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Porous particulate film deposition in the transition regime

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ABSTRACT

We examine the formation of particulate films via Langevin dynamic simulations of monodisperse particle deposition onto flat substrates. In simulations, we consider particle inertia, thermal motion, and a constant advective force directing particles towards the substrate. These conditions effectively mimic the formation of dust cakes during filtration, as well as the production of nanostructured films at the outlet of gas phase synthesis reactors. Parameterization of the equations of motion shows that the movement of each particle depends only upon Kn_D (the diffusive Knudsen number, proportional to the particle's mean persistence distance to its radius) and χ_F (the ratio of the particle's translational energy at the advective velocity to the thermal energy). Use of these ratios is advantageous because one dimensionless number (Kn_D) is independent of the advective velocity, while the second (χ_F) is independent of background gas pressure. This is in contrast with the traditionally employed Peclet and Stokes numbers, both of which are dependent upon the advective velocity and gas pressure. The film structures resulting from simulations are quantified in terms of their porosities, and results are reported for simulations over a wide range of Kn_D (10^{-3}–10^{1}) and χ_F (10^{-4}–10^{3}), deposition areas 1000×1000 particle radii in size, and with more than 10^6 deposited particles. For each simulated deposit, three distinct regions are observed: (I) a region close to the substrate (within 50 primary particle radii), wherein the porosity begins at a minimum and increases to constant value; (II) a region where this constant porosity is maintained (which is dependent on Kn_D and χ_F), constantly increasing in height and encompassing the majority of the film after sufficient deposition; and (III) the outer region, in which the film is still growing, and the porosity increases from the region II value to unity, spanning less than 100 particle radii in height in most circumstances. A regression equation is provided to describe the film porosity as a function of Kn_D and χ_F in region II, facilitating a priori predictions of film porosities from deposition parameters.

1. Introduction

The formation of particulate films by the deposition of aerosol particles onto surfaces is a common occurrence during particle filtration (Endo et al., 1998; Thomas et al., 2001), i.e. the formation of a dust cake, and can also to be used for the synthesis of nanostructured materials (Huisman et al., 2003; Wu & Choy, 2004; Sahm et al., 2007; Thimsen & Biswas, 2007; Hogan & Biswas, 2008; Tolmachoff et al., 2009; Holman & Kortshagen, 2010; Kubo et al., 2013). In nearly all such instances, particles are directed towards a surface via a combination of fluid flow, electrostatic, and thermophoretic motion, and in

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instances when the particles do not deform upon deposition, the resulting film porosity and nanostructure are dependent upon the size distribution of depositing particles, their advective velocity, as well as thermal motion. It is therefore of interest to develop models of deposition processes, which enable a priori prediction of film properties (Tassopoulos et al., 1989; Tassopoulos & Rosner, 1992; Madler et al., 2006). Subsequently, knowledge of film properties enables predictions of film performance, e.g. the pressure drop through a dust cake (Cheng & Tsai, 1998) or the current produced by light absorption in photocatalytic films (Thimsen et al., 2008).

There have been a number of prior efforts to model to the deposition of particles. Meakin (1983, 1984) examined the diffusion controlled deposition of particles in a 2-dimensional lattice onto a surface, but in the absence of any preferential particle advection towards the surface. Kulkarni and Biswas (2003, 2004) examined deposition considering both the influences of particle–particle and particle–surface interactions as well as sintering upon deposition. However, both of these sets of prior work examined deposition in the solely diffusive limit, as such the results apply only in instances where the diffusive Knudsen number, Kn_D, is sufficiently small, where Kn_D is defined as

$$Kn_D = \frac{(kTm_p)^{1/2}}{fa_p} \tag{1a}$$

with kT the thermal energy, m_p the particle mass, f the particle friction factor, and a_p the particle radius. For nanoparticles in the gas phase, this criterion is not valid under all circumstances. Madler et al. (2006) attempted to model deposition without the $Kn_D \to 0$ restriction embedded into the equations of motion. However, in their work, predicted film porosities and thicknesses were again determined only in terms of the Peclet number, Pe, defined as

$$Pe = \frac{U_0}{Kn_D}\left(\frac{m_p}{kT}\right)^{1/2} \tag{1b}$$

where U_0 is the particle advective velocity. Film properties are only functions of Peclet number alone in the $Kn_D \to 0$ limit. For this reason, the description of the most basic instance of particulate film deposition, wherein monodisperse particles are directed towards a surface by a constant external force/velocity and are immobilized upon collision with either the surface or deposited particles, still remains incomplete; film porosities and other nanostructural characteristics have not been predicted from deposition parameters. The purpose of this work is hence to provide a theoretical description of particulate film deposition under this circumstance. This is accomplished via Langevin dynamic simulation (Ermak & Buckholz, 1980) performed in a dimensionless fashion, akin to recent simulations focusing on particle–particle (Gopalakrishnan & Hogan, 2011; Gopalakrishnan et al., 2011; Ouyang et al., 2012; Thajudeen et al., 2012), and particle–ion collisions (Gopalakrishnan & Hogan, 2012; Gopalakrishnan et al., 2013a,2013b) in the gas phase. Deposited film properties, quantified primarily in terms of the porosity, are determined as a function of the diffusive Knudsen number. As in recent Langevin dynamic simulations of particle collection by filter fibers (Hunt et al., submitted for publication), deposited film properties are additionally determined as functions of χ_F, the ratio of the translational kinetic energy to the thermal energy, defined as

$$\chi_F = \frac{m_p U_0^2}{kT} = (PeKn_D)^2 = \left(\frac{St}{Kn_D}\right)^2 \tag{1c}$$

where St is the Stokes number, defined with the particle radius as the normalizing length scale. Although particle deposition is traditionally described using both the Peclet and Stokes numbers, we elect to use Kn_D and χ_F here because only χ_F depends upon the advective velocity and only Kn_D depends upon the background gas pressure (both Pe and St depend upon the advective velocity and gas pressure). In the following sections, the model employed is described in detail, including remarks on the assumptions invoked in its development. Results are then presented for film porosity as a function of Kn_D and χ_F. Influences on film nanostructure are additionally discussed.

2. Theoretical and numerical methods

2.1. Simulation parameters and particle equations of motion

We consider the deposition of particles in the dilute limit, wherein collisions between particles prior to impacting a surface or pre-existing film are neglected. For applications of filtration, this condition is often valid, and in film deposition systems coupled to gas phase synthesis reactors (Tolmachoff et al., 2009; Zhang et al., 2012) deposition often occurs downstream of the non-dilute, high particle concentration region. The motion of particles until deposition is therefore modeled sequentially. Simulations are performed as follows: a particle with a dimensionless radius of one is initially placed at a random (x^*, y^*, where the superscript "*" denotes normalization via particle radius) location above a flat surface of square area of prescribed dimensions (1000×1000 particle radii in most circumstances). For this first particle, the initial vertical (z^*) location is one radius from the surface, hence it is in contact with the surface and immobilized. A second particle is then introduced into the simulation domain at a random (x^*, y^*) coordinate, and its z^* coordinate is selected as 2.1 particle radii above the first deposited particle (such that the new particle cannot be in contact with the previously deposited particle). Over the course of a small dimensionless timestep, $\Delta\tau$ (where time is normalized by m_p/f), the particle is moved from its current coordinate $\vec{x}^*_p(\tau)$ to a new coordinate $\vec{x}^*_p(\tau+\Delta\tau)$ ($\tau=0$ initially) via the dimensionless solution Langevin