Experimental Studies and Modeling of the
Roller Compaction of Pharmaceutical Powders

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John C. Cunningham
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Abstract
Experimental Studies and Modeling of the Roller Compaction of Pharmaceutical Powders
John C. Cunningham
Antonios Zavaliangos, Ph.D.

During roller compaction in the pharmaceutical industry, mixtures of active and inert powders are fed via a screw to counter-rotating rolls, drawn into the nip and compacted under hydrostatic and shear stresses. Experimental studies were conducted using microcrystalline cellulose on a roller compactor that measured feed force, surface roll pressure and shear stress. The following observations were made: densification correlated with maximum roll pressure; increasing feed force increased roll gap; and significant variation in roll pressure and shear stress exists in the transverse and rolling directions. A slab model highlighted the importance of roll friction, feed stress and entry angle on pre-densification in the feed zone. 2-D and 3-D explicit finite element models with adaptive meshing and arbitrary Eulerian-Lagrangian capabilities were developed. A Drucker-Prager/cap model was calibrated using diametrical and simple compression and die compaction tests. The roll friction was estimated using a die instrumented to measure radial stress. The effects of roll friction, feed stress, roll gap to diameter and entry angle on roll force, torque, profiles of roll pressure and roll shear stress, nip angle, neutral angle, and relative density were evaluated. The results indicated increasing entry angle, decreasing roll gap to diameter, increasing feed stress and/or increasing roll friction lead to higher maximum roll surface pressure and attendant relative density at the exit. The results may be explained by the nip angle and amount of pre-densification. Simulations with pressure-dependent frictional coefficients indicated significant difference in
densification. Oscillating feed stress conditions revealed periodic variations in roll pressures and relative densities. Variations in the through-the-thickness were significant in the slip region and diminished in the nip region. The 3-D model predicted lower roll pressure and densities near the edges due to side seal friction. In addition, variable inflow of material along the roll width was related to variation in roll pressure. Overall, the model predictions followed experimental trends. Microcrystalline cellulose experienced higher expansion on release than predicted - related to its non-linear elastic behavior. Various combinations of boundary conditions and geometrical parameters resulted in similar roll pressure profiles and densification thus accurate experimental inputs are essential for model verification.
Chapter 1. Introduction

Roller compaction or roll pressing is used in a variety of industries including chemical, metals, pharmaceuticals, minerals and recycling. In the case of the pharmaceutical industry, roller compaction is a unit operation in the dry granulation process, a commonly used pressure-induced agglomeration technique in which granules with acceptable flowability, compaction properties, compositional uniformity, and chemical stability are formed from feed material consisting of mixtures of active and excipient (inert) powders. During the dry granulation process the dry powders of the active ingredient and excipients, e.g., dry binders, disintegrants, diluents and lubricants, are mixed in a blender. The powder mixtures are then roller compacted and milled to form granules. The resulting granulation is typically blended with a lubricant and either encapsulated or compressed into a tablet.

During roller compaction, the powder mixture is continuously fed – often with the assistance of a screw or auger feeder – to two counter-rotating rolls, which draws the powder between the rolls and compacts the powders into strips or flakes of compacted material. The feed powder is densified by a combination of shear and hydrostatic stresses that develop from the feed stress, friction at the rolls and the confining boundaries, i.e., rolls, screw and lateral side seals.

While roller compaction is often used successfully within the pharmaceutical industry at the production scale, engineers and scientists often rely on an empirical approach to define and optimize the formulation and process. There are a number of reasons for this high reliance on the experimental approach: 1) lack of experimental
measurements of important process parameters; 2) limited efforts on process modeling; and 3) inherent complex mechanical behavior of powder systems. During the process, important process conditions, e.g., surface roll pressure or feed stresses, are typically not measured due to the cost and difficulty in the design and implementation of such measurement systems. These parameters are, however, essential to the understanding and control of the feeding of the powders to the roll and to the development of the compact properties, e.g., the density of the compact is directly correlated to the maximum roll pressure developed. Without measurement of these parameters, researchers must depend on such indirect measurements as the overall roll force. In addition to the lack of process measurements, there has been limited process modeling to date. Process modeling coupled with experimental studies often provides a rational scientific framework to i) probe the underlying physical processes; ii) understand relative importance of various material, equipment and process factors; and iii) predict and optimize formulations and process parameters. While roller compaction appears be a rather simple operation, it is a complex deformation process involving highly non-linear contact and material behavior with complicated boundary conditions, e.g., oscillating screw feeding at the roll entry. Finally, the feed powder undergoes significant evolution in mechanical properties from its rather loose state in feed zone to the compacted ribbon or flake upon exit of the rolls. Capturing of these changes requires determination of an appropriate constitutive model with its accompanying experimental calibration – typically under different states of densification and stress - and its implementation into the process model.
The overall goal of this thesis is to provide greater understanding of the roller compaction process through a series of experimental studies and modeling efforts. More specifically, the objectives of this thesis include:

- Obtain a understanding of the roller compaction process through detailed experimental studies on different model pharmaceutical powders using an instrumented roller compactor capable of measuring the feed force induced by a screw feeder and the roll surface pressure and roll shear stress at different locations across the roll width;
- Determine estimates for experimental inputs of the constitutive and frictional parameters required for the modeling of the roller compaction using mechanical tests and application of elasticity, plasticity and friction theory; and
- Develop process models for the roller compaction process and conduct parametric studies to 1) understand the relative importance of the boundary conditions (feed stress and powder/roll friction), geometry (roll gap to roll diameter and entry angle), initial conditions (initial relative density of the powder) and material parameters of the feed powder and 2) predict the development of contact stresses, i.e., roll pressure and roll shear stress, internal stress and strain fields, densification, and loading conditions, e.g., roll force and roll torque.

The layout of the thesis is as follows: Chapter 2 is a literature search of the use of roller compaction with emphasis on the pharmaceutical application and the review of the detailed experimental studies and process modeling efforts conducted. Chapter 3 provides the results of experimental studies conducted on the instrumented roller
compactor. This includes the measurement of the local roll surface pressure as a function of the rolling angle using a roll instrumented with five discrete strain gage load cells positioned across the width of the roll. The screw feed force is also measured. Chapter 4 focuses on the experimental inputs required for the process models. The experimental results of the characterization of the model powders including their elastic, plastic and frictional properties are discussed. The development of the pressure-dependent frictional elasto-plastic constitutive model is presented. The calibration procedures based on a series of mechanical tests and the results are outlined. Relevant background information on material characterization is also provided. Pertinent derivations are described in Appendices A and B. Chapter 5 is the first of three chapters on the process modeling. More specially, this chapter presents the development of a one-dimensional slab model of roller compaction. A simple porous plasticity model is used to represent the stress-strain behavior of microcrystalline cellulose. The model applies an iterative numerical solution for minimizing the stress gradient in the angular direction. The results of a series of parametric studies on the effects of the boundary and initial conditions, geometrical parameters and material properties of the feed powders are discussed. While there are several limitations to this model, it provides a resource-effective manner to understand the relative importance of several material, geometric and boundary conditions on the development of the roll pressure and attendant densification. Chapter 6 presents the development of the two-dimensional plane strain process model based on the application of the finite element method. The pressure-dependent, friction constitutive model, i.e., the modified Drucker-Prager/cap, is introduced along with roll pressure dependent frictional properties at the roll surface and
oscillating feed stresses used to simulate the feed screw. The results of an extensive series of parametric studies are discussed. The preliminary results of the three-dimensional process model focusing on the effect of the stationary side seal and non-uniform feeding are presented in Chapter 7. Chapter 8 briefly compares the experimental results and modeling results and highlights key considerations when evaluating the predictive capabilities of models of roll compaction. Finally, the overall conclusions and recommendations for future work are covered in Chapter 9.

For clarity and convenience, the definition of symbols are typically noted in the relevant text, however, a comprehensive listing is also provided in Appendix C.
Chapter 2. Background

In this chapter an overview of the roller compaction process is provided with a summary of the detailed experimental studies conducted and process models developed.

2.1. Basic Process and Applications to Pharmaceutical Industry

The roll compaction process dates back to the second half of the 1800s when a roll type briquet machine or Belgian roll press was used to agglomerate coal screenings. The material was fed via gravity to the two counter-rotating rolls that often had pockets or indentations cut into the surface to allow the formation of briquets or compacts. The roller compaction process offered a relatively low cost, continuous means to agglomerate a wide variety of materials.

In a similar manner to calendaring of plastics or rolling of fully dense metals, the basic roller compaction process involves the feeding of material to two counter-rotating rolls, which grip and draw the feed material through a nip region and release upon exit through the roll gap. A schematic of the regions of compaction is provided in Figure 2.2.1. The rolls have been broken up into two main zones: the feed or slip zone and the compaction or nip (stick) zone (Pietsch, 1987). The feed material is delivered to the rolls under gravity or by a force or screw feeder. Under gravity feed, adjustable tongues may control the flow of powder with the distribution across the roll width achieved by rotating devices mounted above the rolls. The screw feeder induces a positive feed pressure to be applied and can be used as part of a control system. The angle of delivery or entry is defined by the angle, $\alpha_{\text{entry}}$. The angular boundary of the feed or slip region is defined
\( \alpha_{\text{entry}} \) and nip angle, \( \alpha_{\text{nip}} \). Within the feed zone the feed pressure induced by either gravity or the force feeder and the friction forces at the roll/powder interface are applied to the powder. These forces drive the powder toward the nip region. The stresses developed are relatively low and densification of the powder in this region is usually dominated by particle rearrangement (Pietsch, 1987). In addition, the peripheral speed of the rolls is higher than the material adjacent to the rolls in this region. The material is thus experiencing slipping.

At the nip angle, \( \alpha_{\text{nip}} \), the powder is gripped by the rolls and drawn between the rolls. The powder adjacent to the rolls moves with the same speed of the peripheral speed of the rolls and is considered sticking to the rolls. It is also referred to as angle of compaction, gripping angle and angle of rolling (Pietsch, 1987). The stress within the powder begins to rise more rapidly and the material undergoes greater densification. This densification may be affected by plastic deformation and/or fracture of the particles. The roll pressure rapidly increases to a maximum in the nip region. At an angle defined by the neutral angle, \( \alpha_{\text{neutral}} \), the direction of the friction force at the roll reverses direction, i.e., frictional force is directed away from the centerline of the rolls, i.e., a rolling angle of zero (\( \alpha = 0 \)). The neutral angle is prior to the centerline of the rolls. After the roll pressure reaches its maximum, it begins to drop as the compacted powder progresses through the roll gap and is released at angle \( \alpha_{\text{release}} \). Pietsch also notes that a third zone may be defined, the extrusion zone, which may occur between the neutral angle and release angle. This zone is related to the possible acceleration of the compacted material with respect to roll, i.e., the compacted material moves faster than the peripheral speed of the rolls.
The roller compaction of powder may appear rather simple at first glance. However, upon further analysis, it is readily recognized that there are many factors that can affect the quality of the compacted material including machine design parameters (e.g., feed system, roll diameter, roll surface configuration, roll gap); process parameters (e.g., roll speed, screw speed); and mechanical properties of the feed materials (e.g., constitutive behavior and frictional properties at the roll surface). In addition, the effect of air within the pores or voids can influence the feeding and the resulting compact properties as it is compressed and flows within the powder bed.

Roller compaction has been applied in many industries including the chemical, food, pharmaceutical, metals, agricultural and environmental fields to agglomerate and densify feed materials, reduce dust, improve solids handling characteristics, prevent chemical segregation and enhance reactions of component materials. In the pharmaceutical industry, roller compaction is a continuous, pressure-induced agglomeration unit operation in the dry granulation process. The primary goal of dry granulation is the production of granules with good flow, compositional uniformity and compaction properties. It is an alternative to wet granulation with aqueous or solvent-based solution and elevated temperatures during subsequent drying that may lead to chemical instabilities of the active ingredient. The pressure agglomeration step in the granulation process may be either through slugging or roller compaction. In the slugging process, large tablets or slugs – often 25 mm or larger in diameter – are formed on a tableting press. While still used by some companies, slugging is usually not used due to higher capital costs compared to roll compaction and because the feeding the tablet press
uniformly at acceptable throughput rates can be difficult for fine, poorly-flowing powders. Roll compactors with screw force feeders have become common.

In the typical dry granulation process, the active pharmaceutical ingredient is mixed with inert excipient powders, e.g., lactose, microcrystalline cellulose and dicalcium phosphate in a ribbon or tumble blender. A disintegrant powder may be also blended into the powder mixture to improve the disintegration and dissolution of the final dosage form. The powder mixture is then usually blended with a lubricant, e.g., magnesium stearate, to minimize adherence to the process equipment, e.g., hopper walls. The relative amount of lubricant and lubrication mixing time can affect the frictional properties of the final feed material. Optimization of the lubrication of the feed material is an important activity in the design of the roller compaction process. Inadequate lubrication can result in sticking of the powder to the rolls. Excessive lubrication can lead to inability of the rolls to grip and draw the feed material between the rolls.

After lubrication the feed powder is fed to a hopper, which in turn delivers powder to the screw feeder. The feed material is conveyed to the counter-rotating rolls and compacted. The compacted ribbons or flakes are milled and, if necessary, classified into various granule sizes for further downstream processing, e.g., fine material may be recycled back to the roller compactor or coarse fraction may be milled smaller. The milled granulation is finally blended with any extra-granular powder and lubricated prior to the encapsulation or tableting operation.
Figure 2.1.1. Schematic of regions of roller compaction.
2.2. Experimental Approaches

2.2.1. Empirical / Design of Experiments

Most of the academic and industrial research on roller compaction has focused on understanding and optimizing the formulation variables and process parameters. Many different formulations and types/scales of roller compactor designs have been employed in these studies. Systematic one-variable-at-a-time experiments or statistical design of experiments are commonly conducted. The formulation variables may be studied in effort to optimize characteristics of the resulting granulation, e.g., flowability, particle size distribution or compactibility, or final product properties, e.g., dissolution behavior. The process variables may include specific roller compactor design features, e.g., specific feed design, dearation, roll design and scale, and/or process parameters, e.g., screw speed, roll speed and gap setting. The experimental studies of roller compaction in the pharmaceutical industry have been reviewed by Miller (1997), Murray (1997) and most recently Kleinebudde (2004).

Systematic one-variable-at-a-time experiments (Sheskey et al., 1994; Sheskey and Dasbach, 1995; and Sheskey and Hendren, 1999); design of experiments (Cohn et al., 1966; Falzone et al., 1992; Rambali et al., 2001) and neutral networks (Inghelbrecht et al., 1997 and Turkoglu et al., 1999) have been used to evaluate materials, formulations, process parameters, scale up and equipment design. Similar experimental studies have been used to evaluate materials (Mollan and Celik, 1993; Inghelbrecht and Remon, 1998(a) and 1998(b); Freitag and Kleinebudde, 2003; Mitchell et al., 2003), process parameters (Hervieu et al., 1994) and scale up (Sheskey et al., 2000). These studies are
often practical and useful, but are limited to the specific materials, process conditions and equipment evaluated. Extending the findings to a broader understanding of roller compaction can be difficult.

2.2.2. *Detailed Studies*

In effort to advance the understanding, optimization and control of the roller compaction process, it is essential that more detailed experimental measurements be made to assess the stresses and deformation that occurs in the various regions, i.e., feeding, slip, nip and release. Several of more detailed experimental studies conducted in roller compaction focused on assessing the feed zone, development of the local roll surface pressure and roll shear stresses and the evaluation of the resulting compacted material. A summary of the studies is provided in Table 2.2.1.
Table 2.2.1. Detailed experimental measurements used in studying of roller compaction.

<table>
<thead>
<tr>
<th>Measurement</th>
<th>Materials (References)</th>
<th>Notes</th>
</tr>
</thead>
<tbody>
<tr>
<td>Roll Force</td>
<td>Corn starch, lactose and Elcema G250 (Jerome et al., 1991)</td>
<td>Related roll force to other process parameters (e.g., speed of feeder and rolls) and quality of compacts</td>
</tr>
<tr>
<td>Roll Pressure</td>
<td>Iron (Chekmarev, et al., 1963); Iron (Kataishinskii and Vinogradov, 1965(b)); Mixtures of iron/chromium carbide and nickel (Kataishinskii and Vinogradov, 1966); iron, lead, copper and nickel (Aksenov and Rebyakin, 1969(a) and 1969(b)); stainless steel, copper, iron and titanium (Mal’tsev, 1971); lignite and sodium chloride (Dec and Komarek, 1991(a)); roller milling of cement clinker (Anderson, 1990); lignite, bentonite and sodium chloride (Dec and Komarek, 1991(b)); alumina (Michel, 1994); roller milling of quartz (Lubjuhn et al., 1994); lactose and microcrystalline cellulose (Zega et al., 1998); lactose (Simon and Guigon, 2000); microcrystalline cellulose/dicalcium phosphate (Cunningham and Zavaliangos, 2002); lactose, alumina and sodium chloride (Simon and Guigon, 2003); microcrystalline cellulose (Bindumadhavan, et al., 2005); organic powder (Lecomposte et al., 2005)</td>
<td>Pressure profiles; relate local maximum roll pressure to compact density; variations in roll pressure in rolling direction and roll width</td>
</tr>
<tr>
<td>Roll Shear Stress</td>
<td>Iron (Chekmarev, et al., 1963); Iron powder (Kataishinskii and Vinogradov, 1965(a)); lead powder (Kuleshov, 1981) roller milling of quartz (Schonert and Sander, 2002)</td>
<td>Profiles of both roll pressure and roll shear stress; roll friction; neutral angle</td>
</tr>
</tbody>
</table>
Table 2.2.1. (continued) Detailed experimental measurements used in studying of roller compaction.

<table>
<thead>
<tr>
<th>Measurement</th>
<th>Materials (References)</th>
<th>Notes</th>
</tr>
</thead>
<tbody>
<tr>
<td>Feed Stresses</td>
<td>Lignite, magnesium oxide and silica fume (Dec and Komarek, 1993)</td>
<td>Feed screw simulator to measure forces within feed zone of screw conveyor; oscillations and spatial variation related to screw design and speed</td>
</tr>
<tr>
<td>X-ray</td>
<td>Aluminum (Vinogradov et al., 1972; Katashinskii, 1981)</td>
<td>Zones of compaction observed; shear bands in feed zone under gravity feed; density through feed and compaction zones</td>
</tr>
<tr>
<td>Transmitted Light</td>
<td>Sodium chloride (Guigon and Simon, 2003)</td>
<td>Related density variation in compact to screw feeding</td>
</tr>
<tr>
<td>Photo-micrography</td>
<td>Iron, copper and stainless steel (Tunderman and Singer, 1969)</td>
<td>Density in feed and compaction zones; density variation across roll width</td>
</tr>
<tr>
<td>Micro-indentation</td>
<td>Magnesium carbonates (Freitag et al., 2004)</td>
<td>Related roll force to compact quality</td>
</tr>
<tr>
<td>Video Imaging</td>
<td>Microcrystalline cellulose (Zega et al., 1998); Lactose (Simon et al., 1999)</td>
<td>Trajectories of particles (markers) in feed and compaction zone; nip angle determination; velocity profiles</td>
</tr>
<tr>
<td>Acoustic Emission</td>
<td>Lactose, microcrystalline cellulose and starch (Hakanen et al., 1993); microcrystalline cellulose and starch (Salonen, et al., 1997)</td>
<td>Monitoring compact quality</td>
</tr>
<tr>
<td>Color Change</td>
<td>Lactose with riboflavin as marker (Funakoshi et al., 1977)</td>
<td>Indicator of uniformity of roll pressure across width of roll</td>
</tr>
<tr>
<td>NIR</td>
<td>Bijlani et al. (2003); Gupta, et al. (2004)</td>
<td>Monitoring compact quality</td>
</tr>
</tbody>
</table>
2.3. **Process Modeling**

The development of a predictive model for roller compaction will significantly advance the understanding, design, optimization and control of the process. A review of these various modeling efforts was recently published (Dec et al., 2003). A brief summary of these models with some additional considerations is provided here.

Modeling of roller compaction has been challenging due to a number of reasons including: (1) complex constitutive behavior of powder; (2) complex frictional conditions at the roll/powder interface and difficulty measuring friction experimentally; (3) complex geometry/material interactions at the feeding zone; (4) significant inhomogeneity of properties in the strip (transverse and rolling directions); and (5) the effect of air under certain material and processing conditions. Despite these challenges, the development of simplified process models can provide important insight into roller compaction.

In approaching any modeling effort, it is important to recognize the purpose of the model and decide on the benefits and cost. Models that account for more of the details of the process will require greater effort to develop. The use of simplifying assumptions can often reduce the overall effort but the impact of these simplifications must be evaluated in light of the goals of the model development. The development of one-dimensional models can often provide elementary understanding of the process and provide a basis for future modeling and experimental efforts. Most of the process modeling to date in roller compaction has been one-dimensional. Some limited effort has been extended to expanding this to two-and three-dimensional models. These models are provided in Table 2.3.1 and are briefly reviewed.
### Table 2.3.1. Summary of process models for roller compaction.

<table>
<thead>
<tr>
<th>References</th>
<th>Model</th>
<th>Key Notes</th>
</tr>
</thead>
<tbody>
<tr>
<td>Johanson (1965(a) and (b))</td>
<td>One-Dimensional</td>
<td>Slip region used Jenike-Shield model and nip region used die compaction data; nip determined by equating pressure gradients for slip and stick; uses simplified method of characteristics</td>
</tr>
<tr>
<td>Johanson (1973)</td>
<td>One-Dimensional</td>
<td>Entrained gas in pores is incorporated in model</td>
</tr>
<tr>
<td>Katashinskii (1966, 1977) and Katashinskii and Shtern (1983(a) and 1983(b))</td>
<td>One-Dimensional</td>
<td>Slab method</td>
</tr>
<tr>
<td>Musikhin (1977)</td>
<td>One-Dimensional</td>
<td>Slab method; simple constitutive model based on difference of maximum and minimum stresses; assumes slipping at roll for all locations except neutral angle</td>
</tr>
<tr>
<td>Dec and Komarek (1991(a))</td>
<td>One-Dimensional</td>
<td>Slab method with “plastic” Poisson’s ratio</td>
</tr>
<tr>
<td>Gun et al. (1986)</td>
<td>Two-Dimensional</td>
<td>Porous plasticity model used; use Galerkin’s numerical technique to solve; calculates 2-D variation in shear deformation, density and velocity fields</td>
</tr>
<tr>
<td>Deshmukh et al. (1998)</td>
<td>Two-Dimensional</td>
<td>Modeling of rolling of sintered porous copper strip; uses upper bound theory</td>
</tr>
<tr>
<td>Domsa and Vasile (1998)</td>
<td>Two-Dimensional</td>
<td>Finite element model of metal powders</td>
</tr>
</tbody>
</table>
Table 2.3.1. (continued) Summary of process models for roller compaction.

<table>
<thead>
<tr>
<th>References</th>
<th>Model</th>
<th>Key Notes</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cunningham and Zavaliangos (2002)</td>
<td>Two - Dimensional</td>
<td>Modified Drucker-Prager/cap model used; finite element technique for solving</td>
</tr>
<tr>
<td>Zavaliangos et al. (2003)</td>
<td>Two - Dimensional</td>
<td>Porous plasticity model to examine roller (smooth) compaction and briquetting; used finite element</td>
</tr>
<tr>
<td>Wang and Karabin (1994)</td>
<td>Three - Dimensional</td>
<td>Porous plasticity model for hot rolling of porous sintered aluminum plates; Eulerian, velocity-based finite element model; “quasi 3-D” model since only one element is used in the thickness direction</td>
</tr>
<tr>
<td>Dec (1995)</td>
<td>Three - Dimensional</td>
<td>Based on slab method results but extended to three dimensions to investigate defects observed during rolling; not a true 3-D analysis</td>
</tr>
<tr>
<td>Pandheeradi et al. (2001) and Erickesen et al. (2001)</td>
<td>Three - dimensional</td>
<td>Analyses related to tape rolling of ceramic powders; uses finite element technique and Drucker-Prager/cap model for powder behavior</td>
</tr>
</tbody>
</table>

Johanson (1965(a), 1965(b)) made a number of significant contributions to the analysis and understanding of rolling of particulate material. Johanson combined the equations of equilibrium with the material models for the feed powder. He divided the regions of compaction into two: the feed region, where the slipping occurred at the roll surface, and nip region, where no slipping occurred at the roll surface. The demarcation of these two regions was defined by the nip angle. Overall, Johanson analyzed the stresses developed each of these regions using two different material models and determined the nip angle based on equating the mean stress gradient between the regions.
In the slip region, he analyzed the equilibrium equations using a pressure-dependent frictional model (Mohr-Coulomb type) and the boundary conditions related to the feed pressure and the frictional conditions at the roll surface. The resulting system of hyperbolic partial differential equations was solved numerically using the method of characteristics. In the nip region, it was assumed, that there is no slip between the material and the roll surface and all material trapped between the rolls at the position of nip angle must be compressed into a strip with the thickness equal to the roll gap. Johanson’s analysis provides two values of the roll pressure gradient at each point (one for slipping conditions and one for sticking conditions). Since determination of the pressure profile requires integration of this gradient from the entry angle forward, Johanson carried the integration by selecting the minimum gradient at each point in an upper bound type of argument. This selection effectively defines two regions: (a) $\alpha_{nip} < \alpha < \alpha_{entry}$ in which slipping occurs since the resulting pressure gradient from the analysis is smaller and (b) $0 < \alpha < \alpha_{nip}$ where the powder is assumed to stick to the rolls since the sticking analysis provides a lower gradient. A series of parametric studies was conducted demonstrating the effects of various parameters including internal friction of the powder, roll friction, gap to roll diameter and compressibility parameter of the powder (Johanson 1965(a) and 1965(b)). Several research groups have applied Johanson’s model including more recently, Yusof, et al., 2005 and Bindhumadhavan et al., 2005. These modeling approaches ignore the elastic unloading and the existence of an extrusion zone.

In 1973, Johanson refined his original continuum model to incorporate the influence of gas within the pores of the compacting powder by including the gas as a second phase and incorporating the continuity equation of the highly compressible gas
and applying Darcy’s law for fluid flow through porous media using an empirically-determined permeability coefficient (Johanson, 1973 and 1977).

Katashinskii (1966, 1977) and Katashinskii and Shtern (1983(a) and 1983(b)) have analyzed the roller compaction of metal powder. In their latest one dimensional continuum analysis, they developed the equilibrium equation based on the roll friction, normal stress acting in the rolling direction and the roll pressure for a slab element undergoing shear deformation and densification. The continuity equation accounted for the compressibility of the powder. The roll friction was based on Coulomb’s law in which the coefficient of friction varied with angular position under the rolls. A constitutive relationship based on the porosity and the yield stress of the individual metal particle was used to describe the stress-strain behavior. The system of equations developed were solved numerically based on the initial feed stress acting in the rolling direction, rolling angle, neutral angle, coefficient of friction, initial relative density of the powder, gap and roll diameter. It should be noted that the model depends on experimental inputs on the positions of the nip and neutral angles, i.e., the model does not predict these based on the other inputs.

