

band branch, line branch, terms in the expansion of the Planck function in a Taylor series, and approximation formula for  $G_{ik}$ , respectively;  $h$ ,  $c$  stand for quantities pertaining to hot and cold layers; ' refers to upper level; " refers to lower level;  $D$ , Doppler profile;  $L$ , Lorentz profile.

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#### APPROXIMATE CALCULATION OF TRANSPORT COEFFICIENTS IN MULTI-COMPONENT MIXTURES

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UDC 533.1/2

An efficient method is developed for calculating transport coefficients in dissociated gas mixtures with a large number of components.

For solution of problems of heat and mass exchange in the presence of hypersonic streamline flow around obstacles [1], data on the various transport coefficients in mixtures of reacting gases is necessary. Application of the expressions for the coefficients of viscosity, thermal conductivity, thermal diffusion, and multicomponent mass diffusion derived from the kinetic theory of gases [2-4] can often be unwieldy because of the large amount of computer time used in the calculation. Significantly simpler expressions can be obtained with the help of the bifurcation approximation to the binary diffusion coefficient [5-7]. This approximation is used in the present paper to obtain a relation for the diffusion current and expressions for the coefficients of viscosity, thermal conductivity, and thermal diffusion.

Specifically, calculations were done for stably dissociated air at temperatures ranging from 2000 to 8000°K and pressures between  $0.1 \cdot 10^5$  and  $10^5$  Pa. Comparison of the results of our calculations for the transport coefficients with those of numerical calculations based on the kinetic theory of gases [2-3] indicates acceptable accuracy for our method.

1. The Chapman-Enskog method for a quasineutral mixture gives the following expressions for the mass diffusion current, heat current and viscous stress tensor [4]:

$$\mathbf{j}_i = \rho_i \mathbf{V}_i = \frac{n^2}{\rho} \sum_{k=1}^N m_i m_k D_{ik} \mathbf{d}_k - D_i^T \nabla \ln T,$$

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$$\mathbf{q} = \sum_{k=1}^N h_k \mathbf{j}_k - \lambda' \nabla T - \frac{p}{\rho} \sum_{k=1}^N \frac{D_k^T}{\alpha_k} \mathbf{d}_k, \quad (1)$$

$$\tau_{ij} = -\frac{2}{3} \eta \operatorname{div} \mathbf{V} \cdot \delta_{ij} + \eta \operatorname{def} \mathbf{V},$$

where

$$\mathbf{d}_i = \nabla x_i + (x_i - \alpha_i) \nabla \ln p; \quad p = \rho \frac{R}{M} T; \quad n = \sum_{k=1}^N n_k; \quad (2)$$

$$x_i = \frac{n_i}{n}; \quad \alpha_i = \frac{\rho_i}{\rho}; \quad \rho_i = n_i m_i; \quad \rho = \sum_{k=1}^N \rho_k.$$

This method in principle allows one to obtain exact values for the coefficients of viscosity  $\eta(\xi)$ , multicomponent mass diffusion  $D_{ik}(\xi)$  ( $D_{ii} = 0$ ), thermal diffusion  $D_i^T(\xi)$ , and the coefficient  $\lambda'(\xi)$  (partial thermal conductivity). This procedure was followed in [2] for partially ionized air and in [3] for stably dissociated air. It was shown in [2, 3] that in the case of dissociation one can calculate the coefficients using the lowest order terms ( $\xi = 1, 2$ ) in the Sonine polynomial expansion of the distribution function for the first-order approximation to the Chapman-Enskog method [4].

