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BARO- AND THERMODIFFUSION OF A GAS MIXTURE IN A CAPILLARY

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In the presence of a solid surfaces confining the flow, the hydrodynamic and diffusional transfer of a gas mixture has a number of peculiarities which distinguish it clearly from the behavior of a mixture in free space. For example, in the analysis of slow flows of a mixture in a capillary, even in the region close to the viscous mode of flow (low Knudsen numbers), it proves important to allow for diffusional slippage at the channel wall, the contribution of the Knudsen boundary layers to the velocity components averaged over a cross section, etc. [1-4]. For this reason, in particular, expressions for the diffusional velocities of the components obtained within the framework of the ordinary kinetic analysis, and valid far from the walls [5, 6], prove to be not fully correct in a description of diffusional transfer inside a capillary.

Below we discuss the derivation of a general expression for the diffusional flux of a gas mixture in a capillary in the presence of longitudinal gradients of concentration, pressure, and temperature. This analysis is confined to the region of low Knudsen numbers ($Kn = \lambda/d \ll 1$, where λ is the effective mean free path of the molecules and d is characteristic transverse size of the channel). Under these conditions the averaged diffusional flux does not depend on the channel geometry in a first approximation with respect to the Knudsen number [4].

Let us consider the flow of a gas mixture in a channel bounded by two infinite parallel planes at $x = \pm d/2$. Let gradients of partial pressure and temperature exist in the z direction. For small $k_\alpha = p_\alpha^{-1} dp_\alpha/dz$ and $\tau = T^{-1}dT/dz$ the linearized kinetic equation for the mixture takes the form [7]

$$v_{\alpha z}\left[k_\alpha + (\beta_\alpha v_\alpha^2 - 5/2)\tau\right] + v_{\alpha x}\partial\Phi_\alpha/\partial x = \sum_\beta \hat{L}_{\alpha\beta}\Phi_\alpha, \tag{1}$$

where Φ_α is a nonequilibrium correction to the distribution function of particles of type α, defined by the equation

$$f_\alpha(\mathbf{v}_\alpha, x, z) = f_\alpha^{(0)}[1 + k_\alpha z + (\beta_\alpha v^2 - 5/2)\tau z + \Phi_\alpha(\mathbf{v}_\alpha, x)],\; f_\alpha^{(0)} = n_{\alpha 0}(\beta_\alpha/\pi)^{3/2}\exp(-\beta_\alpha v^2),\; \beta_\alpha = \frac{m_\alpha}{2kT_0}$$

(the index 0 corresponds to the parameters of an absolute Maxwellian distribution).

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Equations for the moments of the distribution function which follow from (1) are needed below. For a linearized collision operator it is convenient to use the McCormack model [8] for this, which assures that the nonzero moments of the collision integral from the exact and model representations $\hat{L}\Phi$ and $\hat{L}^{(N)}\Phi$ coincide to within the first N moment equations. Then for N = 3 the integration of (1) with a weight $\psi_\alpha(c_\alpha)\exp(-c_\alpha^2)$, where $\psi_\alpha = c_{\alpha z}$, $c_{\alpha x}c_{\alpha z}$, and $c_{\alpha z}(c_\alpha^2 - 5/2)$ for $\mathbf{c}_\alpha = \beta_\alpha^{1/2}\mathbf{v}_\alpha$, leads to moment equations in the form

$$-p_\alpha k_\alpha - \partial\Pi_{\alpha xz}/\partial x = kT\sum_\beta\{(n_\alpha n_\beta/n\,[D_{\alpha\beta}]_1)\qquad(u_{\alpha z} - u_{\beta z}) + \xi_{\alpha\beta}\,[(h_{\alpha z}/m_\alpha p_\alpha) - (h_{\beta z}/m_\beta p_\beta)]\}; \tag{2}$$

$$-p_\alpha\beta_\alpha^{-1/2}\partial Q_{\alpha z}/\partial x = p^2\sum_\beta a_{\alpha\beta}\Pi_{\beta xz}/p_\beta; \tag{3}$$

$$-p_\alpha\tau_\alpha - (2/5)\,\partial P_{\alpha xz}/\partial x\;(p^2/T)\sum_\beta b_{\alpha\beta}h_{\beta z}/p_\beta + (kT/m_\alpha)\sum_\beta \xi_{\alpha\beta}\,(u_{\alpha z} - u_{\beta z}). \tag{4}$$

