Analysis of the saturation line on the basis of Clapeyron–Clausius and Gibbs–Duhem equations

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Abstract. We have considered some modernization of the equation of state for matter (EOS). This modernization is connected with increasing the accuracy of EOS that has a wide area of applicability including the critical region. Our analysis has shown that it is possible to increase the accuracy of such EOS if we prepare a special input array. It has to include heterogeneous reliable data including the isochore heat capacity, gas and liquid densities ρ−, ρ+ and the saturation line pressure ps. The array has to be used as an input array in a statistical processing that allows calculating adjustable coefficients of the EOS. We have set the task to develop models ρ−(T) and ρ+(T) which describe densities ρ−, ρ+ and ps(T) model related to the saturation pressure. In our methodical approach, the Clapeyron–Clausius equation and the Gibbs–Duhem equation participate. We have elaborated structure that is related to these models and meets some requirements of the scaling theory of critical phenomena. Experimental data ρ+, ρ−, ps, T for SF6 are selected to calculate adjustable coefficients of these models. Comparison of models allows us to estimate that models ρ−(T), ρ+(T), ps(T) satisfactorily describe (within the limits of experimental errors) the experimental data obtained in the temperature range from triple point to critical point.

1. Introduction
Our analysis of works [1–11] has shown the following positions:

- The actual problem is the implementation of some modernization of the equation of state for matter (EOS). This modernization is connected with increasing the accuracy of EOS that has a wide area of applicability including the critical region. This has allowed us to estimate that it is possible to increase the accuracy of such an equation if we prepare the special input array. It includes dissimilar reliable data among which isochore heat capacity, gas and liquid densities (ρ−, ρ+) and the pressure ps at the saturation line can be seen. The array has to be located from the triple point with parameters (ρtr, Ttr, ptr) to the critical point with parameters (ρc, Tc, pc). The array has to be used as an input array in statistical processing, which made it possible to calculate the EOS adjustable coefficients.
- It is of interest to elaborate the general approach to the phase equilibrium line description:
  - The equations ρ−(T) and ρ+(T) which can describe densities (ρ−, ρ+)
  - The equation ps(T) related to the saturation pressure.
These models will allow us to analyze the saturation line and give an opportunity to prepare special input arrays \((\rho^+, \rho^-, p_s, T)\) which cover the interval from \(T_{tr}\) to \(T_c\). It is not a trivial task to elaborate these models: firstly, the equations have to have high accuracy in the range of applicability. Secondly, they have to meet some requirements of the scaling theory of critical phenomena (ST).

- Several existing models of \(\rho^-(T), \rho^+(T)\) can be divided into two groups. The first group is considered in [1–3] and describes data \((\rho^+, \rho^-, T)\) in the range from \(T_{tr}\) to \(T_c\) with acceptable accuracy but does not meet the requirements of ST [12–14] which are related to the asymptotic neighborhood of the critical point.

The second group of equations [4–11] is intended to describe data \((\rho^+, \rho^-, T)\) only in the wide neighborhood of the critical point. This group of equations satisfies either the Wegner model [5] (“incomplete scaling”) or the Yang–Yang model [12] (“completed scaling”).

Methodical approach [10] has made it possible to consider jointly models \(\rho^-(T), \rho^+(T), p_s(T)\) on the base of the Gibbs–Duhem equation [15]

\[
v(T) dp_s(T) - d\mu(T) - s(T) dT = 0,
\]

where \(\mu(T_s)\) is the chemical potential on the saturation line; \(s(T_s)\) is the entropy on the saturation line; \(v(T) = 1/\rho(T)\) is the specific volume on the saturation line.

Calculations [8–10] have given some variants of models \(\rho^-(T), \rho^+(T), p_s(T)\).

In this work, we have set a purpose:
- to elaborate a joint group of models \(\rho^-(T), \rho^+(T)\) and \(p_s(T)\) based on the Gibbs–Duhem equation;
- to obtain adjustable coefficients of these models on an example connected with accurate experimental data \((\rho^+, \rho^-, p_s, T)\) of SF\(_6\) in the interval from \(T_{tr}\) to \(T_c\) [16];
- to apply this model group to increase the accuracy of EOS which have to work in the wide range of temperatures and pressures including the critical region.

The last problem of EOS designing has been considered somewhere [17–24]. The authors of papers [20–24] have developed EOS, which are related to some metals. In this case, the critical points are located at high temperatures and high pressures. The authors of works [18, 19] have considered singular behavior associated with functions \((\rho^+(T), \rho^-(T), f_d(T), \ldots)\) and related to EOS of some metals (Al, Cu, U, . . .) in the critical region, here \(f_d = (\rho^+ + \rho^-)(2p_s)^{-1} - 1\) is the mean diameter. Note that this model group has a dimensionless form. This form will make it possible to use our results to describe thermal properties of some metals in the critical region.

