Dynamic modeling of particle size and porosity distribution in fluidized bed spray agglomeration

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Abstract

Population balance modeling is a powerful to tool for the simulation of particle formation processes such as fluidized bed spray agglomeration (FBSA), where agglomerates are formed from primary particles by binary aggregation. In addition to the agglomerate volume, agglomerate porosity is important for the characterization of product particles since it affects various physical properties. In this contribution a new method is proposed to incorporate porosity into a population balance model. To this end an empirical relationship between the volume of solids within the agglomerate and its total volume including voids is utilized. Since this relationship is different for primary particles and agglomerates, respectively, the evolution of the number density distribution for both types of particles is modeled. The proposed population balance equations are validated by fitting three kinetic parameters to experimental data and comparing measured particle size distributions with model predictions, showing good agreement.

**Keywords**: population balance modeling, agglomerate porosity.

* 1. Introduction

Continuous fluidized bed spray agglomeration (FBSA) is a size enlargement process for the production of solid particles. Here, a liquid binding agent is sprayed onto the surface of a fluidized particle bed. After particle collision and binder drying, new agglomerates are formed. In the continuous process configuration (Fig. 1), subject of this contribution, there is a constant primary particle feed and product particle withdrawal.

Figure 1: Process scheme with particle feed, agglomeration in the process chamber and withdrawal

Physical particles properties, such as the volume and porosity of the resulting agglomerates are important, since they determine agglomerate characteristics, such as solubility, mechanical strength and flowability, and thereby the economic value of the product. For process monitoring, control and optimization, it is therefore crucial to have accurate models describing their evolution. Typically, multi-dimensional population balance equations (PBEs) are used as macroscale models (Iveson, 2002), describing the evolution of particle distributions with internal properties by means of partial differential equations. Alternatively, stochastic Monte-Carlo methods can be applied to model certain subprocesses such as particle wetting, collision and drying in detail at the expense of increased computational effort (Singh and Tsotsas, 2019).

In previous contributions, the authors developed one-dimensional population balance models (PBMs) for continuous FBSA processes and fitted parameters to experimental particle size distributions (Otto et al., 2021, 2022). In these contributions however, the porous nature of agglomerates has only been incorporated by assuming an average reduced agglomerate density, resulting in models that do not accurately capture the influence of porosity on the measured particle size distributions. In this contribution a new approach to population balance modeling for the FBSA production of agglomerates described by their volume and porosity is presented. To this end, an empirical relationship between the two descriptors presented in Singh and Tsotsas (2019) and Strenzke et al. (2022) is utilized. In contrast to models in the literature (Iveson, 2002; Poon et al., 2008), the approach presented here allows for the use of only a one-dimensional PBE. Since primary particles and agglomerate have different characteristic porosity distributions, primary particle and agglomerate populations are considered separately, resulting in two modeled distributions. The proposed system of PBEs in validated by fitting its kinetic parameters to experimental particle size distributions. Additionally, model implications for application in process control are discussed.

* 1. Process model
		1. Agglomerate porosity distribution

In the following a model for the relation between agglomerate volume and porosity is presented. It is assumed that an agglomerate consists of a solid and a gaseous (air/void) fraction. Liquid components are neglected, due to their low volume fraction. The porosity of an agglomerate is defined as the ratio between gas volume and volume packed by

solid material . Since the dried binder volume is small compared to the total volume of all primary particles it is neglected and is given by

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| , | (1) |

with the number of primary particles and the primary particle diameter . Here it is assumed, that primary particles are monodispersed and spherical. Although this is a simplifying assumption, the influence of polydispersity of primary particles on agglomerate porosity can be considered as negligibly small (Singh and Tsotsas, 2022). Using , the total volume of the agglomerate (including voids), the porosity is given by

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| , | (2) |

From the above equation, it follows that , or can be used equivalently to fully characterize an agglomerate if is given. Singh and Tsotsas (2019) provide the following empirical relationship between the porosity and the number of primary particles

