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TRANSPORT PROPERTIES OF POLAR GAS MIXTURES

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ABSTRACT

A previously proposed model for the calculation of viscosity, diffusion, and thermal diffusion in dilute polar gases is extended to mixtures. A simple set of combination rules for the potential parameters is postulated and discussed. Available data limit the comparison of calculation and experiment to binary mixtures of a polar and a nonpolar gas. The agreement exhibits a success comparable to that obtained for models and combination rules involving only nonpolar gases. It is concluded that the present model and combination rules can be used to describe the transport properties of mixtures with a fair measure of success, and that any attempts to refine the combination rules should await more experimental results of high accuracy, especially results on thermal diffusion and diffusion.

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TRANSPORT PROPERTIES OF POLAR GAS MIXTURES[1]

I. INTRODUCTION

Recently a simplified model was proposed (Ref. 1)[2] to account for the transport properties of pure polar gases. Briefly, this model assumes that two polar molecules interact according to a Stockmayer potential,

$$\varphi = 4\varepsilon_o \left[\left(\frac{\sigma_o}{r}\right)^{12} - \left(\frac{\sigma_o}{r}\right)^{6} \right] - \frac{\mu_1 \mu_2}{r^3} \zeta , \qquad (1)$$

where μ_1 and μ_2 are the molecular dipole moments and ζ is dependent on the relative orientation of the dipoles (ζ ranges from -2 to +2). This is just the familiar Lennard-Jones (12-6) potential with an added dipole term. The model then approximates ζ throughout a collision as a constant, ζ_o, which is interpreted as the value of ζ at the distance of closest approach. This is equivalent to replacing φ by a multiplicity of central field potentials, corresponding to all values of ζ_o between -2 and +2. The collision integrals were then evaluated in the usual manner and averaged over all relative orientations. These averaged collision integrals were then used to fit the viscosity data for a number of pure polar gases,

[1]Research was supported in part by the U. S. Atomic Energy Commission, and in part under Contract NOrd 7386 with the Bureau of Naval Weapons, Department of the Navy.

[2]References are on pages 39 to 41.

and in this way sets of the potential parameters ϵ_o, σ_o, and $\delta_{max} = 1/2\ \mu^2/(\epsilon_o \sigma_o{}^3)$ were obtained. The parameters obtained were physically more reasonable than those obtained from another model of polar gases (Refs. 2,3) in which only the value $\zeta_o = -2$ is considered. Other properties of pure gases, such as the self-diffusion coefficient, the isotopic thermal diffusion factor, and the second virial coefficient, were calculated with these parameters and agreed satisfactorily with the scanty experimental data available.

In the present paper the foregoing model is extended to mixtures containing polar gases. As in the previous work (Ref. 1) thermal conductivity is not considered because of its strong dependence on the transport of internal molecular energy. Although the results are presumably general, lack of experimental data prevents comparing theory and experiment for any but binary mixtures of a polar and a nonpolar gas. We first postulate a set of combination rules for the calculation of the potential parameters for the interaction of unlike molecules from the potential parameters for the interaction of like molecules. This permits the calculation of mixture properties from the properties of the pure components. If the calculated properties agree satisfactorily with experimental values, then a check is obtained on the postulated combination rules and on the consistency of the original potential parameters for the pure gases.

We have not attempted the inverse problem of calculating the unlike parameters directly from experimental data on mixtures because of the lack of suitable experimental data of sufficient accuracy. The two most useful properties for this procedure are the binary diffusion coefficient and thermal diffusion factor, but practically none of the measurements of these properties for polar gases cover a wide enough temperature range with suitable accuracy. A program of accurate mixture measurements for the determination of unlike potential parameters is most desirable, but in the meantime we must make do with semi-empirical combination rules.

In the next section suitable combination rules are discussed, and in the following section the comparison of theory with experiment is given.

II. COMBINATION RULES FOR POTENTIAL PARAMETERS

The intermolecular potential of Eq. (1) can be written for a general two-body interaction as:

$$\varphi_{ij} = 4\epsilon_{ij} \left[\left(\frac{\sigma_{ij}}{r}\right)^{12} - \left(\frac{\sigma_{ij}}{r}\right)^{6} + 1/2\ \delta_{ij} \left(\frac{\sigma_{ij}}{r}\right)^{3} \zeta_{ij} \right] , \quad (2)$$

where the subscripts i and j denote the molecular species of the interacting pair. δ_{ij} is shorthand for $1/2\ \mu_i\ \mu_j/(\epsilon_{ij}\ \sigma_{ij}^{\ 3})$. Because the dipole moments are known independently, δ_{ij} is not really an independent parameter. In the case that either the i-th or the j-th molecule is nonpolar, δ_{ij} vanishes and φ_{ij} is just the ordinary Lennard-Jones (12-6) potential.

Most of the experimental data have been obtained for pure polar gases, so that potential parameters are available only for the interactions of like molecules. In principle, the potential parameters for the interactions of unlike molecules can be derived from the transport properties, but the experimental data are too meager for this purpose at present. In the case of the Lennard-Jones (12-6) potential, the semi-empirical combination rules (Ref. 4),

$$\epsilon_{ij} = (\epsilon_{ii}\ \epsilon_{jj})^{1/2} \quad (3)$$

$$\sigma_{ij} = 1/2(\sigma_{ii} + \sigma_{jj}) , \quad (4)$$

have been found to be very useful for predicting the transport properties of mixtures of nonpolar gases. Equation (4) is exact for rigid spheres and Eq. (3) is suggested by the theory of the London dispersion forces (Ref. 5). Actually, Eq. (3) is an approximation to more elaborate expressions which have been suggested (Refs. 6 through 9). However, these more elaborate expressions do not always lead to consistently better agreement with experiment, and we therefore prefer for the present to keep Eq. (3) because of its simplicity. As more and better experimental data become available, it may be possible to devise better semi-empirical combination rules than Eqs. (3) and (4).