In the mid-1970s, Musikhin (1977) developed a one-dimensional model for roller compaction following an analogous approach of cold rolling of continuous metals. Only the deformation in the nip is considered. It is assumed that as the particulate material enters this region it behaves as porous solid. The material is assumed to be slipping at all angular position except the neutral angle. The roll shear stress is assumed to be defined by the limiting (slipping) condition defined by Coulomb’s law. Musikhin uses a simple constitutive model for the material based on the difference between maximum and
minimum stresses, which is a modification of a model used for continuous metals. After substitution of the material model and boundary conditions for friction into the equilibrium equation, a differential expression for the roll pressure as function of the geometric, material and density considerations is defined. The differential equation is then solved for the two regions of lag (backward slip) and forward slip. It assumes that the principal stress $\sigma_3$ - assumed to be in the rolling direction - is zero at the bite angle and at the centerline of the rolls. The tapped density is assumed at the bite angle whereas the density at the centerline is provided to the equation. These parameters are supplied to the solution of the differential equations and the plotted with respect to the rolling direction. The point where the curves match represents the neutral plane. The shape of the roll pressure profile is not a smooth bell-shaped-like curve but instead is a combination of monotonic concave upward curves that meet at sharp point for the neutral position. Musiklin proceeded to manipulate the model parameter to match the maximum roll pressure of published data. The limitations to Musiklin’s one-dimensional model includes neglect of the tangential stresses; assumes only slippage occurs at the roll surface except at one point, which leads to a pressure profile that does not have a similar shape of measured profiles; and it relies on manipulation of multiple model parameters to attempt to match experimental data.

Shima and Yamada (1984) combined principles applied in the mechanics of powder compaction and developed an energy-based continuum model using the upper bound theorem to analyze the roller compaction of metal powders. Based on the upper bound theorem, the objective of the solution technique is to determine an admissible velocity field that minimizes energy dissipation due to densification of the powder during
rolling and to the friction at the roll/powder interface under specific boundary conditions. Shima and Yamada applied the yield criterion originally developed by Shima and Oyane (1976) for powders. The constant friction law indicates the frictional shear stress is constant and independent of the normal stress. The model initially assumes values for the gripping angle and velocity field and then, through an iterative process in which the rate of energy dissipation is minimized and the continuity equation is satisfied, the relative density distribution and strain rate distribution can be calculated. Finally using the strain rates and the constitutive model, the stress distributions are calculated. The authors indicated that powder/roll frictional conditions had little effect on the density and the thickness of the strip provided other conditions including the gripping angle could be kept constant. Additional results indicated that for a given strip thickness, the relative density and roll load decreased with roll speed and the relative density increased with flow rate.

Dec (1991(a) and (b)) developed a slab model for roller compaction using the concept of yield criterion for metal powders proposed by Kuhn and Downey (1971). The value of nip angle was taken from the experimental data and the initial stress conditions and density were assumed. The position of the neutral plane was also determined from experimental data. The compacted material density for each step of calculation was determined from compression test data for a corresponding mean stress. The calculation process for assumed conditions at the entry to the pressing zone was repeated until convergence with compacted strip density was achieved. The usage of experimentally measured values of nip angle and the forced matching with the output density limit the predictive capability of the model.
There has been limited two-dimensional analysis to date with notable exception of Gun et al. (1986), Cunningham and Zavaliangos (2002), Dec, et al., (2003), and Zavaliangos et al., (2003). Gun et al. developed a continuum model for the roller compaction of metal powders for plane deformation between smooth rolls. A porous plasticity model is used with ellipsoidal loading surfaces. The boundary value problem is solved using the Galerkin’s method. Gun et al. note variations in the thickness direction for the velocity and density fields. They also highlight the simulation results are in agreement with published experimental data for porous metals, however, opposite of the trends experimentally observed for iron powders, e.g., model predicts a slight non-uniformity through the thickness for density versus a 8 to 15% difference in density observed experimentally with iron powder. In addition the trend was opposite with the experimental data indicating the center was denser than the surface. Gun et al. attributed this to difference in the inlet conditions of the model (uniform velocity) and the experiments (densification in center region commences prior to material at the roll surface).

Cunningham and Zavaliangos (2002) and Dec et al. (2003) introduce the use of a modified Drucker-Prager/cap model to describe the powder behavior over the range of densities from the feeding to nip regions. The plane strain continuum models use the finite element method and examine the effects of the feed stress (constant and oscillatory), powder/roll friction and materials (calibrated for microcrystalline cellulose and dicalcium phosphate). The roll pressure, roll shear stress, nip and neutral angle and densification were calculated. Two-dimensional variations in the velocity fields were observed. The maximum roll pressure oscillated with the oscillation of the feed stress.
Qualitatively the trends followed experimental observations, however, detailed verification was not attempted. Zavaliangos et al. (2003) examined roller compaction and briquetting of coal using a porous plasticity model. The finite element technique was used. The simulations provided informative insight into the flow and densification in the cavity during briquetting. In addition, residual tensile stresses and possible causes for defect formation were discussed.

Additional process modeling efforts are required to improve the knowledge base of roller compaction. While experimental studies will continue to be a valuable to the optimization of a specific formulation and roller compaction operation, a predictive process model will enable the engineer or scientist to improve the design and control of the roller compaction, identify and select the key material properties that influence the process, determine process conditions that achieve the desired level of densification and ultimately reduce the number of experiments (and associated materials, time and effort) required to optimize and scale up the formulation and process. A predictive model of roller compaction, which must incorporate appropriate material properties (constitutive and frictional) based on experimental measurements over the relative ranges of relative densities and stress conditions, should be able assess the effects of material, geometrical and process conditions on the desired properties, e.g., level and spatial uniformity of the density of the compacted strips. The process models developed in this thesis were based on experimental assessment of the roller compaction process using specialized instrumented rolls to measure roll pressure and roll shear stress and experimental calibration of pressure-dependent, porous plasticity constitutive models and Coulomb frictional model for the roll/powder friction as inputs to the process models. The process
models included simple 1-D slab model and 2-D and 3-D finite element-based models that include less simplifying assumptions.
Chapter 3. Experimental Studies of Roller Compaction

3.1. Introduction

During the roller compaction of pharmaceutical powders, the following parameters are typically measured: screw speed, roll speed, roll force and roll gap. These parameters may be used in controlling the process either through direct single loop control or feedback control. A common mode of control for floating roll design roller compactors is to feedback the difference of the roll gap from a pre-set value to the screw motor, which compensates by increasing or decreasing the rate of feeding. The roll force is likewise maintained to a pre-set value through a process controller. While the process may be sufficiently controlled with these parameters, they do not provide detailed information on the stresses developed in the feed zone and between the rolls. Assessment of the feed forces at the entry and the contact stresses at the roll surface would provide valuable insight to the compaction between the rolls. In order to gain greater insight into the roller compaction process, detailed experimental studies were conducted using a roll compactor with three different instrumented roll designs capable of measuring the contact stresses at the roll surface. The overall research goals of the detailed experimental studies included:

- To design and construct an instrumented roll capable of measuring the normal and shear contact stresses at the roll surface;
- To conduct experimental studies on model pharmaceutical powder systems to assess the development of contact stresses between the rolls under varying roller compaction process conditions; and
To develop an understanding of the evolution of contact stresses in the slip, nip and release regions between the roll and the material properties, e.g., frictional properties, process conditions, e.g., roll gap and roll force, and equipment design, e.g., screw feeding.

3.2. Experimental

3.2.1. Materials

The feed materials used in the experimental runs included: microcrystalline cellulose (Avicel PH102, FMC); mannitol (Pearlitol 200DS, Roquette), and magnesium stearate (Mallinckrodt). Specific mixtures of these powders – ranging in scale between 1 to 5 kg - were prepared by blending in an appropriate sized V-shaped tumble blender (Patterson-Kelley). Blending time for the mixtures was 10 to 15 minutes. If magnesium stearate was added as a lubricant, the powder would be blended for 5 minutes.

3.2.2. Roller Compaction

When the original concept to instrument the surface of the rolls was developed in 1995, there was a limited number of small scale roller compactors commercially available in the pharmaceutical industry. In addition, the technical difficulties of adding the instrumentation and process controls led to the decision to design and build a laboratory scale roller compactor with the following capabilities:

- floating roll design with roll dimensions of 100 mm in diameter and 35 mm in width;
- vertical screw feeding system in which the screw speed and feed force were measured;
- measurement of roll speed, roll force, roll torque and roll gap;
- a versatile control system capable of running under single loop control for screw speed, roll speed, and roll force roller or with feedback control loops, e.g., gap control with gap error feeding back to screw motor to adjust screw speed or constant feed force feeding feed force back to screw motor; and
- an instrumented roll that measures the contact stresses under the roll at various locations.

Photographs of the roller compactor are provided in Figures 3.2.1 and 3.2.2. Over the course of this research, the instrumented roll underwent two significant design improvements. The original design included round load cell with a diameter of approximately 3.2 mm using a smooth surface. In improve the spatial resolution, a rectangular design capable of measuring approximately 0.7° of angular position was developed (Figure 3.2.3). The surface was also smooth. The final design was based on an instrumented roll by Schonert and Sander (2002) that enabled measurement of both normal and shear stresses at the roll (Figure 3.2.4). The last roll included a textured roll surface. The detailed design and construction of the roller compactor were completed by Roland Research Devices, Inc. (Trenton, NJ).

Two control schemes were used – feed force control and gap control. In both approaches, the roll force, roll speed and horizontal screw speed were controlled directly on a single loop to maintain the prescribed values. In the feed force control, a pre-set value of vertical feed force was maintained by feedback to the vertical screw motor, which adjusted the screw speed as required, e.g., if the measured feed force fell below the pre-set value, the vertical feed screw speed was increased to feed more material and
generate a higher feed force. In this mode, the gap was allowed to floats. It was observed, however, that constant feed force resulted in a constant gap. In gap control, which is the more common feedback control used with roller compactors with floating roll designs, the pre-defined gap was maintained by adjusted the vertical feed screw speed based on the measure gap distance.

The drive system for the rolls allowed variable rotational speeds between 0 and 10 rpm. The roll speed was set between 1 to 2 rpm depending on the specific experiment. The roll force was controlled to the predefined values through automatic adjustments in the valves of the hydraulic system. The roller compaction was capable of up to 35 kN of roll force. The controllers for the roller compactor were under PID control with the tuning parameters optimized for performance.
Figure 3.2.1. Overview of instrumented roller compactor with key components identified.
Figure 3.2.2. Close up view of the feed screw with feed force load cell and rolls including the instrumented roll on left.
ROLL SURFACE PRESSURE TRANSDUCERS
Locations and Dimensions

Figure 3.2.3. Schematic of roll design using rectangular load cell.
Figure 3.2.4. (a) Schematic of implementation design by Schonert and Sander (2002). (b) Side view of instrumented roll with roll load. (c) View of textured roll surface with five load cells across the roll.
3.3. Results and Discussion

3.3.1. Relationship of Roll Force, Feed Force and Densification

Figure 3.3.1 shows the relation between roll gap and roll force for three different levels of feed force for MCC. Clearly the roll force increases rapidly for decreasing gap as expected. Under force control, the increase of the feed force drives a wider roll gap to maintain the roll force. Equivalently for a fixed roll gap, increased feed force increases the roll force. At the same time the resulting density in the compacted strip depends primarily on the roll force and is only weakly affected by the feed force (at constant roll force). Similar results were obtained for MCC + 0.005% magnesium stearate and 50% MCC + 50% mannitol (see Table 3.3.1). Figure 3.3.3. shows the relative density, $RD$, versus the average maximum roll pressure (average between the various sensors) for the various combinations of process parameters. A one-to-one correlation is observed for each material. Note that a small percent of lubricant may have a strong effect on the rolled density, while the presence of a harder phase (mannitol) in the mixture decreases densification as expected.
Figure 3.3.1. Roll gap as a function of roll force for various feed forces for 100% microcrystalline cellulose.

Figure 3.3.2. Relative density as a function of roll force for various feed forces for 100% microcrystalline cellulose.
Table 3.3.1. Results of experimental studies for (a) 100% microcrystalline cellulose, (b) microcrystalline cellulose with 0.005% magnesium stearate and (c) 50% microcrystalline cellulose/50% mannitol.

<table>
<thead>
<tr>
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<th>Roll Gap</th>
<th>Average Maximum Roll Pressure</th>
<th>Relative Density</th>
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(a)

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<th>Roll Gap</th>
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<th>Relative Density</th>
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(b)
Table 3.3.1. (continued) Results of experimental studies for (a) 100% microcrystalline cellulose, (b) microcrystalline cellulose with 0.005% magnesium stearate and (c) 50% microcrystalline cellulose/50% mannitol.

<table>
<thead>
<tr>
<th>Vertical Feed Force</th>
<th>Roll Force</th>
<th>Roll Gap</th>
<th>Average Maximum Roll Pressure</th>
<th>Relative Density</th>
</tr>
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<td>(MPa)</td>
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<td>0.726</td>
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</table>

(c)
Figure 3.3.3. Relative density as function of the average maximum roll pressure for microcrystalline cellulose (MCC), 50% microcrystalline cellulose/50% mannitol and microcrystalline cellulose with 0.005% magnesium stearate. The individual values are from experiments with varying process conditions.
3.3.2. Determination of Nip Angle, Evolution of Surface Roll Pressure and Estimation of Feed Stress

The pressure profiles during the rolling of MCC are shown in Figure 3.3.4. As can be observed, roll pressure build up starting at approximately 13°, reaches a maximum at approximately 1° prior to the centerline of the rolls and rapidly decays upon exiting. The release angle is of the order of 3° (which is much larger that the width of the sensor). The contribution of the pressure past the minimum separation to the roll force is about 15 to 20%. In addition, the roll pressure varies along the roll width with the edges lower than the center. The nip angle was estimated for MCC by video taping the flow of MCC through the nip using a transparent side seal. Blue MCC markers were added to provide greater contrast with the white powder. The powder contacts the roll at approximately 23°. Based on the angular position at which the powder at the roll surface moves with the same velocity as the roll, the nip angle was identified. For MCC with a smooth roll, the nip angle was approximately 8°. These important angles with a roll pressure profile are shown in Figure 3.3.5. Finally, it is valuable to estimate the range of typical feed stresses on the powder since the feed stress is an important boundary condition for the process modeling discussed in Chapters 5 and 6. Based on measurement of feed forces in the range of 20 to 100 N and the cross-section area in the rolling direction as a function of the contact or entry angle, average feed stresses of approximately 0.025 to nearly 0.40 MPa for entry angles of 20 to 30° (Figure 3.3.6).
Figure 3.3.4. Example of MCC roll pressure profiles at various locations across width (#1 edge, 2-near center, 3- center, 4- near center and 5- edge). Feed force was 100 N and roll force was 3 kN.

Figure 3.3.5. Example of MCC roll pressure profile with entry angle and nip angle noted. Feed force was 100 N and roll force was 3 kN.
Figure 3.3.6. Calculated feed stress for varying feed forces and angles of application. The dashed rectangle indicated range of interest since the feed force is applied over a range of angular positions.

3.3.3. Determination of Roll Normal and Shear Stresses

To study the evolution of the roll pressure and roll shear stress the instrumented roll of Figure 3.2.4. was used. The powder was roller compacted under gap (0.800, 1.2 and 1.7 mm) and roll force of 12.5, 20.0, 27.5 and 35.0 kN. The roll compactor was operated under gap control in which the roll gap measurement was fed back to the screw motor to adjust the screw speed. The roll force and roll speed were directly controlled to maintain prescribed values. Microcrystalline cellulose with either 0.25% or 0.50% magnesium stearate was used.

Figure 3.3.7 provides a plot of the screw feed force, screw speed, roll force and roll gap versus time based on relative scale, i.e., percent of mean values of 52.6
N, 12.7 rpm, 12.5 kN and 1.204 mm, respectively. Both the roll force (relative standard deviation (RSD) of 0.79%) and roll gap (RSD = 2.13%) are tightly controlled during the course of the experimental run. In the gap control mode, the error in the actual roll gap to prescribed roll gap setting is fed back to the screw motor to adjust the speed screw. If the error is positive the motor reduced the speed and thus the amount of powder being fed to the rolls and the gap is reduced. Under steady state conditions, the feed screw speed is fluctuating over a relatively narrow band (RSD = 3.8%). Its periodicity is exactly matching its rotational speed. The corresponding feed force undergoes a wider range of variation (RSD = 12.3%) with a re-occurring pattern. There is a distinct difference in the periodicity of the feed force than the other signals. It is also noteworthy that the periodicity of the roll gap matches the screw rpm, which indicates a slight misalignment of the screw or unsymmetrical loading of the screw. Finally, the periodicity of the feed force is “double” – with one component that matches the screw rpm (small) and a slower one (larger). While it is uncertain what the larger component is related to, one possibility is the time lag between the feed force change (pre-densification) and the arrival of the material at the minimum gap.

An example run (roll force= 27.5 kN, roll speed = 4 rpm, roll gap = 1.20 mm for MCC with 0.50% magnesium stearate) is discussed in detail. A summary of select runs then follows. The profiles of roll pressure and roll shear stresses as a function of position along the roll width is provided in Figure 3.3.8. Several notable observations can be made upon examination of these profiles:

- The roll pressure slowly begin to rise at approximately 10° and reach a maximum at 0° before rapidly returning to zero at approximately 4° on the exit side. Unlike
the smooth roll where the maximum roll pressure occurred prior to the centerline of the rolls, maximum roll pressure appears to be at the centerline. However, it should be cautioned that the sampling rate and roll speed resulted in measurements every $1^\circ$ thus it is difficult to isolate the true maximum since the maximum likely occurs between 0 to $1^\circ$.

- Similar to the smooth roll, there is significant variation in the roll width in terms of position of maximum roll pressure. In this example the maximum roll pressure is off center. The position of the maximum roll pressure along the roll width depends on the distribution of the powder coming into the nip and the feed stress distribution along with the resistance related to the frictional side seals.

- The roll shear stresses gradually build on the entry side of the rolls with local maximum values of 14 to 24 MPa. The shear stress then reduces to zero at the neutral angle and proceed to reverse direction and build rapidly to very high values of 28 to 100 MPa on the exit side of the rolls at approximately $2^\circ$. The neutral angle ranges from 0 to 1.2$^\circ$.

The measurements represent the first time the roll shear stresses were measured for pharmaceutical powders. It is interesting the magnitude of these stresses is very high relatively compared to the shear and tensile strength of typical pharmaceutical compacts. It is possible that these high values of shear stresses are related to the roll texture.

The profiles of roll pressure, $p_r$, and roll shear stress, $t_r$, were used to calculate the corresponding roll force using the following equation:

$$ F_{roll} = \int \left[ (p_r \cos a) + (t_r \sin a) \right] dA $$

(3.1)
where \( a \) is the angular position under the rolls and \( dA \) is the differential area at the local force sensor. Using the local values of \( p_r(a,z) \) and \( t_r(a,z) \), angular position and dividing the area of roll surface in the vicinity of the individual load cells, the roll force was calculated to be 26.5 kN compared to the measured value of 27.4 kN, thus within 4% of the measured value. The relative contribution of the roll shear stress is only 3% so that assumption that the roll pressure contributed primarily to the roll force is valid.

Before leaving this example, it is worthwhile investigating the variation in roll pressure will rolling direction and the frictional conditions at the contact stresses at the roll surface. Contour plots of the local roll pressure with respect to the rolling width and rolling direction for five successive revolutions are shown in Figure 3.3.9. The location of maximum roll pressure varied from across the roll width on both sides of the middle (0 mm). The edges (+/- 17.5 mm) are the lowest likely due to the side seal friction and the lack of sufficient feeding from the screw to the rolls. It is interesting there is no apparent pattern of the movement from one side to the other side of the middle on successive roll revolutions, i.e., time 1, time 2, ..., time 5. Since the load cells are located at one angular position, the measurements represent essentially a snap shot. The movement of the maximum roll pressure is related to the oscillatory nature of the screw feeder. In this example in which the screw speed is continually adjusted as needed as part of the gap feedback control (Figure 3.3.7), the pattern for the pressure profiles is not necessarily repetitive. Potentially if the screw was maintained under constant speed, the pattern may be repetitive if the powder flow and distribution within the feed zone were consistent. There would be a phase shift between the rotating screw and rotating roll. As the powder contacts the roll, there is a transition from a circular cross section of the screw to
rectangular cross section of the rolls. The powder re-distribution across the plane normal
to the rolling direction is complicated and depends on the screw design and relative
position to the rolls. In addition, the frictional resistance at the side seal further reduces
the flow to the edges. These geometrical and boundary effects along with mechanical
behavior of low density powders contribute to mal-distribution of the powder in the feed
zone. These feed effects as also observed by Dec and Komarek (1993) and Guigon and
Simon (2002 and 2003) can directly manifest themselves in the local roll pressure
profiles developed and the local density distribution of the resulting compact.

The ratio of roll shear stress to roll pressure is plotted in Figure 3.3.10 as a
function of the rolling angle at various locations across the roll width. Overall, the ratio
is highly variable and quite large (>1.0) at 10 to 12° on the entry side and >3° on the exit.
As seen in Figure 3.3.10 values of the roll contact stresses are low, thus the ratio is
affected by the arithmetic division of two small values. At angle of 10° where the
contact stresses begin to rise, the ratios are in the range of 0.7 to 1.2. The ratio then
proceeds to monotonically decrease to zero at the neutral angle before increasing again to
large values on the exit side. Although this roll design allows measurement of the roll
shear stress, there is still insufficient information to know what is the coefficient of
roll/powder friction through the process. The Coulomb’s frictional law is often applied,
which simply provides the maximum ratio of shear or normal stress without failure or
slippage at the interface. In our experiment, whether there is slippage at the roll surface
is unknown because of the shape of the roll surface. The combination of the
measurement of roll pressure and roll shear stress along with knowledge of relative
movement at the interface can provide useful information on the frictional contact. Use
of video imaging as conducted earlier with the smooth roll and by Simon et al. (1999) can provide more information as to what is happening at the interface. In this case, the state of the contact conditions at the roll is further complicated surface texture that likely actively grips the powder. While the roll surface is clean, i.e., no adhesion of powder, upon exit thus suggesting the powder failures at the roll/powder interface, it is plausible that the slippage or failure occurs within the powder away from the roll surface. The failure stress would then be defined by the powder’s cohesion strength and angle of internal friction for the given relative density and stress state. It is unclear from the currently available experiments what is specifically occurring at the roll surface. If we assume that there is slippage up to 10°, the coefficient would be in the range of 0.6 to 2. If the powder failed away from the roll surface, these values would suggest internal angles of friction of approximately 30 to 50°. As the powder move deeper into the nip region, the powder will stick to the rolls and the ratio will thus be lower than the coefficient of friction. Regardless if it is the roll/powder interface or within the powder, the high ratio of roll shear stress to roll pressure indicate high effective friction with the texture roll that facilitate the rolling process. This can be viewed in contrast to the smooth roll in which the roll friction was very sensitive to amount of internal lubricant use and the handling of the roll surface, e.g., oils from hands indicated difference in local surface behavior.

Table 3.3.2. summarizes the average maximum roll pressure for various combinations of level of magnesium stearate as an internal lubricant, roll gap and roll force. The following trends can be noted:
- As the roll force increased for a given level of magnesium stearate and roll gap, the roll pressure increased.

- For a given roll force and magnesium stearate level, the roll pressure increases as the gap decreased.

- An increase in magnesium stearate level from 0.25 to 0.5% does not affect the development of maximum roll pressure.

Overall similar trends are observed with the texture rolls as the smooth roll. The textured roll appears less sensitive to the amount of internal lubricant.
Figure 3.3.7. Steady state values for screw feed force, screw speed, roll force and roll gap based on relative scale, i.e., percent of mean values of 52.6 N, 12.7 rpm, 12.5 kN and 1.204 mm, respectively.
Figure 3.3.8. Profiles of roll pressure and roll shear stress for various locations across the roll width. The middle of the roll along its width is designation of 0 mm.

Figure 3.3.9. Contours of roll pressure with respect to rolling directions and roll width.
Figure 3.3.10. The ratio of roll shear stress to roll pressure as a function of rolling angle for various locations across the roll width.
Table 3.3.2. Average maximum roll pressure for various combinations of internal lubricant/magnesium stearate (0.25 and 0.50 % weight); roll gap and roll force.

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3.4. Conclusions

The roller compaction of powders is a challenging process to analyze due to the complexities associated with: feeding of powder using a screw feeder, deformation of the powder under a range of stress conditions and contact conditions developed at the roll/powder interface. In effort to gain greater insight into the physical processes between the rolls, a series of experimental studies were conducted using a instrumented roller compactor. An instrumented roll was designed, constructed and used to study the effect of various process parameters and system response.

Key observations include:

- In a roll force controlled system, the exit $RD$ correlates with maximum roll pressure over range of gap settings studied.
- Increasing feed force increased roll gap for given roll force (floating roll design).
- Relative density was independent of roll gap (over range studied)
- Nip angle for smooth roll with MCC was 8 degrees.
- Neutral angle preceded centerline of rolls (smooth and textured rolls).
- Maximum roll pressure about 1 degree before centerline of rolls (smooth rolls) and near centerline (textured roll).
- Roll shear stress gradually builds on entrance side of roll, reverses at neutral angle and reaches relative high values at exit (possibly resulting in defect formation).
Significant variations in roll pressure were observed in both the rolling and roll width, which is related to oscillatory nature of screw feeder and to friction with the side seals.
Chapter 4. Materials Characterization

The experimental calibration procedures for the constitutive model and Coulomb friction model are outlined in this chapter. The calibration is discussed in the context of die compaction or tableting, which is a common powder compaction operation in the pharmaceutical industry. The modeling of die compaction is generally considered less complicated and thus permitted us an assessment of the calibration procedures and the use of the finite element method for modelling of pharmaceutical compaction operations. A discussion of the finite element modelling and validation is provided by Sinka et al. (2004).

4.1. Introduction

Tableting or die compaction represents one of the most important unit operations in the pharmaceutical industry because the physical and mechanical properties of the tablets, such as density and mechanical strength are often significantly affected by this process. The compaction properties of pharmaceutical formulations can be studied experimentally using a variety of techniques, ranging from instrumented production presses to compaction simulators, and methods of analysis. The results are usually plotted as porosity-axial stress functions (Paronen and Ilkka, 1996). In general the axial stress is determined by measuring the axial force, while the porosity is determined from measuring the tablet dimensions and mass or other methods, such as Helium pycnometry. Porosity-axial stress functions can be used for the comparison of materials. For example, greater slopes of the Heckel plot suggest a “greater degree of plasticity” in the material,
in other words it indicates ease of densification, which is related to the hardening characteristics of the powder.

