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Simulation of the influence of surface tension on granule morphology during spray drying using a simple capillary force model

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ABSTRACT

Spray drying is an important process in powder technology as it transforms fine primary powder into processable granules. While various investigations of granule formation exist, the role of surface tension on the evolution of the granule morphology has not been investigated in much detail. This work closes this gap by using numerical simulations on granule level. In our simulation of suspensions of solid particles, we use the Discrete Element Method (DEM) in combination with a CFD solver. Due to the free liquid surface, a multiphase solver is needed including a force model for the particle-surface contact. As a result, our work gives insight into the granule formation and reveals a clear non-linear dependency relating high surface tension to dense granules.

1. Introduction

Spray drying is an important process in powder technology, e.g. in the ceramic industry. In the latter, the raw material often is available in the form of a very fine primary powder, which is highly adhesive due to the small particle size. Therefore, it has to be transformed to coarser granules, which is often done using the spray drying process [1]. In order to achieve good handling and compacting properties of the granulate, the individual granules are often required to be spherical and dense, allowing for good quality of the final product. Hollow granules, on the other hand, can create voids due to bad compressibility [2], which could create defects inside the sintered product and reduce its strength [3].

Numerous experimental investigations showed that the stability of the suspension has a major influence on the granule morphology evolving during spray drying, i.e. well dispersed suspensions often lead to hollow granules [4–7]. Hence, the interparticle forces and the solvent-particle interaction play an important role in the evolution of the granule morphology. In addition, the capillary force exerted on the primary particles by the receding liquid surface creates a compressive drying stress inside the particle network [8]. Therefore, the capillary force may also have a significant effect on the granule development. However, this force has not been studied experimentally in the context of spray drying of ceramics, which might be caused by the difficulty of varying the surface tension without changing other important suspension parameters.

The experimental investigations have been supplemented by a number of simulation studies on granule formation during spray drying. The different methods proposed in the literature are discussed in the following.

Basic approaches use one dimensional models of heat and mass transfer [9,10] to describe the drying of rotationally symmetric drops depending on the suspension parameters and process conditions. By taking into account transport through porous media in a growing outer zone of the drop, it is possible to even simulate shell formation to some extent [11]. These models can be used to calculate the drying kinetics, which may then be used in process scale simulations for the tracking of drying granules through the drying chamber. However, more detailed information about the evolution of the granule morphology on particle level cannot be gained by such mean-field models.

A more enhanced class of models uses a stochastic description of the suspended particles based on a population balance [12]. This continuum mechanical approach allows e.g. for distinguishing between wet and dry shell and enables the simulation of shell growth [13] and even provides some insight in the granule morphology [14].

In contrast to the above methods, the discrete element method (DEM) may be used to study the morphology of particle based systems in much more detail [15], as it describes the solid content as many individual particles. E.g., Greil et al. [15] used it to simulate the electrophoretic deposit of solids on a substrate and Breinlinger et al. [16] simulated the deposit formation at the drying of suspensions on a substrate using a similar method. Recently, the DEM has been applied to study the granule formation during spray drying [17,18] by coupling it to computational fluid dynamics (CFD). These simulations showed a trend to

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hollow granules for higher temperatures [17] and higher binder content [18].

For the simulation of coupled CFD-DEM problems, the interaction between the fluid and particle phase is described by coupling forces, e.g. drag, buoyancy, etc. If the fluid phase itself consists of two continuous phases, i.e. gas and liquid as in the case of spray drying, additional coupling forces are needed at the liquid surface. This capillary force and the resulting surface curvature has been studied previously and models for thin layers of suspensions on a substrate can be found in literature [19,20]. A full description of the contact between a particle and the liquid surface by this model requires knowledge about either the external conditions (i.e. present forces) or the geometry (i.e. neck curvature, height, length, etc.). However, in our case, the neck geometry and the capillary force are unknown.

None of the aforementioned works – neither experimental nor numerical – investigated the role of surface tension on the formation of spray dried ceramic granules. Therefore, in this work, we aim to investigate this in detail. For this purpose, we implemented a coupled CFD-DEM solver, which is based on OpenFOAM [21], an open source CFD software package. We simplify the coupling conditions by neglecting local curvatures in order to be able to simply derive the required coupling force from the pressure gradient and line tension acting on the particle.

2. Methods

2.1. The Discrete Element Method

In this work, we use the Discrete Element Method (DEM) [22] to describe the granular media. This method describes the motion of an individual solid particle based on Newton's equations of motion

$$\dot{\mathbf{p}} = \mathbf{F}, \tag{1}$$

where $\mathbf{p}$ is the linear momentum and $\mathbf{F}$ is the sum of external forces. The DEM model treats each solid particle as an individual particle and calculates their trajectories by integrating Eq. (1) in time

$$m_i \dot{\mathbf{v}}_i = \sum_{n,j} \mathbf{F}_{ij}^{n} + \mathbf{F}_i^{coupling}, \tag{2}$$

where m_i and $\mathbf{v}_i$ denote the mass and velocity of particle i, respectively, $\mathbf{F}_{ij}^{n}$ represents the n different interaction forces between particles i and j and $\mathbf{F}_i^{coupling}$ is the coupling force for momentum exchange with the fluid phase. Brief general reviews of DEM contact forces are available e.g. from [23] and [24]. In the following, we describe the contact force model used in our approach. The rotation of particles was not taken into account as we suppose that it would be only of minor influence on the particle reorganization and [25] showed that a rotational degree of freedom can actually hinder the development of a steady state system due to numerical reasons. The friction coefficient was used to mimic non-spherical particle shape.