In the case of polar molecules, the above combination rules must be supplemented by a third rule determining δ_{ij}. But since δ_{ij} is determined by μ_i, μ_j, ϵ_{ij}, and σ_{ij}, it is easily found that

$$\delta_{ij} = (\delta_{ii}\,\delta_{jj})^{1/2}\left[\frac{(\sigma_{ii}\,\sigma_{jj})^{1/2}}{\sigma_{ij}}\right]^3. \tag{5}$$

A simplified version of Eq. (5) amounting to $\delta_{ij} = (\delta_{ii}\,\delta_{jj})^{1/2}$ has been suggested by Rowlinson (Ref. 10) in connection with his work on the second virial coefficients of polar gases, but no experimental data are available for mixtures of two polar gases with which to test the combination rule for δ_{ij}. For mixtures of a polar and a nonpolar gas, which are the only ones considered in the present paper because of the data available, we have $\delta_{ij} = 0$ if $i \neq j$. However, if the r^{-6} term in φ_{ij} contains in addition to the usual London dispersion energy a contribution from the (averaged) dipole-induced dipole energy,

$$\varphi_{ij}(\mathrm{ind}) = -\frac{1}{r^6}(\mu_i^2\,\alpha_j + \mu_j^2\,\alpha_i)\ , \tag{6}$$

where α is the (average) polarizability. The combination rules Eqs. (3) and (4) can be corrected for this effect (Ref. 11), but we have not done so for the following reasons. In the first place, this induction energy seldom amounts to as much as 10% of the corresponding dispersion term (Ref. 12), so that its net effect may be presumed to be small. In fact, it usually contributes less to the interaction energy than the ignored next higher term in the dispersion energy (which varies as r^{-8}). In the second place, it affects ϵ_{ij} and σ_{ij} in opposite directions, which tends to reduce the net effect on a calculated transport coefficient. Finally, the simple rules (3) and (4) use experimental values of ϵ_{ii} and σ_{ii} which already contain a contribution from the induction energy. Since the thermal diffusion factor is especially sensitive to the intermolecular forces, however, we repeated all the thermal diffusion calculations with the corrected (Ref. 11) combination rules. The agreement with experiment was not found to be consistently better and so these calculations are not reported here.

The potential parameters for the pure gases which we have used are listed in Table I (page 19). These have all previously been reported elsewhere, of course, but in many cases several sets of parameters have been proposed for a single gas, and it therefore seemed best to record the particular values used.

III. COMPARISON WITH EXPERIMENT

A. Viscosity

The first test of the polar gas parameters and of the combination rules is the prediction of the viscosity of binary mixtures, which can be written in the form (Ref. 13),

$$\left[\eta_{mix}\right]_1 = \left[\frac{x_1^2}{H_{11}} + \frac{x_2^2}{H_{22}} - \frac{2x_1 x_2 H_{12}}{H_{11} H_{22}}\right]\left[1 - \frac{H_{12}^2}{H_{11} H_{22}}\right]^{-1}, \quad (7)$$

$$H_{11} = \frac{x_1^2}{[\eta_1]_1} + \frac{2x_1 x_2}{M_1 + M_2} \frac{RT}{p[D_{12}]_1}\left[1 + \frac{3M_2}{5M_1} A_{12}^*\right],$$

$$H_{12} = -\frac{2x_1 x_2}{M_1 + M_2} \frac{RT}{p[D_{12}]_1}\left[1 - \frac{3}{5} A_{12}^*\right],$$

where x_1 and x_2 are mole fractions, and H_{22} is obtained from H_{11} by interchanging the subscripts 1 and 2. The gas constant R and the pressure p should be in consistent units; e.g., R in cm^3 -atm/mole-^oK and p in atm, or R in erg/mole-^oK and p in dyne/cm^2. This is only a first approximation, and $[\eta_i]_1$ and $[D_{12}]_1$ are only the first Chapman-Cowling approximations to the viscosity of the pure gas and the binary diffusion coefficient, respectively. In practice, however, the experimental value of η_i was substituted for $[\eta_i]_1$ to force agreement with experiment at the ends of the composition range.

The values of $[D_{12}]_1$ and A_{12}^* were calculated from the tabulated collision integrals for the Lennard-Jones (12-6) potential, using the potential parameters determined from the combination rules Eqs. (3) and (4).

In this form, the calculations are really a test of the ability of the model to predict values of D_{12} and A_{12}^*. Since A_{12}^* depends only weakly on the model and its parameters, the test is essentially only through D_{12}. The results are presented in Table II (page20). The deviation from experiment is always less than 5%, frequently less than 1%, and averages about 1 to 2%. That is, the agreement in general is within experimental error and compares favorably with the success of the 12-6 or exp-6 models (Refs. 14,15) for nonpolar gases. The experimental results of Jung and Schmick deviate more from the theoretical than do those of Trautz and his coworkers, even for very similar systems such as NH_3-air (Jung and Schmick) and NH_3-N_2 (Trautz and Heberling). There is indeed some suggestion of systematic error in Jung and Schmick's experiments. The system with the largest deviations, H_2-$(C_2 H_5)_2O$, is one for which experimental viscosities for one component (ether) were not available and calculated values had to be used. Part of the deviation may be due to this. Even so, the marked maximum is correctly predicted. In general, the deviations are not random, the calculated values usually being less than the experimental values.

On the whole, however, the agreement with experiment is satisfactory, and indicates that the combination rules can be used to predict the viscosity of a binary mixture of a polar and a nonpolar gas with some degree of confidence.

Since these calculations are essentially a check on D_{12}, it is interesting to inquire how much error in D_{12} is associated with a given error in η_{mix}. It is not difficult to show from Eq. (7) (the terms in H_{12} are small and can be ignored for this purpose) that a given percentage error in D_{12} appears in η_{mix} reduced by a factor of roughly four or five. The observed deviations in η_{mix} therefore correspond to deviations in D_{12} of roughly 5 to 10%. This figure is consistent with the results for the direct comparison of calculated and experimental values of D_{12}, reported in the following section.

