Towards an Improved Understanding of Strength and Anisotropy
of Cold Compacted Powder

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To my dear parents, Yongming Wang and Qinmei Xu

and

To my dear wife, Er Zhou
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Abstract

Towards an Improved Understanding of Strength and Anisotropy of Cold Compacted Powder

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The strength of powder compacts after cold compaction is known to be anisotropic, which comes from the directionality of microstructure resulting from initial particle morphology and/or from particle deformation during compaction. Current work focuses on multi-scale numerical analysis of powder compaction with emphasis on the role of interparticle cohesion on post-compaction mechanical properties. At macroscopic level, we applied phenomenological model to describe the mechanical behavior of powder, in which the material is considered to be continuum medium. A user subroutine (VUMAT) was successfully developed for ABAQUS/Explicit analysis, in which one of the popular phenomenological models for powder compaction - Drucker Prager/Cap model - is implemented. By studying of pharmaceutical powder die compaction and subsequent diametrical compression test via finite element analysis, the capabilities and limitations of current constitutive models are evaluated on predicting such as density, stress and tool force evolution, as well as the strength and fracture tendency. Our results illustrate that current model has good predictive capability of powder densification (e.g. density evolution) but can not predict post-compaction strength well.

The following studies focus on evaluating the physics and mechanics occurring at particle level. The compaction of granular media was explored by using MPFEM
approach. In the new model, individual particles discretized with a finite element mesh allow for a full description of contact mechanics and local and global particle kinematics. The introduction of a layer of degrading material on the surface of each particle provides the means of introducing variable cohesion and its effect on the final strength of compacts. The simulations show that the unloading creates tensile stresses at the root of the contact necks, which may cause partial or full separation of contact interface when the cohesion developed during loading is not strong enough. These results, which are in agreement with recent strength anisotropy data for cold compaction, bring a new perspective on understanding the interparticle behavior and the origin of the strength and failure of cold compacts.
CHAPTER 1: INTRODUCTION

1.1 Motivation

The compaction of powders into near net-shaped parts has become a successful and well-established process for metals, alloys, polymers, ceramics and composites. Green compacts\(^1\) may be used as final products or are subjected to further processing such as sintering. In either case, green density and strength are important properties and determine the success of cold compaction and possibly that of subsequent operations. Process and properties optimization has always been a major concern in industrial practice. It is usually attempted by detailed considerations of die and punch design, lubrication, and proper selection of powders. The goal is to control the deformation during compaction and ensure minimum variation of density, avoidance of cracking, dimensional control, and elimination of defects.

The process of powder compaction is a complex non-linear problem. When a force is applied on a powder bed, a number of mechanisms become involved in the transformation of the powder into a porous, coherent compact with a well defined shape. Normally, the following processes are involved in a typical compaction of powders:

- particle rearrangement
- elastic deformation of particles
- plastic deformation of particles
- fragmentation of particles

\(^1\) Green compact: powder mechanically pressed into a solid at a low homologous temperature, at which no diffusional mechanisms are contributing to the strength of the compact.
- formation of interparticulate bonds
- elastic recovery of compacts

Under the action of these mechanisms, the pressed powder attains a level of cohesion, which usually increases with applied stress and higher relative density. The properties of green compact, such as relative density and strength, are not only related to the nature of the powders (e.g. composition, particle size and size distribution, particle shape, mechanical behavior of particles, particle surface morphology, existence of oxides or other impurities at the particle surface, interparticle friction coefficient), but also depend on the mode and history of externally applied forces. More specifically, stress loading path affects particle deformation and development of contact areas, which in turn affects the strength and anisotropy of compact.

Since the mid-seventies, modeling of powder compaction has provided an effective way to understand the basic mechanisms and optimize powder compaction. These models are typically continuum mechanics-based phenomenological models. Currently, the use of these models in the finite element analysis has been successful in predicting density inhomogeneity in complex shaped porous compacts [1-6]. Despite the significant progress made, many questions are still open. Understanding of the post compaction mechanical properties is still primitive because practically all existing phenomenological models consider strength as a function of density only.

Early experimental results showed that the strength of powder compact is loading path dependent [7]. Therefore density itself is not enough to describe the mechanical behavior of a green compact. Additionally, all common phenomenological isotropic continuum
models assume that the strength of the green compact approaches the strength of the particle itself, when the relative density reaches one. In reality the strength of green compacts is much smaller than the corresponding strength of the particle. As a result, the prediction of mechanical strength of a green compact with current models is not possible. The formation of defects during unloading and ejection, as well as the residual stresses and material elastic recovery may also have a large effect on the final strength of the green compacts [8, 9].

Recent results on mechanical strength anisotropy of green compact add to the list of topics that we do not have a good handle on the prediction of the strength with current modeling approaches [10]. With these in mind, the goal of this thesis is to make inroads on the understanding of the green strength in powder compacts. In the next section, we review the pertinent literature.

1.2 Literature review

In the past two decades, modeling and simulation play an increasingly important role in the design and optimization of compaction operations (e.g. [4], [11-13]). The most common models are phenomenological and isotropic. In these models, the compact is considered as a continuum medium whose mechanical behavior is a function of the relative density, and its plastic behavior is pressure dependent. Early phenomenological models allowed for a first order understanding of powder compaction operations. These semi-empirical symmetric ellipse models were originally developed for sintered compacts.
This type of model uses associative flow\(^1\) rule and isotropic hardening. Such models were originally proposed by Kuhn and Downey [14] and later by Green [15], Shima and Oyane [16], Weber and Brown [17] and others (Doraivelu et al., [18]). Several variations of the original elliptic yield locus models [19] are still used because they need minimum amount of experimental calibration. For material with low cohesion, however, tensile and compressive response should be asymmetric. Therefore models such as Cam-Clay model (or critical state model) may be more appropriate [20]. In this model, the failure surface is characterized by elliptic arcs with different eccentricity. This type of model was initially used in soil mechanics to describe the deformation of clay under triaxial stress conditions. The most common models used today are based on the Drucker Prager Cap model [21, 22], because they are able to capture the weak response of partially compacted powder under low triaxiality stress and their densification hardening under high compressive triaxialities.

Figure 1.1 shows schematically the three types of models: elliptical model, Cam-Clay model and Drucker Prager Cap model that plotted in \( p - q \) plane. In the figure, \( p \) is the hydrostatic pressure defined as:

\[
p = -\text{trace}(\sigma_y) = \frac{1}{3}(\sigma_{11} + \sigma_{22} + \sigma_{33})
\]

1.1

and is relate to volume change. \( q \) indicates Mises equivalent stress and is defined as:

\[
q = \sqrt{\frac{1}{2} \left[ (\sigma_{11} - \sigma_{22})^2 + (\sigma_{11} - \sigma_{33})^2 + (\sigma_{22} - \sigma_{33})^2 \right] + 3\sigma_{12}^2 + 3\sigma_{13}^2 + 3\sigma_{23}^2}
\]

1.2

where, \( \sigma_y(i,j = 1,2,3) \) are the components of stress in the general stress tensor:

\(^1\) The plastic flow potential function is the same as yield function.
\[
\sigma_y = \begin{pmatrix}
\sigma_{11} & \sigma_{12} & \sigma_{13} \\
\sigma_{21} & \sigma_{22} & \sigma_{23} \\
\sigma_{31} & \sigma_{32} & \sigma_{33}
\end{pmatrix}
\]

As shown in Figure 1.1, in all three models, plastic yielding of the compact is a function of hydrostatic pressure, Mises equivalent stress and relative density. As the overall density increases during consolidation, the yield loci expand, indicating the strong resistance to further plastic deformation of the materials. The cross-hatched symbols in each figure indicate the experimental measurements needed to calibrate the yield surfaces. Comparing these three models, the elliptical curves in the high pressure region of the yield loci are similar. On the other hand, these three models differ substantially in the low pressure region. Therefore, the selection of the model and the design of calibration experiment become very important when the densification of powder involves high shear stresses (e.g., roller compaction of powder) or tensile stresses (defect formation).

The phenomenological continuum models have been successful in predicting density evolution during compaction [1, 2]. Based on that, some attempts have been made to predict crack formation during compaction [23-25]. Although they provide some qualitative information that agrees with experimental observations, their ability to predict strength is still questionable. Practically, all the phenomenological models assume that density is the only internal variable and they predict the same strength for compacts that have the same density. This was proven wrong by the experiments of Koerner [7, 26]. In that work, Koerner produced three specimens of the same density by three different methods, isostatic, die and triaxial compaction. According to the constitutive models discussed above, such specimens should have identical mechanical properties. Koerner’s
experiments showed that the strength of compacts varied by a factor of 2 to 3. The dependence of mechanical properties on the mode of loading is called “loading path dependence” [26-28].

Path dependence and strength anisotropy are topics that have been discussed in the context of soil mechanics for many years. Many soils possess some degree of anisotropy and any additional load causes changes in anisotropy. Early experimental works have used triaxial compression and oedometer consolidation tests to evaluate the strength and deformation behavior of soils as well as the effect of stress path (e.g. [29-33]). Law [34] studied the effect of stress path geometry on the soil brittles. For the particular case of triaxial testing, four typical effective stress paths of shearing have been considered. He found that soil brittleness increases with decreasing effective confining stress or with increasing effective cohesion. For a given soil failing at the same effective confinement, the brittleness decreases with increasing angle between stress path and confining stress axis. Atkinson et al. [35] studied the effects of a change in the direction of the loading path on the stiffness of London Clay. Their results showed that in addition to the overall stress history, the recent stress history - either a sudden change in the direction of the stress path or a period of time at a constant stress state - has a major effect on subsequent stiffness. Bakker et al. [36] showed that the consolidation of agricultural soil varies remarkably under stationary and moving tires. Recently, Garga et al. [37] studied the stress-path dependent behavior of a weathered clay crust. It was observed that the shear strength ratio (vertical versus horizontal) varies from 1.3 to 1.6 for different samples that locate at various depths, for which the in situ stress histories are different.
Other than mechanical measurement such as the triaxial testing, techniques such as the measurement of electrical conductivities of a soil specimen has also been applied to quantify the anisotropic microstructure of soils (e.g. [38-40]). The topics of path dependence and strength anisotropy in soils still remain of interest (e.g. [41-44]).

In the field of powder compaction, path dependence was first recognized importance in seventies [7, 26]. More recently, Galen and Zavaliangos [10] have shown that the mechanical strength of powder compacts produced by closed die compaction is anisotropic. In this work, the cylindrical samples were prepared by means of closed die compaction for both ductile and brittle powders. Diametrical compression tests were conducted to obtain the compact strength in two orientations: perpendicular to and parallel to the direction of compaction. Information regarding strength anisotropy was collected. It was shown that in ductile powders the tensile strength in the prior-compaction direction is lower than in the transverse direction (Figure 1.2). In brittle powders, the opposite behavior was observed. In both cases, strength anisotropy is a function of density and the trends are opposite, with the ductile materials becoming increasingly anisotropic and the brittle material becoming increasing isotropic as density increases. It was also shown that elongated particles, which may exhibit additional morphological anisotropy due to packing preferential, show more anisotropy in strength after compaction than those powders of the same material but in equiaxed form. The extent of the strength anisotropy is also dependent on the compaction path. Isostatically compacted equiaxed powder exhibits no strength anisotropy while acicular powders exhibit less anisotropy when compacted isostatically than when die pressed.
Based on our current understanding, anisotropy and path dependence of green compacts are essentially different manifestations of the directionality of microstructure. Figure 1.3 schematically shows these two concepts: loading path dependence and strength anisotropy. Path dependence reflects the fact that the processing history can affect significantly the resulting microstructure and in turn the properties. In the case of green compacts, it also tells us that the use of relative density alone does not suffice to characterize the microstructure and the corresponding dependence of properties on the processing path. When the strain imposing densification is non-isostatic, the resulting microstructure becomes anisotropic. Anisotropy here indicates that there is an orientational dependence in the microstructure. Path dependence and anisotropy although they appear related are not entirely identical concepts. To clarify this possibility, one can imagine a situation where there may be path dependence without anisotropy. A “rough” example of this maybe the following thought experiment: As shown in Figure 1.4, assume that we have a very loose particle collection that is loaded in two different ways: (A) pure shear loading and followed by isostatic compaction; (B) isostatic compaction only. The shear loading in “A” can rearrange and pack the particles without substantial deformation of the contacts (minor density change). If isostatic compaction causes isotropic deformation of the contacts, of both compacts A and B it is possible that they are both isotropic but with differences in their structure even if their final density is the same. As schematically shown in Figure 1.4, compact A will be the one with a high coordination number and low deformation of the interparticle contact, while compact B will have lower coordination number but higher deformation at the particle contacts. Subsequent
deformation in non isostatic paths is probably going to be different but at this point both compacts are isotropic with a structure that depends on the prior path.

There are few micromechanical models attempt to address loading path dependence and strength anisotropy. The most prominent one is the micromechanical model developed by Fleck [45, 46]. Fleck’s model is a micromechanical model that derives the macroscopic response from local plasticity at the interparticle contacts. The macroscopic strain is related to the particle kinematics via the assumption of affine motion (e.g., isostrain approximation), which renders the macroscopic stress prediction to be an upper bound. A plastic potential \( W \) is defined by summing up the energy dissipation for all contacts. The partial derivative \( \frac{\partial W}{\partial E_{ij}} \) defines the macroscopic stress. The affine motion assumption allows to follow the development of orientation dependence of contacts in a non-isostatic macroscopic strain history. A second order tensor, which evolves with the macroscopic strain rate, is used to represent the orientation dependence of the contact areas.

The presence of an anisotropic microstructure description in Fleck’s model allows it to predict path dependence in powder compaction. Figure 1.5 shows the model prediction of the post compaction yield loci for two microstructures produced by closed die and isostatic compaction respectively. The fact that the predicted yield loci for the same relative density are different is the manifestation of path dependence. Figure 1.5 shows two limiting cases: fully cohesive and non cohesive powders, both of which exhibit path dependence. In either case the predictions are biased by the affine deformation assumption, which exaggerates path dependence [47].
Based on Fleck’s model, one can calculate the strength ratio along loading direction versus transverse direction. As the stress triaxiality\(^1\) moves away from isostatic compression (i.e., become smaller as it moves toward die compaction), the calculated strength ratios\(^2\) are less than one, i.e., the strength in the loading direction is higher than that in the transverse direction. More details about prediction of strength anisotropy from Fleck’s model are given in appendix A. Compared to experiments, Fleck’s model actually predicts the opposite strength anisotropy for ductile material compacted by die compaction. With respect to path dependence, it has been shown that the affine motion is restrictive and artificially amplifies the orientational variation of interparticle contacts \([47, 48]\). As a result, the predicted path dependence is exaggerated. With respect to anisotropy in tension (or low compressive & tensile triaxialities in general) Fleck’s model gives incorrect prediction with the experimental observation for ductile materials. The assumption of affine motion greatly biases the results. For example in die compaction, it predicts a \(\cos^2\theta\) dependence of the area of contact where \(\theta\) is the angle of the contact normal to the axis of the compaction. Therefore, the contacts at 90\(^0\) are assumed not to deform at all. In this way the transverse strength is severely under-predicted and the model predicts opposite behavior than the experiments for the ratio of transverse to normal green compact strength. Another factor that prevents this micromechanical model from explaining the correct strength anisotropy is that it did not take unloading and its effect on strength.

