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Modelling of partially wet particles in DEM simulations of a solid mixing process

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ABSTRACT

It is the basic objective of a solid mixing process to avoid any segregation effects. Especially free-flowing bulk solids with weak interparticle adhesive forces tend to percolating segregation. Liquid addition, which is common in mixing granulation processes, will usually reduce the particle segregation due to pendular liquid bridge networks. However, liquid addition can result in a change of the segregation mechanism. For example: water may accumulate selectively in one component when hydrophilic and hydrophobic solids need to be mixed. Heteroagglomerates that consist mainly of one solid component may arise and degrade the final product quality. In this work the Discrete Element Method (DEM) is applied to investigate mixing and segregation of moist particulate solids in more detail. During particle collision liquid is transferred from one to the other contact partner until the liquid is uniformly distributed in the bulk. The available liquid bridge models assume complete particle wetting or neglect that the mixing components may differ in their contact angle. We developed the liquid contact dispersion model further in order to realize partial particle wetting that includes drop formation on the particle surface. A comparison between the common film model and the proposed model shows an improvement in experimental validation when the partial particle wetting model is included in the DEM simulation. The assessment of these models is carried out by evaluating mixing efficiency and liquid distribution.

1. Introduction

In high shear granulation [1] or tablet coating [2], liquid is often atomized through nozzles and sprayed onto dry particles. To protect the gastric mucosa from aggressive drugs or even for a delayed release of active ingredients, the surface of solid pharmaceutical tablets is often coated in mixing drums. The required tablet quality is significantly influenced by the intra- and inter-uniformity of the coating. Intra-uniformity is the variation of the local layer thickness, while inter-uniformity describes the variation of the coating composition between different tablets. Simulations are becoming increasingly important in order to investigate intra- and inter-uniformity and to optimize coating or mixing processes. Usually, the evaluation of injection and process control is carried out by post-processing algorithms based on particle positioning, orientation and residence time within the spray zone [3,4].

In pharmaceutical high shear granulation, a filling material like lactose and microcrystalline cellulose [5] has to be briefly dry-mixed with a low-concentrated active ingredient in order to increase the particle or granule size by subsequently adding a binder [6]. This improves the processability, for example the filling of capsules. Not only the granule size distribution, but also the distribution of liquid within the powder is influenced by the operating parameters of the high shear mixer. The granule size and the liquid distribution depend on the rate of liquid addition, equally does the product movement behavior in the mixing vessel and the size of the spraying zone [7]. In the case of uneven liquid distribution, over- or under humidified areas may arise in the mixing components, which in turn may result in broad granule size distributions and inhomogeneous agglomerates [8]. In order to improve the uniformity of humidification, formulation or process variables have to be adjusted experimentally. In case of a formulation change the fluid adhesion may be increased by maximizing surface tension, minimizing the contact angle, or decreasing fluid viscosity. This is achieved, for example, by changing the surfactant concentration in the binder or by coating the powders with better wetting materials [9]. Upstream grinding minimizes surface roughness that improves the wetting of particulate solid. A higher process temperature reduces fluid viscosity which contributes to a better fluid distribution [10].

In analogy to solid mixing mechanisms, the liquid distribution is affected by the mixing tool induced convective transport as well as by mechanical contact dispersion [11]. The term 'contact dispersion' defines interparticle transfer of liquid due to mechanical particle contacts. Dry particulate contacts can be calculated on the basis of the Hertz model [12,13] and be simulated numerically with Newton's law of motion (Discrete Element Method). However, the models for DEM simulations of moist cohesive particle systems are still in the early

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stages. Mikami, Kamiya and Horio [14], Willett, Adams, Johnson and Seville [15], Soulie, Cherblanc, El Youssoufi and Saix [16], Butt and Kappl [17], Lian and Seville [18] and Schmelzle and Nirschl [19] developed different liquid bridge models, that have in common that the liquid bridge is stable up to a critical particle separation distance at which the bridge ruptures. Based on the numerical solution of the Young-Laplace equation mathematical expressions for the liquid bridge force and rupture distance were derived for mono- and polydisperse material and materials with different wetting behaviors. As one of the first, Shi and McCarthy [20] presented a thin film model in which the liquid is distributed evenly on the particle surface. After the bridge rupture the liquid is redistributed on the contact partners according to a parabolic bridge profile. Mani, Kadau and Herrmann [21] calculated the shear-induced liquid transport in undersaturated, monodisperse particulate solids based on the thin film model explained with continuum mechanics. Mohan, Kloss, Khinast and Radl [11] compare four models describing liquid transport with variable liquid layer thicknesses and flow rates for dynamic bridge filling within a shear field. In all the mentioned works, the assumption is made that the liquid immediately spreads over the entire particle surface. However, this assumption is only justified for extremely well-wetting materials with low contact angles. In case of a less hydrophilic material wetted with water, a non-ideal liquid film is formed. Depending on the contact angle the size of the droplet and the covered particle surface will be different.

