Motion of a spherical particle in a rarefied gas. Part 1. A liquid particle in its saturated vapour

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The results of a theoretical investigation of the low-speed motion of a volatile spherical particle in its saturated vapour at arbitrary values of the Knudsen number, evaporation coefficient and tangential momentum accommodation coefficient are presented. The problem is solved on the basis of the Bhatnagar, Gross & Krook (1954) linearized kinetic equation with the model collision integral. The variational method developed for the solution of the axisymmetric problems (Cercignani & Pagani 1968) is employed to calculate the drag. Phase changes on the particle surface have been shown to reduce the drag by an order depending on the Knudsen number. Incomplete accommodation of molecules colliding with the particle also results in the reduction of the drag over the whole range of Knudsen numbers.

1. Introduction

A single aerosol particle suspended in a non-equilibrium gas can be affected by different forces due to non-uniformity. They are the isothermal drag, thermal force, photophoretic force, diffusion force and other kinds of forces caused by the combined flows of heat, mass and momentum (Hidy & Brock 1970; Fuchs 1971; Brock 1980).

The purpose of the present paper is to investigate the simplest of the above mentioned phenomena – the isothermal drag. This phenomenon is a subject of considerable interest for general investigations in mechanics of aerosols (Hidy & Brock 1970). The study of the drag and sedimentation velocity is basic for the analysis of processes of coagulation (e.g. in clouds), for the study of the drift of aerosol particles in air flow and their sedimentation velocity, etc.

Optimization of various technological processes (e.g. cleaning of industrial gases, heterogeneous catalysis, motion of drops and dust particles near turbine blades) also requires the detailed analysis of particle motion.

The expression for the isothermal drag of a solid spherical particle moving in a gas at arbitrary degrees of its rarefaction was first obtained by Millikan (1911) after analysing a considerable amount of experimental data for different substance–gas pairs. Among heuristic approximate theories giving a satisfactory description of the particle drag over a wide range of Knudsen numbers (Kn) are the works by Fuchs & Stechkina (1963); Sherman (1963) and Annis & Malinauskas (1972). Fuchs & Stechkina suggested a method of calculating the drag on the basis of 'lacing' the free-molecular and viscous slip-flow solutions near the particle surface (the so called boundary-sphere method). The maximum discrepancy between the results obtained by this method and Millikan's (1923) formula amounts to 5% at $Kn = 3$. The method suggested by Sherman (1963) is similar to the one (the boundary-sphere method) just described. The expression for the drag is also obtained on the basis of the free-molecular and viscous limits. The departure from Millikan's formula amounts to

10 % at intermediate values of Kn. Annis & Malinauskas (1972) determined the drag using the method of 'giant molecules'. Though the authors stress that the results obtained are in a good agreement with those of Millikan (1923) (the discrepancy does not exceed 2 %) the method is not free from certain physical contradictions and includes a great number of adjustable parameters. Waldmann & Vestner (1977) used the method of non-equilibrium thermodynamics and obtained an expression that gives a satisfactory description of the viscous slip-flow and intermediate flow-past regimes.

A strict approach to this problem should be based on solving the kinetic equation with corresponding boundary conditions. The work of Cercignani, Pagani & Bassanini (1968) presents results obtained for the drag at arbitrary Kn. The problem was solved by a variational method on the basis of the model kinetic equation of Bhatnagar, Gross & Krook (1954). Molecules were assumed to be reflected from the particle surface diffusively, the temperature of the particle and the surrounding gas being the same. Calculations showed a good agreement (the discrepancy is within 2 %) with Millikan's (1923) empirical formula. Asymptotic analytical expressions for the drag have been obtained in the free-molecular and viscous slip-flow regimes. Lea & Loyalka (1982) solved the set of integral equations for macroparameters obtained by Cercignani *et al.* (1968) numerically and then calculated the drag and velocity profile near the particle. Their results are in good agreement with Millikan's (1923) data. An analytical expression for the drag at arbitrary Kn was obtained by Phillips (1975). The problem was solved by the moments method for the specular–diffuse scheme of boundary conditions. The solution provides correct results for the viscous and free-molecular limits for the case of perfect accommodation on the surface and gives a satisfactory description (within 10 % accuracy) of the intermediate flow-past regime. In the work of Khlopkov (1975) the drag was calculated on the basis of solving the linearized Boltzmann equation by the Monte-Carlo method. Two schemes of boundary conditions were considered: fully specular and fully diffuse molecular scattering by the particle surface. In the case of diffuse scattering calculations have shown a satisfactory agreement (within 6–10 %) with the results of Millikan (1923) for the transitional and free-molecular regimes. For the viscous slip-flow regime, the discrepancy amounts to 30 %, which must be due to the peculiarities of the numerical method used by the author.

Phase changes on the particle surface and their effect on the drag were analysed only for limiting Knudsen-number regimes in the works by Onishi (1977), Sone & Aoki (1979) and Brock (1964). Sone & Aoki (1979) and Onishi (1977) solved the problem of volatile-spherical-particle motion in its own saturated vapour at small Kn on the basis of the asymptotic method developed by the authors. All molecules incident on the particle surface were assumed to condense and then evaporate with an equilibrium distribution function. The ratio of heat conductivity coefficients of the vapour and liquid phases was assumed to be arbitrary by Sone & Aoki (1979) and to be zero by Onishi (1977). Volatile-particle motion in the free-molecular regime was investigated by Brock (1964). It was assumed that some molecules are specularly reflected, while others are absorbed (with a certain probability of subsequent condensation) on the particle surface. The expression obtained for the drag includes the dependence on the gas–surface interaction coefficients.

Some methodologically significant aspects of the statement of the problem of volatile-particle motion at arbitrary Knudsen numbers were discussed in Brock's (1967) paper. Its estimations, however, cannot substitute for the results of a thorough investigation of the problem.

The purpose of this work is to solve the problem of volatile-particle motion in its

own saturated vapour at arbitrary values of Kn, the evaporation coefficient and the tangential momentum accommodation coefficient.

2. Formulation of the problem

Consider a stationary flow past a particle of radius R_0 in its own saturated vapour. Phase changes on the particle surface are responsible for a number of physical phenomena which require careful analysis. The processes of condensation on the front of the particle and evaporation on its rear produce a temperature gradient along the motion direction. This effect is enhanced owing to the thermal polarization of the particle surface (Deryaguin & Bakanov 1962) caused by the isothermal heat flow in the moving gas. In its turn, a non-uniformly heated particle is affected by the radiometric force contributing to the drag.

