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Thermal Conductivity of Mixtures of Polyatomic Gases at Low and Moderate Density

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The paper discusses the prediction of the thermal conductivity of gas mixtures containing polyatomic components at low and moderate density. The prediction scheme adopted is based upon the rigorous kinetic theory of gases and makes use of other thermophysical properties of the pure gases and binary mixtures in the evaluation of the thermal conductivity. Comparisons with accurate experimental data indicate that only for systems in which the mass ratio of the species is near unity and for which inelastic collisions are rare, is a reliable prediction of the mixture thermal conductivity at low density possible. On the other hand, the density dependence of the thermal conductivity is quite accurately represented for most systems.

KEY WORDS: gas mixtures; polyatomic gases; thermal conductivity.

1. INTRODUCTION

During the last decade a scheme has been developed for prediction of the transport properties of the pure monatomic gases and their mixtures at low density [1, 2]. The procedure is founded upon the rigorous kinetic theory of such gases and a limited, well-chosen set of accurate measurements. As a result, it is now possible to calculate all of the transport properties of the monatomic gases and those of any of their multicomponent mixtures over a wide temperature range with an accuracy comparable with that of direct measurements [3]. Indeed, in the particular case of thermal conductivity this calculation scheme is predictive, in the sense that measurements of thermal conductivity are not required for its application, and the predic-

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tions are considerably more accurate than all but the most recent measurements [3]. However, the fluids of greatest industrial significance contain polyatomic components, and for such systems the development of an analogous calculation scheme is not yet complete. This is because polyatomic molecules interact through anisotropic pair potentials and store energy in internal modes of motion, and these two factors combine to complicate the rigorous kinetic theory analysis of gases containing such species [4]. Considerable progress has been made for both the viscosity and diffusion coefficients of polyatomic systems [5] because the formal kinetic theory results are identical with those for monatomic gases and because the special characteristics of polyatomic gases have only a minor influence on these properties. On the other hand, for the thermal conductivity, the internal energy of the molecules and its relaxation by the mechanism of inelastic collisions have a profound effect on the form of the kinetic theory results and upon the evaluation of the property.

New experimental data for the thermal conductivity of gases have an estimated uncertainty which is at least one order of magnitude less than that of earlier measurements [6]. The results therefore provide an opportunity to examine just how well the available kinetic theory allows us to predict the thermal conductivity using independent experimental information. An examination of this kind for pure polyatomic gases at low density was carried out recently [7], and it revealed that satisfactory predictions can only be made for gases in which inelastic collisions are rare events. In this paper we extend the examination to gas mixtures involving polyatomic components.

2. METHODOLOGY FOR THE LIMIT OF ZERO DENSITY

The most secure foundation for a predictive scheme for the transport properties of gases is a rigorous kinetic theory analysis of the problem. In the case of the thermal conductivity of polyatomic gas mixtures at low density, the best available calculation scheme of this type is based on the analysis performed by Monchick et al. [8] within the framework of the semiclassical kinetic theory of Wang Chang and Uhlenbeck [4]. For the purposes of prediction, the most convenient formulation of this analysis is that given by Monchick et al. [9], which relates the thermal conductivity of the gas mixture to other measurable properties of the system such as the viscosity and thermal conductivity of its components, diffusion coefficients, and internal heat capacities. The expression for the thermal conductivity for a binary mixture at low density, λ^0_{mix}, may be written in the form [5]

$$\lambda^0_{\text{mix}} = \lambda^0_{\text{mix,HE}} + \Delta\lambda \qquad (1)$$

where the principal term, $\lambda^0_{\mathrm{mix,HE}}$, is given by

$$\lambda^0_{\mathrm{mix,HE}} = \lambda^0_{\mathrm{mix}}(\mathrm{mon}) + \sum_{i=1}^{2} \left[\lambda_i^0 - \lambda_i^0(\mathrm{mon})\right] \left[1 + \sum_{\substack{j=1 \\ j \neq i}}^{2} \frac{x_j D_{i\,\mathrm{int},i}}{x_i D_{i\,\mathrm{int},j}} \right]^{-1} \tag{2}$$

