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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 of 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.

Keywords: Granular materials, Particulate processes, Powder technology, Solid mechanics, Mathematical modelling, Numerical analysis

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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) and 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) and by 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 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,b, among others). Simulation results are presented and discussed, pertaining to compression of beds comprising of 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

$$\mathbf{F} = \mathbf{F}^e \mathbf{F}^p, \quad (1)$$

where $\mathbf{F}^e$ and $\mathbf{F}^p$ are the elastic and plastic parts of the total deformation gradient $\mathbf{F}$. From a micromechanical point of view, $\mathbf{F}^p$ 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),

$$\psi = \frac{1}{2}\mu(\text{tr}[\mathbf{b}^e] - 3) + U(J^e), \quad (2)$$

where

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

Here, $\text{tr}[\mathbf{b}^e]$ is the trace of the elastic left Cauchy–Green tensor $\mathbf{b}^e = \mathbf{F}^e \mathbf{F}^{eT}$, $J^e = \det[\mathbf{F}^e]$ 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)

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

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

$$h(J^e) = 3J^e U'(J^e) = 3[\lambda J^e (J^e - 1) - \mu], \quad (5)$$

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),

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

where $\|\text{dev}[\boldsymbol{\tau}]\|$ is the norm of the deviator of the Kirchhoff stress tensor, $\text{tr}[\boldsymbol{\tau}]$ 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

$$a_1(\eta) = c_p(1 - \eta^2) \quad (7)$$

and

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

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 $\text{tr}[\boldsymbol{\tau}]$ 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 $\sigma_y^{\text{eff}} = \sqrt{a_2(\eta)}\sigma_y$ vanishes when η tends to η_c from above.

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

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

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

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

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

$$\frac{d(\ln J^p)}{dt} = \dot{\gamma} \text{tr} \left[\frac{\partial \phi}{\partial \boldsymbol{\tau}} \right] = 2a_1(\eta) \dot{\gamma} \text{tr}[\boldsymbol{\tau}] \quad (11)$$

where $J^p = \det[\mathbf{F}^p]$. 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 non-penetrability 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 finite elements (with quadrilateral surfaces), a symmetric two-pass node-to-surface algorithm was used, in which each surface was considered both as ‘master’ and ‘slave’. A variant of the defence node algorithm (Zhong, 1993) was employed, similar to the one used in Pronto3D and described by Heinsteins et al. (2000). This algorithm may be considered as a hybrid between penalty-type and Lagrange-type contact algorithms, with a ‘penalties’ determined from the amount of penetration (for normal contact) or slip (for frictional contacts).

3. Outline of numerical simulations

Granule characteristics that were kept constant for all simulations are summarized in Table 1. As seen, all granules were assumed to be of a spherical shape, with a diameter of 1 mm. The true density in Table 1 refers to completely nonporous granules, and the effective (or envelope) granule density is thus reduced when pores are present. To avoid the excessively long simulation times that result from the small time steps required for stiff particles (when an explicit solution scheme is employed), the granules were deliberately assumed to be very soft [experimental values of the Young’s modulus are typically 3 orders of magnitude larger (Rowe and Roberts, 1996)]. The Young’s modulus may be considered as the scale for stress, and consistent results will be obtained provided that

all stresses are scaled by the same amount, also when the constitutive model is nonlinear. This invariance has been demonstrated numerically (Kogut and Etsion, 2002). Alternatively, the particle masses could have been artificially increased, to produce a similar end result. Although this scaling may affect the dynamics somewhat, it may be noted that inertial forces – despite this scaling – generally are much smaller than contact forces, indicating that the simulation results should be representative of stiffer particles as well (although all forces and stresses will be scaled down, of course). The value of the critical relative density was estimated from the percolation threshold for microcrystalline cellulose powders, which approximately is 0.2 (Kuentz et al., 1999; Nilsson et al., 2006). All granules were discretized into regular meshes consisting of 1280 hexahedral elements (1501 nodes). Selective Rayleigh damping of non-rigid-body modes was used (Frenning, 2008), with the fraction of critical damping being set equal to 0.5.