The choice of SF\(_6\) can be explained by the fact that data \((\rho^+, \rho^-, T)\) [16] are new and have a small experimental error in comparison with the results of [3].

2. Joint application of the Clapeyron–Clausius and the Gibbs–Duhem equations

The equation for density \(\rho^-(T)\) is developed in paper [25] and based on the Clapeyron–Clausius equation

\[
\frac{dp_s}{dT} = \frac{r(T)}{T(v^-(T) - v^+(T))},
\]

where \(v^+ = 1/\rho^+\) is the specific volume of saturated liquid; \(v^- = 1/\rho^-\) is the specific volume of saturated steam; \(r\) is the heat of vaporization.

Let us introduce into consideration the function \(r^*(T)\) referred to as “a quasi-heat of the vaporization”:

\[
r(T) = r^*(T) \left(1 - \frac{\rho^-}{\rho^+}\right).
\]
It is possible to obtain the gas density expression from equations (2), (3) as
\[ \rho^-(T) = \frac{T \rho'_c(T)}{r^*(T)} \] (4)

The experimental data \( r^*(T) \) can be obtained by direct measurements, e.g., by evaporation method [26]. We have chosen \( r^*(T) \) in the form
\[ r^*(T) = \frac{n_0}{\rho_c} \left( d_0 + d_1|\tau|^\beta + d_2|\tau|^{2\beta} + d_3|\tau|^{\beta+\Delta} + d_4|\tau|^{1-\alpha} + \sum_{i=5}^{M} d_i|\tau|^{m(i)} \right), \] (5)

where \((d_i)\) are the constant coefficients; \(\alpha, \beta\) and \(\Delta\) are the critical indices; \(\tau = T/T_c - 1\).

If in (5) we assume the equality \(d_2 = 0\), then we will get the function \(r^*(T)\) obtained in [25] and used in the calculation of the phase equilibrium line R218 [25, 27]. In this work, the dependence \(m(i)\) is represented by an array of real numbers: \(m(i) = 1 + (i - 5)\beta\).

The model \(p_s(T)\) is taken as the equation suggested in [27]:
\[ p_s(T) = p_c \exp \left( -\frac{a_0 \tau^2}{t} \right) \left( 1 + a_1 \tau + a_2|\tau|^{2-\alpha} + a_3|\tau|^{2-\alpha+\Delta} + \sum_{i=4}^{N} a_i \tau^{n(i)} \right), \] (6)

where \(a_i\) are the constant coefficients; \(n(i)\) is the array of natural numbers; \(t = T/T_c\).

The model \(\rho^-(T)\) is obtained by substituting expressions (5) and (6) into equation (4) in the form
\[ \rho^-(T) = \frac{T \rho'_c(T)}{d_0 + d_1|\tau|^\beta + d_2|\tau|^{2\beta} + d_3|\tau|^{\beta+\Delta} + d_4|\tau|^{1-\alpha} + \sum_{i=5}^{M} d_i|\tau|^{m(i)}}. \] (7)

Let us accept the equality, \(a_1 = d_0\), and decompose:
- the numerator of (7) by a small parameter, \(\tau\);
- the denominator of (7) by a small parameter, \(\tau\).

As a result, we have obtained expressions of the liquid density in the following form near the critical point:
\[ \rho^-(T) = \rho_c \left[ 1 - \frac{d_1}{d_0}|\tau|^\beta - \frac{d_2}{d_0}|\tau|^{\beta+\Delta} - \left( \frac{d_2}{d_0} - \left( \frac{d_1}{d_0} \right)^2 \right) |\tau|^{2\beta} + O(|\tau|^{\beta+\Delta}) \right]. \] (8)

In accordance with the first four components of \(\rho^-(T)\) in (8), we have chosen the expression of the liquid density in the form
\[ \rho^+(T) = \rho_c \left( 1 + D_1|\tau|^\beta + D_2|\tau|^{\beta+\Delta} + D_3|\tau|^{2\beta} + D_4|\tau|^{1-\alpha} + \sum_{i=5}^{K} D_i|\tau|^{k(i)} \right), \] (9)

where \(D_1 = d_1/a_1\), \(D_2 = d_3/a_1\).

