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Computer simulation of granule microstructure formation

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Abstract

The diagenesis (porous microstructure evolution) of granules formed by a layering growth mechanism in a wet granulation process has been modelled. The model includes the packing of primary particles with a given size and shape distribution, and the deposition, spreading, and solidification of binder droplets within the growing granule. The dependence of granule porosity on the binder/solids ratio, primary particle size and morphology, and the rates of binder spreading and solidification has been investigated. The results are presented in the form of structure maps relating volume-averaged microstructure parameters with dimensionless groups including the ratio of droplet spreading and solidification times and the mean time between particle collisions. These graphs can guide the selection of process operating conditions or formulation ingredient properties required to obtain a particular granule microstructure.

Keywords: Agglomeration; Porosity; Microstructure; Diagenesis; Wetting; Product design

1. Introduction

Rational design of structured products and their manufacturing processes requires that a relationship between process and formulation variables and the resulting end-use properties of the product be known (Fung and Ng, 2003). This relationship can be formally represented by the so-called process and property functions (Favre et al., 2002). The process function relates process and formulation variables to a set of parameters that describe the microstructure of the product, and the property function relates the structure parameters to a set of end-use properties. If these functions were known, design of granules would involve inverting the property function for a set of desired end-use properties in order to identify the target granule structure, and then inverting the process function for that structure in order to identify the required granulation process conditions (Rahse and Hoffmann, 2003).

In practice the process and property functions are rarely found in an explicit form and a data-driven approach tends to be adopted (Cryer and Scherer, 2003). The process and property functions are then approximated by correlations between discrete data sets in the formulation-, process-, and end-use property parameter spaces. The size of these data sets is often limited by practical considerations (time constraints, large number of parameters). It would be advantageous to combine the data-driven approach with analytical models or computer simulations that would allow more accurate approximations of the process and property functions, thus more rational product design.

The relation between granule structure and behaviour has been investigated by computer simulations for the case of granule breakage e.g. by Thornton et al. (1999), and for the case of granule dissolution recently by Štěpánek (2004). The relation between the granulation process conditions and granule growth has been investigated computationally e.g. by Talu et al. (2000), and by Goldschmidt et al. (2003). However, the emphasis was on granule size as a product attribute, not on granule structure (i.e., the internal distribution of primary particles, binder, and porosity within the granule). The aim of this work is to demonstrate a methodology for computer simulation of granule structure formation, applicable to wet granulation in the limiting case of growth by layering (i.e., sequential deposition of primary particles and binder droplets on the surface of a growing granule) in the

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absence of deformation due to external mechanical forces (i.e., "low-shear" processes). Such conditions would prevail e.g. in pan granulation (Sherrington and Oliver, 1981), and to certain extent also in drum and fluid-bed granulation.

The elementary processes involved in granulation have been described as nucleation, growth, consolidation, and breakage (Iveson et al., 2001). While these processes are elementary from the point of view of granule size evolution and are suitable for describing granule growth kinetics (population balance modelling), it is preferable to use a different set of "elementary" steps for the purpose of modelling granule structure evolution (diagenesis). In the absence of external deforming forces, these elementary steps are: primary particle packing, binder droplet spreading, and binder solidification. The physics of each of these steps in isolation is relatively well understood and their mathematical models have been independently developed. The structure of random packing of both mono- and poly-dispersed spherical and non-spherical particles has been investigated e.g. by Coelho et al. (1997), Kansal et al. (2002), or Zou et al. (2003). The dynamics of droplet spreading on porous substrates has been studied e.g. by Clarke et al. (2002), Hapgood et al. (2002), or Alleborn and Raszillier (2004). Solidification of droplets has been investigated in detail e.g. by Schiaffino and Sonin (1997) in the case of melts, and Farber et al. (2003) in the case of solutions (both melts and solutions can be used as binders in granulation). The spreading and imbibition of a droplet into a porous medium, accompanied by droplet solidification, has recently been studied by Zadražil et al. (2004). In the present work the simultaneous occurrence of all the three phenomena (particle packing, droplet spreading, and solidification) is considered, and the effect of their rates on the microstructure of the resulting porous medium is investigated.