\(^1\) Stress triaxiality is defined as \(\frac{P}{q}\), where, \(P\) the hydrostatic pressure and \(q\) is Mises equivalent stress.

\(^2\) Strength ratio is defined as \(\frac{\sigma_T}{\sigma_N}\), where \(\sigma_N\) is the strength along prior-compaction direction, \(\sigma_T\) is the strength along transverse direction.
anisotropy into account. The potential for damage at the contacts during unloading is neglected.

The discrete element models (DEM) provide another efficient way to study the behavior of powders during compaction. This method is conceptually identical with molecular dynamics (MD), in which the solid spherical particles play the role of atoms. It was introduced by Cundall and Strack [49] and later on was applied on the analysis of shearing of dense packing [50, 51]. DEM method can provide significant information on particle motion, contact forces and several macroscopic quantities in an assembly of particles subjected to external loads or displacements [47, 48, 52-56]. Discrete element models are an improvement over the Fleck’s model. They can estimate the yield locus for compact after compaction [56], and address particle rearrangement in the early stage of compaction [55]. With regards to unloading, Martin [57, 58] introduced the formulation of Mesarovic and Johnson [59] into his discrete element method (DEM) to model the behavior of powder compacts during loading and unloading. Martin’s model was able to predict failure of contacts and provide a more realistic picture of the behavior of powder compacts under complex loading-unloading-reloading sequences. The predicted yield surfaces for both isostatic and close die conditions are much different from the one predicted by previous DEM and micromechanics models.

Unfortunately, typical DEM simulations are limited by the need to keep the interparticle interaction simple, for example, rotation stiffness of the contacts are oversimplified [47, 55, 56]. When the particles undergo larger plastic deformation, the rotation is restricted because the resistance in rotation at the contact depends on the degree
of prior contact deformation. Early experimental [60] and numerical results [61] showed that particle rolling also has a significant effect on the macroscopic response of powders.

Moreover, DEM is only representative of the true deformation behavior occurring at the particle interface at low densities. It is possible that the particle can be extruded into void spaces due to the large plastic deformation at high relative densities. As the result, the coordination number of a single particle may even exceed the maximum coordination number that DEM allows [62]. Another weakness of DEM is that it ignores the effect of neighbors on the relationship between inter-particle force and center-to-center distance as shown in [63]. Therefore, at the high density level, the assumption of distance and contact area relationship in DEM loses its accuracy. DEM need to be limited to low relative density, where such assumptions are expected to be valid. None of the published DEM analysis has addressed the issue of strength anisotropy.

The recently developed Multi-Particle Finite Element Method (MPFEM) relaxes the assumptions in other computational models discussed above [62-65]. The only assumed constitutive behaviors are the material properties of the particle and the interparticle friction interaction. In this model, the individual particles were discretized by using finite elements. This model offers great flexibility in terms of the shape, mechanical behavior of particles and interaction at the contact, and has the ability to have large contact deformation and to simulate compaction to high relative densities. The major disadvantage of this approach is the computational cost. Therefore current analysis is restricted to 2-D problems. Even the previous MPFEM model of Procopio & Zavaliangos [63], which relaxes many assumptions of Fleck’s and DEM models, predicts the axial direction to be
stronger under the assumption of full cohesion between particles after compaction [62].

We believe that a comprehensive understanding of powder compaction and better prediction of strength anisotropy requires the consideration of both unloading and ejection stages. Back in the sixties, Long brought up an idea of radial crushing to emphasize the importance of unloading stage [66]. In that work, the formation of radial cracks was attributed in the elastic unloading of the deformed die, a concept that is not precise but it does point out that the strength of the compact may degrade during the unloading stage. A similar idea was put forward by Kuroki [67] who attempted to correlated subsequent anisotropy during sintering with excessive axial springback during unloading in die compaction. Kuroki suggested that during axial unloading of the compact, some of the contacts that are normal to the compaction direction may open. Another corroborating evidence comes from the work of Lame et al. [68, 69] who employed x-ray tomography to study the evolution of porosity during sintering. In this work, Distaloy AE powder was compacted at 400 MPa to a relative density of 85%. Examination of the porosity after compaction reveals two categories of pores: (a) regular packing pores coming from the packing of the powders that are partially reduced by compaction, and (b) interfacial pores on the interparticle boundaries of the order of 1 micron. Image analysis of the latter revealed a preferential orientation of interfacial pores on interparticle contacts that are perpendicular to the compaction direction, see Fig. 1.6.

Because we believe that interface cohesion is critical in our problem, we review here some basic ideas. Older adhesion models for contacts formed between previously loaded particles consider only elastic loading, e.g., the so-called JKR and DMT approaches [70,
Only recently, Mesarovic and Johnson (referred to be “M & J” in rest of the thesis) examined the separation of two adhering spheres that have been previously deformed plastically [59]. By analyzing the unloading of compacted particles and considering the details of cohesion/adhesion at the contact, they offer two solutions: (a) an adhesive singular solution and (b) a cohesive solution.

The singular solution considers that adhesion exists within a contact of size \( r \leq a < a_0 \) (where \( a_0 \) is the contact area before unloading and \( a \) is the contact area after unloading) and assumes that deformation is predominantly elastic during unloading, and the contact is small compared to the radii of the contacting spheres. The overall force transmission through the contact consists of the contact pressure in the absence of adhesion and the appropriate adhesive traction. If the pressure distribution at the end of compaction is constant, M&J derived an analytical solution for the contact force versus the size of the contact:

\[ P = P_0 f(\chi, \zeta) \]

where \( P_0 \) is the load at the end of compaction,

\[ \zeta = a / a_0 \]

and \( \chi = \frac{\pi}{2\pi - 4} \frac{wE^*}{P_0^2 a_0} \)

in which, \( w \) = adhesion work, and \( E^* \) is the combined modulus for dissimilar spheres:

\[ \frac{1}{E^*} = \left( 1 - \nu_1^2 \right) / E_1 + \left( 1 - \nu_2^2 \right) / E_2 \]

If the spheres are dissimilar, subscripts 1 and 2 indicate each material and \( \nu \) and \( E \) are the Poisson’s ratio and Young’s modulus respectively. The function \( f(\chi, \zeta) \) has an
extreme at \( \zeta = \zeta_{\text{cr}} = a/a_{\text{cr}} \) where \( \zeta_{\text{cr}} \) is the solution of \( \zeta^3 = \frac{9(\pi - 2)(1 - \zeta^2)}{4} \chi \), which defines the pull-off contact size \( a_{\text{cr}} \). For small \( \chi \) the maximum tensile load carried by the contact is given by \( P_C = 1.09\chi P_0 \). If \( \chi \) is a material property only as claimed by Mesarovic and Johnson, then the predicted strength for small contacts is proportional to the macroscopic compaction stress, and this model fails to predict the experimental results of Galen and Zavaliangos [10], because the radial wall stress in die compaction of a cylindrical sample is always smaller than the axial one. If \( \chi \) is not only a material function but also depends on the compaction process on each compact, it is reasonable to assume that the adhesion work should be lower on contacts with normals along the transverse direction than those along the compaction direction.

The cohesive solution of M&J considers a residual contact \( r \leq c < a_0 \) which is divided in two zones: (a) an inner elastic zone \( r < a \) in which an elastic solution for the pressure is obtained and (b) a cohesion zone \( a < r < c \) in which the material is allowed to separate slightly but can carry an adhesion stress \( \sigma_0 \). The important parameters in this case are: \( \zeta = a/a_0, \quad \eta = c/a_0, \quad S = \sigma_0/p_0 \) and \( \chi \). A complex relation between these parameters exist (equation 29 in [59]), which allows the elimination of either \( \eta = c/a_0 \) or \( \chi \). If \( \chi^{2/3}/S^2 \approx 0 \) then the cohesive solution is approximated quite accurately by the singular adhesive solution. Therefore the same picture emerges with respect to anisotropy in of close-die compacts, i.e., opposite to the experimental observations.

The experimental evidence has shown that preferential damage occurs during unloading on contacts that are perpendicular to the compaction direction [69]. As a result,
the strength in the prior-compaction direction should be less than that in transverse
direction. Unfortunately, there is still no successful modeling framework to describe and
further elucidate this phenomenon.

1.3 Goals of this work

We believe that better understanding the role of cohesion in the interparticle contacts is
the key to better predict the post-compaction mechanical behavior. The major goal of this
dissertation is to address the cohesion between particles and its effect on post compaction
strength and anisotropy. The ultimate goal is to develop a versatile mechanics model that
can better describe mechanical behavior of powder, therefore to provide a design tool to
control and optimize the powder compaction operations. The specific goals of this study
are outline below:

- In chapter 2 we identify the capabilities and limitations of phenomenological models
  on predicting such as the porosity distribution, stress and tool force evolution as well
  as the strength and fracture tendency. To do this, we implemented one of the most
  popular phenomenological models – Drucker Prager Cap model into finite element
  program (ABAQUS/EXPLICIT) and developed a robust user subroutine, which
  provides a useful tool to study the powder compaction in macroscopic level.

- In chapter 3, using the developed VUMAT, we analyzed an example of die
  compaction of pharmaceutical powder and subsequent diametrical compression test.

- Semi-cohesive particle interfaces are modeled and studied with respect to the
  loading path dependence and anisotropy in chapter 4. The evolution of contact and
interparticle cohesion during compaction and decohesion during unloading are analyzed using finite element method of a periodic array of disks. Semi-cohesive contact interfaces were first modeled by applying a layer of decohesion surface elements whose material can degrade. The compaction, unloading and ejection were analyzed using a periodic unit cell problem. This part of the work was conducted to understand the role of various material and mesh design parameters in this problem and is the preparation of the multi-particle analysis.

- Chapter 5 contains a study of the loading path dependence and strength anisotropy of powder with a weak interparticle cohesion. MPFEM simulations were developed to include a semi-cohesive interface. This method was used to study the effect of unloading and ejection on the decohesion of contact interface and on the final strength anisotropy.

- Finally, the conclusions of this work and suggestions for future research are presented in chapter 6.
Figure 1.1 Yield surfaces for phenomenological models. (a) Elliptical model, (b) Cam-Clay model, and (c) Drucker Prager/Cap model.
Figure 1.2 Measured strength in both normal and transverse directions for ductile material compacted by die compaction. (a) Low Alloy Steel (b) Acicular Microcrystalline Cellulose [10]
Figure 1.3 Schematic illustration of loading path dependence and strength anisotropy.

**Path dependence**

Specimens produced by different processing paths have different strength along a specific direction

\[ \sigma_1 \neq \sigma_2 \neq \sigma_3 \]

**Strength anisotropy**

Specimens densified anisotropically exhibit anisotropy in strength

\[ \sigma_1 \neq \sigma_2 \]
Figure 1.4 Schematic representations of compaction modes resulting in isotropic specimens but different microstructure. This example shows that it is possible to have path dependent microstructures without anisotropy.
Figure 1.5 Effect of strain path on the evolution of yield surface. The yield surface is given for both isostatic compaction and close die compaction. Contacts are frictionless and two limits cohesion: full cohesion ($\eta = 1$) and cohesionless ($\eta = 0$) are presented. The powder was compacted to a relative density $D = 0.8$ from initial density $D_0 = 0.64$. [46]
Figure 1.6 (a) Pore morphology in compacted distaloy sample by x-ray microtomography. As shown by arrows, the direction of prior compaction is along the vertical direction in the paper plane. (b) Orientations statistics of the interfaces pores located at interparticle contacts observed in (a). Interfaces pores are mostly perpendicular to the compaction direction. [69]
CHAPTER 2: ISOTROPIC CONTINUUM MECHANICS MODEL OF POWDER AND DEVELOPMENT OF VUMAT

2.1 Introduction

The finite element analysis of many compaction problems faces often difficulties due to the strongly non-linear material behavior including friction which makes convergence difficult in implicit finite element schemes. Such problems can be better addressed within the framework of explicit schemes (such as ABAQUS/Explicit) especially when coupled with a remeshing strategy.

The library of ABAQUS contains several constitutive models including a version of Drucker Prager/Cap (DPC) model, which is the most popular model for powder compaction. Unfortunately the version available in ABAQUS is not versatile enough as a number of parameters are considered to be constant (e.g. material cohesion $d$, friction angle of the materials $\theta$). This problem is addressed in ABAQUS/Standard with the used of field variables and user subroutine “USDFLD” which is not available in EXPLICIT. For this reason we developed a VUMAT user subroutine for the most general version of the Drucker Prager/Cap model for ABAQUS/EXPLICIT. In this chapter, we present a summary of the DPC model and its calibration, as well as an integration model for the DPC model and its validation.

2.2 Drucker Prager/Cap model

The Drucker Prager/Cap model is an extended and modified version of the Mohr-Coulomb model [21, 72, 73]. It is a multi-surface elasto-plasticity model permitting
the representation of densification hardening as well as interparticle friction and was first
developed in soil mechanics. It was then adopted and used to simulate the cold die
compaction of tungsten carbide powder [74, 75] and later other metallic and
pharmaceutical powders [1-4, 11, 13, 76].

It consists of a yield criterion, a flow rule and the evolution of the relative density,
which is the only state variable. In the $p-q$ plane ($p$ - hydrostatic pressure, $q$ - Mises equivalent stress), the yield limit is represented by a yield locus $F(q, p, RD)$, which
demarcates the stress states that cause elastic and plastic deformation. $RD^1$ represents
the relative density of the powder compact. If $F(q, p, RD)<0$, the stress causes only
elastic deformation. Otherwise, this stress will cause plastic deformation. In this model,
the yield locus is the function of relative density. Its shape will shrink or expand as the
relative density decreases or increases. Therefore, a complete yield criterion can be
represented as a series of yield loci in $p-q$ space, as shown in Fig. 1.1c.

In general, when plastic deformation occurs, the plastic flow can be obtained by a flow
potential equation:

$$de_{ij}^p = \lambda \frac{d\Phi}{d\sigma_{ij}}$$  \hspace{1cm} 2.1

It describes the direction of plastic flow, i.e. how much strain distributed in each direction.
If $\Phi = F$, then the flow rule is called associated. Otherwise, if $\Phi \neq F$, then the flow rule
is non-associated, for which we need extra equations to define flow rule. The DPC model
consists of two parts: one associated and one non-associated as discussed below.

---

1 Writing in terms of porosity, $RD = 1 - f$ ($f$ : porosity)
Figure 2.1 shows the schematic diagram of DPC model plotted in the $p-q$ plane. The yield locus (blue line) is described using two parts: a cap surface $F_c$ and a shear failure line $F_s$.

The cap yield surface has an elliptical shape. It addresses the plastic deformation of powders under highly confined stress conditions (high hydrostatic pressure):

$$F_c = \sqrt{(p - P_s)^2 + (Rq)^2} - R(d + P_a \tan(\theta)) = 0$$  \hspace{1cm} (2.2)

where $d$, $\theta$, $R$ and $P_a$ are the material parameters that control the shape of the cap, and are functions of relative density.

