Fundamental equation of state developed on basis of Migdal’s phenomenological theory of critical point

S V Rykov¹, I V Kudryavtseva¹, V A Rykov¹, A V Sverdlov¹ and E E Ustyuzhanin²

¹ ITMO University, Kronvergskiy 49, Saint-Petersburg 197101, Russia
² National Research University "MPEI", Russia, 111250 Moscow, Krasnokazarmennaya, 14
togg1@yandex.ru

Abstract. In this work a fundamental equation of state (FEoS) with account taken of requirements of the scale theory of the critical point has been developed. The behavior of the FEOS in the asymptotic vicinity of the critical point is determined by the characteristics of the scaling function \(a(x)\) of the Helmholtz scale free energy \(F\). The structure of the function \(a(x)\) is calculated on the basis of Migdal’s phenomenological theory and the hypothesis about the identical behavior of the isothermal compressibility coefficient at the critical and near-critical isobars near the critical point (Benedek’s hypothesis). The parameters of the singular component of the FEoS are calculated using the similarity theory. The proposed FEoS has been approved on the example of description of equilibrium properties of the refrigerating agent R1234yf in the range of state parameters: by a temperature from 220 K to 440 K and by pressure up to 40 MPa. The obtained results are discussed.

1. Introduction
For calculation of the equilibrium properties of the perspective environmentally safe refrigerating agent R1234yf (2,3,3,3-tetrafluoropropene), fundamental equations of state (FEoS) are currently used [1–3]. These equations of state have rather a complicated structure since they contain, besides linear coefficients, about 30 individual nonlinear parameters. In addition, the equations of state [1-3] do not take into account the requirements of the scaling theory (ST) of the critical point (CP) of liquid and gas, which narrows the working region of these equations. Disadvantages of the FEoS [1–3] are partially taken into account in papers [4, 5] which propose a technique for building the equation of state for R1234yf which meets requirements of the ST [6] near the critical point. In papers [4, 5] we used an approach to the construction of the FEoS based on the method of pseudocritical points [7] and Benedek’s hypothesis [8]. Within the framework of the approach [4, 5, 7] a scaling function of \(a(x)\) of the Helmholtz free energy is used in which the equations \(x = -x\) determine a set of pseudocritical points on a thermodynamic surface that meet the conditions of [9]:

\[
\left( \frac{\partial \rho}{\partial p} \right)_T = 0 \quad \Leftrightarrow \quad \left( \frac{\partial T}{\partial s} \right)_p = 0, \tag{1}
\]

where \(\rho\) is the density; \(v\) is the specific volume; \(p\) is the pressure; \(T\) is the temperature; \(s\) is the entropy.

Thus, in the case [4, 5, 7] the structure of the fundamental equation of state:
\[ F = F_s(\rho, T) + F_i(\rho, T, a(x)), \]  

(2)

includes three lines of pseudo-critical points, which complicates the structure of the FEoS.

Here, \( F_s(\rho, T) \) is the regular component of the Helmholtz free energy \( F = F_s(\rho, T, a(x)) \) is the singular component \( F \); \( x = \tau / |\Delta \rho|^\gamma \) is the scaling variable; \( \tau = T / T_c - 1 \); \( \Delta \rho = \rho / \rho_c - 1 \); \( T_c \) and \( \rho_c \) are the critical temperature and critical density, respectively; \( \beta \) is the critical index associated with critical indices \( \alpha, \gamma, \delta \) by the Griffith equations [10]:

\[ 2 - \alpha = \gamma + \beta \quad \text{and} \quad \gamma = \beta \delta - \beta. \]

(3)

On the other hand, the works [11, 12] on the basis of the scaling function \( h(x) \)

\[ h(x) = A\left[ (x + x_q)^\beta - (x - x_0)^\beta \right] \]

(4)

of the chemical potential \( \mu(\rho, T) \) have developed fundamental equations of state for SF_{6} [12] and CO_{2} [11]. In (4), \( x_q \) is the saturation line parameter; \( x_0 \) is the individual parameter.

