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The mobilities of flame synthesized aggregates/agglomerates in the transition regime

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ABSTRACT

Mobility analysis is frequently used to evaluate the size distribution of nanoparticle aggregates synthesized in high temperature, gas phase environments, such as flames. Theoretically, the scalar mobilities of such non-spherical entities, whose characteristic sizes are similar to the mean free path of their background gas (λ), are dependent on two size descriptors, the hydrodynamic radius (R_H) and the orientationally averaged projected area (PA) (as well as λ). Unfortunately, the theoretical relationship linking the mobility to R_H and PA for aggregates has not been experimentally tested, and has often been discarded in lieu of simpler yet theoretically invalid expressions. Here, we examine the mobilities of flame synthesized titanium dioxide (TiO_2) aggregates to directly test the link between the mobility, R_H, PA, and λ. Flame synthesized aggregates with mobility equivalent diameters in air in the 45–80 nm range were classified with a differential mobility analyzer (DMA) and deposited on a transmission electron microscope (TEM) grid for subsequent imaging. Probable 3-dimensional structures for each imaged aggregate were constructed, based on the assumption that aggregates were quasifractal in nature and by comparing four 2-dimensional size descriptors calculated for images to those of computationally generated projections of quasifractal aggregates (of prescribed pre-exponential factor, fractal dimension, and number of primary particles). The calculated mobilities for the 3-dimensional structures inverted for each image are found to be in excellent agreement with their mobilities inferred from DMA classification, supporting the theoretically proposed relationship between the mobility and R_H, PA, and λ in the $Kn=\pi\lambda R_H/PA=1.27$–$4.11$ range. Although a wide range of fractal dimensions and pre-exponential factors are inferred for the observed aggregates, the volumes of the reconstructed aggregates are still found to scale with the mobility diameters.

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1. Introduction

Flame synthesis with gas phase precursors is a scalable route to the production of a variety of nanoparticles (Strobel & Pratsinis, 2007; Athanassiou et al., 2010; Camenzind et al., 2010). In the final stages of the flame synthesis process, particle–particle collisions (Thajudeen et al., 2012) are typically the dominant particle growth mechanism. While particles may restructure/rearrange themselves subsequent to colliding, they typically do not fully coalesce; the time scales for viscous

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flow and grain boundary diffusion-driven sintering are slow relative to the characteristic time-scale for collisions, even in elevated temperature environments (Koch & Friedlander, 1990a, 2009b; Schmid et al., 2004; Eggersdorfer et al., 2012b). For this reason, flame synthesis usually leads to the production of aggregates/agglomerates, i.e. ensembles of point-contacting (in the case of agglomerates) or overlapping (aggregates, used henceforth to refer to both types of ensembles in this work) near-spherical primary particles (Xing et al., 1997; Grass et al., 2006; Eggersdorfer & Pratsinis, 2014).

Mobility measurements, with mobility defined as the proportionality constant between a particle's velocity and an external force acting on the particle, can be made with online instruments in near-real time and are used frequently to infer the size distribution functions of aggregates produced in flame synthesis reactors (Jiang et al., 2007). It is thus crucial to provide a link between an aggregate's structure/morphology and its mobility (Sorensen, 2011; Zhang et al., 2012). Unfortunately, efforts to describe the mobility of aggregates theoretically and experimentally have been disconnected. In addition to accounting for their diverse structures, a key challenge in determining the mobilities of aggregates is appropriate consideration of the "transition regime" nature of gas molecule-aggregate momentum transfer, i.e. for aggregates with characteristic sizes similar to the hard sphere mean free path of the background gas (λ), momentum transfer can be analyzed with neither continuum nor free molecular approaches. To address this issue for non-spherical particles, Dahneke (1973) proposed the "adjusted sphere" model, which remains to-date the most tractable approach to non-spherical particle mobility calculations. Recently, we have shown (Zhang et al., 2012) that Dahneke's adjusted sphere model follows directly from dimensional analysis, and that with it the orientationally averaged mobility of a non-spherical particle, B, can be calculated via the equation:

$$B=\frac{1}{f}=\frac{1+(\lambda\pi R_H/PA)\left(1.257+0.4\exp\left(-1.1PA/\lambda\pi R_H\right)\right)}{6\pi\mu R_H} \tag{1}$$