and $\Delta\lambda$ is a small term given by a rather cumbersome expression quoted elsewhere [5, 9]. The term $\lambda^0_{\mathrm{mix}}(\mathrm{mon})$ in Eq. (2) represents the translational contribution to the thermal conductivity and the second term the contribution from internal energy. The term $\Delta\lambda$ of Eq. (1) arises solely from the relaxation processes associated with inelastic collisions. In these expressions λ_i^0 represents the thermal conductivity of the pure gas i and $\lambda_i^0(\mathrm{mon})$ the translational contribution to it. In addition, x_i represents the mole fraction of component i in the mixture and $D_{i\,\mathrm{int},j}$ the diffusion coefficient for internal energy of species i through j [8, 9].

Because we are particularly interested here in the prediction of the mixture thermal conductivity, Eq. (1) has been written so that it automatically reproduces the thermal conductivity of the pure components, but even so it is not an exact result. First, the expression represents only a first-order approximation to the transport coefficient within the Chapman–Cowling solution method for the Wang Chang and Uhlenbeck equation [4]. Second, the Wang Chang and Uhlenbeck theory itself is incomplete because it neglects the spin polarization resulting from imposition of a temperature gradient on a gas in which there are anisotropic interactions [10]. Finally, in deriving Eq. (1), Monchick et al. [9] found it necessary to neglect complex molecular collisions in which both molecules change their internal state or both internal modes of one molecule change in one collision. From the present point of view, the most important of these deficiencies is the first-order nature of the formulation because it is well known that for monatomic mixtures the use of the equivalent first-order formula can lead to substantial underestimates of the mixture thermal conductivity [11].

In order to make use of Eq. (1) for the prediction of the mixture thermal conductivity, it is necessary to employ a number of physical properties. Whenever possible these quantities are obtained from experiment, but in several cases it is possible to do no more than make sensible estimates. Thus, the pure component thermal conductivity $\lambda_i^0(\mathrm{mon})$, as well as the "interaction" thermal conductivity, $\lambda_{ij}^0(\mathrm{mon})$, may be obtained from viscosity data with the aid of the result [5, 9]

$$\lambda_{ij}^0(\mathrm{mon}) = \frac{15(m_i + m_j)k}{8m_i mj}\,\eta_{ij} \tag{3}$$

in which η_{ij} is the "interaction" viscosity [5], and m_i is the molecular mass of species i. The remaining quantities required to evaluate the contribution $\lambda^0_{\mathrm{mix,HE}}$ are the collision integral ratios A^*_{ij} and B^*_{ij} [5] and the diffusion coefficients for internal energy, $D_{i\,\mathrm{int},j}$. In principle, these quantities, which are functionals of the intermolecular pair potentials in the gas, contain contributions from inelastic collisions. However, it is not yet possible to calculate them exactly on a routine basis, so that in most cases it is necessary to employ approximations based on purely elastic collision results. The collision integral ratios A^*_{ij} and $^*_{ij}$ may then be taken from the correlations of the extended law of corresponding states [5] and the diffusion coefficients of internal energy equated to the diffusion coefficients for mass [9], so that

$$D_{i\,\mathrm{int},j} = D_{ij} = \frac{3A^*_{ij}}{5}\,\frac{(m_i + m_j)}{m_i m_j}\,\eta_{ij} \tag{4}$$

For the pure component values of $D_{i\,\mathrm{int},i,}$ it is usually possible to improve upon this assignment because a value may be deduced from an analysis of the pure component thermal conductivity [7].

