Structure of Granular Deposits Formed by Aerosol Particles Conveyed by Fluid Streams

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1 Introduction

Aerosols (particles carried by gas streams) appear in many practical applications and the understanding of their dynamics [1, 4] is needed to control processes such as, for instance, heterogeneous nucleation of vapors on preexisting particles, evolution of clouds, pollution dispersion and production of new materials from powders [3]. In this area, there is a need of controlling and characterizing the structure of granular materials formed by depositing aerosol particles. The main morphological features of these granular deposits as their bulk properties (density, porosity and structure) and interface properties (roughness and thickness of the active region) depend on the way that new particles arrive to form the deposit. The granular structure affects the chemical, optical and mechanical properties of the product and it should be tailored for new materials applications: nanostructured deposits, catalytic surfaces, layered materials and others.

The goal of this work is to relate the structure of the granular deposit to the characteristics of the particle motion near the surface.

2 Monte Carlo Simulation of Particle Motion

The method employed is an on-lattice dynamical Monte Carlo simulation [8,9] for the growth of particle deposits by advection and diffusion of particles towards a (initially clean and flat) surface. The model allows to follow the evolution of deposit formation and to determine the main morphological and structural properties of the generated deposits, depending on the transport properties of the arriving particles.

The Monte Carlo method is used to simulate the particle motion over the deposits until the particle reaches the deposit (becomes in contact with

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any previously deposited particle). Then the new particle attaches there and contributes to the deposit growth. Sintering or restructuring of the deposited particles is precluded so that the analysis focuses on the relevance of the mechanism of particle arrival. For the Monte Carlo simulation, the motion of an aerosol particle is split in two contributions: a mean deterministic velocity (normal to the flat surface) and a random motion. The deterministic contribution can be due to any transport phenomena [1, 4] that drives the particle along well defined trajectories, such as inertia, advection, some phoretic motions (thermophoresis, electrophoresis), particle sedimentation or external fields, whereas the random motion accounts for the particle diffusive motion (Brownian diffusion, turbulent diffusion or the effect of random fields). The Péclet number is the dynamical parameter that measures the relative importance of the deterministic motion to the random contribution,

$$Pe \equiv \frac{va}{\mathcal{D}} \tag{1}$$

relating the average particle velocity toward the wall, v, the particle diffusion coefficient, $\mathcal{D}$, and the particle diameter, a. In these simulations the average particle velocity, v, is considered to be normal to the (initially clean) surface where the deposit builds up. Time and space are discretized and a cubic lattice is used as the basic domain in the simulation with periodic boundary conditions on the lateral walls. A particle is introduced above the deposit structure at a random horizontal location and its motion is tracked until the particle either reaches the deposit or it moves far away from it. Then, a new particle is introduced and the process is repeated until the deposit height reaches a given maximum height.

The purpose of this work is to analyze the structure of deposits formed on attracting surfaces (positive values of v) and on slightly repelling surfaces (negative values of v); that is, when the Péclet number can be taken as negative, in the sense that the particle mean velocity pushes the particles away from the surface, but this repulsion is weak and Brownian diffusion is still able to bring some particle to deposit on the wall.

3 Deposits on Attracting Surfaces

The formation of deposits collected from aerosol particles which are attracted toward a wall (positive values of v in our simulations) has been extensively studied [5, 8–10]. The arriving particles form layered granular deposits with a density profile which depends on the particle Péclet number. Figure 1 shows the density profile for $Pe = 0.1$ and different deposit heights, $h_{\max}$ (which is related to the total number of deposited particles, i.e. to the total growth time). The collected deposits have a (frozen) denser region in contact with the clean wall, a (frozen) middle region of mean porosity and constant mean density, and an (active) upper region where new particles still deposit [8].