This paper concerns the case reflecting the majority of real situations. The particle heating due to viscous friction in the retarding flow was not taken into consideration since it is proportional to the square of the Mach number, while only terms linear in the Mach number were taken into account in the theory. The thermal conductivity of the particle λ_p was assumed to considerably exceed that of the vapour λ_g, i.e. $\lambda_g/\lambda_p \to 0$. In this case the temperature gradient produced inside the particle and thermal polarization of its surface may be ignored, and the particle temperature may be considered constant and equal to the vapour temperature T (Onishi 1977).

The temperature gradient inside the particle and its effect on the drag of a volatile particle in a slip-flow regime was taken into account by Sone & Aoki (1979). Their results show that in this case ($Kn \ll 1$) correction for the drag even for low-conductivity particles ($\lambda_g/\lambda_p \gg 1$) is in proportion to Kn^2. For low-conductivity particles the drag increase due to the thermal polarization of the particle surface amounts to no more than several per cent. For high-conductivity particles this effect is negligible. This was theoretically investigated for the free-molecular regime by Beresnev & Chernyak (1983). Experimental data for the slip-flow regime were obtained by Bakanov & Vysotskiy (1982).

The particle was assumed to retain its spherical shape, which means that a possible deformation of the particle shape due to evaporation–condensation processes is cancelled out by the surface-tension force tending to impart a spherical shape to the particle. This assumption is valid for 'slow' evaporation–condensation processes.

The particle surface-curvature effect causing a difference between saturated vapour pressures above a curved surface and a plane one was not taken into account in this work. This enabled the authors to consider the particle radius to be constant, its lower limit for most substances being $R_0 = 10^{-6}$ m. No substance circulation due to convection was assumed to occur within the particle.

Let the undisturbed values of the number density and the vapour velocity be n_∞ and U_∞ respectively. Let x be a radius vector from the centre of the particle and v the molecular velocity vector (figure 1).

If the flow velocity is small, i.e.

$$u_\infty = U_\infty \left(\frac{m}{2kT}\right)^{\frac{1}{2}} \ll 1,$$

then the distribution function $f(x, v)$ describing the vapour condition can be linearized:

$$f(x, v) = f_\infty [1 + 2c \cdot u_\infty + h], \tag{1}$$

where
$$f_\infty = n_\infty \left(\frac{m}{2\pi kT}\right)^{\frac{3}{2}} \exp(-c^2), \quad c = v\left(\frac{m}{2kT}\right)^{\frac{1}{2}},$$
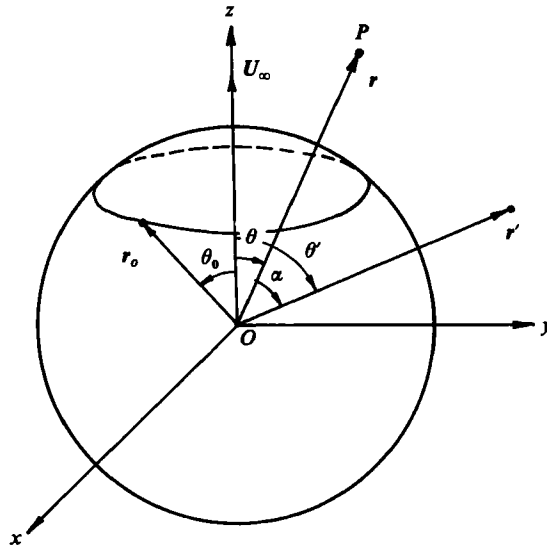
FIGURE 1. Geometry of the problem.

and h is the distribution-function disturbance near the particle. Representation of the distribution function in (1) implies that only linear members of the Mach number were taken into account in the theory.

The linearized kinetic equation with the collision integral in the BGK form (Bhatnagar *et al.* 1954) is the following:

$$\boldsymbol{\Omega}\cdot\frac{\partial h}{\partial \boldsymbol{r}} = 2\boldsymbol{\Omega}\cdot\boldsymbol{W}+\frac{1}{c}\,\nu-\frac{1}{c}\,h, \tag{2}$$

where

$$\boldsymbol{\Omega} = \frac{\boldsymbol{c}}{c}, \quad \boldsymbol{r} = \tfrac{1}{2}\pi^{\frac{1}{2}}\frac{\boldsymbol{x}}{l},$$

and

$$\nu = \frac{n-n_{\infty}}{n_{\infty}} = \pi^{-\frac{3}{2}}\int h \exp\left(-c^2\right) \mathrm{d}\boldsymbol{c}, \\[2mm]
\boldsymbol{W} = \boldsymbol{u}-\boldsymbol{u}_{\infty} = \pi^{-\frac{3}{2}}\int h\boldsymbol{c} \exp\left(-c^2\right) \mathrm{d}\boldsymbol{c}, \tag{3}$$

are the disturbances of the dimensionless number density and macroscopic velocity; l is the mean free path of the vapour molecules defined by the viscosity-coefficient expression

$$\eta_{\mathrm{g}} = \tfrac{1}{2}mn\left(\frac{8kT}{\pi m}\right)^{\frac{1}{2}}l.$$

To solve (2) boundary conditions that disturbance h must satisfy should be set. Here we must take into account the discontinuity in the molecular velocity space character of the velocity distribution function (Kogan 1969):

$$f(\boldsymbol{x}, \boldsymbol{v})|_{|\boldsymbol{x}|=R_0} = \begin{cases} f^{+}, & (\boldsymbol{n}\cdot\boldsymbol{\Omega}) > 0, \\ f^{-}, & (\boldsymbol{n}\cdot\boldsymbol{\Omega}) < 0, \end{cases}$$

where $\boldsymbol{n}$ is the normal unit vector and f^{+} and f^{-} are the distribution functions of the emitted and incident vapour molecules respectively.

In the boundary condition for the distribution function f^{+} a part α_{m} of the

molecules was assumed to evaporate evenly from the surface with a velocity distribution according to Maxwellian law f_∞, and the $(1-\alpha_m)$ part, experiencing no condensation in colliding with the particle, was supposed to be reflected. In this case the emitted molecules' distribution function f^+ can be written in the following form according to Brock (1964):

$$f^+ = \alpha_m f_\infty + (1-\alpha_m)f^r, \qquad (4a)$$

when $|r| = R$, $(n \cdot \Omega) > 0$.

Here $R = \frac{1}{2}\pi^{\frac{1}{2}} R_0/l = \frac{1}{2}\pi^{\frac{1}{2}} Kn^{-1}$ is the dimensionless radius of the sphere related to the Knudsen number $Kn = l/R_0$.

We admit the possibility of the arbitrary accommodation of tangential momentum at the collision of the reflected non-condensed molecules with the particle. This possibility could be taken into account by writing the distribution function of the reflected molecules f^r in the form

$$f^r(R) = f_\infty[1 + A + Bc_\theta], \quad |r| = R, \quad (n \cdot \Omega) > 0, \qquad (4b)$$

according to Cercignani & Pagani (1969), where c_θ is the θ-component of the dimensionless molecular velocity c.