The equations \(\rho^-(T)\) and \(\rho^+(T)\), (7) and (9), correspond to the Yang–Yang model [12] if we accept the condition
\[ D_3 = D_3^{2\beta} = -\frac{d_2}{d_0} + \left( \frac{d_1}{d_0} \right)^2. \] (10)

As a result, we have obtained expressions of the order parameter \(f_s = (\rho^+ - \rho^-)(2\rho_c)^{-1}\) and the mean diameter \(f_d\) in the following forms near the critical point:
\[ f_s = \frac{d_1}{d_0}|\tau|^\beta + \frac{d_3}{d_0}|\tau|^{\beta+\Delta}, \quad f_d = -\left( \frac{d_2}{d_0} - \left( \frac{d_1}{d_0} \right)^2 \right) |\tau|^{2\beta}. \] (11)
Note that equations (7), (9) correspond to the Wegner’s model \( (f_d(T \to T_c) \sim |\tau|^{1-\alpha}) \) [5] if we accept the condition

\[
D_3 = D_3^{1-\alpha} = \frac{d_2}{d_0} - \left( \frac{d_1}{d_0} \right)^2.
\]  

(12)

It is of interest to obtain numerical parameters of equations (6), (7) and (9) under the following conditions:

- models \( p_s(T), \rho^-(T), \rho^+(T) \) must satisfy some requirements of ST;
- the parameters of these models should be evaluated by statistical processing experimental data [16]; the errors of these data are characterized by the limits: \( \delta \rho_{\text{exp}} \leq 0.017\% \) and \( \delta p_{\text{exp}} \leq 0.017\% \) in the temperature range \( T_{\text{tr}} \leq T \leq 0.99T_c \).

In our approach, we have selected the minimization criterion \( U \), which includes some root-mean-square deviations \( S \) (RMS):

- RMS of the liquid density \( (S^+) \);
- RMS of the gas density \( (S^-) \);
- RMS of the saturation pressure \( (S_p) \).

Some conditions have been used:

- critical indexes should be the same for all the models; they follow from ST \( (\alpha = 0.11, \beta = 0.325, \Delta = 0.51) \);
- parameters \( (T_c, \rho_c) \) should be common.

The functional \( U \) has been accepted in the form that includes some components:

\[
U_{p_s} = \sum_{j=1}^{N_1} [Q_{p_s,j}(p_{s,j} - p_{s,j}^{(e)})]^2,
\]  

(13)

\[
U_{\rho^-} = \sum_{j=1}^{N_3} [Q_{\rho^-,j}(\rho_{j}^- - \rho_{j}^{-,\text{e}})]^2,
\]  

(14)

\[
U_{\rho^+} = \sum_{j=1}^{N_3} [Q_{\rho^+,j}(\rho_{j}^+ - \rho_{j}^{+,\text{e}})]^2,
\]  

(15)

where \( Q^*_{X,j} = 1/\delta X_j^{\text{exp}} \) is “the weight” of point \( j \) for a property \( X \), the properties \( X_j, X_j^{(e)} \) and experimental values of the property \( X = p_s, \rho^- \) or \( \rho^+ \) are calculated; \( \delta X_j^{\text{exp}} \) are values of the total uncertainty of the measurements of point \( j \) for the corresponding property \( X \) [16].

We have got the critical parameters: \( \rho_c = 3.754 \text{ MPa}, \rho_c = 742.26 \text{ kg/m}^3, T_c = 318.71 \text{ K} \). As a result of processing, we have obtained two variants which are associated with the parameters and coefficients of equations (6), (7) and (9). If equation (9) with \( D_3^{2/3} \) (variant I) is accepted, we have got the values given in tables 1–3.

If equation (9) with \( D_3^{1-\alpha} \) (variant II) is accepted, we have got the values presented in table 4. The other coefficients and parameters of variant II are the same as the values of variant I.

Our comparison has shown that the calculated values of the properties \( (p_s, \rho^-, \rho^+) \) (variant I) agree with the corresponding experimental data [16] within the specified experimental error (figures 1–3). We have identified some deviations \( S \) of the original data:

- deviation \( S^+ = 0.0012\% \) while local deviations \( \delta \rho^+ = (\rho_{j}^{+,(e)} - \rho_{j}^{+,\text{r}})/\rho_{j}^{+,\text{e}} \times 100\% \) for two points satisfy the condition \( 0.01 < |\delta \rho^+ | < 0.03\% \).
Table 1. Parameters and coefficients of equation (6).

| i | a_i | i | a_i | n(i) |
|---|-----|---|-----|------|
| 0 | 6.0 | 4 | 8.84873064 | 3    |
| 1 | 7.061104518 | 5 | 15.21516103 | 4    |
| 2 | 22.90993576 | 6 | 18.96462416 | 5    |
| 3 | -17.71425160 | — | — | —    |

