Application of reduced variables in problems of analytical thermodynamics

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The article shows, using specific examples, that analytical expressions of equations of state, thermodynamic relations and coefficients, process parameters can be written in terms of the reduced variables. In this case, analogues of thermodynamic parameters used in practice are obtained, which are dimensionless and applicable for all real gases described by a specific equation of state.
Keywords: equation of state, critical parameters, reduced variables, Joule-Thomson effect, inversion curve, thermodynamic coefficients.

В статье на конкретных примерах показано, что аналитические выражения уравнений состояния, термодинамических соотношений и коэффициентов, параметров процессов могут быть записаны в терминах приведенных переменных. При этом получаются аналоги используемых на практике термодинамических величин, являющиеся безразмерными и применимыми для всех реальных газов, описываемых конкретным уравнением состояния.
Ключевые слова: уравнение состояния, критические параметры, приведенные переменные, эффект Джоуля-Томсона, кривая инверсии, термодинамические коэффициенты.

Introduction. Theoretical and numerical studies of the real gases properties have significant practical importance for a variety of engineering and scientific applications. And the basis for these studies is the equations of state for real gases [1]–[9].

One of the elements of comparing the equations of state with experimental data is the consideration of the critical state of a substance, at which the differences in the physical properties of liquid and vapor disappear. On the isotherm graph at a critical temperature, this state corresponds to a single (critical) point, which is the inflection point of the isotherm. Mathematically, this means fulfilling the conditions

$$\left(\frac{\partial P}{\partial V}\right)_{T_{CR}} = 0, \qquad \left(\frac{\partial^2 P}{\partial V^2}\right)_{T_{CR}} = 0. \qquad (1)$$

Relations (1) form a system of equations, the solution of which allows us to express the characteristics of the critical state (critical temperature T_{CR}, critical pressure P_{CR} and critical volume V_{CR}) using the parameters of the equation of state. In many cases, it is convenient to use critical parameters as a unit of measurement of thermodynamic quantities. This means a transition in relationships to reduced, or relative dimensionless variables

$$\widetilde{V} = \frac{V}{V_{CR}}, \qquad \widetilde{T} = \frac{T}{T_{CR}}, \qquad \widetilde{P} = \frac{P}{P_{CR}}. \qquad (2)$$

These variables give the following reduced form of the van der Waals equation [1]

$$\widetilde{P} = \frac{8\widetilde{T}}{3\widetilde{V}-1} - \frac{3}{\widetilde{V}^2}. \qquad (3)$$

In the reduced equation of state there are no parameters a, b, which describe the properties of specific gases. Such an equation describes at once all substances for which the van der Waals equation is valid.

On this basis, the concept of corresponding states (states with the same values of the reduced variables) was introduced for various substances and the law of corresponding states was formulated [1]–[3]: if different gases have the same values for two out of three reduced variables, then the values of the third reduced variable coincide. This makes it possible to obtain results that are common to all substances in thermodynamically similar states, and to study the properties of some gas if the properties of another gas are known. The use of the reduced variables is very convenient, since they give results that are not related to a specific substance, but rather characterize a specific equation of state. This form of writing equations of state uses only dimensionless variables, constant fac-

tors, and functions that depend on dimensionless variables. Another advantage of using reduced (dimensionless) variables is the rare possibility of comparing experimental results expressed in different units of measurement used in the technical literature.

In papers [10]–[14], in the framework of the development of the principle of corresponding states, reduced analogues of various thermodynamic quantities were introduced and their numerical and graphical study was carried out for various equations of state for real gases.

In this paper we provide the review of some problems solved using the reduced variables in the analytical thermodynamics of real gases and the results obtained.

Examples of reduced equations of state for real gases. In this part of the review we will give four examples of reduced equations of state that are currently generally accepted and widely used.