The form of the boundary conditions for the disturbance h is derived from (1), (4a) and (4b):

$$h(r, c) \to 0 \qquad\qquad\qquad \text{when} \quad |r| \to \infty,$$

$$h(r, c) = (1-\alpha_m)[A + Bc_\theta] - 2c \cdot u_\infty \quad \text{when} \quad |r| = R, \quad (n \cdot \Omega) > 0. \qquad (5)$$

The unknown quantities A and B depending on the polar angle θ_0 (figure 1) are derived from the laws of conservation of mass and tangential momentum on the particle surface.

The law of conservation of mass is expressed here in the following form:

$$|N^+| = \alpha_m|N_s| + (1-\alpha_m)|N^-|, \qquad (6)$$

where
$$|N^+| = \int_{(n \cdot \Omega) > 0} (n \cdot v)f^+ \, dv$$

is the full number flow of emitted molecules;

$$(1-\alpha_m)|N^-| = -(1-\alpha_m) \int_{(n \cdot \Omega) < 0} (n \cdot v)f^-(r = R) \, dv$$

is the number flow of incident molecules experiencing no condensation in colliding with the particle; and

$$\alpha_m|N_s| = \alpha_m \int_{(n \cdot \Omega) > 0} (n \cdot v)f_\infty \, dv$$

is the number flow of evenly evaporated molecules.

Equation (6) was derived by multiplying (4) by $(n \cdot v)$ and integration in the half-space $(n \cdot \Omega) > 0$. It must be taken into account that all the non-condensed molecules are supposed to be reflected, that is

$$(1-\alpha_m)|N^-| = (1-\alpha_m)|N^r|,$$

where
$$|N^r| = \int_{(n \cdot \Omega) > 0} (n \cdot v)f^r \, dv.$$

The tangential momentum conservation law for the molecules reflected from the particle surface is expressed in the following form:

$$|P^r_{r\theta}| = (1-\alpha_m)(1-\alpha_r)|P^-_{r\theta}|, \qquad (7)$$

where
$$|P_{r\theta}^{\tau}| = (1-\alpha_{\mathrm{m}}) \int_{(\boldsymbol{n}\cdot\boldsymbol{\Omega})>0} m v_r v_\theta f^{\tau} \, \mathrm{d}\boldsymbol{v}$$

is the radial flow of the tangential momentum of the reflected molecules;

$$|P_{r\theta}^{-}| = - \int_{(\boldsymbol{n}\cdot\boldsymbol{\Omega})<0} m v_r v_\theta f^{-}(r=R) \, \mathrm{d}\boldsymbol{v}$$

is the radial flow of the tangential momentum of incident molecules; α_τ is the tangential momentum accommodation coefficient; α_{m} is the evaporation–condensation coefficient. The normal momentum accommodation coefficient α_{n} was assumed to be equal to 1.

3. Basic equations

The kinetic equation (2), taking into account the boundary conditions (5) was formally integrated along the characteristics (Marchuk 1961) and according to (3) was transformed into a set of integral equations for the number density and macroscopic velocity:

$$\left. \begin{aligned}
\nu &= \pi^{-\frac{3}{2}} \nu_0 + \pi^{-\frac{3}{2}} \int_V [\nu J_1 + 2(\boldsymbol{\Omega}\cdot\boldsymbol{W})J_2] \frac{\mathrm{d}\boldsymbol{r}'}{|\boldsymbol{r}-\boldsymbol{r}'|^2}, \\
\boldsymbol{W} &= \pi^{-\frac{3}{2}} \boldsymbol{W}_0 + \pi^{-\frac{3}{2}} \int_V [\nu J_2 + 2(\boldsymbol{\Omega}\cdot\boldsymbol{W})J_3] \frac{\boldsymbol{\Omega}\cdot\mathrm{d}\boldsymbol{r}'}{|\boldsymbol{r}-\boldsymbol{r}'|^2},
\end{aligned} \right\} \tag{8}$$

where
$$J_n(t) = \int_0^\infty c^n \exp\left(-c^2 - \frac{t}{c}\right) \mathrm{d}c, \qquad \boldsymbol{\Omega} = \frac{\boldsymbol{r}-\boldsymbol{r}'}{|\boldsymbol{r}-\boldsymbol{r}'|}.$$

The integration is carried out over the volume V the points of which can be reached from the point $\boldsymbol{r}$ by straight lines without intersecting the sphere; the argument of the integrals J_n is $|\boldsymbol{r}-\boldsymbol{r}'|$; the expressions for ν_0 and $\boldsymbol{W}_0$ have the following form:

$$\left. \begin{aligned}
\nu_0 &= \int_{\omega_0} \{(1-\alpha_{\mathrm{m}})[AJ_2 + (\boldsymbol{\Omega}_0\cdot\boldsymbol{\tau})BJ_3] - 2(\boldsymbol{\Omega}_0\cdot\boldsymbol{u}_\infty)J_3\} \, \mathrm{d}\omega, \\
\boldsymbol{W}_0 &= \int_{\omega_0} \{(1-\alpha_{\mathrm{m}})[AJ_3 + (\boldsymbol{\Omega}_0\cdot\boldsymbol{\tau})BJ_4] - 2(\boldsymbol{\Omega}_0\cdot\boldsymbol{u}_\infty)J_4\} \boldsymbol{\Omega}_0 \, \mathrm{d}\omega,
\end{aligned} \right\} \tag{9}$$

where $\boldsymbol{\Omega}_0 = \boldsymbol{r}-\boldsymbol{r}_0/|\boldsymbol{r}-\boldsymbol{r}_0|$; $\boldsymbol{r}_0$ is the value of the radius vector on the sphere surface; ω_0 is the solid angle under which the sphere is seen from the point $\boldsymbol{r}$; the argument of the J_n is $|\boldsymbol{r}-\boldsymbol{r}_0|$; $\boldsymbol{n}$, $\boldsymbol{\tau}$ are the normal and tangential to the sphere surface unit vectors. The set of integral equations (8) was completed by two integral relations obtained from (6) and (7):

$$\left. \begin{aligned}
\frac{1}{2}\int_S \left[\frac{A}{\pi^{\frac{1}{2}}} + u_\infty \cos\theta_0\right] \mathrm{d}s &= \pi^{-\frac{3}{2}} \int_{V_0} \mathrm{d}\boldsymbol{r} \int_{\omega_0} [\nu J_2 - 2(\boldsymbol{\Omega}_0\cdot\boldsymbol{W})J_3] \, \mathrm{d}\omega, \\
\frac{1}{2\pi^{\frac{1}{2}}}\int_S \left[\frac{B}{2(1-\alpha_\tau)} + u_\infty \sin\theta_0\right] \mathrm{d}s &= -\pi^{-\frac{3}{2}} \int_{V_0} \mathrm{d}\boldsymbol{r} \int_{\omega_0} [\nu J_3 - 2(\boldsymbol{\Omega}_0\cdot\boldsymbol{W})J_4](\boldsymbol{\tau}\cdot\boldsymbol{\Omega}_0) \, \mathrm{d}\omega,
\end{aligned} \right\} \tag{10}$$

where the integration on the left-hand side of the equations is performed over the whole sphere surface; θ_0 is the polar angle of the problem (figure 1); V_0 is the whole volume around the sphere.

