

Estimation of Brownian diffusion in thermophoretic sedimentation of aerosol onto a surface

D.D. Ryvkin and S.A. Beresnev

Ural State University, Ekaterinburg

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Brownian diffusion of aerosol particles near a sedimentation surface is considered. The generalized coefficient of Brownian diffusion is introduced and calculated. This coefficient depends on the problem's geometry, Knudsen numbers, and accommodation coefficient of gas molecules. The diffusion component of the speed of macroscopic drift of particles towards the surface is analyzed for different values of characteristic parameters using the thermophoretic mechanism as an example.

Introduction

Microphysical theory of aerosol transport and evolution (thermophoresis, diffusophoresis, photophoresis, and coagulation) in a boundless weakly nonequilibrium gaseous medium can be now considered as rather well developed.^{1,2} At the same time, there are significant gaps in understanding of mechanisms and qualitative description of aerosol sedimentation from a gaseous medium onto a surface. Sedimentation (or its prevention) plays an important part in many technological processes, in particular, in production of high-quality optical fibers for aerosol reactors, sedimentation of particles in plane-parallel and round channels (to develop highly efficient gas-purifying equipment), etc. Sometimes, to the contrary, the task is to prevent sedimentation of particles onto a surface from dusty gas flows (such a situation occurs in production of silicon blanks for microelectronics, in nanotechnologies, in motion of a dusty gas along pipe-lines).

It becomes evident that mathematical simulation of such applied problems and, especially, reliable estimation of the effects are impossible without detailed gas-kinetic analysis of the problem. At the same time, most significant mechanisms of non-gas-kinetic nature that accompany sedimentation of aerosol particles should be taken into account as well. Among these mechanisms, the Van der Waals attraction forces between a particle and a surface at a very small distance between them and Brownian diffusion of particles, which is very significant at normal atmospheric conditions for particles of micron and submicron size being near the sedimentation surface, deserve attention in the first turn.

In this paper, the phenomenon of Brownian diffusion is considered using thermophoretic sedimentation as an example. The significance of this phenomenon for different flow conditions and thermal properties of a particle and gas is evaluated.

1. Generalized coefficient of Brownian diffusion

According to the first Fick's law, the flow of particles through a surface perpendicular to the OZ axis is written in the form $j = -D\partial c/\partial z$, where D is the coefficient of Brownian diffusion, and c is concentration of particles. According to the second Fick's law, $\partial c/\partial t = D\partial^2 c/\partial z^2$, under the assumption that the concentration varies in space only along the OZ axis. The coefficient of Brownian diffusion can be calculated in the following way. Let diffusion occur in the OZ direction. Let us consider a straight cylinder whose section equals unity, the axis is parallel to OZ , the lower base coordinate is z , and the upper base coordinate is $z + \Delta z$. Thus, the cylinder volume is Δz . Diffusion of particles in the cylinder generates osmotic forces acting on its bases. The osmotic pressure of a dissolved substance in a solvent (in our case, particles in a gas) is $p = ckT$, where k is the Boltzmann constant, and T is temperature. Assuming that the temperature is constant within the cylinder, let us write the pressure gradient in the form $\partial p/\partial z = kT\partial c/\partial z$. This pressure gradient with the opposite sign is equal to the osmotic force acting upon all particles in the cylinder and referred to unit volume, i.e., $-F_{\text{osm}}/\Delta z$. On the other hand, the osmotic force is equal to the resistance force acting on every particle in the cylinder multiplied by the number of particles: $F_{\text{osm}} = F_{\text{z}}^D c \Delta z$. Let us write the resistance force acting from the gas to a particle moving with the speed U as $F_{\text{z}}^D = \beta U$, where β accounts for the problem geometry, the conditions of flow by the Knudsen number, and the degree of accommodation of gas molecules on the surface. Then $\partial p/\partial z = kT\partial c/\partial z = -\beta c U$. However, $J = cU$ is the diffusion flow along the OZ axis. According to the first Fick's law, we obtain the coefficient of Brownian diffusion

$$D = kT/\beta. \quad (1)$$

2. Rate of diffusion sedimentation

Let us consider stationary diffusion from a zone with constant concentration $c = c_0$ towards a wall on whose surface $c = 0$. According to the first Fick's law, the flow of settling particles to the wall is $J = Dc_0/a$, where a is the distance from the zone with the constant concentration to the wall. Thus, *the rate of stationary diffusion sedimentation of a particle* can be estimated as

$$U_{\text{dif}} = D/a. \quad (2)$$

For non-stationary diffusion, using the second Fick's law with the corresponding initial and boundary conditions, we obtain *the non-stationary diffusion rate* for a particle near the wall at $t > 0$

$$U_{\text{dif}}(a, t) = \left(\frac{D}{\pi t}\right)^{1/2} \exp\left(-\frac{a^2}{4Dt}\right) / \operatorname{erf}\left(-\frac{a}{(4Dt)^{1/2}}\right). \quad (3)$$