We use the well-known result [4] relating the generalized multicomponent diffusion coefficients of the mixture  $D_{ij}(1)$  to the binary mixture diffusion coefficients for different pairs of components  $\mathcal{D}_{jk}(1)$ . Then the following Stefan-Maxwell relation is obtained:

$$\sum_{j=1}^N \frac{n_i n_j}{n^2 \mathcal{D}_{ij}} (\mathbf{V}_j - \mathbf{V}_i) = \mathbf{d}_i - \nabla \ln T \cdot \sum_{j=1}^N \frac{n_i n_j}{n^2 \mathcal{D}_{ij}} \left( \frac{D_j^T}{\rho_j} - \frac{D_i^T}{\rho_i} \right). \quad (3)$$

2. To obtain the exact solution of (3) we use the method of calculating binary diffusion coefficients developed in [5, 6], in which the approximate representation is used:

$$\mathcal{D}_{ij} = \frac{D}{F_i F_j}, \quad (4)$$

where  $D(p, T)$  is a parameter depending on the given multicomponent system as a whole, and  $F_i(T)$  refers to the  $i$ -th component. Since for a mixture consisting of  $N$  components there are  $N(N-1)/2$  distinct binary diffusion coefficients, use of all  $N$  different parameters  $F_i$  leads for  $N > 3$  to an approximate value of  $\mathcal{D}_{ij}$  according to (4), which must then be considered as a correlation relation.

An examination of the accuracy of the correlation given in [5, 6] for various complex mixtures supports the representation of  $\mathcal{D}_{ij}$  in the form (4). In [5, 6] the data on the kinetics in nitrogen-oxygen systems were taken from [8], which was based on the Lennard-Jones potential. However, it is known [1, 3] that application of this potential for calculation of the transport coefficients leads to significant errors at high temperatures. In the present paper we have used a repulsive interaction potential of the form

$$\varphi(r) = \frac{C}{r^\delta}. \quad (5)$$

The resulting expression for the effective scattering cross section takes the form

$$\Omega_{ij}^{(l,s)} = \frac{4(l+1)}{(s+1)! [2l+1 - (-1)^l]} \Gamma\left(s+2 - \frac{2}{\delta}\right) A^l(\delta) \left(\frac{C\delta}{kT}\right)^{2/\delta}, \quad (6)$$

where the  $A^l(\delta)$  integrals are evaluated in [4]. Below we study the five-component system ( $O_2, N_2, NO, O, N$ ), modeling air over the temperature range 2000 to 8000°K and pressure range  $0.1 \cdot 10^5$  to  $1 \cdot 10^5$  Pa. The appropriate values of  $C, \delta, A^l(\delta)$  were taken from [9].

According to [4], the expression for the binary diffusion coefficient, with the help of (6), takes the following form:

$$\frac{\rho \mathcal{D}_{ij}}{M} = a \sqrt{\theta} \frac{\theta^{2/\delta_{ij}}}{\Gamma\left(3 - \frac{2}{\delta_{ij}}\right) A^1(\delta_{ij}) \left(\frac{C_{ij} \delta_{ij}}{kT_0^*}\right)^{2/\delta_{ij}}}; \quad (7)$$

$$\theta = T/T_0^*; \quad a = 2.268 \cdot 10^{-3}; \quad T_0^* = 10^4 k.$$

Assuming further that the approximate relation (4) is satisfied, after some simple transformations we obtain

$$\mathcal{D}_{ij} = \frac{D}{F_i F_j}, \quad D = g^* \frac{M}{\rho} \theta^{0.5 + \frac{2}{\delta^*}},$$

$$F_i(T) = \frac{F_i^0}{\theta^{\omega_i}}, \quad \omega_i = \frac{2}{\delta_i} - \frac{2}{\delta^*}. \quad (8)$$

Values of  $F_i^0$ ,  $2/\delta_i$  for air were found with the help of the method of least squares on the binary diffusion coefficient given by (7). The normalization constants were taken from data on molecular oxygen ( $g^* = 0.1244 \cdot 10^{-4}$  kmole/m $\cdot$ sec;  $\delta^* = 6.68$ ). In Table 1 we show values for  $F_i^0$  and  $\omega_i$  in O-N systems with the interaction potential (5).

