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Theoretically Based Data Assessment for the Correlation of the Thermal Conductivity of Dilute Gases

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This paper presents two schemes for a theoretically based data assessment of the thermal conductivity of dilute polyatomic gases. The first employs the simplified Thijsse expression, combined with accurate experimental data obtained from a transient hot-wire apparatus, as reference. The second makes use of theoretical results for the temperature dependence of the ratio D_{int}/D. Both methods lead to mutually consistent results for linear molecules and to useful criteria for discriminating between experimental data sets. The paper also demonstrates the influence of data burdened with systematic errors upon the final results of different correlation schemes.

KEY WORDS: carbon dioxide; carbon monoxide; nitrogen; polyatomic gas; thermal conductivity of gases; transport properties.

1. INTRODUCTION

The highly developed kinetic theory for dilute monatomic and polyatomic gases [1, 2], on one hand, and increasingly powerful computers, on the other, have formed the basis for extensive work on the correlation and prediction of transport properties, especially in the limit of zero density. Both for "individual" correlations [3–6] and for "universal" correlations [7–11] a critical data assessment must form the third prerequisite for all analyses. Although this is true for the transport properties in general, the thermal conductivity represents a particular case. This is because of the

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sensitivity of this property to the anisotropy of the intermolecular potential and because of the existence of internal degrees of freedom and their interaction with translational degrees of freedom (inelastic collisions).

It rapidly becomes apparent from an analysis of the available thermal conductivity data for fluids that the range of thermodynamic states covered—the temperature range in the limit of zero density—is often very restricted. This fact often necessitates the inclusion of older, sometimes less reliable, data in the basis set if a significant temperature range is to be covered. However, there are often substantial discrepancies between various data sets that have been reported in the literature far exceeding the claimed accuracy. Since it is usually extremely difficult to decide on an objective measure of the accuracy of published data, solely on the basis of information available in the literature, two complementary methods of data assessment have been developed. First, we have employed a comparison with new experimental data of proven accuracy. Here, the simplified Thijsse expression for the thermal conductivity of a dilute polyatomic gas proves of value. Second, some confidence in experimental data can be established by recourse to kinetic theory directly. In this process, the high-temperature behavior of certain effective collision cross sections is deduced from experimental data so that it can be compared with results developed from the kinetic theory for rigid rotors. Since equivalent results are not available for other molecular structures such as spherical tops and chain molecules, we restrict our analysis to linear molecules and employ as examples nitrogen, carbon monoxide, and carbon dioxide.

It is the purpose of this paper to summarize the general results of the assessment procedures and to show the guidance that one can derive from the theory in order to establish reliable primary data sets of thermal conductivity for scientific or engineering purposes.

2. THEORY

2.1. Viehland–Mason–Sandler Formalism

Following a suggestion of Eucken [12] the thermal conductivity of a polyatomic gas in the limit of zero density can always be written as the sum of translational (λ_{tr}) and internal (λ_{int}) contributions

$$\lambda_0 = \lambda_{\mathrm{tr}} + \lambda_{\mathrm{int}} \tag{1}$$

According to the modern kinetic theory developed, in usable form, by Mason, Monchick, and co-workers [13–16], these contributions are [16]

$$\lambda_{\text{tr}} = \frac{5k^2T}{2m\langle v\rangle_0}\left\{\frac{\mathfrak{S}(1001) + r\,\mathfrak{S}\begin{pmatrix}1010\\1001\end{pmatrix}}{\mathfrak{S}(1010)\ \mathfrak{S}(1001) - \mathfrak{S}^2\begin{pmatrix}1010\\1001\end{pmatrix}}\right\} \tag{2}$$

and

$$\lambda_{\text{int}} = \frac{5k^2T}{2m\langle v\rangle_0}\left\{\frac{r\,\mathfrak{S}\begin{pmatrix}1010\\1001\end{pmatrix} + r^2\mathfrak{S}(1010)}{\mathfrak{S}(1010)\ \mathfrak{S}(1001) - \mathfrak{S}^2\begin{pmatrix}1010\\1001\end{pmatrix}}\right\} S \tag{3}$$

