THE EFFECT OF BINDER CONCENTRATION IN FLUIDIZED-BED GRANULATION: TRANSITION BETWEEN WET AND MELT GRANULATION

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Graphical abstracts

Pilot-scale batch fluidized-bed granulator

Highlights

- The effect of the binder concentration on the granulation performance is studied.
- Highly concentrated binders improve the coating and granulation efficiency.
- The temperature limits the binder concentration to be used to avoid agglomeration.
ABSTRACT
According to the binder nature, fluidized-bed granulation (FBG) is usually classified as wet or melt granulation. In particular, the industrial urea granulation performed in fluidized beds, is often called “melt granulation” because a highly concentrated urea solution is used as binder (between 95-97 wt%) (Cotabarren et al., 2012). However, plant disturbances can cause changes in binder urea concentration and thus granulation can shift from melt to wet granulation and vice versa.

In a previous investigation, the granulation system urea (seeds) – urea (binder) was extensively studied in a pilot-scale batch fluidized-bed granulator (Veliz Moraga et al., 2015). Besides, the effect of seed size, bed temperature, binder flowrate and fluidization and atomization air flowrates on process variables as well as on product properties were studied. The aim of this work is to analyze the effect of the binder urea concentration on the urea granulation performance and product properties. This concentration was varied from 87.5 wt% to 98 wt%, while the fluidization air velocity, urea melt flowrate, bed temperature set-point and atomization air flowrate were kept constant. The product properties (percentage of agglomerates and coated particles, crushing strength and moisture content) and granulation efficiency are discussed in terms of the transition from wet to melt granulation. The critical urea content was experimentally found; indeed, urea concentrations lower than the critical one dramatically affect the product quality. Finally, the criterion proposed by Villa et al. (2016) for predicting agglomerates formation is used to determine the minimum allowable binder urea concentration. The prediction is consistent with the trends experimentally observed, indicating the good capacity of the criterion to identify the boundary for agglomeration occurrence.

Keywords: fluidized-bed granulation; coating; agglomeration; urea

7. NOMENCLATURE

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<tr>
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<tr>
<td>A</td>
<td>Agglomerated product mass fraction (-)</td>
</tr>
<tr>
<td>C</td>
<td>Pure coated product mass fraction (-)</td>
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<tr>
<td>cp_u</td>
<td>Urea mass heat capacity (J/kg K)</td>
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<td>Filter-bags fines mass fraction (-)</td>
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<td>Superficial fluidization air velocity (m/s)</td>
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<td>$X_u$</td>
<td>Urea mass concentration (kg$<em>{urea}$/kg$</em>{solution}$)</td>
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**Greek Symbols**

<table>
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<tr>
<th>Symbol</th>
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<tr>
<td>$\Delta H_{fus}$</td>
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INTRODUCTION

Granulation includes a sequence of events to increase the particle size of granular material (Wang et al., 2017). Several technologies are used for particle size enlargement with different aims such as improving handling and flowability, obtaining a certain size, enhancing product appearance, controlling particle moisture content, reducing dusting or material losses, producing structural useful forms, etc. (Barrasso and Ramachandran, 2015; Schmidt et al., 2015).

Granules can be obtained in different equipment, among others, rotary drums, high shear mixers and fluidized beds (Litster et al., 2004; Osborne et al., 2011; Snow et al., 1999). These units basically differ in the way they favor particles mixing. The advantages of a fluidized bed with respect to other granulation systems include coupling of spraying, granulation, drying and cooling stages in one single unit as well as control, within certain limits, of the granules physical properties by manipulation of some operating variables (Heinrich et al., 2005).

When the binder used in granulation is a liquid material, the process is classified into wet or melt granulation. In wet granulation, the liquid binder is distributed on the seeds and, subsequently, the granules are dried to evaporate the solvent. In melt granulation, powders are enlarged by using meltable materials. These last binders are added to the system either as: 1) powders that melt during the granulation process or 2) atomized molten liquids (Zhai et al., 2010). The first melt granulation technique is usually called co-melt or in situ melt granulation (Zhai et al., 2010), while the second method could be referred as spray-on melt granulation (Osborne et al., 2011).

