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The effect of primary particle polydispersity on the morphology and mobility diameter of the fractal agglomerates in different flow regimes

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ABSTRACT

Properties of colloidal and aerosol agglomerates depend on their morphology. Accurate estimation of the mobility-equivalent diameter d_m in different flow regimes is essential in many industrial processes and measurements. Previous work on the hydrodynamic properties of clusters focussed on agglomerates composed of monodisperse primary particles. However aggregates formed in real processes, e.g. soot particles, are usually formed from polydisperse monomers. Using numerically-generated agglomerates it is shown here that the radius of gyration, surface area, and mass of the agglomerates increase with primary particle polydispersity (given constant geometric mean primary particle size d_{pg}). Here, d_m is taken as the projected area-equivalent diameter for the free molecular regime; Stokesian Dynamics is used to compute d_m in the continuum flow regime. For fixed number of primaries and d_{pg}, d_m increases with polydispersity in both free molecular and continuum regimes ($>20\%$ for large particles at high polydispersity). Considering an aerosol population with polydisperse primary particles, this increase is found to depend on whether the variations in primary particle size occur *within* aggregates or *between* aggregates; this can be important in the interpretation of measurements. Finally, mobility diameters are correlated with total number, median diameter and its geometric standard deviation of the primary particles.

1. Introduction

Aerosol and colloidal agglomerates of smaller primary particles are formed in many industrial and natural environments. While some nanoparticle aggregates are potentially useful, soot aggregates formed by incomplete combustion of hydrocarbons are known responsible for adverse health effects Peretz et al. (2008) and climate forcing Hansen and Nazarenko (2004).

Production rate and properties of synthetic particles, aerosol residence time in atmosphere, and its dispersion rate and motion under different flow regimes are influenced by the morphology and hydrodynamic properties of these particles. Reliable measurement of the emission rate and size distribution of fractal aggregates also depend on accurate estimation of the morphology and mobility of these particles.

Great effort has been devoted to the estimation of the hydrodynamic properties of straight chains and fractal agglomerates in different flow regimes. The mobility of the aerosols of different structures is usually expressed by an equivalent

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mobility diameter (d_m). This diameter is equal to the diameter of a virtual sphere experiencing the same drag force under the same flow condition. Experimental measurements have been performed to correlate the mobility diameter of the fractal aggregates to the structure, e.g. radius of gyration (R_g), primary particle diameter (d_p), and projected area of these particles (Binder, Hartig & Peukert, 2009; Gmachowski, 2007; Gwaze et al., 2006; Johnson, Li & Logan, 1996; Rogak, Flagan & Nguyen, 1993; Thajudeen, Jeon & Hogan, 2015; Wang & Sorensen, 1999).

Several simulations have also been carried out to calculate the mobility diameter of clusters in different flow regimes. In the free molecular regime, the mobility diameter of the fractal aggregates, $d_{m,fm}$, is approximately equal to their average projected area equivalent diameter, d_a, (Rogak et al., 1993). This is consistent with the results obtained from Monte Carlo simulations of momentum transfer rate of chains and fractal aggregates in this regime (Chan & Dahneke, 1981; Mackowski, 2006; Thajudeen, Gopalakrishnan & Hogan, 2012). Consequently, if primary particle size variation is to influence particle mobility in this regime, it should be limited to the geometric relation between the number of primary particles and the projected area.