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| --- | --- |
| , | (3) |

with parameters , the so-called fractal dimension, and , the prefactor. In order to obtain this relationship, is defined as the volume of an equivalent sphere with radius based on the gyration radius of the agglomerates. The morphological parameters and are determined empirically by correlating the number of primary particles in an agglomerate to the measured gyration radius for a number of agglomerates. Strenzke et al. (2022) determined and for a continuous FBSA experiment with constant process conditions. Additionally, Singh and Tsotsas (2019) showed that it is possible to linearly correlated the morphological parameters to process conditions, namely the gas inlet temperature and the binder concentration . By rearranging Eqs. (1), (2) and (3), the following power law relationship between and is obtained

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| , | (4) |

Note that the above equation assumes that and can take continuous values, whereas Eq. (1) implies discrete values due to the discrete nature of . Thus, Eq. 4 can be seen as a continuous extension of the underlying discrete map. Furthermore, it is important to remember that Eq. (4) only hold for agglomerates. The primary particle porosity is constant in general, for the FBSA experiments with non-porous glass beads considered here .

*2.2 Population balance equation*

In Otto et al. (2021) a population balance model for the evolution of the particle size distribution during a continuous FBSA process has been presented. Therein, the particle population is represented by the number density distribution (NDD) with solid volume as internal coordinate. In order to describe the agglomeration kinetics, parameters of an empirical agglomeration kernel have been fitted to measurements of particle size distributions (Strenzke et al., 2020), where the particle size distribution is measured by means of image analysis (Camsizer X2, Microtrac Retsch). The measured particle size is represented by a diameter , which can be transformed to volume using the standard assumption of spherical particles. This measured volume, however, includes voids and therefore is not the volume but . To account for this gap between measured NDD and modeled NDD , the literature provides more detailed population balance models. Usually, the NDD is augmented by an additional internal variable, e.g., the agglomerate porosity (Iveson, 2002) or the void/gas volume within the agglomerate (Poon et al., 2008), i.e., . According to the previous subsection, any of the variables , or can be used as second internal variable, since one of these determines the others. In both Iveson (2002) and Poon et al. (2008) it is assumed that the solid volume, the volume of gas and therefore the total agglomerate volume is conserved during binary aggregation which directly determines the resulting agglomerate porosity. However, for the FBSA process considered in this contribution, the nonlinearity of Eq. (4), shows that since is conserved during agglomeration, is not. Furthermore, Eq. (4) shows that and are not independent variables, eliminating the need for a multidimensional PBE. In fact, the derivative of Eq. (4) given by

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| , | (5) |

defines a transformation between and by

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| , | (6) |

which is the desired equation connecting the measured and the modeled NDD for a population of agglomerates.

From the discussion at the end of the previous subsection it is clear that Eq. (5) and thus Eq. (6) only holds for actual agglomerates. For primary particles, on the other hand, the solid volume is equal to the total particle volume, i.e.

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| --- | --- |
|  | (7) |

Thus, for a general particle population, it is necessary to distinguish the NDDs for primary particles and agglomerates with

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| --- | --- |
|  | (8) |

When computing the measured NDD corresponding to Eq. (8), the different transformations have to be considered, resulting in

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|  | (9) |

The different transformations and their effect on the total particle size distribution are showcased in Fig. 2, where example NDDs of a primary particle and an agglomerate population are shown depending on and respectively. While the primary particle NDD is the same in both coordinates, the agglomerate NDD appears flattened and shifted towards higher volumes in the -coordinate.

In order to obtain PBEs for the evolution of and , aggregation between pri-mary particles and primary particles (pp), agglomerates and primary particles (pa) as well as agglomerates and agglomerates (aa) have to be modeled separately, as presented schematically in Fig. 3. The respective population balances are obtained by balancing the particle accumulation at volumes with the respective fluxes

Figure 2: Modelled NDDs with respect to (blue) and measured NDDs with rescpect to .