The equations [11, 12] satisfactorily describe the equilibrium properties of the studied substances in a wide range of state parameters. However, our analysis has shown that the FEoS [11] qualitatively incorrectly describes the behavior of the isothermal compressibility coefficient \( K_T \), isobaric heat capacity \( C_p \), and the sound velocity in the vicinity of the critical point. For example, the behavior of \( K_T \) and \( C_p \) on the critical isotherm near the critical point within the limits of [11, 12] is determined by power-law relations \( K_T(T \rightarrow T_c) \sim \tau^4 \) and \( C_p(T \rightarrow T_c) \sim \tau^4 \). However, according to the ST [6] on the line \( \rho = \rho_c \) at \( T \rightarrow T_c \) the functions \( K_T(\rho = \rho_c, T) \) and \( C_p(\rho = \rho_c, T) \) meet the following power laws: \( K_T(T \rightarrow T_c) \sim \tau^{-\gamma} \) and \( C_p(T \rightarrow T_c) \sim \tau^{-\gamma} \).

In the work of I.V. Kudryavtseva and S.V. Rykov [13] on the basis of Migdal’s theory [14] and Benedek’s hypothesis a large-scaling function of the chemical potential has been developed:

\[ h(x) = A\left[ (x + x_i)^\beta - (x_i - x_0)^\beta \right] \]

(5)

where \( x = -x_i \) is the equation describing the geometric place of the points (1) on the thermodynamic surface.

The function (5), in case \( \gamma = 4 \beta \), meets requirements of (1) and in this respect it favourably differs from the function (4). It should be noted that if you put \( \gamma = 4 \beta \) into (5), then we will get the function (4). In this case, it directly follows from (3) that \( \delta = 5 \), which contradicts the results obtained from the analysis of the Ising three-dimensional model [15, 16]: \( \delta = 4.79 \pm 4.82 \).

In this paper we have considered the possibility of constructing the FEoS on the basis of the approach combining the advantages of methods of equation of state construction proposed by the authors of [4, 5, 7] and [13, 17].

2. Determination of the structure of the fundamental equation of state

The paper [18] shows that on the basis of (3), with the known thermodynamic relation \( F = -\int S \, dT \) and Benedek’s hypothesis [7], it is possible to construct the fundamental equation of state in the following form:

\[ F(\rho, T) = F_s(\rho, T) + F_a(\rho, T) + F_i(\rho, T, a(x)), \]

(6)
where \( F_g(\rho, T) \) is the ideal gas component \( F \); \( F_{\text{res}}(\rho, T) \) is the residual part of the regular function \( F_s(\rho, T) \) (3).

The non-regular component \( F_s \) of the Helmholtz free energy in (6) has a form [18]:

\[
F_s = \psi(\omega) |\Delta \rho|^\delta \cdot \alpha(x). \tag{7}
\]

Here, \( \delta \) is the critical index of the critical isotherm; \( \alpha(x) \) is the scaling function of the Helmholtz free energy; \( \omega = \rho / \rho_c \); \( \psi(\omega) \) is a regular function of \( \omega \).

In this work, the scaling function \( \alpha(x) \) calculated on the basis of the function \( h(x) \) (5) is used. The functions \( \alpha(x) \) and \( h(x) \) are related by the equation:

\[
h(x) = (\delta + 1) \alpha(x) - \frac{x}{\beta} \alpha'(x). \tag{8}
\]

As a result of the solution of differential equation (8) (see, for example, [17]) we obtain:

\[
\alpha(x) = \frac{u_0 \beta}{k \cdot x_1^{3\beta}} (x + x_1)^{2 - \alpha} \left\{ \left[ \frac{1}{2 - \alpha} + \sum_{n=1}^{\infty} (-1)^n \left( \frac{2\beta - 1}{n!(2 - \alpha - n)} \right) \left( \frac{x}{x + x_1} \right)^n \right] - \frac{x}{x_1^{4\beta}} \left[ \frac{1}{2 - \alpha} + \sum_{n=1}^{\infty} (-1)^n \left( \frac{6\beta - 1}{n!(2 - \alpha - n)} \right) \left( \frac{x}{x + x_1} \right)^n \right] \right\} \tag{9}
\]

where \( u_0 \) is the individual parameter of the substance; \( k = (b - 1)/x_0 \).

