac{f_{\mathrm{I}}(\rho)\, T_{\mathrm{I}}(\rho)(1-\alpha)}{T^2\left(1 - \frac{T_{\mathrm{I}}(\rho)}{T}\right)^{\alpha}}. \tag{7}$$

Assuming $\rho = \rho_C$ in the last relation, the behavior of $\overset{\mathrm{I}}{c_V}$ at the critical isochore is investigated:

$$\overset{\mathrm{I}}{c_V}(\rho_C,\ T) = \frac{f_{\mathrm{I}}(\rho_C)\, T_{\mathrm{I}}(\rho_C)(1-\alpha)}{T^2\left(1 - \frac{T_{\mathrm{I}}(\rho_C)}{T}\right)^{\alpha}}. \tag{8}$$

If the function $T_I(\rho)$ is required to satisfy the condition $T_I(\rho_C) = T_C$, then as $T \to T_C$ it is obvious that Eq. (8) yields: $\overset{\mathrm{I}}{c_V}(\rho_C,\ T \to T_C) \sim \Delta T^{-\alpha}$, i.e., the structure of the expression for the internal energy ensures divergence of the isochoric specific heat at the critical point.

If the equation of state constructed by the proposed method is to give both the thermal surface of the material and the caloric properties in quantitatively correct form, then the choice of the structure of the functions $f_I(\rho)$, $T_I(\rho)$, $u_R(\rho,\ T)$ must be based on the analysis both of P–V–T data and the caloric properties. In the present work, in order to simplify the analysis of the general possibilities of the given method, a simplified approach is used in searching for the form of the given functions (which is known to reduce the accuracy of the equation of state constructed). The functional form of $f_I(\rho)$, $T_I(\rho)$, and $u_{2R}(\rho,\ T)$ is chosen according to the results of analyzing the $c_V(\rho,\ T)$ dependence. Thereafter, the equation of state is constructed and, from an analysis of the thermal surface, the form of the function $u_{1R}(\rho)$ is specified; this function appears in the equation of state but not in Eq. (6) for c_V. This search procedure for the structure of the functions $f_I(\rho)$, $T_I(\rho)$, $u_{2R}(\rho, T)$, and $u_{1R}(\rho)$ was realized for argon, taken as the example for which the quantitative possibilities of the given method were investigated. As is known, in this case $u_{2R}(\rho, T)$ may be represented in the form

$$u_{2R}(\rho,\ T) = -\varphi_{2R} f_R(\rho)\frac{1}{T}\left(1+\frac{A}{T}\right);\quad f_R(\rho)=\sum_{i=1}^{6} A_i\left(\frac{\rho}{\rho_C}\right)^i,$$

where $A = 25°K$; $\varphi_{2R} = 3.3465\cdot10^6$; $A_1 = 1.0$; $A_2 = -1.33553$; $A_3 = 0.76133$; $A_4 = -0.29198$; $A_5 = -0.08359$; $A_6 = -0.01194$. Then the following expressions are obtained for the internal energy $u(\rho, T)$ and the isochoric specific heat c_V from Eqs. (4)-(9):

$$u(\rho,\ T) = u_{IG}(T) + u_{1R}(\rho) - \frac{\varphi_{2R} f_R(\rho)}{T}\left(1+\frac{A}{T}\right) - \varphi_I\left(1-\frac{\rho}{\rho_0}\right)^2\left[1-\left(1-\frac{T_I(\rho)}{T}\right)^{1-\alpha}\right], \tag{10}$$

$$c_V(\rho,\ T) = c_V^{IG} + \varphi_{2R} f_R(\rho)\frac{1}{T^2}\left(1+\frac{2A}{T}\right) + \frac{\varphi_I(1-\alpha)\left(1-\dfrac{\rho}{\rho_0}\right)^2 T_I(\rho)}{T^2\left(1-\dfrac{T_I(\rho)}{T}\right)^{\alpha}}. \tag{11}$$

