International Journal of Thermophysics, Vol. 17, No. 3, 1996

Prediction of the Viscosity for Molecular Fluids from the Dilute-Gas Properties via the Inversion Procedure for Spherically Symmetric Pair Potentials[1]

V. F. Nebelenchuk[2] and V. A. Mazur[2,3]

A prediction scheme is presented for the viscosity and translation part of the thermal conductivity ("frozen" thermal conductivity) of molecular fluids via the application of the original inversion algorithm for spherically symmetric pair potentials. The latter allows us to invert the centrally symmetrical effective interaction potential for two types of dilute-gas properties—the pressure second virial coefficient and the low-density gas viscosity. Such a potential is then used for the computation of fluid transport properties. Several weakly anisotropic molecular systems were considered (N_2, O_2, CO_2, CH_4). Analysis of the results obtained reveals that the class of spherically symmetric potentials may still be used for the prediction of fluid transport coefficients from the dilute-gas equilibrium and kinetic characteristics.

KEY WORDS: dense fluid; interaction potential; inversion algorithm; viscosity.

1. INTRODUCTION

The problem dealing with the transport coefficients of molecular gases has a special concern for the dense fluid state. A number of experimental data, obtained by diverse methods, are known for molcular fluids. However, essential discrepancies observed for various sources at high pressures cause difficulties in describing the empirical results by reliable analytical correlations. Lack of rigorous kinetic theory which would allow calculation of the fluid transport properties via the Intermolecular Potential (IP) makes this situation worse.

---

[1] Paper presented at the Twelfth Symposium on Thermophysical Properties, June 19–24, 1994, Boulder, Colorado, U.S.A.

[2] The Odessa State Academy of Refrigeration, 1/3 Peter Velikiy Str., 270100 Odessa, Ukraine.

[3] To whom correspondence should be addressed.

In the present paper we propose a prediction scheme for the fluid viscosity and "frozen" thermal conductivity by using the original inversion algorithm [1]. The latter permits the generation of the sphericalized IP for not strongly anisotropic molecules from two types of dilute-gas properties: the pressure second virial coefficient (SVC) $B(T)$ and the low-density gas viscosity $\eta_0(T)$ (Section 2). These IP are then applied, with the local similarity (LS) method [2], for calculation of the fluid viscosities and "frozen" thermal conductivities (Section 3). A rapidly programmable analytical representation for the results obtained is given in Section 4.

2. THE INVERSION ALGORITHM FOR SPHERICALIZED POTENTIALS

A majority of inversion algorithms supposes that some extension operation $\gamma(\mathbf{r})$ should be applied to an initial approximation of the pair potential $U^{[0]}(\mathbf{r}^{[0]})$ in such a way as to modify the spatial measure $r^2\,dr$ or $b\,db$ (b is an impact parameter) proportionally to the ratio $\beta_\varepsilon(T)/\beta_0(T)$ or $\Omega_e^{(l,s)}/\Omega_0^{(l,s)}$. The function β is associated with the SVC as $\beta = B + T(dB/dT)$ and $\Omega^{(l,s)}$ is the Chapmann–Enskog collision integral. The index "e" refers to the macroscopic properties β or $\Omega^{(l,s)}$ as obtained from experiment, and "0" to the zeroth approximation on the basis of the IP $U^{[0]}(\mathbf{r}^{[0]})$.

In more detail, we consider the concrete inversion algorithm. The method [3] using the SVC $B(T)$ has been chosen here as the starting point for derivation of the sphericalization procedure. We omit a thorough statement of known methods, although the main notions important for a presentation of our algorithm are presented below.

(a) The extension function $\gamma_i^{[k]}(r_i^{[k-1]})$ is introduced by the algorithm [3] on the kth iteration step as follows:

$$\gamma_i^{[k]}(r_i^{[k-1]}) = [\beta_{ei}(T_i)/\beta_i^{[k-1]}(T_i)]^{1/3} = r_i^{[k]}/r_i^{[k-1]} \tag{1}$$

where β_e and $\beta^{[k-1]}$ are the experimental and calculated values of β based on the $(k-1)$th approximation to IP, $U^{[k-1]}(\mathbf{r}^{[k-1]})$. The lower index i denotes the numbering of points in the numerical potential.

