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Fluidized-bed melt granulation: The effect of operating variables on process performance and granule properties

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ABSTRACT

This paper addresses fluid-bed melt granulation (FBMG) for seeds and a binder of the same chemical nature, i.e. with an identical melting point. The purpose of this work is to study the impact of seed size, bed temperature, binder flowrate, and fluidization and atomization air flowrates on process parameters, such as mass balance closure, fines deposited on granulator walls and granulation efficiency, and on product properties, such as particle size distribution, percentages of particles effectively coated or agglomerated and granule crushing strength, among others. This is done to find the operating regions capable of avoiding lump formation or out-of-specification granule production. This work is focused on operating conditions and seed diameter ranges not addressed before. In particular, relatively large seed particles compared to the size of the droplets, high binder/seed mass ratios, and bottom spray are used. The specific method proposed to characterize the granular product into three categories (fines, pure coated particles, and agglomerated/coated granules), allowed to identify the main growth mechanisms for an extended range of operating conditions and seed sizes. The agglomeration rate was found to increase by decreasing the fluidization and atomization air flowrates and the bed temperature, as well as by increasing the binder flowrate. The agglomerated mass fraction presented a non-monotonic behavior as a function of the seed diameter, with a minimum at a seed mean diameter of about 0.26 cm. Even though the operating variables were widely disturbed, the main particle growth mechanism was pure coating for all the tested conditions. In fact, coated particles accounted for more than 68 wt.% of the granular product. As a result, the final mass median size remained almost constant for all the studied cases. On the other hand, the span of the particle size distribution was extremely sensitive to the selected operating conditions. This contribution provides some valuable guidelines to avoid agglomeration in melt granulation processes designed to produce coated granules.

1. Introduction

Among the processes handling particulate systems, granulation is a widely-used unit operation for particle enlargement. Granulation allows the production of granules with well-defined particle size distributions and shapes for many industries such as pharmaceutical, agrochemical, detergent, and food industries, among others [1,2].

Typically, three components are needed to produce granules: initial seeds or nuclei, mixing, and a binder. The seeds are always agitated in order to achieve a good binder distribution. Depending on the mixing principle, granulators are often classified into mechanical (e.g., pans, drums, high shear granulators) or pneumatic (fluidized-bed granulators) agitated units [3]. Fluidized-bed granulators (FBG) offer some advantages with respect to other granulation systems, such as the simultaneous spraying, granulation, drying and/or cooling stages, and control, within certain limits, of the granule physical properties by manipulation of some operating variables [4].

Granulation processes are usually also classified according to the binder nature as: wet, dry or melt. In wet granulation, the liquid binder (a solution or dispersion) is distributed on the seeds and, subsequently, the granules are dried to evaporate the solvent. In dry granulation, fine solid particles are added to the agitated seed bed and the powder adherence is promoted by Van der Waals or electrostatic forces [5]. In melt granulation, powders are enlarged by using meltable materials. These last binders are added to the systems either as: 1) powders that melt during the granulation process or 2) atomized molten liquids [6]. The first melt granulation technique is usually called co-melt or *in situ* melt granulation [7], while the second method could be referred as spray-on melt granulation [8]. Certainly, co-melt granulation is not applicable to those systems in which seeds and binder have similar melting temperatures.

In the past, wet granulation has been widely used. Therefore, the number of articles related to wet granulation processes is quite large.

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In the particular case of FBGs, Smith and Nienow [9], Pont et al. [10] and Hemati et al. [11], among others, studied the influence of process variables and seeds/binders physicochemical properties on particle growth kinetics when aqueous solutions were sprayed on beds of solids fluidized by hot air. These experimental studies revealed that many variables affect particle growth mechanism: binder (composition, viscosity, surface tension, flowrate, droplet size) and seed properties (size, shape, porosity), atomization and fluidization air flowrates, nozzle location, bed temperature, etc. Among other variables, the dependence of particle growth regime (agglomeration or coating) and growth rate on excess gas velocity (i.e., the difference between actual and minimum fluidization velocities), was empirically established. For wet granulation and low excess gas velocities, agglomeration was identified as the main particle growth mechanism. For higher gas velocities, the circulation rate of particles increased, thus improving liquid distribution on seed surface and reducing bed quenching by formation of large agglomerates. Moreover, higher gas velocities increased the frequency and energy of inter-particle collisions and particle-wall impacts, breaking the binder solid bridge that may have been formed between primary seeds [9,11]. The same qualitative effect of the fluidization velocity was observed, through experimental work, for different materials and granulator operating variables. However, growth regimes were found to be sensitive to the product and type of granulation unit [8,12,13]. In order to generalize the findings of many authors about wet granulation, the paper published by Ennis et al. [14] was undoubtedly a major turning point. This author proposed a physical-based model for predicting the granules growth behavior. Even though this model is simple and based on many assumptions, it is still helpful to predict growth regimes using measurable variables.

Nowadays, research in melt granulation has become of interest for many applications. This process is considered a more attractive strategy than wet granulation for those materials that are incompatible with water; completely avoiding the use of solvents and the disadvantages associated with solvent recovery and final disposal, and minimizing the energy cost related to solvent evaporation [7,15].