The explicit effects of inelastic collisions on the mixture thermal conductivity are contained in the term $\Delta\lambda$ whose evaluation requires a knowledge of the internal heat capacities of the pure components $c_{\mathrm{int},i}$ and the four collision numbers for internal energy relaxation in the gas ζ_{ij}. This collision number represents the number of collisions necessary for the relaxation of internal energy of species i by collision with species j. The heat capacities are readily available from standard tabulations [12], and the collision numbers for the pure gases, ζ_{ii}, have usually been determined experimentally [13]. However, the collision numbers for cross-relaxation, ζ_{ij}, must be estimated and, in most cases, the best that can be done is to use the assignment [9]

$$\zeta_{ij} = \zeta_{ii}, \qquad \zeta_{ji} = \zeta_{jj} \tag{5}$$

3. METHODOLOGY FOR ELEVATED DENSITIES

At elevated densities the kinetic theory is even less well developed than for the dilute gas limit. The most successful approach to the description of the transport properties of dense gases is based on a heuristic modification of the Thorne–Enskog equations for the transport properties of a rigid sphere gas by Mason and his collaborators [14]. According to their calculation scheme the thermal conductivity of a dense polyatomic gas mixture is written

$$\lambda_{\text{mix}} = \lambda_{\text{mix}}(\text{mon}) + \lambda_{\text{mix}}(\text{int}) \tag{6}$$

The translational contribution, $\lambda_{\text{mix}}(\text{mon})$, and the internal contribution $\lambda_{\text{mix}}(\text{int})$, correspond to the two terms of the Hirschfelder–Eucken result for the dilute gas with an allowance for the increased collision frequency in the gas which follows from the elevated density. There is no explicit inelastic contribution to the thermal conductivity of the mixture in this formulation, so that it is inherently less accurate than the dilute gas expression.

The effect of increased density on the transport properties of a binary gas mixture is contained within four pseudo-radial distribution functions, $\bar{\chi}_{ij}$, for molecules i and j, which enter into the expressions for $\lambda_{\text{mix}}(\text{mon})$ and $\lambda_{\text{mix}}(\text{int})$. These radial distribution functions can be obtained from the corresponding quantities for the pure gases by means of a combination rule based on the Percus–Yevick equation for hard spheres [15], provided that a hard-sphere collision diameter, σ_{ij}, is defined for each interaction. In turn, the radial distribution functions for the pure gases may be obtained by application of Eq. (6) to the thermal conductivity data for each pure component. Thus, Eq. (6) is employed for interpolation between the thermal conductivity of the pure gases at constant molar density.

It follows from this description that the only additional quantities required for the evaluation of the thermal conductivity of gas mixtures at elevated density are the thermal conductivities of the pure components at the density in question and the rigid sphere collision diameters σ_{ij}. The former are derived from experiment, whereas according to the modified Enskog procedure, the latter may be derived from second virial coefficient data [16] by means of the equation

$$4\pi\sigma_{ij}^3/5 = \gamma_{ij} = 6\left[B_{ij} + T(dB_{ij}/dT)\right]/5 \tag{7}$$

If the interaction second virial coefficients for a particular mixture are not available, an appropriate estimation of the mean-free path shortening parameter, γ_{ij}, may be obtained from the simple empirically verified combination rule [14]

$$\gamma_{ij} = \left(\gamma_{ii}^{1/3} + \gamma_{jj}^{1/3}\right)^3/8 \tag{8}$$

4. MIXTURES SELECTED FOR STUDY

Studies of pure polyatomic gases have shown that the frequency of inelastic collisions in the gas has a large effect upon our ability to predict its

Table I. Quantities Used in the Calculation of the Thermal Conductivity at a Temperature $T = 300.65$ K