For the purpose of subsequent analysis it is convenient to write the set of integral equations (8) in the symmetrized matrix form

$$\boldsymbol{\Psi} = \sigma\boldsymbol{\Psi} + \boldsymbol{S}, \tag{11}$$

where

$$\boldsymbol{\Psi} = \begin{bmatrix} \nu \\ \frac{1}{\sqrt{2}} G_r \\ \frac{1}{\sqrt{2}} G_\theta \end{bmatrix}, \quad \boldsymbol{S} = \pi^{-\frac{3}{2}} \begin{bmatrix} \nu_0 \\ \sqrt{2}\, W_{0r} \\ \sqrt{2}\, W_{0\theta} \end{bmatrix}, \quad \boldsymbol{G} = 2\boldsymbol{W}, \quad \sigma\boldsymbol{\Psi} = \pi^{-\frac{3}{2}} \int_V \boldsymbol{L}\boldsymbol{\Psi} \frac{\mathrm{d}r'}{|r-r'|^2},$$

$$\boldsymbol{L} = \begin{bmatrix} J_1 & \sqrt{2}\,\Omega_{r'} J_2 & \sqrt{2}\,\Omega_{\theta'} J_2 \\ \sqrt{2}\,\Omega_r J_2 & 2\Omega_{r'}\Omega_r J_3 & 2\Omega_{\theta'}\Omega_r J_3 \\ \sqrt{2}\,\Omega_\theta J_3 & 2\Omega_{r'}\Omega_\theta J_3 & 2\Omega_{\theta'}\Omega_\theta J_3 \end{bmatrix},$$

$$\Omega_{r'} = \boldsymbol{n}'\cdot\boldsymbol{\Omega}, \quad \Omega_r = \boldsymbol{n}\cdot\boldsymbol{\Omega}, \quad \Omega_{\theta'} = \boldsymbol{\tau}'\cdot\boldsymbol{\Omega}; \quad \Omega_\theta = \boldsymbol{\tau}\cdot\boldsymbol{\Omega},$$

$$\boldsymbol{n}' = \frac{\boldsymbol{r}'}{|\boldsymbol{r}'|}, \quad \boldsymbol{n} = \frac{\boldsymbol{r}}{|\boldsymbol{r}|}.$$

According to Hidy & Brock (1970), the force acting on the particle at rest is

$$\boldsymbol{F} = \sum_{\mp} \boldsymbol{n}_z\, m \int_S \mathrm{d}S \int_{(\boldsymbol{n}\cdot\boldsymbol{\Omega})\gtrless 0} V_z\, V_r\, f^{\mp}(R)\,\mathrm{d}\boldsymbol{v} = \boldsymbol{n}_z \int_S \mathrm{d}S(\sigma_{rr}\cos\theta_0 - \sigma_{r\theta}\sin\theta_0), \quad (12)$$

where $\quad \sigma_{rr} = \sum_{\mp} \int_{(\boldsymbol{n}\cdot\boldsymbol{\Omega})\gtrless 0} m V_r\, V_r\, f^{\mp}(R)\,\mathrm{d}\boldsymbol{v}, \quad \sigma_{r\theta} = \sum_{\mp} \int_{(\boldsymbol{n}\cdot\boldsymbol{\Omega})\gtrless 0} m V_r\, V_\theta\, f^{\mp}(R)\,\mathrm{d}\boldsymbol{v}$

are the normal and tangential stresses at the sphere surface respectively; $\boldsymbol{V} = (\boldsymbol{v} - \boldsymbol{U}_\infty)$ is the thermal velocity of the molecules; $\boldsymbol{n}_z$ is the unit vector along the axis Oz chosen along the direction $\boldsymbol{U}_\infty$.

Description of the force in the form (12) fully coincides with the definition accepted in fluid mechanics (Batchelor 1967).

Using the expression (12), the distribution-function form (1), the boundary condition (5) and the integral form of the kinetic equation (2), we obtain the following expression for the dimensionless ($F^* = F/(4l^2 P_\infty/\pi)$) drag:

$$F^* = \tfrac{16}{3}\pi^{\frac{1}{2}} R^2 u_\infty - \tfrac{1}{2}(1-\alpha_{\mathrm{m}}) \int_S \left[A\cos\theta_0 - \frac{B}{\pi^{\frac{1}{2}}}\sin\theta_0 \right]\mathrm{d}s$$

$$-2\pi^{-\frac{3}{2}} \int_{V_0} \mathrm{d}r \int_{\omega_0} \mathrm{d}\omega\, [\nu J_3 - 2(\boldsymbol{\Omega}_0\cdot\boldsymbol{W})\, J_4] \frac{(\boldsymbol{\Omega}_0\cdot\boldsymbol{u}_\infty)}{u_\infty}, \quad (13)$$

where $\mathrm{d}s = R^2 \sin\theta_0\, \mathrm{d}\theta_0\, \mathrm{d}\phi_0$ is the dimensionless sphere surface element.

4. Variational method

The set of integral equations (11) with additional conditions (10) was solved by the variational method developed for solving axisymmetric problems by Cercignani *et al.* (1968).

The essence of the method consists in constructing the bilinear functional

$$J(\tilde{A}, \tilde{B}, \boldsymbol{\Psi}) = (E\boldsymbol{\Psi}, \boldsymbol{\Psi} - \sigma\boldsymbol{\Psi} - 2\boldsymbol{S}) + \int_S [\beta_1 \tilde{A}^2 + 2\beta_2 \tilde{A} + \beta_3 \tilde{B}^2 + 2\beta_4 \tilde{B}]\,\mathrm{d}s, \quad (14)$$

where $\qquad\qquad \boldsymbol{E} = \begin{bmatrix} 1 & 0 & 0 \\ 0 & -1 & 0 \\ 0 & 0 & -1 \end{bmatrix}$

and the constants β_i are defined from the extremum condition of the functional J with independent variation of the functions $\tilde{A}$ and $\tilde{B}$.

The functional form (14) differs from the corresponding functional form in the work by Cercignani *et al.* (1968) because the conditions of phase changes and arbitrary accommodation of the tangential momentum on the particle surface are additionally taken into account.