The function $f_I(\rho)$ is approximated by the expression $\varphi_I(1-\rho/\rho_0)^2$ here. The values of the constants φ_I and ρ_0 are taken to be as follows: $\varphi_I = 300$ kJ/kg, $\rho_0 = 1.870\cdot10^3$ kg/m^3. The critical index α is taken to be 0.112, in accordance with the data of [6]. Substituting Eq. (10) into Eq. (3) gives the equation of state

$$p(\rho,\ T) = \frac{p_S}{T_S}T + \rho^2\left\{u_{1R}'\left(1-\frac{T}{T_S}\right) - \varphi_{2R} f_R'(\rho)\, F_1(\rho,\ T) + \varphi_I T\,[F_2(T) - F_2(T_S)]\right\}, \tag{12}$$

where

$$F_1(\rho,\ T) = \frac{1}{2T}\left[1-\left(\frac{T}{T_S}\right)^2\right] + \frac{A}{3T^2}\left[1-\left(\frac{T}{T_S}\right)^3\right],\qquad F_2(x) = \frac{2}{\rho_0}\left[\frac{1}{x} + \right.$$

$$\left. + \frac{1}{(2-\alpha)T_I}\left(1-\frac{T_I}{x}\right)^{2-\alpha}\right]\left(1-\frac{\rho}{\rho_0}\right) - \frac{T_I'}{(2-\alpha)T_I^2}\left(1-\frac{T_I}{x}\right)^{1-\alpha}\left(1+\frac{(1-\alpha)T_I}{x}\right)\left(1-\frac{\rho}{\rho_0}\right)^2.$$

The functions $u_{1R}(\rho)$, $T_I(\rho)$ and also $T_S(\rho)$ and $\rho_S(T_S)$ appearing in Eqs. (11), (12) remain structurally indeterminate.

As already noted, the form of $T_I(\rho)$ is chosen on the basis of analyzing the $c_V(\rho, T)$ dependence in the present simplified approach [7]. The function $T_I(\rho)$ must then satisfy the following requirements: 1) $T_I(\rho_C) = T_C$, which, as noted, ensures divergence of c_V at the critical point; 2) $T_I(\rho\to0) = 0$, when, in Eqs. (10), (11), the transitions, in the limit, to the corresponding expressions valid for an ideal gas are rigorously observed: $c_V(\rho\to0,\ T) = c_V^{IG}(T)$, $u(\rho\to0,\ T) = u_{IG}(T)$; 3) as follows from Eq. (12), the following inequality must be observed for an arbitrary value of the density: $T_I(\rho) \leqslant T_S(\rho)$; 4) the function $T_I(\rho)$ should not have singularities in the given range of densities from 0 to ρ_{tr}^L, including the near-critical region. This condition is satisfied by the function

$$T_I(\rho) = T_C\frac{\rho}{\rho_C}\exp\{-\Delta\rho + B_1^{\mp}\Delta\rho^2 + B_2^{\pm}\Delta\rho^3\}. \tag{13}$$

The constants $B_1^{\pm}$ and $B_2^{\pm}$ are assumed to be: $B_1^+ = 0.47$; $B_2^+ = -1.57$; $B_1^- = 0.50$; $B_2^- = 0.60$. In accordance with [6], the critical parameters are taken to be $T_C = 150.66°K$, $\rho_C = 0.5351\cdot10^3$ kg/m^3. The relation used for $u_{1R}(\rho, T)$ in the present work is

$$u_{1R}(\rho) = \varphi_{1R}\sum_{i=1}^{5} D_i\left(\frac{\rho}{\rho_C}\right)^i, \tag{14}$$

where $D_1 = -160.53$; $D_2 = 3.34435\cdot10^3$; $D_3 = -1.6501\cdot10^4$; $D_4 = 1.9410\cdot10^4$; $D_5 = -3.0278\cdot10^3$; $D_6 = -7.7017\cdot10^2$.

