INVESTIGATRION OF THE GRANULATION PROCESS IN A MULTI-STAGED CONTINUOUS FLUIDIZED BED: INFLUENCE OF THE PROCESS CONDITIONS ON THE PRODUCT PROPERTIES

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ABSTRACT

This work focuses on the particle properties of sodium benzoate granules, which are produced by fluidized bed spray granulation during different process conditions. The drying process has a decisive influence on the kinetics of the growth rate and thus also on the properties of the particles, like the morphological structure, particle moisture and porosity. This influence has been considered in this project in more detail.

In order to compare the produced granules while varying the drying conditions an external product classification is used to achieve a certain product size. The results show that the drying conditions significantly affect the granule morphology and structural properties. Hence, higher spray rates of the suspension and lower drying temperatures lead to higher jaggedness of the product particles.

INTRODUCTION

Fluidized bed spray granulation is an important process to produce granules with a desired particle size and structure. Therefore, it is of great interest in industrial applications, research and literature. This process consists of three main steps until a product granule is generated, shown in fig. 1. In the first step the droplets of the used solution, suspension or melt are usually atomized by two-fluid nozzles. These droplets collide with solid kernels of the hold-up material. Secondly, the liquid droplets are spread on the particle surface, followed by the solidification of the solution. The repetition of these consecutive steps leads to the growth of the kernel layer by layer, until a certain product grain size is achieved. Granulation is typically used for layering an initial particle of the same material as the coating to generate larger particles (Bück et al., 2014).

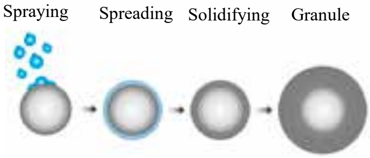

Fig. 1: Principle of the spray granulation process

The particle properties can be influenced by several process parameters, like drying temperature, liquid feed rate as well as droplet size of the atomized liquid feed. There are different granulator types that can be used for granulation and coating processes, briefly summarized by Fries et al (2011). Each type of granulator has its own advantages and disadvantages. In continuous operation mode very often horizontally constructed fluidized beds with rectangular cross sections are used (fig. 2). A main advantage of this type of granulator is the possibility to divide the process chamber by a variable number of plates (weirs) into several stages of different functionalities (e.g. granulation, drying or cooling). Commonly, an external product process chain, consisting of pneumatic conveying, screening, grinding of the oversize granules and recycling of grinded oversize and undersize particles into the fluidized bed granulator, is applied. The resulting internal and external interconnected solids process unit operations as well as gas, liquid and solid flows lead to a complex and dynamic process behavior that affects also the product quality.

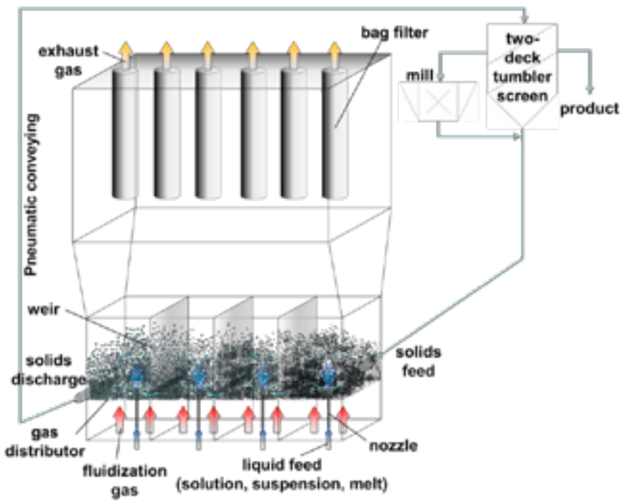

Fig. 2. Scheme of a multi-staged horizontal fluidized bed apparatus

In this contribution the influence of the thermal process conditions on the product quality of granules, produced by a continuously operated granulation process in a horizontal fluidized bed is investigated. The discharged product particles with a diameter between 2.00 – 2.24 mm were used to analyse several different characteristics in order to compare the products that are achieved due to different process conditions during the granulation process.

MATHERIALS AND METHODS

For the experimental studies sodium benzoate is used since granules of this model substance show stable structures over a wide parameter range. The experimental research is done in a horizontal fluidized bed plant (Procell 25) of the company Glatt. The process chamber has a length of 1 m and a width of 0.25 m. The process chamber can be divided into four chambers with an equal length and width of 0.25 m by using weirs, selectively with different configurations (overflow, underflow or lateral flow), as shown in fig. 2.