In the shear region (at low hydrostatic pressure), the yield surface can be represented by a straight line, which is also known as the Mohr-Coulomb shear failure line $F_s$:

$$F_s = q - p \tan(\theta) - d$$  \hspace{1cm} (2.3)

where $\theta$ and $d$ represent the internal angle of friction (or cohesion angle) and the cohesion, respectively.

The flow potential equation in the cap region is associated, i.e., is identical to the cap yield surface equation:

$$\Phi_c = F_c$$  \hspace{1cm} (2.4)

On the shear failure surface the flow is non-associated to $F_s$ and defined by

$$\Phi_s = \sqrt{(\tan(\theta)(p - P_s))^2 + q^2} - \sqrt{\varepsilon(d + P_a \tan(\theta))} = 0$$  \hspace{1cm} (2.5)

where $\varepsilon$ is a scale parameter ($\varepsilon \leq 1$) which is defined below. For $\varepsilon = 1$, $\Phi_s(\varepsilon = 1)$ forms the surface shown in Fig. 2.1, i.e., it passes through the ends of the shear failure surface. For any stress state $(p,q)$ on $F_s$, the value of $\varepsilon$ is adjusted so that the surface $\Phi_s(\varepsilon)$ similar to $\Phi_s(\varepsilon = 1)$ and passes through the stress point $(p,q)$, see Fig. 2.2.
\[ \varepsilon = \frac{q^2}{(P_a \tan(\theta) + d)^2} + \frac{(p - P_a)^2 \tan^2(\theta)}{(P_a \tan(\theta) + d)^2} \]  \hspace{1cm} \text{2.6}

This “peculiar” definition of \( \Phi_s \) is of ad-hoc nature but is fully defined by \( F_s \) and does not introduce new parameters other than those needed for \( F_c \) and \( F_S \). This formulation is therefore advantageous in that it does not increase the experimental calibration requirements. Based on the definition of \( \Phi_S \) and \( \Phi_C \), it is clear that stresses in the cap region cause densification, while stresses in the shear failure region cause volume dilatancy.

The total number of independent parameters in the DPC model is four. Normally, these four independent parameters are calibrated from experiments:

- \( \theta \): friction angle of the materials;
- \( d \): pure shear (cohesion) yield stress;
- \( R \): cap eccentricity parameter (\( 0.0 < R < 1.0 \))
- \( P_h \): hydrostatic compression yield stress

Accordingly, the parameter \( P_a \) can be calculated based on the following equations:

\[ P_a = \frac{P_h - Rd}{1 + R \tan(\theta)} \]  \hspace{1cm} \text{2.7}

The detailed calibration of the parameters of the Drucker Prager/Cap model can be found in [12] for microcrystalline cellulose grade Avicel PH 102, and in [77] for 316L stainless steel powders. Here, we summarize the procedure of calibration for completeness.

The yield surface in the shear failure region is a straight line. It is sufficient to measure the green strength in two types of tests with different degrees of stress triaxiality such as
simple compression test and diametrical compression test. The tension test or the four point bend test would also be appropriate. The former two are the most popular test methods used for measuring the strength of cold compacted samples. With these two sets of data, the parameters $d$ and $\theta$ are then can be determined. An instrumented die is needed during die compaction experiments to provide both stresses in normal axis direction ($\sigma_n$) and radial direction ($\sigma_r$), which in turn define $q$ and $p$. The two unknowns $R$ and $P_b$ require two equations to be solved. $q$ and $p$ for a given RD should satisfy $F_C = 0$ or $F_S = 0$. The second equation comes from the condition of the radial strain potential forms include:

\[ d\varepsilon_{rr} = d\varepsilon_{rr}^{el} + \lambda \left( \frac{\partial \Phi}{\partial \sigma_{rr}} \right) = d\varepsilon_{rr}^{pl} \]

or

\[ d\varepsilon_{rr} = d\varepsilon_{rr}^{el} + \lambda \left( \frac{\partial \Phi}{\partial \sigma_{rr}} \right) = 0 \]

or

\[ d\varepsilon_{rr}^{pl} = \lambda \left( \frac{\partial \Phi}{\partial \sigma_{rr}} \right) = 0 \]

in order of decreasing complexity. The first takes into account the deformation of the die, the second assumes that the die is rigid and the third assumes that the elastic strain is small or negligible compared to the plastic one.

### 2.3 Implementation of DPC model and development of VUMAT

The constitutive model presented in the previous section was implemented in the ABAQUS, a general purpose finite element program [78]. This code provides a general
interface for user programmed constitutive models through a “user subroutine” (VUMAT for ABAQUS/Explicit). As discussed above, we develop our own user subroutine because the version of Drucker Prager/Cap model in ABAQUS/Explicit is not flexible enough.

Figure 2.3 shows schematically the integration procedure in ABAQUS/Explicit with a VUMAT. For each time step, ABAQUS integrates the equations of equilibriums based on the stress state at the beginning of the step at each integration point and provides the deformation gradient for VUMAT subroutine. VUMAT then finishes the integration of the constitutive model and updates the stress and state variable for each integration point. With the information that VUMAT provides, ABAQUS can then continue the calculation for the next time step.

2.3.1 Derivation of the constitutive equations

Elasticity

A basic assumption of elastic-plastic models is that the deformation can be divided into an elastic part and an inelastic (plastic) part. There are mainly two methods of decomposition of kinematics: (a) multiplicative decomposition $F = F^e F^p$, in which it requires that the plastic deformation gradient $F^p$ (9 elements) is stored as a state variable for all integration points. (b) additive decomposition:

$$
\Delta \varepsilon = \Delta \varepsilon^e + \Delta \varepsilon^p
$$

where $\Delta \varepsilon$ is the total strain increment, $\Delta \varepsilon^e$ is the increment of the elastic strain, and $\Delta \varepsilon^p$ is the increment of inelastic strain. The additive decomposition is adequate when the elastic strains are small.
For a linear and isotropic material:

\[ \Delta \sigma = C \Delta e^{pl} = C (\Delta e - \Delta e^{pl}) \text{ with } C \equiv 2GI + (K - 2G/3) \delta \otimes \delta \]

where \( C \) is the forth order elasticity tensor, \( I \) and \( \delta \) are respectively the forth and second order identity tensor, \( G \) and \( K \) are the shear modulus and bulk modulus respectively which are functions of powder porosity. For isotropic materials, \( G = \frac{E}{2(1 + \nu)} \)

and \( K = \frac{E}{3(1 - 2\nu)} \), where \( E \) and \( \nu \) are Young’s modulus and Poisson’s ratio, respectively.

**Plasticity**

The evolution equation for the plastic part of the deformation gradient (“flow rule”) is given by

\[ \Delta e^{pl} = \lambda \frac{\partial \Phi}{\partial \sigma} = \lambda (\frac{\partial \Phi}{\partial q} n - \frac{1}{3} \frac{\partial \Phi}{\partial p}) \]

where, \( n = \frac{3}{2q} S \), \( S \) is the deviatoric part of stress tensor \( \sigma (S = p\delta + \sigma) \), for which, equivalent stress and hydrostatic pressure are defined as: \( q = \sqrt{\frac{3}{2}} S : S \) and \( p = -\frac{1}{3} \sigma : \delta \).

As mentioned before, the DPC model is associated in the cap region (equation 2.4) and non-associated in the shear region (equation 2.5). \( \lambda \) is a scalar that need to be determined. The right side of above equation has two parts, the first part represents the plastic strain associated with distortion (caused by the equivalent stress), while the second part represents the plastic strain associated with volume change. From balance of mass, the evolution for porosity is

\[ \Delta f = (1 - f)tr \Delta e^{pl} \]
Thermal aspects [79, 80] or rate dependent effect [81] can be readily incorporated but this is not done here. The details of time integration procedure are discussed below.

2.3.2 Integration procedure

For a typical time step, our VUMAT uses explicit Euler algorithm (Euler forward) to integrate stresses and internal state variable. The time increment is limited by the overall stability limit of the explicit integration of the equations of motion. This is usually more restrictive than the stability limit of the stress integration in the VUMAT. Other integration algorithms are also can be used, such as implicit Euler algorithm (Euler backward) [82, 83] or semi-implicit Euler algorithm [79]. Since the stability constrain limits the overall time increment, explicit and semi-explicit methods (i.e. not iterative methods) are more efficient than fully implicit which are more appropriate for large time steps and plastic strain increments. The VUMAT uses the stress and internal variables at the beginning of an increment and the strain increment provided by ABAQUS and needs to predict the stress at the end of the increment, as well as the new values of the internal state variables.

The increment of strain across a time step \( \Delta t = t_{n+1} - t_n \) is

\[
\Delta \epsilon_{n+1} = \Delta \epsilon_{n+1}^{el} + \Delta \epsilon_{n+1}^{pl}
\]

from equation 2.10, we have

\[
\Delta \sigma = \sigma_{n+1} - \sigma_n = C(\Delta \epsilon_{n+1} - \Delta \epsilon_{n+1}^{pl})
\]

where, \( \sigma_{n+1} \) is the stress at time step of \( t_{n+1} \) and \( \sigma_n \) is the stress at time step \( t_n \).

From above equation, we have

\[
\sigma_{n+1} = \sigma_{n+1}^{el} - C \Delta \epsilon_{n+1}^{pl}
\]
\( \sigma_{n+1}^* \) is defined as trial stress that calculated based on the total strain increment.

\[
\sigma_{n+1}^* = \sigma_n + C \Delta \varepsilon_{n+1}
\]  

From equation 2.11, the flow rule now becomes:

\[
\Delta \varepsilon_{n+1}^p = \lambda \left( \frac{\partial \Phi}{\partial \sigma_{n+1}} \right) = \lambda \left( \frac{\partial \Phi}{\partial q} n + \frac{1}{3} \frac{\partial \Phi}{\partial p} \delta \right)
\]  

Here, \( n = \frac{3}{2q} S^* \) (\( S^* \) is the deviatoric part of trial stress and \( q^* \) is the equivalent trial stress). By combining equation 2.15, 2.16 and 2.17 we get:

\[
S_{n+1} = S_{n+1}^* - 3G \lambda \frac{\partial \Phi}{\partial q}
\]  

and,

\[
p_{n+1} = p_{n+1}^* - K \lambda \frac{\partial \Phi}{\partial p}
\]  

The equivalent trial stress and equivalent new stress are similarly related:

\[
q_{n+1} = q_{n+1}^* - 3G \lambda \frac{\partial \Phi}{\partial q}
\]  

Equations 2.19 and 2.20 are written according to the specific regime of the flow potential functions. For the shear failure region, we get:

\[
q_{n+1} = q_{n+1}^* - 3G \lambda \frac{q}{\sqrt{\tan(\theta)(p - P_a)^2 + q^2}}
\]  

\[
p_{n+1} = p_{n+1}^* - \lambda K \frac{\tan^2(\theta)(p - P_a)}{\sqrt{\tan(\theta)(p - P_a)^2 + q^2}}
\]  

and for the cap region:

\[
q_{n+1} = q_{n+1}^* - 3G \lambda \frac{R^2 q}{\sqrt{(p - P_a)^2 + R^2 q^2}}
\]  

\[
p_{n+1} = p_{n+1}^* - \lambda K \frac{p - P_a}{\sqrt{(p - P_a)^2 + R^2 q^2}}
\]
We chose to compute \( q_{n+1} \) and \( p_{n+1} \) explicitly. Therefore, the values of \( q \) and \( p \) in the right hand side of above equations are estimated by \( q_n \) and \( p_n \). Also all the model parameters are estimated using the porosity at the beginning of the step. The only unknown in addition to \( q_{n+1} \) and \( p_{n+1} \) is \( \lambda \). Once \( \lambda \) is estimated, the stress in the next time step is determined and therefore the state variable can also be updated. \( \lambda \) is evaluated based on the requirement that the new \( q_{n+1}, p_{n+1} \) and \( f_{n+1} \) (porosity) satisfy the yield condition.

The summary of numerical integration algorithm of Drucker Prager/Cap model is listed as below:

**Step 1: Elastic prediction**

- Assume that the deformation in the step is elastic, we compute an elastic trial stress:
  \[
  \sigma^*_{n+1} = \sigma_n + C\Delta\epsilon_{n+1}
  \]
  using the elasticity tensor \( C \), which is the function of porosity.

**Step 2: Elastic updating**

- If \( \Phi(\sigma^*_{n+1}, f_n) < 0 \) then
  \[
  \sigma_{n+1} = \sigma^*_{n+1} \quad f_{n+1} = f_n
  \]

**Step 3: Plastic correction**

- If \( \Phi(\sigma^*_{n+1}, f_n) > 0 \) then
  - Compute:
    \[
    q_{n+1} = q^*_{n+1} - 3G\lambda \frac{\partial \Phi}{\partial q} \\
    p_{n+1} = p^*_{n+1} - K\lambda \frac{\partial \Phi}{\partial p} \\
    f_{n+1} = f_n + (1-f)\frac{\partial \Phi}{\partial f}
    \]
  as functions of \( \lambda \) and substitute into \( \Phi(q_{n+1}, p_{n+1}, f_{n+1}, \lambda) = 0 \) and solve for \( \lambda \)
analytically.

- Once $\lambda$ is found, compute and update the stresses and state variable $f$.

\textit{Step 4: Return to the main program}

\subsection{2.3.3 Verification of VUMAT subroutine}

The validity of developed VUMAT subroutines was verified by examining the deformation of a unit cell at different loading conditions. As shown in Figure 2.4, six different deformations with different triaxialities were applied on the unit cell: hydrostatic tensile, constraint tensile (the displacement only in x direction), simple tensile, hydrostatic compression, constraint compression and simple compression. The material parameters used in Drucker Prager Cap model are listed in table 2.1. The initial relative density was set as 0.3 (porosity = 0.7). It is extremely difficult to conduct the experimental calibration for the material with low density ($\text{RD} < 0.3$). Therefore, our model assume the parameters to be constant when relative densities is lower than 0.3.

We compared our ABAQUS/Explicit results (with VUMAT subroutines) with ABAQUS/Standard (Implicit) results. Figure 2.5 shows the porosity and stress evolution of the unit cell at hydrostatic tensile conditions. It shows that our ABAQUS/Explicit results have a perfect match to the ABAQUS/Implicit results. The prediction of porosity and stress evolution for other three loading conditions (constraint tension, simple tension and simple compression) also show a perfect match between ABAQUS/Explicit and ABAQUS/Implicit results.