(b) The separation $r_i^{[k-1]}(T_i)$ is defined by a heuristic relation,

$$r_i^{[k-1]}(T_i) = [\beta^{[k-1]}(T_i)/(2/3\pi N)]^{1/3} \tag{2}$$

being analogous to the random-phase approximation when the transport properties are directly inverted [3].

The analysis of known procedures revealed that a convergence to $B_e(T)$, which is really of interest, was always observed only in a narrow vicinity $(\varepsilon_0 \pm \delta\varepsilon)$ around some fixed value of a potential well depth ε_0. The algorithms diverge from $B_e(T)$ beyond this interval, although convergence always occurs for $\beta_e(T)$. Thus, a "single" spherical function $\mathrm{U}(\mathbf{r})$ may be obtained from the information on $B_e(T)$ via the known approaches. However, as a rule, this potential does not yield an adequate description of the dilute-gas transport properties even for weakly anisotropic molecular systems.

To overcome this problem we have introduced a modified extension function,

$$\tilde{\gamma}_i^{[k]}(r_i^{[0]}) = \tilde{r}^{[k]}/r_i^{[0]} \tag{3}$$

where $\tilde{r}_i^{[k]}$ is that value of distance r for which

$$\mathrm{U}_i^{[k]}(\tilde{r}_i^{[k]}) = \mathrm{U}_i^{[0]}(r_i^{[0]}) \tag{4}$$

The separations $r_i^{[0]}$ are defined at the first iteration $(k = 1)$ for the zeroth approximation according to Eq. (2). Thus, an inversion procedure [3] may be carried out formally for any assumed value of ε (though it may be divergent) and the sequence of functions

$$\tilde{\gamma}^{[k]}(r^{[0]}), \qquad k = 1, 2, \ldots \tag{5}$$

then should be determined.

To make the inversion process convergent, we have suggested transforming the function $\tilde{\gamma}(r)$ by a scalar parameter α as follows:

$$\tilde{\gamma}_\alpha(r^{[0]}) = \tilde{\gamma}^{[l]} + \alpha[\tilde{\gamma}^{[m]} - \tilde{\gamma}^{[l]}] \tag{6}$$

where $\tilde{\gamma}^{[l]}(r^{[0]})$ and $\tilde{\gamma}^{[m]}(r^{[0]})$ are the modified extension functions which should be previously determined for the lth and mth iteration steps. Such transform yields the **r**-measure r_α

$$r_a = \tilde{\gamma}_\alpha(r^{[0]})\, r^{[0]} \tag{7}$$

and generates a potential

$$\mathrm{U}_\alpha[\tilde{\gamma}_\alpha(r^{[0]})\, r^{[0]}] = \mathrm{U}^{[0]}(r^{[0]}) \tag{8}$$

This yields a function $\beta_\alpha(T)$ which is always very close to the experimental $\beta_e(T)$ for any α, while the SVC $B_\alpha(T)$ strongly depends on α.

One can "turn back" the initially divergent inversion process by taking the negative value of α. The procedure then becomes convergent. By varying the value of α it is possible to find the approximation $U_\alpha(r_\alpha; \varepsilon)$ which will describe an experimental dependence $B_e(T)$ for any previously chosen magnitude of ε. It becomes clear from the above consideration that the entire multitude of spherical potentials $U(r, \varepsilon)$ is obtained for different values of ε, and all of them will reproduce an experimental curve $B_e(T)$. However, only that function $U(r, \varepsilon_s)$ may be chosen as the sphericalized solution, if it yields an adequate description of transport properties such as the dilute-gas viscosity $\eta_0(T)$.

These considerations were used for deriving a computational algorithm. This algorithm was applied to weakly anisotropic systems such as N_2, O_2, CO_2, and CH_4. The obtained sphericalized potentials were tested first on the dilute-gas data—the SVC $B(T)$ and dilute-gas viscosity $\eta_0(T)$. The comparison of our calculated values $B(T)$ and $\eta_0(T)$ with the experiment [4–9] has shown that the proposed inversion procedure allows us to overcome a problem of inconsistency between the different types of properties (equilibrium and transport) even by using the angular-independent effective potentials. Figure 1 illustrates such a comparison as an example for methane. The procedure is reliable, however, when slightly anisotropic molecules are considered.