B. Diffusion

The diffusion coefficient is a much more sensitive test of the combination rules than is the viscosity because in the first approximation it includes only the 1-2 interaction. To this approximation the binary diffusion coefficient in cm^2/sec is given by (Ref. 16)

$$[D_{12}]_1 = \frac{0.002628\ T^{3/2}}{p\ \sigma_{12}^2\ \Omega_{12}^{(1,1)*}} \left[\frac{M_1 + M_2}{2M_1\ M_2}\right]^{1/2} , \qquad (8)$$

where T is in $^\circ K$, p is in atm, σ_{12} is in A, and M_1 and M_2 are in g/mole. The reduced collision integral for diffusion, $\Omega_{12}^{(1,1)*}$, is a function of $T_{12}^* = kT/\varepsilon_{12}$ and was obtained from the tabulated collision integrals for the 12-6 potential. The calculated and experimental values of D_{12} are compared in Table III (page 32). The average deviation is about 7%, which is consistent with the deviations found for the mixture viscosities. However, it is also approximately the same as the discrepancies between the results of different workers investigating the same system. As a result it is difficult to say whether the model is at fault or whether the experimental accuracy is insufficient for a careful test.

Many of the experimental results involving polar vapors were obtained by the liquid evaporation method, which is often subject to rather large errors. For comparison some results are included which involve a nonpolar vapor (H_2-CCl_4 and O_2-CCl_4) obtained by the same method. Some results are also included for the nonpolar gas pair H_2-N_2 to compare with the results by the same worker (Bunde) on H_2-NH_3 and N_2-NH_3. In all cases it is seen that the discrepancy between theory and experiment is about the same for the nonpolar mixtures as for the polar mixtures. The model for polar gases is thus at least no worse than the corresponding 12-6 model for nonpolar gases.

Use of the second approximation for D_{12} in place of Eq. (8) would not be likely to change the agreement much, since the second approximation usually differs from the first approximation on the order of only a couple per cent.

The parameters for the O_2-H_2O system obtained by the combination rules (σ_{12} = 3.072 A and ϵ_{12}/k = 239°K) differ markedly from those obtained by Walker and Westenberg (Ref. 17) directly from a least mean squares fit of their high temperature diffusion measurements (σ_{12} = 3.335 A and ϵ_{12}/k = 80°K). Their value of 80°K for ϵ_{12}/k seems abnormally low. However, the experimental viscosity data for pure H_2O seemed to indicate that a point dipole model for H_2O was too simplified (Ref. 1). Indeed, it is easily seen by scanning Table III that the discrepancies between theory and experiment are generally larger for mixtures containing H_2O as one component.

C. Thermal Diffusion

A quantity which is more difficult to predict theoretically is the thermal diffusion factor, which describes the degree of separation of a mixture into its components under the influence of a temperature gradient. The sign convention is that a positive coefficient means the heavy component concentrates at the lower temperatures. To be consistent with this convention, the theoretical formulas have to be written so that the subscript 1 refers to the heavy component and the subscript 2 to the light component. The thermal diffusion factor α_T can be written as the product of a temperature-dependent factor, a factor which depends primarily on composition and only weakly on temperature, and a third factor which includes the corrections for higher approximations, as follows:

$$\alpha_T = (6C_{12}^{*}-5) \left[\frac{x_1 S_1 - x_2 S_2}{x_1^2 Q_1 + x_2^2 Q_2 + x_1 x_2 Q_{12}} \right] (1 + \kappa) , \quad (9)$$

where $(6C_{12}^{*}-5)$ is the factor which depends on temperature alone, and κ is the corrections for higher approximations. The dimensionless quantity C_{12}^{*} is a ratio of collision integrals which depends only on T_{12}^{*}, and has been tabulated. Expressions for S_1, S_2, Q_2, and Q_{12} in terms of tabulated or experimental quantities have been given (Ref. 18) but are not written here because of their length.

The largest error in the calculation of α_T comes from the factor $(6C_{12}^{*}-5)$, which is the reason α_T is such a sensitive test of any model. To see why this is so, we write the identity (Ref. 19)

$$6C_{12}^{*}-5 = 2\left[2 - \frac{d \ln [D_{12}]_1}{d \ln T}\right]_{p=\text{const}} . \qquad (10)$$

The magnitude of the derivative is usually near 2 (it is often equal to 2 at a low temperature, called the inversion temperature for thermal diffusion, where α_T changes sign). Thus the calculation of α_T requires first the prediction of the derivative of the diffusion coefficient, and second the difference between the derivative and a number nearly equal to it. These are rather drastic procedures; in view of the admittedly approximate nature of the collision model great accuracy is not to be expected. It is to be hoped, however, that the main features will be properly reproduced by the model.

Before proceeding to the comparison of calculation and experiment, a few qualitative remarks about the behavior of the factors in Eq. (9) for α_T may be helpful. First of all, $*$ is not a very important quantity, although it is often not negligible in comparison with experimental uncertainties. In all cases discussed in this paper, this fact has been verified by direct calculation. Secondly, $(6C_{12}^{*}-5)$ is positive for all cases considered. Errors in the magnitude of α_T or its variation with temperature are attributed mainly to errors in $(6C_{12}^{*}-5)$ due to the causes discussed in the preceding paragraph. Thirdly, the variation of α_T with composition at constant temperature is determined primarily by the factor in brackets in Eq. (6) (which will be called the S, Q factor for short). Furthermore, since $(6C_{12}^{*}-5)$ is constant at a fixed temperature, any change in sign of α_T (inversion) with varying composition must be due to the S, Q factor. Such an inversion is extremely unusual in thermal diffusion work. The reason for this sort of inversion is rather interesting, and has been discussed by Chapman (Ref. 20). The effect depends only on the signs of S_1 and S_2, the signs of Q_1, Q_2, and Q_{12} being always positive. If the masses of the two molecular species are appreciably different, then S_1 is positive and S_2 is negative, which is the normal situation. If the masses are nearly equal, however, the signs of S_1 and S_2 are determined by the relative molecular "sizes" (actually the cross sections). An inversion requires that S_1 and S_2 have the same sign and that $|S_2| > |S_1|$. This can only happen if the cross section for a 1-2 molecular collision bears a peculiar relation to the cross sections for 1-1 and 2-2 collisions, not being a rigid sphere-like average. Thus the whole effect depends in a sensitive way on the relation of the 1-2 intermolecular force law to the 1-1 and 2-2 force laws.

