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Note on the Correction for Diffusion and Drag in the Slip Regime

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Note on the Correction for Diffusion and Drag in the Slip Regime

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ABSTRACT. We present a new equation to describe diffusion, hence drag, of particles in the slip regime that is simple, accurate, and useful.

The purpose of this note is to describe a simple formula for the diffusion coefficient, and hence the drag coefficient, of a particle that applies over the entire Knudsen number range including the slip regime. The Knudsen number Kn is the ratio of the medium molecule mean free path λ to the radius of the particle R , $Kn = \lambda/R$.

The proposed formula is based on the known limits for the diffusion coefficient at $Kn = 0$ and $Kn \rightarrow \infty$. At $Kn = 0$, the continuum limit, the Stokes–Einstein equation applies (Friedlander 1977):

$$D_{SE} = \frac{kT}{6\pi\eta R}. \quad (1)$$

In Equation (1), k is Boltzmann's constant, T is the absolute temperature, and η is the shear viscosity. At $Kn \rightarrow \infty$, the free molecular limit, the Epstein equation applies (Friedlander 1977):

$$D_{Ep} = \frac{3}{8\rho R^2(1 + \alpha\pi/8)} \left(\frac{mkT}{2\pi} \right)^{1/2}. \quad (2)$$

In Equation (2), ρ is the medium mass density, m is the medium molecule mass, and $0 \leq \alpha \leq 1$ is the accommodation coefficient.

We have found that a convenient formula for the diffusion coefficient that applies at all Kn is simply the sum of Equations (1) and (2), viz.,

$$D = D_{SE} + D_{Ep}. \quad (3)$$

Equation (3) is the new idea in this note. This sum can be algebraically manipulated to yield physically appealing forms. To do so, use the relation between the mean free path and the viscosity (Kennard 1938; Allen and Raabe 1982)

$$\eta = \phi\rho\bar{u}\lambda, \quad (4)$$

where the mean speed of the medium molecules is

$$\bar{u} = (8kT/\pi m)^{1/2}, \quad (5)$$

and $\phi = 0.491(1 + \varepsilon)$, where $\varepsilon \ll 1$ (Kennard 1938). For elastic hard sphere molecules with no repulsive force, $\varepsilon = 0.016$ and thus $\phi = 0.499$. As the repulsive molecular field between molecules increases, ε tends to zero and ϕ tends to 0.491. This value is more realistic (Allen and Raabe 1982). Combining Equations (1)–(5) yields

$$D = \frac{kT}{6\pi\eta\lambda}(Kn + AKn^2) \quad (6)$$

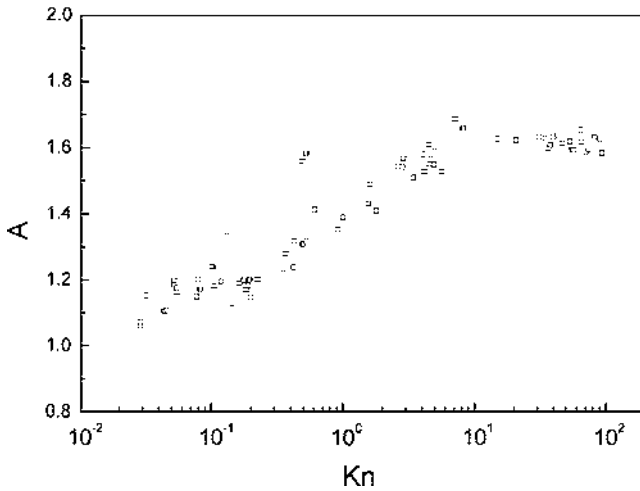


FIGURE 1. The coefficient A as determined from Millikan's data plotted versus the Knudsen number.

or

$$D = \frac{kT}{6\pi\eta R}(1 + AKn), \quad (7)$$

where

$$A = 2.2455/(1 + \alpha\pi/8). \quad (8)$$

This simple formulation applies to the drag on a particle as well, since the drag coefficient f and diffusion are related by the Einstein relation (Friedlander 1977)

$$f = kT/D. \quad (9)$$

The atavistic Equation (7) harkens back to the correction for slip proposed by Cunningham (1910) for the Stokes drag term, $f = 6\pi\eta R$ corrected to $6\pi\eta R/C(Kn)$, where

$$C(Kn) = 1 + AKn. \quad (10)$$

This is identical to the correction in Equation (7). Knudsen and Weber (1911) realized that A was not constant and proposed

$$A(Kn) = \alpha + \beta \exp(-\gamma/Kn). \quad (11)$$

The combination of Equations (10) and (11) is usually called the "Cunningham Correction Factor," and empirical values of α , β , and γ , such as those given by Allen and Raabe (1982) and Buckley and Loyolka (1989), have been determined from the measurements of Millikan (1923a, 1923b).