Simulations were performed for different values of yield stress and initial relative density, with 3 levels for each factor, resulting in 9 combinations. These are summarized in Table 2, which introduces the labels that subsequently will be used to identify different granule types. The yield stress (σ_y) was scaled down in the same manner as the Young’s modulus (E), to achieve E/σ_y ratios between 10 and 100. To investigate how the described material model and parameter values translate into mechanical properties of individual granules, uniaxial compression of single granules was simulated, with the granule being confined by a stationary flat lower surface and a flat upper surface that was lowered at a rate of 5 mm/s. To simulate pharmaceutical tablet manufacture, beds comprising of 1000 granules confined in a cylindrical die cavity (diameter 11.3 mm) were generated via the DE method, as described by Frenning (2008). Particles with random initial velocities were allowed to settle in a gravitational field, with dissipation being introduced gradually to make the particles come to rest. The granule beds were uniaxially compressed by a mobile upper punch moving at a rate of 50 mm/s whereas the lower punch was stationary.

4. Numerical results and discussion

4.1. Response of single granules

The mechanical response of single granules to uniaxial compression is summarized in Fig. 1, which also shows an example of a deformed FE mesh. For the assumed elastoplastic material model, the granule response is seen to be dominated by a relatively linear plastic region, the slope of which increases with increasing yield stress and initial relative density of the granules. Following the work by Thornton and Ning (1998), the slope of this region is often considered to be a measure of an effective granule yield stress, calculated as (Samimi et al., 2005)

$$\sigma_y^{\text{est}} = \frac{4}{\pi d} \frac{dF}{dx}, \quad (12)$$

where d is the granule diameter and dF/dx is the slope of the force–displacement curve. Values of σ_y^{est} were extracted according to the above equation for strains between 0.1 and 0.4. This strain range is larger than typically employed in practical granule testing, and was selected because all curves followed a relatively linear path in this interval. In Table 3, the obtained values are compared to the effective yield stress calculated from the initial relative density: $\sigma_{y0}^{\text{eff}} = \sqrt{a_2(\eta_0)}\sigma_y$. It may be seen that both sets of values increase in a similar manner, but that σ_y^{est} always has about twice the magnitude of σ_{y0}^{eff} . An exact agreement is not to be expected, however, because the estimated and effective yield stresses are defined somewhat differently. As a check, comparison with literature results (Kogut and Etsion, 2002; Jackson and Green, 2005) indicates that the forces presented in Fig. 1 are of the correct magnitude, although no exact comparison is possible, because smaller strains and/or larger E/σ_y ratios are used in these works. Although the curves in Fig. 1 generally follow a linear path, they also exhibit more or less pronounced deviations from linearity, which typically are not seen in practical experiments (Nordström et al., 2008a). These deviations were seen to diminish with increasing mesh refinement (data not shown), and may therefore, at least partly, be attributed to the relatively coarse mesh that was used in the simulations. In addition, some dynamic effects are unavoidable when an explicit algorithm with Rayleigh damping is used, unless the loading rates are considerably reduced, which would increase the simulation times. It may nevertheless be concluded that the force–displacement curves have the same characteristics as those

of real granules, and that a range of slopes was obtained for the investigated parameter values.

4.2. Response of granule beds

The generated initial granule beds had heights of about 9.5 mm, corresponding to packing fractions of approximately 55% (extragranular porosities of 45%). These packing fractions are somewhat lower than those typically obtained for spheres, which generally range between 60% (loose random packing) and 64% (dense random packing) (Scott, 1960). The reason for this most likely is that the top surfaces of the initial granule beds were not completely level; the packing fractions determined by using a representative volume element that did not extend to the boundaries was found to somewhat smaller than 64%, within the typical range.

During the course of the simulation, the upper punch was lowered at a rate of 50 mm/s. It may be noted that the strain rate thus is comparable to the one used in the single granule simulations; although elastoplastic models of the type described in Sec. 2.1 above in principle are unaffected by the strain rate, an explicit solution procedure employing Rayleigh damping will result in some strain-rate sensitivity. Loading continued for 60 ms, resulting in maximal axial strains of slightly more than 30%. The simulations were limited to these moderate strain levels for two reasons. First, each simulation took about 3–4 days to complete, and even longer times would have been required for larger strains. Second, it was found that instabilities sometimes developed at larger strains, and that the numerical results therefore were not always reliable for axial strains above 30%. This undesirable behaviour was never observed for single granules or for regular initial packings, which suggests that the release of previously stored strain energy due to a stick–slip behaviour at the granule contacts may be the culprit. The released energy may produce a significant velocity of one or a few boundary nodes, which subsequently may cause severe distortion of one or a few finite elements. Such mesh distortion is known to adversely affect the accuracy of finite elements and may in the extreme case of inverted elements produce an unstable result. Further developments are needed to completely avoid this behaviour, which is not too uncommon in large-deformation finite-element analysis. Detrimental mesh distortion may be avoided by using an Arbitrary Lagrangian-Eulerian method (Liu

et al., 1986). Elements that may invert and recover gracefully have been proposed by Irving et al. (2006). Such procedures would result in a computational overhead, however, and are beyond the scope of this work.