Calculation of the effect of morphology on continuum regime mobility is not as simple as the free molecule regime. Several methods have been developed for the assessment of the mobility diameter of aggregates in this regime. A group of these methods treats aggregates as porous spheres of homogeneous (Rosner & Tandon, 1994) or variable permeability (Kim & Yuan, 2005). The Lattice Boltzmann Method (LBM) has been used for the investigation of hydrodynamic properties of fractal aggregates by many researchers (Binder et al., 2006; Schlauch et al., 2013; Ulrich, Iglberger & Thurey, 2008). The Brownian dynamics approach has also been used in several studies (Douglas, Zhou & Hubbard, 1994; Gopalakrishnan, Thajudeen & Hogan, 2011; Thajudeen et al., 2012; Zhang, Thajudeen, Larriba, Schwartzentruber & Hogan, 2012) for the estimation of the collision kernels of particles and surrounding gas molecules, and mobility diameters of particles of irregular shapes in the continuum regime. As an another approach the Finite Element Method (FEM) has also been used for the investigation of the hydrodynamic properties of clusters in this regime (Melas, Isella, Konstandopoulos & Drossinos, 2014; Schlauch et al., 2013). Although these methods provide high accuracy, they are usually computationally expensive. Kirkwood–Riseman theory of hydrodynamic interaction of particles is another method commonly used for the calculation of the drag force on clusters of spherical particles (Chen, Meakin & Deutch, 1987; Kirkwood & Riseman, 1948; Lattuada, Wu & Morbidelli, 2003; Meakin, Chen & Deutch, 1985).

One of the most accurate methods commonly used for the simulation of the hydrodynamic interactions between aggregates and flow field in the limiting case of low Reynolds number, creeping flow, is referred to as Stokesian Dynamics (SD) (Binder et al., 2006; Bossis, Meunier & Brady, 1991; Filippov, 2000; Harada, Tanaka, Nogami & Sawada, 2006; Harshe, Ehrl & Lattuada, 2010; Schlauch et al., 2013; Seto, Botet & Briesen, 2011). In this method the flow disturbance caused by the non-overlapping monomers in clusters is modeled through the superposition of point forces on the surface of the monomers. The multipole expansion of the flow velocity in a series of spherical harmonics is used and the hydrodynamic interactions are simulated by a combination of far-field mobility and pairwise additive resistance calculations. Hydrodynamic interactions of two equal and unequal (Ichiki at al., 2013; Jeffrey & Onishi, 1984a, 1984b) spheres in different flow patterns are modeled by the Stokesian Dynamics method through the construction of resistance and mobility matrices which purely depend on the diameter and relative arrangement of primary particles in regards to the flow field. Application of SD to N-body clusters was pioneered by Brady and Bossis (1988) and Durlofsky, Brady, and Bossis (1986). As confirmed by rigorous numerical simulations of Gwaze et al. (2006) and Schlauch et al. (2013), SD provides good accuracy with modest computational time.

In large clusters, it is commonly assumed that the hydrodynamic radius becomes proportional to the radius of gyration (Sorensen, 2011):

$$R_{m,c} = \beta R_g \tag{1}$$

This proportionality was first investigated by Chen et al. (1987) and Meakin et al. (1985). Using Kirkwood–Riseman theory, they obtained $\beta = 0.875$ for particles generated by Diffusion Limited Cluster-cluster Aggregation (DLCA) composed of 50–400 monodisperse primary particles. Studies performed by Rogak and Flagan (1990), Wang and Sorensen (1999), Wiltzius (1987), and Sorensen (2011) suggested β to be in the range of 0.68–0.78 for DLCA aggregates with a fractal dimension (D_f) of 1.78. However, as it will be discussed later in this article, a wide range of values are reported in literature for β.

Although an extensive body of literature is already available for the simulation of the hydrodynamic properties of the fractal aggregates in different flow regimes, previous studies are limited to the clusters composed of monodisperse monomers. However, particles formed in different environments are shown to be composed of monomers of different levels of polydispersity (Dastanpour & Rogak, 2014; Heine & Pratsinis, 2007). Eggersdorfer and Pratsinis (2012) have showen that the fractal dimension of aggregates decreases with primary particle polydispersity. The influence of the primary particle polydispersity on the morphology (i.e. surface area, mass, and radius of gyration), and hydrodynamic behavior of particles in the free molecular and continuum regimes are investigated in the current study.