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Original Research Paper

Quantifying the effect of fillers on the breakage behaviour of needle-shaped particles

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ABSTRACT

Understanding the relationship between the applied stress and the behaviour of packings of breakable granular particles is required in many chemical engineering applications. A pharmaceutical tablet is an example where a composite packing (a packing containing a mixture of several kinds of particles) is compacted in the process of tablet formation. In this work, the unidirectional compaction and the breakage processes in composite packings formed from a mixture of breakable needle-shaped crystals and elastic spherical filler particles were studied using a Discrete Element Method (DEM) simulation. The evolution of crystal size distribution and contacts topology with the increasing stress was computed for different relative size, volume fraction and elasticity of filler particles inside the packing. We show that crystals breakability in a packing is directly related to the number of contacts among particles and that it can be significantly influenced by an appropriate choice of added filler particles. The results can be used for the development of mathematical models that describe the pharmaceutical tablet production and other processes involving composite packings of fragile particles.

1. Introduction

Packings of breakable granular particles are often present in both nature and various industrial processes, including pharmaceutics, food, cosmetics and fertilizers. Because of the enormous richness and complexity of granular motion [1], processes involving the handling of granular materials still present a challenging engineering problem. To design such processes, it is necessary to understand and be able to quantitatively describe how the particle packing responds to various kinds of stress fields and how the particle size distribution changes as a result of external stress fields (particle breakage).

When a particle packing is subjected to stress, the breakage of individual particles depends on the way the forces are transmitted through the packing. It is known that the stress propagates along force chains [2,3], i.e., through just a fraction of contacts that form a percolating network in the packing. The force chains have been observed by both experimental [4,5] and computational [6–9] techniques. The relationship between an externally applied stress and the magnitude of local forces, which are experienced by the particles and eventually cause them to break, is not clear, but most probably strongly depends on the particle coordination number and the contacts topology. The topology changes as particles rearrange and break inside the packing during its compaction. In granular physics research, packings of spheres have been well studied, while considerably less attention has been given to packings of anisotropic particles [10–16] or packings containing a mixture of several different kinds of particles (composite packings) [17,18].

An example of a composite particle packing can be found in a pharmaceutical tablet. Before tablet compaction, the active pharmaceutical ingredient (API), often present in the form of fragile needle-shaped crystals [19], is mixed with other particles called excipients that have different roles in the tablet (e.g. fillers, disintegrants, controlled release polymers, etc.). In order to maintain consistent product properties and defined dissolution profiles of the API, it is essential to understand the changes of particle size distribution that may occur during the material handling and tableting steps. The properties of single-component needle-shaped particle packings, including an investigation of particle breakage, were discussed in our previous papers [20–22]. The presence of an additional kind of particle in the packing can alter its compaction and breakage properties, but the extent of such change is currently not well understood.

Therefore, the aim of the present work is to investigate the breakage processes that occur during the unidirectional compaction of composite packings formed from a mixture of

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fragile needle-shaped crystals and elastic spherical filler particles. Using a Discrete Element Method (DEM) simulation, the evolution of particle size distribution and contacts topology with increasing stress was computed for different relative sizes, volume fractions and elasticities of the filler particles within the packing. We show that the susceptibility of particles to break in a packing is directly related to the number of contacts among the particles and that this parameter can be significantly influenced by an appropriate choice of added filler particles. The results can be used for the development of mathematical models that describe the pharmaceutical tablet production and other processes involving composite packing of fragile particles subjected to external stress fields.

2. Methodology

DEM was used for the preparation of granular packings and for the simulation of their unidirectional compaction between two parallel walls. The packing was formed from two kinds of particles: needle-shaped breakable crystals and spherical non-breakable fillers. The needle-shaped crystals were represented by the multi-element method [23] as compound particles consisting of overlapping spherical elements with radius r_C; relative positions among elements were fixed so the particle moves as a rigid body. During a simulation, the load on the particle is regularly updated and when it exceeds a set threshold, the particle is divided (broken) into two. The algorithm details are presented in our earlier paper [20] and the values of model parameters are reported in Table 1. The second kind of particles, further called fillers, are just spherical particles with a radius r_F and can have different mechanical properties (such as elastic modulus) than the crystals.

The mixed packings were prepared by placing crystals and fillers at random position (but in such a way they initially do not overlap) inside a simulation box and letting the particles settle to the bottom wall under gravity acting along the z axis. Periodic boundary conditions were applied along the x and y axes. The mixed packing can be characterised by two parameters: (i) the relative size of fillers compared to the size of crystals, $R = r_F/r_C$ and (ii) the volume fraction of fillers in the packing $\phi_F = V_F/(V_F + V_C)$, where V_F and V_C represent the volume of all filler and crystal particles in the packing, respectively.