Examples of configurations observed during loading are shown in Fig. 2, whereas compression profiles for all granule types are provided in Fig. 3. Since the granules are considerably softer than their real counterpart, the stress levels are much smaller than for real systems, but the results should otherwise be comparable. Contrary to the results previously obtained for purely elastic particles (Frenning, 2008), the stress at granule surfaces in contact with confining boundaries is seen to be largely unaffected by the strain for the investigated plastically deforming granules.

Compression of granule beds is often analyzed by using the Kawakita equation, which relates the degree of volume reduction (the negative engineering strain) to the applied pressure, as follows (Lüdde and Kawakita, 1966)

$$\frac{P}{C} = \frac{P}{a} + \frac{1}{ab}, \quad (13)$$

where C is the degree of volume reduction at compression pressure P . The Kawakita parameters a and $1/b$ may be interpreted as measures of the maximal engineering strain and the initial compressibility of the granule bed, respectively (Nordström et al., 2008a). Although our simulation results did not extend to the large strain typically encountered in practical applications, it is nevertheless of interest to see how well the Kawakita equation is able to describe the profiles. As may be seen in Fig. 4, an initial curvature was observed in the P/C vs. P plots, but for moderate to large strains ($C \gtrsim 0.25$) a good agreement was seen for granules of type A and B (but not for type C; data not shown). It is in this regard of interest to note that previous DE studies (Hassanpour and Ghadiri, 2004) have indicated that E/σ_y ratios $\gtrsim 30$ are required for the Heckel parameter (Heckel, 1961) to reflect the yield stress. The extracted Kawakita parameters are summarized in Table 3. Despite that only limited strains were used in the simulations, it is evident that the Kawakita $1/b$ parameter rather nicely reflects the (effective) yield stress of the single granules, although the magnitude of $1/b$ is considerably smaller. It should be kept in mind that the results obtained from the Kawakita equation to some extent are affected by the choice of starting height, relative to which the degree of compression is evaluated

(Nordström et al., 2008a). This also affects the shape of the P/C vs. C curves at low pressures. In this work, the starting height was based on the initial height of the granule bed.

From the simulations, it is straightforward to extract micromechanical results as well. Figure 5 displays the evolution of the coordination number (average number of particle contacts) with strain. As may be seen, there is a short region during which the coordination number is very low, indicative of particle rearrangement, followed by, in order, a rapid and a more gradual increase. These three regions have previously been identified in DE (Sheng et al., 2004) and FE/DE simulations (Frenning, 2008). Only relatively modest differences are observed between granule types (see the inset in Fig. 5), and for moderate to large strains there is a tendency for the coordination number to be somewhat larger for the less porous granules.

Similarly, Figs. 6 and 7 show the evolution in average relative density and Heywood (surface–volume) shape coefficient α_{SV} of the granules. The latter is calculated as (Heywood, 1954)

$$\alpha_{SV} = d \frac{S}{V}, \quad (14)$$

where S and V is the surface area and volume of a particle of diameter d . The surfaces and volumes were calculated from the deformed granule shapes, as given by the FE mesh. For a perfect sphere, the Heywood shape coefficient attains a value of 6, but somewhat larger values were obtained also for the undeformed granules, because the FE mesh was unable to fully represent the spherical shape. For the moderate strain levels used here, the average particle density (Fig. 6) is seen to be relatively independent of yield stress for the porous particles (type 1 and 2). Larger differences were observed for the nonporous ones, for which the average relative density is seen to increase with increasing yield stress. Comparison with Fig. 7 reveals that a small increase in relative density generally corresponds to a large increase in the Heywood shape coefficient (and vice versa). These findings may be interpreted as resulting from the extent of plastic contra elastic deformation. Plastic deformation is generally localized, and may thus produce relatively large increases in the Heywood shape coefficient without significantly affecting the average relative density. Elastic deformation tends, on the other hand, to result in a global deformation of the granules, resulting in relatively small increases in the

Heywood shape coefficient also when the average relative density increases significantly. It is emphasized that all values reported here pertain to the loaded state, contrary to experimental investigations, in which the size and shape of unloaded granules have been determined (Johansson and Alderborn, 1996; Johansson et al., 1998). For this reason, elastic deformation affects the pellet size and shape, which in turn makes differences between high and low porosity granules less pronounced. It may also be noted that larger strains would have been required to numerically reproduce the relatively large changes in particle size and shape observed experimentally (Johansson and Alderborn, 1996; Johansson et al., 1998).