Depending on granules requirements, either agglomeration or coating may be preferred; therefore, understanding the mechanisms prevailing in the granulation process is a prerequisite for obtaining proper control over product properties [16]. Unfortunately, the theories developed for wet granulation are not fully appropriate for describing fluidized-bed melt granulation (FBMG). Consequently, many authors have focused on revealing the influence of some of the most important experimental variables on product quality.

For co-melt granulation, Zhai et al. [6,17] together with Walker et al. [7,18,19] explored the effect of granulation time, binder/granule mass ratio, binder viscosity, and size of the seeds on the granule final size and growth mechanisms. Additionally, Mangwandi et al. [20] investigated the effects of fluidization air velocity, fluidization air temperature and granulation time on granule size, particle size distribution, granule homogeneity and strength.

With regard to spray-on melt granulation, Abberger et al. [15], Boerefijn and Hounslow [16], Seo et al. [21] and Tan et al. [22] studied the effects of binder spray rate, droplet size, seed size, bed temperature, atomization air pressure and fluidization air velocity on the performance of fluidized-bed melt granulation. The studies mainly consisted in *in situ* or top spray-on melt granulation based on polyethylene glycol (PEG) or Poloxamer (either melted or as solid particles) as model binder and glass ballotini or lactose as seeds. In addition, all these processes involved seeds of a very small size, mostly from 0.003 to 0.035 cm. Moreover, for spray-on systems, seed sizes were similar to or even smaller than the sprayed binder droplets. Therefore, particles grew preferentially by agglomeration, being the growth by pure coating insignificant. Even though the conclusions of these studies provide valuable insights into the field of melt granulation, they cannot be applied straightforward to the production of relative big granules through coating.

It is important to note that the granulation process is considered as one of the most significant breakthroughs in the fertilizer industry, providing products with higher resistance and lower tendency for caking and lump formation. In particular, granular urea is the most widely consumed nitrogen-based fertilizer, being critical in the modern agriculture scenario [23]. Industrial urea granulation is mainly performed in fluidized beds [2], which use a highly concentrated urea solution as binder (basically molten urea) sprayed from the bottom. Urea seeds are quite large (about 0.2 cm) and, for some technologies, the binder droplets are significantly smaller than the initial nuclei. Due to the high industrial production rates, high urea melt to seed mass ratios are required (about 50%). In the industrial practice, short granulation times are used and coating is the preferred size enlargement mechanism [24–26].

Based on microscopic observations, Chua et al. [27–29] estimated the time scale of granule–granule collision, droplet–granule collision, and droplet spreading in fluidized-bed melt granulation. They found that the relative importance of these characteristic times had an effect on particle growth mechanisms. However, many of the variables involved in the different time scales came from simulations carried out using computational fluid dynamics (CFD), which allowed to obtain droplet and particle trajectories and velocities; or microscopic determinations such as liquid–solid contact angles, restitution coefficients, instantaneous droplet and particle temperature, etc. The determination of these characteristics times and microscopic parameters is important for establishing the kinetics of the different growth mechanisms (i.e., agglomeration, coating, breakage) required to solve the population balance equation together with the mass and energy balances, in order to predict the product particle size distribution.

This paper is rather focused on the analysis of experimental data from a macroscale point of view, with the purpose of elucidating the effect of the main operating variables on growth mechanism and product quality. This approach is selected, as a first step, because industrial urea granulators are mainly operated by trial and error, being undesired shutdowns quite common and frequent due to big lump formation and out-of-specification final product [30]. Granulation upsets cause important production losses; therefore, macroscopic studies about the influence of operating conditions on growth mechanisms can be useful to make quick decisions in the operation of large-scale urea facilities.

Summarizing, it is the aim of this paper to contribute to a better understanding of the melt granulation process in fluidized beds by varying operating conditions and seed properties in ranges different from those previously reported (i.e., relatively large seed particles compared to the size of the droplets, high binder/seed mass ratios and bottom spray FBMG). The present work deals with a urea (seeds)–urea (binder) system in an attempt to study the impact of seed size, bed temperature, binder flowrate, and fluidization and atomization air flowrates on process parameters (mass balance closure, fines deposited on granulator walls, and granulation efficiency) as well as on product properties (among others, particle size distribution, percentage of particles effectively coated or agglomerated and granule crushing strength) in order to distinguish the operating regions to avoid lump formation or production of granules with low crushing strength. Particularly, in this work growth mechanisms are elucidated by the analysis of the final mass fractions of pure coated particles, agglomerated/coated granules and fine particles.

2. Particle size change mechanisms in fluidized-bed melt granulation

Tan and co-workers [31–33] and Chua et al. [27] reported possible growth and breakage mechanisms that can occur during fluidized-bed melt granulation. Based on the acknowledged feasible growth mechanisms and the observed experimental evidences for urea granulation, Fig. 1 shows a scheme of the microscopic events that can occur during the process (considering that urea is sprayed at around its melting temperature, 132 °C, and the bed is maintained at approximately 100 °C