

A SIMPLE FORMULA FOR MULTICOMPONENT GASEOUS DIFFUSION COEFFICIENTS DERIVED FROM MEAN FREE PATH THEORY

E. OBERMEIER and A. SCHABER

Lehrstuhl für Thermodynamik, Gesamthochschule Siegen, Germany

(Received 8 November 1976)

Abstract—A straight forward relation for multicomponent gaseous diffusion coefficients is derived by application of the elementary mean free path theory. The relevant parameters in this relation are obtained by comparison with equations resulting from the kinetic theory of monatomic gases. As a result of this procedure the multicomponent diffusion coefficients are represented explicitly as a function of the binary and self-diffusion coefficients. The accuracy of the new equation is very satisfactory. This is demonstrated by a number of examples.

NOMENCLATURE

D_{ik} ,	multicomponent diffusion coefficient (kinetic theory);
\mathcal{L}_{ii} ,	self-diffusion coefficient;
\mathcal{L}_{ik} ,	binary diffusion coefficient;
D_{ik}^{FP} ,	multicomponent diffusion coefficient (mean free path theory);
$j_{i\beta}$,	diffusional mass flux vector;
k ,	Boltzmann-constant;
$l_{i\beta}$,	mean free path for number density transfer of component "i";
$M_{i\beta}$,	molecular weight of component "i";
\bar{M} ,	molecular weight of gas mixture;
$m_{i\beta}$,	mass of a molecule of component "i";
N ,	number of components;
$u_{i\beta}$,	mean thermal speed of molecules of component "i";
$v_{i\beta}$,	mean total velocity of molecules of component "i";
\bar{v} ,	mass average velocity;
$x_{i\beta}$,	mole fraction of component "i";
$\rho_{i\beta}$,	partial density of component "i";
ρ ,	total density.

1. INTRODUCTION

CONCENTRATION diffusion plays an important role in many technical problems. Typical examples are ablation cooling or combustion processes.

The multicomponent gaseous diffusion coefficients are usually calculated by equations derived from the kinetic theory of monatomic gases [1]. Especially for mixtures with many components the numerical evaluation of these equations is extremely tedious and often not feasible for practical problems. For this reason various authors have derived approximations based on the equations of the kinetic theory [2-5]. These investigations, except those by Schaber *et al.* [4], did not lead to explicit relations for multicomponent diffusion coefficients. Another means of obtaining expressions for the diffusion coefficients or diffusional

mass fluxes is the application of the so called mean free path theory, e.g. [6-8]. The mean free path theory, on the other hand, has certain decisive disadvantages. Depending on the relation adopted for calculating the mean free paths the equations are either simple and extremely inaccurate or nearly as complicated as those derived from the exact kinetic theory [6-8].

For this reason the mean free path theory was never seriously adopted for practical problems involving transport coefficients. The present investigation reveals, that by comparison of relations following from the mean free path theory with those of the "exact" kinetic theory, relatively simple and very accurate approximation equations may be derived for the multicomponent coefficients.

2. BASIC EQUATIONS

Neglecting pressure and thermal diffusion, the following equations for the diffusional mass fluxes j_i of components "i" result from the kinetic theory of monatomic gases in the 1. Chapman-Enskog approximation [1].

$$j_i = \rho \sum_{\substack{k=1 \\ k \neq i}}^N \frac{M_i M_k}{\bar{M}^2} D_{ik} \text{grad } x_k \quad (2.1)$$

$$\frac{\rho}{\bar{M}} \text{grad } x_i = \sum_{\substack{k=1 \\ k \neq i}}^N \frac{1}{\mathcal{L}_{ik}} \left[x_i \frac{j_k}{M_k} - x_k \frac{j_i}{M_i} \right] \quad (2.2)$$

$$\sum_{i=1}^N j_i = 0. \quad (2.3)$$

The diffusional mass flux j_i of component "i" in equations (2.1)-(2.3) is defined by

$$j_i = \rho_i(v_i - \bar{v}) \quad (2.4)$$

with

$$\bar{v} = \frac{1}{\rho} \sum_{i=1}^N \rho_i v_i \quad (2.5)$$

v_i is the mean total velocity of component i and \bar{v} the

mass average velocity. In equations (2.1)–(2.5) M_i is the molecular weight, x_i the mole fraction and ρ_i the partial density of component "i". \bar{M} is the mean molecular weight and ρ the bulk density of the mixture.

