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A Test of the Modified Enskog Theory for the Transport Properties of Liquids

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The modified Enskog theory (MET) has been applied to various fluids in the liquid range (between the triple point and the critical point), and the viscosity, thermal conductivity, and self-diffusion coefficients have been calculated. The temperature dependence of the covolume has been introduced explicitly, bypassing the use of virial coefficients. The agreement is generally acceptable and sometimes good. There is an evident regularity in the results when the reduced temperature is introduced as an independent variable.

KEY WORDS: Enskog theory; self-diffusion; thermal conductivity; viscosity.

1. INTRODUCTION

Research on the transport properties is clearly simplified when a connection can be made with equilibrium properties. In fact, this procedure can sometimes avoid the explicit consideration of a formal theory of transport phenomena. In this context, the Enskog theory [1, 2] holds a privileged position for very good reasons [3].

Although the Enskog theory is a real theory for a fluid of hard spheres, Enskog himself proposed and formulated its application to dense fluids. This transformation, which is known as the modified Enskog theory (MET), presents two modifications with respect to the original Enskog theory. First, the actual pressure P is replaced by the thermal pressure $T(\partial P/\partial T)_V$ in order to take into account the attractive forces among molecules. It can easily be seen that the two quantities coincide for an ideal

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gas. Second, the covolume b is redefined in terms of the second virial coefficient B:

$$b = B + T(dB/dT) \tag{1}$$

The latter step seriously hinders the application of the theory to real systems, because of a lack of knowledge of the virial coefficients for many liquids [4–6]. However, this difficulty can be overcome since the covolume or, equivalently, the hard-sphere diameter and hard-core volume are quantities whose variations with temperature and pressure are well established by perturbation theories for liquids [7].

Since the Enskog formalism is an "ad hoc theory," this consideration can be interpreted as an additional operation which completes the procedure. Therefore, the problem is reduced to the specification of the thermal pressure, which, in turn, involves determining the isobaric expansivity coefficient $(1/V)(\partial V/\partial T)_P$ and the isothermal compressibility coefficient $-(1/V)(\partial V/\partial P)_T$. These properties have been studied for the n-alkanes by Flory et al. [8, 9], and general correlations for a great variety of liquids have been obtained by Brostow and his colleagues [10, 11].

Some years ago, we analyzed the behavior of the internal pressure $(\partial U/\partial V)_T = T(\partial P/\partial T)_V - P$ of liquids [12]. This analysis was extended to several types of liquids. In the present work, we follow the classification developed and the values calculated in this previous paper. The MET has usually been tested [13–15] in the supercritical range and for only a few substances, although there are some recent predictions [16] for the subcritical domain. In the past few years the MET has been applied extensively to mixtures [17, 18]. These papers also contain a considerable number of references to pure fluids. Our test of the validity of the MET includes 15 substances pertaining to atomic liquids, simple molecular liquids, and n-alkane liquids. Each one has been analyzed in the maximum allowable temperature range.

The present paper is organized as follows. Section 2 presents the necessary theoretical background with the expressions for the viscosity, thermal conductivity, and self-diffusion coefficients in the framework of the MET; the results are presented and discussed in Section 3.

2. THEORY

The Enskog theory relates the values of the transport properties to those of the dilute gas [1, 2]. By inserting the pertinent expressions for the viscosity, thermal conductivity, and self-diffusion coefficients for the dilute gas and replacing the pressure by the thermal pressure, one obtains the following expressions for the viscosity coefficient η and self-diffusion coefficient D:

$$\eta_{\mathrm{E}}=\frac{5\sigma}{24V}(\pi MRT)^{1/2}\left\{\frac{1}{(V/R)(\partial P/\partial T)_V-1}+0.8+0.761[(V/R)\right.$$
$$\left.\times(\partial P/\partial T)_V-1]\right\} \tag{2}$$

$$D_{\mathrm{E}}=\frac{\sigma}{4}\left(\frac{\pi RT}{M}\right)^{1/2}\left[\frac{V}{R}\left(\frac{\partial P}{\partial T}\right)_V-1\right]^{-1} \tag{3}$$

Here σ is the hard-sphere diameter, M is the molecular mass, R is the universal gas constant, and P, V, and T are pressure, volume, and temperature, respectively.