The thermal conditions in the granulation process are also influenced by the spraying rate, because of the occurring internal process temperature and gas humidity that results in dependence of these two parameters. For this reason the drying temperature, as well as the spraying rate of the used suspension have been varied. Hence, each of these two influence factors were varied under three different configurations, corresponding to tab. 1.

Tab. 1. Parameter variation of the granulation processes to investigate the influence on the product properties.

parameter	**adjustment**		
drying temperature [°C]	100	150	200
spray rate [kg/h]	40	80	120

For the granulation processes the continuous mode was applied while the fluidization velocity in every experiment has been kept constant at an inlet velocity of 3 m/s, whereby the hold-up mass was also constant at a value of 25 kg. The solids discharge of the process was adjusted to eject solid material automatically when reaching a certain value of the pressure loss over the solid bed material. While setting this pressure loss to be constant the hold-up mass will also be constant, respectively.

During the experiments samples have been taken out of the process chamber in order to analyse the moisture content of the solid material and track the particle size distribution of the hold-up material. Additionally, an inline probe of the company Parsum® for measuring the particle size distribution (PSD) was used. However, since it is admitted only for temperatures up to 100 °C, it could not be integrated in every process. Fig. 3a shows the horizontal fluidized bed with the used sampling devices and the installed inline probe for measuring the PSD. The samples are taken every 20 kg of sprayed in suspension. Based on the spraying rates that were adjusted, sampling rates of 10, 15 and 30 min (spraying rates: 40, 80, 120 kg/h) were chosen.

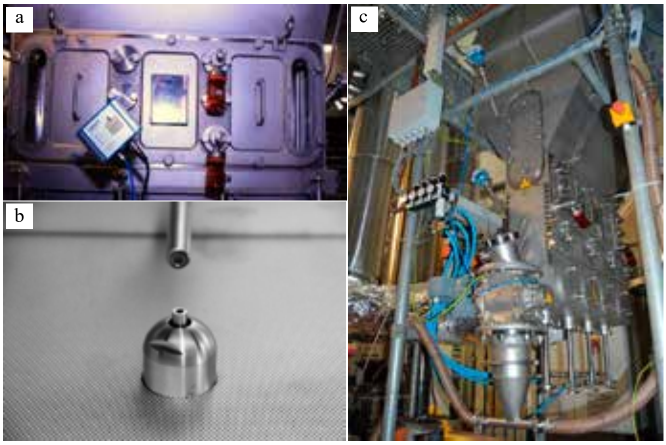

Fig. 3. Photos of the samplers and the installed inline probe (a), the nozzle within the process chamber (b) and the used pilot plant granulator (c)

Fig. 3c shows the used granulator, which is a horizontal fluidized bed, consisting of four separate wind boxes, which allow a separation of the whole process chamber into maximum four stages of equal length and width. This permits different process conditions in every stage of the process chamber, usually separated from each other by weirs, which are applied in over- or underflow configuration. This influences also the residence time behaviour of the solid material inside the process chamber. However, since the aim was to analyse the thermal influence on the product quality, the process conditions in every stage were equal. The used liquid suspension was feed by four two-substance spray nozzles that use compressed air to disperse the liquid solution into fine droplets (fig. 3b). The suspension consists of 35-weight-% of the solid material, which is also used as model substance. Therefore, the produced granules contain only layers of sodium benzoate which are coated under different process conditions onto kernels of sodium benzoate. The kernels within the continuous process are exclusively produced by the comminution process, out of the grinded oversize material. Hence, no external feed of solid material was required in these experiments.

The product granules were discharged after the classification process in a two-deck tumbler screen, in between a size fraction of 2.00 and 2.24 mm. The product of every experiment after at least three times exchange of the bed holdup was used to classify and compare the generated granules according to the following characteristics:

- Surface morphology and roughness
- Solid moisture content
- Solids density and porosity

Surface morphology and roughness analysis

A commonly used method to classify and compare the quality of coating layers and the morphological structure is done by using SEM (*scanning electron microscope*) images or even microscope images, if the resolution is sufficient (Hede et al., 2008; Rieck et al, 2015). These analytical methods allow the visualization of structural differences on the surface of relatively small particles. However, these methods permit only subjective and qualitative evaluation. Additionally, a surface roughness analysis was chosen, which represents the results in a quantitative manner. The roughness was measured by focussing-variation microscopy by an Alicona Infinite Focus. This tool provides a measurement of the real 3D-surface structure and an evaluation based on several roughness parameters, weather line-based or surface-based. While setting manually an upper and lower focussing limit, a 3D-profile of the surface is created that is used to analyse the profile according to roughness parameters.