The right sides of (2)-(4) coincide with the expressions obtained in [6] in Grad's approximation of 13 moments. The values of $[D_{\alpha\beta}]_1$, $\xi_{\alpha\beta}$, $a_{\alpha\beta}$, and $b_{\alpha\beta}$, expressed through the well-known Chapman–Cowling integrals $\Omega_{\alpha\beta}^{lr}$ [5], are also given there. The quantities $u_{\alpha z}$, $\Pi_{\alpha xz}$, and $h_{\alpha z}$ are the macroscopic velocity, the nondiagonal part of the stress tensor, and the relative heat flux of particles of type α, respectively, while for the moments $Q_{\alpha z}$ and $P_{\alpha xz}$, we have

$$Q_{\alpha z} = 2\pi^{-3/2}\int c_{\alpha x}^2 c_{\alpha z}\Phi_\alpha\exp(-c_\alpha^2)\,d\mathbf{c}_\alpha,$$

$$P_{\alpha xz} = 2\pi^{-3/2}p_\alpha\int c_{\alpha x}c_{\alpha z}\left(c_\alpha^2 - \frac{5}{2}\right)\Phi_\alpha\exp(-c_\alpha^2)\,d\mathbf{c}_\alpha.$$

The values of the diffusional fluxes of the components averaged over the channel cross section can be obtained by integrating Eqs. (2) and (4) over x from −d/2 to +d/2, expressing $h_{\beta z}$ from (4), and substituting their values into (2). For a two-component mixture, in particular, we find*

$$J_1 = ny_1y_2(\langle u_{1z}\rangle - \langle u_{2z}\rangle) = -n[D_{12}]_2\;\{dy_1/dz + (y_1/p)dp/dz + [\alpha_T]_1y_1y_2d\ln T/dz + (2/pd)[\Pi_{1xz}(d/2) + \zeta_1P_{1xz}(d/2) + \zeta_2P_{2xz}(d/2)]\}, \tag{5}$$

where $y_\alpha = p_\alpha/p$. Expressions for the coefficient of diffusion $[D_{12}]_2 = [D_{12}]_1/(1 - \Delta_{12})$, where Δ_{12} is the correction of the second approximation, and for the thermodiffusion constant $[\alpha_T]_1$ can be found in [5, 6]; the coefficients ζ_1 and ζ_2 are determined from the equations

$$\zeta_1y_1 + \zeta_2y_2 = \frac{2}{5}[\alpha_T]_1y_1y_2,$$

$$m_1\zeta_2 - m_2\zeta_1 = \frac{2}{5}(m_1 + m_2)\,\Delta_{12}\Big/\left(\frac{6}{5}C_{12}^* - 1\right),$$

where $C_{12}^* = \Omega_{12}^{12}/3\Omega_{12}^{11}$.

To obtain the final result we must find $\Pi_{\alpha xz}$ and $P_{\alpha xz}$ at the channel wall. For this we use Loyalka's approximate method [9, 10], in which the distribution function of particles incident on the wall is assumed to retain the same structure as the asymptotic distribution function valid far from the wall, but with an arbitrary constant determined from the condition of conservation of the tangential component of the total momentum flux in the gas.

We introduce distribution functions of the incident and reflected particles such that $\Phi_\alpha = \Phi_\alpha^+$ for $c_{\alpha x} > 0$ and $\Phi_\alpha = \Phi_\alpha^-$ for $c_{\alpha x} < 0$ ($c_{\alpha x} > 0$ corresponds to the positive direction of the x axis). Using the usual Maxwellian condition of particle reflection at a wall, for the functions $\Phi_\alpha^\pm$ at x = d/2 we have

$$\begin{gathered}\Phi_\alpha^+(\mathbf{c}_\alpha, d/2) = 2\beta_\alpha^{1/2}(a + w_{\alpha z}^{\mathrm{as}})\,c_{\alpha z} + 2p_\alpha^{-1}\times\Pi_{\alpha xz}^{\mathrm{as}}(d/2)\,c_{\alpha x}c_{\alpha z} + \frac{4}{5}\beta_\alpha^{1/2}p_\alpha^{-1}h_{\alpha z}^{\mathrm{as}}c_{\alpha z}(c_\alpha^2 - 5/2),\\ \Phi_\alpha^-(c_{\alpha x}, c_{\alpha y}, c_{\alpha z}, d/2) = (1 - \varkappa_\alpha)\,\Phi_\alpha^+(-c_{\alpha x}, c_{\alpha y}, c_{\alpha z}, d/2),\end{gathered} \tag{6}$$

