Microscopic and macroscopic modeling of particle formation processes in spray fluidized beds

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Summary

The present work deals with modeling of particle formation in batch spray fluidized bed processes, using different methods. First, two process models for coating and layering granulation as well as agglomeration of amorphous particles based on a Monte Carlo method are presented. This method offers great advantages over common modeling approaches. In case of coating and layering granulation, a more detailed description of product properties and their distributions can be obtained. For the first time, the process kinetics for agglomeration of amorphous particles can be described while the influence of process and material parameters is directly taken into account. The presented Monte Carlo models describe layering and agglomeration as a result of processes occurring on the single particle scale such as droplet deposition, binary collisions, droplet drying, size enlargement, and breakage. The influence of operating conditions and material parameters is discussed and the models are validated using theoretical approaches and experimental data. Second, a process model based on population balances and a heat and mass transfer model for spray fluidized beds is presented. This model is used to characterize the border between layering and agglomeration by combining the Stokes criterion and a new dynamic model describing the wet surface fraction of the particles. The influence of operating conditions and material parameters on the dominant size enlargement mechanism is presented in a simulation study. Further simulations and experimental data are used to provide a new classification of the size enlargement mechanisms based on the probability of successful collisions.

Extended abstract

Particulate products represent the majority of all chemical products manufactured in industry. Estimates of the relative amount vary between 60 % for bulk solids according to Schulze [1] and up to 80 % when liquid and solid mixtures, aerosols, and materials that contain gas bubbles are further taken into account, see Merkus [2]. Particles play an important role in many fields such as agriculture, and the chemical, pharmaceutical, and food industries. Examples of particulate products manufactured in these fields are fertilizers, catalysts, detergents, tablets containing an active pharmaceutical ingredient, cosmetics, milk powder, soup and beverage powders. Particulate goods can be produced from liquids containing solid material, which may be solutions, suspensions,

or melts. This process step is performed to reduce transportation costs, simplify handling and post-processing, and to prolong shelf life in comparison to the liquid state of the material.

The present dissertation deals with spray fluidized bed processes such as coating, layering granulation, and agglomeration induced by spraying a solution or suspension onto a fluidized particle bed. In this type of equipment, particle formation and drying can be realized in either batch or continuous mode in a single process step with generally high heat and mass transfer rates. In coating and layering granulation, the main goal is the production of single particles consisting of a core particle covered by a solid layer. The solid layer is built around the core particles by repeated deposition and drying of droplets. In both processes, the size enlargement mechanism is identical and called layering. Generation of dust particles or agglomerates is generally undesired. In agglomeration, small powder particles are transformed into larger particles called agglomerates, consisting of several primary particles bound together by different mechanisms. Since agglomerates are formed by repeated droplet deposition, collisions between (partially) wet particles, and liquid bridge drying in spray fluidized beds, the dominant binding forces are provided by material bridges. Dust generation and layering are undesired since in both cases material for generating liquid and solid bridges is lost. Depending on the molecular structure of the particle material, different mechanisms leading to agglomeration can be observed, see Palzer [3, 4]. If the primary particles are crystalline, typically some binder material needs to be sprayed to create material bridges. If amorphous materials are used, the glass transition temperature may be locally decreased by spraying. The result is a sticky, rubbery material, which is able to form viscous bridges when particles collide at these spots [4, 5]. This is observed in various agglomeration processes used in the food industry, where water or aqueous solutions are sprayed on moving, water-soluble amorphous particles [4].

Important properties defining the product quality of particles produced by coating, layering granulation, and agglomeration are the particle size distribution, shape, density, porosity, flowability, strength, redispersion behavior, and the moisture content [6]. To ensure intended product performance, these properties have to be within certain specifications.

Important properties of coated particles are coating mass uniformity and the morphology of the coating [7, 8]. Coating mass uniformity refers to the variation of coating mass among individual particles and is therefore called an inter-particle property. The variation of the coating mass is especially important when applying active ingredients in the coating. Coating morphology refers to the distribution of the coating thickness on individual particles, which is an intra-particle property. The morphology is further characterized by coating layer porosity and the existence of fissures or gaps. Beyond that, surface coverage is an important property. These properties also influence product performance when the coating is applied for a sustained release application or as a protective layer.