The first case of this type discovered was neon-ammonia, for which Grew (Ref. 21) found an inversion at 75 mole % neon by means of a thermal diffusion column (only the sign of α_T could be found in this way with certainty, not the magnitude). Some years later the system was re-examined by Clusius and Huber (Ref. 22), who used pure isotopes and a new experimental technique by which the magnitude as well as the sign of α_T could be determined (the trennschaukel, or "swing-separator"). Their results for the three systems Ne^{20}-ND_3, Ne^{20}-NH_3, and Ne^{22}-NH_3 are shown in Fig. 1 together with our calculated curves (using Kihara's second approximation (Ref. 18). The agreement is surprisingly good in view of the possible uncertainties of the calculations. For one system α_T is always negative (i.e., the neon concentrates at the hot boundary), but for the other two there is an inversion with composition. The calculations correctly predict these results and also give the correct magnitude of α_T.

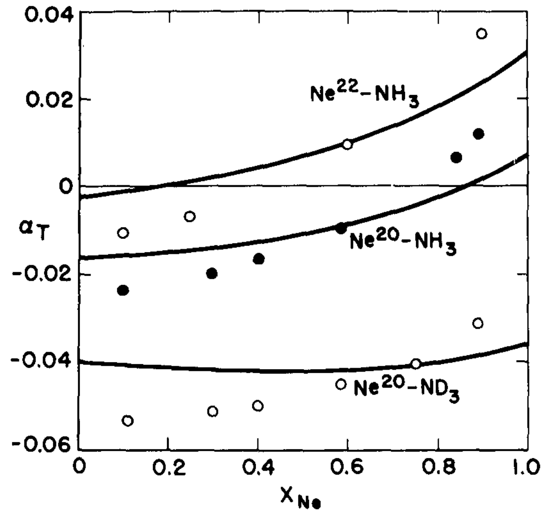

Fig. 1 THERMAL DIFFUSION FACTORS FOR ISOTOPIC NEON-AMMONIA MIXTURES AT 383°K

The trend of α_T increasing with increasing proportion of the heavy gas (neon) is also somewhat unusual. The usual trend is for α_T to increase with increasing proportion of the light gas. This reverse trend occurs when the "size" of the light molecule is larger than usual or the "size" of the heavy molecule is smaller. Here the heavy molecule (neon) is small and the light molecule (ammonia) is large, contrary to the usual order of things. A nonpolar case for which this same effect occurs is a mixture of helium (heavy but small) and hydrogen (light but large) (Ref. 23).

Another known composition inversion occurs in one of the isotopic combinations of argon-hydrogen chloride. Clusius and Flubacher (Ref. 24) have investigated the four isotopic systems Ar^{40}-DCl^{37}, Ar^{40}-HCl^{37}, Ar^{40}-DCl^{35}, and Ar^{40}-HCl^{35}. The first of these shows an inversion. The oddest feature of these four systems is that one would expect α_T to increase steadily as the mass of the hydrogen chloride successively takes on the values of 39, 38, 37, and 36, but the middle two are reversed, so that the order of increasing α_T is 39, 37, 38, 36. It is doubtful whether this reversal of order can be accounted for by any theory which involves intermolecular forces of a central or quasi-central nature which are assumed to be the same for different isotopic species of the same molecule (as our model does — one central force law for each relative orientation of a pair of colliding molecules). It would appear necessary to take into account explicitly the nonspherical nature of the different species of hydrogen chloride molecule. This failure of theory is not confined to polar gases, however, a particularly striking example being that of the nonpolar mixture D_2-HT. Because the molecular masses are equal a central force theory predicts no thermal separation, but a significant separation is found experimentally (Ref. 25), presumably due to the asymmetry of the HT molecule.

Omitting the two reversed systems, the experimental and calculated results for AR^{40}-DCl^{37} and Ar^{40}-HCl^{35} are shown in Fig. 2. The results are not as good as for Ne-NH_3. Although the calculated magnitude of α_T is about right, and the calculated change of α_T from one system to the other is also about right, the inversion is not predicted.

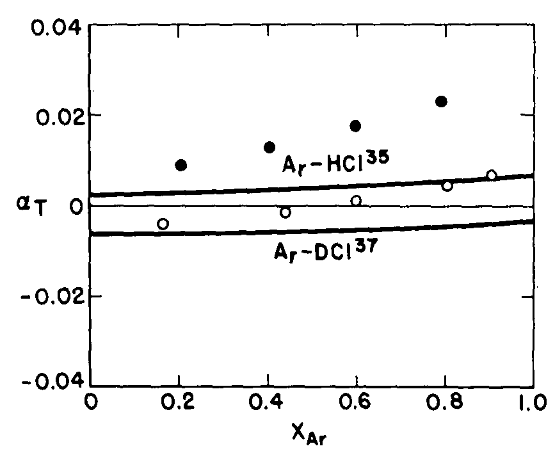

Fig. 2 THERMAL DIFFUSION FACTORS FOR ISOTOPIC ARGON-HYDROGEN CHLORIDE MIXTURES AT 380°K
Upper curve and filled circles -- Ar^{40}-HCl^{35}
Lower curve and open circles -- Ar^{40}-HCl^{37}

The reverse trend of α_T with composition noted for Ne-NH_3 also occurs with Ar-HCl.

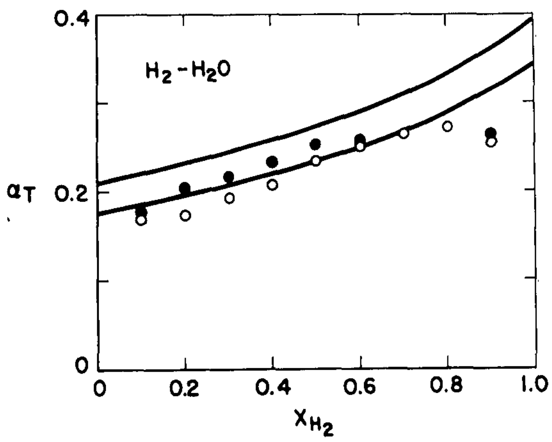

Fig. 3 THERMAL DIFFUSION FACTORS FOR H_2-H_2O MIXTURES

Upper curve and filled circles -- 455°K
Lower curve and open circles -- 368°K

Turning now to more ordinary types of systems, we find experimental results only for H_2-H_2O, H_2-SO_2, and H_2-C_2H_5Cl. The experimental data for H_2-H_2O are not too certain; the results shown in Fig. 3 for two temperatures are based on Whalley's (Ref. 26) recalculation and smoothing of the earlier (1938) measurements of Shibata and Kitagawa. The agreement with the present calculations is not too bad; an adjustment in C_{12}^* of less than 1% would suffice to produce agreement within the rather large experimental uncertainty. These results may be compared with the recent calculations of Saxena (Ref. 27), who calculated the S, Q factor from experimental viscosity and diffusion data (a procedure which should be reasonably accurate), and attempted to evaluate the $(6C_{12}^*-5)$ factor from experimental diffusion data, using Eq. (10). The use of Eq. (10) gives rise to large uncertainties, and Saxena's calculated values of α_T are larger than the results shown in Fig. 3 by about a factor of four.

