

# BOUNDARY CONDITIONS OF SLIP OF A BINARY GASEOUS MIXTURE AND THEIR APPLICATION IN THE DYNAMICS OF AEROSOLS.

## 2. GAS FLOW ALONG A SLIGHTLY CURVED SURFACE. THEORY OF DIFFUSION PHORESIS OF MODERATELY LARGE NONVOLATILE AEROSOL PARTICLES

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*A complete system of boundary conditions for the slip of a binary gaseous mixture along a nonevaporating surface of small curvature is constructed with allowance for all effects that are linear in the Knudsen number. An expression for the rate of diffusion phoresis of a moderately large nonvolatile aerosol particle in the binary gaseous mixture is obtained.*

The surface curvature was taken into account in the boundary conditions of slip of a binary gaseous mixture only in [1, 2]. In them from a solution of the BGK model of the kinetic equation an expression was obtained for the diffusion slip velocity with allowance for Barnett slip and a correction, linear in the Knudsen number  $Kn = \lambda/R$  ( $\lambda$  is the mean free path of the gas molecules;  $R$  is the surface curvature radius), to the coefficient of diffusion slip due to the presence of a radial flow owing to evaporation from the surface. In the case of a nonvolatile particle the latter effect is absent. To construct a theory of diffusion phoresis of moderately large aerosol particles, additional corrections to the slip coefficients for the curvature and also spreading of the mass and heat fluxes and the diffusion flow in the Knudsen layer should be taken into account.

We consider a spherical nonvolatile aerosol particle of radius  $R$  in a flow of a binary gaseous mixture that is nonuniform in temperature and concentration. As in the previous paper [3] we will assume that the variation in the concentration, temperature, and mass velocity of the mixture is small over the mean free path of its molecules and the gradients of the temperature  $T$  and the relative concentration of molecules of the first component of the mixture  $n_{10} = n_1/n$  ( $n = n_1 + n_2$ ) coincide in direction with the mass velocity  $U$ .

We introduce a spherical coordinate system with origin at the center of the particle. The polar axis is directed along  $U$ . All considerations are stated in vector form, i.e., we will consider the distribution function for molecules of the  $i$ -th component of the mixture  $f_i$  as the corresponding component of the vector  $f$ . A similar approach is used, for example, in [4].

We will seek the function  $f$  as a sum of the volume distribution  $\psi$  and a correction  $\Phi$  that describes the influence of the particle:

$$f = f^0 (1 + \psi + \Phi). \tag{1}$$

Here  $f^0$  is a diagonal matrix with the coefficients  $f_{ij}^0 = \delta_{ij} f_i^0$ ,  $f_i^0$  is the equilibrium Maxwell distribution function for molecules of the  $i$ -th component of the mixture.

In [3] the correction  $\Phi_i$  was represented as

$$\Phi_i = \sum_{l=0}^3 a_l^i P_l(c_i). \tag{2}$$

The coefficients  $a_l^i$  were determined from solution of a system of eight differential moment equations, which entails well-known difficulties of a computational character. It is evident that the complexity of computations will increase significantly when computing corrections for the curvature. Therefore, from here on, use will be made of the distribution function

$$\Phi = (a_1 + a_2 \text{sign } c_r) c_\theta \mathbf{W} + (a_3 + a_4 \text{sign } c_r) c_\theta \mathbf{D} + (a_5 + a_6 \text{sign } c_r) c_r c_\theta \mathbf{B}, \quad (3)$$

where  $\mathbf{W}$ ,  $\mathbf{D}$ , and  $\mathbf{B}$  are vectors with the components  $W_i = \sqrt{M_i}$ ,  $D_i$ , and  $B_i$ ;  $M_i = m_i \bar{m} / m_0$ ,  $m_0 = m_1 + m_2$ ;  $D_i$  and  $B_i$  are the corresponding coefficients in the Chapman-Enskog function. This approach corresponds to the basic idea of the method of half-spatial moments [5], according to which the form of the moments of the distribution function  $\Phi$  is chosen in analogy with to the Chapman-Enskog volume distribution.

It can be shown that function (3) yields satisfactory accuracy in computing the coefficients of slip of a mixture along a plane surface. The difference from the values calculated by using function (2) does not exceed 5% over the entire range of mass-diameter relations for the molecules of the mixture components that was considered in [3]. At the same time, using the distribution function in the form (3) enables us to simplify the computations substantially.

