



## The study of granular agglomeration mechanism

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### ARTICLE INFO

#### Article history:

Received 6 May 2009

Received in revised form 5 January 2010

Accepted 25 January 2010

Available online 1 February 2010

#### Keywords:

Agglomeration

Binder

Nucleation mechanism

Consolidation mechanism

Coalescence mechanism

### ABSTRACT

The purpose of this study was to determine the effects of the properties of different binders on the granular agglomeration mechanism for the fabrication of pharmaceuticals. The raw materials included calcium carbonate powders with an average particle size of 32–75  $\mu\text{m}$  and four different grades (4000, 6000, 8000 and 10,000) of polyethylene glycol (PEG) used as binders. The raw material was mixed with the binder in a high shear mixer. The surface structure of the granules was analyzed by low vacuum scanning electron microscope (LV-SEM) examination. Three major agglomeration mechanisms are discussed in this study: nucleation, consolidation and coalescence. The results showed that the agglomeration growth rate increased with increasing binder viscosity during the nucleation stage, but the exact opposite phenomenon occurred in the consolidation stage. Observation of the granular surfaces showed the surfaces to be full of fine powder in the nucleation stage, but PEG crystals appeared on the surface in the consolidation stage. During the coalescence stage, the granules grew quickly due to collisions and the surface structures of the granules became full of binders, as can be seen from the LV-SEM photographs.

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### 1. Introduction

Research in the field of bioscience is important in the modern era due to its relevance to the health of human beings. One important area is pharmaceutical engineering, which is the domain of the study on medicines. The manufacturing of pharmaceuticals (in the form of tablets and capsules) is largely an exercise in powder technology [1]. The main process of this technology is the so-called agglomeration or pelletisation. Agglomeration is the process by which small particles are gathered into larger, comparatively permanent granules in which the initial powders can still be distinguished [2]. The process consists of three phases: pellet formation, homogenous-growth of the pellets, and destruction [3]. Dry particle agglomeration mainly occurs due to adhesive forces (e.g., van der Waals force, electrostatic force and magnetic force) between particles. Wet particle agglomeration is dependent on liquid forces (e.g., static capillary forces and dynamic viscosity forces) between particles [4]. The liquid content played an important role in the wet agglomeration process. Benali [5] observed the granule-surfaces produced by using different liquid to solid ratios. From SEM images four types of granules could be distinguished: nuclei, coarse-granules, spherical granules and granules formed by over-wetting.

Hoornaert et al. [6] described the growth mechanisms of wet particles agglomeration. The process consists of the sequences of nucleation, coalescence, snowballing, crushing and layering, and

abrasion. Iveson and Litster [7] also added a consolidation mechanism between the mechanisms of nucleation and coalescence, which was called rapid-growth stage.

There are a large number of particle-nucleation behaviors that occur in the initial phase of agglomeration. During this stage, most of the particles, which can be called powders, impact each other continuously. Energy is dissipated during interactions with other particles through the viscous force, surface tension and hydrostatic force. Schæfer and Mathiesen [8] considered there to be two basic mechanisms in the nucleation stage: distribution and immersion. Mort [9] reported the occurrence of a distribution mechanism when the binder droplets are smaller than the particles. However, the immersion mechanism can also be found when the binder droplets are larger than the particles.

Agglomerate consolidation through plastic deformation makes the granular structure denser [9]. Iveson et al. [10] investigated the combination process. They observed changes in the granular porosity, which influenced granule deformability and liquid saturation. They also observed that inter-particle friction and viscous dissipation had a great influence on this mechanism. Iveson and Litster [11] looked at the influence of the different properties of liquid binders and the particle size distribution on the consolidation process. They found that capillary, viscous and friction forces all affected the process of granule combination. The surface morphology of granules consolidated by collision among the granules became smoother [12]. Considering the material properties, Mill et al. [13] studied the effect of binder viscosity on agglomeration in a low shear mixer. They mixed silica sand (with a sieved size ranging between 90 and 180  $\mu\text{m}$ ) and silicone oil binders (with viscosities ranging from 20 to 500 mPa s). They

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reported that the granule size continuously decreased with increasing viscosity with binder viscosities > 100 mPa s.

Many researchers have focused on different agglomeration mechanisms and the formation of granules. There have been few studies however, reporting the properties of agglomeration mechanisms. This study mainly deals with the melt agglomeration of calcium carbonate powders and the melting state of polyethylene glycol. The purpose of this work is to illustrate the effect of the operating conditions, and to examine the main forces that lead to the mechanism of agglomeration during high shear mixing. The effects of the physical properties, such as the binder content, the viscosity of the solutions and the surface structure, on the agglomeration mechanisms are discussed.