The quantities $\mathfrak{S}(^{pqrs}_{p'q'r's'})$ are effective collision cross sections [17] [if $pqrs \equiv p'q'r's'$, then $\mathfrak{S}(^{pqrs}_{p'q'r's'}) = \mathfrak{S}(pqrs)$] which incorporate all of the information about the binary molecular interaction and hence the intermolecular pair potential. The aditional symbols introduced in Eqs. (2) and (3) are

$$\langle v\rangle_0 = 4(kT/\pi m)^{1/2} \tag{4}$$

$$r = \, = (2c_{\text{int}}/5k)^{1/2} \tag{5}$$

and

$$S \approx 1 - (5/3)(1 + \lambda_{\text{tr}}/\lambda_{\text{int}})(\Delta\lambda_{\|}/\lambda)_{\text{sat}} \tag{6}$$

Here, c_{int} is the internal heat capacity and m the molecular mass of the gas, respectively. S is a correction factor to the semiclassical Wang Chang–Uhlenbeck kinetic theory that accounts for spin polarization effects [16].

There exist a number of exact relationships between different cross sections [18]. Here we make use of the results

$$\mathfrak{S}\begin{pmatrix}1010\\1001\end{pmatrix} = (5r/6)\ \mathfrak{S}(0001) \tag{7}$$

and

$$\mathfrak{S}(1010) = (2/3)\ \mathfrak{S}(2000) + (25r^2/18)\ \mathfrak{S}(0001) \tag{8}$$

The cross section $\mathfrak{S}(2000)$ is related to the zero-density viscosity by the equation

$$\eta_0(T) = \frac{kT}{\langle v\rangle_0}\,\mathfrak{S}(2000)^{-1} \tag{9}$$

whereas for the (hypothetical) coefficient of self-diffusion we have

$$D = \frac{kT}{nm\langle v\rangle_0}\,\mathfrak{S}(1000)^{-1} \tag{10}$$

and for the collision number for internal energy relaxation [13, 14]

$$\xi_{\text{int}} = \frac{4kT}{\pi\eta_0\langle v\rangle_0}\,\mathfrak{S}(0001)^{-1} \tag{11}$$

Another quantity that has often been used as a quasi-experimental value is the diffusion coefficient for internal energy (D_{int}), which has been defined as [13, 14]

$$D_{\text{int}} = \frac{kT}{nm\langle v\rangle_0}\,[\mathfrak{S}(1001) - \tfrac{1}{2}\mathfrak{S}(0001)]^{-1} \tag{12}$$

This is, in fact, the only quantity of the formalism that cannot be deduced directly from independent measurements. However, for the purposes of data assessment Eq. (12) can be combined with the further kinetic theory result

$$D_{\text{int}}/D = \frac{5}{6A^*}\,\frac{nmD_{\text{int}}}{\eta^0} \tag{13}$$

where A^* is the usual cross-section ratio [1], very weakly dependent on the intermolecular pair potential. This result can be put to good use because it can be shown [18] that for linear molecules (rigid rotors) at high temperatures

$$\mathfrak{S}(1001) \rightarrow \mathfrak{S}(1000) + (35r^2/12)\,\mathfrak{S}(0001) \tag{14}$$

and therefore that

$$D_{\text{int}}/D \rightarrow \frac{\mathfrak{S}(1000)}{\mathfrak{S}(1000) + [(35r^2/12) - \tfrac{1}{2}]\,\mathfrak{S}(0001)} \tag{15}$$

Since at high temperatures $\mathfrak{S}(0001) \rightarrow 0$, then according to the theory D_{int}/D for rigid rotors should approach unity from below at $T \rightarrow \infty$.

2.2. Thijsse Formalism

By employing an alternative set of expansion functions in the solution of the kinetic theory equations, and starting from the isotropic thermal conductivity, i.e., neglecting the effect of angular momentum on the rotational motion of molecules, Thijsse et al. [19, 20] derived the expression

$$\lambda_0=\frac{5k^2T}{2m\langle v\rangle_0}\frac{(1+r^2)}{\mathfrak{S}(10E)}\left[1-\frac{\mathfrak{S}^2\binom{10E}{10D}}{\mathfrak{S}(10E)\,\mathfrak{S}(10D)}\right]^{-1} \tag{16}$$

where the new effective cross sections $\mathfrak{S}(10E)$, $\mathfrak{S}(10D)$, and $\mathfrak{S}\binom{10E}{10D}$ are related to those introduced earlier via linear transformations. Indeed, by using these relationships it can be shown [11, 19–21] that the correction factor in brackets in Eq. (16) is often very close to unity, so that the simpler formula

$$\lambda_0=\frac{5k^2T}{2m\langle v\rangle_0}\frac{(1+r^2)}{\mathfrak{S}_T(10E)} \tag{17}$$