Equation (2.1), which represents the diffusional mass flux j_i explicitly as a function of the concentration gradients $\text{grad } x_i$, contains the multicomponent diffusion coefficients D_{ik} .

These are complex functions of temperature and concentrations. In the first Chapman–Enskog approximation the concentration dependence disappears only for 2-component mixtures.

Equations (2.2), however, usually called Stefan–Maxwell equations, yield the gradients $\text{grad } x_i$ explicitly as function of the implicitly occurring diffusional mass fluxes j_i . The advantage of equations (2.2) is, that only the binary diffusion coefficients \mathcal{G}_{ik} are needed. These may easily be calculated by the well known equations of the kinetic theory [1].

From the elementary mean free path theory [8] one obtains the following relation for the difference of the mean total velocities of the components i and k :

$$v_i - v_k = -\frac{1}{3} [u_i l_i \text{grad} (\ln x_i) - u_k l_k \text{grad} (\ln x_k)] \quad k = 1, 2, \dots, N. \quad (2.6)$$

In equations (2.6) u_i is the mean thermal speed and l_i the mean free path for number density transfer of the molecules of component "i".

Multiplication of equation (2.6) with the partial density ρ_k and a summation over all indices k yields, if one takes account of equation (2.5)

$$\rho(v_i - \bar{v}) = -\frac{1}{3} \rho u_i l_i \text{grad} (\ln x_i) + \frac{1}{3} \sum_{k=1}^N \rho_k u_k l_k \text{grad} (\ln x_k). \quad (2.7)$$

By introducing the relations $\rho_k/x_k = \rho M_k/\bar{M}$ and $\sum_i \text{grad } x_i = 0$ the following equation is obtained:

$$j_i = \frac{\rho}{3\bar{M}} \sum_{k=1}^N \left[\frac{\rho_i}{\rho} M_k u_k l_k + M_i u_i l_i \left(1 - \frac{\rho_i}{\rho} \right) \right] \text{grad } x_k. \quad (2.8)$$

By comparison of the two equations (2.8) and (2.1) the following multicomponent diffusion coefficient D_{ik}^{FP} based on mean free path theory may be defined:

$$D_{ik}^{FP} = \frac{1}{3} \left[x_i u_k l_k + \frac{1}{M_k} (\bar{M} - M_i x_i) u_i l_i \right]. \quad (2.9)$$

The form of equation (2.9) is extremely simple; on the other hand it contains the unknown parameters u_i and l_i . If one calculates u_i and l_i simply by the mean free path theory, the results obtained are extremely unsatisfactory. Even for binary diffusion coefficients equation (2.9) predicts a completely wrong dependence of D_{ik} on concentration, e.g. [8].

3. DERIVATION OF THE APPROXIMATION EQUATIONS

The unknown values $u_i l_i$ in equations (2.8) and (2.9) can be eliminated by comparison of these equations with the relations following from the kinetic theory.

From equation (2.2) one obtains

$$\lim_{x_i \rightarrow 0} j_i = - \frac{M_i}{N} \frac{\rho}{\bar{M}} \text{grad } x_i \quad (3.1)$$

with

$$M' = \lim_{x_i \rightarrow 0} \bar{M}. \quad (3.2)$$

By considering the relation $\sum_i \text{grad } x_i = 0$ a comparison of equations (2.1) and (3.1) yields

$$\lim_{x_i \rightarrow 0} D_{ik} = \frac{\bar{M}' M_k}{\sum_{j=1}^N \frac{x_j}{\mathcal{G}_{ij}}}. \quad (3.3)$$

