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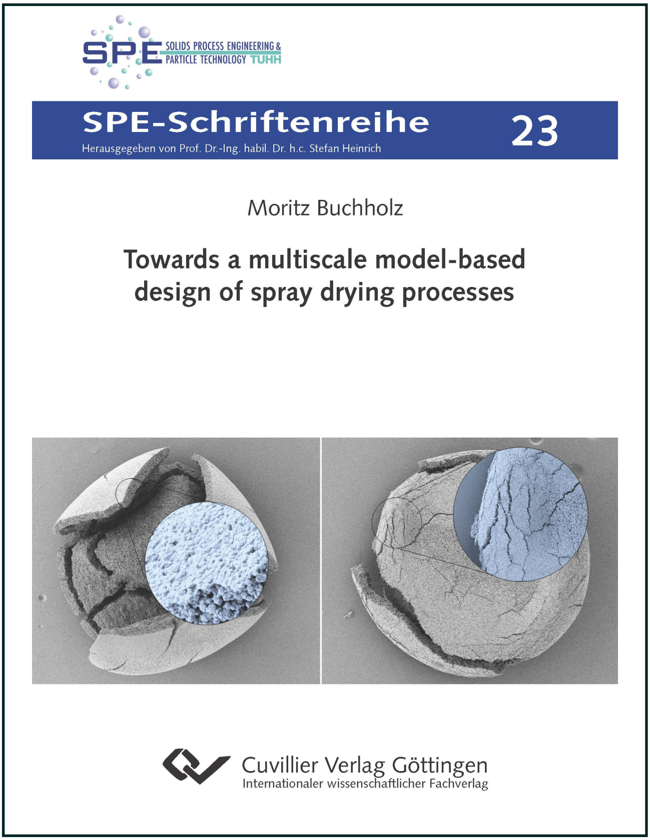
Towards a multiscale model-based design of spray drying processes



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Introduction

1.1 Motivation

Spray drying processes are widely applied to remove solvent from liquid feeds and efficiently produce granular material in large quantities. The first application of spray dryers goes back over a hundred years focusing on dairy and detergent industry (Masters, 1985). Recently, the production of specialized granular material in spray dryers, receives more and more attention. This work focuses on zeolites as an exemplary material with industrial relevance. Zeolites are used in many applications, for example as molecular sieves or as base material for chemical catalysts, due to their structured inner system of pores. For the development of novel product materials, reliably satisfying the high requirements on the product properties, such as the particle morphology, poses a main challenge (Na et al., 2013). This is mainly due to the complex nature of the particle formation process on the single droplet scale (Majano et al., 2012). Another difficulty is related to the scale-up of lab or pilot scale experiments to production scale. This is mostly due to changing flow patterns and long term effects such as wall caking, which affect the local drying conditions and can influence the product properties (Sosnik & Seremeta, 2015).

Despite ongoing research regarding various aspects of the spray drying process, an a-priori prediction of product properties of spray dried granules is challenging and cost-intensive. A crucial property of zeolites is the final particle morphology which influences the stability as well as the functional surface in the inner region

(Na et al., 2013). As the particle formation mechanisms depend both on the applied material and the local drying conditions, an investigation on different time and spatial scales is necessary. When single droplets dry, the suspended primary particles relocate and form a solid particle. Depending on the trajectory of the droplet, particles with different morphology may form, ranging from dense to broken or donut-shaped morphologies, as shown in Fig. 1.