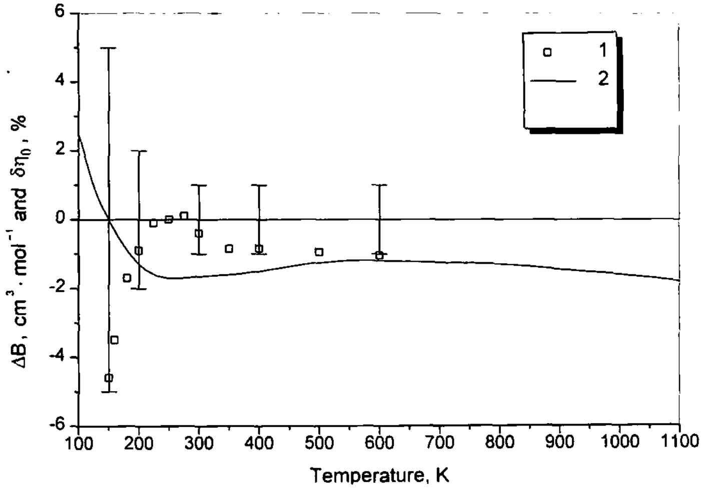

Fig. 1. (1) Absolute deviations $\Delta B = B_{\text{calc}} - B_{\text{expl}}$, relative to the data [4]; (2) percentage deviations $\delta\eta_0 = (\eta_0^{\text{calc}} - \eta_0^{\text{exp}})/\eta_0^{\text{exp}}$ compared to the correlation [8] for methane. The bars denote the experimental errors for the values of SVC $B(T)$ [4].

3. THE BASIC RELATIONS FOR TRANSPORT PROPERTIES

The main conceptions for computation of transport properties in a fluid were presented in Ref. 2, starting from the local similarity hypothesis and the approximation of the "diffusing phonon." The fluid properties are not computed directly via the real IP function U(**r**). The potential U$_b$(**r**) for the reference molecular system is involved in the algorithm too. The kinetic cofficients of real system are calculated through the relations of the Enskog theory modified for the Van der Waals fluid with the IP of Kac et al. [2]. Such a potential consists of a hard-sphere core with a temperature-dependent diameter $d(T)$ and an attractive tail. The latter was chosen in Lennard–Jones (l:6) form. The soft-sphere viscosity η^s and "frozen" thermal conductivity λ^s are defined at the temperature T and density ρ as

$$\eta^s(T,\rho)=\eta_0^s\frac{\beta_P^s\rho}{Y_P}\left[1+\frac{4}{5}Y_P+\frac{4}{25}Y_P^2+\frac{48}{25\pi}Y_P^2\right] \tag{9}$$

$$\lambda^s(T,\rho)=\lambda_0^s\frac{\beta_P^s\rho}{T_P}\left[1+\frac{6}{5}Y_P+\frac{9}{25}Y_s^2+\frac{32}{25\pi}Y_P^2\right] \tag{10}$$

where $\eta_0^s(T)$ and $\lambda_0^s(T)$ are the dilute-gas properties for the hard-sphere system. The value of Y_P is the modified Enskog modulus, which is determined from the cubic equation

$$Y_P=Z^s+\zeta(T,\rho)\,T\left(\frac{\partial Z^s}{\partial T}\right)_\rho-1 \tag{11}$$

According to the "diffusing phonon" approximation, the factor

$$\zeta(T,\rho)=\left[\frac{\beta_s^s\rho(1+\gamma_s^s\rho/\beta_s^s)}{Y_P}\right]^2 \tag{12}$$

simulates the density dependence for the hard-sphere diameter. The numbers β_P^s, β_P^s and γ_P^s, γ_P^s are associated with the virial coefficients for the soft-sphere gas:

$$\beta_P^s(T,\rho)=\tfrac{2}{3}\pi Nd^3(1+3T\zeta d_T'/d_T) \tag{13}$$

$$\gamma_P^s(T,\rho)=0.625(\tfrac{2}{3}\pi Nd^3)^2(1+6T\zeta d_T'/d_\mathrm{T}) \tag{14}$$

where d_T' is the temperature derivative of diameter $d(T)$.