1. Redlich-Kwong equation of state. One of the best versions of the equation of state for real gases up to the present day remains the Redlich-Kwong equation [4], in which the dependence of the term taking into account intermolecular repulsion on temperature was written as $1/\sqrt{T}$. The molar form of the equation and its critical parameters are

$$P = \frac{RT}{V-b} - \frac{a}{\sqrt{T}V(V+b)}; \tag{4}$$

$$V_{CR} = \frac{b}{\xi}, \qquad T_{CR} = \left(\frac{3a\xi^2}{bR}\right)^{2/3}, \qquad P_{CR} = \left(\frac{Ra^2\xi^7}{3b^5}\right)^{1/3}. \tag{5}$$

In formula (5) a constant $\xi = \sqrt[3]{2} - 1 = 0{,}259921 \cong 0{,}260$ is introduced. It satisfies the condition $1 - \xi = \xi(\xi+1)(\xi+2)$. The reduced Redlich-Kwong equation of state has the form

$$\widetilde{P} = \frac{3\widetilde{T}}{\widetilde{V} - \xi} - \frac{1}{\xi\widetilde{T}^{1/2}\widetilde{V}(\widetilde{V} + \xi)}. \tag{6}$$

2. Soave-Redlich-Kwong equation of state. Understanding the structure of the Redlich-Kwong equation [4] as containing a temperature–dependent parameter led to the formulation of the Soave-Redlich-Kwong equation [5]

$$\left(P + \frac{a(\widetilde{T})}{V(V+b)}\right)(V - b) = RT. \tag{7}$$

Here the dependencies on the reduced temperature and the acentric Pitzer factor ω [5] are introduced into parameter a with the same critical parameters and ξ

$$a(\widetilde{T}) = a_{CR}\alpha(\widetilde{T}); \qquad a_{CR} = \frac{0{,}42748\, R^2 T_{CR}^2}{P_{CR}}; $$

$$\alpha(\widetilde{T}) = [1 - m(1 - \sqrt{\widetilde{T}})]^2; \qquad m = 0{,}480 + 1{,}574\omega - 0{,}175\omega^2. \tag{8}$$

Finally, in reduced form, the Soave-Redlich-Kwong equation looks like

$$\widetilde{P} = \frac{3\widetilde{T}}{\widetilde{V} - \xi} - \frac{\alpha(\widetilde{T})}{\xi\widetilde{V}(\widetilde{V} + \xi)}. \tag{9}$$

3. Ishikawa-Chung-Lu equation of state. The use of temperature–dependent parameters was further developed in [6], in which an equation with a new form of the «repulsive» term and two temperature-dependent parameters was proposed. For one mole of gas, this equation is written as

$$P = \frac{RT(2V + b(T))}{V(2V - b(T))} - \frac{a(T)}{\sqrt{T}V(V + b(T))}. \tag{10}$$

In (10) the parameters have the next structure

$$a(T) = \Omega_a \alpha(\widetilde{T})\frac{R^2 T_{CR}^{5/2}}{P_{CR}}, \qquad b(T) = \Omega_b \beta(\widetilde{T})\frac{RT_{CR}}{P_{CR}}, \qquad \alpha(1) = \beta(1) = 1. \tag{11}$$

The functions $\alpha(\widetilde{T})$, $\beta(\widetilde{T})$ in [6] were presented in the form of third–order polynomials in the reduced temperature, the coefficients of which for each substance were selected based on experimental data. In the reduced variables, the Ishikawa-Chang-Lu equation has the form [15]

$$\widetilde{P} = \frac{\widetilde{T}(2\chi\widetilde{V} + \beta(\widetilde{T}))}{\Omega_b \chi\widetilde{V}(2\chi\widetilde{V} - \beta(\widetilde{T}))} - \frac{\Omega_a \alpha(\widetilde{T})}{\Omega_b^2 \sqrt{\widetilde{T}} \chi\widetilde{V}(\chi\widetilde{V} + \beta(\widetilde{T}))} , \tag{12}$$

where

$$\Omega_a = \frac{8(\chi+1)^3}{3(6\chi+1)^2} = 0,46712311 ; \quad \Omega_b = \frac{2}{6\chi+1} = 0,10876233 ; \quad \chi = 2,89812008 . \tag{13}$$

4. Fogel'son-Likhachev equation of state. In paper [7], the molar equation of state for real gas was considered, which containing five parameters and having the meaning of a generalization of the van der Waals equation

$$\left(P + \frac{a}{(V+c)^k T^m}\right)(V - b) = RT . \tag{14}$$

Special cases of equation (14) are the next: van der Waals equation ($c = 0$, $k = 2$, $m = 0$), Berthelot equation ($c = 0$, $k = 2$, $m = 1$), Clausius equation ($k = 2$, $m = 1$). Reduced form of equation (14) is

$$\left[\widetilde{P} + \frac{A}{(\widetilde{V}+\widetilde{c})^k \widetilde{T}^m}\right](\widetilde{V} - \widetilde{b}) = B\widetilde{T} , \tag{15}$$

where following notations are used

$$A = \frac{(k+1)(1+\widetilde{c})^k}{k-1} , \quad B = \frac{4k(1+\widetilde{c})}{k^2 - 1}, \quad \widetilde{b} = \frac{b}{V_{CR}}, \quad \widetilde{c} = \frac{c}{V_{CR}} .$$