The  $\mathcal{D}_{ij}$ (1) were calculated approximately for these systems with the help of the values for  $F_i^0$  and  $\omega_i$  given in Table 2 and the results were compared with the exact values. The results for the quantity  $\rho \mathcal{D}_{ij}/M$  with ( $\omega \neq 0$ ) and without ( $\omega_i = 0$ ) taking into account the temperature dependence of  $F_i$  are given in Table 2 for  $T = 4000^\circ\text{K}$ . Comparison with the results calculated from (7) indicates that the relative error in the value of  $\rho \mathcal{D}_{ij}/M$  is small. The mean relative error does not exceed 3%, while the maximum error in the individual values of the  $\rho \mathcal{D}_{ij}/M$  was 12%. The results also indicate that the temperature dependence of  $F_i(T)$  is weak over the range 2000 to 8000 $^\circ\text{K}$ . Hence for these mixtures one can assume within an error of 3% that the  $F_i$  are constant. We also note that our method leads to a mean absolute error in  $\rho \mathcal{D}_{ij}/M$  which is over an order of magnitude smaller than that in the method based on equal diffusion coefficients; this is in qualitative agreement with the conclusions of [5]. Thus further use of the correlation relation (4) below is completely justified.

3. There remains to find an approximate relation for the mass diffusion current. Substituting (4) into the Stefan-Maxwell relation (3), we find

$$d_i = \frac{M^2}{\rho D} \left( \frac{\alpha_i F_i}{M_i} \sum_{j=1}^N \frac{J_j F_j}{M_j} - \frac{F_i J_i}{M_i} \sum_{j=1}^N \frac{\alpha_j F_j}{M_j} \right). \quad (9)$$

Here for convenience we write the "total mass diffusion current"  $J_i$  as the sum of the molecular diffusion and thermal diffusion currents:

$$J_i = j_i + D_i^T \nabla \ln T. \quad (10)$$

After some relatively straightforward algebraic steps [6] we arrive at the relation

$$d_i = \frac{\alpha_i F_i}{M_i} \sum_{j=1}^N \frac{M_j}{F_j} d_j - \frac{M^2}{\rho D} \frac{J_i F_i}{M_i} \sum_{j=1}^N \frac{\alpha_j F_j}{M_j}. \quad (11)$$

Below for convenience we introduce the new parameters:

$$z_i = \frac{M_i x_i}{\mu_2 F_i}; \quad \mu_1 = \sum_{j=1}^N x_j F_j, \quad \mu_2 = \sum_{j=1}^N \frac{M_j x_j}{F_j}, \quad \mu_3 = \sum_{j=1}^N \frac{\alpha_j}{F_j^2} \frac{dF_j}{dT}. \quad (12)$$

Substitution of (2) and (12) into (11) leads to the following constitutive equation for the diffusion current

$$J_i = - \frac{\rho D}{\mu_1 M} \left[ \mu_2 \nabla z_i + (z_i - \alpha_i) \nabla \mu_2 + \mu_p \nabla \ln p + \alpha_i M \left( \frac{1}{F_i^2} \frac{dF_i}{dT} - \mu_3 \right) \nabla T \right];$$

$$\mu_p = \mu_2 \left[ z_i \left( 1 - \frac{M_i}{M} \right) - \alpha_i \left( 1 - \frac{\sum_{j=1}^N M_j z_j}{M} \right) \right]. \quad (13)$$

This expression replaces the more general relation for multicomponent diffusion (1) and gives precisely the "total mass diffusion current"  $J_i$  of the  $i$ -th component in terms of gradients of the basic gasdynamical quantities of the system as a whole and gradients of the  $i$ -th component only. This functional dependence, obtained in [5] in the case  $\nabla p = 0$ , is an important consequence of the conservation equations and also the correlation relation (4).

TABLE 1. Parameters for Approximate Calculation of Binary Diffusion Coefficients of Dissociated Air

Parameter	O <sub>2</sub>	N <sub>2</sub>	NO	O	N
$F_i^0$	1,0	0,9683	0,9952	0,6152	0,6474
$\omega_i$	0	-0,0114	-0,0904	0,0717	0,0633

TABLE 2. Approximate and Exact Values of  $\rho \mathcal{D}_{ij}/M \cdot 10^5$  kmole/m<sup>2</sup> sec for Air at T = 4000°K