The soft-sphere values of β_s^s, γ_s^s, and Y_s are calculated from Eqs. (11)–(14) with the assumption that $\zeta=1$. The magnitude of ζ tends to unity at zero density and $\zeta\to 0$ as the density increases. The compressibility

Z^s and thermal pressure term $T(\partial Z^s/\partial T)_\rho$ are calculated by the equation of Carnahan–Starling,

$$Z^s = \frac{1+\xi+\xi^2-\xi^3}{(1-\xi)^3} \tag{15}$$

$$T\left(\frac{\partial Z^s}{\partial T}\right)_\rho = 3T\frac{d'_T}{d}\frac{4+4\xi-2\xi^2}{(1-\xi)^4} \tag{16}$$

where $\xi = \frac{1}{6}\pi N d^3$ and N is the particle density.

The real-fluid properties are calculated through the expressions

$$\begin{aligned} \eta(T,\rho) &= \eta_0 + \eta_1(T)\rho + \Delta\eta^s(T,\rho) \\ \lambda(T,\rho) &= \lambda_0 + \lambda_1(T)\rho + \Delta\lambda^s(T,\rho) \end{aligned} \tag{17}$$

where the excess soft-sphere characteristics are

$$\begin{aligned} \Delta\eta^s &= \eta^s(T,\rho) - \eta_0^s(T) - \eta_1^s(T,\rho)\rho \\ \Delta\lambda^s &= \lambda^s(T,\rho) - \lambda_0^s(T) - \lambda_1^s(T,\rho)\rho \end{aligned} \tag{18}$$

The soft-sphere first virial coefficients are

$$\begin{aligned} \eta_1^s(T,\rho) &= (0.8\beta_{\mathrm{P}}^s - \gamma_{\mathrm{P}}^s/\beta_{\mathrm{P}}^s)\,\eta_0^2 \\ \lambda_1^s(T,\rho) &= (1.2\beta_{\mathrm{P}}^s - \gamma_{\mathrm{P}}^s/\beta_{\mathrm{P}}^s)\,\lambda_0^s \end{aligned} \tag{19}$$

It is desirable to use empirical correlations for evaluation of real first density corrections $\eta_1(T)$ and $\lambda_1(T)$, because of the lack of rigorous statistical methods for these purposes. The approximate computations of virial coefficients $\eta_1(T)$ and $\lambda_1(T)$ were carried out for some substances in Refs. 13–15. Those computations were made only for the Lennard–Jones (12:6) model of IP and the data are not available for other potentials. The real dilute-gas properties $\eta_0(T)$ and $\lambda_0(T)$ are calculated directly via the real potential $\mathrm{U}(\mathbf{r})$. The hard-sphere diameter $d(T)$ was computed via the local similarity criteria [2] by applying the obtained inverted IP $\mathrm{U}(\mathbf{r})$ as the real system potentials.

4. THE COMPUTATION OF HARD-SPHERE DIAMETER AND ANALYTICAL REPRESENTATION OF RESULTS

The temperature derivative $d'_T(T)$ is computed from the differential equation

$$q\beta_b(T_b^*;\, l)/\beta_b(T_b^*;\, l) = q\beta^*(T^*) - \gamma^*(T^*)/\beta^*(T^*) \tag{20}$$

where

$$\beta_{\mathrm{b}}(T_{\mathrm{b}}^{*};l)=(d^{*3}+3d^{*2}T^{*}d_{T}')\,B_{\mathrm{b}}(T_{\mathrm{b}}^{*};l)+d^{*3}(1-T_{\mathrm{b}}^{*}\varepsilon_{T}'^{*})\,T_{\mathrm{b}}^{*}\frac{dB_{\mathrm{b}}^{*}}{dT_{\mathrm{b}}^{*}} \tag{20a}$$

$$\gamma_{\mathrm{b}}(T_{\mathrm{b}}^{*};l)=(d^{*6}+6d^{*5}T^{*}d_{T}'^{*})\,C_{\mathrm{b}}(T_{\mathrm{b}}^{*};l)+d^{*6}(1-T_{\mathrm{b}}^{*}\varepsilon_{T}'^{*})\,T_{\mathrm{b}}^{*}\frac{dC_{\mathrm{b}}^{*}}{dT_{\mathrm{b}}^{*}} \tag{20b}$$

$$\beta^{*}(T^{*})=B^{*}(T^{*})+T^{*}\frac{dB^{*}}{dT^{*}},\qquad \gamma^{*}(T^{*})=C^{*}+T^{*}\frac{dC^{*}}{dT^{*}} \tag{20c}$$