Figure 2.6 shows the porosity and stress evolution for hydrostatic compression
condition. There is a disagreement between our explicit results and ABAQUS/Implicit results. Our VUMAT predicts less densification of the material at the same level of deformation, compared to the prediction obtained by ABAQUS/Standard. However, by using constant Young modulus and Poisson ratio, our VUMAT predictions have a perfect match with ABAQUS/Standard. A full understand of the discrepancy is not possible at this point due to the limit access to the original code and integration algorithm of ABAQUS/Standard. We will be aware of this difference between VUMAT and ABAQUS/Standard as we proceed to the following study.
Table 2.1 Parameters used in Drucker Prager/Cap model for Avicel material

<table>
<thead>
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<th>Poisson ratio $\nu$</th>
<th>$d$ (Pa)</th>
<th>$\theta$ (degree)</th>
<th>R</th>
<th>$P_b$ (Pa)</th>
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<td>1.86E+07</td>
<td>66.5</td>
<td>0.90700</td>
<td>1.28E+08</td>
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Figure 2.1 Schematic representation of Drucker Prager/Cap model.

Figure 2.2 Yield surface and flow potential function for Drucker Prager/Cap model. Schematic representation of flow rules at different stress conditions.
Figure 2.3 ABAQUS and VUMAT subroutine.
Figure 2.4 Different loading conditions for unit cell simulation verification.
Figure 2.5 Unit cell verification. (a) porosity and (b) stress evolution at hydrostatic tensile conditions.
Figure 2.6 Unit cell verification. (a) porosity and (b) stress evolution at hydrostatic compression conditions.
3.1 Introduction

The successful compaction of parts/tablets from powders is judged in many cases by their strength. As discussed in chapter 1, the strength of compacts is path dependent and anisotropic. The evaluation of strength is often performed by “simple” tests, preferably directly on parts or tablets without any machining of special geometries. Such tests are useful as quality control tests as they can detect variation of behavior due to changes in processing or feedstock material. Because the nature of green strength is complex, the interpretation of such tests can become difficult. As an example we consider below the diametrical compression test.

The diametrical compression test (also called Brazilian disc test) was originally designed to determine the tensile or fracture strength of brittle materials [84]. This test is characterized by simple specimen geometry (disk, compact, tablet, etc.) and provides a limit force required to cause fracture. It is commonly used in many technological fields, such as rocks, concrete, ceramics and metal composites, as well as materials used in the pharmaceutical solid dosage forms [85-88]. In its simplest form for a flat face disk and a brittle material the Hertz analysis [89] is adequate to understand the results of the test. This analysis predicts that the maximum principal stress is tensile and develops at the center of the disk along the transverse direction to the applied load (Figure 3.1). This stress is given by
\[ \sigma_f = \frac{2F}{\pi Dt} \]

where \( F \) is applied force, \( D \) is the diameter of the disk and \( t \) is the thickness of the disk. This stress is assumed to cause failure in the sample along the diameter connecting the loading points. While this approach is straightforward and useful for brittle materials, complications develop when the material exhibits some ductility before failure [88].

This test is commonly employed in the pharmaceutical industry (and other industries) on shapes other than flat disks (e.g. curved face tablets, capsules, odd shaped tablets etc.). The argument is made that, even if a Hertz like analysis is not known, the measured failure load can serve as a quality measure of the “overall strength” of the part.

Such situations often lead to unexpected results and behavior that is difficult to predict. For example, Sinka et al. in 2004 tested in diametrical compression two types of curved face tablets which were made by die compaction (Sinka 2004). Two different die-wall lubrication conditions were applied during die compaction. The results showed that the lubrication conditions induced strong gradients in density inside the tablets. Tablets compacted under high friction (unlubricated) have a higher density in the periphery, while tablets compacted under low friction (lubricated) have a higher density in the center. The fracture modes observed during subsequent diametrical compression test also differed dramatically. The unlubricated tablets break perpendicularly to the tablet consolidation surface, as normally is the case of brittle disks subject to point loading (Figure 3.2a). On the other hand, lubricated tablets delaminate across the middle along a plane parallel to the tablet prior-compaction direction, as shown in Figure 3.2b. The mode of failure shown in Figure 3.2b is often taken as an “improper” result and is often discarded when it is
encountered in testing practice.

With such phenomena relatively common in practice, engineers are faced with the task of interpreting them in a consistent manner. Efforts have been made to take the shape of the samples into account by developing experimentally calibrated equivalent equations [85]. Attempts to evaluate the effect of shape have been made using finite element analysis. In all these cases, the properties of the material are considered to be uniform. In the example mentioned above [13], not only the properties are non uniform but they vary systematically in ways that depend on processing conditions.

The purpose of this chapter is to examine whether a combined processing and testing simulation can provide useful insight for cases like the one presented by Sinka et al. [13]. Although there are important theoretical questions that need to be fully resolved (path dependence and strength anisotropy), it is important to identify the capabilities and limitations of existing models in green strength prediction. To this end we attempt a simulation approach that employs the Drucker Prager/Cap model for both the processing step and the testing step. The specific objective of the present research is to analyze the fracture of the non-uniform density compacts subjected to diametrical compression test. While the results presented here are validated using pharmaceutical powders, the principles and the corresponding conclusions are general and applicable to any cold compacted powder.

3.2 Modeling and simulation

The simulations were conducted by using finite element software – ABAQUS/Explicit,
in which a user subroutine of VUMAT was developed and the density dependent DPC model was implemented. The description of the DPC model and implementation was given in Chapter 2.

The simulations presented below were carried out in two steps. Since the die compaction geometry is axisymmetric, the compaction step was performed in 2D. The diametrical compression problem is not axisymmetric due to the diametrically applied load. Moreover, only one meridional plane of symmetry exists as the internal density distribution is not symmetric with respect to the equatorial plane (as seen in Figure 3.4). Therefore, a 3D analysis is needed for the diametrical compression test. For this reason the results of the 2D analysis of die compaction were mapped onto a 3D mesh and the diametrical compression analysis was then performed in 3D.

2-D tablet die compaction

The 2D tablet die compaction results presented here are a summary of the work presented in [13, 76] and it serves as a validation of our VUMAT implementation of the Drucker-Prager/Cap model. The full calibration procedure and the extracted material parameters for Avicel (microcrystalline cellulose) are listed in [12] and are also shown in Chapter 2. An axisymmetric tablet was compacted under two curved face punches. The compact was modeled by 30 × 20 axisymmetrical continuum elements (ABAQUS CAX4R type). The geometry of the model was set up for a 25-mm-diameter tablet compressed with punches with a concave surface with a radius of curvature of 19.82mm. The initial relative density of material was 0.25. The punches and die were implemented as rigid surfaces. The bottom punch was stationary. The initial punch separation was
19mm and the simulation was considered to terminate after the top punch moved down by a specific distance that corresponded to a predetermined average relative density, as listed in table 3.1. The friction interaction between powder and die wall and punches is described using Coulomb’s law of friction. The coefficient of friction was measured using a die instrumented with radial stress sensors [12]. The two friction conditions explored in [12] were also considered in the simulations, i.e., high friction in a die cleaned by acetone before the experiment and a low friction in a die in which a magnesium stearate tablet was compressed to apply lubrication at all tool surfaces. In each case, the coefficient of friction varies with the contact pressure. As shown in Figure 3.3, as the contact pressure increases, the friction coefficient decreases. Therefore, a contact pressure dependent friction coefficient has been used in our simulation, as listed in table 3.2. Compared with ABAQUS/Standard, ABAQUS/Explicit takes about 2-5 more CPU time but is devoid of convergence problems.

3-D tablet diametrical compression

The following diametrical compression simulations were conducted using ABAQUS/Explicit, in which the 3-D tablet geometry was modeled by a 22 × 22 × 24 element mesh using element type of C3D8R (8-node linear brick, reduced integration 3D continuum elements). Due to symmetry, only 1/4 of the geometry was modeled. Note that it is not correct to consider 1/8 of the tablet because there is no symmetry along the height of the tablet due to friction and non-symmetric powder placement at the beginning of compaction. The platen that compresses the tablet on its side was implemented as a flat rigid surface. The interface between platen and tablet was assumed to be frictionless. A
vertical displacement was prescribed on the top punch. The compression behavior of material was modeled using the version of the DPC model described in chapter 2 (table 2.1). Note that the initial relative density distribution was not uniform but was mapped from the 2-D axisymmetric die compaction results. Calculations were stopped when excessive mesh dilation occurs.

During the calculation, a constant velocity boundary condition is applied at the top rigid punch with a value of 1mm/s. Mass scaled density of 1.0E6 g/mm$^3$ is employed in ABAQUS/Explicit calculation to improve computational efficiency. Since the application of mass scaling technique will artificially magnify the effect of inertia effect. It was monitored and the value of mass scaling factor is limited so that the ratio of kinetic to internal energy is less than 1.5% for the steady state conditions.

3.3 Results and discussion

2-D tablet die compaction

In this section, we compare two tablets compacted under two different lubrication conditions. The ABAQUS/Explicit simulation results are in agreement with that of ABAQUS/Standard [13, 76] and validate our VUMAT implementation. The wall friction between material and die/punch has a major effect during compaction and reverses the radial density distribution as shown in Figure 3.4 and discussed extensively in [76]. These results are summarized for completeness below.

Figure 3.4 shows the final porosity distribution in the compacted tablets. Despite the fact that their average relative densities are identical, these two tablets exhibit dramatically
different distributions of internal porosity, which is a direct result of the die wall and punch face lubrication. The unlubricated (high wall friction) tablets exhibit high porosity in the center whereas the material in the periphery has been better densified (with low porosity). The lubricated (low wall friction) tablets exhibit an opposite pattern, with a high porosity region around the periphery whereas the material in the center of the tablet show good densification. In earlier work [13, 76], the relative density predictions were validated with experimental density maps obtained from surface hardness tests carried out on cross sections of the tablets. Later the results were also validated using X-ray microtomography and NMR [90]. The agreement between the experimental RD maps and numerical results gives confidence in the predictive capabilities of the model. Figures 3.4c and 3.4d show the comparison of simulation and experimental results of the porosity distribution along the longitude direction of plane AA’, where it can be seen that numerical predictions and experimental data match very well.

The simulation results of force-displacement behavior shows a small difference during compaction between the two lubrication conditions, see Figure 3.5(b). The unlubricated tablet required a larger load to achieve the same overall density.

Figure 3.5(c) shows the stress paths that two points A and B (at the center and edge of the tablet) are subjected. Several differences are observed – some evident and some more subtle. Clearly point B (edge) was densified more (stresses move away from the origin) under high friction. Overall the triaxiality in the low friction case is higher (i.e., the deformation is more constrained. Independent of the frictional conditions, the material at the edge of the tablet (point B) is always subjected to lower triaxialities (more shearing).
Under high friction condition, the radial motion of the material close to the periphery is restricted. As a result, the material in the periphery densifies more. On the other hand, under lubricated conditions, the tendency of the material sliding on the curved face of the punches forces the material at the periphery to move toward to the center. Even though high shear stress condition was also observed at the edge of the tablet, due to the material flow, the final density at the edge of the tablet is lower than that in the center.

**3-D tablet diametrical compression**

As mentioned before, these two types of tablets showed completely different patterns of fracture during subsequent diametrical compression test (see Fig. 3.2). In order to gain insight to these phenomena, a 3D FEM analysis of the diametrical compression test was performed. The inhomogeneous relative density distribution at the beginning of this test was mapped from the result of 2-D die compaction.

Figure 3.6 shows the force displacement curves for the two types of tablets (unlubricated vs. lubricated) at different levels of the average relative density during diametrical compression. To begin with, it can be seen that, similarly to the experimental results, the simulation force displacement curves exhibit a peak indicating some form of failure. The decrease of tool force corresponds to the dilation of the mesh and decrease of the density of the tablet. As shown in the Figure 3.6, the prediction of our simulation in terms of stiffness shows reasonable agreement with experimental observation. We adjusted the experimental result by applied a compliance factor of 1/50000(mm/N). As shown in the Figure 3.6, comparing to the lubricated tablet, the unlubricated tablets are clearly stiffer in this mode of deformation because of the highly compacted material on the
periphery of the tablets (refer to Figures 3.4). Both simulation and experiment results show that the fracture load increases with relative density (Figure 3.7). The prediction of fracture load in the lubricated tablets is quite reasonable, while the fracture load is under-estimated for unlubricated tablets.

As discussed before, the DPC model used in our simulation is density dependent. As the porosity increases (relative density decreases), the yield surface of DPC model shrinks, indicating less strength of the material. The variations of porosity distribution during diametrical compression for both types of tablets are shown in Figure 3.8. Three stages can be distinguished as: early stage, incipient failure and post failure. In both simulations the same failure mode is observed: the maximum decrease of density starts at the center and propagates along a surface parallel to the prior compaction axis. This is the mode of failure observed for unlubricated tablets (Figure 3.2a). For lubricated tablets the experimentally observed mode of failure is different (Figure 3.2b).

We believe that the main cause of the discrepancy is that in the DPC model, failure is accompanied by an isotropic high dilation (volume increase), in contrast to the brittle failure of the tablets. In brittle failure the material progressive weakness only along the fracture plane.

For brittle materials, the Rankine criterion is often used for failure prediction. The Rankine criterion states that failure occurs when the maximum (normal) principal stress reaches a critical stress level. As discussed in the beginning, for uniform material that undergo diametrical compression, the maximum principal stress \( \sigma_f = \frac{P}{\pi R} \). Correspondingly, the hydrostatic pressure \( p = \frac{2}{3} \sigma_f \) and Mises equivalent stress
In the $p-q$ space, this stress condition is corresponding to a straight line with the slope of $3\sqrt{13}/2$, i.e.

$$q = \frac{3\sqrt{13}}{2} p$$  \hspace{1cm} 3.2

This stress state finally reaches the shear failure locus of DPC model when the failure occurs. The equation of shear failure locus of DPC model in p-q plane can be written as:

$$q = p \tan(\theta) + d$$ \hspace{1cm} 3.3

By combining equations of 3.2 and 3.3, we can calculate the critical maximum principal stress when the failure occurs:

$$\sigma_f = \frac{3d}{3\sqrt{13} - 2 \tan(\theta)}$$ \hspace{1cm} 3.4

In the above equation, the variable of $d$ and $\theta$ are both the functions of relative density. Therefore, the calculated critical maximum principal stress $\sigma_f$ will also be function of density. In our VUMAT subroutine, we introduced a parameter $\gamma$ to evaluate how close is the local maximum principal stress, $SP_3$, to the failure state. $\gamma$ is defined as:

$$\gamma = \frac{SP_3}{\sigma_f}$$ \hspace{1cm} 3.5

$\gamma$ provides a cleaner picture as to where the material is about to fail during the diametrical compression test. Positions with a high value of $\gamma$ are expected to fail first.

Figure 3.9 and 3.10 show the maximum principal stresses for those areas with high values of $\gamma$ ($\gamma > 0.98$) at two time steps (before failure occurs) during diametrical compression test, for both unlubricated and lubricated tablets, respectively. The most interesting difference between Figure 3.9 and 3.10 is that the lubricated tablet is subjected to a significant tensile stress along the direction of prior compaction. The origin of this
difference is in the difference of the density distribution produced by die compaction under different friction. It is possible that such a stress may justify the mode of failure shown in Figure 3.2b, especially if one takes into account strength anisotropy as discussed in Galen and Zavaliangos [10]. In [10], it was demonstrated that other than the initial particle morphology, the consolidation path result in the strength anisotropy and strength anisotropy is a function of density. For ductile powders such as Avicel (MCC), the strength along the transverse direction is higher than the corresponding strength along the prior compaction direction. Such an argument together with the predicted tensile stress in the 1-direction (Figure 3.10) appears to justify the fracture mode of Figure 3.2b.