Now consider $T_S(\rho)$ and $p_S(T_S)$. In accordance with scale theory [5], in the asymptotic vicinity of the critical point the temperature dependence of the density at the saturation line is written in the form $\Delta\rho = H|\Delta T|^{\beta}$ sign $(\Delta\rho)$. Therefore, in describing experimental data at the saturation line for $T_S(\rho)$, the following relation may be used: $T_S(\rho) = T_C(1 + C_\beta|\Delta\rho|^{1/\beta} + \sum_{k\geqslant 4} C_k^{\pm}\Delta\rho^k)$. Analysis of the experimental data for argon permits the following relation to be written for the description of the saturation line:

$$T_S(\rho) = T_C(1 + C_\beta|\Delta\rho|^{1/\beta} + C_4^{\pm}\Delta\rho^4 + C_{10}^{\pm}\Delta\rho^{10}), \tag{15}$$

where $C_\beta = -29.1117$; $C_4^- = 7.64$; $C_{10}^- = -0.03$; $C_4^+ = -5.3122$; $C_{10}^+ = 21.58$; $\beta = 0.3514$.

In approximating the elasticity line, it must be understood that in accordance with scale theory, the second derivative d^2p_S/dT_S^2 at the critical point is singular. This requirement, e.g., is satisfied by the equation of the elasticity curve proposed for argon in [8]:

$$\lg\frac{p_S(T_S)}{p_C} = (E_1 - \lg p_C) + E_2 T_S^{-1} + E_3 T_S + E_4(T_C - T_S)^{\varepsilon} + E_5 T_S^3, \tag{16}$$

where $E_1 = 1.83977$; $E_2 = -435.070\,°K$; $E_3 = 2.83062\cdot10^{-2}\,°K^{-1}$; $E_4 = 3.54795\cdot10^{-4}\,°K^{-\varepsilon}$; $E_5 = -4.49101\cdot10^{-7}\,°K^{-3}$; $\varepsilon = 1.90798$.

Thus, all of the parameters appearing in equation of state (12) have been determined, and there is the possibility of estimating the accuracy of the equation of state constructed in calculating the thermal parameters and isochoric specific heat. In the first case, the comparison is with the equation of state of [3] and also with the experimental data used as the basis in determining the coefficients of this equation; in the second case, it is with the experimental data on c_V from [7] and the same equation of [3].

Calculation of c_V from Eq. (11) shows that the mean-square deviation δc_V^m in the whole of the parameter range considered in [7] is not more than 4.5%. The maximum deviation of the results calculated for c_V from the experimental values is observed on the $\rho = 0.3096$ g/cm³ isochore, but is not more than 10%. As a comparison, note that in calculating c_V from the virial equation of state [3], the deviation from the experimental result amounts to 100% and more over a broad region in the vicinity of the critical point. In addition, it is important to stress two factors. First, Eq. (12) ensures that the isochoric specific heat tends to infinity as $T \to T_C$ along the critical isochore according to the law $(1-T/T_C)^{-\alpha}$, which agrees with the conclusions of scale theory [5, 6]. Second, the structure of functions $f_R(\rho)$, $u_{1R}(\rho)$, and $T_I(\rho)$ is such that in Eqs. (10), (11) transition occurs to the corresponding relations for an ideal gas.

Before discussing the accuracy of equation of state (12) in thermal calculations, note the following. The fact that the saturation line is used as the reference curve allows two important problems to be solved. On the one hand, in this case, the problem of consistency of the data in single-phase space and at the saturation line is automatically solved. The use of the equation for $T_S(\rho)$ in the form in Eq. (13) allows the features of the thermal surface of the material in the near-critical region to be more accurately taken into account than in the case of the virial equation. On the other hand, as may readily be shown, the structure of Eq. (12) guarantees the satisfaction of the critical conditions $p(\rho_C, T_C) = p_C$, $(\partial p/\partial\rho)_T^C = 0$, $(\partial^2 p/\partial\rho^2)_T^C = 0$. One other point may be noted. The region of definition of Eq. (2) with respect to the density evidently coincides with the region of definition of the reference curve. Hence, Eq. (12) is defined in the density range $\rho''_{tr} \leqslant \rho \leqslant \rho'_{tr}$.