The reduced temperatures are defined here as $T^{*}=k_{\mathrm{B}}T/\varepsilon$ and $T_{\mathrm{b}}^{*}=k_{\mathrm{B}}T/\varepsilon_{\mathrm{b}}=T^{*}/\varepsilon_{\mathrm{b}}^{*}$ (k_{B} is the Boltzmann constant). The values of ε and ε_{b} are the well depths for the real and reference potentials, correspondingly; $\varepsilon_{\mathrm{b}}^{*}=\varepsilon_{\mathrm{b}}/\varepsilon$ and $d^{*}=d/\sigma$ are the reduced well depth and diameter—the parameters for the reference Kac's potential; σ is the "zero" for the real IP $[U(\sigma)=0]$; the number q is equal to 0.8 when the viscosity is calculated; and $q=1.2$ for the thermal conductivity. The reduced derivatives $d_{T}'^{*}$ [to be found from Eq. (20)] and $\varepsilon_{T}'^{*}$ are meant as the derivatives of $d^{*}(T^{*})$ and $\varepsilon_{\mathrm{b}}^{*}(T^{*})$ by T^{*}. The functions $B^{*}(T^{*})$ and $C^{*}(T^{*})$ are the reduced second and third virial coefficients for the real system and $B_{\mathrm{b}}^{*}(T^{*};l)$ and $C_{\mathrm{b}}^{*}(T^{*};l)$ are the corresponding characteristics for the reference IP.

The value of $\varepsilon_{\mathrm{b}}^{*}$ is determined using the second LS criterion

$$\Omega^{(2,2)*}(T^{*})=d^{*2}\Omega_{\mathrm{b}}^{(2,2)*}(T^{*}/\varepsilon_{\mathrm{b}}^{*}) \tag{21}$$

The derivative $\varepsilon_{T}^{*\prime}$ is obtained from the relation

$$\begin{aligned}\Omega_{1}^{(2,2)*}(T^{*})={}&2d^{*}(T^{*})\,d_{T}^{*\prime}(T^{*})\,T^{*}\Omega_{\mathrm{b}}^{(2,2)*}(T_{\mathrm{b}}^{*};l)\\&+d^{*2}\Omega_{\mathrm{b}}^{(2,2)*}(T_{\mathrm{b}}^{*},l)(1-T^{*}\varepsilon_{T}^{*\prime}(T^{*})/\varepsilon_{\mathrm{b}}^{*}(T^{*}))\end{aligned} \tag{22}$$

for further substitution to Eqs. (20a)–(20c). The magnitude $\Omega_{1}^{(2,2)*}(T^{*})$ is calculated through the transport collision integrals $\Omega^{(l,s)*}$,

$$\Omega_{1}^{(2,2)*}(T^{*})=T^{*}\frac{d\Omega^{(2,2)*}}{dT^{*}}=4(\Omega^{(2,3)*}-\Omega^{(2,2)*}) \tag{23}$$

Equation (22) can be deduced from Eq. (21) by directly differentiating the latter. The initial values of $\varepsilon_{\mathrm{b}0}^{*}$ and d_{0}^{*} are found from Eqs. (20) and (21) at $T^{*}\sim 0.9$ by assuming that the derivatives $\varepsilon_{T}'^{*}$ and $d_{T}'^{*}$ are rather small in magnitude at low temperatures, i.e., these numbers are simply set equal to zero at the initial stage. The value $d_{T}'^{*}$ is calculated at any temperature T^{*} from Eq. (20). This equation is quadratic relative to $d_{T}'^{*}$ and the smallest modulo negative value $d_T'^*$ should be chosen. The numerical procedure for the solution of differential equations is then used to obtain the diameter value at the next temperature $T^* + \Delta T^*$.