Solid moisture measurement

The moisture content of the solid material during the granulation process was measured by taking samples out of the process chamber. Directly after the samples were taken, the moisture content was measured by a thermo-gravimetric method, using a *Precisa EM-120 HR* moisture analyser. The analysis procedure that was used contained a constant drying temperature of 105 °C, using a switch-off criteria that is achieved when the reduction in weight is less than 0.01 mg/s or a total time of 10 minutes.

Solids density and porosity measurement

The density of every product was measured by a helium pycnometer. Therefore, approximately five grams of the produced granules were used to determine the density of each product ten times. This measurement provides the volume of the solid material. Additionally, the weight of the solid material is measured. Afterwards, the observed volume and the weight are combined to a density. In this analysis one should keep in mind that probably existing pores in the solid material change the results.

A second measurement by a geopycnometer is used to calculate the porosity of the granules, produced under different process conditions.

RESULTS

The process parameters of the granulation experiments are explained in the materials and methods section, while the variation parameters are displayed in tab. 1. After the granulation processes under different drying conditions and spraying rates of the suspension, the product material was collected and analyzed regarding several characteristics.

For the analysis of the surface structure, images were taken using a microscope. An overview of these results is given in Fig. 4a. Due to the over-moisturizing of the bulk material within the process chamber, the experiment at the highest spray rate and lowest gas inlet temperature (spray rate: 120 kg/h, temperature: 100 °C) was not accomplished, since this would lead to the collapse of the fluidized bed.

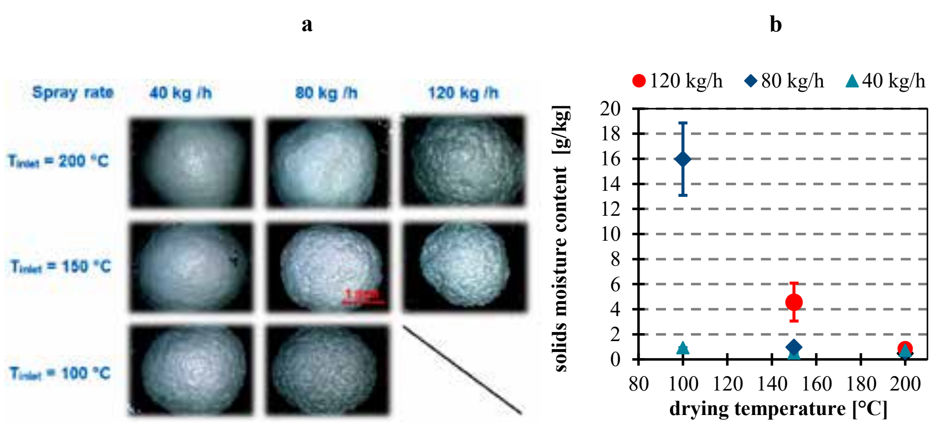

Fig. 4. Images of the particle surfaces of the different products (a) and solids moisture content of the bulk material during the granulation process (b).

The results in fig. 4a clearly show an influence of the process conditions on the particle morphology. An increase in the drying temperature results in smooth-shiny surfaces, while a decrease leads to higher surface roughness's and mat textures. The change in the spray rate results primarily in an increase in the moisture content, which is deposited on the particles (Fig. 4b), resulting in a stronger jaggedness of the surface. The particle moisture was carried out by thermo-gravimetric analysis using a moisture analyzer (Precisa EM-120 HR), directly after taking a sample out of the process chamber. Fig. 4 (right) shows that the particles were relatively dry in all experiments, except for two cases. There seem to be a certain boundary, at which the combination of the drying temperature and the spraying rate do not affect the moisture content of the particles and the moisture content reaches a certain equilibrium value, lower than 1 g/kg. To identify this transition zone the drying potential η, according to Rieck et al. (2015), will be introduced.

$$\eta = \frac{Y_{sat} - Y_{out}}{Y_{sat} - Y_{in}} \quad (1)$$

The drying potential represents the drying conditions of the heated fluidization gas. Hence, dry gas would have a drying potential towards unity, while it would be zero for saturated gas, respectively (Rieck et al., 2015). The drying potential is calculated by the moisture content of the inlet and outlet gas stream of the process. These are measured and recorded by installed moisture and temperature sensors on the plant. The saturation moisture content is calculated from:

$$Y_{sat} = \frac{R_g}{R_v} \frac{\varphi\, p_{sat}}{p - \varphi\, p_{sat}} \quad (2)$$

The corresponding saturation vapor pressure p_{sat} is calculated, according to Wagner (1973). This vapor pressure equation is valid between the triple point and the critical point and has a high reproducibility (Mc Garry, 1983). A detailed description with the used Wagner coefficients for water can be found in Baehr (2012).