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*$J_\alpha = G_\alpha - Gy_\alpha$, where $G_\alpha = n_\alpha\langle u_\alpha\rangle$ is the diffusional flux of component α in the frame of reference where an overall molar flux G of the mixture is absent. We note that $J_1 = -J_2$ for a two-component mixture.

where a is an arbitrary constant; $\varkappa_\alpha$ is the fraction of particles undergoing diffuse reflection at the wall. Knowing $w^{ac}_{\alpha z} = u^{ac}_{\alpha z} - u^{ac}_{z}$ (u_z is the average-mass velocity of the mixture), we determine $\Pi^{as}_{\alpha xz}$ and $h^{as}_{\alpha z}$ through the solutions of Eqs. (2)-(4) which are valid far from the wall, where the distribution function corresponds to the usual approximation of 13 moments, thanks to which we have the conditions [6]

$$Q^{as}_{\alpha z} = \beta^{1/2}_\alpha \left(u^{as}_{\alpha z} + \frac{2}{5} p_\alpha^{-1} h^{as}_{\alpha z}\right),\ P^{as}_{\alpha xz} = \Pi^{as}_{\alpha xz}.$$

The solutions obtained in this region depend formally on the boundary conditions, although the terms connected with them decline exponentially at distances on the order of several mean free paths from the wall, so that far from the wall $w^{as}_{\alpha z}$ and $h^{as}_{\alpha z}$ hardly depend on x, coinciding with the results obtained in [6], while for $\Pi^{as}_{\alpha xz}$ we have

$$\Pi^{as}_{\alpha xz} = -x(\eta_\alpha/\eta)dp/dz,\ \eta = \sum_\alpha \eta_\alpha,$$

where η_α is the partial coefficient of viscosity [6].

We define the value of $\Pi_{\alpha xz}$ (d/2) at the wall as

$$\Pi_{\alpha xz}(d/2) = 2\pi^{-3/2} p_\alpha \int\limits_{-\infty}^{+\infty}\int\limits_{-\infty}^{+\infty} c_{\alpha z} dc_{\alpha z} dc_{\alpha y} \left[\int\limits_0^\infty c_{\alpha x}\Phi^+_\alpha(\mathbf{c}_\alpha, d/2)\exp\left(-c^2_\alpha\right) dc_{\alpha x} + \int\limits_{-\infty}^0 c_{\alpha x}\Phi^-_\alpha(\mathbf{c}_\alpha, d/2)\exp\left(-c^2_\alpha\right) dc_{\alpha x}\right]. \quad (7)$$

We note that from Eq. (2), after summation over α and integration from −d/2 to +d/2, we get

$$\Pi_{xz}(d/2) = \sum_\alpha \Pi_{\alpha xz}(d/2) = -(d/2)\,dp/dz \equiv \Pi^{as}_{xz}(d/2). \quad (8)$$

For simplicity, let us consider the case of $\varkappa_\alpha = 1$ (fully diffuse reflection). Substituting (6) into (7), integrating over the velocities, and using (8), we find

$$a = -\frac{1}{\sum\limits_\alpha p_\alpha \beta^{1/2}_\alpha}\left[\frac{\pi^{1/2}}{4} d\,\frac{dp}{dz} + \sum_\alpha \beta^{1/2}_\alpha\left(p_\alpha w^{as}_{\alpha z} + \frac{1}{5} h^{as}_{\alpha z}\right)\right].$$

Calculating the quantities $\Pi_{\alpha xz}$ (d/2) and $P_{\alpha xz}$ (d/2) with this value of a, we have (α, β = 1, 2)

$$\Pi_{\alpha xz}(d/2) = -\frac{d}{4}\frac{dp}{dz}\left(\frac{m^{1/2}_\alpha y_\alpha}{(m^{1/2})_y} + \frac{\eta_\alpha}{\eta}\right) + \frac{p}{(2\pi kT)^{1/2}}\frac{m_1^{1/2} m_2^{1/2} y_1 y_2}{(m^{1/2})_y}\left[w^{as}_{\alpha z} - w^{as}_{\beta z} + \frac{1}{5}\left(\frac{h^{as}_{\alpha z}}{p_\alpha} - \frac{h^{as}_{\beta z}}{p_\beta}\right)\right],$$

