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The production of binderless granules and their mechanical characteristics

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Abstract

A granulation procedure is described for preparing model binderless granules from spherical polystyrene colloids. The deformation and breakage behaviour of the granules was also studied. Impact and slow diametrical compression experiments were used to simulate the mechanical response of the granules at high and low strain rates, respectively. They were found to deform elasto-plastically before fracturing in a semi-brittle manner. Densification or rearrangement of particle packing in the deformed region was concluded to be the main mechanism for energy dissipation under both impact and diametrical compression. In addition, the surface chemistry of the constituent particles within the granules was found to be one of the factors that govern the strength.

Keywords: Binderless granules; Diametric compression; Impact breakage; Autoadhesion; Humidity; Porous

1. Introduction

The production and handling of particulate solids is important for pharmaceutical, chemical, agricultural, detergent, ceramic and mineral processing industries as powders form a significant proportion of the products. Wet granulation is a widely used size enlargement process where fine powder is agitated with a liquid binder in a mechanical mixer to form larger aggregates known as granules. Generally, they will be dried causing the liquid binder to harden forming solid bridges that hold the fine particles together. Granulated products may offer improved performance since ingredients, which tend to segregate, can be bound together so providing more uniformity. Moreover, granules may dissolve more effectively since they tend to readily disperse compared to the primary particles that may 'clump' in contact with a fluid. Improved flowability is exhibited by granular material, which leads to easier handling and hence minimises material loss through dust emission. In the pharmaceutical industry, some drugs are designed so that they may be delivered directly in the form of dry powder to the human body through inhalation (Boerefijn et al., 1998; Takano et al., 2002, 2003). Therefore, it is desirable to manufacture these drugs in a granular state that can be readily disintegrated and dispersed to form an aerosol but yet sufficiently strong to survive the mechanical stresses during handling and transportation. There is a similar requirement in the ceramic processing industries where granules are disintegrated to avoid inclusions that may create defects in the final products (Kendall and Weihs, 1992). In these cases, dry binderless granules are beneficial compared to those based on liquid or solid binders, which although contributing additional strength to the granules, result in less efficient disintegration.

Recently, the feasibility of granulating pharmaceutical lactose powder without binder addition using a pressure swing fluidised bed was demonstrated by Takano et al. (2002, 2003) and the potential of binderless granulation was reviewed by Horio (2003). Small primary particle sizes are important for achieving an acceptable mechanical strength as shown in the work of Takano and co-workers (2002). The compressive fracture strength of dry binderless limestone pellets was also related to the pellet diameter and powder surface area from the analysis of Kapur and Fuerstenau (1967). The experimental work of Coury and Aguiar (1995) suggested that the fracture stress under compression of

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dry mineral granules could be predicted reasonably using Kendall's fracture mechanics approach, which accounts for surface adhesive forces between the constituent particles (Kendall, 1988). A reduction in the compressive fracture strength was reported by Vervoorn (1987) when dry titanium dioxide granules were conditioned in an environment with increasing moisture content. The reason for this phenomenon was attributed to the decrease in the effective van der Waals attractive forces between the primary particles as a result of adsorbed water at the interparticle contacts.

There is an extensive literature on the deformation and breakage of dry granular solids under slow compression. However, only limited work on the impact behaviour of dry granules, particularly porous binderless granules, is documented. Impact damage of granules is frequently encountered in process equipment and handling operations, for instance, during the discharge of granules from a storage vessel onto a conveyor belt and during pneumatic conveying. The impact study of Boerefijn et al. (1998) revealed that binderless amorphous lactose granules show extensive deformation and failed in a ductile manner. Nevertheless, embrittlement of the lactose granules was found in a humid environment. In the presence of moisture, the transformation of amorphous lactose to the monohydrate state was initiated, which was postulated to cause the formation of solid bridges at the interparticle contacts. Finally, Subero et al. (2000) attempted to study systematically the impact breakage of very porous granules produced using a special moulding technique but with the addition of a polymeric binder.

Most of the granular assemblies with autoadhesive contacts mentioned above were composed of irregular constituent particles. Since the interactions at the interparticle contacts are complex, theoretical modelling of the mechanical behaviour of these systems is complicated and sometimes not possible. In the current paper, a granulation procedure for making porous model granules with spherical constituent particles (polystyrene colloids in the present case) held together by van der Waals forces is presented. This model system may allow the existing micromechanical models of granular assemblies (Kendall et al., 1987; Thornton, 1993; Adams et al., 1997) to be evaluated or refined. Furthermore, the study of this model system is more relevant to real industrial processes as the resulting internal granule structure is produced in a mechanical mixer rather than moulding.

For the purpose of mechanical characterisation, the polystyrene granules were subjected to impact and slow diametrical compression. It is recognised that the relative humidity of the environment has a pronounced effect on the mechanical responses of some granular solids. Thus, the granules used in the current experimental work were conditioned at different relative humidities before testing. The deformation and breakage behaviour of granules impacted at increasing impact velocities were characterised in terms of the coefficient of restitution and also the breakage patterns.

2. Experimental aspects

2.1. Granule preparation

Monodispersed polystyrene colloidal particles were chosen to be the constituent particles of the granules in this investigation owing to their spherical shape and autoadhesive nature. Emulsion polymerisation of styrene monomer (Fisher Scientific UK Ltd.) with potassium persulphate (Fisher Scientific UK Ltd.) as the initiator was employed to synthesise surfactant free colloids. The styrene monomer was filtered through a packed bed of aluminium oxide powder (Fisher Scientific UK Ltd.) to remove the stabiliser before reaction whereas the potassium persulphate was used as received. The particles were prepared using the method described by Goodwin et al. (1973). Distilled water (4 l) was placed in a 5 l three-neck vessel and purged with nitrogen for 30 min, whilst the temperature was raised to 70 °C using a water bath. Styrene monomer (400 g) was added and allowed to thermally equilibrate. Potassium persulphate (0.1 g) was dissolved in 50 ml water and added to the reaction vessel, which appeared milky white after about 10 min. The reaction was allowed to proceed for 12 h under nitrogen. The resulting colloidal suspension was dialysed for 7 days, in order to remove the unreacted monomer, with two changes of water per day. The cleaned suspension was concentrated up to about 30% w/w of particles using rotary evaporation and finally the particles were oven-dried at 60 °C to a powder form. The mean diameter of the polystyrene particles was found to be approximately 516 nm with a polydispersity of 0.09 as measured by dynamic light scattering (ZetaPlus Particle Sizer, Brookhaven Instruments Corp.). The surface of the particles was negatively charged due to the presence of the sulphate groups as indicated by electrophoresis measurements.

Wet granules were produced by mixing polystyrene powder with water in a high-speed food processor (Bosch, MCM 5380) with a modified impeller. During the granulation process, 100 g polystyrene powder and 70 ml water were first agitated in the mixer at an impeller speed of 100 rpm to achieve the required distribution. The water was introduced into the mixer as drops with the aid of a burette. When the water addition was completed, the impeller speed of the mixer was increased to 300 rpm in order to promote compaction and coalescence. After granulating for 1 h, wet granules with a near spherical shape in the sieve range of 3.35–4 mm were produced. For subsequent experiments, binderless granules were obtained by drying the sieved wet masses in an oven at 60 °C. The drying process was terminated when there was no detectable weight loss of the granules. An electron micrograph of a typical granule is shown in Fig. 1. In order to examine the influence of granule size on the diametrical fracture strength, granules within the sieve range of 2–2.35 mm were produced separately by terminating the granulation process 15 min earlier, followed by the drying process described above.