Pure Gases							
	λ_i^0 $(\mathrm{mW \cdot m^{-1} \cdot K^{-1}})$	$\lambda_i^0(\mathrm{mon})$ $(\mathrm{mW \cdot m^{-1} \cdot K^{-1}})$	c_{int}^0/k	A_{ii}^*	ζ_{ii}	$D_{i\,\mathrm{int},i}/D_{ii}$	γ_{ii} $(\mathrm{m^3 \cdot mol^{-1}})$
H_2	188.8	138.7	0.968	1.165	200	1.0	2.60×10^{-5}
$H_2{}^a$	192.2	141.0	0.968	1.165	200	1.0	2.60×10^{-5}
He	155.9	155.9	0	1.131	∞	1.0	1.25×10^{-5}
D_2	134.3	98.75	1.012	1.165	200	1.0	2.50×10^{-5}
CH_4	34.93	21.72	1.806	1.094	12.3	1.003	8.02×10^{-5}
Ne	49.45	49.45	0	1.119	∞	1.0	2.18×10^{-5}
N_2	26.03	19.95	1.003	1.094	5.0	0.94	6.31×10^{-5}
Ar	17.74	17.74	0	1.092	∞	1.0	5.43×10^{-5}
Ar^a	18.20	18.20	0	1.092	∞	1.0	5.43×10^{-5}
CO_2	16.85	10.65	1.980	1.116	2.5	0.89	1.76×10^{-4}
CO	24.83	19.95	1.005	1.094	4.0	0.79	6.31×10^{-5}

Unlike interaction quantities

	λ^0_{12}(mon) ($mW \cdot m^{-1} \cdot K^{-1}$)	A^*_{12}	B^*_{12}	ζ_{12}	ζ_{21}	$D_{i\,\mathrm{int},j}/D_{ij}$	γ_{ij} ($m^3 \cdot mol^{-1}$)	m_1/m_2
N_2–CO	19.95	1.094	1.159	5.0	4.0	1.0	6.31×10^{-5}	1
CO_2–Ar	14.48	1.094	1.128	2.5	∞	1.0	1.04×10^{-4}	1.13
N_2–Ne	31.54	1.150	1.150	5.0	∞	1.0	3.89×10^{-5}	1.4
N_2–CH_4	21.57	1.092	1.133	5.0	12.3	1.0	7.13×10^{-5}	1.75
He–H_2	145.5	1.165	1.136	∞	200	1.0	1.84×10^{-5}	2
D_2–H_2	120.5	1.165	1.094	200	200	1.0	2.55×10^{-5}	2
N_2–He	66.06	1.099	1.218	5.0	∞	1.0	3.13×10^{-5}	7
CO_2–He	56.91	1.116	1.100	2.5	∞	1.0	6.22×10^{-5}	11
N_2–H_2	74.13	1.102	1.078	5.0	200	1.0	4.18×10^{-5}	14
Ar–H_2[a]	79.54	1.096	1.096	∞	200	1.0	3.84×10^{-5}	20

[a] $T = 308.15$ K.

thermal conductivity from other properties [7]. Furthermore, it has been established by work on monatomic gas mixtures that for systems in which the mass ratio of the species differs substantially from unity, the first-order kinetic theory formulas underestimate the thermal conductivity [11]. We therefore expect that for polyatomic gas mixtures, the accuracy of the prediction of the thermal conductivity will be influenced by both of these factors, and accordingly we select binary mixtures for study, which encompass as wide a range of them as possible.

The complete list of the 10 systems studied, for each of which thermal conductivity and viscosity data with an uncertainty of less than ±0.3% are available, is given in Table I, together with all of the values used for the prediction of the thermal conductivity. All of the experimental data have been taken from the results of our own work [6, 17–25]. At one extreme, He/H_2 and D_2/H_2 represent cases in which inelastic collisions are rare and the mass ratio is near unity. At the other extreme, CO_2/He represents a system in which the mass ratio is very different from unity and inelastic collisions among the CO_2 molecules are frequent (ζ_{11} small).