We chose the regular component of the FEoS (10) in accordance with the recommendations [5]:

\[
F_{\text{res}}(\rho, T) = RT \omega \sum_{i=0}^{n_1} \sum_{j=0}^{n} C_{ij} \tau_i^j (\Delta \rho)^j, \tag{10}
\]

where \( C_{ij} \) are the coefficients which are determined on the basis of the experimental data on the equilibrium properties of the studied substance; \( \tau_i = T_i / T - 1 \); \( R \) is the gas constant.

The fundamental equation of state developed on the basis of (6) with the component \( F_s(\rho, T, \alpha(x)) \) (7) and scaling function \( \alpha(x) \) (9) describe the power laws of the ST only if the following conditions are fulfilled [19]:

\[
\left( \frac{\partial \rho^s}{\partial \rho^s} \right)_{T=T_i, \rho=\rho_i} = 0, \left( \frac{\partial p}{\partial \rho} \right)_{T=\rho=\rho_i, \tau=\alpha} \sim \omega(\tau), \tag{11}
\]

where \( n=1, 2, 3, 4 \), \( \omega \) is the Landau symbol [20].

3. Thermal equation of state and isochoric heat capacity

Our analysis has shown that the problems in [11, 12] while describing the region of the critical states of the substance are related to the fact that the FEOS proposed in these works do not meet the conditions of (11). Therefore, we have transformed (10) taking into account the conditions (11) and condition \( p(\rho = \rho_c, T = T_c) = \rho_c \). As a result, on the basis of FEoS (6), we have obtained the thermal equation of state (EoS) in the following form [21]:

\[
\]
Z = 1 + y_1 \omega^2 + y_2 \omega + \left( y_3 \omega^2 + y_4 \omega \right) \left( Z_c - 0.2 \right) + \omega \sum_{i=0}^{3} \sum_{j=0}^{3} C_{i,j} \tau_i^{(\beta)} (i \omega + \Delta \rho) \\
+ D_i \tau_i \left( 2 \omega^3 - 3 \right) + D_2 \omega^2 \tau_i \left( 3 \omega - 4 \right) + D_3 \omega \left( y_1 \omega + y_3 \omega - y_5 \right) \\
+ \omega |\Delta \rho|^{(\beta - 1) \psi} \left( \psi(\omega) \right) \left( \Delta \rho \right)^{\beta - 1} \text{sign}(\Delta \rho) \h(x) + \psi(\omega) \left( \Delta \rho \right)^{\beta} a(x),

where \( Z = p/RT \rho \) is the compressibility; \( p_c \) is the critical pressure; \( Z_c = p_c/(RT_c); \) \( t = T/T_c; \) \( y_2 = 7.7/6 + 2.9/6 \Delta \rho - 1.1/6 \Delta \rho^3 + 0.05 \Delta \rho^3, \) \( y_3 = 5 - 4 \Delta \rho + 3 \Delta \rho^2 - 2 \Delta \rho^3 + \Delta \rho^4, \) \( y_4 = 4 - 3 \Delta \rho + 2 \Delta \rho^2 - \Delta \rho^3 + \Delta \rho^4. \)

We will choose the function \( \psi(\omega) \) in (4) according to the results of [4] in the form of an expression:

\[
\psi(\omega) = \exp \left[ -2(\Delta \rho)^2 \sqrt{\omega} \right].
\]

Our analysis has shown the following:

- in the region of discharged gas the thermal equation of state calculated on the basis of the EOS (4) with the function (7) goes over into the virial form equation [22]:

\[
p / (\rho RT) = 1 + \omega B(T) + \omega^2 C(T) + ..., \]

where \( B(T) \) and \( C(T) \) are the second and third virial coefficients;

- in the vicinity of the critical point, the FEsos (6) with the function (9) ensures the performance of the ST power laws [23].