The effects of process variables and material properties of the solid and the liquid on particle size enlargement in fluidized beds were reported by quite a large number of articles (Ennis et al., 1991; Hemati et al., 2003; Iveson et al., 2001; Pont et al., 2001; Smith and Nienow, 1983; Tan et al., 2005, among others). These contributions are valuable for understanding and quantifying the mechanisms that control granule attributes (Suresh et al., 2017).

Melt granulation presents some advantages with respect to wet granulation: a) the use of solvents is avoided, eliminating the disadvantages associated with solvent recovery and final disposal and minimizing the energy cost related to solvent evaporation (Abberger et al., 2002; Mielke et al., 2016; Walker et al., 2006); b) the amount of liquid added during granulation is better controlled since uncontrolled solvent evaporation will not occur (Parikh, 2010) and c) for moisture-sensitive materials, granulation can be carried out without organic solvents (Parikh, 2010).

The industrial urea granulation process is considered one of the most significant breakthroughs in the fertilizer manufacture and, thus, of great interest (Cotabarren et al., 2012; Heffer and Prud'homme, 2016). Urea is the most widely consumed nitrogen-based fertilizer, being critical in the modern agriculture scenario. Industrial urea granulation is performed in fluidized beds, by spraying from the bottom a highly concentrated urea solution. Urea seeds are quite large (about 2 mm) and, for some technologies, the binder droplets are significantly smaller than the initial nuclei (about 40 times smaller than the urea seeds). In the industrial practice, short granulation times are used and coating is preferred over agglomeration for size enlargement.
In this context, and taking into account that it is difficult to establish the differences between wet and melt granulation because these processes are usually carried out with different solid cores (seeds), urea is an attractive system to study both types of granulation. The aim of this paper is to explore the effects of the binder concentration on process performance and product quality. Particularly, the influence of the binder urea concentration (from 98% to 87.5%) on the fraction of agglomerated granules is evaluated. Besides, the impact of binder concentration on granulation efficiency as well as on granule moisture content, size, morphology and crushing strength is studied.

2. MATERIALS AND METHODS

2.1. Equipment

A schematic diagram of the experimental device is shown in Fig. 1. The experiments were carried out in a batch fluidized-bed granulator (see geometrical parameters in Fig. 1) constituted by a stainless steel bottom conical vessel (1), and a cylindrical column (6) on top of it. The air distributor is a stainless steel perforated plate (2). The fluidization air was supplied by a centrifugal blower (3). Before entering the bed, the fluidization air flowrate was measured by an orifice flow-meter (4) and preheated by an electrical heater (5) to maintain the bed temperature at the desired level. The bed and grid pressure drops were manually measured by water U-tube manometers. The elutriated fine particles were collected by a set of three filter bags located at the top of the fluidized-bed freeboard (6). These filters were periodically blown back by air pulses to disengage the particulate matter. The feed (urea melt) was prepared in an oil-heated tank (7) by typically adding 1 kg of urea, the required small volume of water to reach the desired urea concentration and a tiny amount of food dye to easily monitor the fluidized-bed granulation through the unit observation window. The urea melt tank (of about 0.002 m$^3$) was placed on a scale and kept at constant temperature ($\approx$130 ºC) by means of an oil reservoir (operating at 140 ºC). The urea melt was delivered to an internal mixing two-fluid spray nozzle (8), which was located just above the air distributor, by means of a given compressed air flowrate (atomization air) that was preheated up to 130 ºC before entering into the urea solution tank (9). The atomization air was also preheated and its flowrate was controlled and measured by a valve and a rotameter, respectively. The external tube-skin temperature of the urea line (from the hot container to the spray nozzle) was controlled through an electric heat tracing system. A Programmable Logic Control system (PLC) was used to register and control process variables (Veliz Moraga et al., 2015).

2.2. Experimental Procedure

For each run, a batch of approximately 2 kg of urea seeds was initially charged into the bed chamber. The seeds had diameters between 2.38 and 2.80 mm, which correspond to ASTM meshes #8 and #7, respectively. Thus, the seed arithmetic mean diameter was 2.59 mm. The seeds were fluidized with hot air until the desired bed temperature level was achieved. Then, the urea binder was sprayed at a constant mass flowrate until 1 kg of binder has been added. This flowrate (5 g/s) was computed from the linear time profile of the recorded urea container weight. Once the binder spraying was stopped, the
granules were immediately cooled down using air at room temperature. The collected granular product was set aside for further characterization. After each experiment, the powder deposited onto the granulator walls and the fines collected in the filter bags were weighted for mass balance closure calculations. To identify the binder concentration for which the transition between wet and melt granulation takes place, experiments at different urea concentrations (87.5, 90.5, 91.8, 92.6, 96.0 and 96.7%) were performed. The urea concentration was measured immediately after granulation finished by determination of the water content in the urea melt binder that remained unsprayed in the container with a moisture analyzer (Ohaus MB45).