Interacting molecules	Exact (7)	Approximate ( $\omega_i = 0$ )	Approximate ( $\omega_i \neq 0$ )	Interacting molecules	Exact (7)	Approximate ( $\omega_i = 0$ )	Approximate ( $\omega_i \neq 0$ )
O <sub>2</sub> -O <sub>2</sub>	0,2663	0,2988	0,2988	N <sub>2</sub> -N	0,5022	0,4545	0,4766
O <sub>2</sub> -N <sub>2</sub>	0,2892	0,3118	0,3086	NO-NO	0,3265	0,3560	0,3017
O <sub>2</sub> -NO	0,3341	0,3267	0,3003	NO-O	0,4516	0,4965	0,4880
O <sub>2</sub> -O	0,5202	0,4548	0,4857	NO-N	0,4761	0,4754	0,4637
O <sub>2</sub> -N	0,4940	0,4355	0,4615	O-O	0,7092	0,6923	0,7895
N <sub>2</sub> -N <sub>2</sub>	0,3036	0,3254	0,3187	O-N	0,6678	0,6630	0,7502
N <sub>2</sub> -NO	0,3388	0,3404	0,3101	N-N	0,6032	0,6348	0,7128
N <sub>2</sub> -O	0,5167	0,4747	0,5016				

Application of (13) allows significant reduction in the calculation time for gasdynamical systems and hence allows one to make use of certain special features of algorithms used earlier.

It was noted above that in several cases of interest (for example in dissociated air) one can put to a good approximation  $dF_i/dT = 0$  (or  $F_i = F_i^0$ ). This leads to some simplification in (13):

$$j_i + D_i^T \nabla \ln T = - \frac{\rho D}{\mu_1 M} [\mu_2 \nabla z_i + (z_i - \alpha_i) \nabla \mu_2 + \mu_p \nabla \ln p]. \quad (14)$$

4. Another approximation method of calculating transport coefficients is the approach used in [10, 11] for the coefficients of viscosity and thermal conductivity. Use of the correlation dependence (4) allows further simplification of the procedure for calculating the coefficients of viscosity, thermal conductivity, and thermal diffusion in multicomponent non-ionized gas mixtures.

4.1. Viscosity. The following expression for the coefficient of viscosity  $\eta_m$  (1) in a gas mixture was derived in [4]:

$$\eta_m = \sum_{i=1}^N \frac{x_i}{\frac{x_i}{\eta_i} + \sum_{\substack{k=1 \\ k \neq i}}^N \frac{x_k}{\rho \mathcal{D}_{ik}} \frac{M}{M_i} b_{ik}}; \quad (15)$$

$$b_{ik} = \frac{2M_i}{M_i + M_k} \left[ 1 + \frac{3}{5} \frac{M_k}{M_i} A_{ik}^* \right],$$

where

$$\eta_i = \frac{5}{6} \frac{M_i}{M} \frac{\rho \mathcal{D}_{ii}}{A_{ii}^*}; \quad A_{ik}^* = \frac{\Omega_{ik}^{(2,2)}}{\Omega_{ik}^{(1,1)}}. \quad (16)$$

Expression (15) represents the formulation of the Wilke method of calculating the coefficient of viscosity. The above result was obtained using the Lennard-Jones interaction potential, and  $b_{ik} = 6/5 A_{ik}^* = 1.385$ . However, it was noted above that this interaction model leads to poor results at high temperatures. Therefore, it is necessary to refine the value of the parameter  $b_{ik}$ . This was carried out for dissociated air and it was found that within an error of 5% one can put  $b_{ik} = b = 1.473$ . This value will be used in the calculations below.