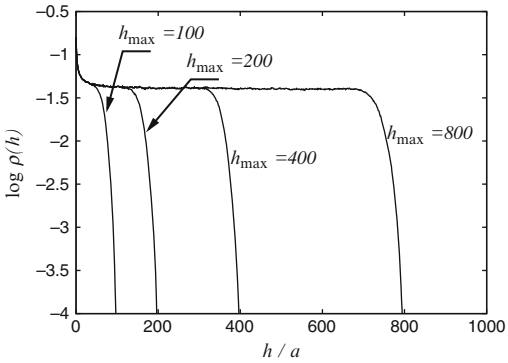

Fig. 1. Density profile versus height for different values of the maximum height, for particles with $Pe = 0.1$

In this active region, the mean density decreases from the middle density, vanishing at the top, $h = h_{\max}$.

The average deposit density in the frozen middle region (relative number of lattice sites occupied by particles in this consolidated region) shows a dependence with the Péclet number given by [8,9]

$$\bar{\rho}(Pe) = \rho_\infty \left(1 + \frac{Pe_0}{Pe}\right)^{-B} \tag{2}$$

With $\rho_\infty = 0.302$, $Pe_0 = 4.8$, and $B = 0.52$. This region presents a fractal-like structure on the short length scales [8], up to a Pe-dependent scale given by the quantity inside the brackets in (2), with a fractal dimension $D_F = 3 - B$. The limit of large Péclet numbers corresponds to ballistic deposition when particles drift towards the wall and Brownian diffusion is absent. Then, the deposits are denser and the porosity is low. However, for small values of Pe, the deposits are fractal with "particle trees" of all the allowed sizes being limited by lattice size, growth time or the Pe-dependent scale.

Moreover, the pure diffusion limit (vanishing Pe) when the particle motion is purely diffusive and the mean velocity vanishes, corresponds to a singular limit as indicated by (2) because the fractal structure of the deposit extends to all scales and the fractal cut-off (Pe-dependent) scale goes to infinity. The open but highly branched structure of the deposit restricts the penetration of new incoming particles deep into the deposit. In this limit, although some reminiscence of the three regions for attracting surfaces still remains, the deposit density decreases continuously with height.

4 Deposits on Weakly Repelling Surfaces

Even in the case of a mean (weak) particle motion away from the wall, diffusion may bring some particles to the surface [2, 4, 7] and form a granular deposit. A *negative value of the Péclet number* characterizes these deposits, with the minus sign indicating that the mean particle velocity v is directed away from the deposit.

In the limit of small (and negative) Péclet numbers, the formed deposits are initially fractal (as in the pure diffusion limit, $Pe = 0$). But, as the deposit evolves in time the dispersion in the deposit height increases, and a new structure appears dominated by the presence of relatively large particle trees. The larger trees are more effective in collecting the particles that approach to the surface by diffusion, avoiding the growth of the shorter trees. Finally, isolated trees emerge from a fractal deposit baseline.

Therefore, at long times, these deposits present two characteristic regions (Fig. 2): a base region which retains the same structure of the pure diffusion deposits, and a second region with spikes emerging from the base. The baseline structure is shorter for stronger repulsion fields, thus the height of the base region decreases as $|Pe|$ increases, according to the law

$$h_c = \left(1 + \frac{4.8}{Pe}\right)^{1/4} \tag{3}$$

Indicating that the same crossover length (the bracketed quantity) remains for repelling surfaces, see (2). On the other hand, the spike becomes thinner as $|Pe|$ increases.