This approach is deficient because it considers only the average stress along the direction of compaction, it ignores the radial stress transmission and friction, and is incapable of addressing the three dimensional character of the stress field and the density inhomogeneity that is present in shapes other than the low profile (i.e., low thickness to diameter ratio) flat tablets. This problem has been widely recognized in other industries that employ powder compaction operations (metals, ceramics etc.) (Trasorras et al., 1998). Over the last 15 years a comprehensive approach has been developed for the analysis of compaction using continuum mechanics principles. This approach is based on the following components:

- equilibrium equations (balance of forces transmitted through the material);
- continuity equation (conservation of mass);
- geometry of the problem;
- constitutive behavior of the powder (stress-strain behavior);
- boundary conditions including loading (e.g., displacement and velocity) and friction between the tooling and the powder; and
- initial conditions (e.g., initial relative density of powder).

Due to the significant non-linearity in material properties and contact stresses, a typical powder compaction problem cannot be solved analytically without major simplifications and thus a numerical approach is required. Today numerical modeling is possible due to the proliferation of powerful inexpensive computers and the existence of commercial finite element software. Therefore, one of the main tasks of a
researcher/engineer is to develop experimental inputs for such modeling efforts, e.g., the constitutive model for the material and die wall friction.

The modeling approaches commonly employed in compaction analysis are: (a) phenomenological continuum models, (b) micromechanically-based continuum models, and (c) discrete element models (e.g., Zavaliangos 2002). Although discrete element models offer a unique insight to the problem of compaction, they have extreme computational requirements (Zavaliangos 2003, Martin and Bouvard 2003). Phenomenological models with a connection to micromechanical models are the most efficient modeling techniques and have been successfully applied in a number of industrial compaction problems (e.g., see the review papers: Trasorras et al., 1998, Federzoni et al., 1999, Cocks, 2001, Zavaliangos, 2002, Ewsuk, 1997).

In the field of pharmaceutics, modeling of die compaction, i.e., tableting, may assist the development scientist or engineer to: i) understand the formulation and compositional effects on the compaction process including the axial loading and unloading along with ejection; ii) determine the stress distributions within the powder compact including the residual stresses; iii) to optimize the tablet tooling design; iv) estimate the density distributions within a tablet that may influence dissolution or mechanical properties; v) estimate the compaction force necessary to obtain tablets having given properties, vi) take into account the effect of the tablet material on the stress distribution on tooling to aid tool design, vii) assess the origin of defect or crack formation, and viii) optimize more complex compaction operations such as bi-layer and tri-layer tablets or compression coated tablets.
The overall goal of this research work is to demonstrate the validity of the continuum-based modeling approach to tablet compaction. The specific objectives of the present research are: i) to describe a simple, straightforward approach to calibrate a commonly used pressure-dependent, frictional plasticity model for microcrystalline cellulose using relative density as the internal state variable; and ii) to implement the elasto-plastic constitutive model and frictional properties into a finite element-based analysis of die compaction to predict density distributions. In this paper the relevant concepts of continuum models applied powder mechanics are reviewed and the general framework for compaction models is presented. A systematic methodology for the identification of material parameters for a linear elastic, modified Drucker-Prager/cap (Drucker et al., 1955, Drucker et al., 1957) model is also presented. With this information the development of a finite element model for density inhomogeneity in convex tablets was analyzed separately (Sinka, et al. (2004)).

4.2. Background

4.2.1. Powder as a mechanical continuum

When the powder is fed into the die, the packing of the particles is generally loose. The early stages of compaction are characterized by particle rearrangement, breakdown of the stable arches of particles and deformation or breaking of the granules for agglomerated powders. Further densification during compaction takes place due to mechanical interactions at the contact between neighboring particles. The nature of these interactions, which range from fragmentation to plastic deformation of the particles depends on the mechanical properties of the powder material. Microcrystalline cellulose,
for example, densifies through plastic deformation at the interparticle contacts. In ceramic materials, such as calcium phosphate, densification typically occurs as a result of crushing or fracturing of the particles.

Regardless of the nature of the powder, the size of the particles is usually more than two orders of magnitude smaller than the dimensions of typical tablets. A representative volume element of a sufficiently large number of particles can, therefore, be defined such that i) it represents properly the macroscopic response of the material and ii) its mechanical response is insensitive to the statistical variation present at the particle level. In this case the powder aggregate can be regarded as a continuum medium. Within this framework the specifics of the interaction between particles, such as cohesion and inter-particulate friction, are combined and included into density dependent material parameters of a macroscopic constitutive model that relates the average of stresses and strains over the representative volume.

Understanding continuum models requires familiarity with fundamental concepts of applied mechanics (Timoshenko and Goodier 1970, Lubliner 1998). In such models the stresses and strains are considered at the level of the representative volume element. Inside the representative volume element, stresses vary within individual particles and from particle to particle. Averaging out such quantities over the representative volume element essentially smooths out their variation to such a degree that a continuum representation is possible.

Within the continuum mechanics framework, powder compaction can be viewed as a forming event during which large irrecoverable deformation takes place as the state of the material changes from loose packing to near full density. From a continuum point
of view, the properties of the material evolve, e.g., the material stiffness increases during compaction. Therefore, the material properties at any one stage during compaction should be expressed as a function of some measures of its state, i.e., one or more state variables.

Although applications of continuum models are relatively new for compaction of pharmaceutical powders (Sinka et al., 2001, Sinka et al., 2002, Michrafy et al., 2002, Sinka et al., 2003) this modeling technique has been applied successfully in a very large variety of engineering problems with microstructure of characteristic dimensions of the order of 1-100 µm, (e.g., deformation processing of metals (Lia et al., 2001), mechanical properties of fiber reinforced composites (Tenek and Argyris, 1998), soil engineering (Rashidi and Arulanandan, 1995), powder metallurgy (Trasorras et al., 1998) and ceramics processing (Ewsuk, 1997)).

4.2.2. Elastoplastic Constitutive Models

Incremental theory of plasticity has been used successfully in describing the deformation of a wide range of materials including fully dense metals, soils and powders. In the case of materials that undergo microstructural changes during the deformation process, e.g., powder under die compaction, there are three components of the constitutive model, i.e., stress-strain, that need to be defined: i) the yield criterion, which defines the transition of elastic to plastic deformation; ii) the plastic flow potential, which dictates the relative amounts of each component of the plastic flow; and iii) the evolution the microstructure, which in turn defines the resistance to further deformation. In the section, these components are further discussed and described in the context of the constitutive model used in this analysis.
The total strain increment resulting from the application of stress consists of two components, a reversible and an irreversible one. It is assumed here that their decomposition is additive\(^1\).

\[
d\varepsilon_{ij} = d\varepsilon_{ij}^e + d\varepsilon_{ij}^{pl}
\]  

(4.1)

where subscripts \(i\) and \(j\) vary from 1 to 3 with reference to the coordinate system axes and indicate the three dimensional nature of the strain. Upon release of the external loads, the reversible (elastic) part of the strain is recovered. The relationship between macroscopic stress and elastic strain and their increments is approximated to the first order by a linear Hooke’s law (Timoshenko and Goodier, 1970):

\[
d\sigma_{ij} = \sum_{k,l=1..3} L_{ijkl} d\varepsilon_{kl}^e, \quad i=1..3, j=1..3
\]  

(4.2)

This is the generalization of the one dimensional form \(\sigma=Ec\) to a three dimensional form. In a simple isotropic case, the 81 (3x3x3x3) components of the elastic stiffness \(L_{ijkl}\) contain only 2 independent parameters (e.g., Young’s modulus and Poisson’s ratio).

The second term on the right hand side of equation (4.1) represents the irreversible (plastic) part of the deformation and reflects a variety of underlying irreversible mechanisms such as particle rearrangement, fragmentation, and plastic deformation at the particle level. The basis of a continuum model for compaction is the yield locus, i.e., a stress based criterion that defines the limits of elastic deformation in the form of an inequality:

\[\]

\(^1\) A multiplicative decomposition of the strain has been developed and is more accurate than the additive decomposition, but more complex. For this reason we present here only the additive decomposition of strain.
In the general case the yield function $F(\sigma_{ij}, km,...)$, which can be geometrically described as a surface in stress space, may be a function not only of the stress state but also internal state variables ($km, m=1...n$), that characterize all aspects of the material that affect yielding. When the yield condition is satisfied, a flow potential is postulated to exist and determines the three-dimensional character of plastic deformation.

$$d\varepsilon_{ij}^{pl} = d\lambda \frac{\partial G(\sigma_{ij}, km,...)}{\partial \sigma_{ij}}$$

The plastic flow potential can also be a function of the stress state, internal state variables, strain rates, deformation history and temperature. In principle a large number of parameters may be required to describe faithfully all physical and geometrical aspects of the material and its microstructure, we will confine our attention to a single internal state variable – the relative density. Although a framework that takes into account temperature and strain rate is available (Zavaliangos and Anand, 1993) we will not include such dependencies in this a first order approximation of the constitutive model.

Our goal is to present a simple yet rigorous approach that will minimize the number of experiments required for calibration of the model. For isotropic materials, the representation of the stress state can be simplified by use of stress invariants (Spencer 1971). The definition and a brief discussion on stress invariants are presented in the Appendix A. Here we will consider functional forms of the yield locus and the plastic potential that depend on two stress invariants, the hydrostatic pressure ($p$) and Mises...
equivalent stress \((q)\), which are associated with volumetric plastic strains and distortion, respectively:

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

For an isotropic material, i.e., for a material with properties that do not vary with direction, it is logical to represent the yield locus and flow potential as functions of these stress invariants\(^2\). In addition, the yield criterion is often affected by the changes in the structure of the powder compact. Plastic (irreversible) deformation leads to a permanent change in the structure of the compacted powder. Similar to research in powder metallurgy (Federzoni, et al., 1999), a simple scalar quantity is often used, namely the relative density \((RD)\) of the material, defined as the ratio between the density of the material at a given state and the density of the fully dense material. This selection implies that the elastic properties, the yield condition and the flow rule are function of the relative density.

\[
L_{ijkl}(RD), \quad F(q, p, RD) = 0 \quad \text{and} \quad G(q, p, RD) = 0
\]  

Equations (4.4.1), (4.4.2) And (4.4.4) are written in an incremental form because the state of the material evolves during compaction. The state of the microstructure of the compact determines the material response and must be tracked during the process. Its evolution is given on the basis of geometrical terms as (Gurson, 1977):

\[
d(RD) = -RD \sum_{i=1}^{3} d\varepsilon_{ii}^{pl}
\]  

\(^2\) It is assumed that the third invariant does not affect plastic deformation of porous materials (Mosbah et al., 1997).
The complete continuum mechanics constitutive model consists of equations (4.1), (4.2) and (4.4). The material properties (4.6) that are determined experimentally are: (a) the elastic constants, (b) the yield surface and (c) the plastic potential. The model is then completed with the evolution of the state variable according to equation (4.7), which represents the hardening rule.

Several types of constitutive models have been developed and applied in the analysis of compaction of metal and ceramic powders, including Gurson (Gurson, 1977), Cam-Clay (Schofield and Wroth, 1968), and Drucker-Prager/cap (Ducker et al., 1955, Drucker et al., 1957, DiMaggio and Sandler, 1971). The latter model, which is the one employed in current analysis, has gained more wide-spread use since it captures important aspects of the physics of compaction such as i) the fact that a compact is stronger in compression than shear and tension and ii) it includes commonly used material parameters such as cohesion, angle of internal friction and yield pressure, and their dependence with internal variables such as \( RD \). The detailed characteristics of the Drucker-Prager/cap model are discussed in the next section.

4.2.3. Linear Elasticity Model

Although early compaction models for metal powders (e.g., Kuhn and Downey, 1971) ignored elasticity, this aspect of material behavior is a necessary component in the model in order to understand ejection, during which most strains are elastic. In some soft macromolecular excipients the compaction stresses are relatively large with respect to the modulus and are the origin of the expansion of the tablet dimensions from die size after ejection.
In this work, the behavior of the compacting powder is assumed to be isotropic (same in all directions) and the elastic response to be linear\(^3\). Thus equation (4.2.) reduces to:

\[
\varepsilon_{ii} = \frac{\sigma_{ii}}{E} - \frac{\nu}{E} \sum_{j\neq i} \sigma_{ij} \quad \text{and} \quad \gamma_{ij} = (1 + \nu) \frac{\sigma_{ij}}{G}
\]  

(4.8)

where \(\varepsilon_{ii}\) and \(\gamma_{ij}\) are the normal and shear strains, respectively, \(E\) is the Young’s modulus and \(\nu\) is the Poisson’s ratio. The behavior of linear isotropic materials is described fully by these two elastic constants (e.g., Timoshenko and Goodier, 1970). Other elastic constants, such as the bulk modulus \((K)\) and shear modulus \((G)\), which describe the elastic behavior under hydrostatic pressure and shear stress, respectively, are related to Young’s modulus and Poisson’s ratio:

\[
K = \frac{E}{3(1-2\nu)}, \quad G = \frac{E}{2(1+\nu)}
\]  

(4.9)

Complete description of the elastic response necessitates that explicit dependence of Young’s modulus \(E(RD)\) and \(\nu(RD)\) Poisson’s ratio on the relative density is determined.

4.2.4. Modified Drucker-Prager/Cap Plasticity Model

The permanent deformation of powder compacts can be divided broadly in two modes. When the applied stress state is highly confining (such as in the later stages of die compaction) then the material densifies if the stresses exceed certain limit. When the

\footnote{\textsuperscript{3} In general the elastic behaviour of powder compacts is neither perfectly linear nor isotropic. Usually deviations from linearity are present at low relative density compacts, and anisotropy develops at high densities. Within the scope of this work, however, linearity and isotropy are considered to be reasonable assumptions.}
stress field is close to simple shear, the degree of confinement is low and the compact will exhibit shearing failure. These two types of behavior are represented in the form of the yield locus of the Drucker-Prager cap (DP/C) model.

In the following we adopt the DP/C model, which is the most accepted model at present in powder metallurgy and ceramic industries. This is a phenomenological model that has been adapted from soil mechanics. It is popular in compaction modeling because it contains features that are in accordance with the physical response of particulate compacts.

The DP/C model at low hydrostatic pressures is a shear failure model, similar to those used in granular flow, and reflects the dependence of the strength on the confining pressure. It predicts that the strength in tension is smaller than that in compression, a concept that is common for rocks, brittle materials and pressed powder compacts. In its simplest form it is represented by a straight line in the $p - q$ plane, which is also known as the Mohr-Coulomb shear failure line $F_S$:

$$F_S(q, p) = q - d - p \tan(\beta) = 0$$

(4.10)

The two parameters, are termed cohesion “$d$” and internal friction angle “$\beta$”. If the stress state is such that the corresponding Mises equivalent stress and hydrostatic pressure result in a value of $F(q, p) < 0$ then the stress causes only elastic deformation. If the stresses are such that equation (4.10) is satisfied, the material “fails” in shearing.

At high hydrostatic pressures the yield surface is described by a cap surface $F_C$:

$$F_C(q, p) = \sqrt{(p - p_a)^2 + \left[\frac{Rq}{1 + \alpha - \alpha / \cos \beta}\right]^2} - R(d + p_a \tan \beta) = 0$$

(4.11)
This form is consistent with the DP/C model implemented in the finite element package ABAQUS/Standard (Hibbitt et al., 1998). The parameters $p_a$ and $R$ are obtained from compaction experiments as described in the following section. The parameter $\alpha$ does not have a physical meaning but ensures a smooth transition between the cap and the shear failure regions. Typically a small value for $\alpha$ (0.01-0.05) is used to avoid the situation of $\alpha = 0$ for which a corner forms at the intersection of $F_c$ and $F_s$ which may lead to numerical problems (Hibbitt, et al., 1988). Because this is simply a numerical “trick” which affect the efficiency but not the accuracy of the computation, in the discussion that follows we will assume $\alpha = 0$. The geometric representation of the complete yield locus is represented in the $p-q$ plane as a limiting curve $F(q,p,R,D)=0$, see Figure 4.2.1(a).

In addition to the yield locus, the flow potential needs to be defined. It has proven experimentally and postulated theoretically that the functional forms of the yield locus and the flow potential coincide ($F = G$) when densification problems, i.e., in the cap region. This type of behavior is termed associated flow rule or associated plasticity (Hill, 1950).

$$G_c = F_c = \sqrt{(p - p_a)^2 + (Rq)^2} - R(d + p_a \tan \beta) \quad (4.12)$$

The plastic flow potential function in the shear region, $G_s$, is non-associated ($F_s \neq G_s$), and is given by:

$$G_s = \sqrt{[(p_a - p) \tan \beta]^2 + q^2} \quad (4.13)$$

The mathematical forms of $G_C$ and $G_s$ predict densification and porosity increase respectively in the corresponding domains of hydrostatic pressure above and below $p_a$. 
see Figure 4.2.1(a). The specific form of $G_s$ in equation (4.11) is such that it is defined completely by parameters that are present in the yield locus. Therefore the amount of characterization experiments needed is limited.

Both the yield surface and the flow potential are unique for a given level of relative density. Description of the complete behavior of the material at all relative densities requires a family of yield surfaces and flow potentials. The model is fully calibrated by expressing its parameters (cohesion, friction angle, size and position of cap surface) as function of the state of the material (i.e., relative density). As the compact densifies during compaction the local stresses on the material are always on the yield locus that corresponds to the current relative density. Since the material densifies the current yield surface becomes large (i.e., the compact hardens), see Figure 4.2.1(b). The limiting behavior as $RD \rightarrow 1$ should be consistent with the fully dense material behavior, i.e., no pressure dependence of the yield locus at high confining pressure (see. Sinka, et al., 2003 for more details).
Figure 4.2.1. (a) The Drucker-Prager/cap model parameters. (b) Family of DP/C yield limits for different levels of relative density over the whole range of compaction (data for microcrystalline cellulose), (c) family of DP/C yield limits for various relative densities at the early stages of compaction.
Figure 4.2.1. (continued) (a) The Drucker-Prager/cap model parameters. (b) Family of DP/C yield limits for different levels of relative density over the whole range of compaction (data for microcrystalline cellulose), (c) family of DP/C yield limits for various relative densities at the early stages of compaction.
4.3. **Model Calibration**

4.3.1. **Material**

Microcrystalline cellulose (MCC) grade Avicel PH102 (manufactured by FMC BioPolymer, Cork, Ireland) was used as a model powder in this study. MCC particles are irregular, with a nominal particle size of 100 µm and size distribution between 20-200 µm. The bulk and full density of the powder is 300 kg/m³ and 1520 kg/m³, respectively. Although this validation study is based on MCC, the experimental techniques presented here and numerical methodologies described in the companion paper are general and can be applied to any powder system.

4.3.2. **The experimental setup**

The calibration of the modified Drucker-Prager/cap model requires the following experiments:

- A die compaction in a fully instrumented die which will provide the material parameters for the cap yield surface, $F_C$, along with the elastic parameters and the coefficient of powder/die wall friction.

- A series of experiments at different relative densities at two levels of hydrostatic pressure each that will determine the shear failure surface $F_S$ (equation (4.10)). Example of such tests are: simple tension, pure shear, diametrical compression and simple compression where the $q/p$ ratios are: $-3$, infinity, $3\sqrt{12}/2$ and $3$, respectively (Procopio et al., 2003), see Figure 4.3.1. Any two of the above tests are sufficient to fully determine the terms of Equation (4.10). For practical purposes diametrical compression tests and simple compression tests were employed on powder compacts. The first
experiment is performed using a compaction simulator and a die instrumented with radial stress sensors. The compaction simulator is a single station press instrumented to monitor and control the pressing parameters. It accommodates dies and punches used on production presses. The powder is poured into a die and compacted between two punches to form a tablet. The displacements of the punches can be programmed to mimic the loading sequences and speed of virtually any kind of production press. Particularly useful for the early stages of formulation design and development, where there is usually only a small amount of material available, is that a limited number of tests yield considerable information. The experimental program described in this work is designed accordingly.

The die compaction experiment is carried out using a standard die with circular cross section having a nominal diameter of 9.525 mm. For each experiment 0.350 gram of microcrystalline cellulose was compacted by flat faced punches in the loading sequence for which the bottom punch is maintained stationary with respect to the die and the compaction is imposed by the movement of the top punch, (see Figure 4.3.2). The velocity of the top punch is constant, 1.0 mm/s during compression. The tablet is then ejected using the bottom punch.

The following parameters are measured during compaction:

- displacement of the top and bottom punches $U_T$ and $U_B$ with respect to the die table are measured using linear variable differential transformers (LVDTs). The difference of the two displacements provided the change in specimen height which in turn is connected with the current relative density

$$RD = RD_0 \frac{H_0}{H_0 - U_T + U_B} \quad (4.14)$$

where $RD_0$ and $H_0$ are the initial relative density and initial specimen height
respectively. The axial strain is then computed as:

\[ \varepsilon_z = \ln((H_0 - U_T + U_B)/H_0) \]  \hspace{2cm} (4.15)

- forces at the top and bottom punch, \( F_T \) and \( F_B \) are measured using load cells.

The stress at top and bottom punch are calculated by dividing the respective forces to the cross-sectional area of the die as follows:

\[ \sigma_T = -\frac{F_T}{A}, \quad \sigma_B = -\frac{F_B}{A} \]  \hspace{2cm} (4.16)

where: \( A = \pi D^2/4 \) is the specimen cross section area. The negative sign indicates compression.

- radial stress \( \sigma_r \) is measured directly using a piezoelectric sensor. The sensor is of 2.5 mm diameter and is mounted in the die wall in contact with the powder compact and are ground cylindrically so that disturbance to the compact is minimized.

The force and displacement data recorded during a typical experiment (Figure 4.3.3(a)) are converted to stress and strain measures as presented in Figure 4.3.3(b). For the pressing schedule considered here where the bottom punch is stationary, we have \( F_T > F_B \) due to the effect of friction between powder and die wall. Consequently the vertical stress distribution in the specimen is not uniform, \( \sigma_T > \sigma_B \). If the aspect ratio of the tablet is small the inhomogeneity is kept to a minimum.

Friction depends on the compacted powder, the die material, and the surface morphology of the die and is traditionally described by Coulomb’s law:

\[ \tau = \mu \sigma \]  \hspace{2cm} (4.17)
where $\tau$ and $\sigma$ represent the tangential and normal stresses on the powder/die interface, respectively and $\mu$ is the friction coefficient. For powder compaction, the coefficient of friction is not constant. A more detailed review of the factor influencing friction and the experimental facilities used to characterize it are presented elsewhere (Sinka et. al, 2001). Here we use a simple approximation that allows the determination of friction from the same data obtained from an instrumented compaction simulator. The height of the specimen is known at any one stage and the diameter and position of the sensor are fixed with respect to the bottom punch, which is maintained stationary. It is shown in Appendix B that the friction coefficient can be estimated by:

$$
\mu = \frac{D}{4H} \frac{\sigma_B}{\sigma_{rr}} \left( \frac{\sigma_T}{\sigma_B} \right)^{\frac{z}{H}} \ln \frac{\sigma_T}{\sigma_B}
$$

(4.18)

where $\sigma_T, \sigma_B, \sigma_{rr}, z, H$, and $D$ are the stress at the top and bottom punches, the radial stress read by the wall pressure sensor, the distance of the sensor from the top punch, height and diameter of the specimen. It is noted that the coefficient of friction decreases during compaction and its variation with the contact pressure is presented in Figure 4.3.4. Die wall lubrication was achieved by compressing a tablet of pure magnesium stearate in the die before the experiment. In can be observed that the coefficient of friction was reduced considerably. We will show below that for the specimen dimensions used in this work this level of friction does not introduce significant inhomogeneity in the samples.

4.3.3. Parameter Identification for an elasto-plastic DP/C model

The cap surface (equation (4.11)) is calibrated from the sequence of Mises equivalent stress and hydrostatic pressure for each relative density during die compaction...
with no wall friction. The loading path in effective stress – hydrostatic stress space is illustrated in Figure 4.2.1(a) using a dotted line. The final stress state “A” is on the yield surface. For this point the strain increment direction is also known because during die compaction the radial strain (or strain increment) is zero. Therefore the size and position of the ellipse that described the compaction surface can be obtained by solving the system of equations that describe the yield condition and plastic potential. Using a procedure that is explained in detail in Appendix A, we show that under low friction we can obtain the parameters \( p_a \) and \( R \) of the model using the following equations:

\[
\begin{align*}
p_a &= \frac{3q - 4d \tan(\beta) + \sqrt{9q^2 - 24d \tan(\beta)q - 24 \tan(\beta)^2 pq - 16 \tan(\beta)^2 q^2}}{4 \tan(\beta)^2} \\
R &= \sqrt{\frac{2}{3q}(p - p_a)}
\end{align*}
\]  

(4.19)  

(4.20)

where \( p = \frac{1}{3}(\sigma_{ax} + 2\sigma_{rad}) \), \( q = \sigma_{ax} - \sigma_{rad} \), and \( \sigma_{ax} = \frac{\sigma_T + \sigma_B}{2} \) and \( \sigma_{rad} \) is directly measured by the wall pressure sensor.

Although a single experiment on the instrumented compaction simulator can determine \( F_c \), in this work we produced a series of flat face tablets having various relative densities. For each level of relative density we conducted two tests: (a) a diametrical compression and (b) a simple compression experiment. The diametrical compression test, also known in the pharmaceutical industry as “hardness” test is carried out by subjecting a disk (i.e., a tablet with a low height to diameter aspect ratio) to compression across its diameter between two rigid platens. As a result, a tensile stress state develops in the center of the tablet, which will result in failure at a given load \( P \). Assuming that the material is linear elastic up to failure and that point contact between the tablet and platen
is maintained (i.e., the contacts do not “flatten”), the stress state in the tablet can be estimated analytically using the Hertz solution. The tensile strength of the material is given by:

\[ \sigma_T = \frac{2P}{\pi Dt} \]  

(4.21)

A more detailed examination of the application of diametrical compression tests, including the effect of specimen plasticity is presented elsewhere (Procopio et al., 2003).

Uniaxial tests are carried out by compressing a cylinder axially between two rigid platens. For this tests tablets having a larger height to diameter aspect ratio of between 1:1 and 2:1 to minimize end effects at the interface between the compact and loading platens. The uniaxial compressive strength is then calculated as:

\[ \sigma_c = \frac{F_c}{A} \]  

(4.22)

Using these two failure stresses, and the stress field at failure in each of these two experiments, the parameters \( d \) and \( \beta \) of the shear failure part of the DP/C model are obtain (see Appendix A, for derivation)

\[ d = \frac{\sigma_c \sigma_f (\sqrt{13} - 2)}{\sigma_c - 2\sigma_f} \text{ and } \beta = \tan^{-1}\left( \frac{3(\sigma_c - d)}{\sigma_c} \right) \]  

(4.23)
Figure 4.3.1. Simple test procedures for strength properties (1) uniaxial tension, (2) simple shear, (3) diametrical compression, (4) uniaxial compression.