On the other hand it follows from equation (2.9) that

$$\lim_{x_i \rightarrow 0} D_{ik}^{FP} = \frac{1}{3} \frac{\bar{M}'}{M_k} \lim_{x_i \rightarrow 0} u_i l_i. \quad (3.4)$$

We now postulate, that the coefficients D_{ik} and D_{ik}^{FP} are identical. A comparison of equations (3.3) and (3.4) then yields

$$\lim_{x_i \rightarrow 0} u_i l_i = \frac{3}{\sum_{j=1}^N \frac{x_j}{\mathcal{G}_{ij}}}. \quad (3.5)$$

For the limiting case $x_i \rightarrow 0$ the term $u_i l_i$ in equation (3.5) is expressed by relations of the kinetic theory, i.e. by the binary diffusion coefficients. No further information may be gained by calculating the other limiting case $\lim_{x_i \rightarrow 1} u_i l_i$ from kinetic theory.

On the other hand, the mean free path theory yields a relation between $u_i l_i$ and the self-diffusion coefficient \mathcal{G}_{ii} .

$$\lim_{x_i \rightarrow 1} u_i l_i = 3\mathcal{G}_{ii}. \quad (3.6)$$

In accordance with equations (3.5) and (3.6) the following empirical equation for $u_i l_i$ is proposed:

$$u_i l_i \equiv \frac{3}{\frac{x_i}{\mathcal{G}_{ii}} f_i + \sum_{j=1}^N \frac{x_j}{\mathcal{G}_{ij}}}. \quad (3.7)$$

The occurrence of the self-diffusion coefficient \mathcal{G}_{ii} in equation (3.7) is justified by the mean free path theory but is contradictory to the equations of the exact kinetic theory.

For this reason the factor f_i in equation (3.7) was introduced in order to allow an adaption to the relations of the kinetic theory.

Equation (3.7) satisfies the limiting case of $x_i \rightarrow 0$ in equation (3.5) and of $x_i \rightarrow 1$ in equation (3.6). By introducing equation (3.7) into equation (2.9) one obtains

$$D_{ik}^{FP} = \frac{x_i}{\frac{x_k}{\mathcal{G}_{kk}} f_k + \sum_{j \neq k} \frac{x_j}{\mathcal{G}_{kj}}} + \frac{1}{M_k} \frac{(\bar{M} - M_i x_i)}{\frac{x_i}{\mathcal{G}_{ii}} f_i + \sum_{j \neq i} \frac{x_j}{\mathcal{G}_{ij}}}. \quad (3.8)$$

The factors f_i must now be determined. One obtains for a binary mixture:

$$D_{ik}^{FP} = \frac{x_i}{\frac{x_k}{\mathcal{D}_{kk}} f_k + \frac{x_i}{\mathcal{D}_{ik}}} + \frac{x_k}{\frac{x_i}{\mathcal{D}_{ii}} f_i + \frac{x_k}{\mathcal{D}_{ik}}} \quad (3.9)$$

As is well known, the binary diffusion coefficient of the first Chapman–Enskog approximation is independent of concentration. If one postulates, that $D_{ik}^{FP} = D_{ik}$, then equation (3.9) at once results in the condition:

$$f_i f_k = \frac{\mathcal{D}_{ii} \mathcal{D}_{kk}}{\mathcal{D}_{ik}^2} \quad (3.10)$$

For a binary mixture, therefore, the factors f_i and f_k have to satisfy the condition (3.10) in order to gain independence of concentration for the binary diffusion coefficient.

