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Stochastic modelling of particle aggregation

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ABSTRACT

Aggregation is an inter-particle process which involves a multitude of different physicochemical mechanisms. In the present work, particles in the nano-scale are considered, with such concentration that renders their direct simulation as individual particles intractable. A stochastic aggregation model is presented for large particle populations in a Lagrangian framework. The model allows for simultaneous collisions between numerical parcels present in a certain volume of interaction (e.g. computational cell) and can be directly coupled to an unsteady numerical solver of a continuous flow. The model performance is evaluated against analytic solutions for a sum (Golovin) and constant aggregation kernel.

Introduction

The intersection of particle paths may lead to a collision event and an inelastic collision event to aggregation. A particle of volume v (or mass m) is formed by the *aggregation* of two particles with volumes v' and $v - v'$. However, its resulting volume is subject to the level of coalescence (also termed 'sintering' for solids), i.e. the extent to which the colliding-pair volumes merge. Depending on the phase of the particle (solid, liquid, or gaseous), the local thermodynamic conditions (e.g. temperature), and the collision process (angle of attack, relative velocity, etc.) the particle will attach to the 'receiver' particle differently, forming fractal aggregate structures (such as soot) or spherical aggregates. In the present study, particles in the nano-scale are considered and instantaneous coalescence of the colliding pair is assumed.

Binary droplet and particle collisions are an area of interest with applications ranging from meteorological to industrial processes. Meteorological phenomena such as cloud formation and raindrop formation drove research on experimental studies of aqueous coalescence in atmospheric air, see works by Adam and Lindblad (1968); Ashgriz and Poo (1990). Maximova and Dahl (2006) review the environmental implications of aggregation phenomena in the context of wastewater and gas emission treatment.

In the context of spray atomization, Gavaises et al. (1996) concluded that coagulation greatly influences the Particle Size Distribution (PSD) and consequently controls the particle relaxation times and velocities during injection.

Following the reports by Brauer et al. (2002); Finlayson-Pitts and Pitts (1997); Montes et al. (2004); Oberdorster (2001) on the effects of the size of aggregates on health, modelling of soot aggregates from combustion has also become an area of great importance, see Zucca et al. (2006).

Another example of industrial-scale nanoparticle aggregation is spray drying technologies (e.g. for the manufacturing of detergents and water-dissolvable powders), wherein aggregation governs the properties of the particles.

Aggregation can be both wanted and unwanted depending on the application. Aggregation is promoted in mining and spray drying applications to reduce wastage and make separation easier (e.g. enhance settling rates in gravity based separations). However, aggregation may reduce product quality by widening the PSD. In order to control the PSD, there has been an increasing effort to combine numerical modelling and experiments to investigate the aggregation process.

The objective of the present work is to develop a model for the aggregation process in the post-spray region using the spray probability density function (pdf) method of Williams (1958). The model focuses on applications particles with large number of particles, $> \mathcal{O}(10^8)$, such as nanoparticle formation, where one-to-one collision approaches are intractable and macroscopic statistical models are hence sought. The aim is to model macroscopic aggregation

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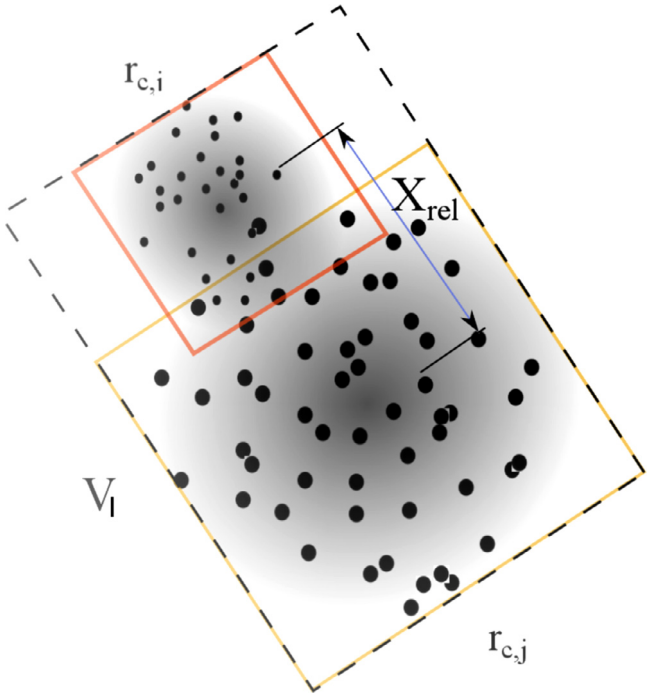

Fig. 1. Interaction volume of two interacting parcels *i, j*. The interaction volume is indicated by the dashed line.

kernels in a Lagrangian framework in an implementation that represents the physics involved in the aggregation process, yet at the same time retains the advantages of the spray-pdf methodology. The proposed model can be directly coupled to an unsteady representation of the continuous flow field, such as Large Eddy Simulation, without any modification. In the present paper, the model is presented and validated against analytical solutions for different aggregation kernels.