where f is the particle's scalar friction factor, R_H is the particle's orientationally averaged hydrodynamic radius (Douglas et al., 1994; Lattuada et al., 2003), PA is the particle's orientationally averaged projected area (Mackowski, 2006; Isella & Drossinos, 2011; Larriba & Hogan, 2013), and μ is the background gas dynamic viscosity. The numerator of Eq. (1) is the slip correction factor, defined with the coefficients provided by Davies (1945) and with the Knudsen number, Kn (Zhang et al., 2012), defined as $\lambda\pi R_H/PA$. For measurements made at room temperature and atmospheric pressure in air, $\lambda\approx 67$ nm, and for aggregates produced in flame synthesis reactors and downstream, Kn is commonly of order 0.1–10.

To date, Eq. (1) predictions have been found to be in good agreement with direct simulation Monte Carlo (DSMC) inferred mobilities for selected aggregate particles in the 0.3–10Kn range (Zhang et al., 2012), mobilities determined by alternative calculation approaches (Melas et al., in press), as well in good agreement with measurements on small aggregates (2–5 primary particles) (Cheng et al., 1988; Cho et al., 2007), larger straight chain aggregates (lower Kn, but with appreciably high aspect ratios) (Kasper, 1982), and gold nanorods with aspect ratios from 1 to 14 and Kn in the 1–6 range (in both air and carbon dioxide) (Gopalakrishnan et al., in review). Nonetheless, comparison of Eq. (1) predictions to measurements of morphologically diverse flame synthesized aggregates remains unreported, and has been hindered because Eq. (1) contains size parameters as inputs which are difficult to determine by direct means. To date, the most successful study linking flame-made aggregate mobility to aggregate structure is that of Rogak et al. (1993), who made measurements of the electron micrograph observed projected areas of mobility selected (via a differential mobility analyzer, DMA) TiO_2 aggregates at intermediate Kn (0.2–2.0). They showed good agreement between the projected area equivalent diameter and an aggregate's mobility diameter, d_m, inferred from mobility via the relationship:

$$B=\frac{1+(2\lambda/d_m)\left(1.257+0.4\exp\left(-0.55d_m/\lambda\right)\right)}{3\pi\mu d_m} \tag{2}$$

Based on this result, the equivalence of d_m and the projected area based diameter has been assumed in recent analysis of flame synthesized aggregates (Eggersdorfer et al., 2012a). However, while a one-to-one correspondence of d_m and the projected area based diameter has been observed in numerical simulations (Zhang et al., 2012) as $Kn\rightarrow\infty$ (the free molecular limit), Eq. (1) suggests that this correspondence should break down at intermediate Kn, unless $\pi R_H^2\approx PA$ for the aggregates in question. Along with examination of the validity of Eq. (1), the link between the projected area based diameter and the mobility diameter requires further scrutiny. Alternative to equating d_m to the projected area diameter, aggregates have been assumed to be straight chains with their mobilities equivalent to the free molecular limit mobility (Lall & Friedlander, 2006; Wang et al., 2010), i.e. their mobilities are assumed inversely proportional to the number of primary particles per aggregate (Chan & Dahneke, 1981). Again, use of this approach could only apply as $Kn\rightarrow\infty$, and further invokes the unproven (and contradicted by computations (Larriba & Hogan, 2013)) claim that the mobility of aggregates does not depend on actual aggregate structure. Finally, there is strong experimental support for describing most flame synthesized aggregates as quasifractal entities (Cai et al., 1995; Koylu et al., 1995; Huang et al., 1998; Filippov et al., 2000; Wang & Sorensen, 2002), approximately obeying the relationship:

$$N_p=k_f\left(\frac{R_g}{a_p}\right)^{D_f} \tag{3}$$

where N_p is the number of primary particles in an aggregate, k_f is the pre-exponential factor, R_g is the radius of gyration, a_p is the primary particle radius, and D_f is the fractal dimension. Based on this scaling, McMurry and coworkers have attempted to scale the mass of mobility selected particles with their mobility diameters (Scheckman et al., 2009; Shapiro et al., 2012),

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