With the help of (4) and (16), the coefficient of viscosity can be written in a different form:

TABLE 3. Values of the Coefficients of Viscosity and Partial Thermal Conductivity in Dissociated Air

T, K	$\eta \cdot 10^4, \text{ Pa} \cdot \text{sec}$		$\lambda, \text{ W/m} \cdot \text{deg}$		T, K	$\eta \cdot 10^4, \text{ Pa} \cdot \text{sec}$		$\lambda, \text{ W/m} \cdot \text{deg}$	
	exact [3]	approximate	exact [3]	approximate		exact [3]	approximate	exact [3]	approximate
$p = 0,1 \cdot 10^5, \text{ Pa}$									
2000	0,6890	0,7294	0,1257	0,1291	2000	0,6889	0,7299	0,1257	0,1292
3000	0,9847	1,017	0,1853	0,1854	3000	0,9678	1,013	0,1810	0,1838
4000	1,306	1,309	0,2518	0,2429	4000	1,289	1,303	0,2471	0,2409
5000	1,583	1,573	0,3088	0,2948	5000	1,572	1,572	0,3048	0,2933
6000	1,850	1,839	0,3743	0,3553	6000	1,849	1,832	0,3641	0,3467
7000	2,038	2,128	0,4304	0,4244	7000	2,103	2,110	0,4298	0,4114
8000	2,278	2,415	0,4858	0,4845	8000	2,307	2,406	0,4868	0,4796
$p = 1 \cdot 10^5, \text{ Pa}$									
2000	0,6889	0,7299	0,1257	0,1291	2000	0,6889	0,7299	0,1257	0,1292
3000	0,9847	1,017	0,1853	0,1854	3000	0,9678	1,013	0,1810	0,1838
4000	1,306	1,309	0,2518	0,2429	4000	1,289	1,303	0,2471	0,2409
5000	1,583	1,573	0,3088	0,2948	5000	1,572	1,572	0,3048	0,2933
6000	1,850	1,839	0,3743	0,3553	6000	1,849	1,832	0,3641	0,3467
7000	2,038	2,128	0,4304	0,4244	7000	2,103	2,110	0,4298	0,4114
8000	2,278	2,415	0,4858	0,4845	8000	2,307	2,406	0,4868	0,4796

TABLE 4. Values of the Coefficient of Thermal Diffusion  $D_T \cdot 10^{10}, \text{ Pa} \cdot \text{sec}$  in Dissociated Air ( $p = 0,1 \cdot 10^5 \text{ Pa}$ )

T, K	O <sub>2</sub>		N <sub>2</sub>		NO		O		N	
	exact [3]	approximate	exact	approximate	exact	approximate	exact	approximate	exact	approximate
2000	4156	4835	4048	4208	-76,61	-516,5	-31,56	-111,0	-0,0002	-0,0002
3000	3853	7315	1687	1480	758,1	-1229	-6294	-20890	-3,802	-4,750
4000	267,8	805,9	18420	66750	1874	1011	-20080	-67880	481,7	-678,6
5000	21,57	54,80	31300	96700	1013	813,5	-22780	-83190	-9558	-14370
6000	5,935	10,45	52560	125100	667,8	727,7	-14483	-64010	-38750	-61810
7000	1,997	3,106	20070	46280	250,8	332,4	-183,5	-256,4	-40140	-20980
8000	0,716	1,176	3339	7809	73,17	103,2	3,882	9807	-7294	-17720

$$\eta_m \approx \frac{\rho D \mu_2}{\mu_1 M b} \quad (17)$$

Calculations using (17) for stably dissociated air at  $p = 0.1 \cdot 10^5$  and  $1 \cdot 10^5$  Pa are displayed in Table 3. The air composition was calculated according to the method of [12]. Comparison with results based on the more rigorous kinetic theory [2, 3] indicated good agreement of the approximate approach (within around 3%) over the entire temperature range  $T = 2000-8000^\circ\text{K}$ .

**4.2. Heat Conductivity.** An approximation formula similar to (15) is known for the coefficient  $\lambda_m^{0'}$  in a mixture of monatomic gases. In [11] the following expression is suggested:

$$\lambda_m^{0'} = \frac{\sum_{i=1}^N x_i \lambda_i^0}{x_i + 1.065 b \frac{M}{M_i} \eta_i \sum_{j=1}^N \frac{x_j}{\rho \mathcal{D}_{ij}}}; \quad \lambda_i^0 = \frac{15}{4} \frac{R}{M_i} \eta_i \quad (18)$$