5. RESULTS IN THE LIMIT OF ZERO DENSITY

Figure 1 contains a plot of the deviations of the experimental thermal conductivity data for the systems He/H_2 and Ar/H_2 from the predictions of Eq. (1). In both cases the Hirschfelder–Eucken result ($\Delta\lambda = 0$) is identi-

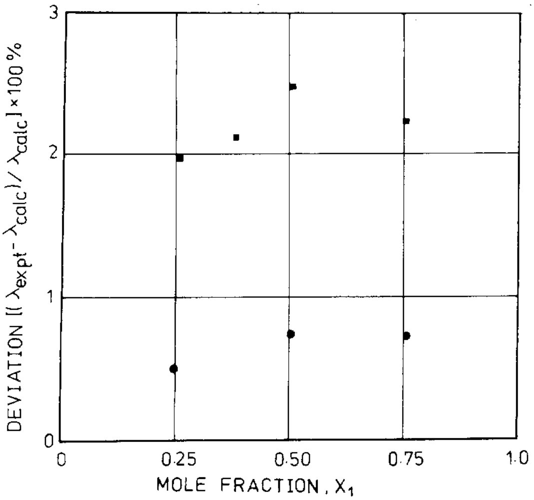

Fig. 1. Deviations of the experimental thermal conductivity from the predicted values. ●; $He-H_2$ [19], $T = 300.65$ K; ■; $Ar-H_2$[21], $T = 308.15$ K.

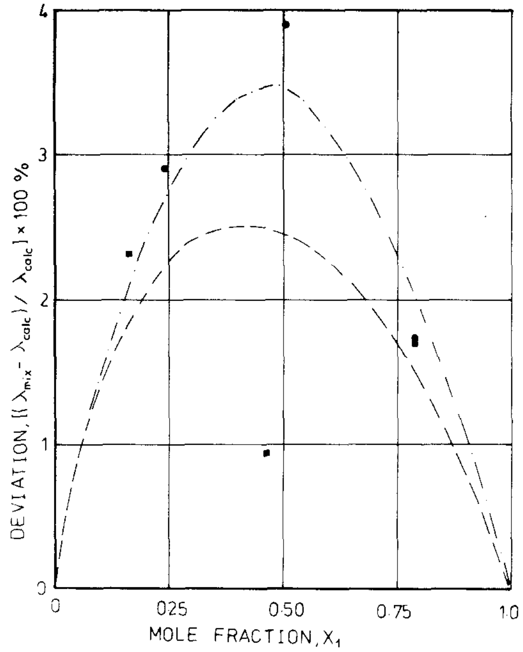

Fig. 2. Deviations of the experimental thermal conductivity from the predicted values. N_2–Ne: ●, experimental data [20]; ——·——, Hirschfelder–Eucken result. N_2–He: ■, experimental data [20]; — — —, Hirschfelder–Eucken result.

cal with that for the full form of Eq. (1) owing to the rarity of inelastic collisions. For He/H_2 the calculation underestimates the thermal conductivity by no more than 0.7%, whereas for Ar/H_2 the discrepancy amounts to 2.5%. Because inelastic collisions are rare in both cases, the difference is attributed principally to the effect of the disparate masses in the second case. Figure 2 contains a comparison with the results for mixtures of neon and helium with nitrogen for which inelastic components are relatively frequent. In these cases the predictions of the full form of Eq. (1) are worse than in the previous examples. For the helium-nitrogen system it seems likely that this is again attributable to the effect of the species mass ratio, but for neon-nitrogen it is much more likely to be a result of the failure to treat inelastic collisions properly. It is interesting to note that the Hirschfelder–Eucken results, also included in Fig. 2, more nearly reproduce the experimental data than do the results of the full Eq. (1). Finally, among the monatomic-polyatomic systems, Fig. 3 contains the results for mixtures of helium and argon with carbon dioxide, for which inelastic collisions are frequent. For the CO_2/Ar system the deviations of the experimental data