3.4 Concluding statement

In this chapter, the Drucker Prager/Cap model was implemented into finite element simulation to provide the numerical solution for the tablet die compaction and subsequent diametrical compression test. This model was calibrated from experimental work by using simple test, such as die compaction and uniaxial compression. The die compaction of the pharmaceutical tablets with convex faces was analyzed by applied the density dependent DPC model. The effect of lubrication conditions between powder and tooling was examined. It was shown that modifying the lubrication conditions between powder and die wall results in opposing relative density distribution trends. The predictions of the model in terms of relative density distribution show good agreement with experimental result and previous work. Two lubrication conditions induce different density distributions inside the tablets, which in turns, induce the different fracture modes during subsequent diametrical
compression test.

The 3D diametrical compression simulation results show reasonable prediction of the model in terms of stiffness and fracture load. The largest difference between experiments and simulation is observed in unlubricated tablets. On the other hand, Drucker Prager/Cap model is an isotropic model, whose yield surface only depends on the relative density, so that it can neither capture the dependence of the loading history nor the evolving anisotropy. The overall load displacement in diametrical compression is reasonably predicted but the mode of failure based on the evolution of porosity distribution is not captured by Drucker Prager/Cap. One of the limitations in this study is that the residual stress after close-die compaction has not been taken into account in the subsequent diametrical compression simulation. A Rankine criterion for failure (max principle tensile stress) gives better insight into fracture mode of two types of tablet that made under different lubrication conditions, but still no definite prediction of the fracture mode can be made unless anisotropy is somehow taken into account.
Table 3.1 Displacement boundary conditions of tablets used in diametrical compression simulation. [13]

<table>
<thead>
<tr>
<th>Tablet</th>
<th>Top Punch Displacement (mm)</th>
<th>Final Relative Density</th>
<th>Die Condition</th>
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<tbody>
<tr>
<td>hf1</td>
<td>12.04</td>
<td>0.59</td>
<td>Clean/no lubrication</td>
</tr>
<tr>
<td>hf2</td>
<td>11.65</td>
<td>0.56</td>
<td>Clean/no lubrication</td>
</tr>
<tr>
<td>hf3</td>
<td>10.83</td>
<td>0.505</td>
<td>Clean/no lubrication</td>
</tr>
<tr>
<td>hf4</td>
<td>10.095</td>
<td>0.464</td>
<td>Clean/no lubrication</td>
</tr>
<tr>
<td>hf5</td>
<td>9.046</td>
<td>0.416</td>
<td>Clean/no lubrication</td>
</tr>
<tr>
<td>lf1</td>
<td>12.29</td>
<td>0.612</td>
<td>Lubricated</td>
</tr>
<tr>
<td>lf2</td>
<td>11.64</td>
<td>0.559</td>
<td>Lubricated</td>
</tr>
<tr>
<td>lf3</td>
<td>10.91</td>
<td>0.51</td>
<td>Lubricated</td>
</tr>
<tr>
<td>lf4</td>
<td>10.23</td>
<td>0.471</td>
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</tr>
<tr>
<td>lf5</td>
<td>9.17</td>
<td>0.421</td>
<td>Lubricated</td>
</tr>
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</table>
Table 3.2 Contact pressure dependent friction of coefficient used in simulation. [12]

<table>
<thead>
<tr>
<th>Contact pressure (Pa)</th>
<th>Coefficient of Friction (Unlubricated)</th>
<th>Coefficient of Friction (Lubricated)</th>
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<tr>
<td>1.20E+08</td>
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Figure 3.1 Schematic representation of diametrical compression test. (a) Hertz point loading condition; (b) Stress state in $p - q$ space for each standard test method. [88]
Figure 3.2. The failure mode of pharmaceutical tablets during diametrical compression test. (a) Unlubricated friction condition; (b) Lubricated friction condition. [13]
Figure 3.3 Variation of the coefficient of friction with applied forces. [12]
Figure 3.4 Tablet porosity distributions after die compaction. (a) Porosity contour of tablet compacted under high friction condition (unlubricated die/punch); (b) Porosity contour of tablet compacted under low friction condition (lubricated die/punch); (c) Porosity distribution along center line of the tablet under high friction condition; (d) Porosity distribution along center line of the tablet under low friction condition. (SDV1 = porosity)
Figure 3.5 Stress state evolutions during die compaction. (a) Representative position “A” and “B”, (b) Force displacements curve during die compaction, (c) Stress paths under high friction condition (unlubricated) and low friction condition (lubricated).
Figure 3.6 Force displacement curves during diametrical compression. (a) Unlubricated condition; (b) Lubricated condition.
Figure 3.7 Break forces as a function of relative densities (a) unlubricated tablet (b) lubricated tablet.
Figure 3.8 Porosity contours evolution during diametrical compression test simulation.  
(a) Originally, (b) before and (c) after failure. Elements with RD < 0.64 were eliminated.
Figure 3.9 Maximum principal stresses at selected critical area during diametrical compression for tablets made high under wall condition. The size and direction of the arrows indicate the magnitude and direction of maximum principal stress.
Figure 3.10 Maximum principal stresses at selected critical area during diametrical compression for tablets made under lubricated condition. The area marked A shows a region with high tensile stresses along the direction of prior compaction which is not presented in the high friction case (Fig. 3.9).
CHAPTER 4: DECOHESION OF PARTICLE INTERFACE DURING UNLOADING AND THE EFFECT ON STRENGTH ANISOTROPY

4.1 Introduction

Recognition of structure and property anisotropy in powder compact is crucial for design and optimization of powder compaction process. For example, the diametrical compression test (see discussion in Chapter 3.1) is used in pharmaceutical industry to evaluate the “strength” of tablets. When load applied on flat face disks it measures the radial strength which according to [9, 10] can be different than the axial strength (Fig. 4.1). This particular test is only a quality control test, so its absolute value does not matter. If in a particular problem the axial strength matters (e.g., abrasion), the fact that in many cases axial and radial strength both vary in the same way (i.e. both increase or both decrease in response to certain processing change) renders this test a valuable control test. In powder metallurgy, mechanical strength is often evaluated by flexural testing [91] (Fig. 4.1). In this test, the limiting tensile strength is measured along a direction normal to compaction. If this “number” is used as a quality control variable, similar thinking to diametrical compression test applies as discussed above. If, however, this number is used as a design criterion, then its applicability can be questionable as it is not conservative with respect to strength in other directions. This practically is “covered” by the use of larger than usual safety factors which in turn may be “damaging” in terms of the perceived quality in the mind of designers. In such case, better knowledge of anisotropy is a requirement for more rational design.

Although the existence of strength anisotropy in cold-compactd powders has been
recognized, the understanding of its cause is very limited. As discussed in Chapter 1, there are some indications that damage that forms during unloading may play a crucial role in determining the strength of the green compact. For example, German in [92] mentions that ejection from a die under pressure may eliminate the formation of cracks. Unloading also plays an important role in the final part dimensions. Die wall friction and density gradients can lead to cracking and lamination [8, 93]. Therefore, understanding the unloading and ejection of the green compact is as crucial as the loading step to the design and optimization of powder compacts.

Despite the indications for the origin of anisotropy, there is neither a clear understanding nor a model that can shed light to this phenomenon. Our goal in this chapter is to explore a numerical 2D approach to this problem. Specifically the problem of compaction, unloading, ejection and post-compaction strength will be examined using a 2D FEM analysis of a periodic arrangement of particles. The particles are connected with a thin layer of a material which can degrade (primarily) under tension. This degrading material is used as a means to simulate an imperfect interface. The 2D periodic results will allow us to understand the implications of this model in a framework which is less expensive computationally than the MPFEM model which will be presented in Chapter 5.

4.2 Constitutive equation for decohesion element

In the simulation, we introduce a layer of decohesion elements on the particle surface to model the weak bonding of contact interface. The concept of decohesion element is not new. Hillerborg et al. [94] first applied it to analyze the crack development in concrete. It
has also been used by Needleman to describe the weak interfacial strengths in composite laminates by using a traction-separation law [95-97]. Decohesion elements have also been used in the study of delamination of composites [98-101].

The selection of the constitutive equation is crucial for the description of the evolution of interfacial decohesion element from initial debonding to subsequent separation. There are two types of models: (a) traction-separation and (b) continuum-based models. In traction-separation based model, the constitutive equations for the interface are basically phenomenological mechanical relations between the traction and interfacial separation. With increasing interfacial separation, the traction across the interface reaches a maximum, decreases and vanishes when complete decohesion occurs. There are several traction-based constitutive equations such as linear elastic-perfectly plastic, linear elastic-linear softening, linear elastic-progressive softening and linear elastic-regressive softening [102]. A comparison of these constitutive equations is shown in Figure 4.2(a).

Continuum-based models describe the degradation behavior of an element, in which the progressive damage and failure of materials is governed by the specific constitutive equations of the model. Figure 4.2(b) shows a typical stress-strain response of degradation material that can be used for interface failure.

Traction-separation models are typically used in situations where a predefined interface exists and is infinitesimally thin. Continuum-based models are naturally used when a bonding layer with a specific thickness, such as when an adhesive exists. While interparticle interfaces do not contain a “bonding” layer, they do possess a length scale, dictated by their surface roughness, and the subsequent plastic deformation. By itself this
length scale does not justify the use of a continuum based model as this length scale is not easily determined. We selected to use a continuum based model primarily for the ease of implementation in ABAQUS/Explicit especially because of the three stages of the problem (compaction, unloading and ejection), which require smooth transitions.

ABAQUS/Explicit offers a general framework for material failure modeling [78]. As shown in Figure 4.3, the model used here includes an undamaged response, a damage initiation criterion and a damage evolution response. The undamaged material is modeled as elastic perfect plastic material model. The properties of the undamaged material are identical to the bulk of the particle. In this way the compaction stage is easily simulated and transitions from undamaged to damaged condition are “smooth”.

**Damage initiation**

Damage begins at the point marked $D = 0$ on Figure 4.3 and initiation is based on an equivalent strain criterion as will be discussed in detail in the following section. The criterion for damage initiation is met when the following condition is satisfied:

$$\omega_D = \int \frac{d\bar{\varepsilon}^{pl}}{\bar{\varepsilon}_0^{pl}} = 1$$  \hspace{1cm} (4.1)

where $\omega_D$ is a state variable that increases monotonically with plastic deformation; $\bar{\varepsilon}_0^{pl}$ is the equivalent plastic strain, which typically is a function of stress triaxiality for ductile materials.

A ductile damage initiation criterion was used [103-105], in which the specific level of the damage initiation criterion is assumed to have a dependence on stress triaxiality $(\bar{\varepsilon}_0^{pl}(\eta))$. Figure 4.4 shows the damage initiation strain as the function of stress triaxiality.
A low level of damage initiation strain is used in tensile conditions and a much higher one in compressive triaxialities. Essentially, the interface does not fail at compressive stress condition but will degrade at tensile or shear stress conditions. There is no doubt that the selection of the damage initiation criterion is ad hoc. We are not aware of any specific reference in the literature that addresses such a criterion explicitly for compacted powders. Our selection was guided by intuition that whether the damage is related to crack like oxide residues or micro-pores due to surface roughness of the powders, damage initiation should have a triaxiality dependence with the high tensile triaxialities more effective in initiating damage.

**Damage evolution**

After damage initiation, the behavior of the materials is controlled by damage evolution law. In this model, the damage manifests itself in two forms: softening of the yield stress and degradation of the elasticity (stiffness) [78]. ABAQUS/Explicit assumes that the degradation of the stiffness associated with each active failure mechanism can be modeled using a scalar damage variable, D [78]. As shown in Figure 4.3, beyond the damage initiation point, the solid curve represents the damaged stress-stain response, while the dashed curve is the response in the absence of damage. At any given time during the analysis, the stress tensor in the material is given by the scalar damage equation

\[
\sigma = (1 - D) \bar{\sigma}
\]  

where \( D \) is the overall damage variable and \( \bar{\sigma} \) is the stresses that would exist in the material in the absence of damage.
The stiffness is given by:

\[ E = (1 - D)E_0 \]  

where, \( E_0 \) is the stiffness for undamaged material.

The material loses its load-carrying capacity when \( D = 1 \). In ABAQUS, mesh-independent measures - either plastic displacement or damage dissipation energy can be used to drive the evolution of damage after damage ignition. Based on the Hillerborg fracture energy approach [94], a material parameter, \( G \), the energy required to open a unit area of crack, can be used to control the softening response of the material after damage initiation. This approach in ABAQUS/Explicit input file is employed by using the following command:

\*DAMAGE EVOLUTION, TYPE=ENERGY, SOFTENING=EXPONENTIAL

The level of the damage dissipation energy \( G \) (units: energy per unit length) needs to be specified. It can be normalized with a quantity such as the product of the yield strength by the particle radius: \( G/(\sigma_y R) \). Small values of \( G \) were selected in our simulation to model the weak cohesion of contact interface. The area under the stress-strain curve after damage initiation corresponds to the fracture energy.

In summary, the properties required to define the progressive damage of interfacial element behavior are the initial Young’s modulus and Poisson’s ratio, the yield strength which is identical to the bulk material, the equivalent plastic strain at the onset of damage \( \varepsilon_{pl}^d(\eta) \) (function of stress triaxiality) and the damage dissipation energy \( G \) or \( G/(\sigma_y R) \).
4.3 Compaction of particle with semi-cohesive interface

4.3.1 Unit cell finite element mesh design

We chose to analyze the unloading of prior-compacted particles using finite element analysis (FEA). FEA is capable of analyzing (a) semi-cohesive interfaces, (b) interface contact mechanics with no geometrical compromises or ad-hoc simplifications that are common in DEM, (c) elastoplastic or more complex constitutive behavior for the particles. Detailed modeling of the interface requires a very fine mesh, as discussed below. For this reason, we restrict the analysis to a 2D unit cell geometry with periodic boundary conditions, see Figure 4.5.

Due to the symmetry, only 1/4 of the particle was analyzed. The mesh design for one quarter of the particle is also shown in Figure 4.5. In this case, the number of outer layer of elements is 120. Much finer mesh designs have also been used to capture the details of the interface cohesion during unloading. To model the semi-cohesive interface, the outer layer elements (also called the degradation zone) of the particle are modeled by using degrading elements. The material in the degradation zone is modeled with the continuum-based constitutive equations as discussed in the previous section. The material for the main body was modeled to be linear elastic-perfectly plastic. The selection of $E/\sigma_y$ was identical to that used in previous MPFEM studied by Procopio [62] and is chosen to be 100 and Poisson’s ratio ($\nu$) is set as 0.3. The mass scaled density of the bulk material is set as 0.0001. We use a single value of equivalent strain at damage initiation, as shown in Table 4.1 [103]. Instead, we vary parametrically the damage evolution parameter
To simulate different levels of cohesion of the interface, we use $G/(\sigma_y R)$.