It can be shown (Cercignani *et al.* 1968) that the stationary value of the functional (14) is directly connected with the reduced drag value

$$D = (8+\pi)^{-1}\left[8 - \frac{3J_{\text{stat}}}{2\pi^{\frac{1}{2}}u_{\infty}^2 R^2}\right],\tag{15}$$

where $D = F(\alpha_r, \alpha_m)/F_{\text{FM}}(\alpha_r = 1, \alpha_m = 0)$; F_{FM} is the free molecular drag value (equation (18)).

The problems pertaining to the features of the functional of type (14) and the estimation of lower and higher limits of the drag D have been discussed in the works by Cole (1981, 1982).

The trial functions $\tilde{A}, \tilde{B}, \varPsi$ in (14) were chosen so as to ensure the correct hydrodynamic limit of the solution (the same was done by Cercignani *et al.* 1968). Exact free-molecular values of the parameters to be determined are contained in the free members (9) of the integral equations (8).

It was shown that in the case of phase changes and incomplete accommodation of the momentum on the particle surface the trial functions can be chosen in the following form:

$$\varPsi = u_{\infty}\frac{R}{r}\begin{bmatrix} \dfrac{a_1}{r}\cos\theta \\[2ex] -\sqrt{2}\left[a_5 + a_2\left(1 - \dfrac{R^2}{r^2}\right)\right]\cos\theta \\[2ex] \dfrac{\sqrt{2}}{2}\left[a_5 + a_2\left(1 + \dfrac{R^2}{r^2}\right)\right]\sin\theta \end{bmatrix},\tag{16}$$

$$\tilde{A} = a_3 u_{\infty}\cos\theta_0,\quad \tilde{B} = a_4 u_{\infty}\sin\theta_0,$$

where a_i are constants chosen in such a way that the functional (14) attains a stationary value.

The angular dependence of trial functions (16) is exact (Cercignani *et al.* 1968). The choice of the dependence on r was based on the solution of a corresponding hydrodynamic problem obtained by the authors.

In this case the stationary value of the functional (14) may be written as

$$3J_{\text{stat}} = \sum_{i=1}^{5}\left[\alpha_{ii}a_i^2 - 2\alpha_i a_i + 2\sum_{k>i}^{5}\alpha_{ik}a_k\right].\tag{17}$$

In what follows the problem solution is reduced to calculation of the coefficients α_{ij}, α_i, which is a labour-consuming problem involving multiple integration of composite subintegral functions.†

5. Discussion of results

In limiting cases of free-molecular ($Kn \gg 1$) and viscous slip-flow ($Kn \ll 1$) regimes analytical expressions were obtained for the drag.

† As the algebraic expressions for the α_{ij} and α_i coefficients are rather cumbersome, they are not included in the present paper. Copies of these expressions together with the asymptotic values of α_{ij}, α_i at $R \gg 1$ may be obtained by writing to the Editor of J.F.M. or to the authors.

In the free-molecular limit

$$F_{\text{FM}} = \tfrac{16}{3}\pi^{\frac{1}{2}} \left(\frac{m}{2kT}\right)^{\frac{1}{2}} P_\infty R_0^2 \, U_\infty \left\{1 + \tfrac{1}{6}\pi(1-\alpha_{\text{m}}) \left[1 - \frac{4}{\pi}(1-\alpha_r)\right]\right\}. \tag{18}$$

Under corresponding assumptions (perfect tangential momentum accommodation and equality of the particle and vapour temperatures) this result coincides with the result of Brock (1964). For a non-volatile particle ($\alpha_{\text{m}} = 0$) at perfect accommodation ($\alpha_r = 1$) the well-known result (Epstein 1924) follows from (18). Note that (18) can be used at $Kn \gtrsim 10$, the error being within 5 %.

The expression for the drag in a viscous slip-flow regime has been obtained by asymptotic expansion of the coefficients from (17) and (15):

$$F_{\text{s}} = 6\pi\eta_{\text{g}} \, U_\infty \, R_0 [1 - a \, Kn], \tag{19}$$

where a is a numerical coefficient dependent on α_{m} and α_r in a complex manner. The formula (19) is valid with an accuracy of within 5 % at $Kn \lesssim 0.2$.

For a non-volatile particle ($\alpha_{\text{m}} = 0$) at $\alpha_r = 1$, $a = 1.1366$ which coincides with the result of Cercignani *et al.* (1968). For a volatile particle at $\alpha_{\text{m}} = 1$, $a = 1.4167$ which is in a good agreement (the discrepancy is within 0.4 %) with the results of Onishi (1977) and Sone & Aoki (1979) for the case when the heat conductivity of the particle considerably exceeds that of the vapour, and with the result of Kucherov & Rikenglaz (1960) (the discrepancy is within 2.7 %).

The dependence of a on the coefficients α_{m} and α_r is shown in table 1.

In what follows the brief outline of the physical mechanism of the effect of the phase changes on the drag is given. As shown by (12) the drag is determined by the normal and tangential stresses. The existence of the phase changes affects these components in different ways. From the boundary condition (4) it follows that the evenly evaporated molecules contribute to σ_{rr} but do not affect the $\sigma_{r\theta}$. Then, with the increase of α_{m}, the contribution to the force from $\sigma_{r\theta}$ decreases, owing to the increase of the evaporated molecules' share. Normal stresses σ_{rr} are less affected by the phase changes as the evaporated and the reflected molecules contribution to the normal momentum flow is approximately the same. Phase changes, therefore, reduce the drag owing to the decrease of the tangential stresses $\sigma_{r\theta}$. The minimum value of the drag is at $\alpha_{\text{m}} = 1$. At fixed value of α_{m} it is possible to analyse the dependence of the drag on the tangential momentum accommodation coefficient. It could be shown that α_r changes affect only $\sigma_{r\theta}$, but do not affect σ_{rr}. With the reduction of α_r the contribution to the force decreases, producing a drag reduction thereby. In this case the maximum value of the drag is at $\alpha_r = 1$. These qualitative inferences are valid for the whole range of Knudsen numbers.

Numerical calculations were carried out at intermediate Kn and various values of α_{m} and α_r. To calculate multiple integrals in α_{ij}, α_i the Korobov (1963) method was used. The results for $Kn \approx 0.09$ and 8.86 are presented in table 2, and those for $Kn \approx 0.22$, 0.44 and 0.89 are available from the editor or the authors on request.

It should be noted that calculation of α_{ij} and α_i with the required accuracy proved to be a labour-consuming procedure and required cumbersome transformations for 'improving' the subintegral function form. The calculation accuracy of triple integrals determines the accuracy of calculating coefficients α_{ij}, α_i and, finally, the drag calculation accuracy was not below 0.4 %. The calculation results and comparison with other theories are shown in figures 2 and 3.