2.3. Granular Product and Process Characterization

2.3.1. Particle size distributions

The particle size distributions (PSDs) were evaluated for the solids collected at the end of the experiments (i.e., final granular product). A riffle splitter was used to accurately divide the total sample (about 3 kg) into representative samples. A batch of 0.8 kg was sieved in a vibratory sieve shaker (Zonytest, Argentina). A series of 9 ASTM standard sieves (# 4, 5, 6, 7, 8, 10, 12, 14 and 16) was employed. The sample was shaken for 20 min at about 2400 strokes per minute.

2.3.2. Process fines mass fractions and mass balance closure

As aforementioned, in each granulation experiment the binder was sprayed onto the urea seeds. At the end of each experiment, three types of particles were identified: granules (product), powder deposited onto the granulator walls and powder retained in the filters. Therefore, the urea mass balance is given by:

\[ M_{u0} + \dot{M}_{binder} X_u t = M_{uf} + M_f + M_w \]  \hspace{1cm} (1)

where \( M_{u0} \) is the initial mass of urea seeds, \( \dot{M}_{binder} \) is the binder mass flowrate, \( X_u \) is the urea mass fraction in the binder and \( t \) is the granulation time. \( M_{uf} \) represents the collected granular product, \( M_w \) is the mass of powder deposited onto the granulator walls and \( M_f \) is the mass of powder retained in the filters.

To determine which fraction of the atomized binder actually contributes to the granules growth, the granulation efficiency is defined as:

\[ \eta = \frac{M_{uf} - M_{u0}}{\dot{M}_{binder} X_u t} \]  \hspace{1cm} (2)

According to Eq. (2), \( \eta \) can vary between 0 and 1, where \( \eta = 1 \) indicates that all the binder is deposited onto the granules while \( \eta = 0 \) indicates that the binder does not contribute to the granules size enlargement, being therefore distributed among \( M_f \) and \( M_w \). Eq. (1) can be rewritten as:

\[ 1 = \eta + F_f + F_w \]  \hspace{1cm} (3)

where \( F_f \) and \( F_w \) represent the mass fractions of powder retained in the filters and deposited onto the granulator walls, respectively, with respect to the total sprayed urea.
2.3.3. Granular product size fractions

As schematized in Fig. 2, the granules obtained as product at the end of the experiments are classified as coated particles, agglomerates and product fines. For this reason, the following three mass fractions were defined:

- **Product fines mass fraction** ($F_p$): represents the mass of particles smaller than the seeds with respect to the total mass of granular product. Laser diffraction analysis of the collected product fines (base case) revealed that their sizes were between 32-75 µm; i.e. a very fine powder.
- **Product agglomerated mass fraction** ($A$): represents the mass fraction of agglomerated particles with respect to the total mass of granular product. When agglomeration took place, the total mass retained on some sieves corresponded to agglomerated particles while other meshes retained both, pure coated particles and small agglomerates. For these cases, a riffle splitter was used to subdivide the retained mass, and the agglomerates were recognized by visual inspection and quantified by weighting.
- **Product pure coated mass fraction** ($C$): denotes the mass fraction of pure coated particles, and was calculated as follows:

$$C = 1 - A - F_p$$  \hspace{1cm} (4)

2.3.4. Granule moisture content (MC)

Representative granule samples of 10 g (collected immediately after each granulation run) were dried at 95 °C for 9 minutes in an Ohaus MB45 moisture analyzer. This treatment profile was defined by using some selected samples and in order to reproduce the moisture content measured by the Karl-Fisher method. The moisture content was determined by triplicate.

2.3.5. Granule crushing strength (CS)

A testing machine Instron model 3369 in compression mode at a speed of 2 mm/min was used. For each experiment the crushing strength, which is affected by the granule size, was measured for the product particles that were trapped in the apertures of mesh #7 to ensure the evaluation of equal-size granules. For each sample, twenty measurements were performed.