The main property of particles produced by layering granulation, influencing product performance is the particle size distribution [9]. In case of fertilizers, usually a narrow size distribution is preferred to ensure uniform distribution of the fertilizer on the field. Additionally, segregation effects are minimized when producing mixtures of fertilizers [9]. The dissolution behavior depends on the

size and porosity of the particles. The porosity should be relatively low to achieve a certain strength minimizing abrasion [9] and to ensure slow dissolution behavior [6].

Important properties of agglomerates are the particle size distribution, redispersion behavior, and compressibility. Fundamentals of redispersion of agglomerates have been stated by Pfalzer et al. [10]. The process of redispersion can be divided into wetting, sinking, and breakup of the agglomerates into their primary particles. In order to achieve fast and complete redispersion, agglomerates should consist of a large number of bridges characterized by relatively weak strength. The bridges should be weak to facilitate dissolution, while a large number of bridges ensures mechanical stability of the agglomerates, e.g., during transport or packaging. Powders are agglomerated prior to tableting to improve compactibility and the strength of the resulting tablets [4, 11]. The flowability of the agglomerates needs to allow accurate dosing when tableting particles including active ingredients and minimize segregation prior to tableting to ensure tablet uniformity [12].

In particle formation processes in spray fluidized beds, usually both size enlargement mechanisms (layering and agglomeration) occur simultaneously. However, in order to achieve the required product quality, only one mechanism depending on the application of the product is desired. In this case, the material properties as well as the process parameters need to be adjusted to favor either layering or agglomeration. The amount of properties and parameters influencing the dominating mechanism opens up a wide field of investigation. Although several theoretical concepts are available in literature [13–16], experimental investigations expose the need for more complex criteria [17, 18].

Design of spray fluidized bed processes requires detailed knowledge of the relationship between operating conditions, material parameters, equipment design, and the final product properties. This can be achieved by extensive experimental work as well as mathematical modeling of the underlying phenomena (i.e., particle formation and heat and mass transfer). For this purpose, different computational methods exist, ranging between macroscopic and microscopic approaches. Macroscopic methods are usually simplified based on assumptions and fast, while microscopic approaches tend to be more detailed, e.g., by considering each particle individually. At the same time, the increase in resolution leads to higher computational cost, limiting their application for systems at industrial scale.

In macroscopic models, usually the transient behavior of a property distribution (e.g., the particle size distribution) due to different particulate processes is modeled. A well-established concept used to model the change of property distributions is the population balance introduced by Hulburt and Katz [19]. This concept is widely used to model particle formation processes such as crystallization [20, 21], layering granulation and coating [22–24], and agglomeration [25–27]. Applications of population balance models are the prediction of property distributions, the design of operating conditions to achieve a desired property distribution, and control of particulate processes [28].

In case of layering granulation and coating, population balance models can be used to calculate particle size or coating mass distributions for batch or continuous processes. Calculation of the macroscopic process kinetics is possible, however, a more detailed description of particle properties

such as intra-particle layer thickness distributions or information about the surface coverage of the coating has not been presented in literature with this approach.

In order to describe agglomeration processes with population balances, usually birth and death terms are used. Layering, which may occur simultaneously, is often neglected since its influence on the kinetics is small in comparison. The complexity of the birth and death terms depends on the number of micro-processes, which need to be included. In contrast to layering, the agglomeration kinetics cannot be directly calculated from process parameters and particle properties. Instead, mathematical formulations of agglomeration kernels are necessary. Although several kernels exist in literature, selection of one still remains a difficult problem since a single kernel considering all governing factors does not yet exist [28]. As a result, one has to rely on experimental data fitting, which is also known as the inverse problem in literature. Empirical kernels may provide an acceptable level of model prediction [29], but the obtained values are restricted to the used material, experimental setup, and process parameters.