The coefficients  $a_l$  are determined from the solution of the Boltzmann kinetic equation, which in matrix form, appears as

$$\hat{f}_{ij}^0 (\mathbf{v} \nabla) \Phi_i = L_{ij} \Phi_i, \quad (4)$$

where  $L_{ij}$  is the matrix operator of collisions:

$$L_{ij} \Phi_i = -\delta_{ij} n_i^2 I_i(\Phi_i) - n_1 n_2 I_{12}(\Phi_i).$$

To form the system of moment equations, we scalar-multiply (4) by the vectors  $\mathbf{W} c_\theta$ ,  $\mathbf{W} c_\theta \text{sign } c_r$ ,  $\mathbf{D} c_\theta$ ,  $\mathbf{D} c_\theta \text{sign } c_r$ ,  $\mathbf{B} c_r c_\theta$ ,  $\mathbf{B} c_r c_\theta \text{sign } c_r$  and integrate over the entire velocity space. Omitting awkward expressions, we note that in a linear approximation with respect to  $\text{Kn}$  the solution of the obtained system of six differential equations appears as

$$a_l = \sum_{\nu=1}^2 C_\nu (\alpha_l^\nu + \text{Kn } \beta_l^\nu) \exp \{ -(\gamma_\nu + \text{Kn } \tau_\nu) r \}, \quad (5)$$

where  $\gamma_\nu$  and  $\tau_\nu$  are determined from the condition of consistency of the system. The coefficients  $\alpha_l^\nu$  and  $\beta_l^\nu$  are expressed in terms of determinants of the fifth rank. The constants of integration  $C_\nu$  are determined from the kinetic boundary conditions of reflection of gas molecules from the particle surface

$$\begin{pmatrix} \nu_{1r} & 0 \\ 0 & \nu_{2r} \end{pmatrix} \mathbf{f}^+ = \Omega \mathbf{f}^-, \quad (6)$$

which that are also represented in a moment form. To do this, equality (6) is scalar-multiplied by the vectors  $\mathbf{W} \nu_\theta$ ,  $\mathbf{D} \nu_\theta$ , and  $\mathbf{B} \nu_r \nu_\theta$  and integrated over the positive velocity half-space. By solving the obtained system of equations we find the unknown constants of integration and the tangential component of the mass velocity for the mixture:

$$U_\theta = D_{12} \left[ \frac{1}{R} (K_{DSl} + \text{Kn } K_{DSl}^R) \frac{\partial n_{10}}{\partial \theta} + \text{Kn } K_{DSl}^B D_{r\theta} \right] + \\ + \frac{\eta}{\rho T} \left[ \frac{1}{R} (K_{TSl} + \text{Kn } K_{TSl}^R) \frac{\partial T}{\partial \theta} + \text{Kn } K_{TSl}^B T_{r\theta} \right] + \text{Kn } R K_{Sl} \Pi_{r\theta}; \quad (7)$$

$$D_{r\theta} = \frac{\partial^2 n_{10}}{\partial r \partial \theta} - \frac{1}{r} \frac{\partial n_{10}}{\partial \theta}; \quad T_{r\theta} = \frac{\partial^2 T}{\partial r \partial \theta} - \frac{1}{r} \frac{\partial T}{\partial \theta};$$

$$\Pi_{r\theta} = r \frac{\partial}{\partial r} \left( \frac{U_\theta}{r} \right) + \frac{1}{r} \frac{\partial U_r}{\partial r}.$$

We do not take into account corrections to the Barnett and isothermal slip coefficients for the curvature, since their contribution to the slip velocity is proportional to the square of Kn.

In addition to the boundary conditions for the tangential component of the velocity it is necessary to obtain the boundary conditions for the normal components of the mass and heat fluxes and the diffusion flow. To do this, we make use of the laws of mass conservation for each component of the mixture and the law of energy conservation.

If there is no adsorption or desorption of molecules of the gaseous mixture on the aerosol particle surface and the surface is impermeable, the normal components of the spatial macroscopic velocity of the  $i$ -th mixture component on the surface must be equal to zero, i.e.,

$$V_{ir} = U_{ir} + \tilde{v}_{ir} = 0 \quad \text{at} \quad r = R. \quad (8)$$

And the spatial velocity itself must satisfy the continuity equation:  $\text{div } \mathbf{V}_i = 0$ . Here  $U_i$  is the macroscopic velocity of the  $i$ -th component of the mixture in the hydrodynamic sense, i.e., the velocity satisfying the gasdynamic equations provided they are assumed valid up to the particle surface;  $\tilde{v}_i = 1/n_i \int f_i^0 \Phi_i v_i dv_i$  is the correction that occurs in taking account of the Knudsen layer.