The experimental results (Ref. 28) for H_2-SO_2 and H_2-C_2H_5Cl are shown in Fig. 4 together with the calculated curves. The calculated curves have the right shape but are

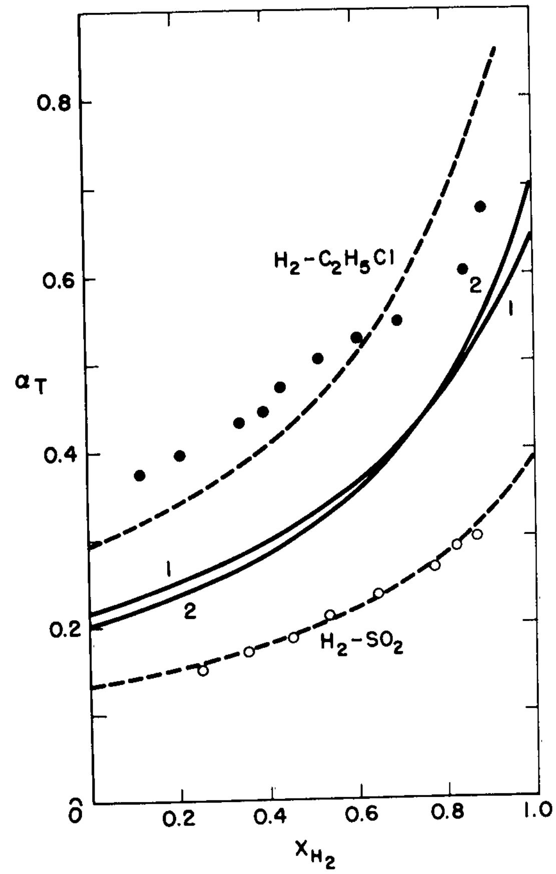

Fig. 4 THERMAL DIFFUSION FACTORS FOR H_2-SO_2 AND H_2-C_2H_5Cl MIXTURES

Curve 1 and open circles -- H_2-SO_2

Curve 2 and filled circles -- H_2-C_2H_5Cl

Dashed curves show how calculated results can be changed by a variation of less than 4% in theoretical quantity C_{12}^*.

appreciably off in magnitude. By permitting the luxury of an adjustment in C_{12}^* of 3.5% for H_2-SO_2 and 3.7% for H_2-C_2H_5Cl, one obtains agreement essentially within experimental uncertainty, as shown by the dashed curves in Fig. 4.

The fact that large variations in α_T can be produced by comparatively minor changes in C_{12}^* is essentially the basis for the familiar remark that α_T is much more sensitive to the intermolecular force laws than are the other transport coefficients. Small changes in the force laws can radically alter α_T without altering the other transport coefficients appreciably.

IV. DISCUSSION

The agreement between the present calculations and experimental measurements is encouraging. It seems probable that the present polar gas model together with the simple combination rules can describe the transport properties of mixtures with a fair measure of success. The poorest agreement occurs in the case of the thermal diffusion factor, α_T. This is not surprising since α_T is the most sensitive to the molecular model. Furthermore, recent work (Ref. 29) has indicated that the thermal conductivity of polyatomic and polar molecules depends to a noticeable degree on the interchange of translational and internal energy. Since α_T is intimately connected with the thermal conductivity in the kinetic theory of gases, it can be expected that α_T will also contain contributions from such processes. Recent calculations on the special model of a dilute suspension of large spheres in a non-uniform gas confirm this expectation (Ref. 30).

We do not believe that an attempt to refine the combination rules is justified at this time. Although such combination rules are suggested by theory, they are still semi-empirical at best, and are ultimately justified only by comparison of calculated and experimental properties. The experimental data presently available do not justify such an attempt. More experimental results of higher accuracy are necessary first. Of these, thermal diffusion would clearly be the most valuable, and ordinary diffusion the next most valuable.

Table I

Potential Parameters Used

Gas	δ_{max}	σ_o, A	ϵ_o/k, ^oK	Ref.*
H_2O	1.2	2.71	506	a
NH_3	0.7	3.15	358	a
HCl	0.34	3.36	328	a
SO_2	0.42	4.04	347	a
H_2S	0.21	3.49	343	a
CH_2Cl_2	0.2	4.52	483	a
C_2H_5Cl	0.4	4.45	423	a
C_2H_5OH	0.3	4.31	431	a
$(C_2H_5)_2O$	0.08	5.49	362	a
$(C_2H_5)_2CO$	0.06	4.50	549	a
H_2	0	2.928	37.00	b
He	0	2.556	10.22	b
Ne	0	2.789	35.7	b
Ar	0	3.418	124	b
N_2	0	3.681	91.5	b
O_2	0	3.433	113	b
Air	0	3.617	97.0	b
CO_2	0	3.996	190	b
CCl_4	0	5,881	327	b
CH_4	0	3.796	144	b
C_2H_4	0	4.232	205	b

* a Reference 1.

b MTGL, pages 1110-1112.