2.2. Contact force model

Contact forces between two individual DEM particles with radius r are applied whenever these particles have a positive overlap

$$h_{ij} = x_{ij} - (r_i + r_j) \tag{3}$$

where x_{ij} is the magnitude of the distance $\mathbf{x}_{ij} = \mathbf{x}_i - \mathbf{x}_j$ between the center points $\mathbf{x}_i$ and $\mathbf{x}_j$ of the particles. The contact model used in this work consists of elastic repulsion, repulsion dissipation, cohesion and friction. The elastic repulsion is described following [26] as

$$\mathbf{F}_{ij}^{e} = \left(2/3\bar{E}\sqrt{\bar{r}_{ij}}h_{ij}^{3/2}\right)\hat{\mathbf{x}}_{ij}, \tag{4}$$

in normal direction $\hat{\mathbf{x}}_{ij} = \mathbf{x}_{ij}/|\mathbf{x}_{ij}|$. Here $\bar{r}_{ij} = r_i r_j/(r_i + r_j)$ represents the effective particle radius and $\bar{E} = E/(1-\nu^2)$ is the effective Young's modulus with the Poisson number ν. The viscous part of the collision, which represents dissipation by plastic deformation and noise emission, is modeled as a velocity dependant damping force [27]

$$\mathbf{F}_{ij}^{v} = -\left(\gamma\sqrt{\bar{r}_{ij}h_{ij}}\mathbf{v}_{ij}\cdot\hat{\mathbf{x}}_{ij}\right)\hat{\mathbf{x}}_{ij}. \tag{5}$$

The damping parameter γ is determined empirically. Cohesion between the particles is modeled based on the overlap area A of the contact

$$\mathbf{F}_{ij}^{c} = -(\omega_A A)\hat{\mathbf{x}}_{ij}, \tag{6}$$

where ω_A is the cohesive force per unit area or work per unit volume. Friction between the DEM particles is modeled following [22] as a tangential force

$$\mathbf{F}_{ij}^{f} = -min\left[k\sqrt{\frac{h_{ij}}{\bar{r}_{ij}}}|\xi_{ij}|, \mu\left|\mathbf{F}_{ij}^{e} + \mathbf{F}_{ij}^{v}\right|\right]\frac{\xi_{ij}}{|\xi_{ij}|} \tag{7}$$

based on the relative displacement ξ_{ij} of the initial contact points, its magnitude is governed by the tangential spring constant k and the friction parameter μ.

2.3. The volume of fluid method

The volume of fluid method (VoF) for multiphase flow [28] can be used to describe the motion of two immiscible fluids. It can be combined with the discrete element method by using locally averaged coupling [29] which will be described in Section 2.4. In this context, the motion of two incompressible fluid phases, e.g. water and air, carrying solid particles is governed by the averaged Navier–Stokes equations

$$\nabla\mathbf{v} = 0 \tag{8}$$

$$\rho\frac{D\mathbf{v}}{Dt} = -\nabla p + \eta\nabla^2\mathbf{v} + \sigma\kappa\frac{\nabla\alpha}{|\nabla\alpha|} + \rho\mathbf{g} + \mathbf{f}_c, \tag{9}$$

where $\mathbf{v}$ is the velocity, p is the pressure and $\mathbf{g}$ is the gravity. Surface tension is included by the term $\sigma\kappa\nabla\alpha/|\nabla\alpha|$, where σ is the surface tension coefficient and κ is the curvature of the liquid surface. The term $\mathbf{f}_c$ represents volumetric coupling forces from the solid particles and will be discussed later. The average density ρ and dynamic viscosity η in each cell are computed as

$$\rho = \rho_l\alpha + \rho_g(1-\alpha), \quad \eta = \eta_l\alpha + \eta_g(1-\alpha), \tag{10}$$

based on the phase function α, which determines the ratio of water in each cell. The indices l and g represent the liquid and gas phase, respectively. The phase function ranges from 0 to 1 and is balanced by its own transport equation

$$\frac{\partial\alpha}{\partial t} + \nabla\cdot\mathbf{v}\alpha = \dot{\alpha}_{ev}, \tag{11}$$

where the $\dot{\alpha}_{ev}$ is a source term which extends the established VoF scheme to incorporate evaporation during the drying process (see Section 4). This source term has the form

$$\dot{\alpha}_{ev} = -\frac{1}{2}D\kappa\frac{(c_f - c_\infty)}{\rho_l}|\nabla\alpha|, \tag{12}$$

where D is the diffusion coefficient and c_f and c_∞ are the vapour concentrations in the evaporation film and at ambient conditions, respectively. For the sake of boundedness of the phase function and stability of the solution, the application of $\dot{\alpha}_{ev}$ was limited to regions where $0 \le \alpha \le 1$. The