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High shear granulation of binary mixtures: Effect of powder composition on granule properties

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ABSTRACT

The granular product being designed in this work required the use of two different powders namely limestone and teawaste; these materials have different bulk and particle densities. The overall aim of the project was to obtain a granular product in the size range of 2 to 4 mm. The two powders were granulated in different proportions using carboxymethylcellulose (CMC) as the binder. The effect of amount of binder added, relative composition of the powder, and type of teawaste on the product yield was studied. The results show that the optimum product yield was a function of both relative powder composition and the amount of binder used; increasing the composition of teawaste in the powder increased the amount of binder required for successful granulation. An increase in the mass fraction of teawaste in the powder mix must be accompanied by an increase in the amount of binder to maintain the desired product yield.

1. Introduction

High shear wet granulation process (HSWG) is a size enlargement process that involves conversion of primary powder particles into larger entities with improved characteristics. Some of the advantages of HSWG include increased bulk densities, improved flow properties, reduced powder segregation and better handling properties. HSWG has been applied in several industries, for example, detergent, food, pharmaceutical and agricultural, to enhance the powder processibility [1–5].

Granulation of a mixture of powders is very common especially in the pharmaceutical industries where several powder components are required to perform different functionalities. Some of the common mixtures used in the pharmaceutical industries are lactose/micro crystalline cellulose (MCC) [6–8], lactose/mannitol [9] and starch/lactose [10–14]. In most granulation applications the raw materials are of comparable bulk densities.

Granulation of binary mixtures of powders has always been proven to be a challenge especially if the powders have differing physical properties. Some of the challenges encountered include segregation of the powder particles during the mixing and preferential nucleation of the component. This often leads to a granular product with inhomogeneous composition. The interaction between the powder particles and the binder particle is crucial. In-fact wettability of the powder components has a significant influence on the success or failure of the granulation process as it impacts the nucleation process. Contact angle measurements are used to indicate the degree of wetting of solid surface by liquid droplets. The high values of contact angle show low wettability whilst small values of contact angle show high wettability. For instance, if one of the components of the powder is hydrophobic this could result in preferential granulation of the hydrophilic component [15]. It has also been shown that as the hydrophobicity of formulation increases the average size of the granules decreases and the granule structure depend on the formulation wettability [16].

Percolation theory has been used to describe wet agglomeration of binary mixtures in high shear mixer in by other researchers [17]. According to their theory it was concluded that if the components exhibit similar growth properties the growth properties would be additive. However, if the components show different growth properties the mixing ratio of the components would control the growth properties of the mixture or intermediate growth properties would be exhibited.

In the current research project, granulation of two different powder materials was necessary to produce a product for application as an improved soil conditions. Each of the components, limestone and teawaste, performs different roles. Limestone modifies the pH of acidic soils whilst teawaste improves the soil texture and introduces organic matter in the soil. In the current study the challenge faced was that the two powders have huge difference in the density which often leads to segregation of the components; limestone (~2700 kg/m^3) is denser than teawaste (~800 kg/m^3).

The aim of this research was to investigate the effect of the relative compositions of the teawaste and limestone on product yield, attrition strength and granule mass mean size. Optimum binder requirements of the different formulations were also investigated. The effect of

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washing teawaste on binder requirement and other granules properties was also studied.

2. Experimental methods

2.1. Materials characterisation

2.1.1. XRD and particle size analysis of the powders

Limestone powder supplied by Kilwaughter Chemicals Ltd., UK, was used as one of the raw materials. X-ray diffraction (XRD) analysis of the powder limestone that it was mainly composed of calcite and quartz minerals [4]. Value tea purchased from Tesco was used as the second powder component in the granulation experiments. It was used in two forms, first as received referred to as fresh tea (FT) and second it was washed in fresh hot water and dried to simulate used tea (UT). The particle size distributions of limestone and used-tea measured by Malvern Mastersizer are shown in Fig. 1. The mass median diameters (d_{50}) of limestone, FT and UT powders were 31.5 ± 4.5 μm; 311 ± 10 μm and 413 ± 31 μm respectively. The other attributes of the particle size distributions are summarised in Table 1.

Carboxymethylcellulose (CMC) salt supplied by Calbiochem, UK was used to make binder in this research project. A homogenous mix was obtained adding the CMC powder to the vortex formed by stirring the deionised water with an impeller rotating at high speed (490 rpm) over a period of 30 minutes. The CMC concentration in the solution was 5 g/L. The binder solution was stored in sealed jars until use. The viscosity of the binder solution was measured with Brookfield Viscometer RVDV-II Pro (Brookfield Engineering Laboratories, USA) and was found to be ~6600 mPa s.

The comparison between the specific pore volume and surface area of teawaste and limestone particles obtained from literature [18–20] is summarised in Table 2. The table shows that the surface area and specific pore volume of teawaste (UT) are higher than that of limestone.