is a good approximation to Eq. (16). The subscript T attached to $\mathfrak{S}_T(10E)$ distinguishes it from the exact cross section $\mathfrak{S}(10E)$. Thus, the final formula for λ_0—at least for simple molecules such as N_2, CO, and CO_2—is similar to that for a monatomic gas and to that for the viscosity of a gas in the limit of zero density. Consequently, a simple means of representing the thermal conductivity data is afforded by defining a reduced cross section by

$$\mathfrak{S}_T^*(10E)=\mathfrak{S}_T(10E)/\pi\sigma^2 \tag{18}$$

following an analogy with the treatment of monatomic gases [7–9] in which σ is a length-scaling parameter [1, 3–5]. The same analogy suggests that the temperature dependence of the cross section could be represented in the form

$$\ln \mathfrak{S}_T^*(10E)=\sum_{i=0}^{n} a_i(\ln T^*)^i \tag{19}$$

or, more simply, in the form

$$\mathfrak{S}_T^*(10E)=\sum_{i=0}^{n} b_i/T^{*i} \tag{20}$$

Here $T^*=kT/\varepsilon$ is the reduced temperature and ε is an energy-scaling parameter [1, 3–5].

3. EXPERIMENTAL DATA

3.1. Thermal Conductivity

The substantial discrepancies between different reported data sets always necessitate commencing a correlation procedure by dividing the experimental data into primary and secondary data [3–5, 22, 23]. In general *primary data* are the results of measurements made with an apparatus of high precision for which a complete working equation and a detailed knowledge of all corrections is available. Strictly, for the thermal conductivity this means that only measurements made with the transient hot-wire technique are included. However, because this would reduce the temperature range studied unduly, it is usually necessary to include data in the primary data set that have been obtained by other techniques.

The primary data finally selected for N_2 and CO [5] and CO_2 [23] have been given elsewhere, so that because we are interested here in demonstrating schemes for the assessment of experimental data, we include other data sets in our considerations. They are Refs. 24 and 25 for nitrogen, Ref. 26 for carbon monoxide, and Refs. 27–30 for carbon dioxide.

3.2. Further Data Needed in the Kinetic Theory Analysis

In Section 2 it was illustrated that a kinetic theory analysis of thermal conductivity must necessarily include information on further experimental data. The sources of data for the viscosity, heat capacity, collision numbers for internal energy relaxation, and fractional change of the thermal conductivity due to a magnetic field parallel to the temperature gradient are given elsewhere [5, 23, 31] for each of the gases discussed here so they are not repeated.

4. RESULTS AND DISCUSSION

4.1. D_{int}/D

From Eqs. (2), (3), (7), and (8) it becomes immediately obvious that the thermal conductivity in the limit of zero density is related to just three effective collision cross sections, $\mathfrak{S}(2000)$, $\mathfrak{S}(0001)$, and $\mathfrak{S}(1001)$. Furthermore, Eqs. (9) and (11) demonstrate that the two first-mentioned cross sections are directly obtainable from independent measurements. To analyze thermal conductivity data via D_{int}/D and the related theoretical predictions, it is therefore necessary to solve Eqs. (2) and (3) for $\mathfrak{S}(1001)$ using the experimental λ_0 values and additional data mentioned in Section 3. The "experimental" D_{int}/D then follows from Eqs. (12) and (13) and can be compared with theoretical predictions as well as results established on the basis of other data sets for λ_0.

The results of such an analysis are shown in Fig. 1 for carbon dioxide in the form of a plot of D_{int}/D. This fluid seemed to be well studied because there are no fewer than 40 data sets available in the open literature. However, the differences between these data sets were often very large [23]. To illustrate some general trends, Fig. 1 displays only selected experimental results. The solid line in this figure represents the correlated temperature function [23] for D_{int}/D deduced from the final, selected primary data [32–37].

Among those data which have been classified as secondary data, two trends become obvious. First, there are data which confirm the temperature function deduced from primary data but are burdened with a systematic error, so that the related curve is therefore shifted. This effect is revealed by the data of Haarman [30] for carbon dioxide in Fig. 1. The data of Le Neindre et al. [27, 28] above 400 K show similar behavior, whereas the same authors' results between 300 and 400 K completely disagree with theoretical predictions.