2.3.6. Particle morphology

The morphology of the external surface and cross-sectional cuts of some selected granules were assessed in an EVO 40-XVP, LEO Scanning Electron Microscope (SEM). The samples were previously metalized with gold in a PELCO 91000 sputter coater.

3. RESULTS AND DISCUSSIONS

3.1 Experimental Results
In this section, the effect of the binder urea concentration on the urea granulation performance and product properties is presented. Table 1 reports the process conditions for the base case, which is characterized by high granulation efficiency and granules growth by pure coating. The binder urea concentration was varied from 87.5 wt% to 98 wt%, while keeping the other process conditions constant. For all experiments, it was found that \( \eta + F_f + F_w = 1 \pm 0.05 \), indicating that almost all the atomized binder was collected at the end of the operation either as granular product, wall fines or filter-bag fines (i.e., the mass balance closure was satisfied).

Fig. 3 shows the granular product size fractions (fines and coated and agglomerated granules) as a function of the binder urea concentration. At high urea concentrations, coating is the dominant growth mechanism while a combined process of coating/agglomeration takes place when the urea concentration is 87.5%. For the selected operating conditions (Table 1), agglomeration becomes an important enlargement mechanism as the urea concentration in the atomized binder droplets decreases. Indeed, below 92%, changes in the urea concentration cause simultaneously large variations in the percentages of agglomerates and coated particles that constitute the product. However, above 92%, \( C \) and \( A \) size fractions have a minor dependence on the urea concentration. For the explored conditions, the product fines mass fractions are negligible, representing less than 1.3 wt. % of the total granular product.

Fig. 4 shows the granulation efficiency as a function of the binder concentration. For urea concentrations below 92%, the granulation efficiency is lower than 80%, indicating that a significant amount of the sprayed material is not collected within the granular product. According to Eq. (3), a decrease in the granulation efficiency should be accompanied by an increase in the wall and filter-bags fines. In fact, Fig. 5 shows that the wall and filter-bags fines increase as the concentration of urea in the binder decreases. Besides, the wall fines \( (F_w) \) are higher than the filter-bag ones \( (F_f) \). These results suggest that the lower the urea concentration, the wetter the particles and the greater the loss of material by sticking on the granulator walls.

Fig. 6 presents the experimental number-volume mean diameter of the product as a function of the binder concentration. These values are compared with the theoretical \( d_{nv} \), which was calculated under the assumption of pure coating. Indeed, and according to Saleh and Guigon (2007), the theoretical \( d_{nv} \) is computed as:

\[
d_{nv,calc} = d_0 \left( \frac{\mu_f}{\mu_a} \right) ^{\frac{1}{3}}
\]  

As it can be seen, the experimental \( d_{nv} \) increases as \( X_u \) decreases, in agreement with the agglomerates formation trend (see Fig. 4). This tendency is not followed by \( d_{nv,calc} \). Therefore, the difference between the experimental and calculated \( d_{nv} \) is a good indicator for deviations from pure coating as the dominant growth mechanism in the granulation system.

Fig. 7 shows the particle moisture content and crushing strength as a function of the binder concentration. The moisture content decreases as \( X_u \) increases, basically due to the diminution in the water content of the droplets deposited on the granules. Regarding the crushing strength, it exhibits an opposite trend to that observed for the moisture.
content. In agreement with the results reported by Walker et al. (1997), the fertilizer granules with higher water content have lower crushing strength. Nevertheless, the coated particles fulfill the fertilizer granule requirements, i.e., moisture content lower than 0.5 wt% and crushing strength higher than 1.5 kgf (UNIDO/IFDC, 1998).

Concerning the granules morphology, Fig. 8 presents SEM micrographs showing the internal structure of the granules obtained with two different binder concentrations. Fig. 8.a) corresponds to $X_u = 0.89$ while Fig. 8.b) to $X_u = 0.93$. As it can be seen, an increase in the binder water content increases the porosity of the layer of deposited binder. This result is in agreement with the decrease in crushing strength observed for the granules produced under lower binder urea concentrations.