Figure 4.3.2. Compaction simulator pressing sequence. The reference point is the top of the die table.
Figure 4.3.3. Compaction simulator data for the compression stage, (a) force – displacement at the top of the powders, (b) Stress – axial strain response of the powder
Figure 4.3.4. Variation of the coefficient of friction with normal stress for lubricated die.
4.4. Results and Discussion

Experimental identification of the material parameters was performed by extracting the data from compaction experiments in which the die was fully lubricated before compaction. The evolution of Young’s modulus and Poisson’s ratio are presented in Figure 4.4.1. It can be observed that Young’s modulus increases from very small values to around 10 GPa for the final relative density reached in the experiment. Young’s modulus increases rapidly towards the end of compaction as the deforming powder bed approaches full density. The elastic behavior of fully dense materials originates from the interactions at the atomic level. Therefore Young’s modulus and Poisson’s ratio have unique values for a single crystal, though they may be different in different directions (anisotropic). This directionality will average out for a powder particle that is a collection of a large number of randomly oriented crystalline grains, i.e., the particle is isotropic and has a unique modulus. If the powder particles are single crystals, when considering an aggregate of a large number of particles, the behavior is isotropic if the particles are randomly distributed. Random distribution is not always the rule. For example, if the compacted powder particles have a platelet morphology and are single crystals, then packing does not produce a random distribution of orientations. The particles tend to rest on their flat side and the powder compact may have an overall morphological texture and anisotropic mechanical properties. For the purpose of this work, however, we consider the material to be isotropic, which is a good approximation for MCC. In this case the two independent elastic properties (Young’s modulus and Poisson’s ratio) fully describe the elastic behavior. Elastic behavior is particularly important during unloading and ejection as it governs the elastic recovery of the tablets and influences the stress states that have a
potential of inducing defects in the tablets. The elastic properties are observed to change significantly during densification according to Figure 4.4.1. Thus, any attempt to model ejection must include the elastic properties as function of density.

It was observed that $\sigma_T < \sigma_c$ for all levels of relative density – this is consistent with the fact the shear failure line of the model has always a positive slope. For $RD = 0.3$, corresponding to a relatively loose packing of powders we have $\sigma_T = 0.083$ MPa and $\sigma_c = 0.152$ MPa, and the ratio between the compressive strength and tensile strength is 1.816. The strength in compression increases faster during densification, and the ratio $\sigma_c / \sigma_T$ increases to 61.6 for $RD = 0.90$. These values can be used in equation (4.23) to compute the cohesion and friction angle, are also function of the relative density as presented in Figure 4.4.2. A loose state powder aggregate has very little load bearing capacity. This is reflected by very small values of cohesion during the early stages of compaction. Similar to Young’s modulus, the cohesion increases rapidly towards the end of compaction. The friction angle for loose powders is around 40 degrees, value which is consistent with low pressure triaxial test data presented in the literature for similar materials (Li and Puri, 1996). As the material is densified towards $RD = 1$, the friction angle increases towards a value of around 70 degrees. Because the measured strength in simple compression is much higher than the cohesion of the material, this limiting value is approximated by:

$$\beta \approx \tan^{-1}\left(\frac{q_{sc}}{p_{sc}}\right) = \tan^{-1}(3) = 71.6^\circ$$
where \( q = |\sigma_c| \) and \( p = -\sigma_c / 3 \) are the equivalent stress and hydrostatic pressure in simple compression (see Appendix A). This value is consistent with similar measurements of (Coube and Riedel, 2000) and (Glass and Ewsuk, 1997) for metallic and ceramic powders respectively.

Figure 4.4.3. shows the evolution of the cap eccentricity parameter \( R \), and the hydrostatic yield stress \( p_h = p_a + R(d + p_a \tan(\beta)) \). Die compaction of low apparent density powders is characterized by large plastic deformations at very small stresses during the initial stage but the hydrostatic yield stress approaches infinity asymptotically at \( RD = 1 \). The corresponding families of Drucker-Prager cap surfaces in hydrostatic stress – effective stress space are presented in Figure 4.2.1(b) for various relative densities. When the density of the material is increased, the size of the yield surface expands in stress space. The curves are not self similar, as the cohesion, internal friction angle increase during compaction and there is also an evolution of the shape and size of the ellipse. For the early stages of the compaction (Figure 4.2.1(c)) the stress state corresponding to closed die compaction is positioned closer to the shear failure line. The results presented in Figure 4.2.1(b) give a condensed description of the plastic behavior of the material during compaction.

Overall the number of experiments required to calibrate the DP/C model is manageable and the required equipment is usually standard in a compaction laboratory. Compaction in an instrumented die, diametrical and uniaxial tests can even be performed on the same machine. Diametrical and uniaxial tests are simple and require minimal specimen preparation procedures. In all case care must be exercised for the proper selection of specimen sizes. For example – the calibration of the cap as well as
diametrical compression must be performed with low profile specimen to minimize inhomogeneities of density due to friction. Simple compression specimens need to be of an aspect ratio of ~1. Finally the diametrical compression test has been developed for brittle materials and the interpretation of the data can become difficult if significant plastic deformation takes place before fracture. Also the result even for simple compression exhibits some size dependence especially for more brittle materials (Doremus, et al., 2001)

The model presented here is not a first principles model. This may be considered as a weakness in the sense that there is no intrinsic aspect of the model that discriminates the behavior of materials that densifies by fragmentation vis-à-vis those compacting by plastic deformation of the interparticle contacts. The specific functional forms of the model parameters $p_a, R, d$, and $\beta$ versus relative density are different in the two classes of materials and this difference reflects albeit indirectly the underlying physical mechanisms.

The key advantage of the DP/C model is the explicit recognition of the three dimensional character of the strain and stress fields in compaction. Even in the simple geometry of a flat face cylindrical sample, the DP/C model recognizes the importance of the transverse stress. This stress in combination with wall friction determines the frictional forces on the surface of the compact which are responsible for the development of density inhomogeneity. Moreover the DP/C model recognizes that a large variety of stress states other than that of uniaxial strain (frictionless die compaction) maybe present in compaction of complex shape tablets. Above all, the DP/C model offers a complete
three dimensional form appropriate for implementation in a finite element code that can
analyze effectively general compaction problems (Sinka et al., 2004).

The powder/die wall friction is other essential experimental input to the finite
element model. Figure 4.3.4 presents the coefficient of friction as a function of the
contact pressure, i.e., radial stress, for the lubricated case. The coefficient is initially high
and decays to a limiting value, e.g., approximately 0.1. It is thus important to recognize
that the coefficient of friction changes during the compaction process. While the
coefficient of friction is plotted as a function of the normal pressure, i.e., the non-linear
Coulomb friction law, it is uncertain whether this dependency is directly related to
normal pressure or the relative density that is increasing during the compaction event.
Similar trends have been observed in the compaction of iron powder (Wikman et al.,
1997). Several experimental studies have been conducted to characterize friction for
powder compaction based on velocity (Sinka et. al., 2001, Wikman et al., 1997, Pavier
and Doremus, 1997), density (Pavier and Doremus, 1997, Wikman et al., 2000),
temperature (Turenne et al., 2000), and displacement (Pavier and Doremus, 1997).
Figure 4.4.1. Young’s modulus (a) and Poisson’s ratio (b) as functions of relative density.
Figure 4.4.2. Cohesion (a) and internal friction angle (b) as functions of relative density
Figure 4.4.3. Characteristics of cap surface, (a) cap eccentricity parameter, (b) hydrostatic yield stress ($p_h$)
4.5. Conclusions

The elasto-plastic constitutive model and the definition of the frictional contact conditions of the powder/die wall are two required experimental inputs to the modeling of die compaction or tableting of pharmaceutical powders. A linear elastic, modified Drucker-Prager/cap plasticity model was selected and calibrated for microcrystalline cellulose powder. The calibration procedures for estimating the elastic and plastic material parameters, $E$, $\nu$, $d$, $\beta$, $R$ and $p_b$ were described using simple mechanical tests - diametrical compression, simple compression and die compaction. These material parameters are found functions of the internal state variable, $RD$, which is directly related to the evolution of microstructure of the deforming powder bed. The resulting modified Drucker-Prager/cap model can be geometrically represented by a family of yield surfaces and plastic flow surfaces in hydrostatic pressure-Mises equivalent stress space described by the evolving material parameters. Each yield surface is composed of a Drucker-Prager shear failure line and an elliptical cap. In addition, the powder/die wall friction was estimated based on the die compaction of microcrystalline cellulose in an instrumented die. In a similar manner, the frictional coefficient was observed to change with densification. These experimental studies highlight that the material parameters do change with compaction and the use of constant material values, which is often applied, is not necessarily appropriate given the evolving microstructure of the deforming powder. The experimental characterization and accompanying analysis allows these material properties to be evaluated within the comprehensive framework of continuum mechanics, which can be useful in analyzing and predicting the effects of constitutive behavior,
friction, geometry, loading schedule and initial condition, e.g., initial relative density and powder fill configuration. The discussion is conducted using microcrystalline cellulose as model powder, however, the experimental procedures are applicable for any powder system in general.
Chapter 5. One-Dimensional Process Model

5.1. Introduction

The goal of the one-dimensional (1-D) modeling effort was to develop a simple continuum-based model of roller compaction to gain greater physical insight of the roller compaction process and to provide a basis for understanding the relative influences of the numerous parameters involved in roller compaction. In addition, it may guide more advanced modeling efforts without the significant investment of time and effort. More specifically, a series of parametric studies were conducted to investigate the effects of the roll friction, entry angle, feed stress, initial relative density, gap to roll diameter setting, and mechanical behavior of the feed powder on the development of internal and contact stresses, deformation and densification and position of the nip and neutral angles during the loading phase. A plane strain elastic analysis was conducted to assess the unloading. The material constitutive parameters were based on experiments on for microcrystalline cellulose discussed in Chapter 4.

5.2. Development of Slab Analysis

In this section a description of the slab method is provided including the assumptions; geometrical considerations; equilibrium equations (von Karman equation of rolling and force balances); equation of continuity; constitutive (stress-strain) relationship of the powder; boundary conditions (friction and feed stress) and initial conditions (initial relative density) used in the analysis of the of the loading stage of roller compaction. The algorithm of the solution technique for the slab analysis used in the rolling simulations
also presented. Finally, a simplified, one-dimensional elastic analysis of the unloading is described.

5.2.1. Slab Method and Assumptions

The slab method is a 1-D analysis technique that considers the equilibrium force balance on a thin slice, or slab, of material with a differential thickness. The slab technique has often been used to study the rolling, extrusion and drawing of continuous metals (Slater, 1977 and Dieter, 1986).

The basic steps in the slab analysis of rolling include (Hosford and Caddell, 1993 and Wagoner and Chenot, 1997): i) identification of the direction with the most significant variation in stress and strain, i.e., the rolling direction in a rolling process; ii) consideration of the equilibrium of a slab of material normal to the rolling direction; iii) derivation of the differential equation for the 1-D variation of stress; iv) introduction of the plasticity (constitutive) model to reduce the number of unknowns; v) application of the appropriate initial and boundary conditions; and vi) solution of the ordinary differential equation to determine stress and strain as a function of the rolling direction.

A number of assumptions is made in the current slab analysis of the roller compaction of powders including:
<table>
<thead>
<tr>
<th>Assumption</th>
<th>Comment</th>
</tr>
</thead>
<tbody>
<tr>
<td>One-dimensional variation in field quantities, e.g., stress and strain</td>
<td>stresses and strains only vary in the rolling direction and not in either the thickness or width directions</td>
</tr>
<tr>
<td>Deformation is considered plane strain, i.e., strain in the roll width direction is zero</td>
<td>thickness &lt;&lt; width and no lateral spread of powder in width direction</td>
</tr>
<tr>
<td>Homogeneous deformation state exists in the slab element</td>
<td>typically acceptable at low $\mu_{\text{roll}}$, $a_{\text{entry}}$, and $2h_o/D$</td>
</tr>
<tr>
<td>Constant friction coefficient following Coulomb’s friction law</td>
<td>simplifies analysis</td>
</tr>
<tr>
<td>Constant circular arc of contact</td>
<td>implies rolls are rigid for a specific prescribed entry angle</td>
</tr>
<tr>
<td>Negligible elastic deformation</td>
<td>plastic strains &gt;&gt; elastic strains</td>
</tr>
<tr>
<td>Constitutive model is porous plasticity</td>
<td>a rate-independent, pressure-dependent comprised of symmetrical ellipses in hydrostatic pressure-Mises effective stress space that expand with increasing relative density</td>
</tr>
<tr>
<td>Body forces can be ignored, e.g., gravity</td>
<td>gravitational forces are small relative to frictional and internal forces</td>
</tr>
<tr>
<td>Inertial effects are ignored</td>
<td>assumes process is sufficiently slow that the inertial forces are much less than the forces associate with the internal stresses developed in the powder, i.e., quasi-static process</td>
</tr>
</tbody>
</table>

5.2.2. Geometrical Considerations

A schematic of the rolls outlining the feed (slip) and nip (stick) regions is shown in Figure 5.2.1. Important geometrical parameters, i.e., roll radius ($R$), roll gap ($2h_o$), entry or delivery angle ($a_{\text{entry}}$), nip angle ($a_{\text{nip}}$) and neutral angle ($a_{\text{neutral}}$) along with the location at which the feed stress ($\sigma_o$) is imposed are noted. Typically the powder is fed to the rolls at a certain entry or delivery angle with or without a feed stress imposed. The powder initially experiences slipping at the roll surface, i.e., the roll moves faster than the adjacent powder, until it begins to stick at the nip angle. The powder then moves with the roll. Prior to release from the rolls the compacted powder may move faster than the roll.
roll and slip on the roll occurs again. The angle at which the frictional shear stress at the roll surface reverses direction is defined as the neutral angle. Within these regions, the powders undergo primarily plastic (irreversible deformation) deformation since the elastic modulus of the powder is much lower than the corresponding yield stresses (Cunningham, et al., 2004). The irreversible deformation of the powder may involve particle re-arrangement and plastic deformation or fragmentation of particles. During the unloading stage the powder experience primarily elastic recovery. Upon passing through the gap, the compact powder expands and is released at an angle noted as the release angle ($a_{\text{release}}$).

### 5.2.3. Equations of Equilibrium and Continuity

The stresses acting on an element in the roll gap on the entry side of the neutral plane is shown in Figure 5.2.2, where the roll pressure, $p_r$, and the roll shear stress, $t_r$, are acting at the roll surface defined by the angular increment $da$.

Based on equilibrium of forces the in the rolling direction, i.e., $x$-direction, for a unit length in the roll width direction, i.e., $z$-direction, and manipulation of the resulting equation the following expression results:

$$\frac{d(\sigma_x h)}{da} + R(p_r \sin a \pm t_r \cos a) = 0$$  \hspace{1cm} (5.1)
where ‘+’ is used when $a > a_{neutral}$ and ‘−’ is used when $a < a_{neutral}$. This expression is referred to as the von Karman equation of rolling (Dieter, 1986) and is independent of the material properties.

With the consideration of the equation of continuity, a fundamental difference between incompressible materials, e.g., a fully dense metal, and compressible materials, e.g., a powder, becomes apparent. The general form of the equation of continuity is given by $\frac{dm}{dt} = d(\rho V)/dt = 0$. For an incompressible material, the density is constant and the continuity equation can be re-written as follows:

$$\frac{dm}{dt} = \rho \frac{dV}{dt} = 0 \Rightarrow \frac{dV}{dt} = \frac{d\epsilon_x}{dt} + \frac{d\epsilon_y}{dt} = 0 \Rightarrow \frac{d\epsilon_x}{dt} = -\frac{d\epsilon_y}{dt} \quad (5.2)$$

For an incompressible material, since the through thickness strain is determined by the roll geometry, velocity along the rolling direction is fully determined by geometry. As a result the velocity of the slab continuously changes with position and it matches that of the roll at a single location – the neutral angle. Before that point the slab moves slower and slips with respect to the roll while after the neutral angle the slab moves faster, i.e., it is extruded by the rolls. However, for a compressible material, the equation of continuity can be written as follows:

$$\frac{dm}{dt} = \frac{d(\rho V)}{dt} = 0 \Rightarrow \frac{d\rho}{\rho dt} = -\left( \frac{d\epsilon_x}{dt} + \frac{d\epsilon_y}{dt} \right) \quad (5.3)$$
While the deformation of the incompressible material must be achieved with the strain rate in the \( x \)- and \( y \)-directions being of equal magnitude and opposite in direction, the deformation of the compressible material in the \( x \)- and \( y \)-directions may be accommodated with attendant changes in density.

The \( RD \) in the slip and nip regions can be obtained, respectively:

\[
RD(a) = RD_o \exp(-\varepsilon_{vol}), \quad a_{\text{entry}} \leq a \leq a_{\text{nip}} \tag{5.4}
\]

\[
RD(a) = \frac{RD(a_{\text{nip}}) \cdot \cos(a_{\text{nip}}) \cdot \left( ho + R - R \cdot \cos(a_{\text{nip}}) \right)}{\cos(a) \cdot \left( ho + R - R \cos(a_{\text{nip}}) \right)}, \quad a_{\text{nip}} \leq a \leq 0 \tag{5.5}
\]

In the slip region, the \( RD \) of the element is thus related to \( RD_o \) and the accumulated volumetric strain, which is determined by the constitutive behavior of the material. The relative density in the nip region is determined by \( RD(a_{\text{nip}}), \ a_{\text{nip}} \) and \( 2h_o/D \).

5.2.4. Material Models: Constitutive Behavior of the Powder and Friction

The analysis in this chapter was performed using a symmetric ellipse yield function and the normality condition. Mathematically, the yield function, \( F \), can be expressed by:

\[
F = A(RD)q^2 + B(RD)p^2 - 1 = 0 \tag{5.6}
\]

where \( A \) and \( B \) are coefficients, which are functions of \( RD \). \( q \) and \( p \) are defined in equation (4.5). In general any yield function can be used instead of (5.6).
The porous plasticity model was calibrated for microcrystalline cellulose (MCC) based on data from die compaction experiments using a die instrumented with radial stress sensors, according to the procedure outlined in Chapter 4. The yield surfaces are shown in Figure 5.2.3.

Based on the associated flow assumption and the normality principle (e.g., Lubliner, 1998), the incremental plastic strains can be determined based on the following equation:

\[
d\varepsilon_{ij}^{pl} = d\lambda \cdot \left( \frac{\partial F}{\partial \sigma_{ij}} \right)
\]  

(5.7)

where \(d\lambda\) is the non-negative plastic strain multiplier.

The densification rule for MCC is defined by the hydrostatic pressure versus volumetric plastic strain, which can be directly related to the relative density, i.e., \(RD = RD_0 \exp (-\varepsilon_{pl}^{vol})\). The hydrostatic yield pressure and corresponding relative density are plotted in Figure 5.2.5.

Coulomb friction law is used in the simulation:

\[t_r = \mu_{roll} \cdot p_r\]

(5.8)

where \(\mu_{roll}, p_r\) and \(t_r\) are the coefficient of friction of the roll/powder interface, roll pressure, and roll shear stress, respectively. This line represents the maximum allowable shear stress supported at the roll surface before slipping occurs at a specific roll pressure. Combinations of shear and normal stresses such that \(t_r < \mu_{roll} p_r\) are possible and correspond to sticking conditions.
5.2.5. Plane Strain Considerations

Plane strain deformation during rolling implies that the displacements of the slab element occur in the $x$-$y$ plane, i.e., rolling direction-thickness direction, and that the $z$-direction represents a principal direction, thus the shear stresses are also zero. Based on the assumption that the material is isotropic, i.e., the principal directions of stress and strain coincide, the plastic shear strain increments are also zero. Therefore, $\tau_{xz} = \tau_{yz} = 0$ and $d\varepsilon_{xz}^{pl} = d\gamma_{xz}^{pl} = d\gamma_{yz}^{pl} = 0$. Using the associated flow rule and normality principle (Lubliner, 1998), the plastic strain increment in the $z$-direction is given by:

$$d\varepsilon_{z}^{pl} = d\lambda \cdot \left( \frac{\partial F}{\partial \sigma_{z}} \right) = 0 \Rightarrow \frac{\partial F}{\partial \sigma_{z}} = 0$$  \hspace{1cm} (5.9)

Therefore, upon substitution of definitions of the effective stress and hydrostatic pressure into the porous plasticity model (equation 5.6) and differentiation with respect to $\sigma_{z}$, we obtain the following equation:

$$\sigma_{z} = \frac{1}{2} \left( \frac{9A(RD) - 2B(RD)}{9A(RD) + 2B(RD)} \right) (\sigma_{x} + \sigma_{y})$$  \hspace{1cm} (5.10)

The stress in the lateral, i.e., roll width, direction is a function of the stresses in the thickness and rolling directions along with the material parameters, $A(RD)$ and $B(RD)$. It is interesting note that in several of the previous analyses of roller compaction (Johanson (1965(a)); Katashinskii and Shtern (1983(a) and 1983(b)); and Dec and Komarek (1991)) involving the plane strain assumption, the stress in the roll width direction is assumed to be equal to $(\sigma_{z} + \sigma_{x})/2$ based on the assumption of incompressibility for a plastically deforming material for which the hydrostatic pressure is:
\[ p = \frac{(\sigma_x + \sigma_y + \sigma_z)}{3} = \frac{(\sigma_x + \sigma_y)}{2} \]  

(5.11)

Obviously this assumption is not valid for a compressible material like a powder. We have made this correction here by using (5.10).

5.2.6. Solution Algorithm

The iterative numerical solution was used to solve the system of equilibrium, continuity and yield equations. At every point the following equilibrium (equations 5.12 and 5.13), continuity (equation 5.14) and yield (equation 5.15) conditions are satisfied:

\[ \frac{\partial (h \sigma_x)}{\partial a} + 2R(p, \sin a \pm t, \cos a) = 0 \]  

(5.12)

\[ \sigma_y = -(p_r \mp t \tan a) - \tau_{xy} \tan a \]  

(5.13)

\[ \frac{d\varepsilon_x}{dt} + \frac{d\varepsilon_y}{dt} + \frac{d\rho}{\rho dt} = 0 \]  

(5.14)

\[ F(\sigma, p, RD) = 0 \]  

(5.15)

Four regions were identified for the element to be located under the roll – on entry or exit side of the neutral angle and either slipping or sticking. The neutral angle for a specific setting of simulation conditions, e.g., initial relative density, entry angle, feed stress and material properties, is determined by an iterative process that assures the stress in the rolling direction at the exit (centerline of the rolls) is zero. Whether sticking or slipping occurs is decided on which condition provides the minimum stress gradient of the stress in the rolling direction with respect to the angular position, \( d\sigma_x / da \). This is
similar to the approach used by Johanson (1965(a)) and it is a minimum upper bound type of argument (i.e., among two compatible strain fields which produce loads larger than the true loads, the one that produces the lower upper bound is selected). Johanson did not consider the possibility that slipping may occur close to the exit of the rolls. Here we select slipping or sticking based on the minimum stress gradient argument throughout the rolling zone. This suffices to pick up the slipping in the extrusion zone as discussed later in this chapter. Typically the powder undergoes slipping then sticking on the entry side followed by sticking on the exit side. This analysis is applicable up to the minimum roll separation position.

The roll force is the force between the centers of the rolls that acts to push the rolls apart, i.e., the roll separating force and can be determined from the stresses at the roll surface. Namely, it is the integral over the roll surface of the normal and shear stress projected onto the normal of the centerline direction:

$$F_{roll} = \int [(p, \cos a) \pm (t, \sin a)] \, dA$$

(5.16)

For a unit length in the roll width direction, the above equation can be expressed as:

$$F_{roll} = \int [(p, \cos a) \pm (t, \sin a)] \cdot (1) \, R \, da$$

(5.17)

where $R$ is the roll radius. This integral is numerically evaluated. ‘+’ is applied in the region prior to the neutral angle while ‘-’ is used after the neutral angle.
5.2.7. Elastic Analysis of Unloading

In traditional slab analysis elastic unloading at the roll exit is ignored. Because many pharmaceutical powders have rather low bulk modulus, elastic unloading must be taken into account. In an effort to assess the release angle and consider the effect of elastic unloading on the roll force, an elastic analysis was conducted. If it is assumed that all stresses and forces within the compacted strip from the centerline of the rolls to the release angle are due to elastic unloading, then one can estimate the additional contribution to roll pressure and the angle of release. Because the release angle is small, it is possible to assume that $t_r$ and $\sigma_c \approx 0$. Based on this assumption an estimate of the roll pressure from $a = 0^\circ$ to $a_{\text{release}}$ is given by:

$$p_r(a) = p_r(0) + \frac{E}{\nu^2 - 1} \ln \left[ \frac{h_o}{R \cos a - R - h_o} \right]$$

(5.18)

Thus the estimate of the roll pressure profile from the centerline of the rolls to the angle of release can be defined by the elastic parameters, the roll gap and roll radius and the roll pressure at the centerline of the rolls. The angular position at which the roll pressure reduces to zero corresponds to the angle of release, i.e., $p_r(a) = p_r(a_{\text{release}}) = 0$ in equation 5.18. The release angle, $a_{\text{release}}$, can therefore be analytically expressed as:

$$a_{\text{release}} = \arccos \left[ 1 - \frac{h_o}{R} \left( 1 - \exp \left( \frac{1 - \nu^2}{E} p_r(0) \right) \right) \right]$$

(5.19)
Figure 5.2.1. Schematic of feed and nip regions with relevant parameters.

Figure 5.2.2. Force balance on slab element during roller compaction.
Figure 5.2.3. Family of elliptical yield surfaces for microcrystalline cellulose using porous plasticity model. Each surface represents a common relative density. The outermost ellipse is for $RD = 1.0$. Each successive curve towards the origin is decremented in values of 0.05 for $RD$. The pressure is in the positive $x$-axis is compressive.

Figure 5.2.4. Yield surfaces for porous plasticity model with modified material parameters. The material parameters, $A(RD)$ and $B(RD)$, are modified by a constant, $\alpha$ or $\beta$. 

\[ F = \alpha A(RD)q + \beta B(RD)p - 1 = 0 \]
5.3. Results and Discussion

As discussed there are numerous factors that may influence the rolling process. These factors can interact with one another. In reviewing the results of the present series of simulations, categorization of the studied parameters into following four groups allowed more useful physical insight into the process:

i) powder/roll friction (coefficient of friction);

ii) roll gap and roll diameter;

iii) factors affecting feed state and conditions (entry or delivery angle, feed stress, and initial relative density in the feed zone); and

iv) material properties.

**Figure 5.2.5.** The densification characteristics of microcrystalline cellulose based on relative density versus hydrostatic pressure.
The simulations presented below are based on a relative density at the entry of $RD_o = 0.20$.

### 5.3.1. Roll/Powder Friction

Since friction draws the powder into the nip region, the friction coefficient at the roll/powder interface has a significant effect on the roller compaction process. As seen in Figure 5.3.1(a) and (b), the relative density at the exit progressively increases while the corresponding maximum roll pressure increases slowly and then rapidly as the coefficient of friction at the roll/powder interface increases. The rapid, non-linear rise in the maximum roll pressure observed as the frictional coefficient is increased is related to the densification characteristics of the microcrystalline cellulose, i.e., relative density (plastic volumetric strain) vs. hydrostatic yield pressure. As seen in Figure 5.2.5, the initial densification occurs with relatively small changes in the yield pressure while the latter densification at higher relative densities is accompanied with significant increases in yield pressure. When higher densification is achieved with higher frictional coefficients, the corresponding yield pressures dictate that higher stresses must be developed in order for the greater densification to occur.