Microscopic models describe the particle formation processes as a result of phenomena occurring on the single particle level. No grid or discretization is needed as the particles are treated as discrete entities. Two well-known methods are discrete element methods (DEM), often coupled with computational fluid dynamics (CFD), and Monte Carlo (MC) methods.

Particle formation in spray fluidized beds has been studied using CFD-DEM simulations by Goldschmidt et al. [30] and Fries [31], who focused on modeling agglomeration and evaluating particle dynamics, respectively. Since CFD-DEM models are computationally expensive, the simulated process time is short and ranged between 2.5 s and 10 s in the above discussed studies. The application of CFD-DEM models for simulating real process times in the range of minutes or hours is therefore not yet possible. Nevertheless, they can be used to extract important properties such as residence time distributions, collision frequencies, or particle velocities for macroscopic models.

Another way of discrete modeling of particulate processes is given by Monte Carlo methods. Metropolis and Ulam [32] introduced this method, which is essentially a statistical approach to study differential equations. The solution is in this case performed by a stochastic sampling experiment involving random numbers [33]. Monte Carlo methods have been used to model different particulate processes such as agglomeration during crystallization [33], agglomeration of droplets in clouds [34], agglomeration of particles in spray fluidized beds [35–37], twin-screws [38], high-shear granulators [39], and coating in spray fluidized beds (Wurster) [40] and pan coaters [41]. In comparison to population balances, this simulation method offers several advantages. No mathematical formulation of the macroscopic process kinetics is needed and the discretization of the property domain is unnecessary. The consideration of several internal coordinates or properties can be easily performed, while in case of macroscopic modeling a multidimensional population balance must be solved, which may lead to large computation times, see Barrasso and Ramachandran [42]. In order to use a Monte Carlo method to model particle formation in spray fluidized beds, the processes occurring on the single particle level need to be modeled. They are then applied sequentially to a sample of the particle population changing its properties over time.

The foundation for the Monte Carlo methods derived in this thesis has been presented in the works of Terrazas-Velarde [43], Dernedde [44], and Hussain [45]. All of them considered spray fluidized bed agglomeration by spraying a binder solution. However, Monte Carlo modeling of layering as well as agglomeration of amorphous particles due to glass transition in spray fluidized beds have not been addressed.

In order to model particle growth by layering using Monte Carlo, only one micro-scale event, the deposition of droplets on the particle surface, needs to be included. Subsequently, the deposited droplets dry and leave a solidified droplet behind. Repeated deposition of droplets and solidification then builds a layer around the initial particles. The lifetime of a deposited droplet is calculated using a simplified heat and mass transfer model. The layer thickness generated by a single dried droplet follows from a new calculation method, including curvature of the particle surface and porosity of the dry droplet. The model is shown to provide information about a variety of important properties such as the intra-particle layer thickness distribution (for each particle considered in the simulation), inter-particle layer thickness distribution, particle size distribution, coating mass distribution, coating time distribution and the coated surface fraction. A comprehensive simulation study was performed to investigate the influence of process and material parameters on the resulting product properties. The transient behavior of the coated surface fraction as well as the coefficient of variation of both the intra-particle layer thickness distribution and the coating mass distribution were compared to analytical models (own models and available in literature [46, 47]), showing good agreement. Additionally, the simulation results were compared to experimental data on intra-particle layer thickness distributions and particle size distributions, showing good agreement as well.

In comparison to macroscopic approaches, which use population balances to calculate the transient behavior of property distributions, the presented Monte Carlo model for coating and layering granulation is able to predict a variety of property distributions at once, which has not been available in literature before. The simulation results were validated both theoretically and experimentally. The presented model can therefore be used to predict intra-particle and inter-particle properties, which is useful for process design. However, an application for process optimization and control is still limited due to higher computational cost in comparison to macroscopic models.