The given drying potentials for the granule production processes in tab. 2 confirm the results regarding the solids moisture content. Consequently the two experiments that show higher moisture content of the holdup material have a drying potential less than 0.5, while the others exceed this value. An increase of the drying potential, from 0.541 does not seem to have an influence on the moisture content of the solids, since the values of these experiments are all less than unity.

Tab. 2. Drying potentials of fluidization air for each granulation experiment, sorted in descending order.

drying potential η [-]	drying temperature T_{inlet} [°C]	spray rate $\dot{M}_{spray}$ [kg/h]
0.411	100	80
0.421	150	120
0.541	200	120
0.562	150	80
0.696	200	80
0.702	100	40
0.803	150	40
0.845	200	40

To compare the produced granules according to their morphology, not only qualitatively based on the microscope images, but also in a quantitative manner, the roughness of the particle surfaces were measured. This was evaluated, using an optical method which creates a 3D-geometrical structure of the surface by focus-variation microscopy, see fig. 5a. After measuring and creating a three-dimensional geometry of the surface, the profile was analyzed according to the height profile (fig. 5b) and characterized by means of several surface based roughness parameters. These roughness parameters are explained in detail by Whitehouse (2012). Every surface based evaluation included a surface of 500 x 500 µm (see fig. 5b). These surfaces were characterized by the ratio of the real surface to the projected surface, to compare the products according to their morphological structure.

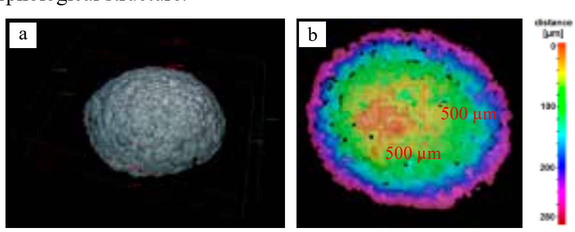

Fig. 5. Example for a created 3D-structure of a focus-variation measurement (a) and the height profile of the corresponding particle surface (b).

Based on the morphological structure of the granule surfaces in fig. 4a, it is obvious that the real surface area is always higher than the projected, because of the waviness of the surface structure. According to DIN 4760 an evaluation regarding the waviness of the profile, using a profile filter with a wavelength of λ = 150 μm for the primary profile (see fig 5b). The results of this roughness evaluation are given in fig. 6.

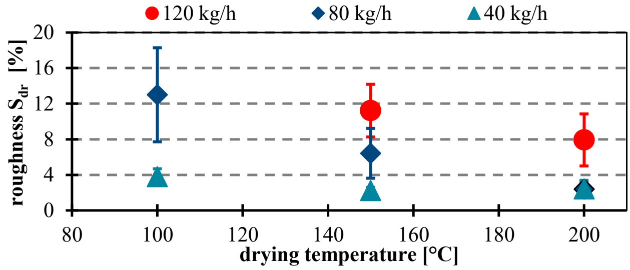

Fig. 6. Characterization of the surface roughness based on the ratio of the true volume to the projected volume S_{dr} of the granules, produced under different process conditions.

The results of the surface roughness in fig. 6 show a deep influence on both variation parameters. The morphology of produced particles with spray rates higher than 40 kg/h reveal a significant increase in the jaggedness of the surface. A reduction in the drying temperature also causes more rugged textures on the granules. These results are consistent with the findings that are shown by the surface images, given in fig. 4. This means, the more rugged and non-uniform the surfaces are, the higher is the roughness parameter S_{dr}. Lower temperatures could be expected to result in a slower drying of the droplets, resulting in a more uniform surface since the drying time is longer and thus the droplets can spread more on the surface of the particles. However, the results show the opposite effect. Rieck et al. (2015) has also observed this phenomenon, namely that lower drying temperatures lead to more rugged surfaces. He explains this effect with the longer drying times of the droplets that are expected to form larger crystals and therefore rough structures with higher porosities.

Additionally, the solids density of every product was measured by a helium pycnometer. In combination with a geopycnometer the porosity of the granules was carried out. Tab. 3 shows the results of these measurements.