We calculated the expression for isochoric heat capacity on the basis of (12) using the known equality \( C_v = -T \left( \partial^2 F / \partial T^2 \right)_\rho; \)

\[
C_v(T, \rho) = C_{v_0}(T) - R \frac{\partial^2}{\partial T^2} \sum_{i=0}^{22} \sum_{j=0}^{6} C_{i,j} \left( j - 1 \right) \tau_i \Delta \rho^j + R \psi(\omega) \Delta \rho^{\beta} f(x).
\]

where \( C_{v_0}(T) \) is the ideal gas isochoric heat capacity whose calculation formula is given in the paper [3]:

\[
C_{v_0}(T) = R \left[ v_0 + \sum_{i=1}^{4} v_i \left( u_i / T \right) \exp \left( -u_i / T \right) \left( 1 - \exp \left( -u_i / T \right) \right)^2 \right],
\]

where \( R = 72.9073665143 \) kJ/(kg-K) [3]; \( v_i \) and \( u_i \) are constant coefficients [3]: \( v_0 = 4.944; \) \( v_1 = 7.549; \) \( v_2 = 1.537; \) \( v_3 = 2.030; \) \( v_4 = 7.455; \) \( u_1 = 718.0; \) \( u_2 = 877.0; \) \( u_3 = 4463.0; \) \( u_4 = 1755.0. \)

The scaling function of the isochoric heat capacity \( f(x) \) in (15) has the following form [17]:

\[
f(x) = -a^\alpha(x) = \frac{u_0 \beta \gamma_1}{k \xi_1} \left( \Delta \rho \right)^{\beta} \left[ \int \left( \frac{x}{t} \right)^{2\beta-1} dt - \frac{\gamma_2 \xi}{\xi_1} \left( x/t \right)^{2\beta-1} \int \frac{\gamma_2 \xi}{\xi_1} \left( x/t \right)^{2\beta-1} dt \right]
\]

\[
= -\frac{u_0 \beta \gamma_1}{k \xi_1} \left( x + x_1 \right)^{-\alpha} \left[ \frac{1}{\alpha} \sum_{n=0}^{\beta} \left( -1 \right)^n \left( \frac{2 \beta - 1}{n} \right) \left( \frac{x}{x + x_1} \right)^n \right]
\]

\[
- \frac{\xi_1}{x_1 \gamma_1} \left( x - x_1 \right)^{\beta} \left( x + x_1 \right)^{-\alpha} \left[ \frac{1}{\alpha} \sum_{n=0}^{\beta} \left( -1 \right)^n \left( \frac{6 \beta - 1}{n} \right) \left( \frac{x}{x + x_1} \right)^n \right],
\]

where \( u_0, \beta, \gamma_1, \xi_1, x_1 \) and \( \alpha, \beta, \gamma_1, \xi_1, x_1 \) are constants.
Table 1. Coefficients $C_{i,j}$ of the EoS (12).

| $C_{i,j}$ | 0 | 1 | 2 |
|-----------|---|---|---|
| 0         | 0 | 0 | 1.2920078081779 |
| 1         | 0 | 0 | -0.82564085211376 |
| 2         | 0 | 0 | -3.8043067473467 |
| 3         | 0 | -1.5134391854777 | 5.2670682863985 |
| 4         | 0 | -0.11546312337717 | -0.63950656145714 |
| 5         | 0 | 1.4228470240074 | -3.1189570014178 |
| 6         | -0.1506968841333 | -1.5661259458511 | 2.011634917453 |
| 7         | 0.063793419580932 | 0.6640890214364 | -0.1149641042878 |
| 8         | 0.2040794569502 | 0.423494085427 | 0 |
| 9         | -0.23476893437614 | -0.64015813703559 | -0.15700032917052 |
| 10        | 0.020671016935083 | 0.34711947539339 | 0 |
| 11        | 0.105450400309007 | -0.20130991917162 | 0.03481040901032 |
| 12        | -0.075153993176463 | 0.140671133558584 | 0 |
| 13        | 0.021754060639002 | -0.056049285517653 | -0.002612336350452 |
| 14        | -0.0024103908857265 | 0.0084206951441226 | 0 |

where $\varepsilon_i = (\gamma - 4\beta)(\gamma - 4\beta - 1)/\gamma_i$.

