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Influence of process variables on internal particle structure in spray fluidized bed agglomeration

M. Dadkhah *, E. Tsotsas

Thermal Process Engineering, Otto von Guericke University Magdeburg, Universitätsplatz 2, Magdeburg 39106, Germany

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ABSTRACT

In order to enhance and control properties of aggregates and be able to predict the structure of final products, it is essential to establish a link between the product properties and the operating conditions. Experimental investigations have been conducted on agglomerates produced in spray fluidized beds using different primary materials (non-porous glass, porous ceramic) and HPMC (hydroxyl-propyl-methyl-cellulose) as the binder, under varying operating conditions, inlet gas temperature and binder concentration. X-ray μ-computed tomography, μ-CT, was utilized to evaluate the three dimensional micro-structure of the agglomerates and spatial distribution of each constituent primary particle in the aggregate. Values and the behavior of evaluated morphological descriptors are presented and discussed for different agglomerates.

1. Introduction

Wet agglomeration occurs when a wet particle collides with another particle and gets bound with it by a liquid bridge which solidifies by drying. When conducted by spraying a liquid – usually an aqueous binder solution – on fluidized particles, the process is called spray fluidized bed agglomeration. Spray fluidized bed agglomeration is widely used in the chemical, pharmaceutical and food industry. It is known from practice and reported that process conditions can have a significant influence on the final properties of the agglomerates, such as flowability, strength, rehydration and active ingredient release [1]. Moreover, it can be postulated that such influences arise from changes in the internal structure of agglomerates with changing process conditions [2]. However, the transition from process conditions to internal agglomerate morphology as a basis for final properties has not been investigated before. The main reason for this is that internal agglomerate morphology is difficult to measure. Therefore, the few existing publications refer to specific aspects, such as agglomerate shape (aspect ratio, circularity, roundness) [3], and porosity [4]. Recently, X-ray tomography was proposed as an appropriate method to overcome this lack of data by thorough characterization of the internal structure of spray fluidized bed agglomerates [5]. However, even in Ref. [5] the influence of process parameters on morphological descriptors was not discussed, because all characterized agglomerates were produced at the same operating conditions.

Hence, the purpose of the present work is to continue the research started in Ref. [5] by investigating the influence of process conditions on the internal structure of spray fluidized bed agglomerates. First, experimental methods and evaluation techniques will be described. Then, the influence of varied process parameters (gas inlet temperature, initial binder mass fraction, use of porous instead of compact primary particles) on morphological descriptors will be presented and discussed. Morphological descriptors investigated are in the first place the fractal exponent and pre-factor, average coordination number, and average porosity. Additionally, coordination angle distribution, the distribution of primary particles in space, and the spatial distribution of porosity have been evaluated. Conclusions are presented at the end of the paper.

2. Experimental methods and evaluation

2.1. Agglomerate production

A spray fluidized bed with an inner diameter of 300 mm was used in Ref. [5] for producing the agglomerated particles. This equipment is rather large and laborious to operate. To make agglomerate production easier, smaller equipment has been used in the present investigation. This was a lab scale batch fluidized bed granulator with a transparent, cylindrical fluidization chamber with 152 mm inner diameter and 450 mm height (Glatt GmbH, Germany).

The solution was injected in top spray configuration and consisted of water with hydroxyl-propyl-methyl-cellulose (HPMC, trade name Pharmacoat 606, from Shin-Etsus, Japan) as the binder. The nozzle provided by Düsen-Schlick GmbH, Untersiemau, Germany, model 940, was a two-fluid nozzle, placed at a height of 150 mm from the plate used to distribute the heated air flow. Nozzle throughput was controlled by a piston pump; relative air pressure was 0.5 bar. Compressed air from

* Corresponding author. Tel.: +49 391 67 58267; fax: +49 391 67 11160.
E-mail address: m.dadkhah@yahoo.com (M. Dadkhah).

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the laboratory supply was the fluidizing gas. This air was heated by an electrical heater before being supplied to the fluidized bed. Process parameters such as the fluidization air flow rate and the spraying rate were kept constant by equipping all relevant parts of the plant with frequency converters and using a programmable logic controller, PLC. Gas inlet temperature was also adjusted via PLC, the temperature sensor being mounted directly below the chamber inflow. The cross section of the upper side of the process chamber was expanded to reduce gas velocity and, thus, enforce the return of entrained particles in the fluidized bed. Gas humidity was measured at the inlet and outlet of the fluidized bed by infrared spectroscopy. The data acquisition software DasyLab was used, and measured signals were recorded every 2 s. Binder and nozzle were the same as in Ref. [5].

In order to study the influence of process parameters, six experiments were carried out, denoted by A to F in Table 1, where they are summarized together with some main results. Experiment A was the reference experiment, so that process parameter variations were conducted in respect to it. The first process parameter varied was the inlet air temperature, set at 30, 60 and 90 °C, so that the transition from experiments B to A and, then, to C represents conditions of increasing inlet air temperature. Secondly, the effect of initial binder mass fraction was studied by using 2 wt.% (low viscosity of 5.7 cP), 6 wt.% (viscosity of 70.2 cP) and 10 wt.% (high viscosity of 292.2 cP) solutions of HPMC in water. In this case, transition from experiments A to D and E corresponds to increasing binder mass fraction. As Table 1 shows, the material used in five out of six experiments was non-porous glass. Respective primary particles (Sigmund Lindner GmbH, Germany) had a very high sphericity of 0.98 and a narrow range of diameters between 450 and 631 μm, with an average diameter of 520 μm (measured by Camsizer, Retsch Technologies GmbH, Germany). Transition from experiment A to experiment F was meant to realize the exchange of compact primary particles with porous ones under otherwise the same conditions. However, it is known from literature [6] that it is not possible to find porous primary particles with otherwise the same properties as given non-porous primary particles. A reasonable selection was to use in experiment F the same porous primary particles as in Ref. [5], which are made of γ-Al_2O_3 (Sasol GmbH, Germany) and have a sphericity of 0.97, and diameters between 580 and 650 μm, with an average diameter of 616 μm.

