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Compression mechanics of granule beds: A combined finite/discrete element study

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ABSTRACT

Compression of three-dimensional beds comprising 1000 plastically deforming initially spherical granules is investigated by using the combined finite/discrete element (FE/DE) method. The material model is formulated within the framework of multiplicative plasticity, and utilizes a density-dependent elliptic yield surface that allows porous particles to both deform and to densify plastically, whereas only volume-preserving plastic deformation is possible for nonporous ones. Granules with different characteristics (yield stress and initial porosity) are studied, and the relationship between the single-granule properties and the global compression behaviour of the granule bed is investigated. It is demonstrated that the FE/DE method may shed light on the deformation and densification behaviour of individual granules, since the size and shape of each granule are continually determined as an integral part of the solution procedure, and that the method thus provides a comprehensive picture of the processes occurring during confined compression of granular materials.

1. Introduction

There has been a considerable interest in mechanistic models for pharmaceutical processes during recent years (see, e.g., the reviews by Kremer and Hancock, 2006; Ketterhagen et al., 2009). Since the active pharmaceutical ingredient and various excipient materials typically are handled and processed in powdered form during pharmaceutical development and manufacturing, it is evident that powder technology underlies many important unit operations, especially for the generally preferred solid oral dosage forms (tablets and tablet-like delivery vehicles). The most versatile computational procedures that may be used to model these processes are based on the distinct-particle approach, using either the classical discrete (or distinct) element (DE) method (Cundall and Strack, 1979) or the combined finite/discrete element (FE/DE) method (Munjiza et al., 1995). Whereas forces essentially are considered to be functions of the particle "overlap" in the DE method, the combined FE/DE method uses a more involved particle description, with each particle being discretized into finite elements. The additional internal degrees of freedom enable a superior representation of particle deformation at the price of a significantly higher computational cost.

The DE method has been used to investigate various aspects of powder compression, such as particle rearrangement (Martin et al., 2003), the effects of particle–size ratios (Skrinjar and Larsson, 2004), and the relationship between single-particle properties and the global compression mechanics of the powder (Sheng et al., 2004; Hassanpour and Ghadiri, 2004). Its main limitation is that the simplified particle description may be unable to capture the deformation behaviour of the particles at large strains.

The use of the FE/DE method in powder compression simulations was pioneered by Ransing et al. (2000) and Gethin et al. (2001). Since the size and shape of each particle are continually determined as an integral part of the solution procedure, this method has the potential to be very useful for studying the deformation and densification of individual particles, as has previously been done experimentally (Johansson and Alderborn, 1996; Johansson et al., 1998). Knowledge of the bonding surfaces between particles and the stresses/pressures that act at these surfaces, opens up the possibility to study the formation of coherent compacts in detail (Nyström et al., 1993). However, the FE/DE method generally requires quite extensive computational resources, and most studies (also recent ones, such as those by Procopio and Zavaliangos, 2005; Choi and Gethin, 2009) have therefore been limited to two-dimensional systems.

We have recently described an efficient FE/DE procedure that enables simulations of three-dimensional systems to be performed, but have hitherto only reported results for elastic particles (Frenning, 2008). The purpose of this work is to extend this procedure to more realistic plastically deforming granules. To this end, we first describe an appropriate constitutive model for the granule behaviour, based on the elliptic yield surface proposed by Doraivelu et al. (1984) and Oliver et al. (1996), which allows porous particles to both deform and to densify plastically, whereas only volume-preserving plastic deformation is possible for

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nonporous ones. The reason for introducing this rather elaborate model is that porous granules often are encountered in pharmaceutical applications. Of particular relevance to this work are granules produced by extrusion and spheronization, typically using microcrystalline cellulose as starting material, that have been extensively used by Alderborn and co-workers in experimental studies of compression and compaction (Johansson and Alderborn, 1996; Johansson et al., 1998; Nordström et al., 2008a, 2008b, among others). Simulation results are presented and discussed, pertaining to compression of beds comprising 1000 granules with varying characteristics (yield stress and porosity).

2. Theory

2.1. Material model

2.1.1. Multiplicative plasticity

In conformity with the assumptions underlying multiplicative plasticity (see, e.g., the textbook by Simo and Hughes, 1998, which is a good source for relevant background information, or the article by Frenning, 2007, where a brief account of the theory is given), a multiplicative decomposition of the deformation gradient into elastic and plastic parts is postulated from the outset. This decomposition takes the form

$$\boldsymbol{F}=\boldsymbol{F}^e\boldsymbol{F}^p, \tag{1}$$

where $\boldsymbol{F}^e$ and $\boldsymbol{F}^p$ are the elastic and plastic parts of the total deformation gradient $\boldsymbol{F}$. From a micromechanical point of view, $\boldsymbol{F}^p$ may be considered as an internal variable that is related to the amount of slipping, crushing, yielding, and plastic bending of the constituents of the granules. The elastic response derives from a free-energy function ψ, whereas the onset and direction of plastic flow are controlled by a yield function ϕ together with an appropriate flow rule, as described below.