It is interesting to note as can be observed in Figure 5.3.1, the increase in the maximum roll pressure and the corresponding increase in relative density at the exit can be limited by the entry or delivery angle. The nip angle versus coefficient of roll/powder friction is plotted in Figure 5.3.2. The nip angle increases with roll friction. In the case of the 15 and 20° entry angle, the powder sticks to the roll immediately upon entry when
the frictional coefficient is approximately 0.30 and 0.40, respectively. The level of final
densification is thus limited by the angle at which the powder is delivered to the rolls.
Since the powder immediately sticks to the rolls, its final density only depends on the roll
geometry (see equation 5.5). Thus any additional increase in roll/powder friction does
not increase the achievable densification. It is therefore advantageous to deliver the
powder at a sufficiently high delivery or entry angle such that the full beneficial effects of
the roll/powder friction can be achieved. For example, in the case where the entry angle
does not limit the nip angle, e.g., at 25°, the nip angle monotonically increases from
approximately 4 to 22° as fiction coefficients of 0.10 to 0.44.

To further examine the slip and nip (sticking) regions, a plot of the ratio of roll
shear stress to roll pressure versus rolling angle is shown in Figure 5.3.3 for an entry
angle of 25° and a friction coefficient of 0.40. The ratio is equal to 0.40 between angles
of 25° to approximately 20°. In this region the Coulomb slip condition is reached and
slipping is occurring at the roll, i.e., there is relative movement between the roll and the
powder in contact with the roll. Although the powder is moving toward the nip region,
the roll is moving faster than the powder in this region. The roll/powder interface is
“failing” with slippage occurring. The relationship between the roll pressure and roll
shear is thus defined by the prescribed coefficient of friction, i.e., \( t_r = \mu_{\text{roll}} p_r \). At the
angle below 20°, the ratio of roll shear stress to roll pressure is less than 0.40 and the
powder sticks to the roll surface. Once the powder is gripped, the roll shear is less than
the product of the coefficient of friction and the roll pressure, i.e., \( t_r < \mu_{\text{roll}} p_r \). In the nip
region, the ratio of roll shear stress to roll pressure monotonically decreases until it
reaches the neutral angle in which case the direction of the shear stress reverses. It then continues to decrease as it approaches the centerline of the rolls at the rolling angle of $0^\circ$.

For lower coefficients of friction, the nip angle is lower and the region of slipping is longer for given entry angle. The corresponding nip regions of sticking are consequently shorter. It has been incorrectly stated or assumed in some previous rolling analyses that the roll shear stress - throughout both the feed (slip) and nip (no slip) regions - is defined by the coefficient of friction (Katashinskii and Vinogradov, 1965(a); Musikhin, 1977).

The roll shear is defined by $\mu_{\text{roll}} p_r$ only in the slip region.

The roll pressure and shear stress profiles for various levels of friction coefficients are shown in Figures 5.3.4 and 5.3.5, respectively. As mentioned above the strongly non-linear effect of friction can be clearly seen in both figures. The raise in pressure is often taken to indicate the nip angle, e.g., a specific percent of the maximum pressure (e.g., Kashinskii and Vinogradov, 1965(b)) or a specific level of pressure (e.g., 2 MPa, Aksenov and Revyakin (1969(b)). This may lead to inconsistent results. Our analysis indicates that while for $\mu_{\text{roll}} = 0.20$ the nip angle occurs at a pressure that is 2.74% of the maximum pressure. The corresponding results for $\mu_{\text{roll}} = 0.44$ is 0.0003%.

Experimentally, the nip angle should be established by examining the relative velocity of the roll and the powder at the roll/powder interface, e.g., video imaging (Zega, et al., 1998) instead of the initial rise in roll pressure.

As can be observed in Figure 5.3.5, the roll shear stress reaches a maximum prior to the neutral angle. After this maximum the roll shear stress decreases until it is suddenly reversed direction at the neutral angle. Immediately following the neutral angle the roll
shear stress further decreases in magnitude as it moves to the exit. The gradient of roll shear stress with respect to rolling angle is higher as the frictional coefficient at the roll/powder interface increases. The values of roll shear stresses also increase with increasing friction coefficients.

Figure 5.3.5 also reveals the location of the neutral angle, which is the angle at which the shear stress at the roll reverses direction. Overall, the neutral angle is approximately 1° over the range of frictional conditions studied. The friction initially facilitates the movement of the powder with the roll upon entry and then resists the motion of the compacted strip upon the exit. The neutral angle is often defined by the location of the maximum roll pressure. This is based on the assumption that the location of the maximum roll pressure coincides with the neutral angle (e.g., Pietsch, 1987). This is not necessarily valid. Throughout the nip region, the equilibrium, continuity and yield conditions must all be satisfied. In the present slab analysis, the maximum roll pressure typically occurs at the centerline of the rolls, i.e., 0°, whereas the neutral angle is in the range of 0.5 to 1.5°. In experimental data in which the roll shear stress is measured, the reversal of the shear stress is prior to the centerline of the rolls (Chekmarev, et al., 1963; Katashinskii and Vinogradov, 1965 (a); Kuleshov, 1981; and Schonert and Sander, 2002). In some cases the maximum roll pressures appeared to coincide and in other cases it did not. In the case of experimental studies in which the roll pressure is measured, the location of the maximum roll pressure varies from the centerline to 1 to 2° prior to the centerline. In terms of the roller compaction model efforts, Johanson does not predict the neutral angle and the maximum roll pressure is at the centerline of the roll (Johanson,
1965(a)). This is related to the simplified material model used in the nip region. In the case of Kataskinskii and Vinogradov (1983) and Dec (1991(b)), the neutral angle is a model input rather than a model prediction.

Based on earlier studies by Chekamrev, et al. (1963) in which the normal and tangential roll stresses were measured and it was observed that the maximum roll pressure virtually coincided with the neutral angle, Mal’tsev (1971) estimated the neutral angles based on the angle at which the roll pressure is maximum.

Aksenov and Revyakin (1969(b)) defined the neutral angle as the angle at which the maximum roll pressure is achieved. The production of higher density strips coincides with lower neutral angles. These authors also observed a significant effect of the hardness and plasticity of the constituent particles. For powder with harder particles, the roll pressure profile had a higher and sharper increase in roll pressure that was located closer to the centerline of the rolls, e.g., approximately 1°. In the case of lead powder, which has soft particles, the roll pressure was lower and more rounded with a peak located at about 3°.

Kuleshov (1981) studied roll contact stress using a photoelastic technique in which lead powder was compacted between rolls containing optically sensitive elements. The tangential and normal roll stresses were measured. The normal stress increases to a maximum and then falls while the shear stress increases to a local maximum then decreases through zero and rises to a local negative maximum before falling back to zero at the exit. For the experimental conditions used the maximum normal roll pressures were 200 to 320 MPa. The corresponding shear stresses were much lower with the
positive maximum ranging from 2 to 6 MPa and a similar range on the negative side. The shape of the shear stresses is similar to the one predicted by the current slab method with the exception of the change from positive to negative shear stresses being less sudden compared to the slab method. The maximum roll pressure does not coincide with the zero roll shear stress. The absolute ratio of the shear to normal roll stress begins at about 0.5 and essentially decreases linearly to zero before rising again linearly. There is no experimental evidence indicating if the powder or compacted powder is sticking or slipping. Kuleshov, however, indicates that the region in which the ratio linearly decreasing and increasing is the expected region of sticking. It is noted that at no region is the ratio constant thus suggesting the coefficient of friction is “inevitably” linked to the changes in density of the powder during rolling. The assumption that the coefficient of friction is a constant over range of conditions and material states during rolling is indeed unlikely given the experimental data on die compaction and other frictional test, which show the frictional coefficient dependent on density and/or stress state of the material.

Due to the cost and complexity of instrumenting a roll, the roll surface pressures are not usually measured. However, the overall roll force is more commonly measured. The roll force is the force between the centers of the roll that acts to push the rolls apart, i.e., the roll separating force. The roll forces were calculated for a range of conditions in which the coefficient of friction varies. A plot of the roll force, normalized per unit cm of roll width, and relative density at the exit vs the coefficient of friction is shown in Figure 5.3.6. At low frictional coefficient in which the powder does not experience extensive densification, the roll force per unit length is low. The roll force per unit length
rapidly rises as the friction increases and the powder is able to densify to increasingly greater extent.

To further understand the development of the internal stresses within the deforming powder, it is useful to plot the stress path with respect to the yield surfaces as shown in Figure 5.3.7. In this example, the stress paths in the hydrostatic pressure or mean stress vs Mises effective stress \((p - q)\) space are plotted for the conditions in which the friction coefficient is 0.4. For reference the family of elliptical yield surfaces corresponding to various relative densities is also plotted. In addition, the stress path for die compaction is provided for comparison. As observed the two conditions follow the same path with the lower friction condition ending sooner, i.e., at a lower final relative density (approximately 0.65) as expected from the earlier results. The stress path of the higher friction condition approach the yield surface corresponding a relative density of 0.90. It is also interesting to note that the rolling process – as one would anticipate by consideration of the loading conditions in the two compaction operations– is predicted to exhibit lower a triaxiality stress state than die compaction. The stress path of the roller compaction is closer to the Mises effective stress axis, i.e., pure shear state, thus the powder is undergoing greater shear compared to die compaction. Die compaction has a higher triaxiality relative to rolling. The high shear conditions would be expected in rolling given the geometry of deformation compared to die compaction in which the powder is constrained by the essentially rigid die wall.
Figure 5.3.1. (a) Relative density and (b) maximum roll pressure vs coefficient of friction at roll/powder interface (constant) for varying entry angles (feed stress = 0, gap/roll diameter = 0.02, $RD_o = 0.225$).
Figure 5.3.2. Nip angle vs coefficient of friction at roll/powder interface (constant) for varying entry angles (feed stress = 0, gap/roll diameter = 0.02, $RD_o = 0.225$)

Figure 5.3.3. Ratio Roll Shear Stress to Roll Pressure vs angular position for Entry angle = 25°, Roll/Powder Friction = 0.4, Feed stress = 0, Gap/Roll Diameter = 0.02 and $RD_o=0.225$)
Figure 5.3.4. Roll pressure vs angular position for varying coefficients of friction. (Entry angle = 25°, Feed stress = 0, Gap/Roll Diameter = 0.02 and $RD_o$=0.225)

Figure 5.3.5. Roll shear stress vs angular position for varying coefficients of friction. (Entry angle = 25°, Feed stress = 0, Gap/Roll Diameter = 0.02 and $RD_o$= 0.225)
Figure 5.3.6. Roll force per unit roll width and relative density at exit vs coefficients of friction of roll/powder interface. (Entry angle = 25°, Feed stress = 0; Gap/Roll Diameter = 0.02, RD₀ = 0.225)

Figure 5.3.7. Stress path of roller compaction simulation and die compaction experimental data in hydrostatic pressure-Mises effective stress space.
5.3.2. *Roll Gap to Roll Diameter*

The effect of roll diameter (100 and 200 mm) was evaluated using varying roll gaps (1.00 to 6.00 mm) with an entry angle of 20°, initial relative density of 0.250, and roll/powder friction coefficient of 0.20. The nip angle, neutral angle maximum roll pressure and relative density at the exit are provided in Table 5.3.1. Overall, there is a slight decrease in the nip angle as the increasing roll gap-to-roll diameter. The neutral angle moves closer to centerline of the rolls as the roll gap decreases. The maximum roll pressure and corresponding relative density at the exit increased significantly as the roll gap-to-roll diameter decreases. Since the relative density is directly related to the volumetric plastic strains, the rapid rise in the maximum pressure is related to the densification of the powder. Once the feed material reaches the nip angle, the additional densification and associated build up of roll pressure are dependent on the geometry and mechanical behavior of the powders.

Analyses by other researchers likewise, show a similar dependency of the rolling parameters to this normalized ratio (Johanson, 1965(a)). This relationship is evident by simply considering Equation (5.5). Once the element reaches the nip angle, the volume of the element reduces in accordance to the geometry due to the no slip/sticking condition at the roll surface. For the relative density at the exit, \(RD(0)\), where \(a = 0\), the equation (5.5) reduces to the following in terms of the roll diameter, \(D\), and the total roll gap, \(2h_0\):

\[
RD(0) = RD(a_{nip}) \cdot \cos(a_{nip}) \cdot \left(1 + \left(D / 2h_0\right) - \left(D / 2h_0\right) \cdot \cos(a_{nip})\right) \tag{5.20}
\]
Thus if the nip angle and the relative density at the nip angle are the same for two different roll diameters, i.e., the feed conditions pre-densify the powder to the same level prior to the same nip angle, the same level of final densification will be achieved for a given ratio of gap to roll diameter. These conditions exist when the initial relative density, feed stress, entry angle and roll friction are equivalent for the combination of roll diameter and gap settings the results in the same gap to roll diameter ratio. In the simulations conducted, as the gap to roll diameter decreased from 0.03 to 0.01, the relative density at the nip angle and at the exit increased from 0.305 to 0.351 and from 0.457 to 0.927, respectively.

Table 5.3.1. Nip angle, neutral angle, maximum roll pressure and relative density at the exit for simulations at various roll gaps to roll diameter ratios.

<table>
<thead>
<tr>
<th>Gap/Diameter (-)</th>
<th>Nip Angle (°)</th>
<th>Neutral Angle (°)</th>
<th>Maximum Roll Pressure (MPa)</th>
<th>Relative Density at Exit (-)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.0100</td>
<td>10.54</td>
<td>0.68</td>
<td>124.6</td>
<td>0.927</td>
</tr>
<tr>
<td>0.0125</td>
<td>10.48</td>
<td>0.88</td>
<td>53.1</td>
<td>0.784</td>
</tr>
<tr>
<td>0.0150</td>
<td>10.42</td>
<td>1.00</td>
<td>33.2</td>
<td>0.689</td>
</tr>
<tr>
<td>0.0200</td>
<td>10.34</td>
<td>1.16</td>
<td>16.9</td>
<td>0.572</td>
</tr>
<tr>
<td>0.0250</td>
<td>10.24</td>
<td>1.24</td>
<td>9.75</td>
<td>0.503</td>
</tr>
<tr>
<td>0.0300</td>
<td>10.14</td>
<td>1.32</td>
<td>6.15</td>
<td>0.457</td>
</tr>
</tbody>
</table>

5.3.3. Feed Conditions

The feed conditions are known to be critical in the optimization of roller compaction processes. In this section, the effect of feed stress, entry angle and initial relative density in the feed zone are presented. In the delivery of the powder to the rolls,
the powder may undergo densification, i.e., the powder is pre-densified in the feed zone prior to the nip region. This pre-densification may be offset by escaping air flowing counter-current to the powder. The air has sufficient pore pressure to expand the bed of feed powder thus reducing its relative density. Ultimately, the powder will be delivered at a specific entry angle with a certain relative density and feed stress imposed by a gravity or a screw feeder.

The effect of feed stress on the maximum roll pressure and relative density at the exit can be examined in Figure 5.3.8 for various roll/powder friction conditions. Even small values of the feed stress result in significant increases in the maximum roll pressure and corresponding relative density at the exit. The nip angle was found to be not dependent on the feed stress over the range of conditions studied. The increasing feed stress increased the relative density at the nip angle highlighting final achievable densification is affected by the early densification before the nip angle.

Experimentally, Aksenov and Rebyakin (1969(a)) used an inflatable rubber bladder in the feed hopper to induce feed stresses in the range of 0 to 0.05 MPa on iron and nickel powders. The application of the relatively small feed stresses increased the maximum roll pressure achieved and the resulting density and thickness of the compacted strip. In the case of the nickel powder the application of 0.05 MPa increased maximum roll pressure from approximately 180 MPa to 450 MPa, the thickness from 1.2 to 1.8 mm and density from 4.84 g/cc to 5.85 g/cc. Given nickel has a much higher yield strength than microcrystalline cellulose (approximately 750 MPa versus 200 MPa), it can be expected
that every 0.01 MPa may make a difference for microcrystalline cellulose compared to 0.05 MPa for nickel.

In the next series of parametric studies, the effect of the entry angle of the feed powder on maximum roll pressure and relative density at the exit was analyzed. Plots of these parameters as function of entry angle for various frictional conditions are provided in Figure 5.3.9. Overall, as the entry angle increases from 10 to 30 °, the maximum roll pressure and corresponding relative density at the exit increase significantly. As shown in Figure 5.3.10, the nip angle, which highly dependent on the friction coefficient at the roll/powder interface, is independent of the entry angle. At low entry angle and high frictional conditions, e.g., at 10 ° for $\mu_{\text{roll}} = 0.20$ and 0.30 and at 15 ° for $\mu_{\text{roll}} = 0.30$, the powder, however, sticks to the roll immediately upon entry and thus the achievable densification is limited by the entry angle. The entry angle is thus an important consideration when designing a feed system for roller compaction. Pietsch (1987) highlighted the importance of having a adequately high entry or delivery angle. The definition of entry or delivery angle is straightforward for gravity feed systems, however for screw feeder system it is more difficult to delineate the entry angle since the tip of the feed screw often penetrates further into the feed zone than what may defined by the entry angle based on where the first contacts the roll.

At higher entry angle, the amount of densification that can occur in the slip region increases. This behavior can be illustrated in Figure 5.3.11, which is a plot of the evolution of relative density of the compacting powder during the rolling process. This graph shows the evolution of relative density versus the rolling angle for five different
entry angles, for a fixed initial density \((RD_o = 0.25)\) and friction coefficient \((\mu_{roll} = 0.3)\). As shown earlier (Figure 5.3.9) the nip angle for all cases is practically constant (15.5°). Recall that the densification past the nip angle is only dictated geometrically (Equation 5.5) and the final density is proportional to the relative density at the nip angle. For \(\alpha_{entry} < 15.5^\circ\) there is no slip region, i.e., the powder is gripped immediately by the rolls upon entry. For \(\alpha_{entry} < 15.5^\circ\), the amount of pre-densification that the powder receives in the slipping region increases with the entry angle. The importance of this pre-densification is evident because it has a pronounced effect on the final density and maximum roll pressure.

Tunderman and Singer (1969) studied the evolution of density between the rolls was experimentally evaluated for the case of rolling of iron powders in which a rolling experiment was stopped. A section of powder mass between the rolls in the compaction zone was removed and impregnated with an acrylic monomer, which was subsequently cured using ultraviolet light. This sample preparation allowed the powder compact to be polished. Image analysis was then used to analyze the densification through the compaction zone as function of the rolling angle. Although the materials and process conditions are different, similar trends were observed experimentally as is predicted in this slab analysis with a region of pre-densification and increasing densification to a maximum near the centerline of the roll. Tundermann and Singer note that the initial densification is related to the mechanical behavior of the iron and the compaction cycle at low relative densities in which the primary mode of densification is related to the restacking and reorientation of particle. They also highlight that the higher densification
is primarily caused by plastic deformation at inter-particle contacts and within the particles.

To examine the effect of the initial relative density in the feed zone, a series of parametric studies was conducted allowing the density at the entry angle to vary. The effect of the initial relative density of the feed powder on roll pressure and relative density at the exit can be observed in Figure 5.3.12. While initial relative density of the feed powder has little effect on the nip angle (Figure 5.3.13), the initial relative density of the feed powder has a dramatic effect on the maximum relative density and attendant relative density at the exit as shown in Figure 5.3.12.

The pre-densification of the feed powder within the feeder is critical to the resulting compaction under the rolls. This is further illustrated in Figure 5.3.14 in which the evolution of relative density under the rolls is plotted against the rolling angle for simulation in which the initial relative density varies between 0.200 to 0.300. As the initial relative density increased the relative amount of pre-densification increased from 0.5 to 22.3%. In addition, the final densification achieved increased substantially as the initial relative density increased.

Once the powder reaches the nip angle and then sticks to the roll, the evolution of the relative density is driven by the geometrical considerations. The relative density at the nip angle and the importance of the pre-densification in the slip region can be further illustrated in Figure 5.3.15. In this plot the exit relative density as a function of the nip angle is provided for varying values of relative density at nip angle. This plot can be generated based on Equation (5.5) in which a range of nip angles and relative densities at
the nip angle are substituted and the resulting relative density at the exit is calculated. As noted previously, the nip angle is primarily influenced by the frictional conditions between the powder and the roll surface. The relative density at the nip angle depends on the initial relative density of the powder as the powder enters the slip zone from the feeder and the pre-densification that occurs in the slip region. The initial relative density in turn depends on the densification that is induced during the conveying of the powder to the feed zone.

The feed stress, entry angle and initial relative density control the pre-densification that occurs within the slip zone prior to the nip angle. To further illustrate how these three factors influence the densification, a series of simulations were conducted. The base simulation included an entry angle of 25°, initial relative density of 0.250, feed stress of zero with a roll gap to roll diameter of 0.02 and roll/powder friction of 0.30. A plot of the stress in the rolling direction, $\sigma_x$, and relative density as a function of the rolling angle is plotted in Figure 5.3.16. $\sigma_x$, which at the entry angle represents the feed stress, gradually increases to a peak at approximately 2° and decays to zero at the exit. The relative density increases to a maximum of approximately 0.84 at the exit. Under the same gap and frictional settings, two additional simulations were conducted at 15 and 10° entry angles. Guided by the results of the base simulation, an initial density of 0.3208 and 0.4836 and a feed stress of 0.0025 and 0.109 MPa were used for the 15 and 10° entry, respectively. The resulting development of stress in the rolling direction and densification was the same as the base simulation. These simulations illustrate how the effect of entry angle, feed stress and relative density at the entry are interrelated and can
be used to optimize the design and process to achieve the desired final densification. The ability to pre-densify the powder is critical to the final densification. The use of higher entry angle, higher feed stress and relative density at the entry allows greater pre-densification going into the nip. It should be noted, however, the relative density at the entry is not necessarily a controllable parameter and is dependent on a number of factors including the material properties, e.g., ease of rearrangement and the densification characteristics at low stress states; the ability of the feed system to densify prior to delivery to the roll; the influence of the escaping air from the compacting powder, which may reduce the density; and the relative rates of feeding to drawing through the rolls, i.e., the ability to deliver sufficient mass of powder to the powder for a given roll speed and gap setting.
Figure 5.3.8. (a) Relative Density at the exit and (b) maximum roll pressure vs feed stress for various coefficients of friction at the roll/powder interface. $2ho/D = 0.020$, $\alpha_{entry} = 20^\circ$ and $RD_o = 0.250$
Figure 5.3.9. (a) Relative density at exit and (b) maximum roll pressure versus entry angle for various coefficients of friction at the roll/powder interface. $2h_o/D = 0.020$, $\sigma_o = 0$ MPa, and $RD_o = 0.250$. 
**Figure 5.3.10.** Nip angle versus entry angle for various coefficients of friction at the roll/powder interface. $2h_o/D = 0.020$, $\sigma_o = 0$ MPa and $RD_o = 0.250$

**Figure 5.3.11.** Evolution of relative density with rolling direction for various entry angles. $2h_o/D = 0.020$, $\sigma_o = 0$ MPa and $RD_o = 0.250$
Figure 5.3.12. (a) Relative density at the exit and (b) maximum roll pressure as function of the initial relative density for various coefficients of friction at roll/powder interface. $2h_o/D = 0.020$, $\sigma_o = 0$ MPa and Entry angle $= 20^\circ$
Figure 5.3.13. Nip angle versus of the initial relative density for various coefficients of friction at roll/powder interface. $2h_o/D = 0.020$, $\sigma_o = 0$ MPa and Entry angle = 20°

Figure 5.3.14. Evolution of relative density as function of rolling angle for various initial relative densities. $\mu_{roll} = 0.30$, $2h_o/D = 0.020$, $\sigma_o = 0$ MPa and Entry angle = 20°
Figure 5.3.15. Relative density at the exit for various nip angles and relative density at the nip angle.

Figure 5.3.16. Evolution of stress in rolling direction and relative density as function of rolling angle for various combination of initial relative densities and feed stresses.
5.3.4. Relationship between Maximum Roll Pressure and Densification

Figure 5.3.176 shows the variation of the relative density at the exit of the roll versus the maximum roll pressure for a large variety of friction, gap to roll diameter, entry angle, feed stress and initial relative density. On the same graph the plane stress solution \(\varepsilon_{33}^{pl} = 0, \sigma_{11} = 0\) for the exit is also plotted. It is noteworthy that all the simulation data collapse onto one curve, which is identical to that of the plane strain solution. This is similar to the experimental observations of Katahinskii and Vinogradov, 1965 (b); Aksenov and Revyakin, 1969(b); and Mal’tsev, 1971, and is not unexpected. With no variation through the thickness, the stress state at the exit is that of plane strain. Given that \(p_r^{max}\) occurs very close to the minimum separation of the rolls, \(p_r^{max}\) is an excellent approximation of the vertical stress at the exit.

If conditions are such that the assumptions of the slab analysis are valid then the plane strain approximation is acceptable. For these conditions to be valid, \(2h_o/D \ll 1\) and \(a_{entry}\) should be small. It is noteworthy that in early work (e.g., Johanson, 1965(a)) the die compaction data are directly introduced into the rolling model without modification. In principle this is incorrect. Die compaction and roller compaction stress paths are shown in Figure 5.3.7 and are clearly different with roller compaction exhibiting higher shear than die compaction.

A direct comparison of the vertical stress in plane strain compression versus the axial stress in die compaction shows that the latter is larger numerically (Katahinskii and Vinogradov, 1965(b) and Mal’tsev, 1971) who claimed that roller compaction is more “efficient” than die compaction. In any event the difference between these two stresses is
relatively small at low density (e.g., 2% at $RD = 0.60$ for MCC) but increases as $RD$ approaches 1.0 (e.g., 14% at $RD = 0.90$ for MCC). Therefore the assumption that a common roll pressure in roller compaction and die compaction will yield the same densification should be used with caution.

**Figure 5.3.17.** Maximum relative density versus maximum roll pressure for range of friction, gap, feed stress, entry angle and initial relative densities.
5.3.5. **Influence of Constitutive Behavior**

To examine the effect of material behavior on the roller compaction process, the material parameters of the porous plasticity model, $A(RD)$ and $B(RD)$, were varied from its base behavior representing microcrystalline cellulose by adjusting $A(RD)$ by multiplying by either 0.5 ($\alpha = 0.5$ and $\beta = 1.0$) or 5.0 ($\alpha = 5.0$ and $\beta = 1.0$) in the equation presented in Figure 5.2.4. The parameter $A(RD)$ is associated with the shear strength of the material and with the hydrostatic pressure equal to zero, $A(RD)$ represents the inverse of the square root of the cohesive strength, i.e., shear strength under pure shear conditions, for the powder at the specific relative density. In addition, the shape of the yield surface changes as shown in Figure 5.2.4. Modifying these parameters in this manner provides a simple way to evaluate the effect of the material properties. It should be noted that the hydrostatic yield pressure – volumetric strain, remain unchanged.