The calculation results have shown a good agreement (the discrepancy does not exceed 2 %) with those obtained by Cercignani *et al.* for a non-volatile particle at

α_m \\ α_r	0	0.1	0.2	0.3	0.4	0.5	0.6	0.7	0.8	0.9	1.0
0.0	∞	19.160	9.178	5.863	4.219	3.244	2.604	2.159	1.836	1.599	1.421
0.1	19.141	9.684	6.319	4.602	3.566	2.879	2.396	2.041	1.775	1.574	1.421
0.2	9.140	6.300	4.731	3.742	3.064	2.576	2.211	1.933	1.717	1.550	1.421
0.3	5.807	4.562	3.721	3.117	2.667	2.320	2.048	1.833	1.661	1.526	1.421
0.4	4.140	3.505	3.021	2.643	2.343	2.100	1.902	1.740	1.609	1.503	1.421
0.5	3.140	2.793	2.508	2.272	2.074	1.909	1.770	1.654	1.558	1.481	1.421
0.6	2.472	2.282	2.116	1.973	1.849	1.742	1.651	1.573	1.509	1.459	1.421
0.7	1.996	1.896	1.806	1.727	1.657	1.595	1.542	1.498	1.463	1.437	1.421
0.8	1.640	1.595	1.556	1.521	1.490	1.464	1.444	1.429	1.419	1.417	1.421
0.9	1.362	1.354	1.348	1.345	1.345	1.347	1.353	1.363	1.376	1.396	1.421
1.0	1.137	—	—	—	—	—	—	—	—	—	1.417

TABLE 1. Dependence of coefficient a on values of α_m and α_r in (19). The given a-values were obtained numerically by comparison of (19) with the calculated drag values. The values $a(\alpha_m = 0, \alpha_r = 1) = 1.1366$; $a(\alpha_m = 1) = 1.4167$ were derived separately (and with a higher degree of accuracy)
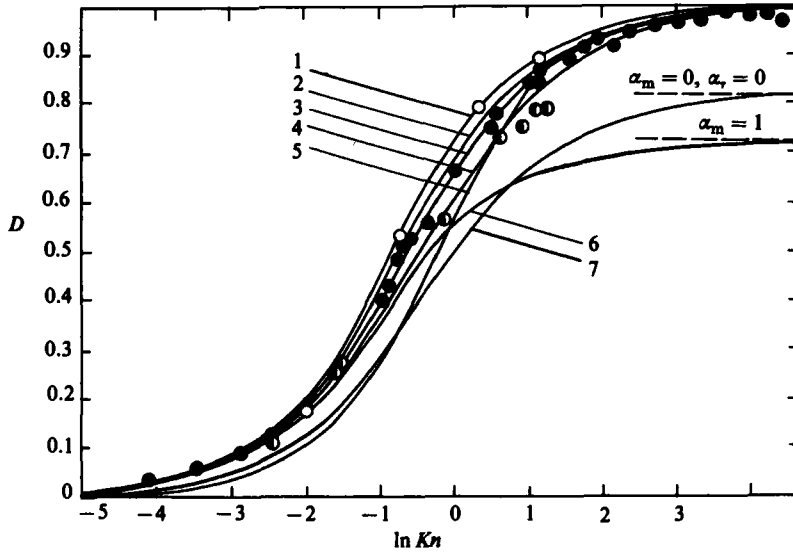
FIGURE 2. Drag D as a function of the Knudsen number, Curve 1, Phillips' (1975) result for the case of diffuse scattering of molecules; 2, Millikan's (1911) emperical formula; 3, the result of Cercignani *et al.* (1968); 4, Sherman's (1963) result; 5, Khlopkov's (1975) result for the case of diffuse scattering of molecules; 6, the result of this work at $\alpha_m = 0$, $\alpha_r = 0$; 7, the result of this work at $\alpha_m = 1$. Millikan's (1911, 1923) experimental data: ●, watch-oil drops in air; ◐, shellac particles in air; ○, mercury drops in air.
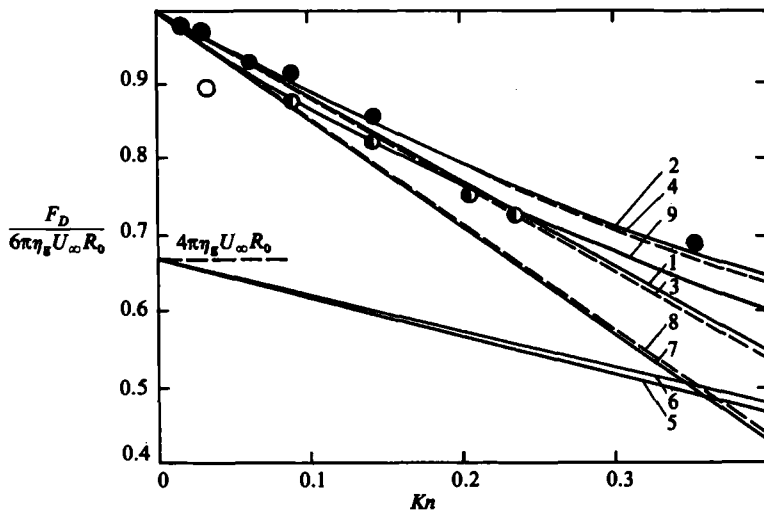

FIGURE 3. Drag at small Knudsen numbers. Non-volatile particles ($\alpha_m = 0$): curve 1, asymptotic expression for the drag (equation (19)) at $\alpha_r = 1$; 2, calculation results at $\alpha_r = 1$; 3, the result of Sone & Aoki (1977) with the first-order members for Kn taken into account; 4, the result of Sone & Aoki with the second-order members for Kn taken into account; 5, asymptotic expression from this work (equation (19)) at $\alpha_r = 0$; 6, calculation results at $\alpha_r = 0$. Volatile particles ($\alpha_m = 1$): curve 7, asymptotic expression (equation (19)); 8, the result of Onishi (1977) and of Sone & Aoki (1979); 9, calculation results. Millikan's (1911, 1923) experimental data: ●, oil drops in air; ◐, Shellac particles in air; ○, mercury drops in air.