5. Conclusions

The results presented in this work demonstrate that the combined FE/DE method may be successfully applied to investigate compression of beds comprising of ~ 1000 plastically deforming granules. The method may be used to investigate the relationship between single-particle properties and the global compression behaviour of the granule bed, as quantified via the Kawakita equation, for instance. In analogy with the results of previous studies, it was found that the Kawakita $1/b$ parameter reflects the (effective) yield stress of the granules provided that the E/σ_y ratio was sufficiently large ($\gtrsim 30$), whereas the results obtained for smaller ratios were inconclusive. It has also been shown that the FE/DE method may shed light on the deformation and densification behaviour of the granules. However, in order to be able to fully exploit the advantages of the method, more work is needed to develop procedures that allow more extensive granule deformation to be simulated, and to reduce the simulation times, if possible.

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Appendix A: The return mapping algorithm

This appendix describes the steps required for the numerical integration of the elastoplastic model in Sec. 2.1. The finite-element procedure naturally leads to a strain-driven

integration of the elastoplastic constitutive equations. The basic problem to be solved may be formulated as follows: Given a database of known values at time t_n (consisting of $\mathbf{b}_n^e$ and either $J_n = \det[\mathbf{F}_n]$ or $\eta_n = \eta_0/J_n$) and a prescribed incremental displacement field $\Delta \mathbf{u}_n$ (that translates into a known relative deformation gradient $\mathbf{f}_{n+1} = \mathbf{F}_{n+1} \mathbf{F}_n^{-1}$ that maps the configuration from time t_n to time t_{n+1}), determine the stress $\boldsymbol{\tau}_{n+1}$ at time t_{n+1} and update the database.

As mentioned, this problem is generally solved by using a predictor–corrector procedure, with an elastic predictor and a plastic corrector (when needed). Notice that the definition of the relative deformation gradient furnishes updates of J and/or η ,

$$J_{n+1} = \det [\mathbf{f}_{n+1}] J_n , \quad (\text{A.1})$$

$$\eta_{n+1} = \frac{\eta_0}{J_{n+1}} = \frac{\eta_n}{\det [\mathbf{f}_{n+1}]} , \quad (\text{A.2})$$

regardless of whether the deformation is purely elastic or not.

First an elastic trial state is determined by freezing plastic flow:

$$\mathbf{b}_{n+1}^{e \text{ tr}} = \mathbf{f}_{n+1} \mathbf{b}_n^e \mathbf{f}_{n+1}^T , \quad (\text{A.3})$$

$$J_{n+1}^{e \text{ tr}} = (\det [\mathbf{b}_{n+1}^{e \text{ tr}}])^{1/2} , \quad (\text{A.4})$$

$$\boldsymbol{\tau}_{n+1}^{\text{tr}} = \mu \mathbf{b}_{n+1}^{e \text{ tr}} + \frac{1}{3} h(J_{n+1}^{e \text{ tr}}) \mathbf{1} . \quad (\text{A.5})$$

If $\phi_{n+1}^{\text{tr}} = \phi(\boldsymbol{\tau}_{n+1}^{\text{tr}}, \eta_{n+1}) \leq 0$, deformation is purely elastic and the trial values are the correct updates.

If $\phi_{n+1}^{\text{tr}} = \phi(\boldsymbol{\tau}_{n+1}^{\text{tr}}, \eta_{n+1}) > 0$, deformation is plastic, and the algorithmic counterpart of the flow rule (9),

$$\mathbf{b}_{n+1}^e = \mathbf{b}_{n+1}^{e \text{ tr}} - 2\gamma \left(\frac{\partial \phi}{\partial \boldsymbol{\tau}} \mathbf{b}^e \right)_{n+1} , \quad (\text{A.6})$$

is invoked.