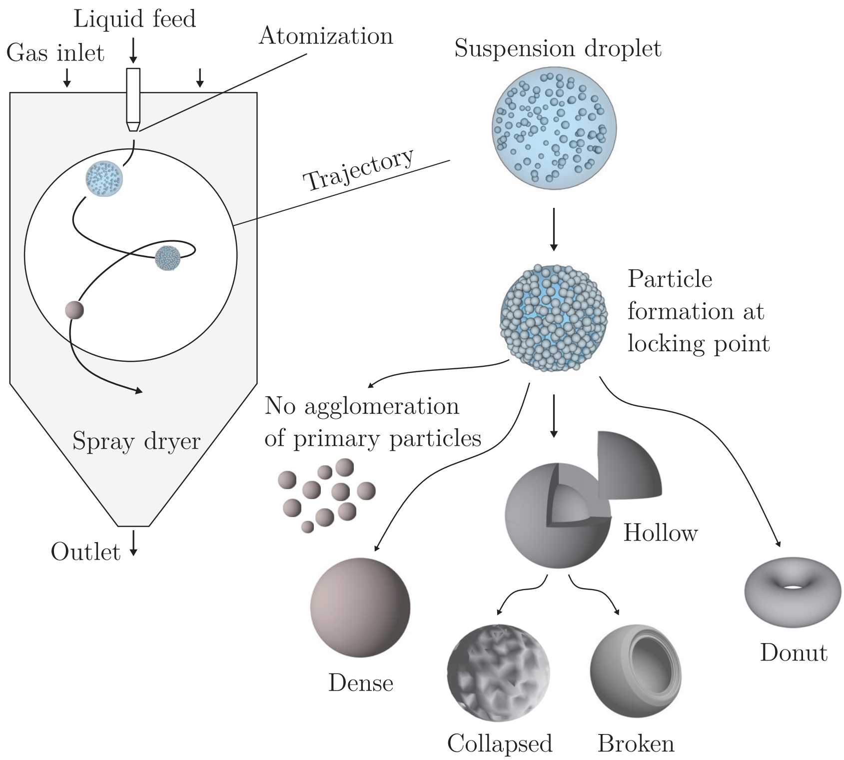

Figure 1: Schematic illustration of the drying process of a single suspension droplet in a co-current spray dryer leading to different particle morphologies depending on the experienced drying conditions.

This work combines different experimental and numerical methods to gain insights to the spray drying process and to capture the relevant mechanisms for an efficient design of the spray drying process for the production of zeolite particles.

Here, the focus lies on co-current spray drying processes, which are characterized by a unidirectional movement of the gas and the drying matter through the drying chamber. Drying experiments of single suspension droplets are used to investigate the impact of different parameters on the resulting particle morphologies. For this reason, acoustic levitation is used to allow for a contact-less investigation of single droplets that levitate in an acoustically induced pressure field. The experimental investigation is supported by a novel simulation approach for single suspension droplets, which relies on an unresolved coupled CFD-DEM simulation model (Computational Fluid Dynamics coupled with Discrete Element Method). This model captures the main drying mechanisms as well as the solidification of the droplet to allow for a prediction of the particle morphologies for different drying conditions. The drying conditions of the single droplets are determined by a large scale spray dryer simulation using CFD simulation. Finally, a reduced order model of the spray drying process is applied to allow for a simulation of the whole production process to account for interdependencies between different unit operations in the process chain.

1.2 Outline

In chapter 2, the fundamentals of the spray drying process are presented. An overview of the different modeling and simulation approaches for the drying process, found in the literature, is given.

In the following chapter 3, the findings of the spray dryer experiments and the single droplet drying experiments are presented. The sections regarding the single droplet experiments give insights into the determination and interpretation of the drying kinetics in the acoustic levitator both before and after the solidification and how they can be modeled. Additionally, the impact of different drying parameters on the particle morphology is discussed.

The drying conditions, which the droplets experience in the spray dryer, are determined via CFD simulations, which are presented in chapter 4. The simulation results are validated by the spray drying experiments and the approach for the data extraction is introduced.

This information is used in the unresolved CFD-DEM simulation of single suspension droplets. Due to the novelty of the model, the governing model equations are presented in detail. The numerical behavior of the model is analyzed regarding the physical plausibility and the impact of different model parameters on the particle formation process is evaluated.

The final chapter introduces the reduced order model for the spray drying process, which is based on a multi-compartment and population balance approach to describe the most important mechanisms in the spray dryer. The model is validated with the spray dryer experiments, shown in chapter 3. Finally, a production scale spray dryer is simulated using additional information from a detailed CFD simulation to further improve the model accuracy.

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Fundamentals of the spray drying process

A spray dryer is generally used to remove single or multiple solvent components of a solid-laden liquid feed. The solid components are either suspended as so-called primary particles or solved as a solute in the liquid material and remain after the drying process as solid particles. The evaporated solvent component is carried out by the drying gas. Depending on the composition of the liquid feed, the solidification of the droplet may be either due to a precipitation or crystallization of the solute material, an agglomeration of suspended primary particles, or a combination of both sub-processes. The focus in this work lies on the investigation of suspensions from anorganic zeolite particles. Solute material, the so-called binder material, is only added in small concentrations to the watery suspension to increase the intergranular adhesive force of the zeolite primary particles during the solidification.