However, in the present work, the equation of state of argon in the form in Eq. (12) was constructed in the narrower density range 0.035 g/cm³ $\leqslant \rho \leqslant 1.02$ g/cm³. The lower limit of this range is determined by the boundary of the range of application of equation $T_S(\rho)$ for the saturation line. The choice of the upper limit is associated with the use of a simplified procedure in seeking the coefficients of Eq. (12), where some of the structural functions – $T_I(\rho)$, $f_R(\rho)$, $f_I(\rho)$ – were determined solely on the basis of a mass of c_V data. For argon there are only reliable data on c_V for densities less than 1.02 g/cm³.

Calculation of the thermal parameters of argon in the single-phase region and comparison of the results with experimental data shows that, on average, the deviation $\delta\rho$ in the given range of parameters of state is 1.1%. Hence, in the accuracy of calculating the density in the region of the single-phase state, the equation constructed is inferior to the analytical equation of state of [3]. However, if the simplification of the approach to finding the coefficients of Eq. (12) and the small number of fitting parameters of Eq. (12) in comparison with the equation of [3] are taken into account, the result obtained here may be regarded as satisfactory.

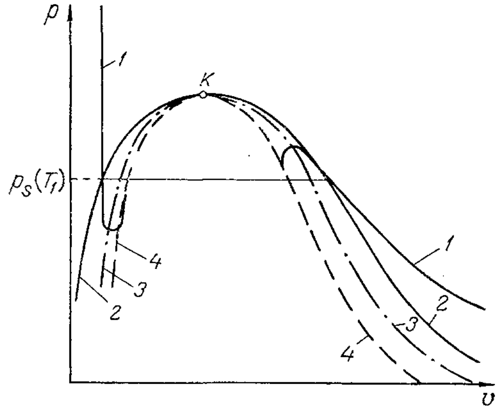

Fig. 1. Description of the metastable states using Eq. (12): 1-1) isotherm $T = T_1$ obtained from Eq. (12); 2-2) liquid–vapor equilibrium line; 3-3) spinodal; 4-4) line $p_I(v) = p(v, T = T_I(v))$; $p_S(T_1)$ is the saturation pressure at temperature T_1. p, Pa; v, m^3/kg; T, °K.

In calculating the density at the saturation line from the virial equation of [3], the deviation from the experimental data rises on approaching the critical point, and reaches tens of percent and above. In the case of the procedure proposed here, the accuracy of the equation obtained in calculating the density at the saturation line is determined by the accuracy of the expression for $T_S(\rho)$. If Eq. (15) is used for $T_S(\rho)$, the mean-square deviation $\delta\rho^m$ is not more than 0.35% over the whole of the density range considered, including the near-critical region.

Before obtaining the basic conclusions, the question of the possibility of describing the region of metastable states by means of the equation obtained may be considered. In Fig. 1 an arbitrary isotherm $T = T_1$ (in the projection on the p–v plane) obtained from Eq. (12) is shown. It is evident that the spinodal obtained from Eq. (12) – $(\partial p/\partial v)_T = 0$ – lies between the liquid–vapor equilibrium line $p_S(v)$ and the line $p_I(v) = p(v, T_I(v))$. Thereby, Eq. (12) depicts the form of the dependence $p(\rho, T)$ in the region of metastable states qualitatively correctly. Consideration of the quantitative prospects for description of the metastable region using Eq. (12) falls outside of the scope of the present work.