The parameter l for the Lennard-Jones $(l{:}6)$ tail of the reference potential is obtained from the requirement for the van der Waals constants:

$$C_l\left(\frac{1}{l-3}-\frac{1}{3}\right)=\frac{2}{3}\int_{0.8}^{1.3} T^*[1-B^*(T^*)]\,dT^* \tag{24}$$

were

$$C_l = i^{(i-1)/i}/(i-1), \qquad i = l/6$$

This is the third LS criterion allowing for the given real IP to match the function $U_{\mathrm{b}}(\mathbf{r})$ having a conformal attracting tail. The data for the functions $\Omega_{\mathrm{b}}^{(2,2)*}(T_{\ddot{\mathrm{b}}53}{}^*, l)$, $\Omega_{\mathrm{b}}^{(2,3)*}(T_{\mathrm{b}}^*, l)$, $B_{\mathrm{b}}^*(T_{\mathrm{b}}^*, l)$, and $C_{\mathrm{b}}^*(T_{\mathrm{b}}^*, l)$ are presented in Ref. 2 for some numbers l.

The calculated values of the reduced diameter d^* were approximated by simple expressions:

$$d^*(T^*) = d_0^*(T_0^*) - 0.02 \sum_{i=1}^{n} [\delta_i \tau^{i+2}/(i+2)] \tag{25}$$

where $\tau = \sqrt{(T^* - T_0^*)}$. If $T^* \leqslant T_0^*$, then $d^* = d_0^*$.

The reduced collision integral was represented for this purpose via the expression [10]

$$\Omega^{(2,2)*}(T^*) = \frac{w_0}{T^{*w_1}} + w_2 \exp(-w_3 T^*) + w_4 \exp(-w_5 T^*) \tag{26}$$

The reduced first density correction η_1^* was evaluated according to [11]

$$\begin{aligned} 6\eta_1^* &= 1.504 - 6.49\ 10^{-2} T^* - 0.792/T^* + 0.155/T^{*2} \\ \eta_1 &= 9.118\ 10^{-9} \sigma \sqrt{\frac{\varepsilon}{m}}\, \eta_1^* \end{aligned} \tag{27}$$

The coefficients for Eqs. (25)–(27) are given in the Appendix. The proposed results on the fluid viscosities were tested over a wide density range. Comparison with experiment was made up to the upper limits on the pressure, which were different for various substances—from 0.1 GPa for O_2 to 1 GPa for CH_4. Figure 2 demonstrates such a comparison as an example for methane.

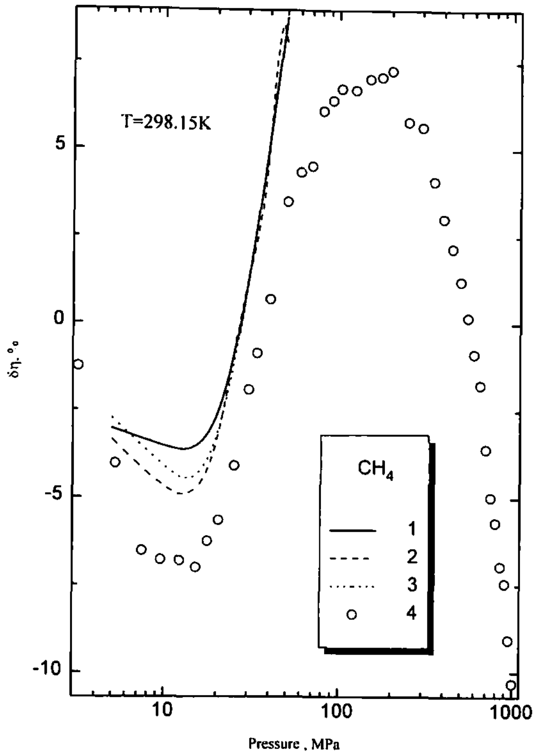

Fig. 2. Percentage deviations $\delta\eta = (\eta^{\mathrm{calc}} - \eta^{\mathrm{calc}})/\eta^{\mathrm{exp}}$ of the calculated viscosities for methane from the data of Ref. 16 (curve 1), Ref. 8 (curve 2), Ref. 17 (curve 3), and Ref. 9 (circles; 4) at the isotherm 298.15 K.

5. CONCLUSIONS

The density dependence for the rotational and vibration internal molecular contributions in the case of thermal conductivity is not given by this method. However, the results obtained for the viscosities allow hope that the proposed prediction scheme will be reliable for weakly anisotropic molecular systems. One should add here that the LS approach was also tested earlier on the viscosities and thermal conductivities for the noble gases up to 1 GPa by using the accurate atom–atom potentials [2].

The correlations obtained for the hard-sphere diameters, Eq. (25), together with Eqs. (26) and (27) are rather simple and may be widely used for generating databases on the transport properties of fluids.