For a mixture of three components i, k, l it follows from equation (3.7) that

$$D_{ik}^{FP} = \frac{x_i}{\frac{x_k}{\mathcal{D}_{kk}} f_k + \frac{x_i}{\mathcal{D}_{ki}} + \frac{x_l}{\mathcal{D}_{kl}}} + \frac{1}{M_k} (\bar{M} - M_i x_i) + \frac{\frac{x_i}{\mathcal{D}_{ii}} f_i + \frac{x_k}{\mathcal{D}_{ik}} + \frac{x_l}{\mathcal{D}_{il}}}{\frac{x_i}{\mathcal{D}_{ii}} f_i + \frac{x_k}{\mathcal{D}_{ik}} + \frac{x_l}{\mathcal{D}_{il}}} \quad (3.11)$$

The equation for the same 3-component mixture, if derived by the kinetic theory, has the same structure as equation (3.11). By transformation of the equations presented by Hirschfelder *et al.* [1] the following relation is obtained:

$$D_{ik} = \frac{x_i}{\frac{x_k}{\mathcal{D}_{ik}} \mathcal{D}_{kl} + \frac{x_i}{\mathcal{D}_{ki}} + \frac{x_l}{\mathcal{D}_{kl}}} + \frac{1}{M_k} (\bar{M} - M_i x_i) + \frac{\frac{x_i}{\mathcal{D}_{ik}} \mathcal{D}_{kl} + \frac{x_k}{\mathcal{D}_{ik}} + \frac{x_l}{\mathcal{D}_{il}}}{\frac{x_i}{\mathcal{D}_{ik}} \mathcal{D}_{kl} + \frac{x_k}{\mathcal{D}_{ik}} + \frac{x_l}{\mathcal{D}_{il}}} \quad (3.12)$$

The similarity in structure of equations (3.11) and (3.12) is quite surprising. The two equations are identical, if the following conditions are fulfilled:

$$f_i = \frac{\mathcal{D}_{ii} \mathcal{D}_{kl}}{\mathcal{D}_{ik} \mathcal{D}_{il}} \quad f_k = \frac{\mathcal{D}_{kk} \mathcal{D}_{il}}{\mathcal{D}_{ik} \mathcal{D}_{kl}} \quad (3.13)$$

From equations (3.13) one again obtains the condition (3.10). Equation (3.10) is therefore a necessary condition for the diffusion coefficients D_{ik}^{FP} of binary and ternary mixtures to be identical with the coefficients D_{ik} defined by the kinetic theory.

Equation (3.10) is now assumed to be generally valid for a definition of the factors f_i and f_k , although this cannot be rigidly confirmed for mixtures with more than three components.

On the other hand such relations as equations (3.13) cannot be generalised for arbitrary multicomponent

mixtures, since the terms f_i and f_k are not solely dependent of the properties of components i and k .

For this reason f_i and f_k were approximated by empirical relations. Numerical investigations of mixtures with 5–17 components showed, that the best results were obtained with

$$f_i = f_k = \frac{(\mathcal{D}_{ii} \mathcal{D}_{kk})^{1/2}}{\mathcal{D}_{ik}} \quad (3.14)$$

Equation (3.10) is satisfied by equation (3.14). Substitution of equation (3.14) in equation (3.8) yields the final approximation formula for the multicomponent gaseous diffusion coefficients:

$$D_{ik}^{FP} = \frac{x_i}{\frac{x_k}{\mathcal{D}_{ik}} \left(\frac{\mathcal{D}_{ii}}{\mathcal{D}_{kk}} \right)^{1/2}} + \sum_{j \neq k} \frac{x_j}{\mathcal{D}_{kj}} + \frac{(\bar{M} - M_i x_i)/M_k}{\frac{x_i}{\mathcal{D}_{ik}} \left(\frac{\mathcal{D}_{kk}}{\mathcal{D}_{ii}} \right)^{1/2} + \sum_{j \neq i} \frac{x_j}{\mathcal{D}_{ij}}} \quad (3.15)$$

Equation (3.15) satisfies the following conditions:

(i) The limiting values of D_{ik}^{FP} for $x_i \rightarrow 0$ and $x_i \rightarrow 1$ are identical with the corresponding values D_{ik} derived by the kinetic theory. From equations (3.3) and (3.15) follows:

$$\lim_{x_i \rightarrow 0} D_{ik}^{FP} = \lim_{x_i \rightarrow 0} D_{ik} \quad (3.16)$$

Furthermore equation (3.15) yields

$$\lim_{x_i \rightarrow 1} D_{ik}^{FP} = \mathcal{D}_{ik} \quad (3.17)$$

It can be shown, that this is also in accordance with the kinetic theory.