The thermal-conductivity coefficient, on the other hand, consists of two terms: one represents the translational motion of the molecules, and the other the transfer of energy due to changes in the internal energy of the molecules. Its final expression is

$$\lambda_{\mathrm{E}}=\frac{25\sigma}{32V}R\left(\frac{\pi RT}{M}\right)^{1/2}\left\{\frac{1}{(V/R)(\partial P/\partial T)_V-1}+1.2+0.755\right.$$
$$\left.\times\left[\frac{V}{R}\left(\frac{\partial P}{\partial T}\right)_V-1\right]\right\}+1.1\frac{\sigma}{4V}(\pi MRT)^{1/2}\left(C_{\mathrm{P}}-\frac{5}{2}R\right)\left[\frac{V}{R}\left(\frac{\partial P}{\partial T}\right)_V-1\right]^{-1} \tag{4}$$

where C_{P} is the ideal-gas molar specific heat. Obviously, the second term is zero for monoatomic substances.

All the parameters in the above formulas are directly available (the thermal pressure from our earlier work) except the hard-sphere diameter σ. Since this quantity is fundamental to our analysis, we have performed a detailed study of it. The hard-sphere diameter can be derived from perturbational and variational theories of the liquid state or from simple considerations about the packing of hard spheres. In the latter case, σ is related to the random closed-packing volume. There are a number of empirical procedures for determining σ and its value varies slightly when different methods are employed. We have verified that all the predictions are compatible with the errors involved in the transport properties. Therefore, we have applied the most adequate for each case.

3. RESULTS AND DISCUSSION

The analyzed substances have been classified in three groups: (a) atomic liquids—Ne, Ar, Kr, and Xe; (b) simple molecular liquids—N_2, O_2, and CH_4; and (c) *n*-alkanes—$n=6$, 7, 8, 9, 11, 12, 15, and 16. In these chemical species we have selected five experimental values for the three transport properties mentioned above in the greatest available range of temperatures. This selection is certainly sufficient for the purposes of this study.

For groups a and b, practically all the range from the triple point to the critical point is included. Then we have evaluated the corresponding values by means of the MET and the percentage deviation has been defined as

$$\delta X(\%) = 100 \frac{|X_{\mathrm{exp}} - X_{\mathrm{MET}}|}{X_{\mathrm{exp}}} \tag{5}$$

where X_{exp} is the experimental value and X_{MET} is the value calculated by the MET. For Ne, we have obtained the molar volume V_{m} of the liquid at the freezing point from Rabinovich et al. [19] and we have applied the correlation of Easteal and Woolf [20] for the hard-sphere diameter,

$$\sigma(T) = 0.11611 V_{\mathrm{m}}^{1/3} \tag{6}$$

where $\sigma(T)$ is in nm and V_{m} is in $\mathrm{cm}^3 \cdot \mathrm{mol}^{-1}$, together with the experimental measurements of the viscosity [19, 21–23], thermal conductivity [19, 24], and self-diffusion [25–27] coefficients. For Ar, Kr, and Xe the correlations of Easteal and Woolf [20] and Nezbeda and Aim [28] are very suitable for determining σ. Experimental measurements of the viscosity [29–38], thermal conductivity [19, 29, 30, 35, 39–42], and self-diffusion [43–46] coefficients have also been utilized. Since Ar is the substance with the most experimental information available, we have recalculated the three transport properties following the original version of

Table I. Comparison Between Experimental Values and MET Values for the Viscosity, Thermal Conductivity, and Self-Diffusion Coefficients for Atomic Liquids

	Ne	Ar	Kr	Xe
$\delta\eta$	56–25	51–14	50–24	52–7
$\delta\lambda$	7–0	20–11	15–12	14–1
δD	135–34	70–17	31–3	72–2
$\Delta T_{\mathrm{r}\eta}$	0.56–0.81	0.56–0.93	0.56–0.85	0.56–0.87
$\Delta T_{\mathrm{r}\lambda}$	0.56–0.67	0.56–0.93	0.59–0.85	0.60–0.87
ΔT_{rD}	0.58–0.85	0.56–0.93	0.59–0.85	0.56–0.87
$T_{\mathrm{r}\eta}$	0.81	0.93	0.85	0.87
$T_{\mathrm{r}\lambda}$	0.56	0.73	0.59	0.80
T_{rD}	0.85	0.73	0.76	0.77