In order to incorporate partial wetting in DEM Simulations, comprehensive model extensions like local liquid storage on the particle surface have to be carried out. Washino, Miyazaki, Tsuji and Tanaka [22] used a discretization method [23] to investigate the intra- and inter-uniformity of the coating of monodisperse particles in a mixing drum. In this work the same discretization method is used to further develop liquid dispersion in DEM simulations. Partial wetting and the formation of drops are implemented, which should lead to physically improved liquid distributions and higher accuracies in experimental validation.

2. Materials and methods

2.1. Mixer and sampling

Experiments and simulations are carried out in a self-made vertical mixer (Fig. 1). The mixing vessel has a diameter of 112 mm resulting in a maximum filling volume of approximately 0.7 l. During experiments the radial fixed sampling holes (Fig. 1 (left)) are masked and not considered in the simulations. At the bottom of the mixing vessel a three-bladed mixing tool operates at different rotational speeds. The experimental validation (Section 4.2) is carried out at 36 rpm.

The procedure for statistical evaluation of a mixture requires an adequate sampling method (Fig. 2). This involves a minimum number of particles in the sample, in order to detect a low-concentrated additive with a given probability in the sample volume. The minimum number of particles per sample can be estimated [24]. In our application the following applies: with $S = 95\%$ confidence level, the sample composition does not deviate more than $y_s = 7.5\%$ from the target composition if a particle number Z of at least

$$Z = \left(\frac{z(S)}{y_s}\right)^2 \cdot \frac{1-q_{target}}{q_{target}} = \left(\frac{1.65}{0.075}\right)^2 \cdot \frac{1-0.5}{0.5} \approx 484 \tag{2.1}$$

appears in the sample. The limit factor $z(S)$ depends on the selected confidence level. Due to the low number of particles ($N_P = 100{,}000$) a higher probability is not achievable in the context of validation. Additionally the number of samples N_s has to be defined. If the empirical variance s^2 has a probability of $S = 95\%$ to be maximally 1.75 times larger than the true variance σ^2, the required number of samples $N_s = 16$ can be determined with the Chi-square distribution χ_S^2:

$$\frac{16}{26.3} = \frac{f}{\chi_S^2(S;f)} > \frac{\sigma^2}{s^2} = \frac{1}{1.75}. \tag{2.2}$$

The values of the Chi-square distribution χ_S^2 and the corresponding degree of freedom f were taken from the textbook by Stieß and Ripperger [25]. In addition to the sample size $Z_{N,\,min}$ and sample number N_s, the location and frequency of sampling must be defined as well. For a representative determination of the mixing efficiency, the samples have to be distributed randomly over the entire mixing vessel. This guarantees a holistically assessment of the batch and excludes any systematic signs of segregation which would falsely suggest inferior product qualities. For this purpose, 16 sampling points were randomly distributed over the entire mixture (Fig. 2a)). The positions were noted and then applied for each sampling time. Depending on the sample position, the rotation of the vessel cap (Fig. 2b)) and the insertion depth of the sampler must be adjusted (Fig. 2c)). With this combination of random and regular sampling, the holistic assessment of the batch is guaranteed in both, experiment and simulation. Due to the high effort, the frequency of sampling is significantly lower in the experiment but nevertheless high enough to eliminate systematic sample errors.

The sample compositions are determined by sieving and counting analysis. If the sample composition q_i is known, the relative standard deviation RSD can be calculated as a mixing efficiency measure according to:

$$RSD = \frac{\left(\frac{1}{N_s}\sum_{i=1}^{N_s}\left(q_i(t)-q_{target}\right)^2\right)^{0.5}}{q_{target}}, \tag{2.3}$$

where $q_{target} = 0.5$ is the predefined number composition. Ideal homogeneity cannot be reached because of random particle motions occuring in solid mixing processes. The best achievable quality of mixing is characterized by the stochastic mixture RSD_Z, which is function of the sample size:

$$RSD_Z = q_{target}\left(1-q_{target}\right)\frac{1}{Z}. \tag{2.4}$$

2.2. Materials

Monodisperse glass beads are used as a model product. The filling fraction has a particle radius of approximately $r_i = 0.92$ *mm* and is transparent. The additive component is half as large as the filler ($r_j = 0.46$ *mm*) and colored red. A different wetting behavior with water arises from the red coating of the individual particles. The static contact angles θ can be measured by microscope images of a liquid bridge between two particles in contact (Fig. 3 (left)). For this purpose, the toroidal approximation was adapted manually to three images and evaluated using the intersection angles between particle and liquid bridge (Fig. 3 (right)). The red particle shows a contact angle of

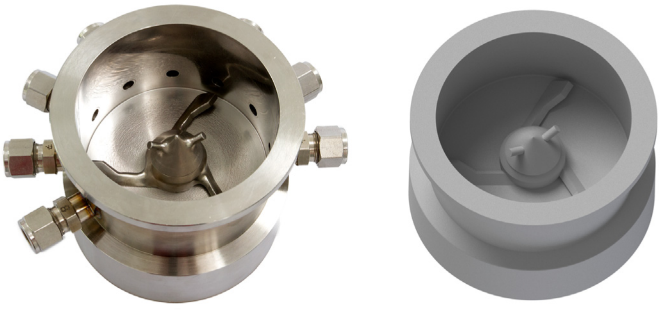

Fig. 1. The vertical mixer (left) and its CAD model (right) used for the experimental validation and the simulation.