Description of the Joule–Thomson effect in the reduced variables. Numerous engineering applications are based on the use of a subsystem–equilibrium isenthalpic ($dW = 0$) process of pumping real gas through a porous partition [1]–[3]. During this process, a change in the temperature of the real gas is observed (Joule-Thomson effect). Mathematically, this change is characterized by the Joule-Thomson coefficient. In work [10] in determining this coefficient

$$\mu_{JT} = \left(\frac{\partial T}{\partial P}\right)_W = -\widetilde{\lambda} \frac{P_{CR}}{C_P}\left(\frac{\partial V}{\partial P}\right)_T \tag{16}$$

a dimensionless quantity $\widetilde{\lambda}$ is highlighted, which we will call the reduced Joule-Thomson parameter, and which is expressed in terms of reduced variables

$$\lambda = P_{CR}\widetilde{\lambda} ; \quad \widetilde{\lambda} = \widetilde{V}\left(\frac{\partial \widetilde{P}}{\partial \widetilde{V}}\right)_{\widetilde{T}} + \widetilde{T}\left(\frac{\partial \widetilde{P}}{\partial \widetilde{T}}\right)_{\widetilde{V}} . \tag{17}$$

This parameter has the same value for gases in the corresponding states. In the physical range $\left(\frac{\partial V}{\partial P}\right)_T < 0$, and the isobaric heat capacity $C_P > 0$, it means that the signs of μ_{JT} and $\widetilde{\lambda}$ in (16) are the same. When pressure decreases ($d\widetilde{P} < 0$), which follows from the condition of the Joule-Thomson process, two options for changing the temperature are possible depending on the sign of the parameter: $\widetilde{\lambda} > 0 \rightarrow d\widetilde{T} < 0$ or $\widetilde{\lambda} < 0 \rightarrow d\widetilde{T} > 0$. The first option corresponds to the positive Joule-Thomson effect (the gas cools down), the second – to the negative one (the gas heats up).

Therefore, to determine the sign of the Joule-Thomson effect, it is enough to know the sign of the reduced parameter $\widetilde{\lambda}$. Accordingly, the reduced inversion temperature $\widetilde{T}_i$ can be obtained from condition $\widetilde{\lambda} = 0$.

The reduced Joule-Thomson coefficient $\widetilde{\lambda}$ for the Redlich-Kwong equation was obtained in [10] in the form

$$\widetilde{\lambda} = \frac{5\widetilde{V} + 3\xi}{2\xi\widetilde{T}^{1/2}\widetilde{V}(\widetilde{V} + \xi)^2} - \frac{3\xi\widetilde{T}}{(\widetilde{V} - \xi)^2} . \tag{18}$$

Then taking into account the condition $\widetilde{\lambda} = 0$, we found expressions for the reduced inversion temperature and the reduced pressure at the inversion points through the reduced volume

$$\widetilde{T}_i(\widetilde{V}) = \left[\frac{(5\widetilde{V} + 3\xi)(\widetilde{V} - \xi)^2}{6\xi^2\widetilde{V}(\widetilde{V} + \xi)^2}\right]^{2/3} \tag{19}$$

$$\widetilde{P}_i(\widetilde{V}) = \frac{3^{1/3}(5\widetilde{V}^2 - 4\xi V - 5\xi^2)}{[4\xi^4\widetilde{V}^2(5\widetilde{V} + 3\xi)(\widetilde{V} + \xi)^4(\widetilde{V} - \xi)^2]^{1/3}} . \tag{20}$$

When considering the reduced Ishikawa-Chang-Lu equation, the reduced Joule-Thomson coefficient was obtained in paper [12] in the form (new parameter $x = \chi \widetilde{V}$)

$$
\begin{aligned}
\widetilde{\lambda} = &\frac{3(6\chi + 1)}{(\chi + 1)^3} x(x + \beta(\widetilde{T}))^2 \left[\widetilde{T}^2 \left(\frac{d\beta}{d\widetilde{T}} \right) - \widetilde{T}\beta(\widetilde{T}) \right] + \\
&+ (2x - \beta(\widetilde{T}))^2 (x + \beta(\widetilde{T})) \left[\frac{3\alpha(\widetilde{T})}{2\sqrt{\widetilde{T}}} - \sqrt{\widetilde{T}} \left(\frac{d\beta}{d\widetilde{T}} \right) \right] + \\
&+ (2x - \beta(\widetilde{T}))^2 \left[\sqrt{\widetilde{T}} \alpha(\widetilde{T}) \left(\frac{d\beta}{d\widetilde{T}} \right) + \frac{x}{\sqrt{\widetilde{T}}} \alpha(\widetilde{T}) \right] = 0.
\end{aligned}
\tag{21}
$$

