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Overestimation of the fractal dimension from projections of soot agglomerates

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ABSTRACT

It is known that the exhaust stream of internal combustion engines contains soot agglomerates with fractal geometries. The samples of real soot agglomerates can only be analyzed from projections observed with powerful microscopes. The fractal dimension of an agglomerate is estimated from the combination of its volume and a characteristic radius. The radius of gyration is often considered as the characteristic radius and, in case of being composed of uniform primary particles, the volume of the agglomerate can be represented by the number of primary particles. However, none of them (radius of gyration or number of primary particles) can be determined from projected images. As previous works have pointed out, there is an intrinsic error in the estimation of the real radius of gyration, which cannot be measured directly from the microscopy projected images. In this work, an algorithm is proposed to generate three-dimensional synthetic agglomerates that mimic the fractal geometry of real ones. The real three-dimensional radius of gyration is measured and compared to the one found from projections. Also, the number of primary particles is compared to that estimated from the projected area. The consequence of both estimation errors on the determination of the fractal dimension is analyzed. It is shown that the fractal dimension is overestimated when it is calculated from projected images.

1. Introduction

Knowledge about the morphology of the soot agglomerates emitted by combustion devices such as internal combustion engines, e.g. diesel engines and direct-injection spark-ignition engines, is fundamental to understand their impact on the environment and on human health (e.g. on the cardiovascular system [1], and on the respiratory system [2]), and also on the aftertreatment systems of the exhaust gas [3]. The fractal dimension is one of the most significant parameters, and quantifies the irregularity of the agglomerates, (see Mandelbrot [4]).

The morphology of fractal agglomerates is described by the power-law relationship [5]. This relationship results from the integration of the autocorrelation function [6], which expresses the decreasing probability of primary particles to exist at a distance of the center of gravity of the agglomerate, for increasing distance.

$$n_{po} = k_f \left(\frac{r_g}{r_{po}} \right)^{D_f} \tag{1}$$

where n_{po} is the number of primary particles, k_f is the dimensional prefactor, r_g is the radius of gyration of the agglomerate, r_{po} is the radius of the primary particles and D_f is the fractal dimension.

The soot agglomerates emitted by a diesel engine have sizes in the nanometer scale. Therefore, a transmission electron microscopy is necessary to observe them. However, only projected images of the real agglomerate can be processed. Normally, the radius of gyration of the real agglomerates is assumed to be equal to that determined from its projected image. But the radius of gyration of the real agglomerate is always higher than the estimated one [7]. Therefore, to find the radius of gyration of real agglomerates, a reconstruction of the morphological parameters of the real agglomerates should be done. One option is to relate the gray level of the images to the thickness of the agglomerate [8]. However, the spatial arrangement cannot be precisely determined. Another option is to use tomography images [9]. Nevertheless, with this technique there is always an unavoidable overlapping between different parts of the agglomerate. Another way is to generate synthetic agglomerates similar to real ones.

Alternative methods for generating agglomerates whose shapes resemble those of the real ones have been extensively used to study their physical properties, such as light scattering [10–13], Van der Waals interaction [14] and aggregation kinetics [11]. These methods have also been used in geometrical studies, such as those evaluating the effect of the size of primary particles [15] and of the anisotropic growth [16] on the fractal dimension of the agglomerates. The effect

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that the external conditions have on the diffusion coefficient and the sticking probability of primary particles can be found in the literature too [17].

Simulations may have different targets: to calculate the solid volume fraction of the agglomerate [10,18], to obtain the potential energy when agglomerates are formed by colloidal primary particles [14], to determine the dynamic structure factor [11]; to determine some geometrical parameters as the fractal dimension or the prefactor [15,19], to study the effect of flattening or sintering in the contact area between primary particles [19], or to study the impact of the evaporation rate [17].

The different algorithms for agglomerate generation can be gathered into two different categories: methods based on simulating the physical phenomena conducting to final agglomerates and others based on geometrical parameters. Among the first ones, the methods found in the literature are based on: Diffusion-Limited Cluster Aggregation (DLCA), whereby the particles undergoing a random path due to brownian motion cluster together with the onset of aggregates of such particles [10,14,15,17]; or Reaction-Limited Cluster Aggregation (RLCA), where the interaction potential exhibits a barrier [11,20]; and via evaporation-condensation method [18]. The second category of methods follow the approach found in [21], where the geometrical parameters such as the fractal dimension, D_f, and the prefactor of power-law relationship, k_f, are imposed as inputs conditions. These methods are frequently referred to as "tunable fractal dimension methods". However, such methods have some drawbacks. The most important is the accuracy to determinate the location of the particles inside the agglomerate. This problem arises from algorithms developed for creating agglomerates using DLCA or RLCA approaches. However when algebraic calculations are used to create agglomerates [13], the particles are located at contact points and, therefore, the location errors are removed. The time is another important parameter to take into account when using algorithms for generating agglomerates. For example, the time for the generation of a synthetic agglomerate, with >1000 particles, can take from hours to days [22]. In the current work, a new algorithm for the generation of synthetic agglomerates is proposed. This algorithm is faster than others previously published, although it is not tunable but collisions are fully random. Thus, our algorithm needs from seconds to minutes to generate complex synthetic agglomerates in a laptop.