This expression asymptotically tends to $U_{\text{dif}} = D/a(1 - a^2/4Dt)$ as $t \rightarrow \infty$ what corresponds to the estimate (2). The equation of diffusion of aerosol particles contained between two walls spaced by the distance L from each other at the same initial and boundary conditions has the solution³

$$c(z, t) = \frac{4c_0}{\pi} \sum_{n=0}^{\infty} \exp\left(-\frac{(2n+1)^2\pi^2Dt}{L^2}\right) \times \\ \times \frac{1}{2n+1} \sin\left[(2n+1)\frac{\pi z}{L}\right].$$

As $t \rightarrow \infty$, the decisive part is played by the first term of the series $c^{(0)}(z, t) = \frac{4c_0}{\pi} \exp\left(-\frac{\pi^2Dt}{L^2}\right) \times \sin\left(\frac{\pi z}{L}\right)$, and the diffusion flow at the distance a from the wall $z = 0$ is equal to

$$J^{(0)}(a, t) = \frac{4Dc_0}{L} \exp\left(-\frac{\pi^2Dt}{L^2}\right) \cos\left(\frac{\pi a}{L}\right), \quad (4)$$

wherefrom the diffusion rate of an aerosol particle between plates is

$$U_{\text{dif}}(a) = \left(\frac{\pi D}{L}\right) \cotan\left(\frac{\pi a}{L}\right). \quad (5)$$

If $a \ll L$, this expression transforms into Eq. (2), i.e., it corresponds to the stationary case in the presence of a single plate.

3. Main conclusions

Equation (1) for the coefficient of Brownian diffusion includes the coefficient β accounting for the dependence of the resistance force on the distance

between the particle and the plane, particle size, flow conditions, and accommodation properties. The ratio of diffusion and thermophoretic sedimentation rates $\gamma = U_{\text{dif}}/U_T$ is interesting for evaluation of the significance of Brownian diffusion. In particular, under viscous conditions with sliding, using the results of, for instance, Ref. 4, we obtain

$$\gamma = \frac{kT_0}{a} \frac{\rho T_0(2 + \Lambda)}{9\pi\eta^2 R |\nabla T|} \frac{1}{\Psi(\alpha, \Lambda)}, \quad (6)$$

where $a/R = \cosh \alpha$, η is gas viscosity, ρ is gas density, Λ is the thermal conductivity ratio of the particle and gas, $|\nabla T|$ is the absolute value of the external temperature gradient, and $\Psi(\alpha, \Lambda)$ is the function tabulated in Ref. 4. One can see that the part of Brownian diffusion increases with increasing pressure, temperature, and molecular mass of the gas and decreasing viscosity.

Brownian diffusion is more significant for small (submicron) particles as they approach the surface ($a \sim R$). Let us consider an example. Let $T_0 = 300$ K, $\rho = 1$ kg/m³, $\eta = 10^{-3}$ kg/m·s, $|\nabla T| = 10^3$ K/m, $R = 0.1$ μm , $a = 2R$. Then $\gamma = 2.2(2 + \Lambda)/[\Psi(\Lambda, \alpha \approx 1.25)]$. For such particles, the rate of Brownian diffusion to the surface far exceeds that of thermophoretic sedimentation near the wall.

Under free-molecule conditions,⁵ the coefficient is

$$\gamma = \frac{kT_0}{a} \frac{15}{32R^2\lambda_g |\nabla T|} \left(\frac{8kT}{\pi m}\right)^{1/2}. \quad (7)$$

Here λ_g is thermal conductivity of the gas and m is mass of a gas molecule. For estimation, let us take $T_0 = 300$ K, $\lambda_g = 10^{-2}$ W/m K, $|\nabla T| = 10^3$ K/m, $R = 0.1$ μm , $a = 100R$. Then for a gas with molar mass $M = 0.016$ kg/mole, we obtain $\gamma \approx 4$. Thus, under the free-molecule conditions at a sufficient distance from the wall, Brownian diffusion plays an important part for submicron particles as well. However, since $\gamma \approx 1/r^2$, the influence of diffusion decreases quickly with increasing particle radius and almost vanishes for $R \approx 1$ μm .

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