The factor 1.065 was chosen from the best fit to the experimental data [11]. In order to obtain an expression suitable for later calculations, we substitute (4) into (18), and finally we have

$$\lambda_m^{0'} \approx \frac{15}{4} R \frac{\rho D}{\mu_1 M b} \sum_{i=1}^N \frac{x_i F_i}{1.065 - \frac{x_i F_i}{\mu_1} 0.065} \quad (19)$$

In the calculation of the thermal conductivity for nonmonatomic gases it is necessary to include contribution of the internal degrees of freedom of the molecules  $\lambda_{in}$ :

$$\lambda = \lambda_m^{0'} + \lambda_{in} \quad (20)$$

An expression for the corresponding correction, which was first introduced in [4], is, using (4),

$$\lambda_{in} = \frac{\rho R}{M} \frac{\sum_{i=1}^N \frac{x_i c_{in,i}}{\sum_{j=1}^N \frac{x_j}{\mathcal{D}_{ij}}} = \frac{\rho D}{\mu_1 M} R \sum_{i=1}^N \frac{x_i}{F_i} c_{in,i} \quad (21)$$

The results of the calculations for the thermal conductivity of stably dissociated air at  $p = 0.1 \cdot 10^5$  and  $1 \cdot 10^5$  Pa from (19)-(21) are shown in Table 3, where the corresponding data from [3] is also shown. Comparison of these results indicates that the approximation formulas developed in this section are highly accurate.

**4.3. Thermal Diffusion.** There have been only a small number of papers devoted to approximate calculations of the thermal diffusion coefficient in multicomponent systems. From a detailed analysis of the exact expression for  $D_i^T(2)$ , Bartlett, Kendall, and Rindal [6] suggested the use of the simple expression

$$D_i^T = c_i \frac{\rho D}{\mu_1 M} \mu_2 (z_i - \alpha_i) \quad (22)$$

The value of the constant  $c_t$  was chosen to be  $-0.5$ ; this is used in subsequent calculations.

The thermal diffusion coefficient was calculated using (22) for stably dissociated air with  $p = 0.1 \cdot 10^5$  Pa. The air composition in this case was taken from [12]. The results are shown in Table 4, along with data taken from [3] for comparison. In spite of notable discrepancies, there is qualitative agreement and it is expected that the quantitative disagreement will not greatly influence gasdynamical calculations. Apparently a more exact approximate method for calculating  $D_i^T$  has not yet been worked out.

In summary, the method considered here allows the efficient calculation of the transport coefficients in complex mixtures with an accuracy that is acceptable for practical work.

#### NOTATION

$\rho$ , density;  $\mathbf{V}$ , velocity;  $m$ , mass of a molecule;  $x_i$ ,  $\alpha_i$ , molar and mass concentrations of the  $i$ -th component;  $p$ , pressure;  $T$ , temperature;  $M$ , molecular weight.

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METHOD OF ANALYZING AND GENERALIZING EXPERIMENTAL DATA ON THE  
FREE MOTION OF TOLUENE

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An equation describing the heat transfer in the free motion of toluene about a horizontal tube and at supercritical pressures is proposed.

The results of analyses of experimental data by various authors show that the laws of convective heat transfer in free convection at supercritical pressures of the heat carrier differ from the laws of heat transfer in the subcritical region of states of the material. In a series of works, the influence of individual factors on the heat-transfer coefficient has been noted [1, 2]. Therefore, generalization of experimental data at supercritical pressures of different heat carriers by means of a single critical equation with free convection is very difficult. The basic difficulties are associated with taking account of the influence of change in physical properties of the given fluid on the heat-transfer coefficient. The means of taking account of this phenomenon adopted by individual researchers have been different. Many have taken the well-known relations obtained for the Nusselt number ( $Nu_0$ ) and added corrections that take account of the variability of the physical properties.

At present, there exist a series of critical relations for calculating the heat-transfer coefficient with free convection at supercritical pressures. One was proposed in [3] on the basis of the results of investigating the heat transfer of carbon dioxide with free convection in horizontal tubes, in the form

$$Nu = 0.152 Ra^{1/3} (Pr_c/Pr_f)^{0.25} \quad (1)$$

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