The second group of data sets yields a totally different temperature function compared with that of the primary data. This is exemplified in Fig. 1 by the data of Gupta and Saxena [29]. The curve clearly demonstrates the disagreement of these data with the expected behavior of

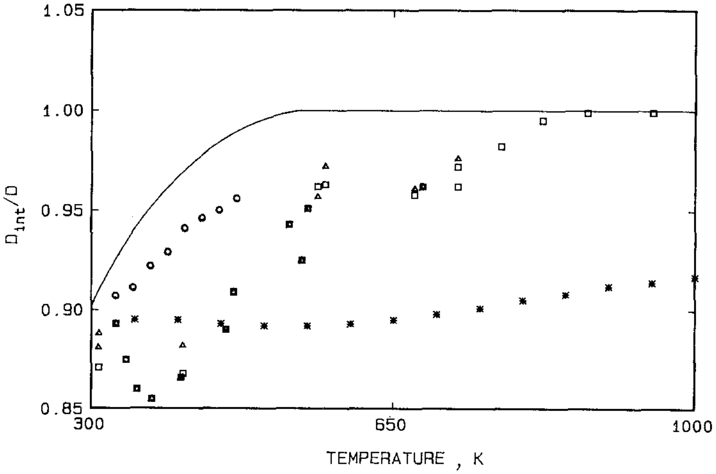

Fig. 1. Values for D_{int}/D for CO_2 calculated using the Viehland–Mason–Sandler formalism. (——) Primary correlation [23]; (○) Ref. 30; (△) Ref. 27; (□) Ref. 28; (∗) Ref. 29.

D_{int}/D over the entire temperature range. The results of the same group of workers for nitrogen confirm the general trends formed for CO_2, although the displacement of Haarman's data [30] and those of Le Neindre and co-workers [27, 28] is noticeably smaller, so that it has been possible to include those in the primary data set. In fact, this finding proved valuable because the available temperature range could be significantly enlarged in this way (cf. Section 4.3).

4.2. $\mathfrak{S}^*_T(10E)$

Equations (17) and (18) enable the reduced effective collision cross section $\mathfrak{S}^*_T(10E)$ to be calculated from experimental thermal conductivity data because the only additional quantity required is the internal heat capacity. In this case, the method of assessment is based upon the observation that, to a good approximation, the temperature dependence of effective cross sections has been found to be linear in $1/T$ or $1/T^*$. Thus,

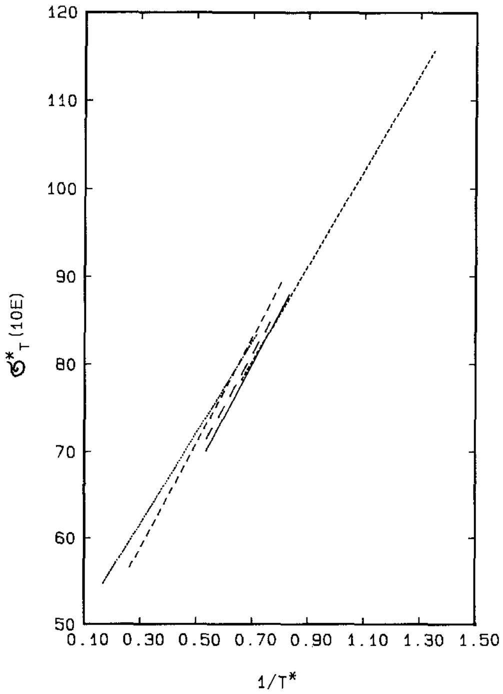

Fig. 2. A comparison of different data sets for CO_2 using the reduced Thijsse cross section $\mathfrak{S}^*_T(10E)$. (——) Transient hot-wire data [32–35]; (---) Ref. 41; (–––) Refs. 27 and 28; (———) Ref. 30; (· · · ·) Ref. 29.

different thermal conductivity data sets are analyzed using Eq. (20) with $n=1$ and the results are compared with the function established from transient hot-wire data, in a graph $\mathfrak{S}_T^*(10E)$ vs $1/T^*$. It will be seen that in such a plot some then give parallel lines, the displacement of the lines being characteristic of systematic errors associated with the data.

