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Quantitative validation and analysis of the regime map approach for the wet granulation of industrially relevant zirconium hydroxide powders

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ABSTRACT

The objective of this work was to study the granulation behavior of three different types of zirconium hydroxide, each varying in particle size and density. Different concentrations of yttrium (III) nitrate hexahydrate ($Y(NO_3)_3 \cdot 6H_2O$) solutions were used as a doping agent and also acted as binder. Experiments were performed using a high shear wet granulation process by adjusting two parameters, 1. liquid to solid mass ratio and 2. impeller speed, to obtain four parameter settings (referred as bounds in the paper) for a two factorial design of experiment for each powder. To understand the granule growth behavior, a regime map analysis using the growth regime map first proposed by Iveson and Litster (1998) , was carried out on the bounds. The granule growth behavior observed experimentally was compared with the regime map results. Different growth behavior was observed for different powders. Furthermore, an attempt was made to obtain a steady growth for those parameter settings that initially resulted in an induction growth. A surfactant, SDS (Sodium dodecyl sulfate), was used to improve the wetting properties of the powder and its addition to the binder solution resulted in a steady growth that is more controllable for granulation manufacturing operations compared to induction growth.

1. Introduction

Wet granulation is a common particle design and size enlargement process used as a downstream processing step in various industries dealing with particulates, such as food, pharmaceutical, catalysts and detergents. In the pharmaceutical industry for instance, to overcome poor flow and compaction characteristics of the active pharmaceutical ingredient, wet granulation is a suitable means of producing tablets. In the chemical industry, surface area and porosity are important properties of catalysts and can be altered by wet granulation [1]. In the detergent industry, gradual interest in granulation arose due to strict environmental regulations [2]. Generally, the granulation process consists of addition of a liquid binder to the powder to further agglomerate and form larger particles, which are easier to handle and also possess superior properties (e.g., flowability, uniform composition, strength, porosity, surface area, low dusting). This mixture is typically agitated using a rotating drum, high shear mixer or fluidized bed, depending upon the process requirements for granulation [3–5]. In the present study a high shear mixer granulator was used.

The granulation process is divided into various mechanisms, which are wetting and nucleation, aggregation and consolidation, layering, breakage and attrition [3]. The early stage of wet granulation is marked by wetting and formation of nuclei as a result of decrease in the surface energy of the system as the liquid wets the solid particles. Furthermore, granule growth takes place either by aggregation and consolidation or layering depending on the initial powder and binder properties. Layering includes the bonding of additional finer particles onto existing granules resulting in granules that are typically weak due to layering occurring in the predominantly capillary regime. Fine particles can form granules that also undergo aggregation and consolidation in the viscous regime wherein excess liquid squeezed out on the surface is available to form bridges between the two granules resulting in stronger larger granules. The granules formed by this mechanism are usually irregular and rough textured [5]. During granulation, granules may also undergo breakage and attrition.

Several parameters are known to affect the granule dynamics making the process operationally complex. The liquid to solid ratio and the impeller speed, however, have been found to have the most significant impact and were thus considered in this study [6,7]. The liquid to solid (LS) ratio is critical since the liquid induces granulation and an excess of it leads to over massing and slurry formation and the lack of liquid results in weak granule formation or the formation of only nuclei particles.

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The LS ratio also dictates the pore saturation of granules and consequently controls the deformability of the granules and hence the degree of coalescence [8]. Previous studies have concluded that a higher LS ratio results in shorter granulation time with similar product granule size distribution [9]. They also observed that the shape or circularity distribution profile of granules remains similar at different LS ratios by altering the granulation time, however with an increase in LS ratio, strength of the granules decreases [9]. The effect of impeller speed on granule growth has been studied by Schaefer et al. and Knight et al. [10,11]. The impeller speed increases the amount of energy input in the system. Its influence depends on the response of granules to the energy input. It can either increase granule size if the increase in impeller speed results in more deformation of granules or a decrease in the granule size if granule breakage occurs [11–13].

The theoretical aspects of the mechanisms of granulation and granule growth have been studied in great detail with the support of experimental results. However they have not always been sufficient to quantitatively predict the effect of material properties and operating parameters on granule growth. Optimization, scale-up and design are largely dependent on the effects of these parameters, hence it is very important to study dynamics of wet granulation using a quantitative engineering approach. One of the approaches is using statistical design of experiments but that involves several experiments at all levels of process development. Another approach is to predict the behavior by mathematical modeling tools, but it is still a topic of current research for granulation process. An intermediate alternative in terms of number of experiments to be performed is to develop a regime map [14,15]. Regime maps are based on granule deformation and the liquid content in the granule where the granule deformability during collision, a ratio of granule impact energy to the plastic energy absorbed per unit strain is given by the deformation number and the liquid content is measured as maximum pore saturation [16]. This is determined by the respective dimensionless groups of deformation number (De or St_{def}) on the y-axis of Fig. 1 and maximum pore saturation ($s_{\max}$) on the x-axis of Fig. 1. As described by Iveson et al. [16–17], the maximum pore saturation, S_{max} given by Eq. (1) and the deformation number, St_{def} given by Eq. (2) are defined as follows:

$$S_{\max} = \frac{w\rho_s(1-\varepsilon_{\min})}{\rho_l\varepsilon_{\min}} \tag{1}$$

$$St_{def} = \frac{\rho_g U_c^2}{2Y_g} \tag{2}$$

where, w is the binder-to-solids ratio, ρ_s is the solid density, ρ_l is the liquid density and ε_{min} is the minimum porosity which is given by Eq. (1).