### 4.3.2 Convergence study

A convergence study was conducted to verify the sensitivity of the cohesion/decohesion behavior of the interface on the mesh size. In the simulation, a particle is compacted until the contact area reaches 0.3 of the radius and is then subjected to simple tensile load along the same direction. Two types of convergence studies were carried out. In the first case, the thickness of the degrading elements was kept constant (the thickness of the surface element is $1/30$ of the particle radius) and the number of elements along the hoop direction was changed from 120 to 360 and then to 1080 (coarse, medium, and fine). In the second case, the number of elements in the hoop direction was kept to 1080 and the thickness of the surface element was varied from $1/30$, $1/15$ to $1/10$ of the particle radius. The use of a continuum degradation layer may introduce mesh sensitivity, which needs to be evaluated.

The comparison was made in terms of two results. First, we consider the force displacement curve during post compaction and then the predicted strength anisotropy.

Figure 4.6(a) shows the force displacement curves for the first case study. The fine mesh does a reasonable job to capture the quasi-ductile decohesion behavior of the interface. The coarse and medium meshes show a brittle behavior (as can be seen in the figure there is a quick drop of the force after it reaches the maximum), which may imply the mesh density at the surface may not be adequate. In other words, there are not enough elements at the tip of the neck to resolve the gradual propagation of the crack. The force displacement curves for the second convergence study are shown in Figure 4.6(b). All
three simulations predict similar results for the maximum force but they vary in term of interface “ductility”. The mesh dependence of the results originates from the use of a continuum model of the weak interface. Therefore, not only the results are different but there is no convergence at the limit of zero thickness. The critical question is whether the thickness of the surface element affects the end result. The end result in our case is not the ductility of the interface per se but the strength and more importantly the strength anisotropy. To explore whether mesh dependence has a direct effect on the predicted strength anisotropy, two levels of surface element thickness were compared: 1/30 and 1/15 of particle radius for particles compacted at different densities. The details of the anisotropy calculations will be discussed later in this chapter. The result was shown in Figure 4.6(c) and it shows that the thickness of the surface element has very little effect on the predicted strength anisotropy.

4.3.3 Simulation procedure

The process of close die compaction followed by unloading and ejection is simulated on the fine mesh discretization of the unit cell selected from the convergence study. A non-separation boundary condition is applied on the contact between particle and rigid surfaces during unloading and ejection. Therefore, under tensile or shear stress conditions, the surface degrades. As shown in Figure 4.7, a complete simulation procedure includes four steps as listed in the following.

- Close-die compaction: A displacement is applied on the top rigid surface, while the horizontal deformation is restricted by a vertical rigid surface.
In-die unloading: the top rigid surface is moved upwards until the resultant reaction force at the top surface reaches zero, while the side rigid die remains its position.

Ejection: removal of side rigid die horizontally until the reaction forces at the interface become zero.

Reloading: it is conducted by applying a displacement on either vertical or horizontal direction. In this case, the other rigid surface is free to move. This effectively simulates tension.

During close-die compaction, a constant velocity boundary condition with the value of 0.2 is applied at the top punch. At this stage, decohesion is not activated. The surface and main body elements have the same material properties. Simulation stops when the designed density is achieved. The calculation step time is about 1.0.

The in-die unloading simulation model is rebuilt from the close-die compaction simulation. The information such as node coordinates, velocities at each node and the stress conditions at each integration point are collected from the previous loading calculation. Interface decohesion is activated in this stage and hereafter. By adjusting the materials properties of the degradation zone and the contact surfaces boundary conditions, our model are able to model three different cohesion scenarios: 

- no cohesion (cohesionless), which was achieved by allowing particles to separate at the interface;
- full cohesion, which was accomplished by applying non-separation boundary and non-degrading surface material;
- partial cohesion, which was achieved by applying non-separation boundary conditions and decohesion elements at the particle surface as discussed before. A velocity boundary condition is applied at the top punch with the value of 0.1, while the side rigid
die is fixed. Total time of simulation depends on the reaction force evolution at the top punch. A typical step time is about 0.2.

The calculation will restart after the reaction force at the top punch reaches zero during the previous unloading stage. A new boundary condition is applied on the side rigid die to simulate the ejection step. During this step, the top punch is free to move. To avoid the quick change of kinetic energy, an acceleration boundary condition with the value of 0.05 is used. Again, total step time depends on the reaction force evolution at the side rigid die. The step time is 0.6.

During reloading, a constant velocity boundary condition (0.05) is applied at corresponding rigid surface while the transverse rigid surface is free to move.

4.4 Results and discussion

4.4.1 Close-die compaction

The simulation conditions during die compaction were described in the previous section (Section 4.3.3). The particle will go beyond elastic deformation and will experience large plastic deformation during compaction. Since the horizontal expansion was restricted by the side rigid die, the contact area and reaction force in the transverse direction also increase as the deformation continues. Figure 4.8(a) and 4.8(b) show respectively the evolution of contact area and compaction stress at both contact interfaces as the top punch moves down during die compaction. Since the wall stress is always smaller than the compaction stress, it is expected that the particle contact area should exhibit an orientational dependence. Figure 4.8(c) plot the reaction stress versus the
corresponding contact area at both interfaces. Two lines are very close to each other but reach different levels.

Figure 4.9(a) shows the compressive stress distribution at top contacts at different stages during compaction. Positive value indicates that the stress is compressive. Due to the large plastic deformation at each contact interface, the pressure becomes quickly nearly uniform during compaction. This nearly uniform stress distribution is observed when the contact area is less than 50% of particle radius. Similar behavior has been observed experimentally by Johnson on compressed spheres of hard drawn copper [106], and in other FEM simulations [59]. Obviously this is quite different than the parabolic pressure distribution described in Hertz’s equation for elastic contacts. As compaction continues, the contact areas increase further and the pressure distributions are no longer uniform. The stress distribution on the side contact at different stages of compaction is also shown in Figure 4.9(b).

4.4.2 Evolution of stress distribution at interface during unloading

In cold compacted powders, the cohesion/bonding at the contact interface is weak. Even a low tensile stress may cause the decohesion and separation of the interface. Therefore, it is important to know the exact stress condition at the contact interface and the effect of unloading procedure. In this section the detailed stress evolutions at the interface during unloading are analyzed. Three different cohesion scenaria are compared: cohesionless, full cohesion and partial cohesion. In order to make the comparison with analytical solutions [59, 89, 107], the unit cell model created in this case is axisymmetric.
The deformation of the particle along radial direction is not restricted. The particle is initially compacted up to the top contact radius reaches 31% of particle radius (at this point, the pressure distribution at the contact interface is quite uniform). The exact stress distribution on interface is strongly dependent on the cohesion model. A detailed discussion is presented next.

**Cohesionless**

Figure 4.10(a) shows the simulated pressure distributions at different stage during unloading for the contact in the absence of adhesion. As can be seen from this figure, as the load at the contact decreases from $P_0$ during unloading, the residual compressive pressure ($p/p_0$) at the contact also decreases. At the same time, the residual contact area ($r/a_o$) decreases too. For comparison, the residual compressive stress distributions calculated based on Hertz’s equation [89] and the singular solution in M & J model [59] are also shown in Figure 4.10(b). Our simulation results have good agreement with the adhesive singular solution in M & J model. Both results show that there is a transition of residual pressure from near uniform pressure to near quadratic (Hertz) distribution. A plot of unloading force as the function of residual contact area (top interface) is also shown in figure 4.10(c). Our FEM result shows a good match with singular solution in M & J model.

**Full cohesion**

To simulate full cohesion, a non-separation contact boundary condition is applied during the unloading stage. Figure 4.11(a) shows the computed stress distribution at different stage during unloading. As can be seen from the figure, as the load is removed
during unloading, the overall residual compressive pressure \((p/p_0)\) at the contact decreases. Perfect cohesion results in a tensile stress at the edge of contact, even though the overall stress at the contact interface is compressive. Even when the load is fully released, the interface is not stress free in the full cohesion scenario. A residual stress field exists on the interface with compressive pressure in the center of the neck and tensile stress close to the periphery of the neck. Such stress indicates the possibility of damage at the neck tips. This type of stress distribution can not be captured by either singular solution [107] or cohesive solution developed by Mesarovic and Johnson [59]. Compared to the cohesive solution of M & J, we observe that in the numerical solution, there is a tensile stress concentration at the contact tip [108]. On the other hand, M & J cohesive solution models the adhesive tension acting in the annulus of the neck to be constant with a value of \(\sigma_0\), as shown in Figure 4.11(b). The value of \(\sigma_0\) in the M & J model is a material property and is related to the parameter \(S = \sigma_0 / p_0\). Detailed discussion can be founded in [59]. Therefore, the cohesive M & J solution is not a good approximation in the fully cohesive case.

**Partial cohesion**

In reality, the mechanism of development of cohesion between two surfaces is complicated. Cohesion at the interparticle interfaces after cold compaction is often weak. Secondary (weak) bonding is often present, while primary bonding is difficult to achieve due to the roughness of the interface and the presence of surface impurities, oxidation etc.

The application of decohesion elements allows us to model weak bonded interfaces. As has been shown previously in the case of full cohesion interface, there is a tensile stress
at the edge of contact interface during unloading. If the cohesion between two surfaces is not strong enough, it is possible that the contact tip opens due to this tensile stress. This damage can be captured by the degrading elements on the surface. As can be seen in Figure 4.12(a), the stress at the edge of interface becomes zero which indicates that decohesion elements fail and the material loses the load carrying capacity in those areas. As a result, the interface opens slightly at the contact tip. The remaining contact interface has a similar stress distribution: a tensile stress concentration is observed at the edge of the intact part of the interface. A similar decohesion behavior has also been modeled by using cohesive solution developed by Mesarovic and Johnson (Figure 4.12(b)). Again, the M & J solution limits maximum tensile stress to $\sigma_0$ while the numerical gives a strong stress concentration.

### 4.4.3 Stress and geometry of contacts during unloading

Figure 4.13(a) shows the evolution of average normalized stress $F/R\sigma_y$ on each face of the unit cell during unloading (vertical) and “ejection” (horizontal unloading) from a compacted density of 97.9% for all cohesion values examined. For the fully cohesive interface case, the reduction of the vertical load (“punch”) is practically linear. At the other extreme, the non-cohesive interface shows a large nonlinearity with an elastic springback that is 33% larger than that of the full cohesive case. The semi-cohesive interface shows an intermediate nonlinearity. Therefore these models attribute the nonlinearity in unloading, which is commonly observed experimentally [62] to the opening of previously closed interfaces.
At the end of unloading, significant side load remains (on the “die wall”). The magnitude of this residual “wall” pressure does not depend on the details of the interface behavior. During further unloading (“ejection”), nonlinearities develop but the overall magnitude of expansion in the prior compaction direction is about three times the side expansion. Figure 4.13(b) shows the change of overall dimensions during “ejection”. The slope of the curves is proportional to the effective Poisson ratio \( \nu \). It appears that the cohesion model does not affect \( \nu \) which at this level is about the same with the Poisson ratio of the particle.

We turn our attention now to the difference between the two contacts that lead to the final strength anisotropy. Figure 4.14 shows the extent of interface failure after complete unloading for the semi-cohesive case examined. The extent of damage is significantly larger on the contact along the prior compaction direction.

The size of the contact and the damage that develops during unloading determine the final properties of the compact. To a first approximation, the ratio of contact area \( A \) over the size of the unit cell on the same side: \( A_y/L_y \) and \( A_x/L_x \) determine the corresponding strength in that direction. Figure 4.15(a) schematically shows the way how we normalize the contact area. Assuming that the strength developed at the contact is proportional to the contact area ratio, \( R_x \) and \( R_y \) values are roughly proportional to strength in both vertical and horizontal directions. Figure 4.15(b) and (c) show the variation of \( R_x \) and \( R_y \) with relative density. We see a sizeable reduction of \( R_x \) in Figure 4.15(b), while \( R_y \) remains the same after unloading. It tells us that the top interface suffers more damage than the side interface during unloading. At the highest
density, we even see that $R_y$ is greater than $R_x$ for the contact interface with a weak cohesion ($G/(\sigma, R) = 0.0006$). Therefore, the unloading stage has an important effect on the final strength and strength anisotropy.

### 4.4.4 Strength and anisotropy

Reloading was achieved by simulating a tensile test on the compact after unloading. Figure 4.16(a) shows a typical force-displacement curve of the unit cell during reloading. A sharp drop is observed right after the force reaches the maximum. While the intact interface fails “gracefully” in tension (see Fig. 4.6 and relevant discussion). The contacts after unloading are partially detached and a crack essentially forms at the corner of the pore. The presence of this crack leads to an abrupt reduction of load when this crack propagates.

Figure 4.16(b) shows the calculated strengths at different levels of density. In this case, a weak interfacial cohesion was chosen with the damage dissipation energy of $G/(\sigma, R) = 0.0006$. The interface degradation during unloading has a major effect on the achieved strength. At relative low densities, the unloading stage does not cause severe damage of the interface. At those low densities, the strength along the prior compaction direction is higher than the transverse strength. As the relative density increases, the contact, which is normal to the compaction direction, begins to degrade significantly during unloading. As a result, a remarkable reduction of strength occurs. The transverse strength continues to increase. Therefore, the trend of anisotropy reverses and becomes similar to the experimentally observed.
Table 4.2 shows the calculated strength and strength anisotropy at different level of cohesion of the contact interface for the particle compacted to density of 97.9%. The level of cohesion has a major effect on the damage of the interface during unloading and ejection. Low cohesion interface results in large damage of interface and more severe damage is observed in the direction of prior-compaction direction.

The strength ratio (transverse strength vs. normal strength) versus relative density with different level of interfacial cohesion is shown in Figure 4.17. For full cohesion case, the strength ratio increases from about 0.80 at the density of 87% to about 1 when the particle was compacted to nearly fully dense. This is consistent with the fact that the fully cohesive case regresses to the isotropic matrix/particle properties as relative density approaches to 1. In the case of weak cohesion, for the low densities, the behavior is similar to the fully cohesive one. As the unit cell is compacted further, the contact normal to the prior compaction becomes significantly damaged during unloading and the ratio of strength along the transverse to prior compaction direction increases following the experimental result.
4.5 Concluding statement

In this chapter, the unit cell loading, unloading and reloading was studied with emphasis on the role of interparticle cohesion on post-compaction mechanical properties. The introduction of decohesion elements allows us to simulate the partial cohesion interface. The process of die compaction followed by unloading and ejection was studied on a periodic unit cell problem. During unloading, a tensile stress concentration was observed at the tip of the contact interface. This stress will cause damage of the interface if the cohesion between two surfaces is not strong enough. At the end of first step unloading (in-die unloading), a nonlinear force displacement behavior was observed for the contact interface with partial cohesion or cohesionless. This is consistent with the previous MPFEM prediction and experimental observations. Particles with semi-cohesive interface will have certain extent of damage at the contact during unloading, depending on achieved density during compaction. When the particle is compacted to high density, a severe damage of interface is observed in the prior-compaction direction during unloading. As a result, we see a lower strength in the prior-compaction direction than transverse direction. Our model is the first to predict the right trend of strength anisotropy for weak bonded cold compacted powder and gives a reasonable explanation for the experimental observation. This explains the experimental observation of the lower strength in the prior-compaction direction during die compaction for material of microcrystalline cellulose [10]. X-ray microtomography observation by Lame et al. [69] also prove that there is “damage” in contacts normal to compaction direction after unloading.