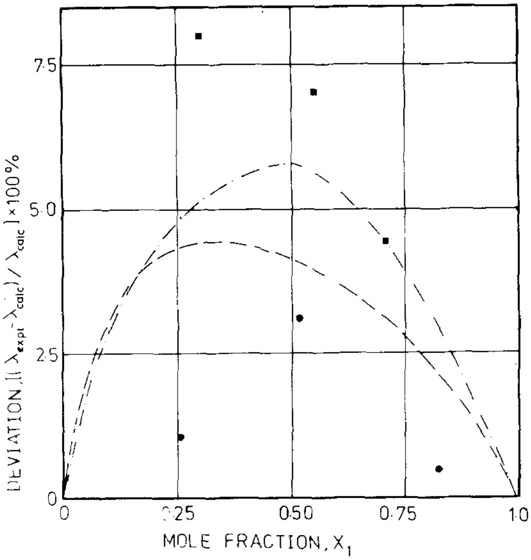

Fig. 3. Deviations of the experimental thermal conductivity from the predicted values. CO_2–Ar: ●, experimental data [23]; ——·——, Hirschfelder–Eucken result. CO_2–He: ■, experimental data [23]; ———, Hirschfelder–Eucken result.

from the predictions are quite large (~3%), and even their qualitative behavior cannot be described by Eq. (1) [26]. For He/CO_2, a system with a species mass ratio of 11, the deviations are even more pronounced. Again the Hirschfelder–Eucken result more closely reproduces the experimental data.

Turning to mixtures of two polyatomics, Fig. 4 contains results for D_2–H_2 and N_2–H_2. For the former system, in which the mass ratio is quite close to unity and inelastic collisions are rare, the agreement of prediction and experiment is good. However, for the latter system, the species' masses are very different, N_2 undergoes quite frequent inelastic collisions, and the predictions are very much worse. Finally, in Fig. 5 we consider mixtures in which the mass ratio is very close to unity and inelastic collisions are relatively frequent. Here, the deviations from the predictions are much smaller than for the other systems considered.

A number of general observations may be made about these results. The full form of Eq. (1) almost always underestimates the experimental thermal conductivity, and the extent of the underestimation grows as the frequency of inelastic collisions in one or both of the gases increases and as the mass ratio of the species departs from unity. Only for cases in which the

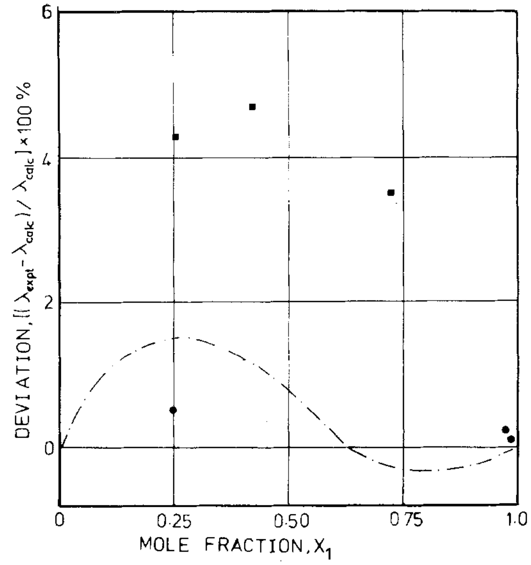

Fig. 4. Deviations of the experimental thermal conductivity from the predicted values. D_2–H_2: ●, experimental data [17]; N_2–H_2: ■, experimental data [22]; ——·——, Hirschfelder–Eucken result.

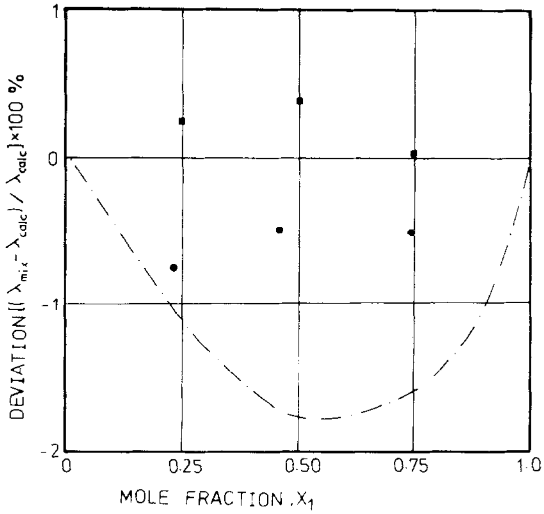

Fig. 5. Deviations of the experimental thermal conductivity from the predicted values. N_2–CH_4: ●, experimental data [24]; ——·——, Hirschfelder–Eucken result. N_2–CO: ■, experimental data [25].