Thus, at a fixed temperature, the condition $\widetilde{\lambda} = 0$ leads to a cubic equation in the parameter x.

Now we present the graphical results of the numerical analysis:
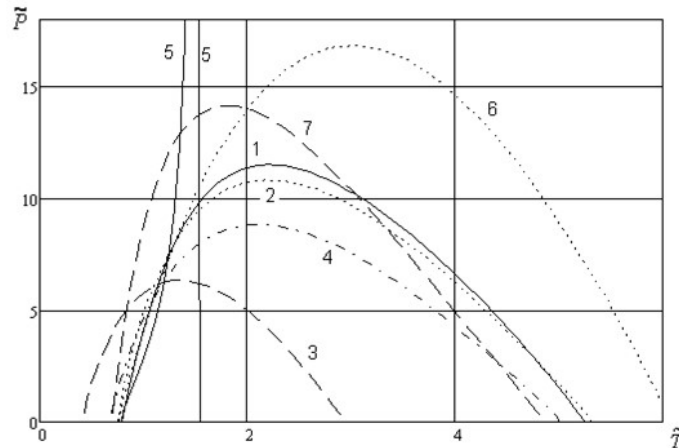
Picture 1 – Graphs of inversion curves at $\omega = 0$: 1 – based on the results of processing experimental data [15]; 2 – for the Redlich-Kwong equation; 3 – for the Soave-Redlich-Kwong equation; 4 – for the Peng-Robinson equation (additional one); 5, 6, 7 – for the Ishikawa-Chang-Lu equation with various parameters [12]

The theoretical inversion curves in the reduced variables were compared with the data of [15], in which a generalized experimental inversion curve in the reduced variables was constructed for substances with a small acentricity factor (argon, methane, nitrogen, oxygen, xenon, krypton, carbon monoxide) in the form:

$$
\widetilde{P} = \sum_{k=0}^{6} \beta_k \widetilde{T}^k .
\tag{22}
$$

In (22) generalized curve parameters are the following: $\beta_0 = -32.5209374$, $\beta_1 = 65.6922312$, $\beta_2 = -39.738430$, $\beta_3 = 12.9300299$, $\beta_4 = -2.46176904$, $\beta_5 = 0.25378553$, $\beta_6 = -0.0109865$.

Picture 1 shows the inversion curve plots obtained from various equations of state in reduced form. Each curve limits its area of positive effect from above. The best agreement with experiment is observed for the theoretical inversion curve obtained on the base of Redlich-Kwong equation.

Reduced thermodynamic coefficients of real gases. Thermodynamic (thermal) coefficients [2] characterize the thermal and elastic properties of substances. The mathematical definition of isobaric volume expansion coefficient, isochoric thermal pressure coefficient and isothermal compressibility has the form, respectively, [2]

$$
\alpha_P = \frac{1}{V} \left(\frac{\partial V}{\partial T} \right)_P ; \qquad \beta_V = \frac{1}{P} \left(\frac{\partial P}{\partial T} \right)_V ; \qquad k_T = -\frac{1}{V} \left(\frac{\partial V}{\partial P} \right)_T .
\tag{23}
$$

Well–known relation for derivatives $(\partial P / \partial V)_T (\partial V / \partial T)_P (\partial T / \partial P)_V = -1$ and definition (23) give a mathematical connection between them

$$
\alpha_P = \beta_V k_T P .
\tag{24}
$$

Along with the coefficients listed above, we can also express the difference between the isobaric and isochoric heat capacities of the gas (analogue of Mayer's relation), determined by the relation

$$\Delta c = c_P - c_V = T\left(\frac{\partial P}{\partial T}\right)_V \left(\frac{\partial V}{\partial T}\right)_P = -T\left(\frac{\partial P}{\partial T}\right)_V^2 \left(\frac{\partial P}{\partial V}\right)_T^{-1}. \tag{25}$$