On the other hand, inconsistent data sets show a remarkably different temperature dependence of $\mathfrak{S}_T^*(10E)(T^*)$. Figures 2 and 3 contain plots of the kind for carbon dioxide and nitrogen, respectively. The transient hot-wire data have been obtained from Refs. 32–35 for carbon dioxide and from Refs. 33, 35, 38, and 39 for nitrogen. It can be seen that these results generally confirm our conclusions drawn from the behavior of D_{int}/D. First, the data of Haarman [30] are always shifted by an amount depending on the gas with respect to the most accurate values. This finding, together with similar results for the monatomic gases [40], leads to the conclusion that the uncertainty ascribed to these data should be increased to ±1% instead of the figure ±0.3%, as originally assumed by the author.

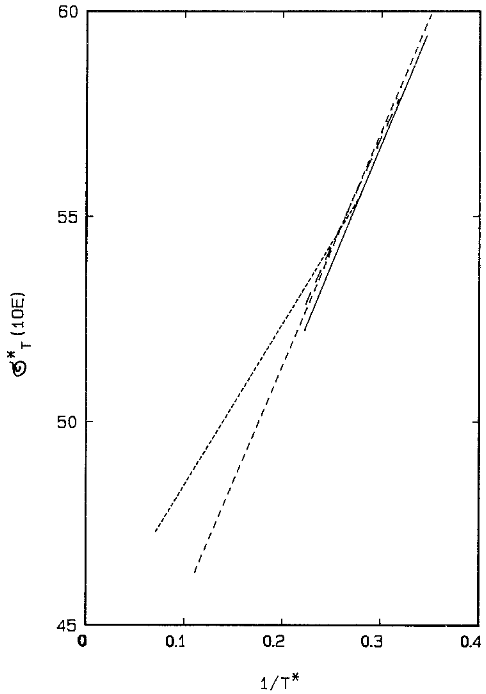

Fig. 3. A comparison of different data sets for N_2 using the reduced Thijsse cross section $\mathfrak{S}_T^*(10E)$. (——) Transient hot-wire data [33, 35, 38, 39]; (— — —) Ref. 30; (- - -) Refs. 27 and 42; (----) Refs. 24 and 25.

Second, the data of Johnston and Grilly [41] for carbon dioxide agree well with the transient hot-wire data and they were therefore included in the primary data set in order to extend its temperature range. The data of Le Neindre et al. for both carbon dioxide [27, 28] and nitrogen [27, 42] seem to be influenced mainly by the values above 400 K and seem therefore only shifted in the plots in Figs. 2 and 3.

On the other hand, the data of Saxena and co-workers for nitrogen [24, 25] as well as for CO_2 [29] and CO [26] show a completely different temperature dependence. For these reasons, these data were not included in primary data sets for the correlation of thermal conductivity because the systematic error associated with them could influence the results of the correlation dramatically.

4.3. The Influence of Systematic Errors in Data on the Correlation

In the previous sections, two procedures for the assessment of gas-phase thermal conductivity data were discussed. If a selection of data on the basis of these or similar analyses is not made, then obviously any final representation must be burdened with some of the systematic errors in the data. Thus, for example, it is always possible to represent any set of low-density thermal conductivity data by one of the forms

$$\lambda_0(T) = \sum_{i=1}^{n} a_i (R/T)^i \tag{21}$$

where R is a suitable reduction parameter, or by

$$\lambda_0(T) = T^{1/2} \Big/ \left[\sum_{i=1}^{n} a_i T^{*-i} \right] \tag{22}$$

which is a form derived by analogy with the result for monatomic gases. In neither case does such a representation have any theoretical foundation so that they cannot be employed in any data assessment. In this section we consider how the neglect of such a data assessment can alter the final correlation of data.

Figure 4 shows the deviations of various correlations of the thermal conductivity of nitrogen from the theoretically based representation of Ref. 5. Curves 1 to 3 are the representations secured by Eqs. (19), (21), and (22), respectively, when the data set employed is that established as primary in Ref. 5. Within the temperature range 300–900 K all three representations are equivalent within the uncertainty of the final correlation. On the other hand, when other experimental information is included in the data set to be fitted, such as that from Refs. 24 and 25, which was