α_m \ α_τ	0	0.1	0.2	0.3	0.4	0.5	0.6	0.7	0.8	0.9	1.0
0.0	0.08905	0.09820	0.1049	0.1100	0.1140	0.1172	0.1198	0.1218	0.1236	0.1250	0.1261
	0.6615	0.6678	0.6741	0.6806	0.6870	0.6935	0.7001	0.7067	0.7134	0.7201	0.7269
0.1	0.09865	0.1047	0.1094	0.1132	0.1163	0.1188	0.1208	0.1225	0.1240	0.1251	0.1261
	0.6982	0.7008	0.7035	0.7062	0.7090	0.7118	0.7147	0.7177	0.7207	0.7238	0.7269
0.2	0.1057	0.1098	0.1132	0.1159	0.1183	0.1202	0.1218	0.1232	0.1244	0.1253	0.1261
	0.7350	0.7359	0.7329	0.7319	0.7310	0.7302	0.7294	0.7297	0.7280	0.7274	0.7269
0.3	0.1112	0.1140	0.1163	0.1183	0.1200	0.1215	0.1228	0.1238	0.1247	0.1254	0.1261
	0.7718	0.7670	0.7622	0.7576	0.7530	0.7485	0.7440	0.7396	0.7353	0.7311	0.7269
0.4	0.1155	0.1174	0.1190	0.1204	0.1217	0.1227	0.1236	0.1244	0.1251	0.1257	0.1261
	0.8086	0.8001	0.7917	0.7833	0.7750	0.7668	0.7586	0.7506	0.7426	0.7347	0.7269
0.5	0.1190	0.1202	0.1213	0.1223	0.1231	0.1238	0.1245	0.1250	0.1255	0.1258	0.1261
	0.8454	0.8332	0.8210	0.8090	0.7970	0.7851	0.7933	0.7616	0.7499	0.7384	0.7269
0.6	0.1219	0.1227	0.1233	0.1239	0.1244	0.1249	0.1253	0.1256	0.1258	0.1260	0.1261
	0.8823	0.8663	0.8505	0.8347	0.8190	0.8034	0.7879	0.7725	0.7572	0.7420	0.7269
0.7	0.1244	0.1247	0.1251	0.1254	0.1256	0.1258	0.1260	0.1261	0.1262	0.1262	0.1261
	0.9191	0.8995	0.8799	0.8604	0.8410	0.8218	0.8026	0.7835	0.7645	0.7457	0.7269
0.8	0.1264	0.1266	0.1266	0.1267	0.1267	0.1267	0.1267	0.1266	0.1265	0.1263	0.1261
	0.9560	0.9326	0.9093	0.8861	0.8631	0.8401	0.8172	0.7945	0.7718	0.7493	0.7269
0.9	0.1282	0.1282	0.1280	0.1279	0.1278	0.1276	0.1274	0.1271	0.1268	0.1265	0.1261
	0.9929	0.9658	0.9388	0.9119	0.8851	0.8584	0.8319	0.8055	0.7791	0.7530	0.7269

TABLE 2. Calculated values of the drag D as a function of α_m and α_τ, at three intermediate values of Kn. The first of each group of two numbers refers to $R = 10$, $Kn \approx 0.09$ and the second to $R = 0.1$, $Kn \approx 8.86$.

Substance of the particle and the surrounding gas		a	α_τ
Watch oil (Millikan 1923)	air	1.23	0.95
	CO_2	1.18	0.98
	H_2	1.16	0.99
	He	1.29	0.93
Vacuum oil M300		1.49	0.85
Vacuum oil PH300		1.42	0.88
Vacuum oil PN200	Ar	1.46	0.86
Paraffin (Schmitt 1959)		1.28	0.93
Amorphous particles NaCl–Ar (Jacobsen & Brock 1965)		1.20	0.97
Tricresylphosphate (TCP)–air (Rosenblatt & La Mer 1946)		1.23	0.96

TABLE 3. Coefficient values a and α_τ, derived from experimental data

$\alpha_\tau = 1$. The drag curve constructed by the authors runs between the curves for D constructed by Millikan (1911) and by Cercignani *et al.* (1968) over the whole range of Kn (it is not shown in figure 2). It is obvious that the results obtained by Phillips (1975) at complete accommodation are systematically overestimated while those obtained by Sherman (1963) are underestimated compared with the results of this paper over the whole range of Kn. Khlopkov's (1975) result underestimates the D-values to a great extent (by 30 %) in the viscous slip-flow and transitional regimes.

A good agreement between the results obtained by different methods at small Kn is shown in figure 3. At $Kn \gtrsim 0.2$ the linear dependence of the drag on Kn ceases and correction for higher orders should be taken into account.

The analysis has shown that phase changes on the particle surface decrease the drag by values depending on Kn (figure 4). If at $Kn = 0.1$ the difference between the drag values at $\alpha_m = 0$ and $\alpha_m = 1$ (at $\alpha_\tau = 1$) does not exceed 3 % then in the transition regime this difference grows and in the free-molecular regime is $\approx 30\%$. Thus even in the case of motion in the saturated vapour it is permissible to speak about a considerable effect of volatility on the drag.

Unfortunately the authors are not aware of any experimental results for the measurements of the drag (or velocity) of an aerosol particle in its own saturated vapour. Such experimental data would be of a considerable interest both for theoretical investigations and for practical application.

In complete accommodation of the tangential momentum of the molecules colliding with the particle also leads to a drag decrease (figure 5). In the viscous slip-flow regime the calculation for a non-volatile ($\alpha_m = 0$) particle has confirmed the validity of the well-known result (Basset 1888) that at $Kn \ll 1$ and $\alpha_\tau \to 0$ the drag is

$$F_S = 4\pi\eta_g U_\infty R_0, \tag{20}$$

which is 1.5 times less than the drag at complete accommodation of the tangential momentum. Note that the moments solution of Phillips (1975) does not predict this kind of behaviour of the drag at $\alpha_\tau \to 0$.

At intermediate Kn the effect of α_τ decreases (figure 5) and at $Kn \to \infty$ the difference in the drag at $\alpha_\tau = 1$ and $\alpha_\tau = 0$ (at $\alpha_m = 0$) is about 20 %. Thus in the whole range of Kn there exists a pronounced dependence of the drag on the degree of accommodation of the tangential momentum of the molecules colliding with the
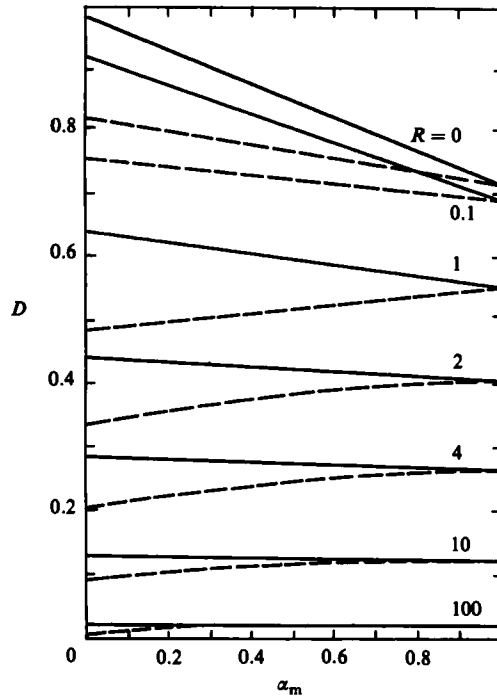
FIGURE 4. Drag as function of α_m for different Knudsen numbers. Solid lines correspond to the value $\alpha_r = 0.9$, dotted lines to $\alpha_r = 0$. Knudsen number $Kn = \frac{1}{2}\pi^{\frac{1}{2}}/R$.
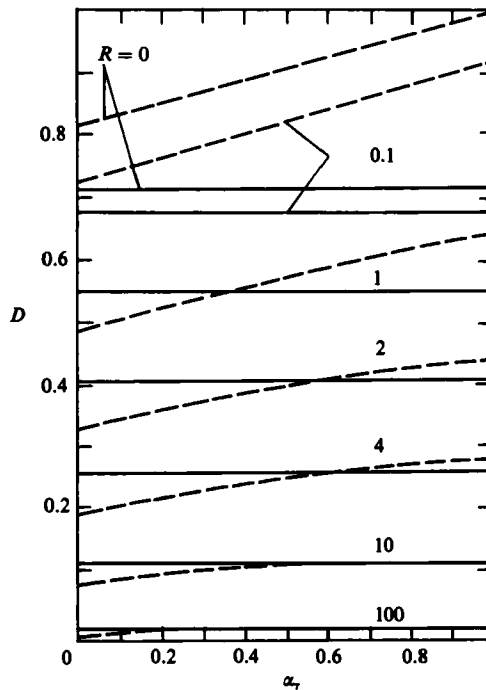

