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The aerosol particle collision kernel considering the fractal model of particle motion

Lech Gmachowski *

Warsaw University of Technology, Institute of Chemistry, 09-400 Płock, Poland

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ABSTRACT

A method is presented in which the fractal model of particle motion, distinct from the Langevin equation of motion, has been used to determine the collision kernel of particles. The collision kernels, valid in the continuum and free molecular regions, are reformulated to be dependent on one sixth of the mean square displacement of a particle divided by time instead of diffusion coefficient and the root mean square displacement divided by time instead of the mean velocity. One generalized formula for collision kernels in the transition regime is obtained by equating the two expressions for kernels. The approaching time is calculated using equation describing the particle trajectory. For condensation and monodisperse aggregation the calculated collision kernel is the harmonic average of limiting kernels valid in continuum and free molecular regimes. It is in agreement with the experimentally verified formula of Fuchs–Sutugin. The results for polydisperse aggregation are close to those obtained by the widely accepted method of Fuchs. The obtained formula can also be used to approximate the drag force on particles in the transition regime.

1. Introduction

The growth of aerosol particles by collisions either between particles (aggregation) or particles and vapor molecules (condensation) changes the particle size distribution and plays an important role in a number of aerosol phenomena and processes, including new particle formation in the atmosphere (Leppä et al., 2011) and nanomaterials synthesis (Buesser & Pratsinis, 2012). The behavior of these systems is governed by the collision rate which for low volume fractions of particles and vapor molecules is given (Friedlander, 2000) as the product of the collision kernel k and the number concentrations of colliding particles n_i and n_j

$$R = kn_in_j \tag{1}$$

The form of the aggregation kernel depends on the region where the process occurs. In the continuum region the kernel is proportional to the product of the sum of diffusion coefficients of aggregating particles and the sum of their radii, according to Smoluchowski theory (Smoluchowski, 1916)

$$k_{diff} = 4\pi(a_i+a_j)(D_i+D_j) \tag{2}$$

Veshchunov (2010, 2012) and Veshchunov & Azarov (2012) consider the applicability of the formula describing particle–particle collisions (Eq. (2)). They show it is relevant for coalescence of small particles with large ones, but for

* Tel.: +48 24 262 36 91.
E-mail address: gmachowski@poczta.onet.pl

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Nomenclature

a	particle radius (m)
D	diffusion coefficient (m^2/s)
f	translational friction factor of the particle (kg/s)
k	collision kernel in transition region (m^3/s)
k_{ball}	collision kernel in free molecular region (m^3/s)
k_{diff}	collision kernel in continuum region (m^3/s)
k_B	Boltzmann constant ($m^2\ kg\ s^{-2}\ K^{-1}$)
Kn_D	diffusive Knudsen number (–)
m	particle mass (kg)
$\langle r^2 \rangle$	mean square displacement of the particle position in three dimensions (m^2)
T	temperature (K)
t	approaching time (s)
u	mean velocity of the particle (m/s)
λ	particle mean free path (m)
τ	momentum relaxation time (s)

equal sized particles applies only in the limit in which the particle mixing time is substantially faster than the collision time, such that concentration gradients do not develop between particles. This takes place, however, for sufficiently dilute systems, in which the particle radius is much smaller than the inter-particle distance.

The free molecule aggregation kernel can be expressed as (Seinfeld, 1986)

$$k_{ball} = \pi(a_i + a_j)^2 \sqrt{u_i^2 + u_j^2} \tag{3}$$

Their mutual relation can be expressed by the formula

$$\frac{k_{ball}}{k_{diff}} = \frac{a_j u_i}{4D_i} \frac{(1 + a_i/a_j)\sqrt{1 + u_j^2/u_i^2}}{1 + D_j/D_i} \tag{4}$$

The mean thermal velocity of the particle, which arises when deriving the ballistic collision kernel considering the full Maxwell–Boltzmann distributions of the colliding particles (Vincenti & Kruger, 1975; Allen, 1992) is described by the equation

$$u_i = \sqrt{(8/\pi)k_B T/m_i} \tag{5}$$

which corresponds, via a set of equations ($D_i = k_B T/f_i$; $f_i = m_i/\tau_i$; $\tau_i = \lambda_i/u_i$) to the diffusive mean free path

$$\lambda_i = (8/\pi)D_i/u_i \tag{6}$$

Using Eqs. (2) and (3), the ratio of collision kernels can be expressed as dependent on the diffusive Knudsen number

$$\frac{k_{diff}}{k_{ball}} = \frac{\pi}{2} Kn_D \tag{7}$$

where

$$Kn_D = \frac{1 + D_j/D_i}{(1 + a_i/a_j)\sqrt{1 + u_j^2/u_i^2}} \frac{\lambda_i}{a_j} \tag{8}$$

The diffusive Knudsen number takes different form dependent on the ratio of particle radii. For

$$a_i = a_j \quad Kn_D = \frac{1}{\sqrt{2}} \frac{\lambda}{a} \tag{9}$$

whereas for

$$a_j \gg a_i \quad Kn_D = \frac{\lambda_i}{a_j} \tag{10}$$

At a constant temperature, according to Eqs. (5) and (6), the diffusive mean free path of a particle is proportional to its diffusion coefficient and to square root of its mass (Dahneke, 1983). The non-fractal particle growth causes the increase of the square root of its mass ($\sqrt{m_i} \sim a_i^{3/2}$) and the reduction of the diffusion coefficient in the slip region ($D_i \sim a_i^{-2}$), so the reduction of the diffusive mean free path ($\lambda_i \sim a^{3/2-2}$) and the diffusive Knudsen number ($Kn_D \sim a_i^{3/2-3}$). For larger particles $D_i \sim a_i^{-1}$ and $Kn_D \sim a_i^{3/2-2}$. For high diffusive Knudsen numbers the aggregation can be approximated by the ballistic model. As the aggregation proceeds Kn_D diminishes. The continuum approximation is adequate for $Kn_D \cong 0$ which corresponds to negligible diffusive mean free path.

In the transition region neither ballistic nor diffusive model can describe the collision kernel since the diffusive Knudsen number is of the order of unity for atmospheric submicrometer particles. Therefore some trials were undertaken to solve the problem.

Fuchs (1934, 1959) first proposed a model, in which a vapor molecule moves diffusively if is far from the particle but starts to move ballistically when the distance becomes less than a critical value. This approach results in an analytical