## 2. Materials and methods

### 2.1. Materials

Calcium carbonate (Meilin White-Stone Chemical, Taiwan) powders were used as the raw material in the experiments. The surface area of the material was 21 cm<sup>2</sup>/g, the average particle size 32–75 μm, the whiteness 96 and the moisture content 0.6%. Four different grades of polyethylene glycol, 4000, 6000 (Showa, Japan), 8000 (Nippon Shiyaku Kogyo, Japan) and 10,000 (Ferak Berlin, Germany) were used as the melt binders. All were used as flakes. The physical properties of the polyethylene glycol are shown in Table 1.

Two types of electronic weight scales were used to determine the weight of the powders and binder particles. One (UWE, Taiwan) with a load capacity of 12 kg was used to decide the initial weight of the powder and binder. The other (Precisa, Switzerland) with a 0.001 g readability was employed to measure the smaller amount of particles after the experiment.

### 2.2. Equipment

The high shear mixer granulator (Yi-Chen Industry, Taiwan, ROC) shown in Fig. 1(a), consists of a bowl, a discharge point, a heater, a temperature probe, an impeller and a chopper. The volume of the inter-bowl was 10 L. The inside temperature was controlled by the heater and was measured by a temperature probe. Fig. 1(b) shows a schematic representation of the four-bladed impeller. The longer blades are 25 cm in diameter and the shorter blades 20 cm. Fig. 1(c) shows the diameter of the chopper dish, which is 10 cm, and the 2 cm wide blades.

### 2.3. The agglomeration procedure

The melting agglomeration process [14] occurred in the high shear mixer granulator. First, 2000 g of calcium carbonate were put into the bottom of the bowl, and then covered by polyethylene glycol. The heating apparatus was turned on and kept at temperature of 70 °C for 1.5 h. After the binder had melted inside the granulator, the impeller and chopper were turned on and the mixing process started. A plastic scoop was used for granule-sampling in the experiment. The sampling

locations selected were in the intermediate zone of the granular bed in the vertical direction, and at the intermediate zone between the centre and the tip of the impeller in the horizontal direction.

### 2.4. Analysis of granular size

The granular size distribution was analyzed using a shaking sieve (W. S. Tyler, USA) following the ASTM (American Society for Testing and Materials) standards. The net of the sieve was made of brass and stainless steel. Thirteen different mesh sizes of net were used in this study: 32 μm, 54 μm, 75 μm, 106 μm, 180 μm, 300 μm, 500 μm, 750 μm, 1 mm, 1.7 mm, 2.8 mm, 4 mm and 5.6 mm. After the granules were sieved, an electronic scale (Precisa, Switzerland) was used to determine the weight of the granules.

### 2.5. Analysis of liquid content

The binder content, which had been distributed throughout the porosity and the surface of granules were analyzed. The particles were put in a hot circulator oven (Channel Corporation, Taiwan) and kept at a temperature of 200 °C for 5 h. After roasting, the particles become dry and yellow in color. This was because most of the polyethylene glycol had boiled away (the boiling point is about 200 °C). However, polymers like PEG have a glass transition temperature below the boiling point. At this transition temperature the small amount of chemical bonding of PEG is destroyed and the PEG remains in a liquid state. This transition absorbs short wavelength light (e.g., blue or purple), making the liquid become yellow. However, only 1–2% of the binder had reached this transition point so it did not affect the results of the analysis.

### 2.6. Photographs

The surface structure of the particles was observed by a low vacuum scanning electron microscope (LV-SEM) (HITACHI, Japan).

## 3. Results and discussion

A set of experiments was performed to analyze the three agglomeration mechanisms: nucleation, consolidation and coalescence. The effect of the binder content on the hardness of the particles and roughness of the surface are discussed in this section. The binders were utilized at temperatures higher than their melting points.

### 3.1. Agglomeration mechanism

Fig. 2 shows the weight retained versus granule size at different times during the agglomeration experiment using PEG 6000 and calcium carbonate. In the first two time slots, 0.5 and 1 min, there are two obvious peaks for weight retained. One denotes that the powder contained few liquid binders before coming into contact with the other powders. After this the powders adhered to each other due to collisions and the liquid cohesive force. This distribution mechanism results in the left-hand peak seen in the first two time slots in Fig. 2. It can also be seen that several fine-particles might adhere to become a larger liquid drop, causing some granules to get bigger than the others. This is the immersion mechanism and causes the right-hand peak in the first two time slots shown in Fig. 2. Both of these mechanisms belong to the nucleation stage, which is the first stage of agglomeration. The powder and binder first come into contact at that time [15].

About 1.5 to 3 min after the nucleation stage the two peaks have combined with each other to become a single maximum value. This clearly indicates what we know as the consolidation stage. The process requires plastic deformation of the granular structure into a

**Table 1**  
Physical properties of the polyethylene glycol binders used in the experiments.

Type of PEG	Melting point (°C)	Boiling point (°C)	Viscosity (mPa s)	
			70 °C	100 °C
4000	53–57	200	92	85
6000	56–63	200	935	890
8000	57–63	200	1044	978
10,000	58–63	200	3622	2625

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