Thus, the approach considered in the present work permits, in principle, the construction of an equation of state of gas and liquid, which, in contrast to the analytical equations of state, more strictly depicts the features of the critical region. In particular, the equation constructed in this way gives a qualitatively correct depiction of the singular character of the isochoric specific heat in the vicinity of the critical point. This, in turn, means that the equation constructed leads to satisfactory accuracy in calculating c_V, both in the regular part of the surface and in the critical region.

Increase in the accuracy of the approach proposed here in calculating the thermal data will in the future be associated with the perfection of the structure of the expression for the internal energy and with optimization of the search for the set of coefficients of the equation of state.

NOTATION

p, pressure; T, absolute temperature; v, specific volume; ρ, density; ρ_C, T_C, ρ_C, v_C, critical parameters; $\Delta\rho = (\rho-\rho_C)/\rho_C$; $\Delta T = (T-T_C)/T_C$: α, β, critical indices of the isochoric specific heat and the saturation line, respectively; $c_V(\rho, T)$, $c_V^{IG}(T)$, isochoric specific heat of real material and ideal gas; $c_V^R(\rho, T)$, $c_V^I(\rho, T)$, regular and irregular components of the isochoric specific heat; $u(\rho, T)$, $u^{IG}(T)$, specific internal energy of a real material and an ideal gas; $u_R(\rho, T)$, $u_I(\rho, T)$, regular and irregular components of the internal energy; $T_{re}(\rho)$, equation of reference curve; $T_S(\rho)$, saturation line; $p_S(T_S)$, elasticity curve; $f_R(\rho)$, $f_I(\rho)$, $T_I(\rho)$, structural functions of the internal energy; $f'_R(\rho)$, $f'_I(\rho)$, $T'_I(\rho)$, derivatives of the corresponding functions with respect to the density; ρ''_{tr}, ρ'_{tr}, vapor and gas density at triple point; $\delta c_V = (c_V^{calc} - c_V^{exp})/c_V^{exp}$; $\delta\rho = (\rho^{calc} - \rho^{exp})/\rho^{exp}$; φ_I, φ_{1R}, φ_{2R}, ρ_0, A_i, D_i, E_i, $B_i^{\pm}$, $C_i^{\pm}$, constants corresponding to the cases $\rho \geqslant \rho_C$ (+) and $\rho < \rho_C$ (−).

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DIFFUSIONAL EXTRACTION OF MATERIAL FROM MODEL POROUS BODIES

R. Rakadzhiev, N. V. Pavlyukevich,
A. Trifonov, S. Radev,
R. Kuzmanova, and B. Nikolova

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The solution and diffusional extraction of solid materials from porous membranes is investigated theoretically and experimentally.

The extraction of materials from porous bodies is widespread in commercial technology and in nature: the separation of inorganic and organic products in porous catalysts, the leaching out of mineral salts, the extraction of oil from cells of plant origin, hydrometallurgical treatment, etc. Similar subjects were investigated, e.g., in [1-3]. However, in describing these processes, mass transfer in a single capillary is most often considered, with a specified density of the solute at the exit, or the mass transfer in the solution and diffusional extraction in a porous body and outside it is not investigated in a consistent formulation. In the present work, results are given of experimental and theoretical investigations of the successive processes of solution of a solid material and its diffusional extraction from model porous membranes, the capillaries of which are entirely filled with material in the form of a solid phase or solution. A mathematical model of the above-noted processes is proposed in the form of a refined version of that described in [4].

As follows from [5], reflective spectroscopy, based on the phenomenon of attenuation of

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Institute of Organic Chemistry, Institute of Mechanics and Biomechanics, Bulgarian Academy of Sciences, Sofia. A. V. Lykov Institute of Heat and Mass Transfer, Academy of Sciences of the Belorussian SSR, Minsk. Translated from Inzhenerno-Fizicheskii Zhurnal, Vol. 45, No. 1, pp. 106-112, July, 1983. Original article submitted April 22, 1982.

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