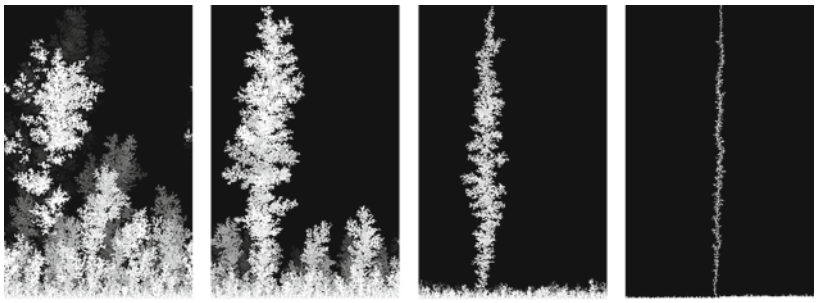

Fig. 2. Side view of the granular deposit, for weakly repealing surfaces and different Péclet numbers (a *grey scale* is used to represent the distance from the frontal wall)

5 Comparison with Experimental Results

In the laboratory, granular deposits are grown using the technique of electrohydrodynamic atomization [6] to disperse a liquid. The liquid is a suspension of carbon nanoparticles in ethanol with a small amount of dispersant to reduce the agglomeration of the nanoparticles. The liquid is pumped through a needle at a fixed flow rate, whereas a high voltage is applied between the needle and a flat collector located below the needle. The electrosprayed liquid forms a quite monodisperse cloud where the size of the primary droplet depends on the liquid properties and flow rate [6]. In our experiments the mean droplet size is of the order of $10\,\mu$m, and each droplet includes $\sim 10^3$ carbon nanoparticles. The fragmentation and evaporation of the charged droplets leaves dry carbon nanoparticles which retain the electrical charge and the nanoparticles are attracted by the collector and form a granular deposit. The charge of the particles in the cloud contributes to the particle dispersion and promotes an effective particle diffusion.

SEM images of the deposit are used to measure the surface roughness as a function of the applied voltage (proportional to the particle Péclet number). A *Shape-from-Focus* (SFF) technique is used to obtain a reconstruction of the deposit surface that can be treated by image processing. The measured roughness decreases with increasing applied voltage, in accordance with the numerical simulations that predict denser deposits as the Péclet number increases. The larger particle agglomerates (with larger inertia and Péclet number) deposit on the central part of the collector whereas at the collector edge some smaller particles deposit. Then, the characteristic particle size is not uniform and a direct comparison with the numerical simulation is not yet possible. To this end, new experiments are being conducted to get sprays containing fewer particles per droplet.

6 Conclusions

The structure of granular deposits formed by the deposition of aerosol particles has been related to the characteristic of the particle motion near the collecting surface. A Monte Carlo method has been implemented to simulate the particle motion [8,9] and the main features of the growth deposits were obtained as a function of the particle Péclet number which measures the relative importance of the mean particle velocity normal to the surface with respect to the particle Brownian motion, see (1).

For attracting surfaces (mean particle velocity towards the wall), the deposit is structured in three differentiated regions: a denser bottom layer in contact with the collecting wall, a middle region with mean constant density and the active region at the top where new particles may still attach. The deposit density decreases as the importance of Brownian diffusion increases (Pe decreases), with the density in the middle region correlated by (2). In the

ballistic limit (large Pe), the deposit is denser, whereas when Pe is reduced the deposit becomes lighter, very porous and highly branched.

In the pure diffusive limit, the branches at the top of the deposit are very effective in collecting particles. Then, the penetration of the incoming particles into the deposit is reduced and longer times are needed to achieve the fractal limited deposits. The deposits grow taller but fragile with open structures, leading to materials which are suitable for some applications where the ratio of area to volume plays a relevant role.

Furthermore, for weakly repelling surfaces the particle Brownian motion is able to bring some particles against the mean drift and a deposit is still formed on this surface. These deposits present two different regions: a base region with the same density profile as the pure diffusive deposit (as the growing mechanism is the same, Brownian diffusion) and large isolated spikes that emerge from this base region. The thickness of the base regions is reduced and the spikes become thinner as the repulsion intensity increases.

Experiments with atomized liquid suspensions are being performed to check the results of the simulations and the preliminary results shows a good qualitative agreement.

The simulation and experimental procedure used in his work open the possibility of making tailored granular materials for specific applications with defined bulk and surface morphologies.

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