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The unipolar diffusion charging of arbitrary shaped aerosol particles

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ABSTRACT

The unipolar diffusion charging of particles, i.e. the net increase in particle charge through ion–particle collisions, is an important process in a number of aerosol systems. Accurate methods are hence needed to predict the unipolar charging rate, not only for spherical particles, but also particles of arbitrary geometry. In this work, the unipolar charging (described by the particle–ion collision kernel) of conducting, arbitrary shaped particles is studied theoretically. Through a combination of dimensional analysis, Brownian dynamics (BD), and molecular dynamics (MD), the collision kernel is found to be described accurately by a simple-to-use expression across the entire diffusive Knudsen number Kn_D range (from the continuum regime to the free molecular regime), where Kn_D is the ratio of the ion mean persistence path to a well-defined particle length scale (proportional to the ratio of orientationally averaged projected area PA to the Smoluchowski radius R_s). In the developed collision kernel expression, the effect of repulsive Coulomb and attractive image potential interactions between the ion and the particle are parameterized by the coulomb potential energy to thermal energy ratio, ψ_E, and image potential energy to thermal energy ratio, ψ_I. It is found that the changes in collision rates due to potential interactions in the continuum ($Kn_D \to 0$) and free molecular ($Kn_D \to \infty$) regimes collapse to particle geometry independent functions, expressed in terms of ψ_E and ψ_I. In the transition regime, the dimensionless collision kernel H is shown to be geometry independent, and is a function of a suitably defined Kn_D only. Comparison is made between the predictions of the proposed expression and the flux matching model of Fuchs; for non-spherical particles, theories available in the literature are examined and commented upon. Finally, sample calculations of the mean charge acquired by selected particle geometries are presented and discussed.

1. Introduction

In unipolar diffusion charging, i.e. aerosol particle collisions with gas phase ions of a single polarity, knowledge of the collision rate between nanoparticles and ions (the charging rate, defined as the number of collisions per unit volume per unit time, with assumed charge transfer from ion to particle upon collision) is paramount, as it is this rate which ultimately determines the charge distribution of nanoparticles upon timed exposure to ions of a given concentration. The charging rate, $R_{p,i}$, for particles with p number of excess charges can be calculated from binary reaction kinetics as

$$R_{p,i} = \beta_{p,i} n_p n_i \tag{1a}$$

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where n_p and n_i are the particle and ion number concentrations, respectively, and $\beta_{p,i}$ is the collision rate coefficient/kernel. Correspondingly, the change in number concentration of particles with p charges over time is described by the equation:

$$\frac{dn_p}{dt} = R_{p-1,i} - R_{p,i} = n_i(\beta_{p-1,i} n_{p-1} - \beta_{p,i} n_p) \tag{1b}$$

for positive ions, with the subscripts $p-1$ replaced with $p+1$ for negative ions. Evident in Eqs. (1a) and (1b), proper collision kernel evaluation is thus required for both charging rate and charge distribution calculations. Collision kernels have been predominantly determined using the flux matching approach of Bricard (1962) and Fuchs (1963). A number of aspects of the charging process are captured by flux matching based analyses, including the combined influences of the Coulomb and image potential, as well as the fact that particle–ion collisions may occur in different transport regimes. The transport regime in which gas-phase entities migrate and collide with one another is determined by the ratio of the colliding entities' persistence distance to a characteristic length scale for the collision. While Fuchs and earlier researchers did not explicitly name this ratio, later authors have referred to the persistence distance to collision length scale ratio as the diffusive Knudsen number, Kn_D (Dahneke, 1983; Pierce et al., 2006; Zurita-Gotor & Rosner, 2002), a term which is also adopted here. In the continuum limit, in which the mean persistence distance is substantially smaller than the collision length scale ($Kn_D \to 0$), moving entities behave diffusively with negligible inertia during collision events, and the collision kernel takes the form of Smoluchowski's hard sphere collision kernel (Chandrasekhar, 1943) augmented by an enhancement factor for the potential between colliding entities (Fuchs, 1963). In the opposing, free molecular limit ($Kn_D \to \infty$), entity motion is inertially driven and ballistic, and the collision kernel can be derived from kinetic theory (Allen, 1992; Mott-Smith & Langmuir, 1926). Flux matching theory is specifically developed for collision kernel calculation in the intermediate Kn_D range, which it accomplishes by treating ion motion as diffusive when the ion and particle are sufficiently far from one another, and treating motion as entirely ballistic when the ion is a distance less than the radius of a predefined "limiting sphere" from the particle center.