mass ratio is near unity and inelastic collisions are rare is an accurate prediction of the thermal conductivity possible. The effect of the mass ratio is likely to be a result of the use of a first-order kinetic theory formulation, whereas the observation concerning inelastic collisions indicates that when such collisions are frequent, it is inappropriate to employ values for A^*_{ij}, B^*_{ij}, and $D_{i\,\mathrm{int},j}$ computed on the basis of elastic collisions only. In general, the simpler and less correct Hirschfelder–Eucken result reproduces the experimental data more successfully, but this is probably fortuitous.

The agreement between calculation and experiment can be significantly improved if empirical adjustments are made to the quantities A^*_{ij} and B^*_{ij}. Indeed, the calculations are particularly sensitive to B^* when the species mass ratio is very different from unity. Such adjustments can serve as a useful mechanism for the correlation of experimental data; however, without theoretical guidance, they are of little value in the development of a predictive scheme.

6. RESULTS AT ELEVATED DENSITIES

In the limit of zero density, the high density formulation of Mason et al. [14] reduces to the Hirschfelder–Eucken result of Eq. (2). Consequently, the predictions of the absolute values of the thermal conductivity at elevated densities suffer from the same errors associated with the predictions of the zero-density thermal conductivity. In order to examine the predictions of the density dependence of the mixture thermal conductivity in isolation, it is therefore preferable to consider the ratio $\lambda_{\mathrm{mix}}/\lambda^0_{\mathrm{mix}}$ for a selection of the systems studied earlier. Figure 6 contains a plot of the ratio as a function of density for the systems neon-nitrogen and methane-nitrogen for which the mass ratio is near unity. The predicted result is in good agreement with that observed experimentally, the greatest deviation amounting to 1% in the thermal conductivity itself. Figure 7 contains a similar plot for the argon-hydrogen system where the agreement is even better despite the disparate masses of the two species. These results suggest that given an accurate prediction of the zero-density thermal conductivity of the mixture, the scheme of Mason et al. [14] may be employed to extend the data to higher densities, even for systems in which inelastic collisions are quite frequent and the species differ considerably in mass.

In Fig. 8 the ratio $\lambda_{\mathrm{mix}}/\lambda^0_{\mathrm{mix}}$ for mixtures of helium and carbon dioxide is plotted as a function of density. In this case there is evidently a qualitative difference between the predicted behavior and that observed experimentally, even though the quantitative discrepancy in the thermal conductivity does not exceed 2%. It is likely that the qualitative difference arises because, under the conditions of interest, carbon dioxide is near its

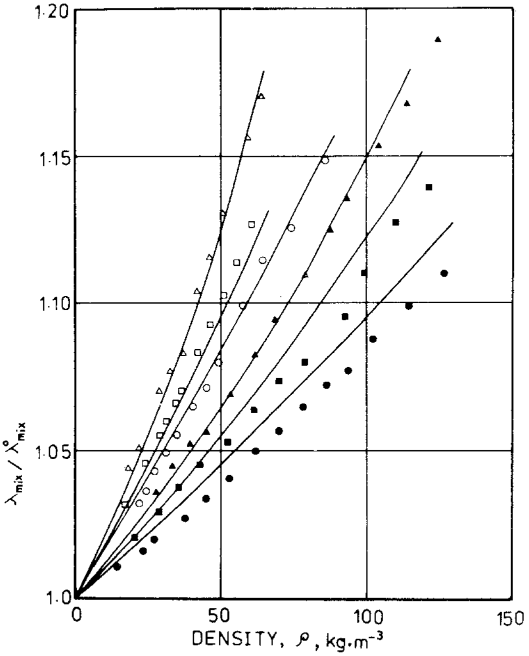

Fig. 6. The ratio $\lambda_{mix}/\lambda^{0}_{mix}$ as a function of density. The predictions are indicated by the solid lines. Experimental data [20] as follows. N_2–Ne: ●, $x_{N_2} = 0.2363$; ■, $x_{N_2} = 0.5063$; ▲, $x_{N_2} = 0.7810$. N_2–CH_4: ○, $x_{N_2} = 0.2564$; □, $x_{N_2} = 0.5432$; △, $x_{N_2} = 0.7707$.