APPENDIX

Table AI. The Coefficients of Eqs. (25), (26), and (27)

	CH_4	CO_2	O_2	N_2
d_0^*	1.05	0.975	1.007	1.0175
T_0^*	1.0375	1.264	1.170	0.980
ε/k_B (K)	221.7424	474.3418	175.2834	141.5
σ (Å)	3.510817	3.521208	3.183727	3.47
m (c.u.)	16.043	44.011	31.9988	28.0134
δ_1	−2.371047e+00	1.873587e−02	9.550195e−05	1.794800e−04
δ_2	3.033080e+01	1.129905e+01	2.584558e+00	9.520699e+00
δ_3	−5.199015e+01	−6.731780e+00	3.986658e+01	2.807697e+01
δ_4	3.693447e+01	−1.206349e+01	−1.365031e+02	−1.171160e+02
δ_5	−1.218862e+01	1.258755e+01	1.874029e+02	1.516621e+02
δ_6	1.548757e+00	−3.182510e+00	−1.372774e+02	−9.956796e+01
δ_7			5.660626e+01	3.605847e+01
δ_8			−1.242260e+01	−6.881681e+00
δ_9			1.130813e+00	5.419823e−01
w_1	3.5583321e−01	7.7848565e−01	4.8284179e−01	5.7619157e−01
w_2	1.0267673e+00	1.5876374e−01	8.4746481e−01	6.2031632e−01
w_3	9.7171392e−01	3.2419474e−01	9.1993584e−01	7.8294878e−01
w_4	1.0783942e−02	1.1803972e−03	1.0678673e−02	7.7561288e−03
w_5	6.7097677e−01	8.3913522e−01	6.2991328e−01	8.1449310e−01
w_6	1.1097162e+00	1.0484874e+00	1.5414915e+00	1.6553697e+00

REFERENCES

1. V. F. Nebelenchuk, *Prediction of Transport Properties of Fluids via Spherical Model of Interaction Potentials*, D.Ph. thesis (Odessa), 1987.
2. V. F. Nebelenchuk and V. A. Mazur, *Physica* **A178**:123 (1991).
3. G. C. Maitland, V. Vesovic, and W. A. Wakeham, *Mol. Phys.* **54**:287 (1985).
4. J. H. Dymond and E. B. Smith, *The Virial Coefficients of Pure Gases and Mixtures* (Oxford University Press, London, 1980).
5. K. Stephan, R. Krauss, and A. Laesecke, *J. Phys. Chem. Ref. Data* **16**:993 (1987).
6. G. P. Mathews, C. M. S. R. Thomas, A. N. Duffy, and E. B. Smith, *J. Chem. Soc. Faraday Trans. I* **72**:238 (1976).
7. V. Vesovic, W. A. Wakeham, G. A. Olchowy, J. V. Sengers, J. T. R. Watson, and J. Millat, *J. Phys. Chem. Ref. Data* **19**:763 (1990).
8. D. G. Friend, J. F. Ely, and H. Ingham, *J. Phys. Chem. Ref. Data* **18**:583 (1989).
9. P. S. Van der Gulik, R. Mostert, and H. R. Van den Berg, *Physica* **A151**:153 (1988).
10. P. D. Neufeld, A. R. Janken, and R. A. Aziz, *J. Chem. Phys.* **57**:1100 (1972).
11. J. Kestin, S. Ro, and W. Wakeham, *Physica* **58**:165 (1972).

12. M. Kac, G. E. Uhlenbeck, and P. C. Hemmer, *J. Math. Phys.* **4**:216 (1963).
13. D. G. Friend and J. C. Rainwater, *Chem. Phys. Lett.* **107**:590 (1984).
14. J. C. Rainwater and D. G. Friend, *Phys. Rev.* **A36**:4062 (1987).
15. E. Bich and E. Vogel, *Int. J. Thermophys.* **12**:27 (1991).
16. A. D. Kozlov, V. M. Kuznetzov, and U. V. Mamonov, in *Reviews on Thermophysical Properties of Substances* (Acad. Sci. High Temp. Inst., Moscow, 1989), N3 (in Russian).
17. B. A. Younglove and J. F. Ely, *J. Phys. Chem. Ref. Data* **16**:577 (1987).