It is evident that the velocity  $U_i$  also must also satisfy the continuity equation, and therefore the mean velocity  $\tilde{v}_i$  is to satisfy this equation too. By integrating the continuity equation, written for  $\tilde{v}_i$  in a spherical coordinate system, over the Knudsen layer and taking account of equality (8) we obtain

$$r^2 U_{ir} = -r^2 \tilde{v}_{ir} = - \int_R^\infty \frac{r}{\sin \theta} \frac{\partial}{\partial \theta} (\tilde{v}_{i\theta} \sin \theta) dr.$$

In a linear approximation with respect to Kn the variable  $r$  can be replaced by  $R$ . After integrating we obtain the boundary condition for the normal component of the mass velocity of the  $i$ -th mixture component. In specific computations it is more convenient to use the boundary condition for the mass velocity of the mixture:

$$U_r = \sum_{i=1}^2 U_i \frac{\rho_i}{\rho} = \text{Kn} \frac{1}{R} \left[ C_v^D D_{12} \left( \text{ctan } \theta + \frac{\partial}{\partial \theta} \right) \frac{\partial n_{10}}{\partial \theta} + C_v^T \frac{\eta}{\rho T} \left( \text{ctan } \theta + \frac{\partial}{\partial \theta} \right) \frac{\partial T}{\partial \theta} \right] \quad (9)$$

and the boundary condition for the diffusion flow:

$$U_{1r} - U_{2r} = - \left( \frac{\partial n_{10}}{\partial r} + \frac{K_T}{T} \frac{\partial T}{\partial r} \right) D_{12} \frac{n^2}{n_1 n_2},$$

$$\frac{\partial n_{10}}{\partial r} + \frac{K_T}{T} \frac{\partial T}{\partial r} = \text{Kn} \frac{1}{R} \left[ C_D^D \left( \text{ctan } \theta + \frac{\partial}{\partial \theta} \right) \frac{\partial n_{10}}{\partial \theta} + C_D^T \frac{\eta}{\rho T D_{12}} \left( \text{ctan } \theta + \frac{\partial}{\partial \theta} \right) \frac{\partial T}{\partial \theta} \right], \quad (10)$$

where  $K_T$  is the thermodiffusion ratio.

Similarly, we can obtain the boundary condition for the normal component of the heat flux:

TABLE 1. Values of the Kinetic Coefficients

Kinetic coefficients	N <sub>2</sub> -H <sub>2</sub> , n <sub>20</sub> = 0.01	N <sub>2</sub> , n <sub>10</sub> = n <sub>20</sub> = 0.5		
		O <sub>2</sub>	CO <sub>2</sub>	Ar
$K_{DSI}$	-0.1852	0.0233	0.1968	0.1031
$K_{DSI}^R$	-0.1468	0.0780	0.1639	0.1714
$K_{DSI}^B$	0.4846	0.0520	-0.6271	-0.1418
$K_{TSL}$	1.1468	1.1474	1.1282	1.1280
$K_{TSL}^R$	-2.4598	-2.4580	-2.4422	-2.4170
$K_{TSL}^B$	-3.5670	-3.5587	-3.5532	-3.5249
$K_{sl}$	1.1498	1.1478	1.1543	1.1524
$C_v^D$	-0.0435	-0.0131	0.0578	0.0006
$C_v^T$	0.4445	0.4445	0.4390	0.4368
$C_D^D$	0.3377	0.3351	0.3410	0.3357
$C_D^T$	0.0021	-0.0030	-0.0116	-0.0083
$C_q^D$	0.0141	-0.0078	-0.0153	-0.0167
$C_q^T$	0.2272	0.2266	0.2260	0.2223

$$\begin{aligned} & \kappa \frac{\partial T}{\partial r} - \kappa_i \frac{\partial T_i}{\partial r} + PD_{12}K_T \frac{n^2}{n_1 n_2} \frac{\partial n_{10}}{\partial r} = \\ & = Kn \frac{P}{R} \left[ C_q^D D_{12} \left( \tan \theta + \frac{\partial}{\partial \theta} \right) \frac{\partial n_{10}}{\partial \theta} + \frac{\eta}{\rho T} C_q^T \left( \tan \theta + \frac{\partial}{\partial \theta} \right) \frac{\partial T}{\partial \theta} \right] \end{aligned} \quad (11)$$

( $T_i$  and  $\kappa_i$  are the temperature and thermal conductivity of the particle;  $P = nkT$ ).

