EQUATIONS FOR CALCULATING THE VISCOSITY COEFFICIENT OF NITROGEN AND HYDROGEN IN THE GASEOUS AND LIQUID STATES

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An equation for describing experimental data on the viscosity of gases and liquids is presented, taking into consideration the peculiarities of the configuration of isochoric sections of the viscosity coefficient surface. The effectiveness of the equation is demonstrated for nitrogen and hydrogen.

The known formulas for calculation of the viscosity of a substance, even those recently presented [1], are valid only for the gaseous or the liquid region. Hence, further effort in the development of an effective equation valid both in the gaseous and the liquid state is valuable.

Bogolyubov's theory [4] permits the writing of a formula relating the viscosity coefficient to the thermal parameters

$$\eta = \eta_0 \left(1 + B_n \rho + C_n \rho^2 + \ldots \right). \tag{1}$$

Here B_{η} , C_{η} are virial viscosity coefficients. Unfortunately, the relatively low accuracy of experimental data on viscosity permits a sufficiently accurate determination of only the second virial coefficient, which complicates direct development of an expression for viscosity in virial form. Therefore, an attempt was made to develop an equation agreeing with the theoretically based form of Eq. (1), but permitting the use of all experimental data in its formulation. Such an equation was found by examination of the configuration of the surface of the viscosity coefficient in a real gas.

Figure 1 presents the isochors of the reference network of the viscosity coefficient for liquid and gaseous nitrogen: 1) the curve $\eta_0(T)$; 2) curve of saturation; 3) data of Golubev [9, 10]; 4) Michels and Gibson [11]; 5) Vermesse et al. [13]; 6) Lazarre and Vodar [12]; 7) Van Itterbeek et al. [14].

From Fig. 1 it is evident that, if we exclude the region of greatly reduced pressures ($\omega > 2$), then for the remaining significant portion of the surface of the viscosity coefficient the equation can be written in the form

$$\eta (\rho, T) = f_1(\rho) + f_3(\rho) \varphi (T)$$
(2)

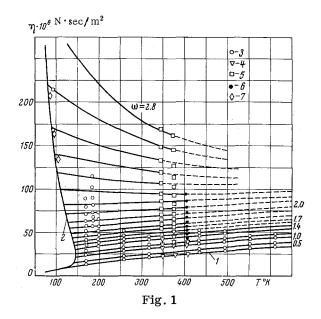
where the function $f_1(\rho)$ accounts for the systematic increase in viscosity along an isotherm with increase in density, and the second term of the equation, presented in the form of the product of two functions with separable variables, reflects the curvature of the isochor. To describe the viscosity in the region $\omega > 2$, where the isochors have a parabolic character, a term $f_2(\rho) T^{-1}$ must be added to the equation. Then the equation for the viscosity surface has the form

$$\eta (\rho, T) = f_1(\rho) + f_2(\rho) T^{-1} + f_3(\rho) \varphi (T)$$
(3)

As a result of the invariance of the equation in the form of Eq. (4), relatively linear transformations of the temperature function [5] can be used for $\varphi(T)$ in the isochor equation. At the same time the common form of the isochor configuration and the curve $\eta_{\eta}(T)$ permits the latter to be used as a temperature func-

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tion. An important argument for the choice of such a $\varphi(T)$ is the fact that values for the viscosity coefficient are easily determined at atmospheric pressure by experimental methods, and such data exist for a large number of substances. Furthermore, kinetic theory permits calculation of the viscosity of rarefied gases to an acceptable degree of accuracy with a limited number of experimental data.

With the choice of $\eta_0(T)$ as a temperature function, the equation for calculating viscosity is written in the form

$$\eta (\rho, T) = F_1(\rho) + F_2(\rho)T^{-1} + F_3(\rho)\eta_0(T)$$
(4)

 $F_1(\rho)$, $F_2(\rho)$, and $F_3(\rho)$ are polynomials in density. It is easy to see that Eq. (5) could be obtained from Eq. (1) by assumptions on the form of the temperature dependence of the virial coefficients. Comparison of the indicated equations shows that the coefficients B_{η} , C_{η} , . . . should be presented in the form

$$B_{\eta} = \frac{a_1}{\eta_0} + \frac{b_1}{\eta_0 T} + c_1, \qquad C_{\eta} = \frac{a_2}{\eta_0} + \frac{b_2}{\eta_0 T} + c_2 \tag{5}$$

Such a form for the virial coefficient expressions agrees sufficiently well with theoretical investigations [7].