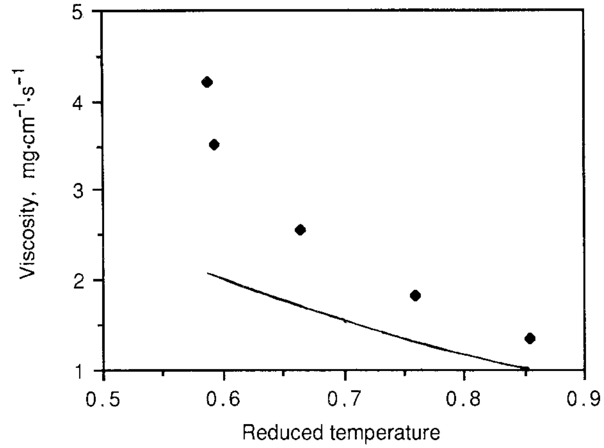

Fig. 1. The viscosity η of krypton as a function of reduced temperature. The points indicate the experimental data and the curve represents the values calculated from Eq. (2).

the MET [13]. In all cases, these calculations showed a worse agreement with the experimental data. This provides us with a validation of our results, which are shown in Table I. In this table $\delta\eta$, $\delta\lambda$, and δD represent the percentage deviation of η, λ, and D, respectively, according to Eq. (5). Maximum and minimum values are shown for each substance. $\Delta T_{r\eta}$, $\Delta T_{t\lambda}$,

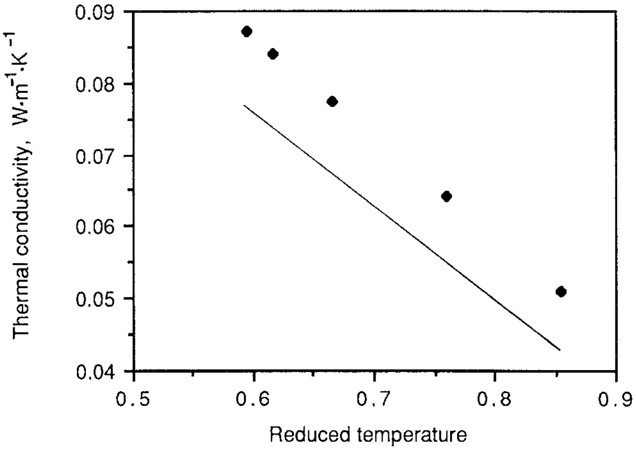

Fig. 2. The thermal conductivity λ of krypton as a function of reduced temperature. The points indicate the experimental data and the curve represents the values calculated from Eq. (4).

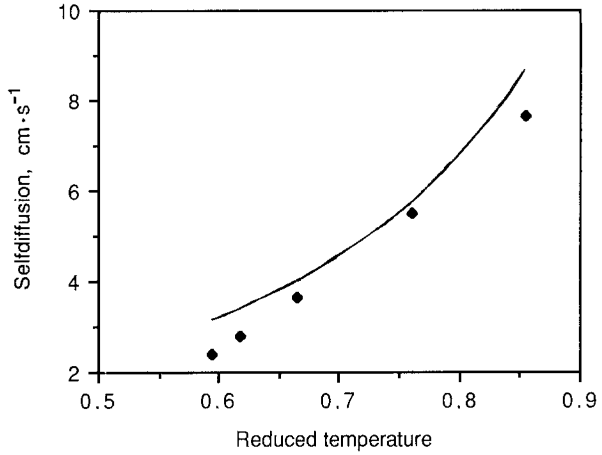

Fig. 3. The self-diffusion D of krypton as a function of reduced temperature. The points indicate the experimental data and the curve represents the values calculated from Eq. (3).

and ΔT_{rD} represent the range in the reduced temperature $T_{\mathrm{r}} = T/T_{\mathrm{c}}$ (T_{c}, critical temperature), which corresponds to the viscosity, thermal conductivity, and self-diffusion coefficients, respectively; $T_{\mathrm{r}\eta}$, $T_{\mathrm{r}\lambda}$, and T_{rD} are the values corresponding to the minimum deviations of η, λ, and D, respectively. Figures 1, 2, and 3 compare the experimental data for the three transport properties of krypton with the predicted behavior.