Conditions (7), (9)–(11) are a complete system of boundary conditions of slip for a nonuniform binary gaseous mixture along a slightly curved nonevaporating surface of small curvature. The values of the kinetic coefficients calculated for specific mixtures for molecules that interact as elastic spheres in pure diffuse reflection of them from the particle surface are given in Table 1.

We use the obtained boundary conditions to compute the rate of diffusion phoresis of a moderately large nonvolatile aerosol particle. We consider a spherical aerosol particle of radius  $R$  in a binary gaseous mixture, in which a constant concentration gradient of the components is maintained. We will assume a small concentration drop over the particle dimensions. This enables us to linearize the diffusion, heat conduction, and Navier-Stokes equations that describe the state of the gas beyond the Knudsen layer. In the stationary case these equations appear as [6]

$$\begin{aligned} \eta \Delta \mathbf{U} &= \Delta P, \quad \text{div } \mathbf{U} = 0, \\ \Delta n_{10} &= 0, \quad \Delta T = 0, \quad \Delta T_i = 0. \end{aligned} \quad (12)$$

At a large distance from the particle the following conditions must be satisfied:

$$\begin{aligned} U_r &= |\mathbf{U}_\infty| \cos \theta, \quad U_\theta = -|\mathbf{U}_\infty| \sin \theta, \\ n_{10} &= n_{10}^* + |(\nabla n_{10})_\infty| r \cos \theta, \quad P = P_0, \quad T = T_0. \end{aligned} \quad (13)$$

TABLE 2. Parameter  $U_D^*$  as a Function of the Knudsen Number

Kn	N <sub>2</sub> -H <sub>2</sub>	N <sub>2</sub> -O <sub>2</sub>	N <sub>2</sub> -CO <sub>2</sub>	N <sub>2</sub> -Ar
0	0.1851	-0.0233	-0.1968	-0.1031
0.05	0.1957	-0.0231	-0.2130	-0.1085
0.10	0.2054	-0.0233	-0.2275	-0.1137
0.15	0.2145	-0.0238	-0.2307	-0.1190
0.20	0.2231	-0.0244	-0.2530	-0.1242
0.25	0.2314	-0.0253	-0.2647	-0.1295
0.30	0.2396	-0.0263	-0.2760	-0.1348

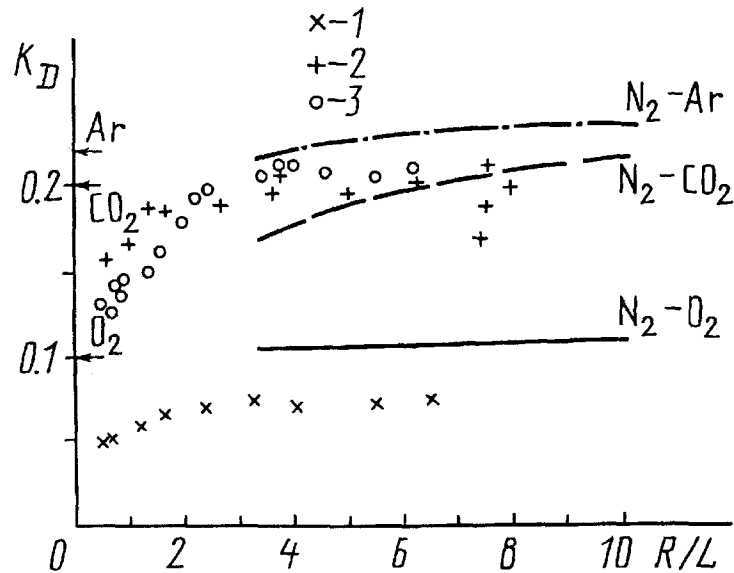


Fig. 1. Parameter  $K_D$  as a function of the particle dimension: 1) N<sub>2</sub>-O<sub>2</sub>; 2) N<sub>2</sub>-CO<sub>2</sub>; 3) N<sub>2</sub>-Ar;  $L = 1/\pi\sqrt{2}n\sigma_{12}^2 \sim \lambda$ .