FIGURE 5. Drag as a function of α_r for different Knudsen numbers. Solid lines correspond to the value $\alpha_m = 1$, dotted lines to $\alpha_m = 0$. Knudsen number $Kn = \frac{1}{2}\pi^{\frac{1}{2}}/R$.

particle. This fact can be used for determination of the accommodation coefficients when comparing theory with experiment. The values a and α_r for different gas–substance pairs obtained after analysing the most reliable experimental data are given in table 3. The particle substances under experimental conditions were assumed to be low-volatile (which is confirmed in the works mentioned). The values of a and α_r were taken from the experimental data at $Kn \lesssim 0.1$.

It is well known that in the viscous slip-flow regime the isothermal slip coefficient (coinciding with coefficient a) depends only on α_r and does not depend on α_n. Therefore, all the discrepancies between the experimental data and theoretical predictions at perfect accommodation should be accounted or by the incomplete accommodation of the tangential momentum.

REFERENCES

ANNIS, B. K. & MALINAUSKAS, A. P. 1972 *J. Aerosol Sci.* **3**, 55.

BASSET, A. 1877 *Phil. Trans. R. Soc. Lond.* B **179**, 43.

BAKANOV, S. P. & VYSOTSKIY, V. V. 1982 *Kolloidn. Zh.* **44**, 1156.

BATCHELOR, G. K. 1967 *An Introduction to Fluid Dynamics.* Cambridge University Press.

BERESNEV, S. A. & CHERNYAK, V. G. 1983 *Inzhenerno Phys. Zh.* **44**, 142.

BHATNAGAR, P. L., GROSS, E. P. & KROOK, M. 1954 *Phys. Rev.* **94**, 511.

BROCK, J. R. 1964 *J. Phys. Chem.* **68**, 2862.

BROCK, J. R. 1967 *J. Colloid Interface Sci.* **24**, 344.

BROCK, J. R. 1980 *Aerosol Microphysics I* (ed. W. H. Marlow), p. 15. Springer.

CERCIGNANI, C. & PAGANI, C. D. 1968 *Phys. Fluids* **11**, 1395.

CERCIGNANI, C. & PAGANI, C. D. 1969 *Rarefied Gas Dynamics* (ed. L. Trilling & H. Y. Wachmann), Vol. 2, p. 269. Academic.

CERCIGNANI, C., PAGANI, C. D. & BASSANINI, P. 1968 *Phys. Fluids* **11**, 1399.

COLE, R. J. 1981 *Rarefied Gas Dynamics* (ed. S. Fisher), p. 1007. American Institute of Aeronaut. and Astronaut.

COLE, R. J. 1982 *13th Rarefied Gas Dynamics Symposium. Book of Abstracts*, Vol. 2, p. 394. Novosibirsk: Institute of Thermophysics.

DERYAGUIN, B. V. & BAKANOV, S. P. 1962 *Dokl. Akad. Nauk SSSR* **147**, 139.

EPSTEIN, P. S. 1924 *Phys. Rev.* **23**, 710.

FUCHS, N. A. 1971 *Topics in Current Aerosol Research* (ed. G. Hidy & J. Brock). Pergamon.

FUCHS, N. A. & STECHKINA, I. B. 1963 *Zh. Tech. Phys.* **33**, 132.

HIDY, G. & BROCK, J. R. 1970 *The Dynamics of Aerocolloidal Systems.* Pergamon.

JACOBSEN, S. & BROCK, J. R. 1965 *J. Colloid Sci.* **20**, 544.

KHLOPKOV, YU. I. 1975 *Uchenie Zapiski ZAGI* **6**, 174.

KOGAN, M. N. 1969 *Rarefied Gas Dynamics.* Plenum.

KOROBOV, N. M. 1963 *Theoretical Numerical Methods in an Approximating Analysis.* Moscow: Fizmatgiz.

KUCHEROV, R. YA. & RIKENGLAZ, L. E. 1960 *Dokl. Akad. Nauk SSSR* **133**, 1130.

LEA, K. C. & LOYALKA, S. K. 1982 *Phys. Fluids* **25**, 1550.

MARCHUK, G. I. 1961 *Methods of Calculations of Nuclear Reactors.* Moscow: Gosatomizdat.

MILLIKAN, R. A. 1911 *Phys. Rev.* **32**, 349.

MILLIKAN, R. A. 1923 *Phys. Rev.* **22**, 1.

ONISHI, Y. 1977 *J. Phys. Soc. Japan* **43**, 1434.

PHILLIPS, W. F. 1975 *Phys. Fluids* **9**, 1089.

ROSENBLATT, P. & LA MER, V. 1946 *Phys. Rev.* **70**, 385.

SCHMITT, K. H. 1959 *Z. Naturforshg.* **14a**, 870.

SHERMAN, F. S. 1963 *Rarefied Gas Dynamics* (ed. J. A. Laurmann), Vol. 2, p. 228. Academic.

SONE, Y. & AOKI, K. 1977 *Rarefied Gas Dynamics* (ed. J. L. Potter), Vol. 1, p. 417. American Institute of Aeronaut. and Astronaut.

SONE, Y. & AOKI, K. 1979 *Rarefied Gas Dynamics* (ed. R. Camparque), Vol. 2, p. 1207. Paris: CEA.

WALDMANN, L. & VESTNER, H. 1977 *Physica* **86A**, 303.