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| --- | --- |
|  | (10) |
|  | (11) |

Here, is the primary particle feed and as well as are modeling the particle withdrawal. For the specific modeling of these terms, we refer the reader to Otto et al. (2021). The variables B and D denote particle birth and death respectively, given by the following equations (Hulburt and Katz, 1964)

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| --- | --- |
|  | (12) |

and

|  |  |
| --- | --- |
|  | (13) |

The agglomeration kinetics are described by the agglomeration kernel , representing the volume dependent rate of agglomeration. In this standard form of the agglomeration term, it is assumed, that the kernel only depends on the solid volume part of the agglomerate and therefore on the agglomerate mass. This is a simplifying assumption, since the rate of agglomeration depends on multiple factors such as the binder coverage of the particle surface which itself may be increased by higher total agglomerate volume.

Various kernel functions, both mechanistic and empirical, with varying complexity are presented in the literature. Kernel identification results from Otto et al. (2022) suggest that the agglomeration behavior is different for the three types of particle interaction described above. Therefore, in this contribution we distinguish between the kernel , and . The simplest kernel model is the constant, i.e., size and time independent kernel function which will be adopted here.

Figure 3: Agglomeration between a) primary particle and primary particle, b) primary particle and agglomerate and c) agglomerate and agglomerate.

*2.3 Model discussion*

The presented correlations of number of primary particles and agglomerate porosity by fractal dimension and prefactor have been applied to FBSA in batch configuration (Singh and Tsotsas, 2019) and one continuous experiment (Strenzke et al., 2022). In both settings process conditions have been kept constant. For the general case of non-constant process

conditions, the morphological descriptors and will change and the power law of Eq. (3) and thus Eq. (4) will generally not be valid. In this case there will be no unique, i.e., time-independent, relationship between and and a population balance equation with two internal coordinates may be required to capture the dynamics of the process. This could be circumvented by assuming a slow and uniform change of and depending on a process condition , e.g.

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| --- | --- |
|  | (14) |

In order to apply the PBEs in a model-based process control context, the state, i.e., primary particle and agglomerate distributions have to be measured. Current measurement technology, however, does not distinguish between primary particles and agglomerates. Therefore, either advanced measurement technology has to be developed, e.g., image-based distinction or the distributions have to be estimated using model-based observer or soft-sensor techniques.

* 1. Model validation

In order to validate the proposed model, the three yet unidentified kernel parameters are fitted to the continuous FBSA experiment presented in Strenzke et al. (2022). In accordance with Otto et al. (2021) the focus is on the steady state particle size distribution measured after 120 minutes. For the parametrization of the feed and withdrawal terms in the population balance as well as the numerical discretization of the PDEs, we refer the reader to Otto et al. (2021). Using the Matlab implemented surrogate optimization method (Gutmann, 2001) to minimize the -error between experimental and calculated distribution, the parameters , and are found. In Fig. 5 the measured and calculated normalized volume distributions

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|  | (15) |

in steady state are presented, showing good agreement.

* 1. Conclusion and outlook

In this contribution a novel population balance model for fluidized bed spray agglomeration has been presented. In contrast to previous contributions, it is distinguished between solid agglomerate volume and total agglomerate volume including voids. The solid volume part of the agglomerate is utilized as internal coordinate of the modeled number density distribution, resulting in a classical one-dimensional population balance equation. In order to derive the measurable particle size distribution which is based on the total agglomerate volume, an empirical correlation between solid and total volume is incorporated. Since this correlation, based on the particle fractal dimension, is different for primary particles and agglomerates, both populations are modeled separately. The presented model is validated by fitting three kinetic parameters and comparing the particle size distribution predictions with the measurements of a continuous experiment. The results are in good agreement for the steady state distributions.

Figure 4: Comparison between experimental (blue) and calculated (red) normalized volume distribution in steady state.

Future research directions are twofold. By using correlations of different process conditions to the fractal dimension, their influence on the agglomerate porosity will be modeled and validated with additional experiments. Furthermore, the presented model will be used as basis for model-based process control of agglomerate size and porosity.

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