Modeling of binder-less agglomeration using Monte Carlo requires two micro-scale events: deposition of droplets on the particle surface and binary collisions between particles. The deposited droplets wet the particles and are imbibed. Depending on the process conditions, the droplets may cause glass transition of the material, leading to sticky, viscous spots on the particle surface. If particles collide at these spots, agglomerates may be formed. Collisions at dry spots will always result in rebound of the collision partners due to the conditions in this thesis (inlet gas temperature is always lower than the glass transition temperature of the dry solid). A simplified breakage model is used to take breakage of viscous and solid bridges into account. Based on the modeled micro-scale events and processes and three agglomeration criteria, the model is able to calculate the transient behavior of the particle size distribution during an agglomeration process. Opposing trends on the microscopic level have been identified in a simulation study. The governing trend on the macro-scale

was then discussed using experimental data on spray agglomeration of amorphous maltodextrin particles and corresponding simulations. In the investigated range, the first two agglomeration criteria (at least one wet droplet must be present at the contact point and the solid temperature must exceed the sticky point temperature) seem to dominate the macroscopic behavior, while the third one (Stokes criterion) is shown to play a minor role. A comparison between experimental data and corresponding simulations shows that the agglomeration behavior is correctly described by the model for varying inlet gas temperatures and spraying rates. However, the model predicts faster agglomeration kinetics for increasing DE values, which is not fully supported by the experiments.

The presented model is the first one to account for binder-less agglomeration due to glass transition. Despite of several assumptions, the model is able to correctly describe the influence of process parameters on the macroscopic agglomeration behavior. The influence of the DE value could not be fully described by the presented version of the model, which is considered to be a result of the assumptions in the drying model (gas-side controlled drying). Nevertheless, if the necessary model parameters are available, the model may be used for process design. However, an application in optimization or process control is unlikely due to the computation times. Instead, an extended macroscopic model similar to the one presented by Hussain et al. [37] for binder-less agglomeration could be derived, which may then be applied to optimization or process control.

Furthermore, a macroscopic model for estimating the dominant size enlargement mechanism for spray fluidized bed processes from the point of view of layering is presented. The dominant size enlargement mechanism is linked to the probability of successful collisions, which can be calculated using the probability of wet collisions and the probability of successful wet collisions (in terms of dissipation of kinetic energy). The probability of wet collisions is calculated from the wet surface fraction, which follows from a new dynamic model and a heat and mass transfer model for spray fluidized beds. The probability of successful wet collisions is then calculated from the Stokes criterion using a new model, which is validated theoretically by a Monte Carlo method. A simulation study is performed, which shows the influence of inlet gas temperature, viscosity, droplet size, and contact angle on the probability of successful collisions. It is shown that the agglomeration tendency decreases when the inlet gas temperature is high and the viscosity, droplet size, and contact angle are decreased. Further simulations based on experimental work published in literature were performed, which are used to classify the experimentally observed dominant size enlargement mechanism based on the calculated probability of successful collisions. The results indicate that layering will be dominant if this parameter is smaller than 0.001. Consequently, agglomeration will become dominant if this value is exceeded. The obtained classification is then used to create regime maps, which are able to show the border between the size enlargement mechanisms for different parameters.

The presented model combines microscopic and macroscopic parameters in order to estimate the dominant size enlargement mechanism for spray fluidized bed processes. The model includes a new dynamic model for calculating the wet surface fraction, taking the geometry of deposited droplets and drying conditions into account. Further, a new theoretically validated model for calculating

the probability of successful wet collisions based on the Stokes criterion is presented. Both models were not available in literature before. An interesting result of the simulation study based on experiments taken from literature is that for agglomeration to become dominant, the probability of successful collisions does not need to be high (i.e., close to unity). Instead, a relatively small value of 0.001 represents the border between the size enlargement mechanisms based on the considered simulations and experiments. The presented model may be used in process design to estimate the dominant size enlargement mechanism under given conditions. Further applications may be a comprehensive macroscopic process model considering particle size enlargement by simultaneous layering and agglomeration including the effect of drying conditions and wetting parameters on the process kinetics.

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