For the series of simulations in which $2h_o/D = 0.02$, $\sigma_0 = 0$ MPa, $\alpha_{entry} = 20^\circ$ and $RDo = 0.250$, the nip angle, the relative density at the exit and corresponding maximum roll pressure are presented in Table 5.3.2 for varying roll/powder frictional coefficients. As the friction increases the densification and maximum roll pressure increase. While the nip angle, with the exception of $\mu_{roll} = 0.1$ for $A(RD) = 5.0$, is essentially the same, the level of densification and maximum pressure is, however, quite different. The differences in degree of densification diminishes as the friction increases with the nip angle being the same for the simulations for $\mu_{roll} = 0.2$ and 0.3. At $\mu_{roll} = 0.30$, the exit density is in fact the same. There is sufficient friction at the roll surface such that the material is immediately sticks to the roll and the densification is thus determined by the
geometrical considerations. In terms of the achievable densification, the material differences evaluated become less influential as the roll friction increases. The maximum roll pressure required, however, to achieve the level of similar level densification is quite different, as seen in the case of the $\mu_{\text{roll}} = 0.30$ in which case the maximum roll pressure varies from approximately 70 to 180 MPa. A plot of the exit relative density versus maximum roll pressure, as shown in Figure 5.3.18, the plots are different for the different materials. This further highlights the significant effect of the material properties on the roll compaction processes.

To examine these effects further the stress path of the three materials are plotted in Figure 5.3.19. The materials with the decreasing value of the parameter $A(RD)$ experience lower triaxiality, i.e., high shear relative to hydrostatic pressure. The stress path initially for all three materials is similar but once the material is gripped and sticks to the roll surface, the stress path is dictated by satisfying the geometrical considerations and the yield condition and the normality principle. The material with a yield locus with the more concave shape is driven to lower triaxiality in effort to satisfy the degree of volume reduction required to maintain the no slip condition in the nip region. If the path was higher in triaxiality, i.e., closer to the hydrostatic pressure axis, the level of volume reduction would be higher than would be required to satisfy the sticking condition. When the yield surface is less concave, the stress path must undergo higher triaxiality.
Table 5.3.2. The nip angle, the relative density at the exit and corresponding maximum roll pressure are for varying values of $A$ and $B$ and roll/powder frictional coefficients. ($2h_0/D = 0.020$, $\sigma_o = 0$ MPa, $\alpha_{entry} = 20^\circ$ and $RD_o = 0.250$)

<table>
<thead>
<tr>
<th>$A$</th>
<th>$0.5$</th>
<th>$1.0$</th>
<th>$5.0$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$B$</td>
<td>$0.2$</td>
<td>$0.3$</td>
<td>$1.0$</td>
</tr>
<tr>
<td>$\mu_r$</td>
<td>0.1</td>
<td>0.2</td>
<td>0.3</td>
</tr>
<tr>
<td>$\alpha_{nip}$ ($^\circ$)</td>
<td>4.8</td>
<td>15.4</td>
<td>20</td>
</tr>
<tr>
<td>$RD_{exit}$ (-)</td>
<td>0.747</td>
<td>0.810</td>
<td>0.943</td>
</tr>
<tr>
<td>$p_r^{max}$ (MPa)</td>
<td>56.0</td>
<td>77.4</td>
<td>180.6</td>
</tr>
</tbody>
</table>

Figure 5.3.18. Maximum relative density versus maximum roll pressure for material models. $2h_o/D = 0.020$, $\sigma_o = 0$ MPa, $\alpha_{entry} = 20^\circ$ and $RD_o = 0.250$
Figure 5.3.19. Stress paths of hydrostatic pressure – Mises effective stress space for various materials. \( 2ho/D = 0.020 \), \( \sigma_0 = 0 \) MPa, \( \alpha_{entry} = 20^\circ \) and \( RD_0 = 0.250 \)

5.3.6. Elastic Unloading

During the loading stages of roller compaction the current slab analysis assumes that the powder is undergoing plastic deformation only, i.e., the elastic deformation is ignored. This is a reasonable first approximation considering the yield stresses are significantly lower than the elastic moduli for the powder at a specific relative density (Cunningham, et al., 2001). During unloading, however, in which the stress and strain are due primarily to elastic behavior, it is important to consider the contribution of the elastic behavior. As indicated in equation (5.18) the unloading portion of the roll
pressure profile is influenced by the elastic properties of the material (Young’s modulus, $E$, and Poisson’s ratio, $\nu$), the gap setting and the maximum roll pressure. The effect of these parameters on the release angle is evaluated analytically using equation (5.19). In addition, the roll pressure profile during unloading and the contribution of the elastic unloading on the roll separating force are discussed.

A plot of the release angle versus the roll gap to diameter is presented in Figure 5.3.20 for various values of the Young’s modulus. The Poisson’s ratio, which causes small changes to the release angle over range of $\nu = 0.1$ to $0.35$, was held at $0.25$. It can be seen that as the roll gap to roll diameter increases the release angle also increases. In addition, as the Young’s modulus decreases, the amount of expansion and consequently the release angle increases. Figure 5.3.21 reveals the effect of maximum roll pressure on release angle. Assuming the elastic properties are the same, as the maximum roll pressure increases the angle of release increases. This increase is also a function of the roll gap to roll diameter as shown in Figure 5.3.21. It should be noted that as the roll pressure increases during roller compaction, the density of the corresponding compacted strip increases and thus the Young’s modulus and Poisson’s ratio typically increase thus these parameters are not independently controlled. The gap setting, however, may be adjusted more independently to allow a range of densification and thickness. For a given level of densification, a thicker compacted strip will have a higher release angle and greater expansion upon release.

To examine the roll pressure profile more closely, equation (5.18) was used to determine the unloading portion of the pressure profile for the simulation in which the
roll diameter is 100 mm, roll gap is 2 mm, the initial relative density is 0.225, and the powder/roll friction coefficient is 0.44. Estimated elastic parameters for microcrystalline cellulose at the relative density corresponding to the relative density achieved at the centerline of the rolls for the specific simulation were used. The pressure profile is shown in Figure 5.3.221. Similar to experimentally measured roll pressure profiles, an asymmetric profile is observed in which there is gradual build up of roll pressure followed by more rapid increase related to the densification properties of the densifying powder. Upon reaching the maximum value, the roll pressure rapidly decreases to zero over 2.3° upon unloading.

The slab analysis conducted ignored the contribution of the elastic unloading to the overall roll force. To assess how significant this contribution, the roll force for elastic unloading was calculated using equation (5.17). The contribution of the roll shear stress was ignored since the component of roll shear stress that contributes to the roll force is small compared to the roll pressure component. Using the earlier example, the total roll force, which includes the plastic loading and the elastic unloading portions of the pressure profile, was 11.6 kN/cm roll width. The components associated with plastic and elastic deformation were 8.6 and 3.0 kN/cm, respectively. Thus the elastic contribution to the overall roll force in this case was significant at about 25% of the total. It should therefore be noted that the roll force calculated by the current slab method underestimates the true roll force since the analysis does not account for the elastic unloading. This simple observation highlights concern with research publications that claim good agreement between experimental data and slab predictions without taking elasticity into
account. It is also worth noting that the unloading elastic behavior of partially compacted powders is highly non-linear (Procopio, 2006). Therefore, ever higher values of the release angle may be present than those predicted by simple linear analysis presented here.

![Graph showing release angle versus roll gap to roll diameter for material with E = 3, 5 and 10 GPa.](image)

**Figure 5.3.20.** Release angle versus roll gap to roll diameter for material with \( E = 3, 5 \) and 10 GPa.  (\( \nu = 0.25; p_r (0) = 100 \text{ MPa} \))
Figure 5.3.21. Release angle versus maximum roll pressure for roll gap to roll diameters of 0.01, 0.02 and 0.03. \( (E = 5 \text{ GPa}; \nu = 0.25) \)

Figure 5.3.22. Pressure profile including elastic unloading. \( \mu_{\text{roll}} = 0.44, 2h_o/D = 0.02, \sigma_o = 0 \text{ MPa}, \alpha_{\text{entry}} = 20^\circ \) and \( RD_o = 0.250 \)
5.4. **Conclusions**

A 1-D model of roller compaction was developed based on the slab method in which the equations of equilibrium and continuity and the conditions of yield were satisfied throughout the slip (backward and forward) and nip regions. The powder was modeled throughout the slip and nip regions using the same constitutive behavior with yield surfaces defined by $RD$-dependent material parameters. A plane strain assumption was used with the transverse stress calculated based on satisfying the yield and normality condition – instead of applying the common incorrect assumption that the transverse stress is one-half the sum of the other two normal stresses. The final densification of a given powder is closely associated with the maximum roll pressure. The maximum pressure developed was highly dependent on the densification that occurs in the slip region, the position of the nip angle and the gap to diameter setting. The densification of the powder in the slip region is dependent on the entry angle, initial relative density, feed stress, and friction at the roll surface. In order to achieve higher final densification, it is essential the powder is sufficiently pre-densified prior to the nip angle. This pre-densification is affected by the equipment design and process conditions. Several combinations of the parameters studied can result in similar densification. This highlights the need for caution in attempting to verify models for roller compaction. It is essential to have the appropriate model inputs to independently verify the predictive capabilities of a model. Currently, additional experimental efforts are needed to measure the relative density of the powder in the feed zone and the stress applied by the feed
system such that relevant initial and boundary conditions can be applied. For the unloading, a simplified elastic model was developed to predict the angle of release and elastic expansion of the compacted material. In the calculation of the overall roll force, it is important to consider the elastic unloading, which can contribute appreciably to the force. In conclusion, the slab model developed provided a resource-effective approach to understanding the relative importance of the various roller compaction and material parameters on the stresses developed and the densification achieved.
Chapter 6. Two-Dimensional Finite Element Model

6.1. Introduction

The development of a predictive model for roller compaction will significantly advance the understanding, design, optimization and control of the process. Several efforts have been made over the past four decades in this area. A review of these various modeling efforts was recently published (Dec, et al., 2003). Modeling of roller compaction has been challenging due to a number of reasons including: i) the non-linear behaviors related to the contact or frictional conditions at the roll surface and to the constitutive behavior of the deforming powder which undergoes significant evolution of its mechanical properties as it is densified; ii) the lack of experimental inputs measured under relevant process conditions, e.g., frictional coefficient of roll/powder under stress states encountered during roller compaction; iii) experimental difficulties of evaluating the actual process including the boundary conditions, e.g., feed stress and friction, and initial conditions, e.g., density of the powder in the feed zone; iv) the numerous interrelated parameters associated with equipment design, material properties and process settings can make the verification of the model very difficult since many of these parameters can have similar effects on the overall behavior, e.g., roll force or density of compact; v) existing experimental observations and measurements indicate that significant variations in behavior can exist in the roll width and rolling directions and even in the thickness direction, e.g., delamination of a compacted ribbon, thus a model that accounts for these two-and three-dimensional effects will be more complicated to develop; vi) the air within the pores of the deforming powder can have significant effects
under certain conditions and thus a coupled two phase solid-gas analysis is needed. Despite these challenges, the development of simplified process models can provide important insight into roller compaction.

The goal of this modeling effort was to develop a two-dimensional, continuum model of roller compaction to gain greater physical understanding of the process by examination of the variation in field variables, e.g., stress, strain and velocity in both the rolling and thickness directions. Due to the complexities associated with the non-linear behavior of the contact conditions at the roll surface and of the mechanical behavior of the powder, the finite element method was used to solve the equations of continuity and energy. Roller compaction was modeled as a quasi-static process using an arbitrary Lagrangian Eulerian framework with adaptive meshing. Several parametric studies were conducted to investigate the effects of the roll friction ($\mu_{roll}$), entry angle ($\alpha_{entry}$), feed stress ($\sigma_o$), initial relative density ($RD_o$), gap to roll diameter setting ($2h_o/D$), and mechanical behavior of the feed powder on the evolution of contact stresses, internal stresses and densification. The input parameters were defined by experimental measures for microcrystalline cellulose (MCC) and dicalcium phosphate (DCP) as discussed in Chapter 4 and Cunningham et al., (2001).

The use of a 2-D finite element analysis here will provide further insight into the stress and displacement fields during roller compaction without the need for ignoring the through-the-thickness variation that limits the slab analysis. In addition, the frictional conditions (slip versus stick) are no longer evaluated by upper bound argument but are directly evaluated so that they satisfy both equilibrium and compatibility.
The key assumption for the 2-D FEM analysis is that there is no variation in the transverse to the rolling direction (plane strain conditions). To keep the analysis tractable the following approximations are made:

- The effect of interstitial air is ignored which implies that single phase porous plasticity models (such as Gurson and Drucker-Prager/cap (DP/C)) are adequate for the description of powder deformation.
- The deformation of the rolls due to the roll pressure is ignored. This is a reasonable assumption for the roll dimensions and properties and loads observed here.
- Coulomb friction is assumed on the powder/roll interface.
- Body forces such as gravity are omitted.
- Quasi-static deformation is assumed (inertia ignored).
- The effect of the screw feeder is approximated by a uniform stress at the entry angle.

Because the conditions before the nip angle are such that significant shearing is expected, we no longer use an ellipse as yield function (which was consistent with the slab assumption). Instead, we employ a modified DP/C model calibrated as described in Chapter 4. Such models are readily available in commercial finite element programs such as ABAQUS, which was used here.

The two common solution techniques used in the finite element method are implicit and explicit formulations. The implicit technique solves the dynamic equilibrium equations by solving the system of equations for all the elements simultaneously. While the implicit technique is, in general, robust, our attempts for using
it to model roller compaction were unsuccessful due to numerical issues related to severe element distortion. The use of explicit finite element formulation, which has found to be advantageous in certain classes of engineering problems including: high-speed dynamic events, complex contact problems, and highly nonlinear quasi-static problems, was assessed.

The explicit formulation also offers significant advantages over the implicit ones in problems with contact and friction (Bathe, 1996). The explicit integration is however limited by a stability condition, which imposes a maximum time increment:

$$\Delta t_{\text{stable}}^{\text{max}} \leq \frac{L^e}{c_d} = L^e \sqrt{\frac{\rho}{E}}$$

(6.1.1)

where $L^e$, $c_d$, $\rho$ and $E$ are the characteristic length of the element, dilation wave speed, density and Young’s modulus of the material, respectively.

Since tracking the deformation within a deforming body is often of primary interest to the engineer conducting stress analysis, a pure Lagrangian framework, i.e., the mesh representing the material moves with the deforming material, is commonly used. This approach is very effective in analyzing problems in which the deformation is relatively small. In cases where there is significant deformation as in forming processes, the Lagrangian approach may result in a severely distorted mesh and leads to inaccurate results or to premature termination due to convergence problems. The initial attempts of modeling roller compaction using the fully Lagrangian framework with either the implicit or explicit formulation resulted in excessive mesh distortion. The implicit simulation terminated due to inability to converge while the explicit simulations resulted in severely
distorted mesh. To address the issues associated the mesh distortion in the roller compaction problem, the arbitrary Lagrangian-Eulerian (ALE) and adaptive mesh capabilities within ABAQUS/Explicit were used.

To avoid restricted stability condition and integration inaccuracies from distorted elements, a re-meshing algorithm was employed (Hibbitt, et al., 1998). During the roller compaction of powder, the feed powder is continuously delivered to the entry side of rolls and then drawn and densified between the counter-rotating rolls. The compacted powder is subsequently released in the exit side of the rolls. In the analysis of this process, the steady state condition for the imposed boundary and initial conditions is of primary interest. Based on these considerations, it is natural to consider the influx of powder to flow through an Eulerian inflow boundary as depicted in Figure 6.1.1.

Similarly on the exit side, an Eulerian outflow is defined. The material mesh represents a control volume in which material enters and exits. At the Eulerian inflow, the powder with a pre-defined initial relative density, flows in the rolling direction into the mesh domain. The powder is subjected to an evenly distributed load applied in the rolling direction, along the thickness direction. This stress represents the feed stress induced by the feed system. Based on the symmetry of the rolling process, a symmetry boundary exists along the centerline in the rolling direction and only the top half of the material mesh is analyzed. In the feed zone material mesh is constrained by the feeder boundary, which is defined using rigid elements. To simplify the analysis, the contact conditions at this interface between the feeder and powder is friction-less. The distance in the rolling distance was defined to allow fully developed flow conditions to develop prior to the powder contacting the roll.
The 100 mm diameter roll is modeled as a rigid body with a reference node at the center to allow determination of roll force and roll torque. The center of the roll is located to allow the prescribed gap setting. The roll/powder is a Lagrangian boundary with the contact defined as a kinematic contact formulation (Hibbitt, et al., 1998). Two-dimensional plane strain with reduced integration elements (CPE4R) were employed to construct the mesh.

To assess the optimal mesh density for the deformable mesh, a series of simulations with varying mesh densities was conducted. The results are listed in Table 6.1.1. As seen in this table the results are similar between the various mesh densities, therefore based on the optimal combination of accuracy, resolution and computational time, the 70 x 12 mesh was used in the simulations.

**Table 6.1.1.** Results of simulations to evaluate optimal mesh density

<table>
<thead>
<tr>
<th>Mesh Density (rolling x thickness) (Total Elements)</th>
<th>70 x 12 (840)</th>
<th>70 x 24 (1680)</th>
<th>140 x 24 (3360)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Roll Force (N/mm)</td>
<td>358</td>
<td>368</td>
<td>365</td>
</tr>
<tr>
<td>Roll Torque (N-mm)</td>
<td>1020</td>
<td>1043</td>
<td>1029</td>
</tr>
<tr>
<td>RD at $\alpha = 0^\circ$ (-)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>At roll</td>
<td>0.82</td>
<td>0.82</td>
<td>0.82</td>
</tr>
<tr>
<td>At axis of symmetry</td>
<td>0.81</td>
<td>0.81</td>
<td>0.81</td>
</tr>
<tr>
<td>Roll pressure – max (MPa)</td>
<td>61</td>
<td>62</td>
<td>65</td>
</tr>
<tr>
<td>Roll shear stress – max (MPa)</td>
<td>-6.3/+18.2</td>
<td>-7.0/+16.8</td>
<td>-6.9/+20.5</td>
</tr>
<tr>
<td>Nip Angle ($^\circ$)</td>
<td>10.0</td>
<td>9.6</td>
<td>9.7</td>
</tr>
<tr>
<td>Neutral Angle ($^\circ$)</td>
<td>1.2</td>
<td>1.2</td>
<td>1.2</td>
</tr>
</tbody>
</table>
To accelerate the speed of the simulation mass scaling of artificial increase of roll rotation can be exploited (Hibbitt, et al., 1998). In this case the speed of the rolls was artificially increased to accelerate the simulation. Because no inherent rate effects exist in the DP/C model used, the only limit that needs to be considered is to avoid inertia effects. This can be done by limiting the total kinetic energy to be less than a small percent of the internal energy and the maximum local speed to be substantially less than the wave speed. Both constraints were satisfied in our simulations, i.e., the kinetic energy was less than 5% of the internal energy and the deformation speed was less than 0.01% of the wave speed.

A series of parametric studies, as noted in Table 6.1.2, were conducted to analyze the effects of geometry (entry angle and roll gap-to-roll diameter), boundary conditions (feed stress and roll/powder friction), initial condition (initial $RD$) and material properties.

To identify when the roller compaction simulation reaches steady state, the simulation is allowed to proceed until the roll force, $F_{\text{roll}}$, and roll torque, $T_{\text{roll}}$, reach relatively constant values. Initially there is a transient period in which the roll begins to rotate, the feed stress is applied and material enters through the Eulerian inflow boundary. Since the $RD$ throughout the mesh - including under the roll and in the feed zone - is initially equal to the prescribed initial $RD$, there is a period of time in which this material densifies and is drawn between the rolls. Once the steady state condition is reach, there is material entering the Eulerian inflow boundary at a constant rate and is subjected to the prescribed feed stress. The material is drawn between the rolls and further densified before it exits through the Eulerian outflow boundary.
Figure 6.1.1. Schematic of finite element mesh for roller compaction with notation of axis of symmetry, Eulerian inflow and outflow, Lagrangian roll and feeder boundaries and rigid bodies for the roll and feeder.
Table 6.1.2. Parameters used in 2-D simulations of roller compaction.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Designation</th>
<th>Range</th>
<th>Comment</th>
</tr>
</thead>
<tbody>
<tr>
<td>Entry Angle</td>
<td>$\alpha_{\text{entry}}$</td>
<td>15 to 25°</td>
<td>The baseline entry angle was 20°.</td>
</tr>
<tr>
<td>Roll Gap to Roll Diameter</td>
<td>$2h_o/D$</td>
<td>0.015 to 0.0325</td>
<td>The baseline roll gap to diameter was 0.02, i.e., 2 mm for 100 mm diameter roll.</td>
</tr>
<tr>
<td>Feed Stress</td>
<td>$\sigma_o$</td>
<td>0.0125 to 0.400 MPa</td>
<td>Constant and oscillatory in time to simulate screw feeder</td>
</tr>
<tr>
<td>Roll/Powder Frictional Coefficient</td>
<td>$\mu_{\text{roll}}$</td>
<td>0.2 to 0.8</td>
<td>Constant and variable (decrease with normal stress)</td>
</tr>
<tr>
<td>Initial Relative Density</td>
<td>$RD_o$</td>
<td>0.22 to 0.37</td>
<td>Represents varying degrees of pre-densification in screw feeder; baseline $RD_o$ was 0.27.</td>
</tr>
<tr>
<td>Young’s Modulus</td>
<td>$E$</td>
<td>5 GPa</td>
<td>Constant</td>
</tr>
<tr>
<td>Poisson’s Ratio</td>
<td>$\nu$</td>
<td>0.3</td>
<td>Constant</td>
</tr>
<tr>
<td>Cohesion</td>
<td>$d$</td>
<td>0 MPa</td>
<td>Constant</td>
</tr>
<tr>
<td>Angle of Internal Friction</td>
<td>$\beta$</td>
<td>45 to 65°</td>
<td>Most simulations used $\beta = 65°$</td>
</tr>
<tr>
<td>Cap Parameter</td>
<td>$R$</td>
<td>0.3 to 0.8</td>
<td>Most simulations used $R = 0.6$</td>
</tr>
<tr>
<td>Yield Pressure</td>
<td>$p_b(\varepsilon_{\text{pl}}^*)$</td>
<td>Varies</td>
<td>Based on MCC and DCP; function of volumetric plastic strain</td>
</tr>
</tbody>
</table>
6.2. Results and Discussion

The current two-dimensional analysis specifically examines the following:

i) effect of friction at the roll/powder interface using both a constant and a variable (with normal stress) coefficient of friction;

ii) geometrical effects ($\alpha_{\text{entry}}$ and $2h_o/D$); and

iii) influence of constant and oscillatory $\sigma_o$;

iv) impact of $RD_o$ in the feed zone; and

v) effect of varying material parameters used to describe the mechanical behavior of the powder.

The results and discussion section is broken into two primary sections – (i) two-dimensional analysis of roller compaction using an example simulation with comparison to corresponding one-dimensional analysis and (ii) parametric studies on the effects of the boundary conditions ($\mu_{\text{roll}}$ and $\sigma_o$), initial conditions ($RD_o$), geometrical considerations ($\alpha_{\text{entry}}$ and $2h_o/D$) and material properties.

6.2.1. Two-Dimensional Analysis of Roller Compaction

6.2.1.1. An example of 2-D analysis

We begin the presentation of the 2-D analysis by considering in detail an analysis of a case for which:

\[
\begin{align*}
D &= 100 \text{ mm} \\
2h_o &= 2 \text{ mm (gap)} \\
\alpha_{\text{entry}} &= 20.5^\circ \\
RD_o &= 0.27 \\
\sigma_o &= 0.025 \text{ MPa} \\
\mu_{\text{roll}} &= 0.4 \text{ (constant)}
\end{align*}
\]

and the DP/C model is calibrated for MCC as a constitutive model (see Chapter 4).
The resulting roll pressure and shear stress are shown in Figure 6.2.1. The shape of the roll pressure curve is similar to the one obtained by the 1-D slab analysis (especially when the elastic unloading was considered in 1-D, e.g., Figure 5.3.22). The peak pressure is also observed to be just before the minimum separation of the rolls (approx. 0.6°). Note that the $RD_{\text{max}}$ is achieved at the centerline, i.e., $\alpha (p_{\text{max}})$ does not equal $\alpha (RD_{\text{max}})$ contrary to common claims (Pietsch, 1987). A gradual increase of the roll pressure in the entry region and the drastic unloading that extends the stressed region beyond the minimum roll separation is observed as in the 1-D analysis. The first major difference from the 1-D analysis that can be observed is in the shear stress on the roll. While the predicted variation from the slab analysis is discontinuous at the neutral angle (see Figure 5.3.3), the prediction of the 2-D analysis does not show any signs of the discontinuity. The transition from negative to positive shear stresses at the neutral angle is “smooth” in agreement with experimentally measured one (Figure 3.3.8). The neutral angle does not coincide with the maximum pressure angle. In fact it occurs just before (approximately 0.5°) the peak roll pressure (in agreement with the 1D result). This result is also supported by experiments (Schonert and Sander, 2002).

The identification of the slip and stick (nip) regions can be easily performed by the comparison of the local speed of the material mesh in contact with the roll against the roll speed. Also the evolution of the regions can be made by studying the ratio of shear to pressure on the rolls. Recall that the friction model implemented in ABAQUS dictates that slipping occurs when $\tau_{\text{roll}} = \mu_{\text{roll}} p_{\text{roll}}$. Figure 6.2.2. shows clearly:

a) a slipping region close to the entry angle where the powder moves slower than the roll (backward slip);
b) a region where the powder sticks to the roll (nip region) since \( \tau_{\text{roll}} < \mu_{\text{roll}} P_{\text{roll}} \); and

c) a second slip region at the exit of the rolls where the powder moves faster than the roll in an extrusion-like fashion (forward slip).

The forward slip is common when the powder is densified to very high relative densities. A fully dense material when rolled slips everywhere in the rolling region except the neutral angle. Therefore, the presence of the forward slipping region close to the roll exit is not a surprise, but is a subject that has not been disclosed earlier in the literature. Note that although the extrusion zone starts here after the minimum roll separation in many cases (especially the more intense densification) the extrusion zone may start even before the minimum roll separation. It is also notable that the peak of shear stresses after the neutral angle is about three times the corresponding peak before the neutral angle, which is in general agreement with our experimental measurement (Figure 3.3.8.). This high shear stress at the exit occurs within the elastic unloading region. Actually this shows that the analytical expression for the elastic unloading presented in Chapter 5 (Equation 5.18) is an approximate one at best because it ignores the shear stresses. It is remarkable that the finite element model has provided a much more realistic picture of the stress variation in roller compaction; a picture that is much closer to the experimental results than any earlier approximate analysis.
Figure 6.2.1. The roll pressure and roll shear stress versus rolling angle for example simulation \((2h_o/D = 0.02; \alpha_{\text{entry}} = 20.5^\circ; RD_o = 0.27; \sigma_o = 0.025 \text{ MPa}; \mu_{\text{roll}} = 0.4)\).