Introducing the normalized deviatoric tensor,

$$\mathbf{n} = \frac{\text{dev} [\boldsymbol{\tau}]}{\|\text{dev} [\boldsymbol{\tau}]\|} , \quad (\text{A.7})$$

decomposing $\mathbf{b}^e$ into spherical and deviatoric parts,

$$\mathbf{b}^e = \text{dev} [\mathbf{b}^e] + \frac{1}{3} \text{tr} [\mathbf{b}^e] \mathbf{1} , \quad (\text{A.8})$$

and noting that the hyperelastic relationship (4) implies that

$$\text{dev} [\boldsymbol{\tau}] = \mu \text{dev} [\mathbf{b}^e] , \quad (\text{A.9})$$

one thus obtains

$$\begin{aligned} \frac{\partial \phi}{\partial \boldsymbol{\tau}} \mathbf{b}^e &= 2 \|\text{dev} [\boldsymbol{\tau}]\| \|\text{dev} [\mathbf{b}^e]\| \mathbf{n}^2 \\ &+ \frac{2}{3} (\|\text{dev} [\boldsymbol{\tau}]\| \text{tr} [\mathbf{b}^e] + a_1 \text{tr} [\boldsymbol{\tau}] \|\text{dev} [\mathbf{b}^e]\|) \mathbf{n} \\ &+ \frac{2}{9} a_1 \text{tr} [\boldsymbol{\tau}] \text{tr} [\mathbf{b}^e] \mathbf{1} . \end{aligned} \quad (\text{A.10})$$

With the aid of Eq. (A.9), it may be seen that the norm of the first term in the above equation equals $2 \|\text{dev} [\boldsymbol{\tau}]\|^2 / \mu \leq 4 a_2 \sigma_y^2 / (3 \mu)$, where the inequality follows from the yield condition (6). Provided that $\sigma_y \ll \mu$, which we assume to be the case, this term may thus be neglected.

For convenience defining

$$\theta = \mu \text{tr} [\mathbf{b}^e] , \quad (\text{A.11})$$

$$\alpha = \text{tr} [\boldsymbol{\tau}] = \theta + h(J^e) , \quad (\text{A.12})$$

$$\beta = \|\text{dev} [\boldsymbol{\tau}]\| = \mu \|\text{dev} [\mathbf{b}^e]\| , \quad (\text{A.13})$$

the algorithmic flow rule (A.6) takes the form

$$\begin{aligned} \mu \mathbf{b}_{n+1}^e &= \mu \mathbf{b}_{n+1}^{e \text{tr}} - \frac{4}{3} \gamma \left[\beta_{n+1} (\theta_{n+1} + a_1 \alpha_{n+1}) \mathbf{n}_{n+1} \right. \\ &\quad \left. + \frac{1}{3} a_1 \alpha_{n+1} \theta_{n+1} \mathbf{1} \right] . \end{aligned} \quad (\text{A.14})$$

By taking the trace of this expression, one finds that

$$\left(1 + \frac{4}{3} \gamma a_1 \alpha_{n+1} \right) \theta_{n+1} = \theta_{n+1}^{\text{tr}} \quad (\text{A.15})$$

and by taking the deviator that

$$\left(1 + \frac{4}{3} \gamma a_1 \alpha_{n+1} + \frac{4}{3} \gamma \theta_{n+1} \right) \beta_{n+1} = \beta_{n+1}^{\text{tr}} , \quad (\text{A.16})$$

$$\mathbf{n}_{n+1} = \mathbf{n}_{n+1}^{\text{tr}} . \quad (\text{A.17})$$

Hence $\mathbf{n}_{n+1}$ may be computed solely from trial values.

Integrating Eq. (11), governing the plastic volume change, using a backward Euler procedure, one finds that

$$(1 - 2\gamma a_1 \alpha_{n+1}) J_{n+1}^p = J_n^p . \quad (\text{A.18})$$

Using the identity $J = J^e J^p$ [obtained by taking the determinant of both members of Eq. (1)], the above expression may be rephrased as

$$J_{n+1}^e = (1 - 2\gamma a_1 \alpha_{n+1}) J_{n+1}^{e \text{ tr}} , \quad (\text{A.19})$$

where we have used the fact that $J_{n+1} J_n^e / J_n = J_{n+1}^{e \text{ tr}}$. As long as $a_1 > 0$ it will be convenient to consider $\epsilon = 4\gamma a_1 \alpha_{n+1} / 3$ (rather than γ) as the unknown variable. The relevant equations then take the form