Modelling aggregation

The aggregation kernel

The aggregation process involves collisions of particles to form new ones with larger volumes. Consequently, both microscopic and phenomenological approaches can be sought.

In microscopic approaches each collision event is considered separately, such as the hard sphere collision model of Sundaram and Collins (1997); Yamamoto et al. (2001) and Yan et al. (2008). Microscopic models are used to understand the physics involved in the aggregation processes. Liao and Lucas (2010) categorise the numerous models for fluid particle coalescence processes and their relation to findings from binary (microscopic) collision experiments. However, in particle-laden flows in engineering applications, the number of particles, hereafter N_{tot}, render microscopic approaches intractable.

Phenomenological approaches treat the collision process in a macroscopic framework. It is usually assumed that a collision event will result in the immediate coalescence of the two particles. Consider that C_{ij} is the number of collisions per time and volume between two size classes i and j that form a new class k with $\upsilon_k = \upsilon_i + \upsilon_j$. The joint probability of a collision to occur (and an aggregate to form) is quantified by the aggregation kernel (or frequency), β_{ij}, and the respective concentrations $N_{c,\,i}$ and $N_{c,\,j}$ such that:

$$C_{ij} = \beta_{ij} N_{c,i} N_{c,j}. \tag{1}$$

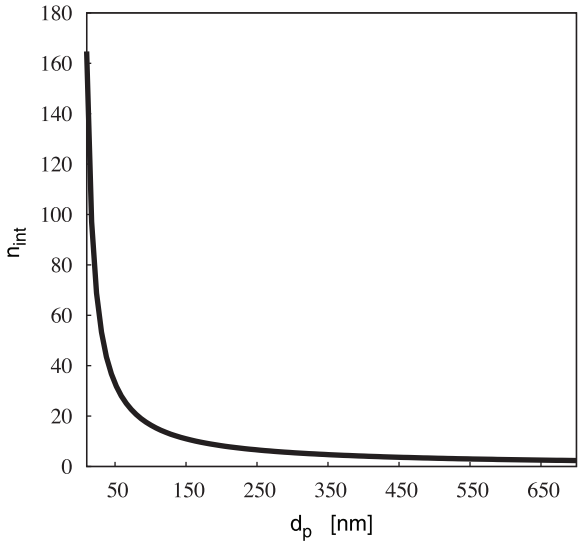

Fig. 2. Maximum number of real particles represented by one parcel as a function of particle diameter. With $\eta_k = 0.28$ mm, Sc = 1 and $\mathrm{Kn}_{f=air} = 170$.

The summation over all possible pairs *i, j* leading to a size class υ_k gives rise to the Smoluchowski (1917) equation, also known as the Population Balance Equation (PBE). Considering only aggregation processes, the equation is:

$$\frac{dN_{c,k}}{dt} = \frac{1}{2}\sum_{i+j=k} \beta_{ij} N_{c,i} N_{c,j} - N_{c,k} \sum_{i=1}^{\infty} \beta_{ik} N_{c,i}. \tag{2}$$

Where, the kernel β_{ij} incorporates the physico-chemical characteristics of the process. Macroscopic parameters of both the continuous and disperse phases may influence β_{ij} depending on the physical mechanism that drives the aggregation process.

Modelling the coupled kernel

Solution methods for the flow-coupled aggregation stem from those for the uncoupled kernel, namely: purely analytic, Method of Moments (MOM) and its variations (QMOM, HMOM and DQMOM), sectional methods, Monte-Carlo (MC) methods, and combinations thereof. These can be solved either in an Eulerian or in a Lagrangian framework.

Purely analytic methods solve the aggregation integrals or the equivalent closed set of moment integrals. Such solutions are confined to a limited number of kernels (e.g. $\beta_{ij} =$ constant, $\beta_{ij} = \upsilon_i + \upsilon_j$, or $\beta_{ij} = \upsilon_i \times \upsilon_j$) for which the simple integrals can be directly evaluated, see for example (Scott, 1968), or used to close the PBE for specific initial PSDs (e.g. log-normal or mono-disperse). However, if the particles have a wide range of initial sizes, the aggregation kernel cannot be considered size-independent and analytic methods become intractable.

Eulerian frameworks used for the population balance solve a set of ODEs that result from a reconstruction of the PBE using a Method of Moments, or a discrete number of size classes. The reader may refer to the review of Yu and Lin (2010) for an overview of moment methods to solve nanoparticle-laden flows.

Other widely used Eulerian approaches for the solution of the flow coupled PBE are discretization methods such as Yeoha and Tub (2006) and Miller and Garrick (2004). The number of 'bins' used for the discretization must represent the PSD accurately and a large number of bins might be required in problems where the PSD covers a wide range or widens significantly due to the particle processes, see Rigopoulos (2010). Azizi and Taweel (2010) suggested to adjust the PSD if its range requires expansion or contraction.