Table II

Viscosities of Binary Mixtures

System	T, °K	x_{polar}	$10^7\eta_{mix}$, g/cm-sec Calc.	Exptl.	% dev.	Ref.*
NH_3-H_2	293.16	0.9005	1004	1004	0.0	e
		0.7087	1045	1047	- 0.2	
		0.5177	1077	1080	- 0.3	
		0.2975	1083	1087	- 0.4	
		0.2239	1067	1072	- 0.5	
		0.1082	1006	1011	- 0.5	
	373.16	0.9005	1299	1299	0.0	e
		0.7087	1333	1333	0.0	
		0.5177	1352	1354	- 0.2	
		0.2975	1326	1329	- 0.2	
		0.2239	1294	1299	- 0.4	
		0.1082	1200	1204	- 0.3	
	473.16	0.9005	1663	1660	+ 0.2	e
		0.7087	1681	1680	+ 0.1	
		0.5177	1677	1676	+ 0.1	
		0.2975	1610	1610	0.0	
		0.2239	1558	1560	- 0.1	
		0.1082	1426	1432	- 0.4	
	523.16	0.9005	1826	1825	+ 0.1	e
		0.7087	1839	1837	+ 0.1	
		0.5177	1826	1823	+ 0.2	
		0.2975	1740	1737	+ 0.2	
		0.2239	1680	1678	+ 0.1	

*See page 31 for References.

Table II (Cont'd)

System	T, °K	x_{polar}	$10^7 \eta_{mix}$, g/cm-sec Calc.	Exptl.	% dev.	Ref.*
NH_3-N_2	293.16	0.8883	1084	1092	- 0.7	e
		0.7147	1236	1254	- 1.4	
		0.5638	1361	1383	- 1.6	
		0.2920	1564	1585	- 1.3	
		0.1111	1681	1690	- 0.5	
	373.16	0.8883	1392	1398	- 0.4	e
		0.7147	1558	1569	- 0.7	
		0.5638	1692	1710	- 1.1	
		0.2920	1904	1920	- 0.8	
		0.1111	2022	2031	- 0.4	
	473.16	0.8883	1766	1768	- 0.1	e
		0.7147	1939	1946	- 0.4	
		0.5638	2075	2085	- 0.5	
		0.2920	2286	2296	- 0.4	
		0.1111	2402	2408	- 0.3	
	523.16	0.8883	1936	1939	- 0.1	e
		0.7147	2111	2112	- 0.1	
		0.5638	2248	2250	- 0.1	
		0.2920	2457	2460	- 0.1	
		0.1111	2569	2572	- 0.1	

*See page 31 for References.

Table II (Cont'd)

System	T, °K	x_{polar}	$10^7 \eta_{mix}$, g/cm-sec Calc.	Exptl.	% dev.	Ref.*
NH_3-O_2	293.16	0.8755	1124	1143	- 1.7	e
		0.7079	1312	1350	- 2.7	
		0.4786	1561	1604	- 2.7	
		0.2986	1745	1783	- 2.1	
		0.1351	1902	1924	- 1.1	
	373.16	0.8755	1444	1459	- 1.0	e
		0.7079	1659	1689	- 1.8	
		0.4786	1938	1972	- 1.7	
		0.2986	2141	2170	- 1.3	
		0.1351	2311	2326	- 0.6	
	473.16	0.8755	1830	1840	- 0.5	e
		0.7079	2067	2085	- 0.9	
		0.4786	2368	2390	- 0.9	
		0.2986	2585	2604	- 0.7	
		0.1351	2766	2773	- 0.3	
NH_3-Air	288.66	0.900	1081	1100	- 1.7	d
		0.800	1172	1203	- 2.6	
		0.700	1261	1306	- 3.5	
		0.600	1349	1403	- 3.9	
		0.500	1434	1492	- 3.9	
		0.400	1516	1575	- 3.8	
		0.300	1595	1618	- 1.4	
		0.200	1670	1713	- 2.5	
		0.100	1742	1764	- 1.3	

*See page 31 for References.

Table II (Cont'd)

System	T, ^{o}K	x_{polar}	$10^7\eta_{mix}$, g/cm-sec Calc.	Exptl.	% dev.	Ref.*
NH_3-CH_4	287.66	0.900	997	1008	- 1.1	d
		0.800	1013	1039	- 2.5	
		0.700	1028	1061	- 3.1	
		0.600	1042	1077	- 3.3	
		0.500	1054	1091	- 3.4	
		0.400	1065	1099	- 3.1	
		0.300	1074	1105	- 2.8	
		0.200	1082	1105	- 2.1	
		0.100	1087	1099	- 1.1	

*See page 31 for References.

Table II (Cont'd)

System	T, °K	x_{polar}	$10^7\eta_{mix}$, g/cm-sec Calc.	Exptl.	% dev.	Ref.*
NH_3-C_2H_4	293.16	0.8867	995	1001	- 0.6	e
		0.8071	1002	1013	- 1.1	
		0.6961	1009	1022	- 1.3	
		0.5172	1016	1030	- 1.4	
		0.2993	1017	1027	- 0.9	
		0.1096	1012	1015	- 0.3	
	373.16	0.8867	1286	1294	- 0.6	e
		0.8071	1289	1301	- 0.9	
		0.6961	1290	1304	- 1.1	
		0.5172	1288	1303	- 1.2	
		0.2993	1279	1291	- 0.9	
		0.1096	1266	1269	- 0.2	
	473.16	0.8867	1640	1647	- 0.4	e
		0.8071	1634	1648	- 0.9	
		0.6961	1625	1639	- 1.1	
		0.5172	1606	1622	- 1.0	
		0.2993	1581	1595	- 0.9	
		0.1096	1556	1561	- 0.3	
	523.16	0.8867	1802	1809	- 0.4	e
		0.8071	1792	1805	- 0.7	
		0.6961	1778	1791	- 0.7	
		0.5172	1752	1764	- 0.7	
		0.2993	1717	1729	- 0.7	
		0.1096	1685	1689	- 0.2	

*See page 31 for References.