In [13], reduced forms of determining these thermal coefficients (23) and the difference (25) are proposed:

$$\alpha_P = \frac{1}{V}\left(\frac{\partial V}{\partial T}\right)_P = \frac{1}{T_{кр}}\tilde{\alpha}_P; \qquad \tilde{\alpha}_P = \frac{1}{\tilde{V}}\left(\frac{\partial \tilde{V}}{\partial \tilde{T}}\right)_{\tilde{P}} = -\frac{1}{\tilde{V}}\left(\frac{\partial \tilde{P}}{\partial \tilde{T}}\right)_{\tilde{V}}\left(\frac{\partial \tilde{P}}{\partial \tilde{V}}\right)_{\tilde{T}}^{-1};$$

$$\beta_V = \frac{1}{P}\left(\frac{\partial P}{\partial T}\right)_V = \frac{1}{T_{кр}}\tilde{\beta}_V; \qquad \tilde{\beta}_V = \frac{1}{\tilde{P}}\left(\frac{\partial \tilde{P}}{\partial \tilde{T}}\right)_{\tilde{V}};$$

$$k_T = -\frac{1}{V}\left(\frac{\partial V}{\partial P}\right)_T = \frac{1}{P_{кр}}\tilde{k}_T; \qquad \tilde{k}_T = -\frac{1}{\tilde{V}}\left(\frac{\partial \tilde{V}}{\partial \tilde{P}}\right)_{\tilde{T}} = -\frac{1}{\tilde{V}}\left(\frac{\partial \tilde{P}}{\partial \tilde{V}}\right)_{\tilde{T}}^{-1};$$

$$\Delta c = \frac{P_{кр}V_{кр}}{T_{кр}}\Delta\tilde{c}; \qquad \Delta\tilde{c} = -\tilde{T}\left(\frac{\partial \tilde{P}}{\partial \tilde{T}}\right)_{\tilde{V}}^2\left(\frac{\partial \tilde{P}}{\partial \tilde{V}}\right)_{\tilde{T}}^{-1}. \tag{26}$$

In this case, formula (24) for the reduced coefficients has the form $\tilde{\alpha}_P = \tilde{\beta}_V \tilde{k}_T \tilde{P}$.

Let us consider the reduced coefficients in the Redlich-Kwong gas model. From (6) we find the partial derivatives appearing in (26)

$$\left(\frac{\partial \tilde{P}}{\partial \tilde{T}}\right)_{\tilde{V}} = \frac{3}{\tilde{V} - \xi} + \frac{1}{2\xi\tilde{T}^{3/2}\tilde{V}(\tilde{V} + \xi)}; \qquad \left(\frac{\partial \tilde{P}}{\partial \tilde{V}}\right)_{\tilde{T}} = \frac{2\tilde{V} + \xi}{\xi\tilde{T}^{1/2}\tilde{V}^2(\tilde{V} + \xi)^2} - \frac{3\tilde{T}}{(\tilde{V} - \xi)^2}. \tag{27}$$

Substituting (27) into definition (26) and taking into account equation (6), we received: two expressions for $\tilde{\alpha}_P$ and $\tilde{\beta}_V$, one expression for $\tilde{k}_T$ and for $\Delta\tilde{c}$. We list them below

$$\tilde{\alpha}_P(\tilde{V},\tilde{T}) = \frac{(\tilde{V} + \xi)(\tilde{V} - \xi)[6\xi\tilde{T}^{3/2}\tilde{V}(\tilde{V} + \xi) + \tilde{V} - \xi]}{2\tilde{T}[3\xi\tilde{V}^2\tilde{T}^{3/2}(\tilde{V} + \xi)^2 - (2\tilde{V} + \xi)(\tilde{V} - \xi)^2]},$$

$$\tilde{\alpha}_P(\tilde{P},\tilde{V},\tilde{T}) \cong \frac{(\tilde{V}^2 - \xi^2)[9\tilde{T} - \tilde{P}(\tilde{V} - \xi)]}{2\tilde{T}[3\tilde{T}\tilde{V}(\tilde{V} + \xi) - (2\tilde{V} + \xi)(\tilde{V} - \xi)(3\tilde{T} - \tilde{P}\tilde{V} + \tilde{P}\xi)]},$$

