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Growth kinetics of nuclei formed from different binders and powders in vertical cylindrical mixing devices

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ABSTRACT

Granulation is the process of forming large aggregates from fine particles using a high shear mixer. This method is used in several industries from pharmaceuticals to chemical and fertilizer production. This research will study the effect of four process variables: speed of mixer rotation in the range 100–200 rpm, powder bed mass (25–40 g), mass of the initial nucleus (0.6–2 g), and binder viscosity (water, carboxymethyl cellulose (CMC) solutions with concentrations in the range 0.5–20 g/L) on single nuclei growth kinetics in low mixing devices. The powders under study were: lactose, tea, sugar, starch, and limestone. The results show the initial size of nuclei, the initial mass of the powder bed and binder viscosity and speed of rotation all influence the rate of nuclei growth. Analysis of the stokes deformation number of the nuclei show that growth rate of the nuclei decreases as the deformation number increases whilst the percentage gain in mass of the nuclei increases with increasing deformation number. The binder viscosity was shown to have the biggest influence of the growth rate of the nuclei. Results show that difference in powder density also has an effect on the growth kinetics of nuclei. The initial position of nuclei was also shown to influence the nuclei growth rate; the closer the starting position of the nuclei to the wall of the vessel the slower the growth rate.

1. Introduction

Granulation is, by definition, the act of agglomerating particles together to form larger, semi-permanent aggregates using a high shear mixer (Mirza et al., 2015). Some of the most important reasons for granulating powder-like material are eliminating dust handling hazards, increasing bulk density for storage, and creating non-segregating blends of different ingredients (Litster and Ennis, 2013). The process of granulation plays a major role in numerous industries such as fertilizers, pharmaceuticals, and fine chemicals. For instance, in the pharmaceutical industry, granulation is used to develop raw material with the required flow properties, compactness, and homogeneity (Monteyne et al., 2016).

There are two types of granulation processes. Wet granulation is the process of mixing a dry powder with a binder whereas dry granulation is the use of pressure (slugging or roller compaction) to form nuclei (Šantl et al., 2011). This project will tackle the process of wet granulation through the introduction of a wet binder into a rotating dry powder bed. This process can be divided into three stages of rate processes: wetting and nucleation, consolidation and growth, and breakage and attrition (Iveson et al., 2001a).

The first stage in the granulation process is wetting and nucleation; when the liquid binder is added to dry powder in order to form the nuclei of the granules. At this stage, a defining factor is the size of the binder droplet with respect to that of a unit particle. When the size

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of the droplet is larger than that of the particle, then a distribution of tiny granules called nuclei are formed. The binder is sprayed onto the powder bed where its drops coalesce and increase their effective size. By capillary action, the drops penetrate the powder bed and create wet clusters that break apart into granules of different sizes as a result of shear forces acting on the system. On the other hand, if the droplet size is less than the particle size, then the liquid is said to coat the particles (Litster and Ennis, 2013).

The second stage is consolidation and growth in the granulation process. It is considered as the period during which the nuclei grow; either by collision of two nuclei, mixing of the granule with dry powder, or collision of nuclei with the equipment used. Under stable conditions of granule viscosity, plasticity, and malleability, compressive and shear forces push particles close together. During this process, the initial nuclei tend to gain weight and grow. This process is dependent on the energy of collisions (Litster, 2003). The third, and final stage in the granulation process is attrition and breakage which refers to the period when the granules lose some of their gained weight due to their collision, with either the walls of the vessel or other granules (Tardos et al., 1997).

Several parameters may affect the granulation process: (i) binder introduction and content play a major role in granule growth kinetics; (ii) the way the binder is introduced changes the degree of wetting and the extent of spreading on the powder bed (Osborne et al., 2011); (iii) in addition to that, the type of binder viscosity induces consolidation and affects particle growth. The binder plays a role in defining the maximum pore saturation within the nucleus (Iveson et al., 1996). Also, the initial size of the particle plays a role in the yield strength of the granule; as the size decreases, the strength increases.

Finally, equipment speed and type is a major factor in nucleation growth kinetics. Mixer rotational speed has been the focus of many studies about granulation processes (Mirza et al., 2015; Börner et al., 2016; Mangwandi et al., 2012, 2013; Rahmanian et al., 2011). It is a source of shear forces created within the studied system. This factor can lead to both growth and breakage of formed granules (Cheong et al., 2007).