However, the unit cell problem has limitations. For example, it does not take into
account particle rearrangement which is a common phenomenon at the initial stage of compaction. The periodic arrangement of the particle can not represent well the real configuration of the particles. It may also exaggerate the discrepancy of contact area evolution in two directions. In the next chapter, we will conduct a similar studied but on multiple particles.
Table 4.1 Damage initiation strain ($\bar{\varepsilon}_0^{pl}$) used in simulation.

<table>
<thead>
<tr>
<th>Triaxiality $\eta = p/q$</th>
<th>$\bar{\varepsilon}_0^{pl}$</th>
<th>Triaxiality $\eta = p/q$</th>
<th>$\bar{\varepsilon}_0^{pl}$</th>
<th>Triaxiality $\eta = p/q$</th>
<th>$\bar{\varepsilon}_0^{pl}$</th>
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<tbody>
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<td>0.002632</td>
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Table 4.2 Strength and anisotropy for the particle that compacted by close-die compaction to density of 97.9% with different cohesion level.

<table>
<thead>
<tr>
<th>Damage dissipation energy ( G/(\sigma_i R) )</th>
<th>Residual top contact area</th>
<th>Residual side contact area</th>
<th>Strength in prior-compaction direction (SN)</th>
<th>Strength in transverse direction (ST)</th>
<th>Strength ratio (ST/SN)</th>
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<tr>
<td>Full cohesion</td>
<td>0.9999</td>
<td>0.9988</td>
<td>0.8382</td>
<td>0.8863</td>
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<td>1.9433</td>
</tr>
</tbody>
</table>
Figure 4.1 Schematic representation of (a) diametrical compression test, (b) four point bending.
Figure 4.2 Constitutive softening equations for decohesion materials. (a) stress displacement relationship for traction-separation based models, [102] (b) stress-strain relationship for continuum based model. [78]
Figure 4.3 Stress-Strain curves with progress damage degradation used in the FEM simulation.
Figure 4.4 Damage initiation strain ($\varepsilon_{0}^{p/l}$) as the function of triaxiality. $p$ represents hydrostatic pressure and $q$ indicates Mises equivalent stress.
Figure 4.5 Unit cell mesh design. (a) Periodic arrangement configuration of particles; (b) 1/4 unit cell particle setup and (c) unit cell mesh design.
Figure 4.6 Stress strain curves for element size convergence study (a) variance number of surface elements, (b) variance thickness. (c) Predicted strength ratio (transverse strength versus compaction direction strength) with two different surface element thicknesses.
Figure 4.7 Simulation procedure: (a) initial position, (b) end of compaction, (c) unloading vertically; (d) unloading horizontally (ejection) (e) reloading along prior-compaction direction, (f) reloading along transverse direction.
Figure 4.8 (a) Contact area evolution during close-die compaction; (b) Compaction stress \((F/R/\sigma_y)\) versus load strain during compaction. (c) Compaction stress versus corresponding contact area \(a\). \(F\) represents the reaction force at contact, \(a\) represents the actual contact area, \(R\) is the particle radius and \(\sigma_y\) is the yield strength of the material.
Figure 4.9 Contact pressure distributions during close-die compaction (a) top interface, (b) side interface. Contact pressure is normalized by yield strength of the material. $p$ represent the actual pressure at the contact interface, $X$ and $Y$ represent the actual coordinates along x and y direction, respectively. $R$ is the particle radius and $\sigma_y$ is the yield strength of the material.
Figure 4.10 (Cont’d)
Figure 4.10 Pressure distribution at the top contact for cohesionless contact interface as the load is progressively removed during unloading.  (a) FEM prediction, (b) Hertz and M & J cohesive model prediction.  $p$ represents the actual pressure at the contact, $p_0$ represents the average pressure at the contact before unloading, $r$ is the actual contact radius and $a_0$ is the contact radius before unloading and (c) Unloading force as the function of residual contact area.
Figure 4.11 Pressure distributions for full cohesion contact interface as the load is progressively removed during unloading. (a) FEM prediction, (b) M & J cohesive model prediction. \( p \) represents the actual pressure at the contact, \( p_0 \) represents the average pressure at the contact before unloading, \( r \) is the actual contact radius and \( a_0 \) is the contact radius before unloading.
Figure 4.12 Pressure distributions for partial cohesion contact interface as the load is progressively removed during unloading. (a) FEM prediction, (b) M & J cohesive model prediction. \( p \) represents the actual pressure at the contact, \( p_0 \) represents the average pressure at the contact before unloading, \( r \) is the actual contact radius and \( a_0 \) is the contact radius before unloading.
Figure 4.13 (a) Normalized stress at both top and side contacts as the loads are progressively removed during unloading. $F$ represents the reaction force at contact, $\delta_y$ represents the actual displacement along y axis during the first step unloading (vertical). $\delta_x$ represents the actual displacement along x axis during the second step unloading (horizontal). $R$ is the particle radius and $\sigma_y$ is the yield strength of the material. (b) The displacement along two direction during second step unloading (horizontal).
Figure 4.14 Geometry of contact interface after unloading and ejection. (failed elements has been removed).
$A_x$: contact area at top interface
$A_y$: contact area at side interface
$L_x$: particle size along x direction (width)
$L_y$: particle size along y direction (height)

Top contact: $R_x = \frac{A_x}{L_x}$
Side contact: $R_y = \frac{A_y}{L_y}$

Figure 4.15: Ratio of contact area as the function of relative density.
Figure 4.16 (a) Typical force displacement curve during tensile reloading, (b) Calculated strength in prior-compaction direction and transverse direction.
Figure 4.17 Calculated strength ratio (ST/SN) for the particles that have been compacted to different densities.
CHAPTER 5: MPFEM ANALYSIS OF POWDER COMPACTION AND UNLOADING

5.1 Introduction

The unit cell study in Chapter 4 has given the first reasonable explanation of the anisotropy result of Galen and Zavaliangos [10]. The main idea in this study was that the introduction of a weak interface leads to the formation of cracks on the contacts normal to the prior compaction direction which in turn makes the normal strength lower than the transverse strength. Despite the positive results of the unit cell, the particular periodic geometry is not a good representation of the true compact. For this reason, in this chapter we perform an analysis of compaction, unloading, ejection and reloading within the framework of the Multi-Particle Finite Element Model (MPFEM approach was reviewed in Chapter 1). The MPFEM allows for a much more realistic geometry than the unit cell despite the 2D nature of the simulation.

5.2 Multi-particle finite element model

Details of the framework of Multi-Particle Finite Element Model (MPFEM) are discussed elsewhere [63]. We only provide a summary of the model here. A number of particles are compacted under quasi-static conditions using a commercial finite element program (ABAQUS/EXPLICIT). When there is a very large number of actual or potential contacts or the material undergoes large plastic deformation, the explicit integration is more appropriate than implicit. There are two major differences from the prior MPFEM work. A degrading layer of elements is added on the surface of each particle
to model partial cohesion. Also, a much finer mesh is needed to capture the details of the interface cohesion during unloading compared to prior MPFEM work. The mesh design used here for a single particle is shown in Figure 5.1. There are 633 nodes and 560 elements per particle, of which 144 nodes and 144 elements are located at particle surface. This mesh design ensures little geometric compromise for the contact. The study of the adequacy of the mesh design can be found in Chapter 4. In this study, a complete simulation cycle also includes compaction, unloading, ejection and reloading.

Each particle is discretized into two regions: the main body zone and the degradation zone, which is one layer of elements on the surface. The thickness of outer layer element is about 1.5% of the particle diameter. The material for the main body is modeled to be linear elastic-perfectly plastic. At the stage compaction, the degradation behavior of the material in the degradation zone is not activated. Therefore, we assume it has the same material properties as the bulk material (Fig. 5.1(a)). During the unloading stage and thereafter, to model the semi-cohesive interfaces, the material in the degradation zone is modeled with the continuum-based constitutive equations as discussed in Chapter 4.2(Fig. 5.1(b)). A convergence study conducted by Procopio [63] showed that for 200 particles some wall effect is present while it is very small for 400 and negligible for 800 particles. Due to the extreme computational requirements of this simulation we kept the number of particles to 200. To avoid pathological “crystallization” in packing, the particles were generated with a radius which be randomly distributed with a standard deviation of 5% of the mean radius, see Figure 5.2.
Boundary conditions and simulation procedure

Rigid surfaces was used to simulate the die and punches by imposing external displacement/velocity boundary conditions via their interactions with particles touching the walls. The particle-wall interaction is set as frictionless in all cases. With a limited number of particles, the ‘wall effect’ comes into the picture. Particles near the rigid boundary tend to form long chains parallel to the wall with little ability for transverse movement, resulting in higher local stress. As a result, it increases the overall rigidity of the subsequent macroscopic response.

The number of contact pairs plays an important role in the computation time. Potential contact pairs are prior-defined by identifying nearest-neighbor particles within a given radial length. Since there is a large amount of particle rearrangement and rotation at the beginning of compaction, a relative large sweep radius should be chosen. In our simulation, this length is set to be 4 times of particle radius. At the end of compaction while the particle rearrangement becomes inactive, a smaller sweep radius may be used to reduce the number of contact pairs and to accelerate the computing.

Close-die compaction was achieved by applying acceleration boundary condition on top and bottom rigid surfaces, while the left and right rigid surfaces remain fixed. Initial acceleration was set as 0.15 on the bottom punch and -0.15 on the top punch. Note that the average particle size is taken as 1. After certain point, a deceleration boundary condition was set with a value of -0.075 on bottom punch and 0.075 on top punch. The transition point from acceleration to deceleration was adjusted so that when the particles are compacted to a designed relative density, the overall kinetic energy (or velocity)
reaches minimum. The total step time was adjusted to simulate the close-die compaction of powder for a range of final relative densities. The typical step time is about 15.0. The simulation is extreme computationally consuming due to large number of nodes, elements and contact interactions involved. To accelerate the calculation, a mass scaling factor may be used to artificially increase the maximum time increment at each time increment. To avoid the inertia effect by introducing the mass scaling, a relative small value (50) has been applied (the density of bulk material is set as 0.0001). At the end of close-die compaction, some elements will have large distortion when they are extruded into the gap of the other particles. To avoid the convergence problems due to the element distortion, we also applied an adaptive mesh technique which is provided by ABAQUS/Explicit to adjust the shape of certain number of elements. Typically, it takes about 40 hours for a desktop PC (3.39 GHz CPU and 3G of RAM) to finish 200 particle close-die compaction simulation.

After compaction, an unloading simulation model was recreated with the information of nodes coordinates and the stress conditions at each integration point that collected from previous loading calculation. A complete unloading stage includes two steps: (i) removal of top and bottom punches until the reaction forces in vertical direction become zero (“unloading”); (ii) removal of horizontal punches until the reaction forces in transverse direction become zero (“ejection”). Decohesion of the contact interface was activated in the first step of unloading. To alleviate the “wall effect”, interfaces that are attached to the rigid surfaces are modeled with cohesionless interface boundary condition. Therefore, the interface damage will only occur at internal particles contact interfaces. An
acceleration boundary condition was applied on the top and bottom rigid die with the same value that was used in the previous compaction simulation (0.075 on the top rigid die and -0.075 on the bottom rigid die). The total step time depends on the reaction force evolution. To accelerate the calculation, a mass scaling factor with the value of 50 was used again. Simulation was stopped when the reaction force at top and bottom punch reach zero. A typical step time is about 2.0. After unloading, “Ejection” simulation was then conducted by restarting the previous unloading calculation at the point when the reaction force at top/bottom punches reach zero. During this step, the top and bottom punch is free to move. The acceleration boundary conditions are applied on left and right rigid dies with a value of -0.075 and 0.075 respectively. The total step time is about 1.0.

After ejection, reloading is then conducted by restarting the previous ejection simulation at the point when the reaction force at side rigid dies reach zero. A velocity boundary is applied on the outer surfaces in either horizontal or vertical directions with the value of 0.05. When reload in one direction, the rigid surfaces in the transverse direction are free to move. In a typical force displacement curve that specimen reloaded with low triaxiality stress condition (e.g. tensile test), the reaction forces at the rigid surfaces first increase until reach the maximum, and then decrease. The strength of the compacted powder, therefore, is calculated by dividing the maximum force recorded during reloading by cross section area. Compaction and unloading/ejection sequences were simulated for a range of final relative densities and the corresponding strengths in the compaction and the transverse to compaction directions were obtained in order to compare the model predictions with the anisotropy experiments of Galen and Zavaliangos [10].
5.3 Results and discussion

Close Die Compaction

The evolution of compaction and side wall stresses are shown in Figure 5.3. Densification is initially achieved by a combination of rearrangement and contact deformation. Rearrangement essentially stops around 0.85 [63]. Further densification is accomplished primarily by particle contact and bulk plastic deformation. Since the wall stress is always smaller than the compaction stress it is expected that the particle contact area should exhibit an orientation dependence. The corresponding prediction of the model is shown in Figure 5.4.

Figure 5.4 shows the accumulated contact areas that were projected into the first quadrant at different densities during compaction. The calculated total contact area between 200 particles was distributed into six ranges based on the direction of the average normal to the interface. Contacts with a normal close to 90 degree from the horizontal are along the prior compaction direction, while those close to 0 degree are along the transverse direction. Contact areas increase as the density increases as expected. The contact areas with a normal along the horizontal direction are always smaller than those in the vertical direction. This effect is exaggerated in the affine motion of the Fleck 1995 model who assumes that the contact area is proportional to $\sin^2 \theta$ which gives a zero contact area in the transverse to compaction direction, which is obviously an exaggeration [109]. In any event, the larger area along the compaction direction may suggest a larger strength along that direction, at least if cohesion is perfect along the interfaces.

Figure 5.5 shows the pressure distribution at individual contacts at the end of
compaction on a representative particle. The particles undergo large plastic deformation during die compaction. The compressive pressures at the contact surface are almost uniform and reaches approximately $2\sigma_y$.

**Unloading**

Unloading is simulated with the gradual withdrawal of top and bottom rigid surfaces (unloading) and followed by removal of the rigid surfaces in transverse direction (ejection). By adjusting the materials properties of the degradation zone and the contact surfaces boundary conditions, our model is able to model three different cohesion scenarios: no cohesion, which was achieved by allowing particles to separate at the interface; full cohesion, which was accomplished by applying non-separation boundary and non-degrading surface material; and partial cohesion, which was achieved by applying non-separation boundary conditions and degrading material model as discussed before.

