Powder Technology 219 (2012) 249–256

Contents lists available at SciVerse ScienceDirect

Powder Technology

journal homepage: www.elsevier.com/locate/powtec

An experimental evaluation of the accuracy to simulate granule bed compression using the discrete element method

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ARTICLE INFO

Article history:
Received 9 August 2011
Received in revised form 21 November 2011
Accepted 25 December 2011
Available online 29 December 2011

Keywords:
Compression
Discrete element method
Contact model
Plastic overlap
Elastic deformation

ABSTRACT

In this work, granule compression is studied both experimentally and numerically with the overall objective of investigating the ability of the discrete element method (DEM) to accurately simulate confined granule bed compression. In the experiments, granules of microcrystalline cellulose (MCC) in the size range 200–710 μm were used as model material. Unconfined uniaxial compression of single granules was performed to determine granule properties such as the yield pressure and elastic modulus and compression profiles of the MCC granules were obtained from granule bed compression experiments. By utilizing the truncated Hertzian contact model for elastic-perfectly plastic materials, the rearrangement and plastic deformation stages of the force displacement curve were found to be in reasonable agreement with experiments. In an attempt to account for the final compression stage, elastic deformation of the compact, a simple modification of the contact model was proposed. This modification amounted to the introduction of a maximal plastic overlap, beyond which elastic deformation was the only deformation mode possible. Our results suggest that the proposed model provides an improved, although not perfect, description of granule bed compression at high relative densities and hence may be used as a basis for future improvements.

1. Introduction

Tablets are today the dominating dosage form available on the pharmaceutical market due to their safety and ease of administration. Compression of powders or granules (secondary particles) is however an intricate procedure. Confined compression of granular materials may typically be divided into a series of stages [1–3]. During initial compression, rearrangement of particles dominates with predominantly elastic particle deformation until a confined particle arrangement is reached and the motion is restrained. At increased pressure, the particles experience plastic deformation and possibly fragmentation that is followed by an elastic deformation of the compact. During the last stage of the compression cycle, unloading, the tablet first recovers elastically and may finally undergo an inelastic deformation at the end. Material properties such as plasticity, elasticity and contact forming ability are hence essential in order to manufacture a tablet with suitable mechanical properties.

By utilizing numerical simulations, an increased understanding of the mechanical properties that govern the compression process may be obtained. In addition, simulation may be a powerful tool in formulation development in order to predict compression properties. Numerical simulations may be based on either continuous or discrete models or a combination of both. The finite element method (FEM) is adopted for simulations at the continuum level and is typically used to determine density and stress distributions within compacts of different shapes [4–7]. The FEM however requires indirect constitutive models for the powder behaviour that make predictions based on particle properties challenging.

The discrete element method (DEM), originally developed by Cundall and Strack [8], enables simulation at a microscopic level where the interparticle forces, particle velocities, etcetera, are calculated for each time step. Compression simulations of large systems comprising several thousand particles are possible using the DEM [9,10]. In the DEM, the interparticle contacts are generally introduced as a function of the particle overlap and the contacts are typically assumed to be independent.

The FEM may be used for investigating local contact behaviours of powders during compaction when used in a combination with a micromechanical model [11]. The combined finite/discrete element (FE/DE) method gives a more thorough description of the particles during compression but is unfortunately also very time consuming. However, compression of plastically deforming granules has been investigated by the FE/DE method [12]. In order to save computational time when simulating contact forces, the extraction of a contact expression from the FE/DE method has proven successful in DEM simulations [13]. In order to simulate more realistic systems, corresponding to experimental compressions, the combined FE/DE method is impracticable. This and the simplicity of the DEM render the latter suitable for investigations of confined granule bed compressions and the DEM was therefore the focus of the current project.

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doi:10.1016/j.powtec.2011.12.054

DEM simulations have been used to investigate various powder compression processes. Both isostatic and closed die compressions have been performed to study particle rearrangement [14,15] and the effect of size ratio in binary powder mixtures [9] and for frictionless spheres [10]. In addition, simulations of uniaxial compressions have previously been evaluated by experiments. By utilizing single granule mechanical properties, the potential of DEM simulations to accurately describe the bulk mechanical response has been proven [16–18].

During compression, the coordination number and contact area change with applied pressure. In order to enable simulations of granules, the formulation of a contact law between particles is critical. Various contact models have previously been derived, e.g. the models developed by Thornton and co-workers [19,20] and Storåkers [21]. The choice of model is dependent on the material properties making the first suitable for elastic-perfectly plastic materials and the latter suitable for visco-elastic materials. As pharmaceutical materials most often display elastic-perfectly plastic behaviour the contact model developed by Thornton and Ning [20] is frequently utilized [16,18]. The model accurately describes the initial particle rearrangement and the following plastic deformation. The model does however not depict the final elastic compact deformation and hence the contact model is not suitable for compressions at high relative densities. Before initiation of compression, the relative density of randomly close packed spheres is approximately 0.64 [22]. At increased compression pressure the interparticle contacts are independent up to relative densities of approximately 0.8. At higher relative densities, the contact deformation is dependent on the influence of neighbouring particles [14], which is not considered in the contact model. However, simulations at relative densities exceeding 0.8 are feasible using a contact law derived from a combined discrete and continuum model in which Voronoi cells are used to estimate particle volumes [13].