Examples of mixed packings with various parameters R and ϕ_F are shown in Fig. 1. When forming the packings, the initial number of crystals n_{C0} and fillers n_F were chosen in such a way that the prescribed packing parameters are met and also that the volume of all particles $V_F + V_C$ is similar in all packings. Thus, the number of crystals n_{C0} varied between 450 and 1150 for packings with high and low filler volume ratio ϕ_F, respectively, and the number of filler particles n_F varied between 0 and 45000. All crystals initially consisted of $N_0 = 20$ elements and had the same initial length $L_0 = (N_0 + 1)r_C$. The ratio between the simulation box size and the initial length of crystal particles is 2. Therefore, even with periodic boundary condition, the boundary effect between longest crystals in prepared packings cannot be completely excluded. The segregation was also observed in packings with some particular combination of R and ϕ_F: the extent of segregation effect will be discussed separately at the end of the Results & discussion section.

Table 1
Parameter values used in simulations.

η_t	Global damping of the translation	158 s^{-1}
g	Gravitational acceleration	10 m/s^2
m_C	Mass of element	8.4 mg
r_C	Radius of element	1 mm
k	Elastic modulus	2005 N/m
b	Particle strength: maximum stress	641.68 or 320.84 kPa

The packing compaction and crystals breakage was then simulated by placing a top wall above the packing. The packing was compressed between the top and bottom walls by gradually increasing the weight of the top wall while keeping the position of the bottom wall fixed. As the stress propagating through the packing increased, the load on individual crystals increased as well, eventually leading to their breakage.

The following parameters were recorded at every time step i during the computational experiment: the number of crystals n_{Ci}, the number of contacts between various kinds of particles (m_{CCi}, m_{CFi} and m_{FFi} for crystal-crystal, crystal-filler and filler-filler contacts, respectively), the mean and the variance of local forces acting at particle contacts, the polydispersity index Z_i which characterises the width of the crystal length distribution, the void fraction (porosity) of the packing ε_i, and the overall dimensionless stress σ_i acting on the packing

$$\sigma_i = \left(\frac{F_i^{top}}{k_{CC} r_C}\right) \Big/ A \qquad A = A'/r_C^2. \tag{1}$$

The symbols F_i^{top} and A' in Eq. (1) are the weight and the area of the top wall and k_{CC} is the elastic modulus (stiffness) of the linear spring characterising the local force at contacts between crystals.

In addition, several packing characteristics independent of the packing size and comparable between different packings were also calculated: The average crystal length L_i

$$L_i = \left(\frac{N_0 n_{C0}}{n_{Ci}} + 1\right) r_C, \tag{2}$$

which is inversely proportional to the number of crystal breakage events ($n_{Ci} - n_{C0}$). The average length between two contacts supporting the crystals, d_i, which is defined as

$$d_i = \frac{(N_0 + 1)\, n_{C0}\, r_C}{2m_{CCi} + m_{CFi}}, \tag{3}$$

where the numerator corresponds to the total length of all crystals and the denominator is the number of crystal contacts. Finally, the filler coordination number

$$E_i = \frac{2m_{FFi} + m_{CFi}}{n_F} \tag{4}$$

characterises the contact network among the filler particles.

3. Results & discussion

The effect of two packing parameters, namely the relative size $R = r_F/r_C$ and the volume fraction $\phi_F = V_F/(V_F + V_C)$ of spherical filler particles admixed to a random packing of needle-shaped crystals, was studied in the course of DEM computational experiments. The maximum stress (shear or bending) a crystal is able to withstand before it breaks was first set to a value that ensured the crystals did not break just due to their own weight in the packing, but were sufficiently fragile to undergo fragmentation during uniaxial compaction of the packing. Apart from packings with the default setting of the crystal strength, also packings consisting of "weak" crystals whose breakage threshold was set to one half of the threshold for regular crystals were prepared.

The results (Fig. 2) show that the addition of fillers into the packing generally postpones crystal breakage to higher compaction stresses σ; the average crystal length L starts to decrease at higher values of σ. In other words, the crystals of a particular length can hold up until higher stresses in composite packings containing fillers than in the packings of crystals only. This difference increases with increasing volume fraction of the fillers and decreasing relative filler size. It can be seen, for example, that weak crystals can withstand similar stresses as regular crystals if they