It is important to note that for the higher water content, the limits of crushing strength and moisture content are reached; therefore, it is expected (if the operating conditions are not changed to accelerate the water evaporation process) that solutions with lower urea concentration will give granules out of specification. According to Veliz Moraga et al. (2015), higher fluidization air velocities give more resistant granules with lower moisture content. These results are in good agreement with the expected heat and mass transfer enhancement for intense mixed systems. Therefore, if binders with lower urea concentration are atomized, it is convenient to operate the granulator at higher fluidization air velocities to ensure granules with acceptable moisture content and crushing strengths.

3.2 Criteria to Predict the Occurrence of Agglomeration

Although several attempts have been made in the past to establish a parameter that allows predicting the growth regimes in different types of granulation processes (Akkermans et al., 1998; Ennis et al., 1991), the one proposed by Villa and co-workers describes the boundary of agglomeration occurrence for melt granulation (Villa et al., 2016). This criterion, which is formulated in terms of dimensionless numbers that depend on process conditions and take into account mass, heat and momentum transfer phenomena, was found to be appropriate to predict the growth regime for fluidized-bed spray-on melt granulation of urea. The criterion postulates the existence of a variable (called Dimensionless Agglomeration Parameter, or $DAP$), which follows an univocous trend with the mass fraction of agglomerates.

The $DAP$ is defined as:

$$DAP = \frac{1}{K M_u} \frac{\dot{m}_a (v_f - v_{mf}) d_a \Delta H_{ fus}^2}{\psi_{mf} 4.6 \left[ X_u c_p u + (1 - X_u) c_p w \right]^{0.3} \left[ X_u \Delta H_{ fus} - (1 - X_u) \Delta H_{ ev} \right]^{1.9} \frac{\pi d_n^2 \mu_a}{4}^{2.6}}$$

where $K$ depends on physical properties, geometric dimensions, the minimal fluidization velocity and the binder urea concentration:

$$K = 0.028 \left[ \frac{4 \pi d_n \Delta H_{ fus}^2 L}{\rho_f \Delta T_{ fus}^2 / \mu_a} \right]^{0.3} \left[ \frac{X_u c_p u + (1 - X_u) c_p w}{X_u \Delta H_{ fus} - (1 - X_u) \Delta H_{ ev}} \right]^{1.9}$$

The $DAP$ takes high values for low mass percentages of agglomerates (i.e., for pure coating as the dominant growth mechanism). In the paper by Villa et al. (2016), the parameter $K$ was constant for all the experiments (i.e., the binder concentration was not modified) while process conditions (binder flowrate, fluidization air flowrate, bed temperature and seeds diameter) were varied. In this work, since the process conditions
were not modified, the DAP only changes with \( K \), which depends on the binder urea concentration.

By means of Eq. (6) and (7), Fig. 9 shows the DAP as a function of the binder concentration. Villa et al. (2016) found that if the maximum allowable value for the mass percentage of agglomerates is chosen as 5\%, the DAP should be greater than 6 whatever the chosen combination of process variables. By using this criterion, and according to Fig. 9, the urea concentration should be greater than 92.2\% in order to promote particle size enlargement by coating (i.e., minimizing agglomerates formation). This result is consistent with the trends experimentally observed, indicating that the criterion given by Villa et al. (2016) is proper to determine the boundary for agglomeration occurrence even when the binder urea concentration is modified.

Taking into account that a decrease in the binder urea concentration may be compensated with an increase in the water evaporation rate by manipulating operating variables, the effect of changes in urea concentration on the size enlargement of the particles is investigated for different fluidized-bed temperatures. In this sense, Fig. 10 shows, for different \( T_{\text{bed}} \) values, the binder concentration \( X_u \) that gives a DAP = 6 (i.e., a mass percentage of agglomerates equal to 5\%) (Eq. 6). As the bed temperature increases, the minimum binder urea concentration necessary to prevent more than 5\% of agglomerates in the product decreases. Therefore, the thermal condition of the granulator limits the water content of the binder to be used to enlarge the granules by coating. For example, if the granulator temperature is 100 °C, a urea concentration higher than 92\% ensures that A<5\%. However, if the temperature of the granulator reduces to 95 °C, the binder concentration should increase to 95.5\% to avoid the undesired agglomerates formation. On the other hand, it should be noted that a bed temperature of 90 °C is too low for a bottom-spray urea granulator since the nozzles should be kept at a sufficiently high temperature (close to the urea fusion temperature, 133 °C) in order to avoid clogging.