In this work, granule bed compression was studied both experimentally and numerically with the general aim of investigating the ability of the DEM to accurate simulate confined granule compression. In order to match the experimental conditions, the properties of unconfined single granules composed of microcrystalline cellulose (MCC) were measured. In addition, granules of varying size were studied. In an attempt to account for the elastic deformation of the compact we propose a simple modification of the contact model for elastic-perfectly plastic materials. The modification amounts to the introduction of a maximal plastic overlap, where elastic deformation is initiated, in a similar manner as in a recent effective medium model of confined compression [23]. The simplicity of the modification was important in order to retain the computational efficiency of the original DEM and hence a perfect correlation to experimental data was not expected.

2. Theory

2.1. Original contact model

The normal contact force between two particles and between a particle and a confining surface was based on the model developed by Thornton and co-workers [19,20]. The classical Hertz analysis is assumed to be valid before any plastic deformation has occurred. Hence the magnitude of the normal force (F_n) increases nonlinearly with normal overlap (δ_n), according to

$$F_n = \frac{4}{3} E_* R_*^{1/2} \delta_n^{3/2}, \tag{1}$$

where R^* and E^* are the effective radius and Young's modulus (see Appendix A). A limiting contact pressure P_y (henceforth referred to as the yield pressure) is introduced, such that plastic deformation commences once the maximal pressure in the contact region equals P_y. From the Hertz analysis it follows that plastic deformation starts at a normal overlap δ_y, calculated as

$$\delta_y = \left(\frac{\pi P_y}{2E_*}\right)^2 R_*. \tag{2}$$

This overlap corresponds to a normal force of magnitude $F_y = (4/3)E^* R^{*\,1/2} \delta_y^{3/2}$, according to Eq. (1). The post-yield behaviour is inferred from a Hertzian contact pressure distribution that is truncated at P_y. As a result, the normal force increases linearly with the normal overlap during plastic loading,

$$F_n = F_y + \pi P_y R_* \left(\delta_n - \delta_y\right). \tag{3}$$

When unloading of plastically deformed particles takes place, the model accounts for plastic deformation by using a contact curvature $1/R_p$ that is smaller than $1/R^*$. The expression for the normal force becomes

$$F_n = \frac{4}{3} E_* R_p^{1/2} \left(\delta_n - \delta_p\right)^{3/2}, \tag{4}$$

where R_p and δ_p are parameters that depend on the amount of plastic loading. The parameter R_p is calculated as

$$R_p = \frac{F_e}{F_m} R_*, \tag{5}$$

where F_e is an equivalent normal force and F_m is the maximal normal force from which unloading starts. Specifically, if unloading starts from a maximal normal overlap δ_m, corresponding to a normal force F_m as inferred from Eq. (3), an equivalent elastic force F_e is determined by using δ_m in Eq. (1). Finally, the parameter δ_p is determined from the condition that Eqs. (3) and (4) must produce the same normal force at a normal overlap of δ_m.

The tangential contact force between two particles and between a particle and a confining surface was based on the modification of the Mindlin–Deresiewicz model proposed by Di Renzo and Di Maio [24]. As long as no slip occurs, the magnitude of the tangential force (F_t) hence increases linearly with the tangential overlap (δ_t) with a constant of proportionality that depends on the normal overlap,

$$F_t = \frac{16}{3} G_* R_*^{1/2} \delta_n^{1/2} \delta_t, \tag{6}$$

where G^* is the reduced shear modulus (see Appendix A). The tangential force was truncated in accordance with Coulomb friction, using a sliding friction coefficient μ_s. In addition, rolling torques were included in the model, as proposed by Zhou et al. [25], with a rolling friction coefficient μ_r.

2.2. Modified normal force model

A shortcoming of the truncated Hertzian model in its original form is that it allows plastic deformation to continue indefinitely. This poses no problems in typical DEM analyses that entail limited particle overlap, but is unsatisfactory for simulations of powder compression, where the overlap becomes much larger. During confined compression, a porous particle may densify plastically, but plastic deformation must come to an end once the particle has become essentially nonporous. To account for this phenomenon in a simplified manner, a maximal plastic overlap was introduced, denoted by δ_e, beyond which elastic deformation was the only mode of deformation possible. Once δ_e was exceeded, the elastic unloading law, Eq. (4), was in effect employed both during loading and unloading (with R_p and δ_p calculated from δ_e rather than δ_m). For contact between