In order to better compare the growth kinetics and the final properties of agglomerates, all the experiments were performed with the same initial mass of bed material, 500 g, and the same total amount of sprayed solid binder, namely 0.5% of the initial mass of dry primary particles (i.e. 2.5 g). Since the amount of binder sprayed to the primary particles was kept constant in all experiments, process duration and solution usage were different, and each experiment was terminated when the specific amount of solute had been sprayed in the bed. The liquid spray rate at an initial binder mass fraction of 2% was 200 g of solution per hour (corresponding to 4 g of binder per hour). Relatively low spraying rates resulted in almost completely dry product at the end, and in only small differences between inlet and outlet gas moisture content during the process. A further process parameter that kept constant in experiments A to E was fluidization gas mass flow rate at 130 kg/h. The corresponding air velocity is 1.91 m/s at 60 °C. With a minimum fluidization velocity of 0.232 m/s for primary particles from glass, this corresponds to a fluidization ratio (actual by minimal fluidization velocity) of 8.25. This was kept constant in experiment F. Therefore, and since minimum fluidization velocity of Al_2O_3 primary particles was 0.139 m/s, experiment F was conducted with an air mass flow rate of 78.2 kg/h.

It should be noted that, due to different primary particles and/or operating conditions, none of the present experiments is immediately comparable to the experiment conducted in a larger spray fluidized bed in [5]. Hence, the issue of a possible influence of scale-up on agglomerate structure has to be addressed separately.

After running an experiment in the fluidized bed, the size distribution was determined by Camsizer. This equipment measures cord length distributions of particles falling in front of a camera and transforms this information to various particle size distributions. The size distributions of agglomerates resulting from all six trials at different conditions are plotted in Fig. 1. Following the usual practice, particle size is defined by the diameter of a sphere of equal volume in this figure.

Moreover, a certain number of agglomerates per trial were selected to be scanned individually for the examination of morphological descriptors. This number ranges from 22 to 28 agglomerates for each trial (Table 1). In total, 148 agglomerates have been scanned and analyzed, which is a necessary compromise between the time and effort needed (cf. Section 2.2) and the statistical significance of the results. Another compromise is necessary concerning the sampling of agglomerates to be analyzed out of the entire holdup of the respective batch. Commercial devices cannot be used for subdividing the holdup to random small samples, because of the danger of damaging the agglomerates during this procedure. Therefore, selection by a person is necessary. Such selection needs to include agglomerates covering the entire range of the particle size distribution, and can hardly be made in random with open eyes and this requirement in mind. The procedure followed was to get an optical impression of the particle size distribution in the holdup – which works quite well with a certain experience – and then pick up about 10% of very small, 50% of relatively small, 30% of relatively large and 10% of very large agglomerates, mimicking with this asymmetry the shape of particle size distributions in Fig. 1. In each of these classes, selection without any additional criterion has been attempted. As already indicated, samples were treated with care, so that breakage during selection and analysis can be excluded.

2.2. X-ray tomography and morphological descriptors

Computed X-ray micro-tomography (X-ray μ-CT) gives volumetric information on the internal microstructure of an object. In case of agglomerates, it allows the acquisition of 3D images of the detailed agglomerate structure with sufficient contrast and resolution to clearly

Table 1
Main experimental parameters in each trial and evaluated morphological descriptors.

	A	B	C	D	E	F
Primary particles	Glass	Glass	Glass	Glass	Glass	γ-Al_2O_3
Temperature [°C]	60	30	90	60	60	60
Binder wt%	2	2	2	6	10	2
Evaluated samples	25	28	22	24	24	25
Growth rate [μm/s]	1.068	1.146	0.630	3.789	8.204	0.599
Fractal dimension, D_f	2.45	2.31	2.94	2.24	2.09	2.45
Pre-factor, K_g	1.76	2.01	0.98	1.96	2.24	1.60
Regression, r^2	0.97	0.99	0.98	0.97	0.95	0.99
Mean coordination number, MCN	3.32	3.10	4.02	2.92	2.87	3.16
Average porosity	0.57	0.62	0.53	0.58	0.63	0.62

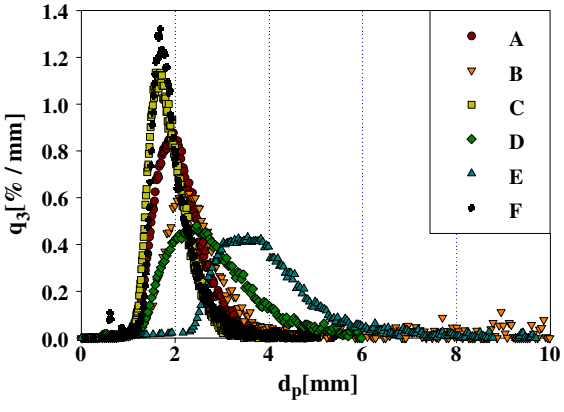

Fig. 1. Density distributions of agglomerate size.