2.1.2. Fourier transform infrared (FTIR) spectroscopy

FTIR was principally employed as a qualitative technique for the assessment of the chemical structure of fresh tea (FT) and the used tea (UT). The IR spectra of the samples were recorded on a PerkinElmer Spectrum 100 spectrophotometer to characterise the change in the functional groups of the material surface before and after washing with hot water (using a KBr disc technique in the range of 400–4000 cm^{-1}). The results for the analysis are shown in Fig. 2. The results do not show any significant difference between the two samples as the main peaks are still present in both samples.

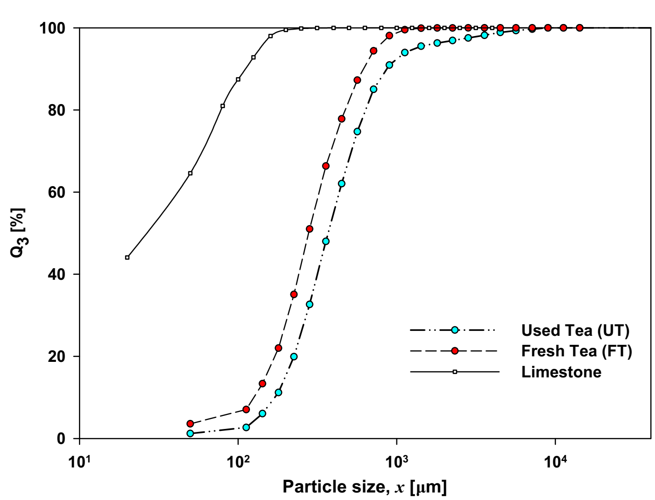

Fig. 1. Particle size distributions of the different powders used in the granulation experiments.

Table 1
Comparison of particle size distribution parameters of UT, FT and limestone powders.

Powder type	Parameter			
	d_{10} [μm]	d_{50} [μm]	d_{90} [μm]	Span
Limestone	4.7 ± 0.1	32 ± 5	114 ± 5	3.49 ± 0.13
UT	197 ± 4	413 ± 31	1065 ± 22	2.00 ± 0.36
FT	143 ± 3	311 ± 10	681 ± 31	1.73 ± 0.03

2.1.3. Contact angle measurements

The interaction between the binder droplets and the powder was analysed using the contact angle measurement. The goniometer method is the most common, simple and practical method to measure the contact angle. The contact angles on the prepared surfaces were monitored using a FTA1000B goniometer instrument (First Ten Angstroms Inc., USA). For each sample, five different readings were recorded and the contact angle values were averages of the five measurements made on different points of the sample surface. Measurements were obtained on loose beds and on compacted beds (tablets). Fifty grams of binary mixtures of teawaste and limestone with teawaste mass fractions of 0, 0.25, 0.5, 0.75 and 1.0 was obtained by mixing teawaste and limestone in the granulator for 5 minutes. For loose bed tests, small homogenous 0.5 g samples were taken from the binary mix and particles were spread on to a glass plate to form a thin layer. The contact angle was then measured by introducing the binder droplet (volume of ~15 μl). For the second set of measurements, samples of the 0.5 g were collected from the binary powder mix and then compressed into tablets by applying a load of 8 kN using a manual hydraulic press (Atlas 15T Manual Hydraulic Press, Specac Inc., USA). Fig. 3 shows the particle distribution on the surfaces of the tablets with different compositions. It can be seen in these images that the number of teawaste particles on the surface increases as the mass teawaste mass fraction is increased as expected. The images also show that the teawaste particles are angular and fibre. Binder droplet was then introduced on the tablet surface to measure the contact angle. Typical images recorded during the measurements are shown in Fig. 4.

2.2. Production of the granules and drying

The granulation experiments were carried out in a small bench scale high shear granulator; Kenwood (KM070, Kenwood, UK). The granulator has a stainless steel mixing bowl with a total capacity of 6.7 L and is also equipped with two blade impeller which also undergoes "planetary mixing" motion around the mixer during granulation. The rotational speed of the impeller can be varied between 100 and 213 rpm.

A parameter λ is defined in the following equation (Eq. (1)) was used to indicate the powder composition:

$$\lambda = \frac{m_{tea}}{m_{tot}} \quad (1)$$

where m_{tea} is the mass of tea and m_{tot} is the total mass of powder (limestone + teawaste), respectively.

This binary mix was pre-mixed for about half a minute at an impeller speed of 103 rpm. At the end of the pre-mixing stage the granulator was stopped to allow addition of the granulating fluid. After addition of the required amount binder the wet mass was mixed for half a minute and the granulator was interrupted at the end of this period to allow

Table 2
Comparison of BET surface area and specific pore volume of teawaste (UT) and limestone.

Material	Specific pore volume	BET surface area	Reference(s)
	$\times 10^{-4}$ [cm^3/g]	[m^2/g]	
Teawaste (UT)	45.1	30.04	[18]
Limestone	3.33–3.76	0.35–1.04	[19,20]