Table 2. Parameters and coefficients of equation (7).

| i | d_i | i | d_i | m(i) |
|---|-----|---|-----|------|
| 1 | 8.96467066052 | 5 | -664.492800470 | 1    |
| 2 | 147.213093868 | 6 | -8.33257701440 | 1.325 |
| 3 | -1654.52758013 | 7 | 24.2184182876 | 1.65  |
| 4 | 2174.91159478 | — | — | —    |

Table 3. Parameters and coefficients of equation (9). Variant I.

| i | D_i | k(i) | i | D_i | k(i) |
|---|-----|------|---|-----|------|
| 4 | 1812.62815707 | — | 9 | -494101.558419 | 2.3   |
| 5 | -3876.03182316 | 1 | 10 | 761286.471195 | 2.625 |
| 6 | 14339.2562002 | 1.325 | 11 | -749980.684616 | 2.950 |
| 7 | -64891.5035288 | 1.65 | 12 | 426369.372177 | 3.275 |
| 8 | 215266.255249 | 1.975 | 13 | -106328.161789 | 3.60  |

Table 4. Parameters and coefficients of equation (9). Variant II.

| i | D_i | k(i) | i | D_i | k(i) |
|---|-----|------|---|-----|------|
| 4 | 293.613921561 | — | 7 | 76.1722789420 | 1.65  |
| 5 | -62.2427517493 | 1 | 8 | -54.5947138003 | 1.975 |
| 6 | -52.8738382067 | 1.325 | 9 | 16.4855138438 | 2.30  |

- deviation \( S^- = 0.011\% \) while local deviations \( \delta \rho^- = \rho^{(-)(e)} - \rho^{-(r)} / \rho^{(-)(e)} \times 100\% \) satisfy the condition \( 0.02 < |\delta \rho^-| < 0.35\% \);
- deviation \( S_p = 0.0012\% \) while local deviations \( \delta p_s = (p_s^{(c)} - p_s^{(r)}) / p_s^{(c)} \times 100\% \) for seven points satisfy the condition \( 0.002 < \delta p_s < 0.0055\% \) in the neighborhood of the critical point.

A significant difference between \( S^+ \) and \( S^- \) is due to the fact that the experimental errors of these data do not coincide (table 5) [16].

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• deviation \( S^- = 0.011\% \) while local deviations \( \delta \rho^- = \rho^{(-)(e)} - \rho^{-(r)} / \rho^{(-)(e)} \times 100\% \) satisfy the condition \( 0.02 < |\delta \rho^-| < 0.35\% \);
• deviation \( S_p = 0.0012\% \) while local deviations \( \delta p_s = (p_s^{(c)} - p_s^{(r)}) / p_s^{(c)} \times 100\% \) for seven points satisfy the condition \( 0.002 < \delta p_s < 0.0055\% \) in the neighborhood of the critical point.

A significant difference between \( S^+ \) and \( S^- \) is due to the fact that the experimental errors of these data do not coincide (table 5) [16].
In variant II, the calculated data \( \rho^+ \) have been defined for several conditions related to the number \( N \) of coefficients \( (D_i) \); even with the increase of \( N \) from 9 to 13, we have failed to describe the data \( \rho^+ \) [16] within the experimental error (figure 4).

The analysis shows that the authors of [16] have used different values of the critical temperature:

- \( T_c = 318.723 \) K is used for \( \rho^-(T) \) and \( \rho^+(T) \);
- \( T_c = 318.706 \) K is used for \( \rho_s(T) \); this value is called pseudo-critical temperature in [16].

Within our approach to describing the coexistence curve of sulfur hexafluoride (7) and (9), we observe the law of rectilinear diameter (figure 5). Note that the law of rectilinear diameter is satisfied in both cases: variant I (10) and variant II (12).

It is interesting to compare models \( \rho^+(T) \), \( \rho^-(T) \) with EOS [3] which have a wide range of applicability (from the melting line to 625 K and pressures up to 150 MPa including the critical region). This EOS has allowed us to determine data \( (\rho^+, \rho^-, T) \) in the interval \( 5 \times 10^{-6} < \tau < 10^{-3} \). Our analysis has shown that the data \( (\rho^+, T) \) [3] are systematically higher than the data \( (\rho^+, T) \) relating to \( \rho^+(T) \). Deviation \( \delta \rho^+_{\text{max}} \) is defined as \( \delta \rho^+_{\text{max}} = 3.1\% \) at \( \tau = 5 \times 10^{-6} \). Calculated data \( (\rho^-, T) \) [3] are systematically lower than data \( (\rho^-, T) \) relating to \( \rho^-(T) \). Deviation \( \delta \rho^-_{\text{max}} \) is defined as \( \delta \rho^-_{\text{max}} = 3.3\% \) at \( \tau = 5 \times 10^{-6} \).