The relationship in the form of Eq. (5) can be constructed from experimental data by the method of base isotherms, which has had wide use in the construction of equations of state [5]. The indicated method permits preliminary development of the net of reference values on the basis of experimental data. Then, using data on viscosity at atmospheric pressure, the function $\eta_0(T)$ is determined and described analytically.

With the known temperature function of the expression for volume functions a solution of the system is obtained, containing on its left side Eq. (5) for the given values of η_0 and T, and on its right side the equations of the base isotherms. For description of the data on viscosity of the liquefied gas, one of the base isotherms is chosen in the liquid region. In order to assure good extrapolation into high-temperature regions, it is helpful to initially determine the values of the second virial coefficient and consider them in formulation of the isotherm equations.

We will illustrate the results of the equation for gaseous and liquid viscosity using two well-studied substances, nitrogen and hydrogen.

Matching of the experimental data on nitrogen viscosity was conducted along isochoric sections in the coordinates η , T. Initially, the data of the various experimenters was graphically processed along isotherms to determine the viscosity values on the isochors. The necessary values for nitrogen density were taken both from the original works and by interpolation from tabular data [5, 6]. The resultant set of reference values for the nitrogen viscosity coefficient, based mainly on the experimental data of [8-14], occupies

a temperature range from the saturation curve to 873°K. For temperatures up to 323°K the reference data have a range of reduced pressures up to 2.9, for higher temperatures the maximum density value up to which experimental data exist is significantly reduced (up to $\omega = 1.56$ for T = 348°K, $\omega = 1.15$ for T = 523°K, and $\omega = 0.52$ for T = 873°K) and the reference values are correspondingly limited over density.

The nitrogen viscosity curve at atmospheric pressure, obtained from the smoothed data of [5], is analytically described over the temperature range 80-1300°K by the equation

$$\eta_0(T) = -0.54232 + 10.514 \tau - 1.5550 \tau^2 + 0.19238 \tau^3 - 0.013388 \tau^4 + 0.00038242 \tau^5 \qquad (\tau = T/T_*)$$
 (6)

The isotherms 77, 323.15, and 773.15°K were selected as bases. The reference data on the 77°K isotherm in the density interval from ω " to ω ' and on the 773.15°K isotherm were obtained by graphic extrapolation of the isochors and subsequent smoothing along the isotherm. In constructing the isotherm equations, values were given to the second virial coefficients which were found from reference data. After solution of the equation system an equation for calculating the viscosity of nitrogen was found in the form

$$\eta(\omega, \tau) = \alpha(\omega) + \beta(\omega) \tau^{-1} + \gamma(\omega) \eta_0(\tau)$$
 (7)

where the function $\eta_0(\tau)$ is described by Eq. (6), and the volume functions have the form

$$\alpha = 1.825 \omega + 18.961 \omega^{2} - 21.770 \omega^{3} + 29.347 \omega^{4} - 23.147 \omega^{5} + 9.072 \omega^{6} - 1.2925 \omega^{7}$$

$$\beta = 0.683 \omega - 0.740 \omega^{2} + 2.451 \omega^{2} - 12.972 \omega^{4} + 22.066 \omega^{5} - 11.344 \omega^{6} + 1.9257 \omega^{7}$$

$$\gamma = 1 + 0.01867 \omega - 0.34468 \omega^{2} + 0.49498 \omega^{3} - 0.67044 \omega^{4} + 0.5482 \omega^{7} \omega^{5} - 0.21122 \omega^{6} + 0.02862 \omega^{7}$$
(8)

In calculations for Eq. (8) the following values for the critical parameters of nitrogen were taken: $T_* = 126.25$ °K, $\rho_* = 0.304$ g/cm³. The dimensions of η were 10^{-6} N·sec/m².