It can be seen that $\delta\eta$ decreases as T increases. The deviation is the same if the reduced temperatures are equal. This feature clearly indicates a behavior in agreement with the law of corresponding states. There is

Table II. Table I but for Simple Molecular Liquids

	N_2	O_2	CH_4
$\delta\eta$	46-4	46-8	40-1
$\delta\lambda$	20-4	17-6	10-2
δD	43-2	—	43-5
$\Delta T_{\mathrm{r}\eta}$	0.55–0.95	0.39–0.91	0.52–0.94
$\Delta T_{\mathrm{r}\lambda}$	0.55–0.95	0.39–0.91	0.52–0.94
ΔT_{rD}	0.61–0.74	—	0.58–0.94
$T_{\mathrm{r}\eta}$	0.95	0.91	0.73
$T_{\mathrm{r}\lambda}$	0.95	0.39	0.63
T_{rD}	0.74	—	0.66

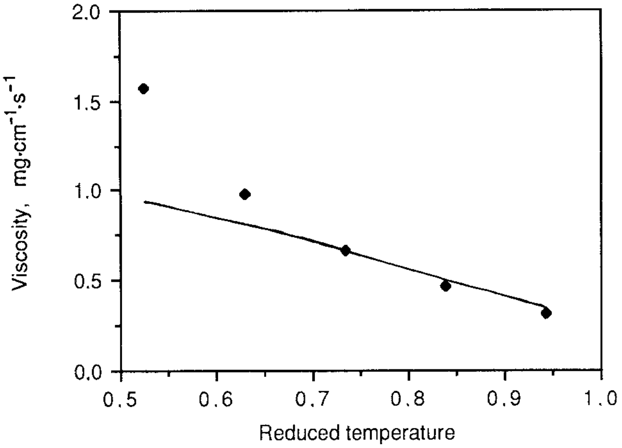

Fig. 4. As Fig. 1 but for methane.

acceptable agreement when T_r exceeds 0.80. The behavior of $\delta\lambda$ is practically uniform and its value is completely suitable in the whole temperature range. In contrast, δD changes abruptly, showing a very low minimum for Ar, Kr, and Xe. Only in the neighborhood of this point is the agreement satisfactory. For Ne, a steady decrease is observed and no minimum appears. However, the few measurements available for this substance are relatively old, which suggests some caution in drawing conclusions.

Consider now the simple molecular liquids in group b. The hard-sphere diameter σ was determined from the correlation of Easteal and

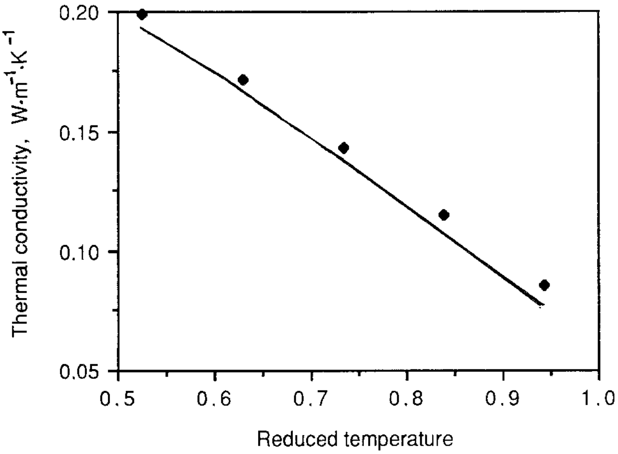

Fig. 5. As Fig. 2 but for methane.

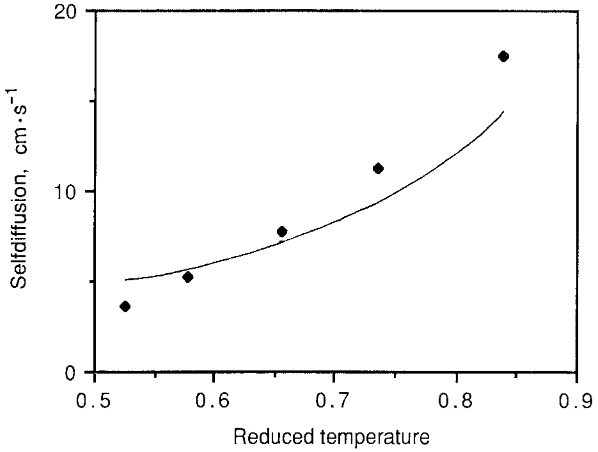

Fig. 6. As Fig. 3 but for methane.