The spray drying process can be partitioned into three main stages, as shown in Fig. 2.1: the atomization of the liquid, the evaporation of the solvent component and the particle formation in the drying chamber and finally the separation of the dried granules from the gas. There are several modes of operation, ranging from a co-current, over a mixed-type to a counter-current operation. In co-current spray driers, the liquid feed and the drying gas flow move through the drying chamber in the same direction. The co-current mode favors the drying of temperature sensitive materials as the feed comes into contact with the hot gas, while it is still in liquid state or at a high moisture content (Masters, 1985). Additionally, the amount of wall deposition is lower than in the counter-current mode, in which the gas and the

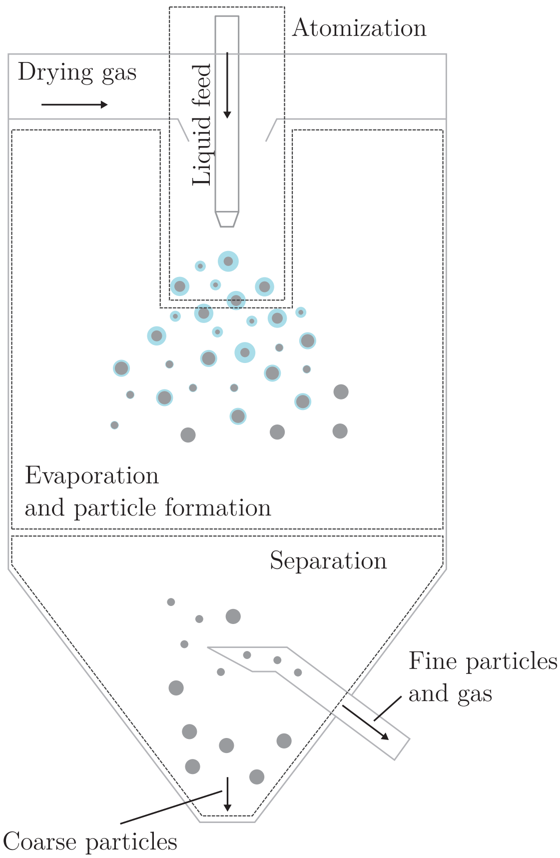

Figure 2.1: Schematic presentation of a co-current spray drying process. The spray drying process can be divided in three main sub-processes: the atomization, the drying of the droplets and particles and the final separation of the particles from the drying gas.

droplets move in opposite direction through the spray dryer. The counter-current mode in return offers a better thermal efficiency as well as a more intense mixing of gas and droplets resulting in high evaporation rates (Bellinghausen, 2019; Masters, 1985). When flammable compounds have to be dried, an inert drying gas like

nitrogen is used, which is oftentimes recycled in a so-called closed-loop configuration including a recycling stage for the humid drying gas. Using air as drying gas in an open-loop configuration, however, is more cost-effective and stable (Sosnik & Seremeta, 2015).
The following chapter gives an overview over the different stages of the spray drying process and the relevant sub-processes. Additionally, the theoretical modeling and simulation approaches related to the spray drying process are presented.

2.1 Atomization

The goal of the atomization stage is the generation of a droplet spray to increase the specific surface area of the drying substance on the one hand and to control the final granular properties by setting the initial conditions, i. e. droplet size and velocities, for the drying stage on the other hand. These properties depend on the choice of the atomizer type and on its operation conditions. Hence, the atomization is pivotal for the resulting dried granule properties (Poozesh et al., 2018).

2.1.1 Droplet formation and collision

The formation process of the droplets after the atomization may be described by two different stages. The so-called primary atomization consists of the formation of a liquid sheet that may result in the formation of ligaments which break up into small droplets (Ashgriz et al., 2011). This mechanism is presented in Fig. 2.2.