Table II (Cont'd)

System	T, ^oK	x_{polar}	$10^7\eta_{mix}$, g/cm-sec Calc.	Exptl.	% dev.	Ref.*
$HCl-H_2$	294.16	0.8220	1456	1461	- 0.3	b
		0.7179	1465	1469	- 0.3	
		0.5042	1467	1471	- 0.3	
		0.2031	1340	1342	- 0.2	
	327.16	0.8220	1619	1626	- 0.4	b
		0.7179	1626	1632	- 0.4	
		0.5042	1621	1625	- 0.2	
		0.2031	1462	1472	- 0.7	
	372.16	0.8220	1846	1848	- 0.1	b
		0.7179	1849	1855	- 0.3	
		0.5042	1830	1831	- 0.1	
		0.2031	1625	1629	- 0.3	
	427.16	0.8417	2103	2099	+ 0.2	b
		0.6989	2101	2104	- 0.1	
		0.5092	2067	2053	+ 0.7	
		0.2409	1871	1866	+ 0.3	
	473.16	0.8417	2307	2311	- 0.2	b
		0.6989	2299	2304	- 0.2	
		0.5092	2254	2261	- 0.3	
		0.2409	2024	2024	0.0	
	523.16	0.7947	2526	2527	0.0	b
		0.6312	2499	2507	- 0.3	
		0.5178	2458	2454	+ 0.2	
		0.2991	2275	2281	- 0.3	

*See page 31 for References.

Table II (Cont'd)

System	T, °K	x_{polar}	$10^7 \eta_{mix}$, g/cm-sec Calc.	Exptl.	% dev.	Ref.*
HCl-Air	298.66	0.900	1474	1489	- 1.0	d
		0.800	1521	1545	- 1.6	
		0.700	1566	1592	- 1.6	
		0.600	1612	1638	- 1.6	
		0.500	1654	1678	- 1.4	
		0.400	1694	1715	- 1.2	
		0.300	1731	1749	- 1.0	
		0.200	1764	1778	- 0.8	
		0.100	1794	1800	- 0.3	
$HCl-CO_2$	291.16	0.900	1445	1459	- 1.0	d
		0.800	1447	1472	- 1.7	
		0.700	1450	1483	- 2.2	
		0.600	1453	1492	- 2.7	
		0.500	1457	1499	- 2.8	
		0.400	1461	1502	- 2.7	
		0.300	1466	1503	- 2.5	
		0.200	1471	1500	- 1.9	
		0.100	1477	1495	- 1.2	

*See page 31 for References.

Table II (Cont'd)

System	T, °K	x_{polar}	$10^7 \eta_{mix}$, g/cm-sec Calc.	Exptl.	% dev.	Ref.*
SO_2-H_2	290.16	0.8215	1291	1293	- 0.2	a
		0.5075	1349	1350	- 0.1	
		0.2963	1360	1370	- 0.7	
		0.2286	1343	1344	- 0.1	
		0.1676	1306	1304	+ 0.2	
	318.16	0.8028	1422	1425	- 0.2	a
		0.5075	1474	1475	- 0.1	
		0.2963	1477	1494	- 1.1	
		0.2286	1454	1453	+ 0.1	
		0.1676	1410	1410	0.0	
	343.16	0.8028	1534	1535	- 0.1	a
		0.6999	1553	1557	- 0.3	
		0.6175	1568	1574	- 0.4	
		0.4823	1586	1587	- 0.1	
		0.2963	1578	1596	- 1.1	
		0.2306	1551	1551	0.0	
		0.1676	1499	1500	- 0.1	
		0.1657	1497	1505	- 0.5	
	365.16	0.8028	1635	1633	+ 0.1	a
		0.6999	1653	1648	+ 0.3	
		0.6175	1667	1675	- 0.5	
		0.4823	1683	1682	+ 0.1	
		0.2306	1636	1640	- 0.2	
		0.1676	1578	1573	+ 0.3	
		0.1657	1575	1577	- 0.1	

*See page 31 for References.

Table II (Cont'd)

System	T, °K	x_{polar}	$10^7 \eta_{mix}$, g/cm-sec Calc.	Exptl.	% dev.	Ref.*
SO_2-H_2 (Cont'd)	397.16	0.6760	1796	1797	- 0.1	a
		0.4698	1818	1814	+ 0.2	
		0.3265	1804	1801	+ 0.2	
		0.1636	1684	1685	- 0.1	
	432.16	0.6760	1950	1942	+ 0.4	a
		0.4698	1967	1960	+ 0.4	
		0.3265	1944	1942	+ 0.1	
		0.1676	1809	1803	+ 0.3	
		0.1512	1781	1748	+ 1.9	
	472.16	0.6760	2120	2118	+ 0.1	a
		0.4905	2131	2121	+ 0.5	
		0.3265	2096	2098	- 0.1	
		0.1512	1906	1953	- 2.4	
SO_2-CO_2	288.96	0.900	1279	1288	- 0.7	d
		0.800	1299	1316	- 1.3	
		0.700	1319	1338	- 1.4	
		0.600	1339	1363	- 1.8	
		0.500	1361	1384	- 1.7	
		0.400	1383	1407	- 1.7	
		0.300	1405	1429	- 1.7	
		0.200	1429	1447	- 1.2	
		0.100	1452	1464	- 0.8	

*See page 31 for References.

Table II (Cont'd)

System	T, ^oK	x_{polar}	$10^7 \eta_{mix}$, g/cm-sec Calc.	Exptl.	% dev.	Ref.*
H_2S-Air	290.36	0.900	1313	1331	- 1.4	d
		0.800	1367	1403	- 2.6	
		0.700	1422	1469	- 3.2	
		0.600	1478	1535	- 3.7	
		0.500	1535	1603	- 4.2	
		0.400	1593	1655	- 3.8	
		0.300	1651	1709	- 3.4	
		0.200	1710	1755	- 2.6	
		0.100	1768	1795	- 1.5	
CH_2Cl_2-CCl_4	293.15	0.8425	1011	1021	- 1.0	f
		0.6985	1001	1016	- 1.5	
		0.5014	992	1013	- 2.1	
		0.3114	986	1000	- 1.4	
		0.1484	983	991	- 0.8	
	353.26	0.6739	1191	1212	- 1.7	f
		0.3649	1178	1192	- 1.2	
	413.43	0.8485	1415	1425	- 0.7	f
		0.7118	1406	1403	+ 0.2	
		0.5262	1393	1411	- 1.3	
		0.2904	1379	1382	- 0.3	
		0.1261	1369	1368	+ 0.1	

*See page 31 for References.