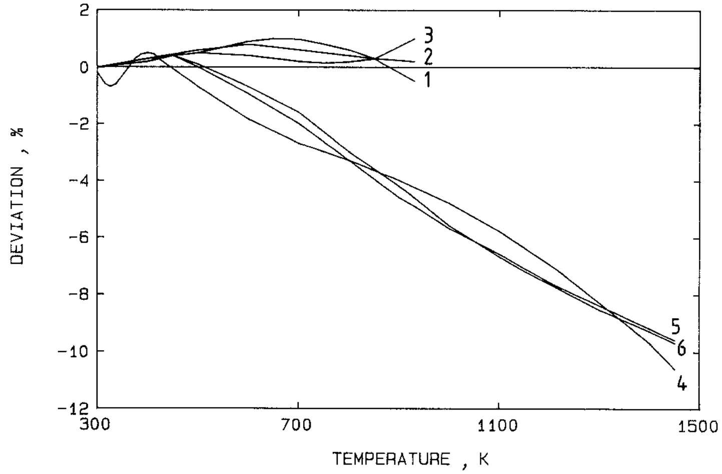

Fig. 4. A comparison of different correlation schemes with the primary correlation for N_2 [5]. Curves 1–3, primary data from Ref. 5; Curves 4–6, primary data + Ref. 24; Curves 1 and 4, Eqs. (17)–(19); Curves 2 and 5, Eq. (22); Curves 3 and 6, Eq. (21).

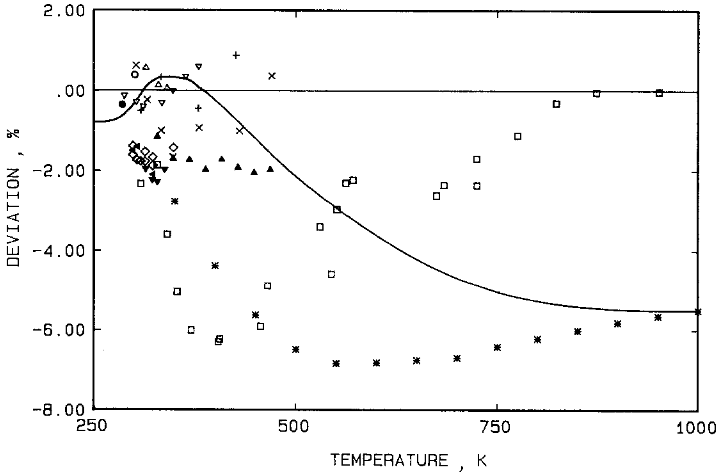

Fig. 5. Deviation plot for primary and selected secondary data for CO_2 from the primary correlation of Ref. 23. (+) Ref. 32; (○) Ref. 33; (×) Refs. 34 and 35; (▽) Ref. 41; (△) Ref. 36; (●) Ref. 37; (▲) Ref. 30; (□) Refs. 27 and 28; (∗) Ref. 29; (◇) Ref. 43; (▼) Ref. 44; (◁) Ref. 45. (——) Polynomial fit using Eq. (22) and the entire data set.

designated secondary in Ref. 5, the effect on the final representation is quite dramatic. Curves 4 to 6 represent the new results obtained with the three correlating procedures of Eqs. (19), (21), and (22). The inclusion of these data sets, which are inconsistent with theory, is thus seen to be responsible for the deviations between the most recent representation of the thermal conductivity of nitrogen and earlier correlations [5].

A very similar result can be seen in Fig. 5 for carbon dioxide. Here the baseline of the deviation plot corresponds to the optimum correlation established with respect to a primary data set on the basis of the assessment procedures set out above [23]. If some of the secondary, data previously rejected is included in a fit with the aid of Eq. (22), the correlation changes dramatically.

Once again, the deviations from the primary correlation are very large, amounting to 7% at the highest temperature. Evidently, the temperature dependence of the thermal conductivity given by the two representations is quite different and emphasizes the need for a theoretically based data assessment before a representation is undertaken.

5. CONCLUSIONS

It has been shown that in order to secure an accurate representation of the thermal conductivity of dilute gases, a very careful assessment of the experimental data is necessary. Two means of carrying out such an assessment have been described and illustrated by means of examples. In the first, the simplified Thijsse expression for the thermal conductivity is used as a vehicle to compare experimental data sets with data of proven accuracy. In the second, the high-temperature limiting behavior of one kinetic theory cross-section ratio, D_{int}/D, known from theory for rigid rotors, is employed as an independent check of data. The first procedure is universally applicable, but the second is confined to linear molecules at present, because the limiting behavior of D_{int}/D for other molecules is not yet known.

Arising from this study, it can be seen that there is a clear need to extend the temperature range over which accurate thermal conductivity data are available for gases. It seems that above 450 K there are no reliable experimental values. As a corollary, it follows that a theoretical study of the temperature dependence of the effective cross sections of the kinetic theory would be useful as a guide to the extrapolation of the current, modest *experimental* data base.

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