The newly formed droplets may be deformed or further broken up into smaller droplets, if the aerodynamic forces are large enough, which is also called secondary atomization (Chryssakis et al., 2011). The breakup is mainly determined by the ratio of the disruptive aerodynamic force and the opposing surface tension, given by the Weber number, and the dissipative nature of the viscous forces inside the droplet, which is taken into account by the Ohnesorge number (Guildenbecher et al., 2011). The Weber number We is calculated by

$$\mathrm{We} = \frac{\rho u^2 L}{\sigma}, \tag{2.1}$$

where ρ is the fluid density, u is the fluid velocity, L a characteristic length and σ the surface tension. The Ohnesorge number is calculated by

$$\mathrm{Oh} = \frac{\mu}{\sqrt{\rho \sigma L}}, \tag{2.2}$$

with the dynamic viscosity of the fluid μ. For example, Weber numbers smaller than 11 indicate for Newtonian liquids only vibrational movement of the droplet without breakup (Guildenbecher et al., 2011).

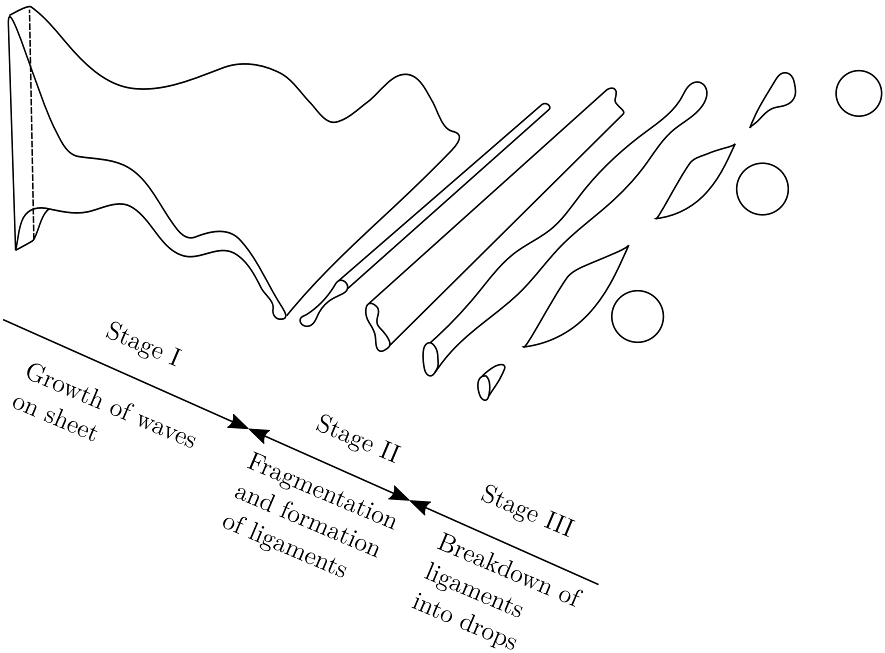

Figure 2.2: Schematic illustration of different stages during droplet formation according to Dombrowski & Johns (1963).

After the formation process, the droplets may change their properties due to collisions with other droplets in the spray. Different modes of droplet collisions ranging from coalescing droplets to droplets breaking up after impact are presented by Brenn (2011). In addition to the Weber number and Ohnesorge number, which play an important role regarding the collision mode similar to droplet breakup, the non-dimensional impact parameter and the droplet size ratio play have to be considered when characterizing the droplet collision (Brenn, 2011). The impact parameter also factors in the directions and velocities of the droplets and takes low values for head-on collisions (Brenn, 2011).

2.1.2 Atomizer types

There are three main types of atomizers that are commonly applied in industrial or scientific spray dryers: pressure nozzles, two-fluid nozzles and rotary atomizers.

Pressure nozzle

In pressure nozzles, droplets are formed by the conversion of pressure energy to kinetic energy without using additional gas flows (Masters, 1985). At the outlet, the spray forms a cone with a cone angle between 5° and 15° (Poozesh & Bilgili, 2019). The main advantage of pressure nozzles is the simple design and operation with the disadvantage of their inability to produce very fine sprays (Poozesh & Bilgili, 2019). The diameters of droplets formed from pressure nozzle are typically in the range of 120 µm to 250 µm (Masters, 1985). In most pressure nozzles, a swirling motion is added to the liquid before entering the process chamber to give additional control over the cone angle, ranging between 40° and 60° (Walzel, 2011), and simultaneously over the degree of gas exposure (Poozesh & Bilgili, 2019). In Fig. 2.3, two different types of pressure nozzles are schematically shown.