In Figure 1 we plot Millikan's A versus Kn taken from Allen and Raabe (1982). We see that A varies from ca. 1.1 to 1.6 as Kn increases from zero. Thus the results of Equations (6) and (7) in which A is constant is in error. Moreover, given the work of Cunningham from long ago, our work appears to be a step backward. Despite this, we advocate a constant A and the forms of Equations (6) and (7) not only because of their simplicity and physical esthetics, but also because of their accuracy and usefulness, as we describe below.

Regarding accuracy, in Figure 2 we plot the correction factor $C(Kn) = 1 + AKn$ of Equation (10) vs. Kn and compare this to Millikan's data. The value of $A = 1.612$ was used as determined from Equation (8) and an accommodation coefficient of $\alpha = 1.0$. This figure shows that the forms of Equations (6) and (7) are quite accurate with at most a 10% error near $Kn \approx 0.5$. Small, reasonable changes in α do not change this conclusion. The error is comparable to other uncertainties endemic in aerosol transport and kinetics studies, hence specification of the drag correction factor to better than 10% is rarely warranted.

The simplicity of Equations (6) and (7) makes them useful. In our work using dynamic light scattering to determine aerosol particle sizes (Olivier et al. 1992; Cai and Sorensen 1994;

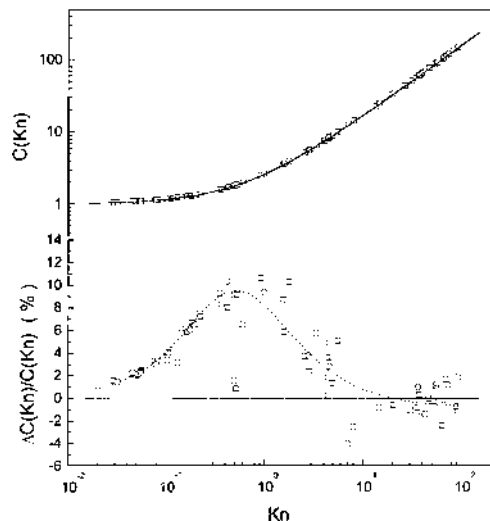


FIGURE 2. a) The line is the correction factor $1 + AKn$, where $A = 1.612$, as proposed in Equation (7) versus the Knudsen number plotted with Millikan's data (square symbols). b) Deviation of the correction factor $1 + AKn$, where $A = 1.612$, and the data of Millikan. The dashed line is the standard Cunningham correction formula, Equations (10) and (11) with $\alpha = 1.155$, $\beta = 0.471$, and $\gamma = 0.596$ from Allen and Raabe (1982).

Wang and Sorensen 1999a), we have found that inversion of Equation (7) with the experimentally determined diffusion coefficient D to calculate the particle size R involves a simple quadratic equation. This is in contrast to inversion of Equations (10) and (11), the standard Cunningham correction, which involves a complex transcendental equation.

In other work involving aggregation kinetics (Wang and Sorensen 1999b), we have found Equation (6) or (7) much easier to use than the standard Cunningham correction formula for both data analysis and scaling analysis. In particular, scaling analysis of aggregation takes advantage of the algebraic homogeneity of many physical aggregation kernels to predict scaling or self-preserving size distributions as solutions to the Smoluchowski equation which governs aggregation (Friedlander 1977; van Dongen and Ernst 1985). Furthermore, the degree of homogeneity determines the temporal functionalities of the aggregation. Use of the Cunningham cor-

rected diffusion coefficient yields a very nonhomogeneous aggregation kernel which does not allow physical insight regarding a scaling solution or the dynamics. On the other hand, the proposed Equation (6), while not homogeneous, is the sum of two homogeneous functions, and this allows for physical insight into both the scaling of the size distribution and the dynamics. We will make use of this in a future publication involving aggregation kinetics.

A final benefit of the formalism we have presented here is in regard to the definition of the Knudsen number for nonspherical and aggregate particles. The problem for these particles is that their radius is ill defined, hence ambiguous, in the definition of $Kn = \lambda/R$. Now, however, use of Equations (1)–(3) forces use of the mobility radius R_m defined as the radius of a solid sphere that has the same diffusion coefficient as the nonspherical particle or aggregate. Then to proceed with the derivation from Equation (3) to Equations (6) and (7) one must use

$$Kn = \lambda/R_m. \quad (12)$$

In the past we have used $Kn = \lambda/R_g$, where R_g is the aggregate radius of gyration for fractal aggregates (Wang and Sorensen 1999a) but in light of the argument above, we believe Equation (12) is better.

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