The critical indices are chosen according to [11, 12, 16]: $\alpha = 0.11$; $\beta = 0.3255$; $\gamma = 1.239$; $\delta = 4.806$.

The critical parameters are assigned values that are consistent with the results of [2, 3]: $T_c = 367.85$ K; $p_c = 3.3822$ MPa; $\rho_c = 475.55$ kg/m$^3$.

When calculating the parameter $u_0$ we used the relationship of the theory of similarity proposed by the authors of [24]:

$$u_0 = \left[ \frac{k}{A_0 + B_0 \rho_e} \right]^\delta,$$

(18)

where $A_0 = 1.274$ and $B_0 = -2.327$ [25].

In the work [25], using the description of the equilibrium properties of argon as an example, it is shown that the relation (18) allows to calculate the isochoric heat capacity of substances poorly studied in the vicinity of the critical point.

We have established the EoS (12) coefficients $C_{i,j}$ on an array of experimental data [26–38] as a result of search for the minimum of the following functional:

$$\Phi = \sum_{n=1}^{N_t} Q_{p,n} \left( p^{(\alpha)} - p^{(\alpha)} \right)^2 + \sum_{n=1}^{N_v} Q_{v,n} \left( v^{(\alpha)} - v^{(\alpha)} \right)^2 + \sum_{n=1}^{N_s} Q_{s,n} \left( s^{(\alpha)} - s^{(\alpha)} \right)^2 + \sum_{n=1}^{N_i} Q_{i,n} \left( i^{(\alpha)} - i^{(\alpha)} \right)^2,$$

(19)
Figure 1. Deviations $\delta p = \left( \frac{p^{(ex)} - p^{(r)}}{p^{(ex)}} \right) \times 100\%$ of pressure $p^{(r)}$ calculated as per EoS (12) from the pressure values $p^{(ex)}$ given in the work [33].

Figure 2. Deviations $\delta \rho = \left( \frac{\rho^{(ex)} - \rho^{(r)}}{\rho^{(ex)}} \right) \times 100\%$ of density $\rho^{(r)}$ calculated as per the EoS (12) and FEoS [3] from the values of density $\rho^{(ex)}$ presented in work [36]: 1 – $\delta \rho$ calculated as per the EOS (12); 2 – $\delta \rho$ calculated as per the FEOS [3].

Figure 3. Deviations $\delta C_v = \left( \frac{C_v^{(ex)} - C_v^{(r)}}{C_v^{(ex)}} \right) \times 100\%$ of isochoric heat capacity $C_v^{(r)}$ calculated as per the equation (15) from the data $C_v^{(ex)}$ [35].
Figure 4. Deviations $\delta w = \left( w^{(ex)} - w^{(r)} \right) / w^{(ex)} \times 100\%$ of the sound velocity $w^{(r)}$ calculated as per the EoS (12) and FEoS [3] from the data $w^{(ex)}$ [37]: 1 – $\delta w$ calculated as per the EoS (12); 2 – $\delta w$ calculated as per the FEoS [3].

Figure 5. Deviations $\delta C_p = \left( C_p^{(ex)} - C_p^{(r)} \right) / C_p^{(ex)} \times 100\%$ of isobaric heat capacity $C_p^{(ex)}$ calculated as per the equation of state (12) from the data $C_p^{(ex)}$ [34]. Isotherms: 1 – 373.15 K; 2 – 393.15 K; 2 – 413.15 K.

Figure 6. Dependence of the second virial coefficient $B(T)$ on temperature: 1 – calculation as per FEoS [38]; 2 – calculation as per FEoS [3]; 3 – calculation as per FEoS [33]; 4 – calculation as per the EoS (12).
Figure 7. Dependence of the third virial coefficient $C(T)$ on temperature: 1 – calculation as per EoS [38]; 2 – calculation as per EoS [3]; 3 – calculation as per EoS [33]; 4 – calculation as per the EoS (12).