Note: \(\alpha(p_{\text{max}})\) does not equal \(\alpha(RD_{\text{max}})\)

Figure 6.2.2. The ratio of roll shear stress to roll pressure for example simulation.
\((2h_o/D = 0.02; \alpha_{\text{entry}} = 20.5^\circ; RD_o = 0.27; \sigma_o = 0.025 \text{ MPa}; \mu_{\text{roll}} = 0.4)\)
6.2.1.2. Through-the-Thickness Variation

Although the majority of the pharmaceutical powder rolling operations occur at very small $2h_o/D$ which should promote minimal through-the-thickness variation at least close to the exit, there may, however, be significant through thickness variation closer to the entry angle. Figure 6.2.3 shows the velocity profile predicted by the FEM simulation for the conditions listed section 6.2.1.1. It is evident that there is significant variation of the velocity in the rolling direction close to the entry angle. The velocity at the center at $\alpha = \alpha_{entry}$ is 20% smaller than the velocity of the powder close to the roll. The velocity becomes more uniform downstream but it is evident that thickness inhomogeneity from the early stage may persist as the powder is compacted further. To examine these phenomena we follow the variation of some key variables at three positions (at the roll surface, mid-way between roll and axis of symmetry at the center and along the line of symmetry at the center) which are plotted in Fig. 6.2.4-6.2.9.

In the simulation discussed here, the variation of the vertical stress (Figure 6.2.4(b)) and the stress along the width (Figure 6.2.5(a)) are minimal. Significant variation in the thickness is present in the stress along the rolling direction, $\sigma_{11}$ (Figure 6.2.4(a)). This variation that reaches a maximum of about 35% directly reflects the through thickness variation of the shear stress $\sigma_{12}$ (Figure 6.2.4(d)), which is by definition zero at the centerline (symmetry) and maximum on the surface of the rolls.

Whether this variation of the shear stress is important is not directly evident. In the context of the model there will be no significant variation unless the shear stresses on the roll become comparable to a “large” fraction of $\sigma_{12}$ is less than 25% of $\sigma_{22}$ and the resulting variation in relative density is small ($< 1\%$).
The corresponding variation of the plastic strains through-the-thickness is shown in Figures 6.2.6 and 6.2.7. The variation of the volumetric strain through thickness is small and therefore the resulting variation of the relative density predicted by the model is limited (Figure 6.2.7(b)). Notable is the immediate densification of the material under the feed stress, i.e., increases from 0.27 to 0.32. Recall that at this stage the powder is very loose, and even low levels of the feed stress will result in significant densification.

Despite the similar density histories through-the-thickness, the straining path that causes the densification is different. The most significant variation is caused by the strong shearing of the material close to the rolls which is subjected to a level of shear which is reversed, i.e., there is first shearing forward and subsequent reversal of shearing direction. This is significant because this intense shearing may be beneficial initially (higher shearing will cause higher strength for the same level of relative density (Koerner, 1971) but the reversal of shear may be detrimental in some materials. The latter is an aspect of the physical behavior of some materials that is not incorporated in the DP/C model, and merits further research.

It is often useful to examine the stress path in $p$-$q$ space as shown in Figure 6.2.8. As discussed earlier, the powder at various locations through-the-thickness initially is under relatively high shear conditions in the backward slip region. As the powder is further drawn into the nip region, the slope of the stress path becomes less steep and more hydrostatic in nature. The powder near the roll experience a slightly greater reduction in slope in the loading region of the nip region. Upon unloading, however, the powder near the roll experiences appreciably more shear than the powder closer to the center. The stress path for powder throughout the thickness hits the shear failure line at the exit of the
Although the shear conditions are high in the backward slip region, the powder through-the-thickness does not fail along the shear failure line as shown in Figure 6.2.8. The effect of feed stress acting in the rolling direction offset some of the shear induced by the friction at the roll. When $\sigma_o$ is reduced, conditions can develop in which the powder fails along the shear failure line leading to localization and shear band formation. Shear bands have been observed in practice and can often be eliminated by applying increasing feed stresses to the powder.

The relative degree of hydrostatic to shear conditions can also be evaluated by examining the triaxiality of the stress state, $X = p/q$, through the slip and nip regions as shown in Figure 6.2.9. Overall, the powder initially experiences lower triaxialities in the slip backward region and then increasing triaxialities in the nip region followed again by lower triaxialities upon unloading and exit from the roll. In the backward slip region, the powder is under higher shear state due the friction at the roll. As the powder sticks at the roll, the stress state becomes more hydrostatic. Upon unloading, the predominance of shear again becomes apparent. Finally, it should be noted that the observed through thickness variation will be higher for larger $2h_o/D$ and $a_{entry}$. 
Figure 6.2.3. (a) Velocity in the rolling direction for powder at the roll, mid and center. Velocity of the roll in the rolling direction is also shown for reference. (b) Contours of velocity in the rolling direction. \(2h_o/D = 0.02; \alpha_{entry} = 20.5^\circ; RD_o = 0.27; \sigma_o = 0.025 \text{ MPa}; \mu_{roll} = 0.4\)
Figure 6.2.4. Stresses for material at center, mid and roll: (a) $\sigma_{11}$ (S11) and (b) $\sigma_{22}$ (S22) as a function of rolling angle. ($2h_0/D = 0.02$; $\alpha_{entry} = 20.5^\circ$; $RD_o = 0.27$; $\sigma_o = 0.025$ MPa; $\mu_{roll} = 0.4$)
Figure 6.2.5. Stresses for material at center, mid and roll: (a) $\sigma_{33}$ (S33) and (b) $\sigma_{12}$ (S12) as a function of rolling angle. ($2h_o/D = 0.02$; $\alpha_{entry} = 20.5^\circ$; $RD_o = 0.27$; $\sigma_o = 0.025$ MPa; $\mu_{roll} = 0.4$)
Figure 6.2.6. Plastic strains for material at center, mid and roll: (a) $\varepsilon_{pl}^{11}$ (PE11) and (b) $\varepsilon_{pl}^{22}$ (PE22) as a function of rolling angle. ($2h/D = 0.02; \alpha_{entry} = 20.5^\circ; RD_o = 0.27; \sigma_o = 0.025$ MPa; $\mu_{roll} = 0.4$)
Figure 6.2.7. Plastic strains for material at center, mid and roll: (a) $\varepsilon_{pl12}^{PE12}$ and (b) $RD$ as a function of rolling angle. ($2h_0/D = 0.02$; $\alpha_{entry} = 20.5^\circ$; $RD_0 = 0.27$; $\sigma_0 = 0.025$ MPa; $\mu_{roll} = 0.4$)
Figure 6.2.8. (a) The stress path in hydrostatic pressure-Mises effective stress \((p-q)\) space for material at roll, mid and center. (b) The stress path in hydrostatic pressure-Mises effective stress \((p-q)\) space for material at roll, mid and center at the low stress states in the feeder and slip regions. \((2h_o/D = 0.02; \alpha_{entry} = 20.5^\circ; RD_o = 0.27; \sigma_o = 0.025 \text{ MPa}; \mu_{roll} = 0.4)\)
**Figure 6.2.9.** Triaxiality of material at center, mid and roll. ($2h_{o}/D = 0.02; \alpha_{\text{entry}} = 20.5^\circ; RD_{o} = 0.27; \sigma_{o} = 0.025 \text{ MPa}; \mu_{\text{roll}} = 0.4$)
6.2.1.3. Comparison to 1-D Analysis

It is rather difficult to achieve a proper comparison between the 1-D slab analysis and the 2-D FEM analysis primarily because of the omission of elasticity in the slab analysis. In order to make such comparison more objective we compared the 2-D FEM with a special 1-D FEM simulation that approximates the uniform through-the-thickness assumption of the slab analysis. To achieve this, single elements were used in the through-the-thickness direction and the elements were prevented from undergoing distortion by placing constraints on the elements in a similar manner as used in a slab method, i.e., there was no relative displacement in the rolling direction between the corners of the element sharing the same position along the rolling direction. For comparison 1-D analyses were conducted using the same parameters (geometrical, initial and boundary conditions and constitutive model) as used for several of the 2-D simulations. Parametric studies examining the effects of $\mu_{\text{roll}}$ and $\sigma_o$ were conducted. In Figure 6.2.10 (a) and (b), a plot of $F_{\text{roll}}$ and $T_{\text{roll}}$ as a function of $\sigma_o$ is presented. These values are normalized for the unit roll width and are presented in terms of N per mm. In these simulations, $\alpha_{\text{entry}} = 20.5^\circ$, $2h_o/D = 0.02$, $\mu_{\text{roll}} = 0.35$, $RD_{o} = 0.27$ and the constitutive model is the DP/C for MCC. $\sigma_o$ is varied from 0.025 to 0.300 MPa. As seen in this figure, $F_{\text{roll}}$ and $T_{\text{roll}}$ are overall higher for the 1-D case than the 2-D case. The difference is, however, relatively small at lower $\sigma_o$. The difference becomes greater as $\sigma_o$ increases. A plot of the $p_{\text{roll}}^{\text{max}}$ and $RD_{\text{max}}^{\text{max}}$ as a function of $\sigma_o$ is provided in Figure 6.2.11 (a) and (b). Similarly $p_{\text{roll}}^{\text{max}}$ and $RD_{\text{max}}^{\text{max}}$ are overall higher for the 1-D case with the difference increasing as $\sigma_o$ increases.
The $p_{roll}^\text{max}$ and $RD_{\text{max}}$ as a function of $\mu_{roll}$ is plotted in Figure 6.2.12 (a) and (b).

In these simulations, the feed stress was held constant at 0.025 MPa. In a similar manner, the values of $p_{roll}^\text{max}$ and $RD_{\text{max}}$ are higher for the 1-D case. The difference becomes significantly larger as the frictional coefficient increases. In both analyses, the kinematic conditions and continuity condition must be satisfied. In the case of the 1-D analysis the elements have an artificial constraint that can lead to a higher stiffness and thus higher stresses developed. This is contributing to the observed differences in the predictions.

$\alpha_{nip}$ predicted by the 1-D and 2-D analyses are quite different as shown in Figures 6.2.13 (a) and (b) in which $\alpha_{nip}$ are shown as functions of $\sigma_o$ and $\mu_{roll}$, respectively. In the case of $\sigma_o$ varying from 0.025 to 0.300 MPa, $\alpha_{nip}$ for the 1-D case is approximately 3° lower than the 2-D case. Likewise the 1-D analysis consistently predicted lower $\alpha_{nip}$ than the 2-D analysis when the roll friction is varied. The difference is about 2 to 5°.
Figure 6.2.10. 1-D vs 2-D simulations. (a) Roll force and (b) roll torque (per unit roll width) of feed stress. ($2h_o/D = 0.02; \alpha_{entry} = 20.5^\circ; RD_o = 0.27; MPa; \mu_{roll} = 0.35$)
Figure 6.2.11. 1-D vs 2-D simulations. (a) Maximum $p_{roll}$ and (b) maximum relative density as function of feed stress. ($2h_o/D = 0.02$; $\alpha_{entry} = 20.5^\circ$; $RD_o = 0.27$; MPa; $\mu_{roll} = 0.35$)
Figure 6.2.12. 1-D vs 2-D simulations. (a) maximum roll pressure and (b) maximum relative density as function of roll/powder friction. ($2h_o/D = 0.02; \alpha_{entry} = 20.5^\circ; RD_o = 0.27; \sigma_o = 0.025 \text{ MPa}$)
Figure 6.2.13. (a) Nip angle as function of feed stress ($2h_o/D = 0.02$; $\alpha_{entry} = 20.5^\circ$; $RD_o = 0.27$; $\mu_{roll} = 0.35$). (b) Nip angle as function of roll/powder frictional coefficient ($2h_o/D = 0.02$; $\alpha_{entry} = 20.5^\circ$; $RD_o = 0.27$; $\sigma_o = 0.025$ MPa)
6.2.2. Parametric Studies

In an analogous manner to the studies using the 1-D slab analysis in Chapter 5, the effects of roll/powder friction, roll gap to roll diameter, and feed conditions (feed stress, entry angle and initial relative density) on the maximum roll pressure, maximum relative density, nip angle, neutral angle and evolution of relative density were assessed. In addition, the effects of a variable coefficient of roll/powder friction and an oscillating feed stress were also evaluated. Finally, the influence of the mechanical behavior of the powder based on the modified DP/C model was determined. The focus of the discussion is on information that provides additional insight into the roller compaction process beyond that derived from the slab (Chapter 5) and 1-D (Section 6.2.1.3) analyses.

In Figure 6.2.14, the impact of the parameters studied are categorized in terms of their influence on the pre-densification in the feeder and slip region, the position of the nip angle and the densification in the nip region. Several of the parameters including roll friction, entry angle, feed stress, initial relative density and constitutive behavior affect the pre-densification that is achieved prior to the nip angle. These parameters primarily affect the pre-densification either by directly influencing the densification prior to the nip angle, e.g., feed stress and initial relative density, or by defining the boundaries of the slip region (entry angle to nip angle), e.g., roll friction and entry angle. In a similar manner to what was observed in the 1-D model the pre-densification has very significant impact on the final achievable densification. The position of the nip angle is significantly affected by the roll
friction and constitutive behavior. The entry angle, feed stress, initial relative density and roll gap to diameter do not affect the nip angle. The densification in the nip region is influenced by the roll friction, the roll gap to roll diameter and the constitutive behavior. Roll friction and constitutive behavior influence the length of the nip region through the position of the nip angle. The roll gap to roll diameter defines the deformation that must be achieved in the nip region. Finally, the constitutive behavior influences the development of stresses and resulting deformation and densification.

**Figure 6.2.14.** List of factors affecting the pre-densification in the feeder and slip region, location of the nip angle and densification in the nip region.
6.2.2.1. Effect of Roll Friction, Entry Angle, Feed Stress and Initial Density

The effect of roll friction in the 2-D FEM model was similar as observed in the 1-D model, i.e., the powder is gripped sooner when the roll friction is higher and greater densification is obtained in the nip region with higher values of roll force, roll torque and roll pressure developed. Interestingly, there were conditions that were not possible due to inability of the feed powder to be gripped and drawn into the nip region. This occurred with $\mu_{\text{roll}} = 0.10$ for $\alpha_{\text{entry}} = 10, 15$ and 25° and with $\mu_{\text{roll}} = 0.20$ for $\alpha_{\text{entry}} = 20$ and 25°. Under these low $\mu_{\text{roll}}$ /high $\alpha_{\text{entry}}$ conditions, the powder continued to slip at entry and did not proceed into the nip. This is analogous to the rolling of fully dense metal slab in which the thicker slab will not be gripped unless the roll friction is sufficiently high (Dieter, 1986). To examine this effect closer a series of simulations were conducted with $\alpha_{\text{entry}} = 20°$ and $\mu_{\text{roll}} = 0.20$. In these simulations, $\sigma_o$ was progressively increased until a critical stress (here when 0.400 MPa is reached) when the powder was gripped and drawn into the nip. Below this critical stress, there was continual slippage at the roll/powder interface. An increase in the feed stress implies an immediate pre-densification of the powder. This is shown in Figure 6.2.15 in which the $\tau_{\text{roll}}$ is plotted against $p_{\text{roll}}$ for these simulations. The region of slipping and sticking are demarcated by the line with the slope equivalent to $\mu_{\text{roll}} = 0.20$. Under lower $\sigma_o$, the ratio of $\tau_{\text{roll}}$ to $p_{\text{roll}}$ remains equal to $\mu_{\text{roll}}$. As $\sigma_o$ increased, greater $p_{\text{roll}}$ develops and the roll is able to grip and draw the powder into the nip, i.e., the combination of contact stresses moves off the slip failure line as shown in Figure 6.2.15. These simulations indicate that low roll friction may be
compensated with higher $\sigma_0$. In practice, however, whether adequate feed stresses can be developed depends on the feeder design and capabilities.

As noted in Chapter 5, different combinations of roll friction, entry angle and feed stress can yield similar roll pressures and densification. An example of this is shown in Figure 6.2.16. This highlights the interplay of some process and material parameters.

As seen in Chapter 4, the friction may not be constant and tends to decrease with normal stress. To assess the significance of varying $\mu_{\text{roll}}$, a series of simulations in which $\mu_{\text{roll}}$ decreased from constant values following either a linear reduction or varying degrees of exponential reduction with the roll pressure as shown Figure 6.2.17(a) and (b). As observed in these figures, the resulting relative density and maximum roll pressure can vary significantly. These results highlight the importance of appropriate definition of $\mu_{\text{roll}}$ under relevant conditions, e.g., normal stress, for modeling roller compaction.

Although smooth rolls are sometimes used in roller compaction, often the roll has a textured surface that enhances the gripping action of the rolls. The contact conditions at the roll/powder interface are more complex. Low pressure shear cell tests using coupon surfaces reflecting the surface properties can be useful for low stress states but higher normal stresses are also relevant in roller compaction. In addition, in some cases the powder may adhere to a textured surface and friction is effectively shearing of a layer powder adjacent to the rolls. Although it was not attempted here, the use of finite element modeling with textured rolls has been conducted with briquetting (Zavaliangos et al., 2003). Given its importance in roller compaction, critical experimental assessment of the frictional properties of the roll/powder over the ranges of stress states and $RD$ is warranted. It is also interesting to highlight that there is a correlation between $p_{\text{roll}}^{\text{max}}$ and
$RD^{max}$ for the various constant and variable $\mu_{roll}$ with varying entry angle falls, which is directly related to the one-to-one correlation of $p_{roll}$ and $RD$ for the low $2ho/D$ in which $p_{roll}$ is approximately equal to $\sigma_{22}$.

Similar to the 1-D model the effect of constant feed stress can be observed in the level of pre-densification in the feeder prior to contacting the roll. At higher values of $\sigma_o$, the powder densifies to increasingly higher levels. The nip angle is not affected by the feed stress. The powder densified higher in the feeder continues to densifies to a greater extent in the slip region and consequently achieves higher final densification. Since the feed stress is directly related to the entry angle and corresponding thickness at the entry, it should be noted that higher entry angles require significantly higher feed forces to be developed to maintain a constant feed stress, e.g., the feed force must be increased by 48% and 110% to apply the same feed stress at entry angles of 20° and 25°, respectively, compared to 15°. Whether a specific roller compactor can achieve this level of feed force depends on the feeder design.

In practice the screw feeder does not induce a constant feed stress but rather it is oscillatory in nature (Dec and Komarek, 1993). To simulate this effect, $\sigma_o$ was prescribed as a sinusoidal boundary condition for the case in which $\alpha_{entry} = 20^\circ$, $2h_o/D$, =0.02, and $\mu_{roll} = 0.40$. The prescribed $\sigma_o$ and resulting roll force and $T_{roll}$, $RD$, and $p_{roll}$ are provided at select time points in Figure 6.2.19. Although there is a lag related to the movement of material already with the nip when $\sigma_o$ changes, the $F_{roll}$, $T_{roll}$, $RD$, and $p_{roll}$ follow similar sinusoidal fluctuations in values. The oscillatory nature of the feed stress is directly reflected in the other process parameters and compact properties.
Parametric studies on the effect of the entry angle revealed similar trends as the 1-D model in Chapter 5. When the entry angle was sufficiently high, the nip angle was not affected by the entry angle. The entry angle mainly affects the length of the slip region and thus the level of pre-densification that can be achieved prior to the nip angle (Figure 6.2.20).

The roll gap to roll diameter, $2h_o/D$, is an important geometric parameter to consider when designing a roller compactor and selecting the optimal process parameters. The roll diameter has a significance influence on the overall size of the roller compactor and the associated drive motors for the rolls and the screw conveyor. Achieving the desired densification and throughput can also be significantly affected by the gap setting. The gap is also often used in process control, e.g., for roller compactors with the floating roll design, the gap setting is measured and maintained within acceptable limits by adjustment of the feed screw motor. Similar to results of the 1-D slab analysis in Chapter 5, the $RD_{max}$ increases steadily as $2h_o/D$ decreases for all feed stress conditions.

A series of simulations were conducted in which $RD_o$ varied from 0.22 to 0.42 with $\sigma_o = 0.0125$ to 0.075 MPa. $\alpha_{entry} = 20^\circ$, $\mu_{roll} = 0.35$, $2h_o/D = 0.020$ were used in the simulations. The effect of the initial relative density depended on the level of pre-densification induced by the feed stress, which can be more clearly be understood by examining the evolution of density. When $\sigma_o = 0.025$ MPa, when the powder with $RD_o = 0.22$ or 0.27 pre-densifies to a similar level as the powder with $RD = 0.32$. These simulation have similar evolution of $RD$. When $RD_o = 0.37$, however, the feed stress of 0.025 MPa is insufficient to pre-densify further and the density of the powder does not increase until the powder is 2-3$^\circ$ into the slip region. The level of pre-densification in the
feeder is dependent on the low pressure yield behavior of the powder. The yield pressure of the powder with $RD_o=0.37$ was higher than that achieved with $\sigma_o = 0.025$ MPa. This feed stress was, however, sufficiently high to allow the powder with $RD_o = 0.22$ and $0.27$ to behave similarly to powder with $RD_o = 0.32$. The densification occurring within the feeder is dependent on the starting density and the constitutive behavior at low pressures.

To further examine this behavior, the evolution of $RD$ for additional simulations in which $RD_o = 0.42$ and $\sigma_o = 0.025$ MPa and $RD_o = 0.27$ and $\sigma_o = 0.075$ MPa are plotted along with the previously discussed simulations, Figure 6.2.21. Similar to $RD_o = 0.37$, the feed stress of 0.025 MPa is insufficient to induce additional densification of the powder and the density does increase until the powder is $4^\circ$ into the slip region. In the case of $RD_o = 0.27$ and $\sigma_o = 0.075$ MPa, the powder densifies to $RD = 0.34$ — higher than the 0.32 achieved when $\sigma_o = 0.025$ MPa. The powder continues to densify to $RD = 0.84$ similar to the simulation in which $RD_o = 0.42$ and $\sigma_o = 0.025$ MPa. It is interesting to note this is higher than that achieved for $RD_o = 0.37$ and $\sigma_o = 0.025$ MPa, although the $RD$ in the feeder was lower (0.34 vs 0.37). This highlights that the feed stress not only contributes to pre-densifying the powder on the feeder but also enhancing the densification occurring in the slip region. These results also emphasize the importance of the $RD$ of the powder as it contacts the rolls, which is directly related to the low pressure yield behavior and stresses developed in the feeder. The influence of varying low pressure yield pressure is further analyzed later.
Figure 6.2.15. Roll shear stress versus roll pressure for varying feed stresses when roll/powder friction = 0.2.

Figure 6.2.16. Roll pressure profiles for two examples. \(2h_o/D = 0.02; RD_o = 0.27; \sigma_o = 0.025 \text{ MPa}\)
Figure 6.2.17. (a) Maximum roll pressure and maximum relative density as function varying definition of roll/powder friction. \(2h_o/D = 0.02; \alpha_{entry} = 20.5^\circ; RD_o = 0.27; \sigma_o = 0.100 \text{ MPa}\) (b) Maximum roll pressure and maximum relative density as function varying definition of roll/powder friction. \(2h_o/D = 0.02; \alpha_{entry} = 20.5^\circ; RD_o = 0.27; \sigma_o = 0.400 \text{ MPa}\)
Figure 6.2.18. Evolution of relative density with rolling angle for various feed stresses. \(2h_o/D = 0.02; \alpha_{\text{entry}} = 20.5^\circ; RD_0 = 0.27; \mu_{\text{roll}} = 0.35\)

Figure 6.2.19. The feed stress (prescribed) and resulting roll force as a function of time. The specific value of roll force, roll torque, maximum roll pressure and maximum relative density are shown. \(\alpha_{\text{entry}} = 20^\circ, 2h_o/D = 0.02, \mu_{\text{roll}} = 0.40\)
**Figure 6.2.20.** Evolution of relative density as a function of rolling angle for varying entry angles. 
\(2h_o/D = 0.02, RD_o = 0.27, \sigma_o = 0.025\) MPa

**Figure 6.2.21.** Evolution of relative density for varying initial relative densities for feed stress = 0.0125 MPa and feed stress of 0.075. 
\(2h_o/D = 0.02, \alpha_{entry} = 20.5^\circ, \mu_{roll} = 0.35\)
6.2.2.2. Material Properties

In this section, the effects of various material parameters of the modified DP/C model are examined. As discussed in Chapter 4 the material parameters that define the modified DP/C model, e.g., cohesion \((d)\), angle of internal friction \((\beta)\) and cap parameter \((R)\), were demonstrated to evolve or change with densification. To incorporate these changing material parameters in the explicit version of ABAQUS a user-defined subroutine, a VUMAT, must be developed. This was outside the scope of the present research. However, to assess the impact of these parameters, 2-D parametric studies were conducted with varying constant values. In one set of studies the effect of \(\beta\) and \(R\) was evaluated while keeping the hardening characteristics, i.e., \(p_b\) versus \(\varepsilon_{vol}^{pl}\), constant. Since cohesion \((d)\) is very low at the initial relative densities, it was assumed to be zero. In additional studies, the effect of changing the hardening characteristics was assessed. Two set of studies were conducted: i) the effect of extrapolating the hydrostatic yield pressure at low \(RD\) as an exponential function, which was used previous simulations, and as a linear function for MCC and ii) effect of different hardening characteristics, i.e., MCC compared to DCP. Finally, results of simulations using a 1-D FEM model in ABAQUS/Standard for which \(d\), \(\beta\), and \(R\) were defined as a function of the \(RD\) are provided to assess the effect of evolving material parameters.

The definition of the cap surface in ABAQUS depends on the values of \(d\) and \(\beta\) thus changing the value of \(\beta\) causes the shape of the cap to shape. To minimize this effect, the value of \(R\) was adjusted in effort to keep the cap similar (Figure 6.2.22). In
these simulations, $\sigma_0$ was varied from 0.025 to 0.400 MPa while $\alpha_{\text{entry}}$, $\mu_{\text{roll}}$ and $2h_0/D$ were set at 20°, 0.35 and 0.020, respectively.

As the angle of internal friction increased, higher roll pressures and relative densities were achieved for a given feed stress (Figure 6.3.23). The nip angles for both $\beta = 55^\circ$ and $65^\circ$ were similar at $9^\circ$. During the analysis of the results of simulations in which $\beta = 45^\circ$, it was observed that the material underwent dramatic decrease in densification near the centerline of the rolls as shown in Figure 6.2.24(a). Examination of the roll pressure and roll shear stress profiles – Figure 6.2.24(b) – reveals the roll pressure reaches a maximum at $3.6^\circ$ and the neutral angle is at $3.8^\circ$, both of which are much higher than other simulations. In addition, the profile of the velocity in the rolling direction of the powder at the roll surface – Figure 6.2.25 – indicates the nip angle is $8.4^\circ$ while the forward slip begins at $2.6^\circ$ - well before the centerline of the rolls. The powder begins to accelerate significantly prior to the centerline of the rolls.

The stress path in loading and unloading (based on position of the maximum roll pressure) – Figure 6.2.26 – indicates that the powder is initially yielding along the shear failure line when it first contacts the roll after being pre-densified in the feeder. This is opposed to the higher values of internal friction where the powder is yielding in the cap region upon entering the rolls – Figure 6.2.27. For $\beta = 45^\circ$, the powder does not experience densification until the stress state develops such that yielding occurs in the cap region deeper into the slip region. Upon unloading the powder yields at shear failure surface and experiences dilation as defined by the flow potential.