### Table 5. Deviation characteristics in the critical region.

| \( T, \text{K} \) | 316.8 | 317.5 | 318.0 | 318.35 | 318.55 | 318.65 | 318.69 |
|-----------------|-------|-------|-------|--------|--------|--------|--------|
| \( |\delta \rho^+,\text{exp}|, \% \), [16] | 0.017 | 0.018 | 0.025 | 0.040 | 0.11 | 0.30 | — |
| \( |\delta \rho^+,\% \), calculated by (9) | 0.007 | 0.005 | 0.006 | 0.0009 | 0.015 | 0.028 | — |
| \( |\delta \rho^-,\text{exp}|, \% \), [16] | 0.027 | 0.037 | 0.051 | 0.094 | 0.21 | 0.54 | 0.73 |
| \( |\delta \rho^-,\% \), calculated by (7) | 0.005 | 0.008 | 0.004 | 0.021 | 0.001 | 0.007 | 0.34 |

![Figure 1. Deviations \( \delta p_s \) of the values \( p_s(T) \) calculated by equation (6) from experimental and calculated values \( p_s(e) \): 1—[16]; 2—[28].](image-url)
Figure 2. Deviations $\delta \rho^-$ of the values $\rho^{-(r)}$ calculated by equation (8) from experimental and calculated values $\rho^{-(e)}$: 1—[16]; 2—[28].

Figure 3. Deviations $\delta \rho^+$ of the values $\rho^{+(r)}$ calculated by equation (9) from experimental and calculated values $\rho^{+(e)}$: 1—[16]; 2—[28].

3. Discussion
The joint group of models $\rho^-(T)$, $\rho^+(T)$, $p_s(T)$ has been developed. We have considered two variants of equations (6), (7) and (9). Variant I can be related to the Yang–Yang model [12]. Variant II can be related to the Wegner model [5].

Our methodical approach allows us to investigate models $\rho^-(T)$, $\rho^+(T)$, $p_s(T)$ jointly due to the Gibbs–Duhem equation. Model $\rho^-(T)$ is developed on the basis of the Clapeyron–Clausius equation and the function $r^*(T)$. This approach allows us to ensure consistency between such heterogeneous thermodynamic functions ($\rho^-(T)$, $\rho^+(T)$, $p_s'(T)$, $r^*(T)$) in the temperature range $T_{tr} \leq T \leq T_c$.

The proposed approach is tested on the example of SF$_6$ phase equilibrium lines description.

We have evaluated regulated parameters of the models $p_s(T)$, $\rho^-(T)$, $\rho^+(T)$ by statistical processing reliable experimental data [16]. Our analysis confirm that deviations of the data from these models do not exceed the experimental error of data [16] in a wide temperature range; thus, deviations can be characterized by RMS in the following form: $S^+ = 0.0012\%$, $S^- = 0.011\%$ and $S_p = 0.0022\%$. 
Figure 4. Deviations $\delta \rho^+$ calculated by equation (9) from experimental values $\rho^{+,(e)}$ [16]: 1—variant II; 2—variant I.

Figure 5. The coexistence curve of the sulfur hexafluoride: 1—the coexistence curve calculated with the help of (7) and (9); 2—the experimental points [16]; 3—the values $f_z = (\rho^+ + \rho^-)/2$ calculated with the help of (7) and (9); 4—the values $f_z$ calculated with the help of [16]; 5—the values $f_z$ calculated with the help of paper [28]; 6—the coexistence curve calculated with the help of [28].

A comparative study of models $\rho^-(T)$, $\rho^+(T)$ with data $(\rho^+\), \rho^-, T)$, which are determined on the basis of EOS [3] in the interval $5 \times 10^{-6} < \tau < 10^{-3}$ is carried out. Systematic deviation of data $(\rho^+,\rho^-, T)$ related to [3] has been estimated. Thus, data $(\rho^+, T)$ [3] are placed higher and deviations $\delta \rho^+$ are placed in the range $0.1 < \delta \rho^+ < 3.1\%$.

4. Conclusion
Our analysis has shown that the model of average diameter of Wegner describes the experimental data about the phase equilibrium line of sulfur hexafluoride with less error than the Yang–Yang model.
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