Figure 8. Dependence of heat capacity $C_v(\rho,T)$. on density over the isotherm $T = 367.8501$ K. 1 – calculation as per the EoS (12); 2 – calculation as per the EOS of Richter et al. [3].

where indices ($ex$) and ($r$) denote, respectively, the experimental [26–38] and calculated values on the basis of the EOS (4), values $\rho$, $C_v$ and the saturated steam pressures $p_s$, $Q_{p,s}$, $Q_{v,n}$ and $Q_{p,n}$ are the weight functions of the $n$-th point from the data array [26–38]; $\mu_{n}^{ex}$ and $\mu_{n}^{r}$ are the values of the chemical potential $\mu$ on the steam and liquid branches of the saturation line.

We compiled the data array $p_{i,n}^{ex}-T_{i,n}-\rho_{i,n}^{ex}$ on the phase equilibrium line of R1234yf in accordance with the method [39].

The EoS coefficients (12) $C_{ij}$ are presented in table 1.

The following values are assigned to other EoS (12) coefficients and parameters: $\chi_{0} = 0.185$; $u_{0} = 4.75130399901207$; $D_{1} = 0.8535442472138$; $D_{2} = 0.8806981483171$; $D_{3} = 0.01082059557366$.

We have compared the values $p-\rho-T$ of the data (figures 1, 2), isochoric heat capacity (figure 3), sound velocity $w$ (figure 4), isobaric heat capacity (figure 5), second and third virial coefficients
(figures 6, 7) calculated by the EoS (12) with the corresponding experimental and calculated data [26–38]. It has been found that the EoS (12) conveys with low uncertainty (within the limits of the experimental data error) both the thermal surface (figures 1, 2) and experimental data on $C_v$ (figure 3). The comparison with the tabular data [3] shows that the accuracy of calculation according to equation (12) meets the requirements for the equations of state while calculating the tables of standard reference data (figures 2, 4, 5, 6). The equation (12), as well as FEOs [4, 5, 40], not only pass into the virial equation of state [22] in the gas region, but also qualitatively correctly convey the behavior of the second $B(T)$ and third $C(T)$ virial coefficients (figures 6, 7). It should be noted that FEOS [3] and the virial EoS [38] describe the behavior of the virial coefficient $C(T)$ in the temperature region $T > 370$ K even qualitatively incorrectly. In contrast to FEOs [1–3], the equation (15) reproduces behavior of $C_v$ in a wide vicinity of the critical point in accordance with the requirements of the theory [6, 23] (figure 8).

4. Findings
The use of A.A. Migdal's phenomenological theory [14] and the equation of state developed on the basis of a new representation of the scaling hypothesis [18] allowed to develop an equation of state (6) with the scaling function (9) which contains only one line of pseudocritical points $x = -x_1$ and meets all power laws of the ST [6, 22] in the asymptotic vicinity of CP. The equation of state (6) has been tested on the example of description of the equilibrium properties of the prospective refrigerating agent R1234yf. It has been shown that the proposed equation of state (6) with low uncertainty corresponding to the uncertainty of the experimental data [26–35] describes the thermodynamic surface of R1234yf in the range of the state parameters: by temperature from 220 K to 440 K and by pressure up to 40 MPa. When calculating the parameters of the scaling function (9), the Lysenkov-Rykov relation (19) [24] was used as a singular component (7) of the fundamental equation (6) which provides, as shown in [25] an increased accuracy of calculation of the isochoric heat capacity in the vicinity of the critical point. Therefore, in contrast to the FEOs [1–3], the equation (6) can be used to calculate the equilibrium properties of the R1234yf both in the regular part of the thermodynamic surface (figures 1–7), and in the wide vicinity of the critical point (figure 8). Further development of our proposed method for constructing FEOs based on the Migdal’s theory [14] is related to the following approximation of the scaling theory [40].

5. Conclusion
The use of the FEOs (6) developed on the basis of a new representation of the scale hypothesis [6] and Kudryavtseva-Rykov scaling function (5) developed using the phenomenological theory of A.A. Migdal, Benedek's hypothesis, and the similarity ratio (19) seems to be perspective, as we showed on the example of R21234yf when describing the equilibrium properties of individual substances that are poorly studied in terms of thermodynamics in the vicinity of the critical point.

6. References
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