(ii) The equation is exact for a binary mixture.

(iii) The equation is exact for a ternary mixture, if the conditions

$$\mathcal{D}_{il} = (\mathcal{D}_{ii} \mathcal{D}_{il})^{1/2} \quad \mathcal{D}_{kl} = (\mathcal{D}_{kk} \mathcal{D}_{il})^{1/2} \quad (3.18)$$

are fulfilled, i.e. the binary diffusion coefficients must be equal to the geometric mean of the corresponding self-diffusion coefficients.

This also makes it understandable physically, why the self-diffusion coefficients appear in equation (3.15). The ratios of the self-diffusion coefficients in equation (3.15) may be regarded as approximations for the ratios of binary diffusion coefficients.

In the following chapter the general applicability and accuracy of equation (3.15) for multicomponent mixtures is demonstrated by some specific examples.

4. EXAMPLES

To test the validity of equation (3.15) the multicomponent diffusion coefficients were calculated for various gas mixtures. The diffusion coefficients obtained by equation (3.15) were compared with the results computed by the "exact" kinetic theory. The binary- and self-diffusion coefficients were calculated by the well known relations of the kinetic theory [1]. For the mixture of dissociated air the potential parameters for a Lennard–Jones 6–12 potential by Schaber *et al.* [4]

were adopted. For the other mixtures these values were taken from Svehla [9].

To begin with, let us examine the results for dissociated air with the five components N_2 , O_2 , NO , N and O . Figure 1 shows multicomponent diffusion coefficients, which were calculated as a function of temperature, once again with equation (3.15) and once by the exact kinetic theory. The corresponding concentrations are the equilibrium values at a pressure of $p = 1$ bar. In Fig. 1 only those six diffusion coefficients out of twenty are presented, for which the largest differences between approximate and exact theory occur.

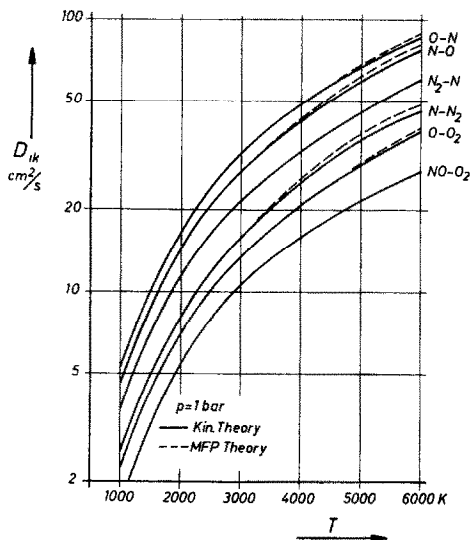


FIG. 1. Diffusion coefficients in the mixture of dissociated air.

The dashed curves show the results of equation (3.15) and the solid curves represent "exact" values. As can be seen from Fig. 1 the agreement between the approximate and exact theory is very good. For the other fourteen values of diffusion coefficients no significant differences occur. The mean relative error for this mixture is less than 3%. Here one has to bear in the mind, that the potential parameters, which are employed in both theories to calculate the binary diffusion coefficients, are certainly less accurate, especially at high temperatures [4].

As a further example a mixture of hydrogen and oxygen with the six components H , H_2 , O , OH , H_2O and O_2 was investigated. This mixture may be regarded as representing the oxygen-hydrogen combustion. This mixture is particularly suited to test the utility of equation (3.15), since the components have extremely different molecular weights. This implies, that also the diffusion coefficients differ largely.