Woolf [20] for N_2, from the correlation of Speedy et al. [47] for O_2, and from the correlation of Nezbeda and Aim [28] for CH_4. The ideal-gas molar specific heat C_P was obtained from Younglove [29] for N_2 and O_2 and from Friend et al. [48] for CH_4. We have employed also experimental data for viscosity [34, 35, 49–54], thermal conductivity [35, 39, 40, 41, 51, 52, 54–59], and self-diffusion [45, 60–64] coefficients. The results are shown in Table II. Figures 4, 5, and 6 compare the experimental data for the three transport properties of methane with the predicted behavior.

The percentage deviation $\delta\eta$ behaves as before for nitrogen and oxygen. For methane there is a minimum, although the rise is very smooth. This behavior is unique among all the substances analyzed. The agreement is acceptable from $T_r = 0.75$. It is found that $\delta\lambda$ also behaves like the simple liquids, although in this case, the expression for λ_E includes the term corresponding to the internal degrees of freedom. In the temperature range considered, its influence amounts to less than 10%.

For N_2, δD decreases with temperature. The agreement is satisfactory when T_r exceeds 0.65. To the best of our knowledge, there is no experimental information available for O_2. The data for CH_4 show a minimum although the subsequent increase is again very smooth. Good agreement is observed for $T_r > 0.58$.

Finally, the normal alkanes in group c are considered. To determine σ we have employed the general correlation for all n-alkanes ($n < 16$), due to Assael et al. [65]. This correlation enables us to determine the close packing volume of spheres V_0, given by $N\sigma^3/2^{1/2}$. Here the correlation for C_P was obtained from Aly and Lee [66]. In addition, we have used the

Table III. As Table I but for *n*-Alkanes

	C_6H_{14}	C_7H_{16}	C_8H_{18}	C_9H_{20}	$C_{11}H_{24}$	$C_{12}H_{26}$	$C_{15}H_{32}$	$C_{16}H_{34}$
$\delta\eta$	27–13	38–8	39–6	47–12	62–24	66–29	68–17	80–33
$\delta\lambda$	32–25	37–23	38–29	43–29	56–32	46–31	46–35	48–35
δD	20–9	26–3	32–5	31–2	—	87–9	—	168–9
$\Delta T_{r\eta}$	0.57–0.65	0.51–0.65	0.52–0.66	0.49–0.66	0.46–0.66	0.45–0.57	0.44–0.58	0.41–0.56
$\Delta T_{r\lambda}$	0.57–0.65	0.51–0.65	0.52–0.66	0.49–0.66	0.46–0.66	0.45–0.60	0.44–0.58	0.43–0.56
ΔT_{rD}	0.57–0.65	0.51–0.65	0.52–0.66	0.50–0.61	—	0.45–0.57	—	0.41–0.56
$T_{r\eta}$	0.65	0.65	0.66	0.66	0.66	0.57	0.58	0.56
$T_{r\lambda}$	0.57	0.51	0.52	0.49	0.46	0.45	0.44	0.43
T_{rD}	0.57	0.58	0.57	0.54	—	0.57	—	0.56

suitable experimental data concerning viscosity [24, 67–78], thermal conductivity [24, 79–99], and self-diffusion [77, 100–110] coefficients.

The results are shown in Table III. Figures 7, 8, and 9 compare the experimental data for the three transport properties of the hexane with the predicted behavior.

It is observed that $\delta\eta$ decreases with T. For $n = 6, 7, 8, 9$ (C_nH_{2n+2}), the value of $\delta\eta$ is encouraging, with the discrepancy being less than 20%. This does not hold true for the other hydrocarbons. However, if we observe the range of reduced temperatures, we see that approximately the same deviation appears for all *n*-alkanes (only the lower value for $C_{11}H_{24}$ is unsuitable) at the same reduced temperature. Thus the behavior is in accordance with the law of corresponding states. This similarity, in spite of

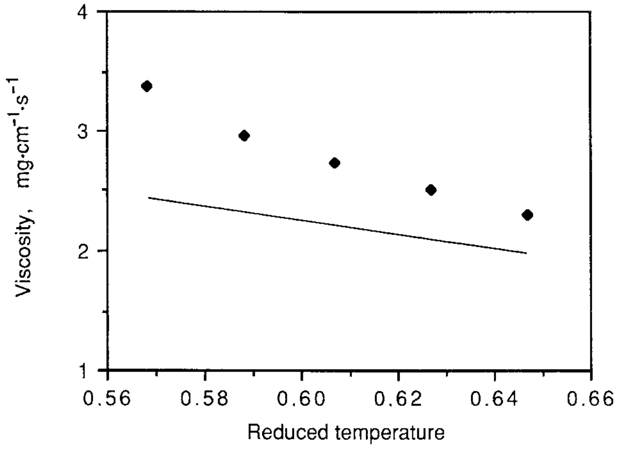

Fig. 7. As Fig. 1 but for hexane.