2.1.2. Elastic response

The elastic free-energy function is assumed to be of the compressible neo-Hookean type (Zienkiewicz and Taylor, 2005, pp. 161–163),

$$\psi=\tfrac{1}{2}\mu(\operatorname{tr}[\boldsymbol{b}^e]-3)+U(J^e), \tag{2}$$

where

$$U(J^e)=\tfrac{1}{2}\lambda(J^e-1)^2-\mu\ln J^e. \tag{3}$$

Here, $\operatorname{tr}[\boldsymbol{b}^e]$ is the trace of the elastic left Cauchy–Green tensor $\boldsymbol{b}^e=\boldsymbol{F}^e\boldsymbol{F}^{e\mathrm{T}}$, $J^e=\det[\boldsymbol{F}^e]$ is the determinant of the elastic deformation gradient, and λ and μ are material constants that for small strains may be identified as the Lamé parameters. The Kirchhoff stress tensor $\boldsymbol{\tau}$ may be determined as (Simo, 1992)

$$\boldsymbol{\tau}=2\frac{\partial\psi}{\partial\boldsymbol{b}^e}\boldsymbol{b}^e=\mu\boldsymbol{b}^e+\tfrac{1}{3}h(J^e)\mathbf{1}, \tag{4}$$

where $\mathbf{1}$ is the second-order unit tensor. To simplify the appearance of the above equation, we have introduced the function

$$h(J^e)=3J^eU'(J^e)=3[\lambda J^e(J^e-1)-\mu], \tag{5}$$

where the prime denotes differentiation with respect to the indicated argument.

2.1.3. Plastic response

We assume an elliptic yield function of the form proposed by Doraivelu et al. (1984) and Oliver et al. (1996),

$$\phi(\boldsymbol{\tau},\eta)=\|\operatorname{dev}[\boldsymbol{\tau}]\|^2+\tfrac{1}{3}a_1(\eta)(\operatorname{tr}[\boldsymbol{\tau}])^2-\tfrac{2}{3}a_2(\eta)\sigma_y^2, \tag{6}$$

where $\|\operatorname{dev}[\boldsymbol{\tau}]\|$ is the norm of the deviator of the Kirchhoff stress tensor, $\operatorname{tr}[\boldsymbol{\tau}]$ is the trace of the same tensor and σ_y is the yield stress. Moreover, $a_1(\eta)$ and $a_2(\eta)$ are functions of the relative density η. For a completely nonporous material ($\eta\geq 1$), these functions attain the values $a_1(\eta)=0$ and $a_2(\eta)=1$, which means that the yield function (6) reduces to the classical von Mises yield condition. To be specific, the functions $a_1(\eta)$ and $a_2(\eta)$ are in this work given by

$$a_1(\eta)=c_p(1-\eta^2) \tag{7}$$

and

$$a_2(\eta)=\frac{\eta^2-\eta_c^2}{1-\eta_c^2}, \tag{8}$$

where c_p and η_c are positive constants (the above expressions apply when $\eta<1$, whereas $a_1(\eta)=0$ and $a_2(\eta)=1$ otherwise, as already mentioned). Since $\operatorname{tr}[\boldsymbol{\tau}]$ is proportional to the (negative) pressure in the material, c_p may be referred to as a pressure coefficient. The constant η_c may be interpreted as the critical relative density (percolation threshold), since the effective yield stress $\sigma_y^{\mathrm{eff}}=\sqrt{a_2(\eta)}\sigma_y$ vanishes when η tends to η_c from above.

The flow rule is assumed to be of the form (Simo, 1992)

$$\mathcal{L}_v\boldsymbol{b}^e=-2\dot\gamma\frac{\partial\phi}{\partial\boldsymbol{\tau}}\boldsymbol{b}^e, \tag{9}$$

where $\dot\gamma$ is a nonnegative consistency parameter and $\mathcal{L}_v$ is the Lie derivative with respect to the spatial velocity field $\boldsymbol{v}$ (Marsden and Hughes, 1994). By straightforward differentiation one finds that

$$\frac{\partial\phi}{\partial\boldsymbol{\tau}}=2\operatorname{dev}[\boldsymbol{\tau}]+\tfrac{2}{3}a_1(\eta)\operatorname{tr}[\boldsymbol{\tau}]\mathbf{1} \tag{10}$$

and the rate of plastic volume change may therefore be determined as (Simo, 1992)

$$\frac{\mathrm{d}(\ln J^p)}{\mathrm{d}t}=\dot\gamma\operatorname{tr}\left[\frac{\partial\phi}{\partial\boldsymbol{\tau}}\right]=2a_1(\eta)\dot\gamma\operatorname{tr}[\boldsymbol{\tau}], \tag{11}$$

where $J^p=\det[\boldsymbol{F}^p]$. When porous particles deform plastically, both $a_1(\eta)$ and $\dot\gamma$ are positive, and the time derivative of the plastic volume change thus has the same sign as the trace of the (Kirchhoff) stress tensor. This in turn implies that a positive pressure (corresponding to a negative trace) results in a plastic volume decrease. On the other hand, only volume-preserving plastic deformation is possible for nonporous particles, since $a_1(\eta)=0$ for these.

Elastoplastic models of the type outlined here are generally solved by using a predictor–corrector procedure, with an elastic predictor and a plastic corrector (when needed) (Simo and Hughes, 1998). This solution procedure is referred to as a return mapping, because the elastic predictor will—for plastic steps—result in an inadmissible state of stress outside the yield surface, which subsequently is mapped back onto the yield surface. The steps required for the numerical integration of the elastoplastic model are described in the appendix.

2.2. Contact detection and enforcement

During the course of multi-particle simulations, it is necessary to detect contact between particles and to enforce the constraints imposed by nonpenetrability and friction (in this work, Coulomb friction was assumed between particles and between particles and confining walls). A two-stage contact-detection algorithm was used, as described in detail previously (Frenning, 2008). During the first stage, spatial screening was used to identify all particle pairs for which contact might occur. The detailed contact check and enforcement were performed during the second stage. Utilizing the fact that all particles were discretized by hexahedral