$$P_{\alpha xz}(d/2) - \frac{1}{2}\Pi_{\alpha xz}(d/2) = -\frac{d}{8}\frac{\eta_\alpha}{\eta}\frac{dp}{dz} + \frac{6}{5}\left(\frac{m_\alpha}{2\pi kT}\right)^{1/2} h^{as}_{\alpha z},$$

$$(m)^{1/2}_y = m_1^{1/2} y_1 + m_2^{1/2} y_2.$$

Substituting $\Pi_{\alpha xz}$ (d/2) and $P_{\alpha xz}$ (d/2) into (5), we can find that the parts of them proportional to $w^{as}_{\alpha z}$ and $h^{as}_{\alpha z}$ make contributions to J_1 corresponding to the second order with respect to the Knudsen number. The remaining terms only affect the value of the coefficient to the pressure gradient, as a result of which J_1 takes the form $J_1 = -n[D_{12}]_2\,[dy_1/dz + \alpha_p y_1 y_2\, d\ln p/dz + [\alpha_T]_1 y_1 y_2 \times d\ln T/dz]$, where $\alpha_p = \frac{1}{2}([\alpha_p]_2 + \alpha_p^{k})$; $[\alpha_p]_2 = [\alpha_p]_1(1 - \Delta_p) - \frac{2}{5}[\alpha_T]_1$;

$$\alpha_p^{k} = \frac{m_2^{1/2} - m_1^{1/2}}{(m^{1/2})_y} - \delta;\ [\alpha_p]_1 = \frac{1}{\eta}\left(\frac{\eta_2}{y_2} - \frac{\eta_1}{y_1}\right);\ \Delta_p = \zeta_2 - \zeta_1;$$

$$\delta = \frac{1}{2}\left(\zeta_1 m_1^{1/2} y_1 + \zeta_2 m_2^{1/2} y_2\right)/(m^{1/2})_y y_1 y_2.$$

The main result distinguishing J_1 from the well-known expressions of [5, 6] consists in a new value of the barodiffusion constant, equal to half the sum of the two values of $[\alpha_p]_2$ and $\alpha_p{}^{k}$. The first of these coincides with the barofiffusion constant in a viscous stream, first calculated in [6] and actually figuring in the expression for the difference between the asymptotic velocity components, $u_{1z}^{as} - u_{2z}^{as}$. The second value, as is easy to ascertain, equals (with the opposite sign) the coefficient of diffusional slip σ_{12} obtained in [1]. As for the thermodiffusion constant, its difference from the usual value $[\alpha_T]_1$ appears only in the next order with respect to the Knudsen number.*

As an illustration of the difference between α_p and $[\alpha_p]_2$ let us consider the case of a mixture having a small relative difference between the masses and the scattering cross sections of the component molecules of the mixture. Then for the model of molecules as rigid spheres with diameters d_1 and d_2, the general expression for α_p takes the form

$$\alpha_p = 1.275(m_2 - m_1)/(m_2 + m_1) - 0{,}597(d_2 - d_1)/(d_2 + d_1). \tag{9}$$

In the expression for $[\alpha_p]_2$ the corresponding coefficients are 1.405 and −1.263. The new barodiffusion constant gives half as strong a dependence on the relative difference in the collision cross sections of the molecules and a fairly close dependence on the ratio of masses of the component molecules. Here we do not give the complete expression for α_p obtained by dropping the assumption $\varkappa_\alpha = 1$. If in addition to the conditions of the particular case considered above, however, we assume the presence of a small relative difference between $\varkappa_1$ and $\varkappa_2$ (with $\varkappa_1 + \varkappa_2 \approx 2$), then an additional term of the type $1.90(\varkappa_2 - \varkappa_1)/(\varkappa_2 + \varkappa_1)$ appears in Eq. (9) for α_p, whereas $[\alpha_p]_2$ does not depend on the character of molecular scattering at the wall.

We note that the values of the baro- and thermodiffusion constants essentially determine the effect of separation of a gas mixture in a capillary [1, 4], and the expressions obtained above can be used to estimate it for $Kn \ll 1$.

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*We do not consider results obtained in the second-order approximation with respect to the Knudsen number, since a description using the distribution function (6) proves inadequate in this case.