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The capacitance and charge of agglomerated nanoparticles during sintering

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ABSTRACT

The electrical capacitance of aerosol particles indicates their diffusion charging level, which is important for their classification by electrical mobility, precipitation (removal or collection) in electrical fields, and morphology characterization. A minimum potential energy method was used to calculate the electrical capacitance for agglomerates composed of equally sized spherical primary particles (PPs). By discretizing the particle surface using finite spherical elements, as net charge only resides on the surface of an isolated conductor, this method was extended to calculate the capacitance of arbitrarily shaped particles. Based on the capacitance, the charge of these particles was obtained by diffusion charging theory. In addition, the dynamics of capacitance and mean charge of agglomerate during sintering or coalescence (at constant particle volume) to aggregates and finally to compact structures was computed and found in agreement with sparse experimental data. Particle morphology strongly affects the capacitance and mean charge of fractal-like particles. For example, both decreased by 60% upon full coalescence or sintering of an agglomerate consisting initially of 128 PPs.

1. Introduction

Electrical diffusion charging of aerosol particles plays an important role in the research of gas borne particles including aerosol instrumentation (Pui, Fruin, & McMurry, 1988; White, 1951), materials production from aerosols (Hogan & Biswas, 2008; Vemury & Pratsinis, 1995), air pollution control (Gentry, 1972), determination of the size distribution of fine particles by measuring their electrical mobility (Kirsch & Zagnit'ko 1981; Reischl, Makela, Karch, & Necid, 1996), and atmospheric aerosol physics (Fuchs, 1963).

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Particle capacitance indicates the ability of a body to store electrical charges. More specifically for aerosol charging, we are interested in self-capacitance of a particle, which is the electrical charge that must be added to an isolated particle to raise its potential by one unit (Greason, 1992), as opposed to the mutual capacitance between two adjacent conductors, such as a capacitor composed of two plates. Chang (1981) proposed expressions for the mean charge of arbitrarily shaped particles as a function of their self-capacitance in unipolar diffusion charging processes. His results were developed based on the work of Laframboise and Chang (1977), who extended the continuum regime diffusion equation to particles of arbitrary shapes. Rogak and Flagan (1992) and Filippov (1994) pointed out that the electric capacitance is affected by particle morphology using an analogy between electrostatics and diffusion.

In addition, the particle capacitance can be used to calculate diffusion charging of particles. Chang (1981) showed, for a given charger, the mean charge per particle is proportional to the particle capacitance in the continuum regime of charging. Shin et al. (2010) showed by simulations that the capacitance of chain-like agglomerates (physically-bonded PPs) is larger than that of spheres with the same mobility diameter and the difference increased with increasing PP number per agglomerate. This is in agreement with his experiments showing that the mean charge per particle of silver agglomerates was about 24% larger than that for fully coalesced silver spheres with the same mobility diameter in the mobility size range of 30–200 nm when the measurement error for both agglomerates and spheres was within 2%. In addition, Oh, Park, and Kim (2004) used an indirect photoelectric charger and showed that TiO_2 agglomerates in the mobility size range of 50–200 nm with a low mass fractal dimension, D_{fm}, had about 30% more charges than spherical particles. Jung and Kittelson (2005) showed that diesel agglomerates acquired more charges than nearly spherical NaCl particles by 15–17%. Wang et al. (2010) observed that the number of charges acquired by compact aggregates (chemically- or sinter-bonded PPs) was in-between those of agglomerates and spheres.

Nonetheless, it is difficult to directly measure the capacitance of airborne particles. Therefore, data for capacitance of aerosols are mainly obtained from analytical solutions (Serway & Jewett, 2009) or numerical studies (Zhou, Szabo, Douglas, & Hubbard, 1994). Brown and Hemingway (1995) used a variational method to calculate the charge distribution for the minimum electrostatic energy, thus obtaining the capacitance. Their method is applicable to agglomerates of spherical PPs in point contact with arbitrary agglomerate geometry and PP size. However, this method is limited to conducting particles with the entire agglomerate being at the same electrical potential. Nevertheless, Brown and Hemingway (1995) also pointed out that it is not necessary for the particle to be a good conductor, only that the charge relaxation time should be shorter than the typical time of observation or life time of aerosols. They argued that this assumption is reasonable for many aerosols, except for those with extremely high resistivity over $10^{11}\ \Omega$ m, as for example insulating polymer particles. Brown and Hemingway's (1995) method has been used to calculate the capacitance of particles in various configurations, including single sphere, doublet, triplet, straight and branched chain (Shin et al., 2010). However, only few 3D numerical simulations have been done for the spatial charge distribution and capacitance of agglomerates with many PPs. Also, little is known about the capacitance of necked aggregates that are often formed at high temperatures by sintering during gas-phase synthesis of materials (Pratsinis, 1998).

Here, 3D agglomerates with up to 512 PPs are investigated. The evolution of capacitance and mean charge for such agglomerates undergoing sintering or coalescence is simulated by generalizing the method of Brown and Hemingway (1995), covering particle morphologies from fractal-like agglomerates to aggregates and finally to compact spheres. The method was modified to relax the restriction on spherical PPs and calculate particles of arbitrary shapes. Unlike agglomerates with spherical PPs in point contact, in aggregates PPs are not well defined. Here a given particle with arbitrary shape was discretized into fine spherical elements to facilitate the calculation of the minimum electrostatic energy. The assumption that the particle is a reasonable conductor is still needed, and any net charge on an isolated conductor resides on its surface according to Gauss' theorem (Serway & Jewett, 2009). This means that discretization is only needed on the particle surface. Under those assumptions, our method could be applied to any type of particles. Validation was carried out by comparison with analytical (Serway & Jewett, 2009) and numerical solutions and experiments (Brown & Hemingway, 1995). By applying our method to agglomerates undergoing viscous flow sintering through multiparticle coalescence simulations (Eggersdorfer, Kadau, Herrmann, & Pratsinis, 2011), we are able to calculate the evolution of aggregate capacitance. So, by applying Chang's (1981) theory for particle charge based on capacitance, we simulate the evolution of aggregate charge during sintering.

2. Methodology

2.1. Agglomerate and aggregate generation

In this study, the capacitance of diffusion-limited cluster–cluster agglomerates (DLCAs) consisting of monodisperse spherical PPs during sintering is investigated. These simulations are validated with experiments of agglomerates produced at room temperature (Shin et al., 2010). Here such DLCAs are generated numerically from initially monodisperse PPs by a hierarchical cluster–cluster algorithm (Botet, Jullien, & Kolb, 1984). The agglomerate generation starts with 2^n individual PPs. Two PPs are randomly chosen from the ensemble and undergo a random walk until they collide and stick to form a dimer. The process is repeated with all pairs until 2^{n-1} dimers are assembled. Then the dimers are combined to 2^{n-2} clusters of four particles and so on until only one single agglomerate consisting of 2^n PPs is obtained. For each PP number (4, 8, 16, 32,