Here  $U_\infty$  is the velocity of the gaseous mixture relative to the particle at infinity;  $P_0$ ,  $T_0$ , and  $n_{10}^*$  are the undisturbed values of the pressure, temperature, and relative concentration of the first component of the mixture.

By solving the system of differential equations (12) with the boundary conditions (7), (9)–(11), and (13) we obtain the following expression for the diffusion phoresis rate, i.e., the velocity of the aerosol particle relative to the mixture at rest at infinity:

$$U_D = -U_\infty = U_D^* D_{12} (\nabla n_{10})_\infty,$$

$$U_D^* = \frac{-1}{(1 - Kn C_D^D)(1 + 2Kn K_{Sl})} \left\{ K_{DSl} + Kn \left[ K_{DSl}^R - \right. \right.$$

$$\left. - K_{DSl}^B (1 + 2Kn C_D^D) - C_V^D (1 + 6Kn K_{Sl}) + \right.$$

$$\left. + K_{TSl} \frac{\eta P}{\rho T} \frac{2}{\kappa_i + 2\kappa} \left( C_q^D - C_D^D K_T \frac{n}{n_1 n_2} \right) \right\}.$$

It is only terms linear in  $K_T$  that are left here since the thermodiffusion ratio is small for the majority of real mixtures.

Table 2 gives the dimensionless parameter  $U_D^*$  as a function of the Knudsen number for a highly heat-conducting particle, when the contribution of the thermal slip to the diffusion phoresis rate can be ignored.

Of special interest is a comparison with data of an experiment [7] in which the velocity of a mixture and drops of silicone oil suspended in it was measured. Figure 1 gives experimental values of the parameter  $K_D$ , which is related to  $U_D^k$  by the relation:

$$K_D = U_D^* + \frac{m_2 - m_1}{m_1 n_{10} + m_2 n_{20}}.$$

The arrow shows the values of  $K_D$  obtained in experiments on measuring the diffusion phoresis force that acts on a fixed large aerosol particle [8]. The solid, dashed, and dot-dash lines are theoretical values of  $K_D$  for  $N_2-O_2$ ,  $N_2-CO_2$ , and  $N_2-Ar$  mixtures, respectively.

For the  $N_2-Ar$  and  $N_2-CO_2$  mixtures the theoretical curves lie within the spread of the experimental data. For the  $N_2-O_2$  mixture we observe qualitative agreement of the character of the dependence of  $K_D$  on the particle dimension but there is a systematic deviation of the theoretical curve from the experimental data. However, the plot shows that for this mixture the data of the above experiment differ substantially from the value obtained in an experiment with a solid large particle [8], the reason for this difference being vague. It is pertinent to note that the difference of the theoretical value for  $K_D$  from the results of the experiment of [8] is under 10%.

## NOTATION

$T$  and  $\rho$ , temperature and density of the mixture;  $D_{12}$  and  $\eta$ , coefficients of interdiffusion and dynamic viscosity of the mixture;  $k$ , Boltzmann constant;  $n_i$ ,  $m_i$ ,  $\sigma_i$ , and  $v_i$ , concentration, mass, diameter, and intrinsic velocity of molecules of the  $i$ -th component of the mixture;  $c_i$ , dimensionless intrinsic velocity of molecules of the  $i$ -th component of the mixture;  $f_i$ , velocity distribution function for molecules of the  $i$ -th component of the mixture;  $f_i^0$ , equilibrium Maxwell distribution function;  $\psi_i$ , distribution function in the gas volume away from the surface;  $\Phi_i$ , correction describing the influence of the surface;  $K_{Sl}$ , diffusion, thermal, and isothermal slip coefficients;  $K_{DSl}^B$  and  $K_{TSl}^B$ , Barnett slip coefficients;  $K_{DSl}^R$  and  $K_{TSl}^R$ , corrections to the slip coefficients for surface curvature;  $C_v^D$ ,  $C_v^T$ ,  $C_D^D$ ,  $C_D^T$ ,  $C_q^D$ , and  $C_q^T$ , coefficients describing spreading of the mass and heat fluxes and the diffusion flow;  $\delta_{ij}$ , Kronecker delta symbol;  $\nabla$  and  $\Delta$ , Hamilton and Laplace operators.

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