The values for the dynamic viscosity coefficient of nitrogen obtained from Eq. (8) were compared with multiple experimental data. Without introducing detailed comparison tables, we will indicate that in the temperature range above 273°K the equation does describe an overwhelming majority of the experimental data of Golubev et al. [8-10] as well as that of Michels and Gibson [11], in the pressure range of 490-980 bar, with an error of less than 1%. Greater departures are observed upon comparison with the experimental data of [12, 13] for the region of very high pressure (up to 4880 bar); nevertheless, at the majority of points (34 out of 42) these departures are no greater than 2%. At temperatures below 273°K, especially in the liquid range, where greater departures between the data of the various authors may be seen, the differences between the calculated viscosity values and those determined experimentally increase. However, a satisfactory (essentially in the range 2-3%) agreement is maintained with the data of Van Itterbeek et al. [14], Goldman [15], Flynn et al. [16], and the majority of the data of Shepeleva and Golubev [9].

In establishing the viscosity coefficient for parahydrogen, use was made of the detailed data of Diller [17], with consideration of the corrections thereto recently accomplished by Lyusternik and Mamonov [18]. The data indicated are for a temperature range of 14-100°K and pressures of 1-345 atm (ω = 2.8), including the saturation curve, eliminating the need of constructing reference value sets. This parameter range is most interesting from the viewpoint of equation formulation, since it contains liquid and dense gas, as well as gas at moderate and low pressure.

Analogously to the nitrogen case, initially the data [17] on parahydrogen viscosity at atmospheric pressure were analytically described with an error less than 1% with the help of the equation

$$\eta_0\left(T\right) = -0.11532 + 57.973 \frac{T}{100} - 20.185 \left(\frac{T}{100}\right)^2 + 4.9626 \left(\frac{T}{100}\right)^3 - 0.45754 \left(\frac{T}{100}\right)^4$$

The volume functions obtained thereupon by the base isotherm method for the equation for viscosity of liquid and gaseous $p-H_2$ in the form of Eq. (5) appear as

$$\begin{split} \alpha &= 29.954 \cdot 10 \; \rho - 436.586 \cdot 10^2 \; \rho^2 - 1307.05 \cdot 10^3 \rho^3 + 307.50 \cdot 10^4 \rho^4 \; + \; 4342.5 \cdot 10^5 \; \rho^5 - 4524.4 \cdot 10^6 \; \rho^6 \\ \beta &= -224.010 \cdot 10 \; \rho - \; 5842.6 \cdot 10^2 \; \rho^2 \; + \; 26136.0 \cdot 10^3 \; \rho^3 \; + \; 2225.0 \cdot 10^4 \; \rho^4 \; - \; 120460 \cdot 10^5 \; \rho^5 \; + \; 126085.6 \cdot 10^6 \; \rho^6 \\ \gamma &= 1 \; - \; 0.26273 \cdot 10 \; \rho - \; 5.33450 \cdot 10^2 \; \rho^2 \; + \; 1.07 \cdot 10^3 \; \rho^3 \; + \; 75.340 \cdot 10^4 \; \rho^4 \; - \; 191.49 \cdot 10^5 \; \rho^5 \; + \; 139.85 \cdot 10^6 \; \rho^6 \end{split}$$

In this equation the dimensions are

$$\eta \ [10^{-6} \frac{\text{N} \cdot \text{sec}}{\text{m}^2}], \quad \phi \ [\frac{g}{\text{cm}^3}], \quad \textit{T} \ [^{\circ}\text{K}]$$

A detailed comparison of the equation obtained with Diller's experimental data, as corrected in [18], showed a mean square departure of calculated from experimental values of 0.8%, with a maximum difference of 3% occurring at only a few points.

These calculations permit the conclusion that the form of equation described herein is a useful one for the analytic description of experimental data on viscosity of gas and liquid within the limits of experimental error.

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