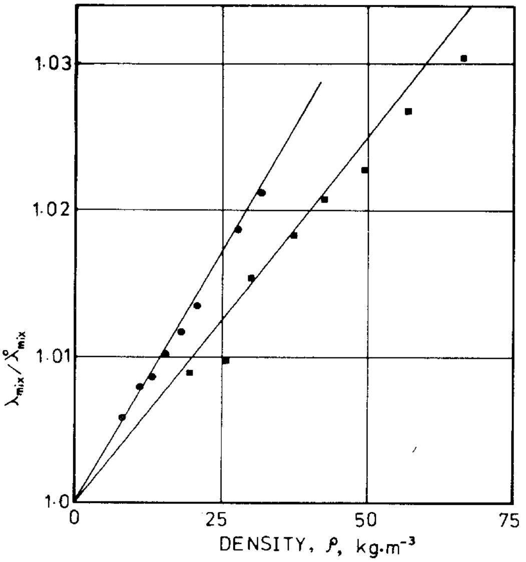

Fig. 7. The ratio $\lambda_{mix}/\lambda^{0}_{mix}$ as a function of density for Ar–H_2 mixtures. The predictions are indicated by the solid lines. Experimental data [21] as follows. ●, $X_{Ar} = 0.2496$; ■, $X_{Ar} = 0.7386$.

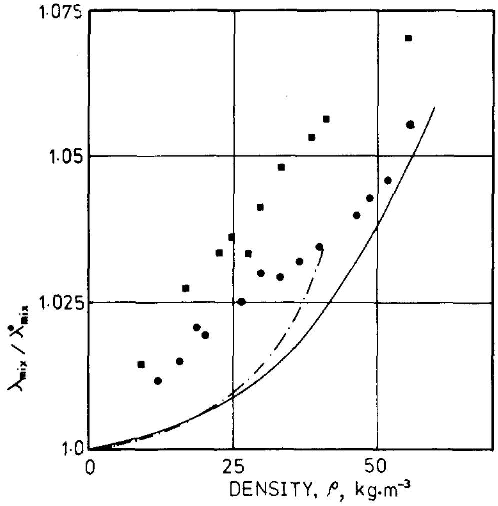

Fig. 8. The ratio $\lambda_{mix}/\lambda^{0}_{mix}$ as a function of density for CO_2–He mixtures. Experimental data [23] as follows. ●, $X_{CO_2} = 0.5385$; ———, prediction. ■, $X_{CO_2} = 0.3044$; ——·——, prediction.

critical state where the thermal conductivity of the pure gas exhibits anomalous behavior. This behavior is not incorporated into the formalism of the Thorne–Enskog equations, and it is therefore to be expected that the theory will not be adequate in such circumstances [23].

7. CONCLUSIONS

For binary gas mixtures containing polyatomic components in which the species have a similar mass and for which inelastic collisions are rare, it is possible to perform reliable predictions of the thermal conductivity which have an accuracy comparable with that of direct measurement. As the frequency of inelastic collisions increases and the masses of the species become more disparate, the accuracy of the predictions decreases. In the worst case studied, the deviations from accurate data amount to 7%, which could be significant in engineering design calculations. Moreover, for all of the systems studied here, a considerable body of accurate data on other thermophysical properties is available which may be utilized in the prediction scheme. For other systems of industrial importance, this is rarely the case, and the predictions in these cases will be correspondingly worse.

The results of the present examination indicate that future effort should be devoted primarily to the improvement of the kinetic theory description of polyatomic gas mixtures in the limit of zero density. The methods available for prediction at elevated density yield essentially the correct density dependence of the thermal conductivity except for systems involving a component near the critical state.

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