$$\theta_{n+1} = \frac{\theta_{n+1}^{\text{tr}}}{1 + \epsilon} , \quad (\text{A.20})$$

$$J_{n+1}^e = \left(1 - \frac{3}{2}\epsilon\right) J_{n+1}^{e \text{ tr}} , \quad (\text{A.21})$$

$$\alpha_{n+1} = \theta_{n+1} + h(J_{n+1}^e) , \quad (\text{A.22})$$

$$\beta_{n+1} = \frac{\beta_{n+1}^{\text{tr}}}{1 + \epsilon + \delta} , \quad (\text{A.23})$$

where we have let

$$\delta = \frac{4\gamma\theta_{n+1}}{3} = \frac{\epsilon\theta_{n+1}}{a_1\alpha_{n+1}} . \quad (\text{A.24})$$

It may be noted that the above equations yield the correct response also when $a_1 \rightarrow 0$ (in which case $\epsilon \rightarrow 0$), provided that γ is considered as the unknown variable. When $a_1 > 0$, the consistency condition,

$$\phi_{n+1} = \beta_{n+1}^2 + \frac{1}{3}a_1\alpha_{n+1}^2 - \frac{2}{3}a_2\sigma_y^2 = 0 , \quad (\text{A.25})$$

provides a nonlinear equation for ϵ that may readily be solved by Newton's method. When $a_1 \rightarrow 0$, the classical radial return map is recovered.

Updated values of the stress and strain are finally determined as

$$\boldsymbol{\tau}_{n+1} = \beta_{n+1} \mathbf{n}_{n+1}^{\text{tr}} + \frac{1}{3}\alpha_{n+1} \mathbf{1} , \quad (\text{A.26})$$

$$\mathbf{b}_{n+1}^e = \frac{\beta_{n+1}}{\mu} \mathbf{n}_{n+1}^{\text{tr}} + \frac{1}{3} \frac{\theta_{n+1}}{\mu} \mathbf{1} , \quad (\text{A.27})$$

and $\mathbf{b}_{n+1}^e$ and J_{n+1} or η_{n+1} are stored for later retrieval.

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Figures

Figure 1: Results from simulations of uniaxial compression of single granules (labelled according to Table 2). An example of a deformed FE mesh is also shown.

Figure 2: Examples of configurations during compression of 1000 initially spherical granules (of type B2; Table 2) at three instants of time (corresponding to axial strains of about 10, 20 and 30%).

Figure 3: Compression profiles for beds of granules (labelled according to Table 2): axial strain vs. applied pressure.

Figure 4: Compression profiles for granules of type A and B (Table 2) plotted according to the Kawakita equation.

Figure 5: Coordination numbers for granules of different porosity (1–3 according to Table 2). The inset shows a magnification of the behaviour at large strains.

Figure 6: Evolution in average relative granule density with axial strain. The curves are labelled according to Table 2.

Figure 7: Evolution in Heywood shape coefficient with axial strain. The curves are labelled according to Table 2.

Tables

Table 1: Fixed parameters used in the simulations.

Symbol	Meaning	Value
d	Particle diameter	1 mm
ρ	True density	1.5 g/cm ³
E	Young’s modulus	1 MPa
ν	Poisson’s ratio	0.3
c_p	Pressure coefficient	0.3
η_c	Critical relative density	0.2
μ_{pp}	Friction coeff. (particle–particle)	0.1
μ_{pw}	Friction coeff. (particle–wall)	0.1

Table 2: Variable parameters used in the simulations: Yield stress σ_y and initial relative density η_0 . Granule types are labelled as A1–C3, as indicated in the table.

σ_y (kPa)	η_0 (–)		
	0.6	0.8	1.0
10	A1	A2	A3
30	B1	B2	B3
100	C1	C2	C3

Table 3: Effective yield stress σ_{y0}^{eff} , estimated yield stress σ_y^{est} , and the Kawakita parameters a and $1/b$.

Granule type	σ_{y0}^{eff} (kPa)	σ_y^{est} (kPa)	a (–)	$1/b$ (kPa)
A1	5.77	16.1	0.636	3.72
A2	7.91	20.4	0.577	4.44
A3	10.0	23.7	0.510	4.92
B1	17.3	40.6	0.603	8.17
B2	23.7	51.7	0.568	10.4
B3	30.0	59.9	0.538	12.3
C1	57.7	130	–	–
C2	79.1	178	–	–
C3	100	214	–	–