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Analysis of mesoscale effects in high-shear granulation through a computational fluid dynamics–population balance coupled compartment model

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ABSTRACT

There is a need for mesoscale resolution and coupling between flow-field information and the evolution of particle properties in high-shear granulation. We have developed a modelling framework that compartmentalizes the high-shear granulation process based on relevant process parameters in time and space. The model comprises a coupled-flow-field and population-balance solver and is used to resolve and analyze the effects of mesoscales on the evolution of particle properties. A Diosna high-shear mixer was modelled with microcrystalline cellulose powder as the granulation material. An analysis of the flow-field solution and compartmentalization allows for a resolution of the stress and collision peak at the impeller blades. Different compartmentalizations showed the importance of resolving the impeller region, for aggregating systems and systems with breakage. An independent study investigated the time evolution of the flow field by changing the particle properties in three discrete steps that represent powder mixing, the initial granulation stage mixing and the late stage granular mixing. The results of the temporal resolution study show clear changes in collision behavior, especially from powder to granular mixing, which indicates the importance of resolving mesoscale phenomena in time and space.

Introduction

In a recent review on high-shear-granulation modeling by Kumar, Gernaey, De Beer, and Nopens (2013), the special need for mesoscale resolution was highlighted by addressing the coupling between local flow-field information and particle property evolution. This issue was highlighted further in a granulation review by Iveson, Litster, Hapgood, and Ennis (2001), which indicates the need for flow-pattern analysis for different types of granulators and how such an analysis can be coupled to the mixing behavior and impact velocities of individual granules. A tool to solve flow properties for vessels of all types and sizes and relate them to the evolution of particle properties would be of great value for scale up and process control.

Population-balance modeling is the main modeling tool that is used to follow the evolution of particle properties in a granulation process. Often, the models are based on the assumption of a well-mixed vessel with experimentally determined averaged rate constants for growth and breakage. There has been a development towards flow-dependent mechanistic kernels as pointed out in the recent review by Kumar et al. (2013). For example, Tan et al. (2004) demonstrated how a growth kernel can be derived based on the kinetic theory of granular flow (KTGF). This includes local random particle motion as a time-dependent component compared with the more classical size-dependent aggregation kernel that was developed by Hounslow (1998). A mechanistic model for breakage of a wet granule was derived by Ramachandran, Immanuel, Stepanek, Litster, and Doyle (2009) where they provided a detailed account of all forces that act on a granule and compared these

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results to the internal forces that hold the aggregate together. Such a comparison of internal and external forces has also been applied by Liu, Litster, Iveson, and Ennis (2000) where they used a Stokes deformation number to classify the type of aggregation that occurs at a certain collision energy. This type of analysis shows the importance of resolving stress peaks and estimating local collision velocities because one average value will only capture one of the potentially many regimes that are present in a vessel. Such information could be collected experimentally or by simulation. Nilpawar, Reynolds, Salman, and Hounslow (2006) examined the aggregation efficiency by observing surface velocity fluctuations experimentally to estimate collision frequencies and obtain kernel information.