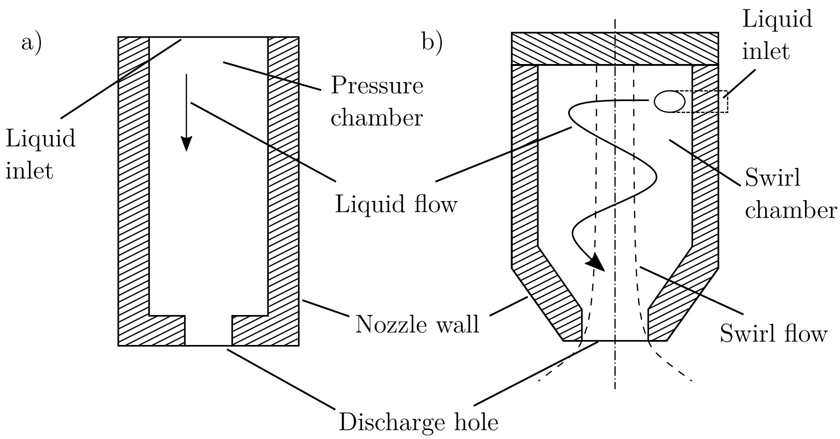

Figure 2.3: Different types of pressure nozzles: a) plain-orifice and b) pressure-swirl atomizer (Lefebvre, 1989; Walzel, 1990).

Two-fluid nozzle

In two-fluid nozzles, both a high speed gas and the fluid are transported separately to the nozzle head and are mixed at this location, either on the inside (internal mixing) or on the outside (external mixing) of the nozzle head (Masters, 1985). The different nozzle types are shown in Fig. 2.4. The shear forces between the high-speed gas and the liquid result in the sheet, ligament and finally the droplet formation (Poozesh & Bilgili, 2019). The droplet sizes that are generated by two-fluid nozzles lie commonly in the range of 10 µm to 1000 µm (Heng et al., 2011), giving the two-fluid nozzles a wide applicability at the cost of higher clogging probability.

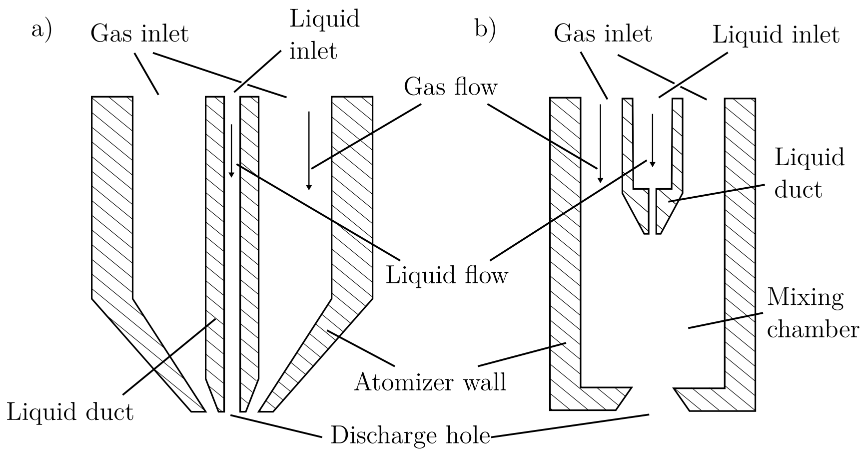

Figure 2.4: Different types of two fluid nozzles: a) external mixing and b) internal mixing of liquid feed and atomization gas (Walzel, 1990).

Rotary atomizer

Rotary atomizers create droplets by feeding the liquid onto a rotating disc, wheel or cup accelerating it to the periphery, where it disintegrates into droplets (Masters, 1985). The main advantage of rotary atomizers is its reliability regarding the ease of operating, because of the absence of clogging and the handling of fluctuating feeds, as well as the possibility to operate with high feed rates (Masters, 1985). A disadvantage of rotary atomizers is the inherent tendency to accelerate the feed to the walls, which has to be mitigated by the use of wider drying chambers to