Tab. 3. Process and product properties of each granulation experiment.

spray rate $\dot{M}_{spray}$ [kg/h]	drying temperature T_{inlet} [°C]	drying potential η [-]	density ρ [kg/m³]	porosity ε_P [%]
120	150	0.421	1530	27.46
	200	0.541	1536	31.55
80	100	0.411	1472	27.59
	150	0.562	1486	29.73
	200	0.696	1504	28.39
40	100	0.702	2048	26.42
	150	0.803	1803	24.12
	200	0.845	1736	23.48

Based on tab. 3 an increase in the drying temperature, while keeping the spray rate constant, changes the density as well as the porosity of the granules. In qualitative agreement with the results of Hoffmann et al. (2015), a decrease in the porosity with increasing drying potential η of the fluidization air was observed, neglecting the two experiments with drying potentials less than 0.5. Obviously, this boundary seems to have not only a significant influence on the moisture content of the granules during the granulation process, but also on the porosity, leading to considerably lower values. Before the density analysis every product sample was measured, these values were all between a range of 7-11 g/kg. Thus, the contained moisture can be excluded as reason for the lower porosity values.

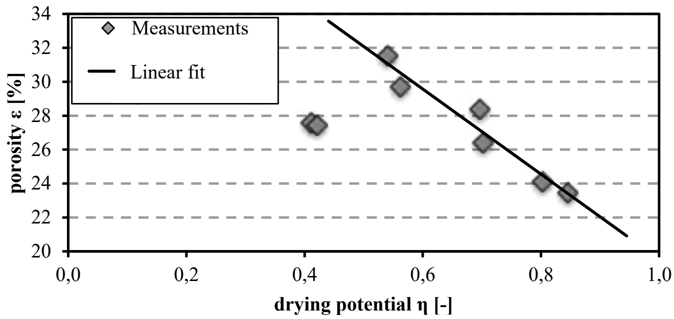

Fig. 7. Measured granule porosities in dependence of the drying potential of each granulation experiment. Neglecting the two experiments with drying potentials η less than 0.5 an additional linear fit was added.

SUMMARY

In this work, spray granulation experiments in continuous processing of a pilot-scale horizontal fluidized bed were performed at different spray rates and drying conditions. The aim was to analyze the product granules of these experiments and to examine the influence of the variation parameters on the product quality. A certain set of the described parameters is used to derive models of property coordinates for the generated products according to a specific parameter configuration that is chosen.

The experimental investigation of process parameters influencing the property coordinates of the product showed that it is possible to generate different kinds of granule surface morphologies with the same material. In general, very compact as well as porous granules can be produced by the choice of the thermal process conditions. The results showed that the spray rates, as well as the drying temperature have an influence on the granule structure. It can be concluded that the increase of the spray rate causes jugged surfaces, while the thermal conditions during the granulation process lead to smoother and more homogeneous surfaces with rising drying temperatures. With decrease of the introduced drying potential mat and rough surface structures are formed on the layer structure of the growing particles. To compare the surface structure of the granule, microscope images were produced. Additionally, the layer surfaces were characterized according to surface roughness parameters by focus-variation-microscopy in order to describe the granule structure also in a quantitative manner. Combined with density and porosity measurements the granule properties are described with reference to the used process conditions. It could be verified that the observed trends are consistent with the results of Rieck et al. (2015) and Hoffmann et al. (2015). Therefore, the porosity is decreasing with increasing drying potential.

Furthermore, the solids moisture content was measured during the granulation processes. The particle moisture content is a major factor that influences the granulation, since this affects the flow behavior of the granules. Due to the hydrophilic character of sodium benzoate the particle moisture content was determined directly after sampling during the granulation process. However, it was found that only granulation processes with drying potentials less than 0.541 did affect the moisture content of the solids significantly.

The results of this investigation are used for a comprehensive description of the complex dynamics of the spray granulation process in continuously operated horizontal fluidized beds to derive physically based models for the resulting property coordinates of the generated products in dependence of the process conditions.

NOTATION

Symbols

p absolute pressure, bar
p_{sat} saturation vapor pressure, bar
R_g specific gas const. for air, kJ/(kg K)
R_v specific gas const. for water vapour, kJ/(kg K)
Sdr
Y_{in} gas inlet moisture content, g/kg
Y_{out} gas outlet moisture content, g/kg
Y_{sat} gas saturation moisture content, g/kg

Greek letters

ε porosity, %
η drying potential, -
σ compression strength, N/mm²
φ relative humidity, %

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