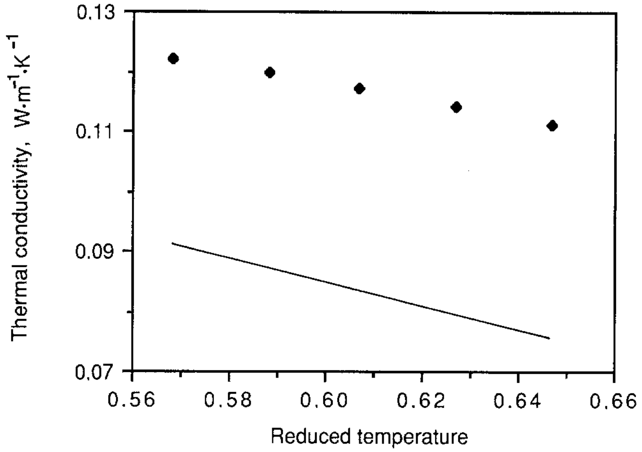

Fig. 8. As Fig. 2 but for hexane.

the different structures, again reveals the fact that the viscosity coefficient does not depend on the internal degrees of freedom.

In contrast to $\delta\eta$, the percentage deviation $\delta\lambda$ increases with T. The variation is smooth just like the other groups but with larger discrepancies (40%, compared to 15% in the other cases). This behavior can be explained by the fact that the thermal-conductivity coefficient depends on the internal degrees of freedom. For very high temperatures, the second term in Eq. (4) may play a more important role and the discrepancy could diminish.

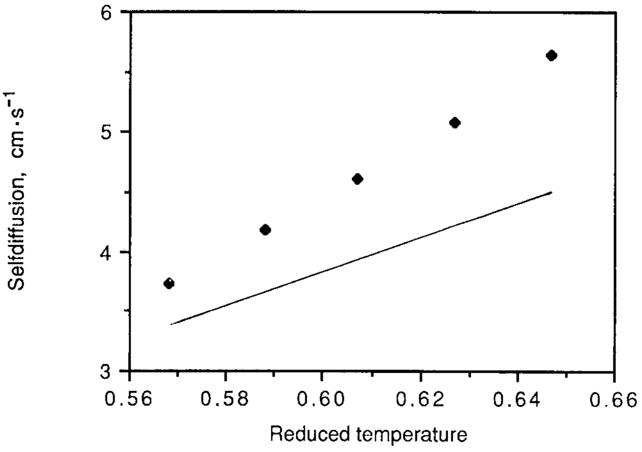

Fig. 9. As Fig. 3 but for hexane.

With respect to δD, the conclusions must be very cautious because the data are scarce. The value of departure for T_r governs the variation, which shows an initial decrease for δD when T_r increases, a minimum for a certain value of T_r, and a later increase. The ΔT_{rD} range for the higher hydrocarbons (dodecane and hexadecane) is completely within the first region. The next hydrocarbons (heptane, octane, and nonane) show the minimum mentioned and hexane shows only the increasing branch. Consequently, the discrepancy with the experimental values depends essentially on the reduced temperature but not on the analyzed substances, just as in the case of the viscosity coefficient, and probably for the same reason. The agreement is satisfactory if the reduced temperature lies between 0.53 and 0.63.

In summary, the three coefficients display common features for all substances. For the viscosity and self-diffusion coefficients, the discrepancies are due exclusively to their distinct reduced temperatures. For the viscosity coefficient, good agreement is observed from a given value of T_r, whereas there is satisfactory agreement only in a range of T_r for the self-diffusion coefficient. For the thermal conductivity coefficient, the variation is smooth everywhere but the agreement is acceptable only for the atomic liquids and the simple molecular liquids.

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