2. Methodology of granule diagenesis simulation

2.1. Granule structure encoding

The structure of the granule as well as the morphology of primary objects (particles and droplets) is encoded using the volume-of-fluid method (Kothe et al., 1996) generalised to n phases. The granule structure is maintained in a computational unit cell consisting of a cubic grid of $N_x \times N_y \times N_z$ voxels (voxel is an elementary volume element, or spatial "point", defining the resolution at which the structure is represented). Each voxel carries information about the phases present in the volume it covers. Formally, we define the phase function $f_{i,j} \in \langle 0; 1 \rangle$ as the volume fraction of phase j (where $j \in [G, L, S_1, S_2, \ldots]$, meaning gas phase, liquid phase, solid phase 1, solid phase 2, etc.) in voxel with index i. The boundary between a given phase and its surroundings is defined by an isosurface, i.e., a set of points in which the phase function attains a chosen value, typically $f_j = 0.5$.

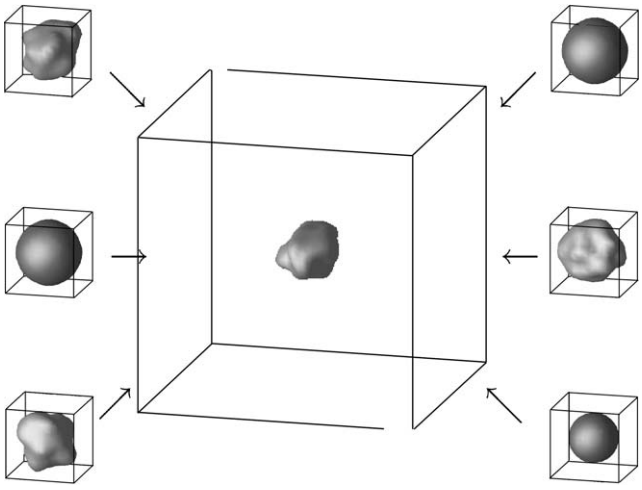

Fig. 1. Computational unit cell in which a granule is constructed by random sequential deposition of binder droplets and primary particles onto a seed particle.

2.2. Granule construction sequence

As mentioned in the Introduction, granule growth by layering is considered in this work. The granule is constructed within the computational unit cell by sequential deposition of primary particles and binder droplets onto a seed particle located in the centre of the unit cell, as illustrated in Fig. 1. The growing granule is stationary within the unit cell but the unit cell itself can be thought of as representing a moving coordinate frame that follows the trajectory of a seed particle through a granulator (or a zone thereof), characterised by a local set of parameters such as the inter-particle collision frequency, temperature, etc.

The introduction of the moving coordinate frame concept means that an abstraction from any particular granulation equipment is made, i.e., we do not specify whether the granule is formed in a pan, drum, fluid-bed, or some other type of granulator. For granule microstructure formation simulation the particle/droplet collision sequence, experienced by the seed particle in the granulator, is important. The collision sequence ("granulation script") is a list of the times of all collisions that the particle experiences during its residence time in the granulator, and the identity of the colliding objects (primary particles and binder droplets—their size, shape, and material). Computationally the granulation script can be obtained e.g. by an explicit DEM simulation of granular flow (Goldschmidt et al., 2003) within a particular type of granulation equipment, or it can be generated as a pseudo-random sequence by assuming certain statistics of inter-particle collisions derived from theory (e.g., from the kinetic theory of granular flow). In this work a parametric study with respect to the mean time between particle collisions is performed.

Overall the granule diagenesis simulation proceeds according to the algorithm shown in Fig. 2. At time $t = 0$ an initial seed particle is placed into the centre of the unit cell,