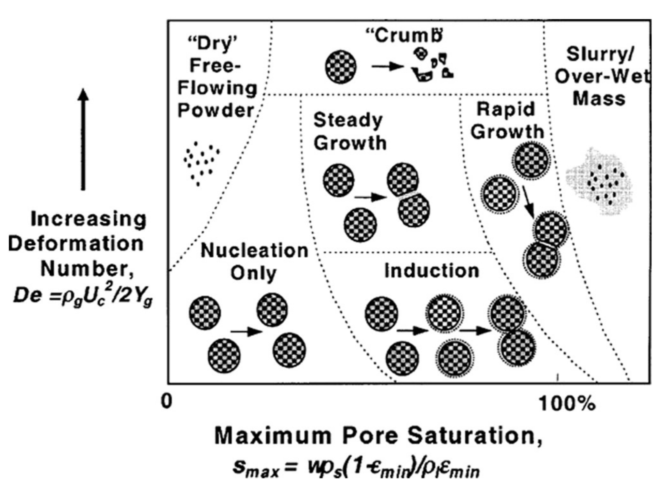

Fig. 1. Granule growth regime map adapted from [16].

ρ_g is the granule density, U_c is the particle velocity and Y_g is the dynamic yield stress of the granule. By measuring the above-mentioned granule properties, one can then compute the dimensionless groups and ascertain what kind of regime the granulation is expected to follow.

The regime map accounts for granule growth regimes such as steady growth, induction, dry free flowing powder, nucleation only, crumb, rapid growth and slurry (Fig. 1). Conceptually, it is well understood that for optimal granulation operation, the region of steady growth is desired since in that regime, the growth and increase in granule size is steady and controllable from a process point of view. However, oftentimes, due to a lack of process understanding the selection of operational parameters leads to induction growth, which is categorized by a period of little or no growth followed by a period of sudden exponential growth. This then leads to uncontrollable granule growth behavior, which is undesirable and sub-optimal. The other regimes are typically not encountered since there is sufficient understanding to eliminate combinations of operational parameters that lead to the more extreme regions in the regime maps. Therefore, this paper will focus on the steady and induction growth regimes, which as will be seen by the experimental data, are also the regions of interest and applicability in this study.

Each region on the regime map can be obtained from demarcation of experimental data points. During granulation, the liquid is distributed in the powder bed and nuclei are formed. When the amount of binder is insufficient, there is no further nucleation. When there is sufficient binder, the distribution of binder takes place and the granules start growing. If the yield strength of the granules dominates the relative particle velocity and there is slow distribution of binder, granulation is delayed (induction). This behavior is characterized by a sudden growth in granule size once its critical porosity has been achieved. At this stage the granule consolidation eventually squeezes liquid binder to the surface and rapid coalescence of the granules follows. The sudden growth in granule size makes the process difficult to control and to predict its end point [18]. When the granules are weak and easily deformable, they deform creating large surface area during impact, which promotes aggregation (steady growth). At this stage, granules size increases linearly with time. When the pore saturation is more than a hundred percent, the granules are formed within a short time from the start of process (rapid growth). The size of granules gradually keeps increasing and eventually reaches a stable size [19–25]. A key factor that determines the regime separation between induction and steady growth is the granule yield strength. This is because as the yield strength increases, granules are less deformable and form less surface area when two granules collide. The formation of this surface area is critical for sustained and steady aggregation/growth. Therefore, as yield strength remains high, induction growth is seen where the granules are not aggregating/growing sufficiently but as a result of the collisions, the granules consolidate and this results in a sudden event where the pore saturation increases past the maximum and this leads to sudden liquid surface coverage leading to rapid aggregation/growth, hence this collective behavior is known as induction growth.

Various efforts have been made to develop predictive tools for granulation by regime map analysis. An attempt to understand and predict the granule growth behavior by regime map was initiated by Iveson et al. [16]. The study proposed a regime map for granule growth to qualitatively explain the variations in the granulation behavior. Further development in the validation and applicability of regime map proposed by Iveson et al. [17] was also carried out. Regime maps have been studied for different formulation types. Rough et al. [19] validated the growth regime map using an ultra-high viscous binder [18]. Bouwman et al. [8] used Stokes deformation number to predict the material exchange behavior during granulation and effect of material properties and process conditions on exchange [15]. The applicability of regime map was investigated by studying the effect of impeller speed and LS ratio on granulation behavior by Tu et al. [4]. Certain formulations that show nucleation or induction growth were found to be highly governed