Since there are several variables affecting granulation, the purpose of this project was to develop a mathematical relationship that correlates the different variables of a wet granulation process to the growth rate of a single nucleus. In order to eliminate the effect of breakage, a low shear mixing system without an impeller was used. Powder mixing was achieved by the rotating the mixing vessel. In this research, several parameters were changed in order to find the optimal conditions that enable proper granulation. Different powder materials, namely lactose, tea, sugar, limestone, and starch were used as starting materials. The parameters that were investigated are rotational speed (150 rpm, 200 rpm, 250 rpm, 300 rpm, 400 rpm), initial mass of powder bed (25 g, 30 g, 35 g, 40 g), initial mass of granule (0.6 g, 0.8 g, 1.2 g, 1.6 g, 2 g), and binder viscosity (water, CMC (0.5 g/L), CMC (1 g/L), CMC (2 g/L), CMC (10 g/L), CMC (20 g/L)).

2. Modelling

Here, it is proposed that once a nucleus is formed it grows through a layering mechanism by capturing the primary powder particles from the moving powder bed. The rate of mass transfer between the nuclei and the bed is a function of the amount of binder available on the surface, the rolling action of the nuclei on the bed (influenced by the process conditions i.e. speed of rotation of the vessel or impeller). Assuming that the conditions in the mixer prevail such that there is no breakage of the nuclei, each nucleus will achieve a maximum size which is influenced by the availability of binder for layering.

The hypothesis is that the rate of nuclei growth decreases as the amount of binder available for nuclei growth is depleted. It is assumed to be proportional to the difference between the current nuclei size M(t) and the maximum achievable nuclei is M_{max}, hence one can write:

$$-\frac{dM}{dt} = k\,(M_{max} - M) \tag{1}$$

Re-arranging Eq. (1) gives:

$$\frac{dM}{(M_{max} - M)} = -k dt \tag{2}$$

Integrating LHS and RHS of Eq. (2) with respect to mass and time respectively gives:

$$\ln(M_{max} - M) = -kt + const. \tag{3}$$

Applying the boundary condition when t = 0, M(t) = Mo (initial mass of the nuclei), allows the value of the constant of integration to be determined:

$$const. = \ln(M_{max} - M_0) \tag{4}$$

Hence;

$$\ln(M_{max} - M(t)) = -kt + \ln(M_{max} - M_0) \tag{5}$$

Re-arranging gives:

$$\ln\left[\frac{M_{max} - M(t)}{M_{max} - M_0}\right] = -kt \tag{6}$$

The mass of a nucleus at any given time can be obtained from re-arranging Eq. (6) to make M(t) subject of the formula:

$$M(t) = M_{max} - (M_{max} - M_0)e^{-kt} \tag{7}$$

where M_{max} and M_0 are the maximum mass the nucleus can reach and the initial mass of the nucleus respectively; k is the rate of nuclei growth in s^{-1} and t is time is seconds. The parameter is introduced into the equation to take cognisance of the fact that the nuclei will not experience perpetual growth. The growth rate constant is influenced by material variables such as binder and powder properties and process variables.

3. Materials and methods

3.1. Material and equipment

The α-Lactose monohydrate, and starch were supplied by Sigma–Aldrich, UK. Limestone powder was supplied was a gift from Kilwaughter Chemicals Ltd. UK. Teawaste powder was collected from cafeteria at Queen's University Belfast. Carboxymethyl cellulose was sourced from Acros Organics US.

The particle densities of the material were determined using helium pyconometery (AccPyc II 1340 Pyconometer, Micromeritics USA) and results are reported in Table 1. It can be note that particle densities of starch and lactose were similar whilst limestone had the highest particle densities. Teawaste was the lightest material. The particle size distribution was determined using Microtrac S3500 Particle Sizer Laser diffraction system. The surface-volume diameters (d_{3-2}) of the materials are summarised in Table 1. As can be seen from Table 1 limestone, starch and lactose had similar average particle sizes.

In order to make the nuclei visible in the powder bed, they were coloured by adding methylene blue to the binder. The methylene blue that was used was produced by Alfa Aesar. In addition to that, the binder used in the experiments was