The flow field in a granulator has been modeled using two different approaches: the discrete-element method (DEM) and continuum modeling of the granular phase. A comparison of the two modeling frameworks exists in Kumar et al. (2013). The DEM model solves a force balance at each time step for individual or small groups (parcels) of particles and resolves individual particle collision dynamics. Gantt, Cameron, Litster, and Gatzke (2006) used DEM simulations to extract collision rates and energies in a disc-impeller granulator for population balance model (PBM) kernels. A disadvantage of the DEM method is that it becomes impractical for industrial-size granulators where the number of particles may exceed 10^6 by orders of magnitude. For dense sheared granular systems with a high number of particles, the number and frequency of collisions leads to unfeasible calculation times unless the collision model is simplified significantly. The continuum approach to particle flows in high-shear granulation was first used by Darelius, Rasmuson, van Wachem, Björn, and Folestad (2008) and Ng, Ding, and Ghadiri (2009), and has been studied further by Abrahamsson, Björn, and Rasmuson (2013); Abrahamsson, Sasic, and Rasmuson (2014a); Abrahamsson, Sasic, and Rasmuson (2014b). Further promising developments, using a rheology model, were made by Khalilitehrani, Abrahamsson, and Rasmuson (2013); Khalilitehrani, Abrahamsson, and Rasmuson (2014); Khalilitehrani, Gómez-Fino, Abrahamsson, and Rasmuson (2015). High-shear granulation is a continuously changing system in which continuous changes in particle properties affect the flow field and vice versa. To resolve collision properties locally at every instance in time would be too computationally expensive even for the use of continuum models. Some discretization (compartmentalization) of the system is needed in time and space. Abrahamsson et al. (2013) investigated the effects of changing particle properties on the flow field of a disc-impeller granulator to study the time discretization of granulation. Compartment models have been developed for spatial discretization and these have been used within many other chemical engineering applications (Bezzo, Macchietto, & Pantelides, 2003; Bezzo, Macchietto, & Pantelides, 2004; Iliuta, Leclerc, & Larachi, 2010; Guha, Dudukovic, Ramachandran, Mehta, & Alvare, 2006; Alexopoulos, Pladis, & Kiparissides, 2013; Le Moullec, Gentric, Potier, & Leclerc, 2010; Gresch, Brügger, Meyer, & Gujer, 2009). A theoretical framework and an automatic zoning process for hybrid compartmental-computational fluid dynamics (CFD) modeling were developed by Bezzo et al. (2004). Bezzo and Macchietto (2004) applied this framework to a batch bioreactor to account for imperfect mixing with regard to viscosity. Another approach was developed by Iliuta et al. (2010), and used compartments to separate gasification and combustion during incineration. Guha et al. (2006) modeled a single-phase stirred-tank reactor and discretized the tank based on the characteristic reaction time scale, which captured the essential features of macromixing. In crystallization, Kougoulos, Jones, and Wood-Kaczmar (2005) continued the work by Bermingham, Kramer, and van Rosmalen (1998), Kramer, Dijkstra, Neumann, ÓMeadhra, and Van Rosmalen (1996), and ten Cate, Bermingham, Derksen, and Kramer (2000) by creating compartments with respect to energy dissipation rate, temperature distribution, and solid concentration from CFD simulations. The well-mixed assumption is commonly used for high-shear granulation. However, the lack of incorporation of flow-field and mesoscale information makes the predictability of models for new types of equipment limited. Yu et al. (2017) attempted to compartmentalize a high-shear granulator. A simple method for finding compartments for high-shear granulation was achieved by using similar heuristics as were used for a crystallizer by Bermingham et al. (1998).

Table 1
Particle properties in CFD cases (Abrahamsson et al., 2013).

CFD case no.	1	2	3
Particle average diameter, d (μm)	59	400	800
Particle–particle restitution coefficient, e	0.9	0.9	0.9
Angle of internal friction, φ (°)	35	32.5	30
Boundary slip, *B.C.*	1	10	100
Maximum packing limit, $\alpha_{s,max}$	0.65	0.65	0.65
Density, ρ (kg/m^3)	750	750	750

We developed a model framework that can discretize (compartmentalize) high-shear granulation based on process relevant parameters in time and space. The model is built up by a sequentially coupled flow field and population-balance solver. We use one flow case to investigate the influence of compartmentalization on the PBM solution. We also look at three flow cases that represent different stages in a granulation, to study changes in predicted collision dynamics of the model over the progress of a granulation. For a well-mixed system, there would be no influence of a spatial discretization of the system. The aim of this work was to determine whether the model shows the influence of spatial and temporal mesoscale phenomena and thereby test the assumption of a well-mixed system.

Model theory

Here we explain the different parts of the developed model, starting with the necessary information on the system that is modeled, and thereafter motivating the choice of a population-balance equation, kernels, and solver method. The CFD model is described and the compartmentalization method that has been developed is presented and the coupling between the CFD and PBM solver is addressed by discussing the relevant flow parameters and their relationship with the PBM kernels.

System to be modeled

The system is based on a 4-L laboratory-scale DIOSNA high-shear granulator with a three-bladed beveled impeller that rotates at 600 rpm. The vessel outline and computational mesh set up are shown in Fig. 1. The properties of the granular material for the CFD cases is set to mimic those of microcrystalline cellulose granules at different stages of granulation (Table 1) (Abrahamsson et al., 2013).

Variations in the properties were made to mimic changes in particle properties as the granulation progresses. This was done according to Abrahamsson et al. (2013) where the CFD cases are set up to mimic the evolution of the stated properties above.

Population-balance modeling

Granulation behavior has been described by a number of competing mechanisms, but has been condensed more recently into three main areas, which are summed up in the review by Iveson et al. (2001). The first area is wetting and nucleation by liquid binder addition; this process is modeled by relating the droplet addition rate from the spray rate to the events of either rewetting a granule