Table II (Cont'd)

System	T, ^oK	x_{polar}	$10^7\eta_{mix}$, g/cm-sec Calc.	Exptl.	% dev.	Ref.*
$(C_2H_5)_2O-H_2$	288.16	0.2650	930	900	+ 3.3	c
		0.1330	976	937	+ 4.2	
	373.16	0.2650	1164	1119	+ 4.0	c
		0.1330	1198	1146	+ 4.5	
	423.16	0.2650	1295	1252	+ 3.4	c
		0.1330	1324	1262	+ 4.9	
	486.16	0.2650	1452	1391	+ 4.4	c
		0.1330	1471	1403	+ 4.9	

*See page 31 for References.

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Table III

Diffusion Coefficients of Binary Mixtures at One Atmosphere

System	Temp.	D_{12}(1 atm), cm^2/sec Calc.	Exptl.	% dev.	Ref.*
H_2O-H_2	19.95 °C	0.830	0.850	- 2.3	c
	49.5	0.989	1.012	- 2.3	
	92.4	1.239	1.236	+ 0.2	
			1.256	- 1.4	
	99.34	1.281	1.282	- 0.1	
	34.1 °C	0.905	1.020	-11.3	e
	55.4	1.022	1.121	- 8.8	
	79.5	1.161	1.200	- 3.3	
	34.0 °C	0.904	0.915	- 1.2	h
	55.5	1.023	0.961	+ 6.5	
H_2O-He	34.0 °C	0.916	0.902	+ 1.6	e
	55.3	1.027	1.011	+ 1.6	
	79.3	1.160	1.121	+ 3.5	
	25.0 °C	0.870	0.908	- 4.2	f
H_2O-N_2	34.4 °C	0.242	0.256	- 5.5	e
	55.4	0.275	0.303	- 9.2	
	79.0	0.314	0.359	-12.5	
	55.8 °C	0.276	0.313	-11.8	h
	76.0	0.309	0.354	-12.7	

*See page 37 for References.

Table III (Cont'd)

System	Temp.		D_{12}(1 atm), cm^2/sec Calc.	Exptl.	% dev.	Ref.*
H_2O-O_2	34.9	°C	0.245	0.282	-13.1	e
	55.8		0.278	0.318	-12.6	
	79.2		0.318	0.352	- 9.7	
	400	°K	0.405	0.48	-14.6	j
	500		0.612	0.68	-10.3	
	700		1.125	1.20	- 6.3	
	900		1.748	1.84	- 5.0	
	1050		2.280	2.39	- 4.6	
H_2O-Air	42	°C	0.254	0.288	-11.8	b
	16.7	°C	0.217	0.244	-11.1	c
	92.4		0.338	0.357	- 5.3	
				0.360	- 6.1	
	99.3		0.350	0.377	- 7.2	
	25	°C	0.229	0.260	-11.9	f
H_2O-CO_2	22.95	°C	0.160	0.164	- 2.4	c
	92.4		0.243	0.248	- 2.1	
	99.38		0.252	0.259	- 2.7	
	34.3	°C	0.172	0.202	-14.9	e
	55.4		0.197	0.211	- 6.6	
	79.2		0.226	0.245	- 7.8	
	55.5	°C	0.197	0.198	- 0.5	h

*See page 37 for References.

Table III (Cont'd)

System	Temp.		D_{12}(1 atm), cm^2/sec Calc.	Exptl.	% dev.	Ref.*
H_2O-CH_4	34.5	°C	0.241	0.292	-17.5	e
	55.6		0.274	0.331	-17.2	
	79.1		0.314	0.356	-11.8	
$H_2O-C_2H_4$	34.6	°C	0.171	0.204	-16.2	e
	55.3		0.194	0.233	-16.7	
	79.4		0.223	0.247	- 9.7	
NH_3-H_2	25	°C	0.780	0.783	- 0.4	g
	55		0.926	0.943	- 1.8	
	85		1.081	1.093	- 1.1	
	263	°K	0.622	0.57	+ 9.1	i
	328		0.925	0.90	+ 2.8	
	395		1.284	1.35	- 4.9	
	473		1.755	1.86	- 5.6	
NH_3-N_2	25	°C	0.218	0.230	- 5.2	g
	55		0.261	0.285	- 8.4	
	85		0.308	0.328	- 6.1	
SO_2-H_2	12.4	°C	0.530	0.525	+ 1.0	a
	263	°K	0.457	0.43	+ 6.3	i
	323		0.661	0.61	+ 8.4	
	473		1.287	1.23	+ 4.6	

*See page 37 for References.

Table III (Cont'd)

System	Temp.		D_{12}(1 atm), cm^2/sec Calc.	Exptl.	% dev.	Ref.*
SO_2-Ar	263	°K	0.089	0.077	+15.6	i
SO_2-N_2	263	°K	0.0999	0.104	- 3.9	i
SO_2-CO_2	263	°K	0.0675	0.064	+ 5.5	i
	343		0.1137	0.108	+ 5.3	
	473		0.209	0.195	+ 7.2	
C_2H_5OH-H_2	67	°C	0.655	0.586	+11.8	c
C_2H_5OH-He	25	°C	0.491	0.494	- 0.6	f
C_2H_5OH-Air	42	°C	0.132	0.145	- 9.0	b
	67		0.153	0.154	- 0.7	
	67	°C	0.153	0.153	0.0	c
	25	°C	0.119	0.135	-11.9	f
C_2H_5OH-CO_2	67	°C	0.107	0.106	+ 1.0	c
$(C_2H_5)_2O$-H_2	19.9	°C	0.377	0.354	+ 6.5	c

*See page 37 for References.

Table III (Cont'd)

System	Temp.	D_{12}(1 atm), cm^2/sec Calc.	Exptl.	% dev.	Ref.*
$(C_2H_5)_2O$-Air	14.8 °C	0.0809	0.0863	- 6.3	c
	19.9	0.0836	0.0896	- 6.7	
$(C_2H_5)_2O$-CO_2	19.9 °C	0.0578	0.0632	- 8.5	c
$(CH_3)_2CO$-H_2	23 °C	0.464	0.424	+ 9.4	d
H_2-N_2	25 °C	0.762	0.784	- 2.8	g
	55	0.896	0.908	- 1.3	
	85	1.040	1.052	- 1.1	
CCl_4-H_2	23 °C	0.354	0.345	+ 2.6	c
CCl_4-O_2	23 °C	0.0703	0.0749	- 6.1	d

*See page 37 for References.

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