$$\tilde{\beta}_V(\tilde{V},\tilde{T}) = \frac{6\xi\tilde{V}\tilde{T}^{3/2}(\tilde{V} + \xi) + \tilde{V} - \xi}{2\tilde{T}[3\xi\tilde{V}\tilde{T}^{3/2}(\tilde{V} + \xi) - \tilde{V} + \xi]},$$

$$\tilde{\beta}_V(\tilde{P},\tilde{V},\tilde{T}) = \frac{3}{\tilde{P}(\tilde{V} - \xi)} + \frac{1}{2\xi\tilde{P}\tilde{T}^{3/2}\tilde{V}(\tilde{V} + \xi)},$$

$$\tilde{k}_T(\tilde{V},\tilde{T}) = \frac{\xi\tilde{V}\sqrt{\tilde{T}}(\tilde{V} - \xi)^2(\tilde{V} + \xi)^2}{3\xi\tilde{V}^2\tilde{T}^{3/2}(\tilde{V} + \xi)^2 - (2\tilde{V} + \xi)(\tilde{V} - \xi)^2},$$

$$\Delta\tilde{c}(\tilde{V},\tilde{T}) = \frac{[6\xi\tilde{V}\tilde{T}^{3/2}(\tilde{V} + \xi) + \tilde{V} - \xi]^2}{4\xi\tilde{T}^{3/2}[3\xi\tilde{V}^2\tilde{T}^{3/2}(\tilde{V} + \xi)^2 - (2\tilde{V} + \xi)(\tilde{V} - \xi)^2]}.$$

The coefficients and the analogue of Mayer's relation for Soave-Redlich-Kwong reduced equation of state (9) in the optimal form for calculations can be written using additional function $D(\tilde{V},\xi)$ as follows:

$$D(\tilde{V},\xi) = \frac{\tilde{V} - \xi}{\tilde{V} + \xi};$$

$$\tilde{\alpha}_P(\tilde{V},\tilde{T}) = \frac{(\tilde{V} - \xi)[3\xi\tilde{V} - \alpha'(\tilde{T})D(\tilde{V},\xi)]}{3\xi\tilde{V}^2\tilde{T} - \alpha(\tilde{T})(2\tilde{V} + \xi)D^2(\tilde{V},\xi)};$$

$$\tilde{\beta}_V(\tilde{V},\tilde{T}) = \frac{3\xi\tilde{V} - \alpha'(\tilde{T})D(\tilde{V},\xi)}{3\xi\tilde{V}\tilde{T} - \alpha(\tilde{T})D(\tilde{V},\xi)};$$

$$\tilde{k}_T(\tilde{V},\tilde{T}) = \frac{\xi\tilde{V}(\tilde{V} - \xi)^2}{3\xi\tilde{V}^2\tilde{T} - \alpha(\tilde{T})(2\tilde{V} + \xi)D^2(\tilde{V},\xi)};$$

$$\Delta \widetilde{c}(\widetilde{V}, \widetilde{T}) = \frac{\widetilde{T}\left[3\xi\widetilde{V} - \alpha'(\widetilde{T})D(\widetilde{V}, \xi)\right]^2}{3\xi^2\widetilde{V}^2\widetilde{T} - \alpha(\widetilde{T})\xi(2\widetilde{V} + \xi)D^2(\widetilde{V}, \xi)}.$$

Conclusion. In the paper we consider reduced forms of several equations of state for real gases, introduce the reduced coefficient of the Joule–Thomson effect, and obtain evident reduced forms for the inversion curves of the Joule–Thomson effect. Definitions of the reduced thermal coefficients (isobaric volume expansion coefficient, isochoric thermal pressure coefficient, isothermal compressibility) and analogue of Mayer's relation are proposed as well. Explicit reduced expressions for them are obtained for the Redlich–Kwong and the Soave–Redlich–Kwong equations of state.

This approach based on the systematic use of the reduced variables was also implemented in [14], where the numerical values of the reduced parameters of the boundary state, which determines the occurrence of the stretched liquid phase, were obtained for several most used two–parameter equations of state for real gas.

The results obtained in the paper show that almost all analytical expressions for thermodynamic relationships and parameters of gases and liquids can be formulated in terms of the reduced variables. In this case, reduced analogues of thermodynamic quantities used in practice are obtained, which are dimensionless and applicable for all real gases described by any equation of state. This approach, in a certain sense, expands the field of applicability of the law of corresponding states and can be used as a basis for analyzing the applicability of various equations of state for real gases to the description of experimental data, as well as for predicting the physical characteristics of new substances based on the law of corresponding states.

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