Figure 5.6 shows the evolution of stresses in both directions during unloading and ejection for the particles that have been compacted to high density (RD=96.6%). Three interface cohesion conditions were examined: full cohesion, partial cohesion and cohesionless. Similar to that in the unit cell study, the nonlinear stress-strain behavior is observed at the end of unloading. Particles with semi-cohesive and cohesionless interfaces show a large nonlinearity, comparing to the one with fully cohesive interfaces. This observation is consistent with our previous unit cell study and experimental observations [62]. Figure 5.6 also shows that there is a significant residual wall stress remaining at the end of unloading (approximately 26% of the compaction stress). This stress together
with the wall friction is responsible for the ejection force in actual compaction.

In order to understand what happens at unloading (which may affect the final properties), we need to consider first the stress distribution at unloading of fully cohesive contacts. The stress distribution at a typical contact interface during unloading for full cohesion contact interface is shown in Figure 5.7. Because there are no external loads, the average contact force carried by the interfaces is zero (which some statistical spread to both net tensile and compressive loads). An interesting pattern of local stresses is observed at the contacts of the particle shown in Figure 5.7. A tensile stress was observed at the tip of contact. The fully cohesive model is capable of carrying this stress. Therefore, the fully cohesive model predicts that the strength of the green compact approaches the strength of the particle itself, when the relative density reaches one. This is also the underlying assumption of all common isotropic continuum models. In reality, the strength of green compacts is much smaller than the corresponding strength of the particle. The contact surfaces are in reality far from perfect. Local roughness, surface oxides and other surface impurities reduce significantly the ability of the contact to carry load. If the cohesion between two surfaces is low, it is possible that the contact tip opens due to the tensile stress. As a result, not only the contact tip opens, but also the load carrying capability is reduced due to the development of local cracks.

In order to study the spatial characteristics of degradation, we follow a parameter called “SDEG”, which is provided by ABAQUS to quantify the amount of degradation of the material. When SDEG value reaches 1, the material is completely failed and the loading carrying capacity is completely lost. We assume those elements with a SDEG reaches 1.0
are failed. Therefore, we are able to calculate the residual contact area during and after unloading. Assuming those contact interfaces that has a 70% of elements failed are the damaged interfaces. This simulation predicts damage not only in unloading but in ejection as well. As can be seen in Figure 5.8, we have about 3% lose of contacts in prior compression direction (75~90°) during the first step of unloading and 7% lose after ejection. On the other hand, the contacts in the transverse direction (0~15°) only have less than 1% of contact damaged during the unloading stage. The failure at these contact interfaces may serves as small crack and has a major effect on the strength and anisotropy of compacts.

**Reloading and strength probing**

Reloading was achieved by applying displacement on the rigid surfaces in prior compaction or transverse directions after complete unloading. A maximum tensile force was recorded during reloading. The strength of the compact was calculated by dividing the maximum force by cross section area in that direction. Figure 5.9 (a) and (b) shows the final strength and strength anisotropy for the compacts of different relative densities. The results are very revealing. Fully cohesive results indicate that the strength of the compact reach a large fraction of the yield strength of the particles at high densities which is uncommon in regular green compacts. Also higher strength along the prior compaction than the transverse direction is predicted. (Figure 5.9b). This directly reflects the orientational dependence of contact areas (Figure 5.4), but is opposite of experimental observations [10]. Semi-cohesive interfaces, on the other hand predict strength that are
significantly lower than the yield strength of the particles (~5-10%) and predict the correct sense of anisotropy. In addition, the semi-cohesive model predicts an increase of anisotropy at higher density in direct agreement with the experiments. Although the picture of cracks at the tips of the contacts may not be perfect, it is not far from reality. X-ray microtomography observations by Lame et al. [69] also prove that there is “damage” in contacts normal to compaction direction (Figure 1.6). It is possible to assume that any imperfection along the particle contacts may give rise to local crack and damage.

As shown in Figure 5.10, our model predicts that such damage is more severe along contacts normal to the prior compaction direction. This prediction is key because it brings the model predictions for strength level and anisotropy in green compacts in agreement with experimental observations.

5.4 Concluding statement

Within the framework of MPFEM, the effect of loading path and interparticle cohesion on the achieved strength anisotropy has been analyzed. The introduction of a layer of degrading material on each particle surface allows us to model the semi-cohesive contact interfaces. Our simulation results show that the unloading and ejection stages play a very important role on the strength anisotropy of compact. After removal of punch in the prior-compaction direction, the large residual stress in the transverse direction results in elastic recovery of compacted particles and local stress concentration. If the interparticle cohesion is weak, our simulation results show that die compacted powder has a higher strength in the transverse direction than in the prior-compaction direction. As the final
relative density increases, this difference becomes stronger. The main cause is the damage and debonding of the contact interfaces due to the local tensile stresses that develops at particle interface during unloading and ejection stages. As the result, the final strength of the compact is much lower than the bulk material itself. On the other hand, full cohesion model predict a much higher strength of compacted powder. The full cohesion model predicts a die compacted powder with a higher strength in the prior-compaction direction. This result is consistent with the orientational contact distribution caused by die compaction but contradicts the experimental observation for ductile powders.

Our decohesion model is the first that explains why close-die compacted material has weaker strength in the prior-compaction direction than along the transverse direction. However, our model also has some limitations. 200 particles may not well represent the actual microstructure of powders. The parameters used in the degrading material model need experimental calibration. A detailed discussion of the limitations of current model and the future work will be presented in the next chapter.
Figure 5.1 Representative mesh used in MPFEM simulation. (a) Initial circular particle (633 elements) (b) Deformed particle, the outer blue layer represents the degradation zone.
Figure 5.2 Initial configurations of 200 particles within four rigid surfaces (die/punches).
Figure 5.3 Normalized reaction stresses \( (\sigma / \sigma_y) \) versus relative density during loading.

(\( \sigma_y \) - yield stress).
Figure 5.4 The distribution of accumulated contact area that was projected into the first quadrant. The contact areas are summed up for those contact angle locates in $\theta_1 < \theta < \theta_2$ ($\sum A_i$) and then normalized by total surface area for 200 particles $\sum_{i=1}^{200} \pi D_i$. $D_i$ represents the diameter of each particle.
Figure 5.5 Pressure distributions on contact interface at the end of die compaction.
Figure 5.6 Normalized die (horizontal) and punch (vertical) stresses at rigid surfaces during unloading. Semi-cohesive interfaces were used and the out of die relative density was 96.6%.
Figure 5.7 Pressure distribution on contact interface at the end of unloading (full cohesion contact interface). This stress may exceed the yield stress because of the multiaxial stress conditions or small numerical errors from the simulation.
Figure 5.8 The evolution of contact area for semi-cohesive interface during unloading. The contact interface areas are also projected into first quadrant and plotted in six ranges starting from horizontal (0-15) to vertical directions (75-90). The damage dissipation energy is $G/(\sigma_c R) = 0.0006$. The 200 particles were initially compacted to 96.6% out of die relative density.
Figure 5.9 Normalized strength in the prior-compaction direction (normal strength) and transverse direction (transverse strength); (b) Strength ratio (transverse versus normal) as the function of relative density.
Figure 5.10 Contact geometry after unloading. (Red area represents the failed contact interface, blue area represents the intact contact interfaces)
CHAPTER 6: CONCLUSIONS AND FUTURE WORK

6.1 Conclusions

Our major goal in this study is to better predict the strength and strength anisotropy of cold-compacted powders. The following conclusions are based on the numerical analysis of powder compaction at two length scales.

At macroscopic level, one of the most popular phenomenological models, the Drucker Prager/Cap model has been implemented into ABAQUS/Explicit and a user subroutine (VUMAT) has been developed. This model was used in order to determine whether peculiarities observed in the experimental results of Sinka et al. can be predicted [13]. In these results, two tablets were compacted to the same final shape and average density under different wall lubrication conditions. Subsequent diametrical compression of these tablets gave different fracture loads and modes of fracture. The predictions of the DPC model in terms of load displacement curve are reasonable but the prediction of the failure mode was not correct. The use of maximum principal stress criterion gives better insight to the different fracture modes observed but without invoking implicitly anisotropy, the experimental results can not be well explained.

The difficulty of the DPC model to predict green strength is due to its isotropic nature. While the assumption of isotropy is reasonably acceptable for the compaction stage, fracture by nature is not isotropic. Therefore, although the DPC model predicts the load to failure, this is achieved mainly due to the experimental calibration. When it comes to failure mode, the isotropic Mohr-Coulomb shear failure line ($F_\sigma$) is not capable of detecting the direction of crack propagation. This is why the maximum principal stress
(Rankine) criterion gives better insight to the mode of fracture. But even in this case, we only get an indication that a second mode is possible. The local normalized strength index $\gamma$ (Eq. 3.5) gives similar chances for the two modes to occur (Fig. 3.10). Only if we invoke the potential anisotropy we can rationalize the experimental results.

Our particle level study focuses on addressing the physics and mechanics of interparticle interface. The introduction of a layer of degrading elements on the particle surface provides the means of modeling different levels of interfacial cohesion. The analysis of close-die compaction, unloading, ejection and reloading was conducted on both unit cell and multi-particle models. Our results show that at the stage of compaction, the stress at the contact interface quickly reaches uniform due to the large deformation. The developed contact interfaces show an orientational distribution due to the restricted movement in the transverse direction during die compaction. During the stage of unloading, a tensile stress concentration was observed at the root of the contact necks. This stress may cause partial or full separation of contact interface when the cohesion developed during loading is not strong enough. We also observed a non-linear elasticity at the end of in-die unloading for weak bonded interfaces. This is consistent with experimental observations. This non-linear behavior becomes stronger as the interparticle cohesion becomes weaker. In the simulation, we observed the damage of interface and crack propagation during unloading and ejection stages. Both the periodic boundary condition and MPFEM simulation predict that contacts with a normal along the prior compaction direction develop more damage during unloading and ejection. As a result, the final compact after ejection shows a higher strength in the transverse direction than in
compaction direction. It was the first time that a model predicts the right trend of strength anisotropy for weak bonded cold compacted powders and gives a reasonable explanation for the experimental observations. Our results bring a new perspective on understanding the interparticle behavior and the origin of the strength and failure of cold compacted powder.

Further systematical analyses of powder compaction and prediction of strength and anisotropy are also possible. The loading path dependence, interparticle cohesion and unloading stage can all be taken into account in the framework of MPFEM simulation, as well as other important factors such as the effect of material hardening.

Limitations of the proposed model do exist. The geometry of the unit cell is not representative of powders and this shows in the predicted trend of the strength and anisotropy versus relative density. Many of these limitations are alleviated by the MPFEM model which provides a better geometric picture of the compact and its microstructure. The MPFEM model is, however, a 2D model and its predictions were compared with experiments only in terms of trends. Extension to 3D models is not practical with currently available computer power. Even the 2D problem solved utilized a limited number of particles (200), which is lower than what is required for statistical representation of the compact (\( N_p = 400 \sim 800 \)). Therefore, a “wall effect” is present and we had to deal carefully with it. Another limitation of the current formulation of the MPFEM model is that all contacts at the end of compaction are given the same “strength” (cohesion) properties. In reality the history of stresses on each contact should determine the individual strength of each contact. This should be addressed in the future. Despite these
limitations, we believe that the proposed model is a major step forward towards understanding the strength and anisotropy of powder compacts in the green state.

6.2 Future work

Within the current MPFEM framework, several further simulation studies can be readily implemented. First, the effect of particle shape and initial packing/damping on the strength anisotropy of compact can be studied. Currently, MPFEM approach can model elliptical or acicular particles. Elliptical or acicular particles will cause an orientational microstructure during packing. This anisotropic microstructure will affect the particle deformation, evolution of contact areas and interface cohesion during subsequent loading and unloading condition, which in turns will affect the final strength and strength anisotropy of green compact. Secondly, the mixture of hard and soft particles can also be studied. Soft particles tend to deform more than hard particles. As the result, the orientational contact area distribution observed in Fig. 5.4 may be biased, which in turn will also affect the final strength anisotropy. Other studies can also be performed such as: (a) the compaction of porous particles; (b) Effect of material hardening and effect of particle fragmentation etc. These studies would help to build a more comprehensive picture of the strength of green compacts. The biggest challenge is to introduce more reality in the decohesion model that control the final properties. Obtaining directly the properties from contact measurements (e.g. through AFM experiments [110]) is conceivable but difficult. Perhaps the most promising way is to adjust the properties to match the results of a macroscopic test (e.g. diametrical compression test, fracture
toughness etc.). The only problem there is the need of 3D simulation, which at this point is only possible with DEM but not MPFEM.
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APPENDIX A: STRENGTH ANISOTROPY PREDICTION BY FLECK’S MODEL

The 3D prediction of yield surfaces by Fleck’s model is shown in Figure A.1. The red color surface represents the yield surface at hydrostatic compression condition. The elongated blue one represents the yield surface at close-die compaction condition, in which the prior compaction direction is along 3-direction. Under axisymmetric strain rate condition, both 3D yield surfaces can be simplified and plotted in 2D plane as has been shown in Figure 1.5.

After compaction, the virtual diametrical compression tests were conducted in two directions (along and transverse to prior compaction direction) for two cases, one for hydrostatic compression and one for close-die compaction. Figure A.2 shows the way we virtually cut two cylindrical specimens after close die compaction, which was initially compacted in the third direction. In the first case, the axis of the cut specimen is parallel to the prior-compaction direction. Therefore the measured strength is along transverse direction. During diametrical compression test, the maximum tensile stress that causes failure is in the center of the sample and is perpendicular to the compression direction with the value of 1/3 of that of the compression stress. Therefore, as shown in Figure A.2, the stress condition is $S3 = 0, S1/S3 = -3$ for this specimen during the diametrical compression test. This stress path is also shown by a magenta arrow in Figure A.1. The measured strength, therefore, is corresponding to the point at which this stress condition reaches the yield surface in that direction. In the second case, as shown in figure A.2, the axis of the specimen is perpendicular to the prior-compaction direction. The strength measured during the test is along prior-compaction direction (normal direction). Under
this condition, the stress condition is \( S_1 = 0, S_2/S_3 = -3 \), as also shown by a green arrow in Figure A.1.

The calculated strengths based on the yield loci predicted by Fleck’s model are listed in Table A.1. The parameter of \( \eta \) represents the cohesion between two particles, while the values of 1 and 0 indicate full cohesion and cohesionless respectively. The calculated strength ratios are also listed in the table. According to these data, we have the following conclusions:

- Under hydrostatic compaction, there is no strength anisotropy.
- Under die compaction condition, strength anisotropy depends on the cohesion. Low cohesion results in high anisotropy.
- Fleck’s model predicts that the strength along loading direction is always higher than that in the transverse direction (\( SN > ST \)) for the samples compacted under close-die compaction condition. This originates from the \( \cos^2 \theta \) distribution of contact area, but is opposite to experiment.
- Fleck’s model implies that powder compacted with hydrostatic compaction will show higher strength than that compacted with die compaction during diametrical compression test.
Table A.1 Calculated strength and anisotropy during diametrical compression test predicted by Fleck’s model.

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Figure A.1 3D yield surfaces predicted by Fleck’s model. Red color represents yield surface at hydrostatic compression condition, blue color represents yield surface at close die compaction condition.
Figure A.2 Schematic representation of diametrical compression specimens cut from previous close-die compaction sample.
Vita

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