In industrial fluidized granulators, typically the seeds (which come in a recycle stream constituted by product out of specification) are continuously incorporated to the granulator while a concentrated urea solution (~96\%) (Nijsten and Starmans, 1998) is sprayed from the bottom of the unit. Industrial granulators often have several growth chambers, which allow the particle residence time distribution to be moved toward that of plug flow (Bertín et al., 2007). The thermal condition of the growth chambers limits the recycle operating range of industrial granulators for urea production. The bed temperatures have to remain higher than 100 °C (Kayaert and Antonus, 1997) in order to avoid melt solidification in the spray nozzles, but should not be close to the melting point (about 132 °C) to avoid partial or total quenching of the bed (Kayaert and Antonus, 1997). In particular, the granulation unit first chamber has a bed temperature lower than the next one because the recycled seeds (coming from the screen that classifies the granulator product in oversize, undersize and on specification and the crusher that reduces the oversize particles) enter the granulator at a much lower temperature. Downstream from this compartment, the temperature increases as a consequence of the binder addition that is sprayed at relatively high temperature (Bertín et al., 2010). In case the granulator chambers operate at temperatures higher than 100 °C and the atomized binder has a urea content of 96\%, there will be no considerable agglomeration according to Fig. 10. However, if due to an upset condition the binder urea concentration decreases to 90\%, the chamber temperatures should be higher than 106.5 °C to avoid agglomeration. Then, for the fluidized-bed temperature control loop,
it would be worthy to consider the urea concentration value to define the chamber temperatures’ set point to exert the control action.

4. CONCLUSIONS

This paper contributes to a better understanding of the transition between wet and melt granulation processes in fluidized beds by varying the binder concentration. For a urea (seeds) - urea (binder) system, the binder urea concentration greatly affects the granulation efficiency and granules quality. Indeed, for the tested process conditions and binder urea concentrations above 92%, the mass fraction of agglomerates in the product was lower than 10%, with a relatively high granulation efficiency. Thus, it is preferable to operate with highly concentrated solutions (almost molten material) as binder to obtain granules grown by coating without losing much material within the granulation unit. It is important to note that the critical value of 92% is a consequence of the set of operating conditions chosen to carry out the experiments. The criterion proposed by Villa et al. (2016) was found to be useful for predicting this critical value. Consequently, operating conditions and control strategies to minimize urea lumps formation in industrial urea granulators could be better defined by applying the criterion developed by Villa et al. (2016).

8. ACKNOWLEDGEMENTS

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REFERENCES


Figure 1. Schematic representation of the experimental set-up (adapted from Veliz Moraga et al., 2015).
Figure 2. Types of particles found at the end of the experiments.
Figure 3. Product mass fractions as a function of the binder urea concentration.
Figure 4. Granulation efficiency as a function of the binder urea concentration.
Figure 5. Wall and filter-bags fines as a function of the binder urea concentration.
Figure 6. Calculated $d_{nv}$ for pure coating and experimental number-volume mean diameter as a function of the binder urea concentration.
Figure 7. Moisture content and crushing strength as a function of the binder urea concentration.
Figure 8. SEM micrographs of granule internal surface for a) $X_u = 0.89$ and b) $X_u = 0.93$. 
Figure 9. Dimensionless Agglomeration Parameter as a function of the binder urea concentration.
Figure 10. Binder concentration $X_u$ for $DAP = 6$ in Eq. (8), as a function of the bed temperature.
Table 1. Process Conditions

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<td>$d_0$</td>
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</tr>
<tr>
<td>Bed temperature (°C)</td>
<td>$T_{bed}$</td>
<td>100</td>
</tr>
<tr>
<td>Urea fusion temperature (°C)</td>
<td>$T_{fus}$</td>
<td>133</td>
</tr>
<tr>
<td>Initial mass (kg)</td>
<td>$M_{u0}$</td>
<td>2</td>
</tr>
<tr>
<td>Maximum sprayed urea mass (kg)</td>
<td>$M_{us}$</td>
<td>1</td>
</tr>
</tbody>
</table>

(*) at standard conditions, (*)& at 130 °C and 3 bars.