2. Generation of 3-D agglomerates

2.1. Algorithm

This section is devoted to detail the different steps of the algorithm to simulate random cluster-cluster collisions to build the synthetic three-dimensional agglomerates with fractal geometry. This algorithm can be applied to agglomerates with and without sintering. The term sintering should be understood here as the geometrical effect of different phenomena such as flattening, partial coalescence, surface growth, etc., but not necessarily sintering. The sintering coefficient is the ratio between the average radius of the primary particles and half of the distance between the particle centers. This concept is taken from Lapuerta et al. [23], and the sintering coefficient was defined by Oh and Sorensen [19]. In this study, only point contacts between primary particles are considered (sintering coefficient equal to one). The steps of the algorithm are:

- *Step 0*: The algorithm starts with the introduction of the number of primary particles to be generated, n_{po}, that will form the agglomerate as well as their diameter, d_{po}. Both values can be fixed by the user or randomly selected by the algorithm within a given interval.
- *Step 1*: The first step of the algorithm is the stochastic determination of all the directions for every collision. These directions will be used in the latter steps of the algorithm to determine the direction from which the clusters will collide each other. The three components of the impact will be given as a result of this step. The initial positions of these clusters (composed of only one primary particle at the beginning) are kept throughout all the simulation. Every cluster is saved as a matrix where the rows represent the displacements of every coordinate (x, y, z) of the geometrical center of the particle with respect to the center of gravity of the cluster. The coordinates (0, 0, 0) are assigned to this center of gravity. These coordinates will be moved later after every new collision. The radius of the maximum sphere, representing the sphere which circumscribes the cluster is also saved.
- *Step 2*: In this step the collision is simulated. Fig. 1 shows two clusters before the collision, and also the impacting trajectory. There are several sub- steps involved in this process: *Step 2.1*: First, there is a draw whereby two clusters are selected among the population of clusters already generated. *Step 2.2*: A second draw determines which one of the clusters selected in the previous step will be the colliding cluster and which one will be the collided cluster. For coherence's sake, they will be referred to as impacting and impacted, respectively. *Step 2.3*: The center of gravity of the impacted cluster is reallocated at the global center of coordinates (0, 0, 0).
- *Step 3*: The surface area of the impacted cluster is projected in a direction which is perpendicular to the impact trajectory (see Fig. 1). The resulting area is enlarged in a quantity given by the radius of circumscribing sphere of the impacting cluster. A random point is selected among this enlarged surface.
- *Step 4*: From the point determined in the previous step and following the trajectory of the impact, a point is defined where the center of gravity of the impacting cluster will be located. This point has to be a sufficiently far away from the impact point.
- *Step 5*: A pre-screening process is carried out in order to determine particle candidates for the collision. The criterion for this screening is to select those pairs of particles (one from the impacting cluster and other from the impacted one) whose minimum distance between centers is equal or less than the sum of their respective radii. Among the candidate pairs the one with the minimum distance will be selected. In case there is no pair fulfilling this condition, the collision will be rendered as failure and the process will start again (Step 2). Should a collision between clusters be rendered as a failure, the direction of the collision will be kept until the collision becomes successful.
- *Step 6*: The particle(s) enclosed in the impacting agglomerate are added to the one(s) already existing in the impacted one. The new center of gravity of the resulting cluster is calculated.
- *Step 7*: All the coordinates of the particles are referenced to the new center of gravity determined in the previous step. The radius of the circumscribing sphere and radius of gyration are again calculated.
- *Step 8*: The process is repeated from Step 2 until there is only one cluster, which is the final agglomerate.

The flow chart of the algorithm proposed is illustrated in Fig. 2.

2.2. Generation of sample agglomerates

For the present study, 500 random agglomerates were generated. This number of random agglomerates is a compromise between the

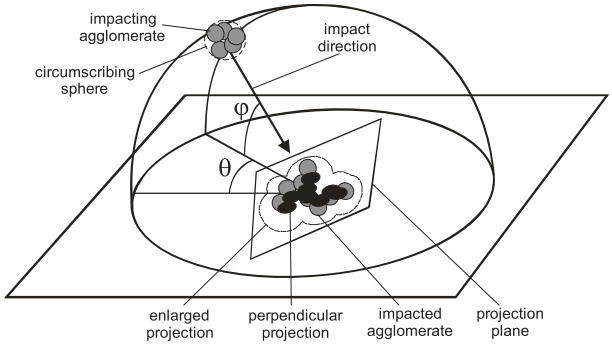

Fig. 1. Scheme of the cluster-cluster collision simulation.