Figure 2 shows those multicomponent diffusion coefficients of this mixture as a function of the hydrogen concentration for which the largest differences occur. The temperature is $T = 1500$ K and the pressure $p = 1$ bar. In order to demonstrate the concentration dependence of the diffusion coefficients the concentration of hydrogen was varied between $0 < x < 1$ whereas the concentrations of the other com-

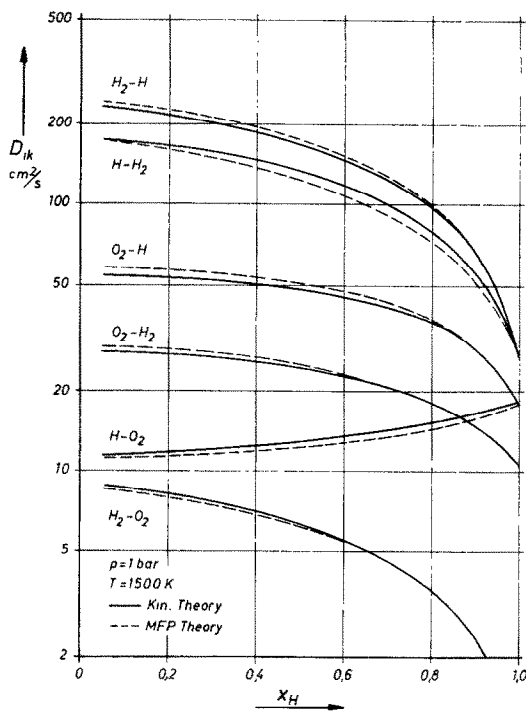


FIG. 2. Diffusion coefficients in a mixture of hydrogen and oxygen.

ponents were taken to be equal, i.e. $x_k = (1 - x_H)/5$. As may be seen in Fig. 2, equation (3.15) permits the calculation of multicomponent diffusion coefficients with excellent accuracy even for mixtures with components of very different molecular weights.

Figure 3 shows a number of diffusion coefficients for the same mixture; in this case, however, the concentration of oxygen, the component with the largest molecular weight, was varied between $0 < x_{O_2} < 1$.

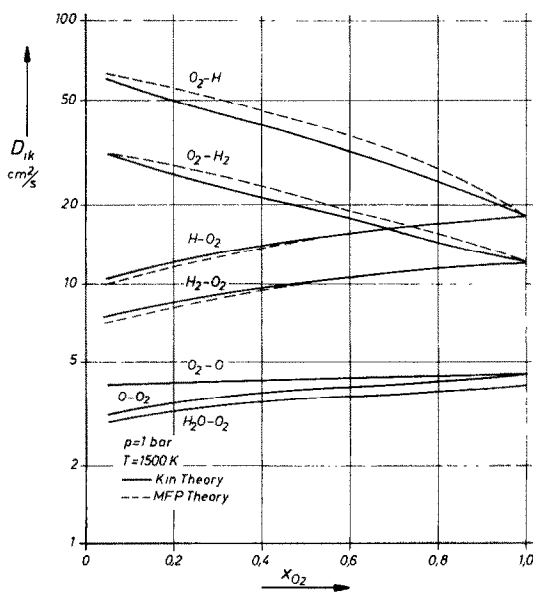


FIG. 3. Diffusion coefficients in a mixture of hydrogen and oxygen.

In addition to the above two examples various other mixtures were investigated. These mixtures and the observed mean relative deviations of the coefficients D_{ik}^{FP} from those of the kinetic theory are listed in Table 1. The temperatures were varied between $1000 < T < 7000$ K. The concentrations of the components with the smallest and largest molecular weight were systematically varied as described above for the H-O mixture.