In the absence of potentials, with an appropriate limiting sphere radius (Wright, 1960) the predictions of flux matching theory agree extremely well with more rigorous derivations of the collision kernel in the transition regime (Loyalka, 1973; Sahni, 1966; Takata et al., 1998). Furthermore, the modifications to flux matching theory proposed by Hoppel & Frick (1986) do not alter its predictions for unipolar charging, wherein the particle is either uncharged or is of the same polarity as the ion. Nonetheless, improvements can be made to flux matching theory derived collision kernels for unipolar charging; in Fuchs's flux matching approach at the point when an ion approaches the limiting sphere, the ion's speed is equated with the mean thermal speed, leading to incorrect collision kernel evaluation when the potential interactions between ion and particle are significantly larger than the background thermal energy (Filippov, 1993; Gopalakrishnan & Hogan, 2012; López-Yglesias & Flagan, 2013). Moreover, while in earlier studies selected nonspherical particle collisions with ions have been analyzed close to the $Kn_D \to 0$ and $Kn_D \to \infty$ limits (Han & Gentry, 1993; Han et al., 1991; Mayya, 1990; Wen et al., 1984), and Biskos et al. (2004) applied a flux matching-like approach to simulate the unipolar charging of selected nonspherical particles, no simple-to-implement expression for the unipolar charging of arbitrary shaped particles under all possible background gas conditions is hitherto available.

The purpose of this work is hence to develop an expression for the collision kernel for the unipolar diffusion charging of conducting aerosol particles, where the assumptions made in deriving the expression are minimal. Specifically, our goal is to develop a collision kernel expression which is a function of ion and particle properties, accounts for both repulsive Coulomb and attractive image potentials between ion and particle, and can be extended reasonably to particles of arbitrary size and shape. We utilize an approach distinct from flux matching theory to develop a collision kernel expression for unipolar charging, involving combination of dimensional analysis, molecular dynamics, and Brownian dynamics calculations (Ermak & Buckholz, 1980; Narsimhan & Ruckenstein, 1985). To date this approach has been successful in developing collision kernel expressions for collisions between spheres (Gopalakrishnan & Hogan, 2011), vapor molecule condensation onto non-spherical particles (Gopalakrishnan et al., 2011), collisions between spheres considering the Coulomb potential (Gopalakrishnan & Hogan, 2012), collisions between two nonspherical particles (Thajudeen et al., 2012), and collisions between spherical entities in the presence of singular contact potentials (Ouyang et al., 2012), throughout the entire Kn_D range in all cases. The results presented here demonstrate that the dimensionless collision kernel expression for unipolar charging can be collapsed to a function solely of the diffusive Knudsen number irrespective of the shape of the particle, as it has been for the aforementioned gas phase collision processes.

2. Theoretical and numerical approach

The development of a collision kernel for unipolar charging requires (1) treatment of the electrostatic interactions involved, (2) analysis in the continuum limit, (3) analysis in the free molecular limit, and (4) analysis in the transition regime. Furthermore, to examine nonspherical particles, an appropriate set of particle test geometries is necessary. For this purpose we use ensembles of point contacting spheres which are quasi-fractal in geometry (i.e. they satisfy the relationship $N = k_f(R_g/a)^{D_f}$, where N is the number of primary spheres in each particle, a is the primary sphere radius, k_f is a pre-exponential factor, and D_f is the fractal dimension (Friedlander, 2000)). We discuss the computational generation of a wide range of quasi-fractal aggregate test geometries first, followed by discussion of (1–4).