Table 1

Mixture	Number of components	Mean relative error (%)
N ₂ , O ₂ , NO, N, O	5	<3
H, H ₂ , O, OH, H ₂ O, O ₂	6	<3
C, N, O, CN, CO, C ₂ , N ₂ , O ₂ , CO ₂	9	<3
H, H ₂ , CH ₃ , CH ₄ , O, OH, H ₂ O, CO, N ₂ , NO, O ₂ , H ₂ O ₂ , N ₂ O, CO ₂	14	<1.5
H, H ₂ , C, N, O, OH, H ₂ O, CH ₄ , N ₂ , O ₂ , CN, CO, NO, C ₂ , C ₂ H ₂ , HCN, CO ₂	17	<1

As one can see in Table 1, the approximation equation (3.15) yields excellent results. Only in exceptional cases greater differences between D_{ik}^{FP} and D_{ik} can occur. These are for a mixture of two excess- and some trace components, if the diffusion coefficient of an excess-component with extremely large molecular weight into a trace-component with extremely small molecular weight is computed. The reason is, that the numerator of the right hand side of equation (3.15) contains the ratio of the molecular weights of the heavy and the light component and therefore a small error in the denominator on the right hand side may be increased strongly by this ratio.

This deviation, however, has no effect on the resulting diffusional mass flux j_i of component i since for its

calculation D_{ik} is multiplied by the extremely small concentration gradient $\text{grad } x_k$ of the trace component k and therefore becomes negligible in comparison with the other terms.

5. SUMMARY

The approximation equation (3.15) permits a simple and accurate calculation of multicomponent gaseous diffusion coefficients. The dominant advantages are the simple and explicit representation as compared with the computational complexity of the exact theory.

The greatest accuracy is achieved if either the molecular weights of the components do not differ strongly or if the number of components is large. However, the accuracy is very satisfactory for mixtures with components of extremely different molecular weights. This was demonstrated for various mixtures with up to 17 components.

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UNE FORMULE SIMPLE, DERIVEE DE LA THEORIE DU LIBRE PARCOURS MOYEN, POUR LES COEFFICIENTS DE DIFFUSION DANS LES GAS A PLUSIEURS COMPOSANTS

Résumé—Par application de la théorie élémentaire du libre parcours moyen, on dérive une relation directe pour les coefficients de diffusion dans les gaz à plusieurs composants.

Les paramètres intervenant dans cette relation sont obtenus par comparaison avec des équations qui résultent de la théorie des gaz monoatomiques. Il résulte de cette procédure que les coefficients de diffusion sont représentés explicitement comme fonctions des coefficients binaires et d'autodiffusion. La précision de la nouvelle formule est très satisfaisante. Ceci est démontré par de nombreux exemples.

BEZIEHUNG ZUR BERECHNUNG DER POLYNÄREN GASDIFFUSIONSKOEFFIZIENTEN

Zusammenfassung—Aus der elementaren Weglängentheorie wird eine einfach strukturierte Beziehung zur Berechnung der polynären Gasdiffusionskoeffizienten hergeleitet. Die in dieser Gleichung auftretenden unbekannt Parameter werden durch einen Vergleich mit den aus der kinetischen Theorie einatomiger Gase folgenden Beziehungen bestimmt. Auf diese Weise können die polynären Diffusionskoeffizienten explizit als Funktion der binären Diffusionskoeffizienten und der Selbstdiffusionskoeffizienten dargestellt werden. Die Genauigkeit dieser Beziehung ist sehr gut. Dies wird anhand einiger Beispiele demonstriert.

ПРОСТАЯ ФОРМУЛА ДЛЯ КОЭФФИЦИЕНТОВ МНОГОКОМПОНЕНТНОЙ
ДИФФУЗИИ ГАЗА, ПОЛУЧЕННАЯ С ПОМОЩЬЮ ТЕОРИИ СРЕДНЕЙ
ДЛИНЫ СВОБОДНОГО ПРОБЕГА

Аннотация — Выведено простое соотношение для коэффициентов диффузии многокомпонентной смеси газов с помощью элементарной теории средней длины свободного пробега. Параметры в этом соотношении получены посредством сравнения с уравнениями кинетической теории для одноатомных газов. Коэффициенты диффузии многокомпонентной смеси представлены в явной форме как функция бинарных коэффициентов диффузии и коэффициентов самодиффузии. Точность нового уравнения весьма удовлетворительна, что подтверждается целым рядом сопоставлений.