A series of simulations were conducted in which $R$ was varied between 0.3 to 0.8 while $\beta = 65^\circ$. In these simulations, $\alpha_{\text{entry}}$, $\sigma_0$, $\mu_{\text{roll}}$ and $2h_0/D$ were set at 20°, 0.025
MPa, 0.35 and 0.020 mm, respectively. The value of $R$ affects the shape of the elliptical cap region and the associated plastic flow potential surface. As $R$ increases, the $\alpha_{nip}$, $p_{roll}^{\text{max}}$ and $RD^{\text{max}}$ decrease significantly (Table 6.2.1). To examine this closer, the evolution of $RD$ with the rolling angle is presented in Figure 6.2.28(a). As seen in this graph, the powders with the various cap parameters experience similar densification in the feed zone. The level of densification achieved in the slip region, however, varies significantly with $R$. The powder with values of $R$ densify to higher levels in the slip and also reaches the nip region sooner and thus achieves higher final density. This behavior may be explained in the context of the $p$-$q$ stress path and yield and plastic flow surfaces as shown in Figure 6.2.28(b). Although the overall stress path has a lower triaxiality, i.e., higher effective (shear) stress to hydrostatic pressure, for $R = 0.3$, the relative degree of volumetric plastic strain is higher (compared to effective plastic strain) thus powders with lower values of $R$ undergo greater densification. Finally a plot of the maximum densification and maximum roll pressure is provided in 6.2.29. The densification behavior are similar to previous simulations in which $R = 0.6$. This highlights the densification curve is primarily a function of the hardening characteristics of the powder.

As previously discussed, densification and stress state that develops in the feeder and in the slip region can have a profound effect on the maximum achievable $RD$. The stresses in the feed region are low, however the amount of deformation that occurs in the powder is relatively high. To examine this effect, $p_b$ versus $\varepsilon_{vol}^{pl}$ for $RD$ below 0.35 were modified. The experimental data used to calibrate the DP/C model for MCC were limited to relative densities of higher than 0.35. For the $RD$ less than 0.35, it was assumed that $p_b$ exponentially decayed. Parametric studies were conducted using $\alpha_{entry} = 20^\circ$, $2h_o/D =$
shows the $p_{roll}^{\text{max}}$ and $RD_{D}^{\text{max}}$ for varying $\sigma_o$ (0.025 MPa to 0.075 MPa). The values of $RD_{D}^{\text{max}}$ and corresponding $p_{roll}^{\text{max}}$ are appreciably higher for the simulations in which $p_b$ decays exponentially at the lower relative densities compared to a linear decay (Figure 6.2.30(a) and (b)). To better understand this behavior, a plot of the evolution of $RD$ for $\sigma_o$ of 0.075 MPa is shown in Figure 6.2.31(a). Although there is small differences in $\alpha_{nip}$, the level of densification in the feed zone and slip zone is quite different in the two cases. The powder with the linear decay in $p_b$ undergoes less densification for the given $\sigma_o$ since its yield pressure for the given $RD$ is higher. The powder with the exponential decay in $p_b$ undergoes greater pre-densification in the feeder (0.28 vs 0.37), thus the powder with the exponential decay contacts the roll with a higher $RD$ and is further compacted to higher density in the slip and nip regions. This highlights the importance of the low pressure behavior and the need for appropriate experimental calibration of the constitutive model at these low pressures. Finally, it is interesting to plot the $RD_{D}^{\text{max}}$ vs $p_{roll}^{\text{max}}$ for these two powders – Figure 6.2.31(b). The plots are superimposable indicating the $RD_{D}^{\text{max}}$ and the corresponding required $p_{roll}$ is affected primarily for the higher pressure behavior.

A series of simulations was conducted in which the hardening characteristics were modified to reflect the differences in densification behavior of MCC and DCP. While $d$, $\beta$, and $R$ were the same, $p_b - \varepsilon^{pl}_{vol}$ relationship was different (Cunningham et al., 2001). The key difference in the material behavior is the significantly higher pressures required to densify DCP to comparable $RD$. This difference can be related to the inherent differences in the soft, polymeric nature of MCC and the hard, ceramic-like DCP. The simulations used $\sigma_o = 0.025$ MPa, $\alpha_{entry} = 20^\circ$, and $2h_o/D = 0.020$. $\mu_{roll}$ varied from 0.30
to 0.40. \( \alpha_{nip}, p_{roll}^{max} \) and \( RD_{max}^{max} \) as a function of \( \mu_{roll} \) are presented in Table 6.2.2. \( \alpha_{nip} \), which is generally higher for the MCC, contributes to the higher densification for MCC. 

\( p_{roll} \) developed for a given \( \mu_{roll} \) is significantly higher for the DCP. These differences are readily apparent in \( p_{roll} (\alpha) \) profiles presented in Figure 6.2.32(a). Finally, the significant differences in the hardening characteristics of DCP and MCC are readily apparent in the plot of \( RD_{max}^{max} \) versus \( p_{roll}^{max} \) in Figure 6.2.32(b). Unlike the previous plot for MCC in which the various combination of \( \sigma_o, \mu_{roll}, \alpha_{entry} \) and gap resulted a superimposable curve, the densification behavior is dramatically different for these two powders.

Finally, two series of 1-D FEM simulations in the implicit version of ABAQUS were conducted using \( RD \)-dependent material parameters as described in Chapter 4: i) \( \mu_{roll} \) was varied between 0.30 and 0.70 with \( \sigma_o = 0.025 \) MPa, \( \alpha_{entry} = 20^\circ \) and \( 2h_o/D = 0.020 \) and ii) \( \sigma_o \) was varied between 0.025 to 0.300 MPa with \( \mu_{roll} = 0.35, \alpha_{entry} = 20^\circ \) and \( 2h_o/D = 0.020 \). The values of \( F_{roll}, T_{roll}, \alpha_{nip}, \alpha_{neutral}, p_{roll}^{max} \) and \( RD_{max}^{max} \) are presented in Table 6.2.3. It is observed that the \( \alpha_{nip}, p_{roll}^{max} \) and \( RD_{max}^{max} \) increase with \( \mu_{roll} \).

Interestingly, the nip angles are lower than observed in the 1-D model using constant material parameters (Figure 6.2.13) reflecting the importance influence of the material parameters on the nip angle. As discussed earlier the initial stress path is low triaxiality and the powder deforms and densifies based on satisfying the equilibrium and yield condition at or near the shear failure line. Figure 6.2.33(a) reveals the stress path during loading for the frictional case (\( \mu_{roll} = 0.50 \)) with the initial shear failure line, i.e., \( RD = 0.31 \), and with \( RD = 0.75 \). Since the shear failure line evolves with densification, i.e., cohesion and angle of friction increase with \( RD \), these shear failure lines represent the
boundaries for this simulation in which the maximum $RD$ is 0.75. Initially the stress path moves close to the failure line while remaining in the cap, i.e., densification region, - Figure 6.2.33 (b). While similar trends are found with the increasing feed stress as observed in the previous 1-D FEM and 2-D FEM models, the level of densification is overall lower. These simulations highlight the importance of implementing a constitutive model that appropriately captures the evolution of the material properties as the powder densifies.
Figure 6.2.22. Example yield surfaces for various combinations of angle of internal friction ($\beta$) and cap parameter ($R$). The yield pressure is the same.
Figure 6.2.23. (a) Maximum roll pressure and (b) Maximum relative density as a function of feed stress for $\beta = 55$ and 65°. ($2h/D = 0.02; \alpha_{entry} = 20.5^\circ; RD_o = 0.27; \mu_{roll} = 0.35$)
Figure 6.2.24. (a) Evolution of relative density as a function of rolling angle for beta = 45°. (b) Roll pressure and roll shear stress for β = 45°. (2h_o/D= 0.02; α_{entry} = 20.5°; RD_o = 0.27; MPa; μ_{roll} = 0.35)
Figure 6.2.25. Velocity of powder near roll surface as function of rolling angle. \((2h_o/D=0.02; \alpha_{entry}=20.5^\circ; RD_o=0.27; \text{MPa}; \mu_{roll}=0.35)\)

Figure 6.2.26. Stress path in loading and unloading for \(\beta=45^\circ\). \((2h_o/D=0.02; \alpha_{entry}=20.5^\circ; RD_o=0.27; \mu_{roll}=0.35)\)
Figure 6.2.27. Initial stress paths during loading for $\beta = 55^\circ$ and $65^\circ$. ($2h_o/D = 0.02$; $\alpha_{entry} = 20.5^\circ$; $RD_o = 0.27$; $\mu_{roll} = 0.35$)

Table 6.2.1. Table of Nip Angle, maximum roll pressure and maximum relative density for varying cap parameters.

<table>
<thead>
<tr>
<th>Cap Parameter, $R$ (-)</th>
<th>Nip Angle ($^\circ$)</th>
<th>Maximum Roll Pressure (MPa)</th>
<th>Maximum Relative Density (-)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.3</td>
<td>11.9</td>
<td>268.7</td>
<td>0.95</td>
</tr>
<tr>
<td>0.4</td>
<td>10.1</td>
<td>125.0</td>
<td>0.90</td>
</tr>
<tr>
<td>0.5</td>
<td>9.5</td>
<td>71.6</td>
<td>0.83</td>
</tr>
<tr>
<td>0.6</td>
<td>8.3</td>
<td>44.7</td>
<td>0.77</td>
</tr>
<tr>
<td>0.7</td>
<td>7.8</td>
<td>31.3</td>
<td>0.72</td>
</tr>
<tr>
<td>0.8</td>
<td>7.2</td>
<td>21.9</td>
<td>0.67</td>
</tr>
</tbody>
</table>
Figure 6.2.28. Effect of cap parameter, $R$. (a) Relative density profiles. (b) $p$-$q$ stress paths.
Figure 6.2.29. Maximum roll pressure and maximum relative density.
(a) Maximum roll pressure and (b) maximum relative density as function of feed stress for different low pressure yield behavior – exponential and linear.

**Figure 6.2.30.** (a) Maximum roll pressure and (b) maximum relative density as function of feed stress for different low pressure yield behavior – exponential and linear.
Figure 6.2.31. (a) Relative density profile and (b) maximum relative density versus maximum roll pressure for different low pressure yield behavior – exponential and linear.
Table 6.2.2. Nip angle, maximum roll pressure and maximum relative density for DCP and MCC for varying values of roll friction.

<table>
<thead>
<tr>
<th>$\mu_{roll}$ (-)</th>
<th>$\alpha_{nip}$ (°)</th>
<th>$p_{roll}^{max}$ (MPa)</th>
<th>$RD_{max}$ (-)</th>
<th>$\alpha_{nip}$ (°)</th>
<th>$p_{roll}^{max}$ (MPa)</th>
<th>$RD_{max}$ (-)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.30</td>
<td>5.14</td>
<td>107.1</td>
<td>0.63</td>
<td>6.75</td>
<td>22.4</td>
<td>0.67</td>
</tr>
<tr>
<td>0.35</td>
<td>7.26</td>
<td>170.6</td>
<td>0.67</td>
<td>7.81</td>
<td>33.3</td>
<td>0.73</td>
</tr>
<tr>
<td>0.40</td>
<td>7.81</td>
<td>7.81</td>
<td>0.70</td>
<td>10.05</td>
<td>46.0</td>
<td>0.77</td>
</tr>
</tbody>
</table>
Figure 6.2.32. MCC and DCP. (a) Roll pressure profiles for varying roll/powder friction. (b) Maximum relative density as function of maximum roll pressure.
Table 6.2.3. 1-D FEM Results for RD-dependent material parameter. Roll force, roll torque, nip angle, neutral angle, maximum roll pressure and maximum relative varying values of (a) roll friction and (b) feed stress.

<table>
<thead>
<tr>
<th>$\mu_{\text{roll}}$ (-)</th>
<th>$F_{\text{roll}}$ * (N)</th>
<th>$T_{\text{roll}}$ * (N-mm)</th>
<th>$\alpha_{\text{nip}}$ (°)</th>
<th>$\alpha_{\text{neutral}}$ (°)</th>
<th>$p_{\text{roll}}^{\text{max}}$ (MPa)</th>
<th>$\text{RD}^{\text{max}}$ (-)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.30</td>
<td>33.8</td>
<td>111</td>
<td>9.40</td>
<td>3.0</td>
<td>5.72</td>
<td>0.468</td>
</tr>
<tr>
<td>0.35</td>
<td>77.1</td>
<td>212</td>
<td>9.97</td>
<td>1.0</td>
<td>14.2</td>
<td>0.556</td>
</tr>
<tr>
<td>0.40</td>
<td>142</td>
<td>373</td>
<td>10.72</td>
<td>1.0</td>
<td>26.1</td>
<td>0.634</td>
</tr>
<tr>
<td>0.50</td>
<td>291</td>
<td>789</td>
<td>12.22</td>
<td>0.9</td>
<td>52.4</td>
<td>0.756</td>
</tr>
<tr>
<td>0.60</td>
<td>457</td>
<td>1235</td>
<td>14.27</td>
<td>0.8</td>
<td>83.9</td>
<td>0.841</td>
</tr>
<tr>
<td>0.70</td>
<td>659</td>
<td>1662</td>
<td>15.11</td>
<td>0.8</td>
<td>129.2</td>
<td>0.900</td>
</tr>
</tbody>
</table>

* per mm roll width

(a)

<table>
<thead>
<tr>
<th>$\sigma_0$ (-)</th>
<th>$F_{\text{roll}}$ * (N)</th>
<th>$T_{\text{roll}}$ * (N-mm)</th>
<th>$\alpha_{\text{nip}}$ (°)</th>
<th>$\alpha_{\text{neutral}}$ (°)</th>
<th>$p_{\text{roll}}^{\text{max}}$ (MPa)</th>
<th>$\text{RD}^{\text{max}}$ (-)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.025</td>
<td>77</td>
<td>212</td>
<td>9.97</td>
<td>1.0</td>
<td>14.2</td>
<td>0.556</td>
</tr>
<tr>
<td>0.050</td>
<td>121</td>
<td>316</td>
<td>9.38</td>
<td>1.1</td>
<td>22.1</td>
<td>0.608</td>
</tr>
<tr>
<td>0.075</td>
<td>167</td>
<td>447</td>
<td>9.59</td>
<td>1.0</td>
<td>29.8</td>
<td>0.658</td>
</tr>
<tr>
<td>0.100</td>
<td>217</td>
<td>594</td>
<td>9.70</td>
<td>1.0</td>
<td>39.0</td>
<td>0.704</td>
</tr>
<tr>
<td>0.200</td>
<td>508</td>
<td>1378</td>
<td>10.21</td>
<td>1.1</td>
<td>93.7</td>
<td>0.858</td>
</tr>
<tr>
<td>0.300</td>
<td>918</td>
<td>2290</td>
<td>9.49</td>
<td>1.1</td>
<td>177.3</td>
<td>0.955</td>
</tr>
</tbody>
</table>

* per mm roll width

(b)
Figure 6.2.33. (a) Stress path in loading. (b) Initial stress paths during loading for RD-dependent material parameter simulations.
6.3. Conclusions

A two-dimensional, plane strain continuum model of the roller compaction of pharmaceutical powders was developed. To address the high shear conditions and accompanying deviatoric strains, an explicit formulation of the finite element method was used with adaptive meshing and Arbitrary Eulerian Lagrangian (ALE) capabilities. The model incorporated a pressure-dependent, frictional plasticity model, i.e., a modified DP/C model, to describe the stress-strain behavior of the powder over the range of relative densities experienced in the feed, slip and nip regions. The material parameters, e.g., cohesion, internal friction, cap parameter and $p_b$, were based on calibration studies for MCC. Simulations with $p_{roll}$-dependent frictional coefficient indicated appreciable differences in development of $p_{roll}$ and densification of the powder depending on the changes in roll friction with roll friction. The oscillating $\sigma_o$ conditions indicated anticipated periodic variations in $p_{roll}$ and exit relative densities. Variations in the through-the-thickness were significant in the slip region and diminished as the powder moved through the nip region. The two dimensional model predicted lower $F_{roll}$ and $p_{roll}$ than the corresponding one dimensional analyses. The model predictions followed similar trends observed in the published experimental studies (e.g., roll shear stress). The results indicate various combinations of boundary conditions and geometrical parameters can result in similar $p_{roll}$ (\(\alpha\)) profiles and densification. It is therefore important that accurate experimental inputs are used to verify of the model, i.e., simply adjusting several parameters to match a roll pressure profile does mean not the model is accurately representing the process.
Chapter 7. Three-Dimensional Finite Element Model

7.1. Introduction

Experimental results presented in Chapter 3 show that there is variation in stress along the transverse to the rolling direction. This variation may be due to the side seal friction and/or non-uniform feeding of powder to the rolls by the screw feeder. Such variation was - by definition - ignored in the 1-D and 2-D analyses. To explore the transverse variation of stress in roller compaction a 3-D FEM analysis is set up and is discussed in this chapter.

7.2 Three-Dimensional Finite Element Model

The development of the 3-D model was built from the 2-D analysis discussed in the last chapter. The mesh representing the deforming powder was defined by 20 x 9 x 54 array using 3-D continuum elements. Only one quarter of the geometry needs to be modeled due to symmetry. Figure 7.2.1 provide the mesh and the key boundary information. The roll was removed to allow viewing of the mesh representing the powder. The mechanical behavior of the powder was described using the Drucker-Prager/cap with the same material parameters used for MCC in Chapter 6 with $RD_o$ of 0.27. The explicit formulation within ABAQUS with the arbitrary Lagrangian Eulerian (ALE) and adaptive meshing features were used to handle the non-linear contact conditions and severe distortion of the mesh due to the high shear conditions induced in rolling. Similar to the 2-D model, Eulerian inflow and outflow boundaries were employed. The roll, seal side and screw housing were represented by rigid surfaces. The
contact between the deformable mesh and these surfaces were defined by Coulomb’s frictional law \( \mu_{\text{screw}} = 0 \) on screw housing and variable on roll/powder and powder/side seal interfaces). The entry angle was 20°.

The following sets of 3-D simulations were run:

- Three simulations with the Coulomb coefficient of friction between powder and side seal at \( \mu_{\text{side seal}} = 0, \mu_{\text{side seal}} = 0.2 \) and \( \mu_{\text{side seal}} = 0.35 \). In this simulation the feed stress was kept at 0.1 MPa.
- Two simulations with constant (35 mm/s) and variable (32 mm/s at the sidewall to 38 mm/s at the center in a linear manner) inlet velocity to “simulate” the effect of non-uniform feeding due to the presence of the screw. The friction coefficient between powder and side seal was 0.35.

For all the simulations above, \( 2h/D = 0.02 \) and \( W/D = 0.375 \).

For the simulations in which the effect of side seal friction was investigated, the powder entered the Eulerian inflow boundary with an initial RD of 0.27 and was immediately subjected to uniform feed stress of 0.100 MPa. In the simulations involving of non-uniform feeding from the screw, the inflowing material was assigned an initial inlet velocity. In one case this was uniform and in another case, the inlet velocity decreased linearly along the roll width from a maximum in the center to the minimum at the side seal. For simplicity and to isolate the effect of the uniform feeding, the side seal friction was zero.
**Figure 7.2.1.** Mesh, screw housing and side seal for 3-D simulations. Roll was removed for clarity.
7.3. Results and Discussion

In this section, we compare numerical predictions the two sets of simulations examining the effect of side seal friction and inlet feed velocity. All the results presented correspond to steady state conditions.

7.3.1. Effect of Side Seal Friction

Figure 7.3.1. shows the maximum roll pressure along the transverse (roll width) direction for the three frictional cases for the seal side. In the frictionless case, the maximum roll pressure is uniform across the transverse direction. As the side seal friction increased the roll pressure decreases as the side seal friction counteract the effect of the feed stress. The roll pressure also is reduced close to side seal with the highest side seal friction creating the largest reduction. The corresponding maximum relative density is plotted in Figure 7.3.2. It is evident that increasing the side seal friction results in lower densification and greater variation in the transverse direction. This is the reason why side seals are typically manufactured with materials with low friction properties.

7.3.2. Effect of Variable Inlet Velocity

In these simulations in which the side seal friction is zero and the inlet feed velocity is either uniform or linearly decreasing from the center to the edge. By adjusting the inlet velocity of the feed powder, the flux of powder entering rolls varies. This simulation is an attempt to understand aspects of inhomogeneity that may be associated with the presence and operation of the screw. In the case of uniform inlet velocity the
maximum roll pressure and corresponding maximum relative density was the same along the transverse direction as shown in Figures 7.3.3 and 7.3.4, respectively. Since there is no side seal friction, the uniform inlet velocity results in a uniform development of roll pressure. When the inlet velocity varies, i.e., the powder is preferentially fed to the center versus the edge, higher maximum roll pressures are developed toward center than the edge near the side seal. Figure 7.3.5 (a) and (b) provides the profiles of roll pressure and roll shear stresses across the roll width, respectively. The nip angle was the same across the roll but higher roll pressure and corresponding shear stresses developed in the center where material was preferentially fed, i.e., the high inlet velocity resulted in higher amount of material being fed to the center and consequently high density and roll pressures. In Figure 7.3.6, the profiles of the normalized roll pressure with respect to the maximum roll pressure are plotted. The profiles are super-imposable.

The results of these simulations indicate that both the side seal friction and the non-uniform feeding of the powder to the rolls may be contributing to the non-uniformity in roll pressure and density across the roll width. Lower densification at the edges has been attributed to the frictional resistance of the side seal to the flow of powder. Improvements in the densification of the edges have been identified through 3-D FEM simulation by in which the side seal rotated (Wang and Zavaliangos, 2004). The screw feeder may also contribute to the non-uniformity across the transverse. The powder transitions from a circular cross-section related to the screw housing to a rectangular cross section associated with the rolls. The powder must re-distribute in this region of the feed zone. In addition, the oscillating screw is delivering material in a non-uniform manner across the roll width. These variations have been observed especially when the
tip of the screw is very close to the nip where there is inadequate opportunity for the powder to rearrange. These variations are reflected in the maximum roll pressures generated and the density developed along the width. The screw feeder not only conveys the powder to the rolls but also generates a feed stress that varies depending on the screw relative position and the material distribution within the feed zone. The information on the spatial distribution of the material in terms of the relative density in the feed zone along with the feed force distribution in the feed zone are important initial and boundary conditions to be considered in understanding roller compaction in three dimensions.

With the exception of Dec and Komarek (1993), there has not been sufficient research to understand the details of the screw feed systems.
Figure 7.3.1. Maximum roll pressure along the transverse direction based on distance from the center (with 0 mm at the center and 37.5 mm at the side seal) for side seal friction of $\mu_{\text{side seal}} = 0$, 0.20 and 0.35.

Figure 7.3.2. Maximum relative density along the transverse direction based on distance from the center (with 0 mm at the center and 37.5 mm at the side seal) for side seal friction of $\mu_{\text{side seal}} = 0$, 0.20 and 0.35.
Figure 7.3.3. Maximum roll pressure along the transverse direction based on distance from the center (with 0 mm at the center and 37.5 mm at the side seal) for uniform and non-uniform inlet velocity.

Figure 7.3.4. Maximum roll pressure along the transverse direction based on distance from the center (with 0 mm at the center and 37.5 mm at the side seal) for uniform and non-uniform inlet velocity.
Figure 7.3.5. (a) Profile of roll pressure versus rolling angle at various positions along the transverse direction from the center. (b) Profile of roll shear stress versus rolling angle at various positions along the transverse direction from the center.
Figure 7.3.6. Profile of normalized roll pressure versus rolling angle at various positions along the transverse direction from the center.
7.4. Conclusions

A 3-dimensional finite element model was developed based on the 2-dimensional plane strain model. The 3-D model allows examination of three-dimensional effects like side seal friction and non-uniform feeding that could not be examined using the previous 2-D model. High friction at the side seal resulted in lower pressure generation. Non-uniform influx of material to the rolls results in non-uniform stress development and densification across roll width.
Chapter 8. Comparison of Experiment to Modeling

In this chapter a brief comparison of the model predictions with the experimental results is discussed – both qualitatively and quantitatively. The roller compactor used in the experimental set up included a screw feeder system, which better emulates typical roller compactors used in practice. The feed system has a major effect on the initial relative density of powder, $RD_o$, as it is fed to the rolls and the feed stress, $\sigma_o$, developed – two essential initial and boundary conditions for the model. The effect of the screw is evident in the dramatic variations in the roll pressure profiles across the roll width and rolling direction and quality of the formed compact, e.g., density distribution. Direct one-to-one comparison of the experimental and model is thus difficult due to lack of experimental information on the spatial and time information for $RD_o(x, y, t)$ and $\sigma_o(x, y, t)$ related to the feed system. The use of a more simplified feeding system including gravity feed and use of a constant head pressure using a plate in contact with the feed powder with various dead weights were attempted with microcrystalline cellulose (MCC) without success. MCC did not have sufficient inherent flowability to be fed by gravity alone. In the case of the constant head pressure, MCC would compact within the feed hopper and form a compacted plug that could not be fed further into the rolls. A more controlled application of the head pressure with a feedback loop may have overcome these issues with MCC but such a design was not developed within the scope of this research. A design similar to Guigon. et al., (2002 and 2003), i.e., use of a plunger-type feeder in which the powder was delivered to the rolls with a more uniform loading across the transverse direction, would have enabled a more well-defined feed condition to be
established. To objectively assess any process model, it is essential to have appropriate experimental input, e.g., constitutive model and initial and boundary conditions. This is particularly important with roller compaction since various factors can have similar effects on important parameters like roll pressure and density of resulting compact. Nevertheless, the following comments can be made when comparing the experimental results outlined in Chapter 3 and the modeling results discussed in Chapter 5, 6 and 7:

- The experimental determination of the nip angle was approximately 8° for MCC for the smooth roll configuration. The 2-D model predicted nip angles in this range when the roll/powder frictional coefficient was approximately 0.3 to 0.4, which is reasonable for MCC with low internal lubrication.

- The predicted and experimental observations indicated that the neutral angle was prior to roll centerline.

- The maximum roll pressure was predicted and observed experimental to occur just prior to roll centerline (smooth roll).

- When the 2-D model included an oscillating feed force, the resulting maximum roll pressure and corresponding densification oscillated, which was similar to that observed in the experimental studies.

- The 3-D model predicted that side seal friction and variable inflow of feed powder results in corresponding variation in densification in the roll width. These effects may be contributing to the similar in trends seen in the experimental studies.
Experimentally, increasing the feed force increased the roll gap for a given roll force. This trend is similar to the model prediction that increasing the feed stress increased the roll gap for a given feed force.

Both the experimental studies and model prediction indicated that – for a given material – the maximum relative density was related to the maximum roll pressure regardless of the specific roller compactor or process settings.

Since the roll force is directly related to the roll pressure, a close correlation of the maximum relative density and the roll force existed for both the experimental and modeling results.

While the models predicted several of the experimental trends, it did overestimate the level of densification achieved at a given maximum roll pressure (Figure 8.1). The experimental relative density for MCC is compared to that predicted by the 2-D FEM model. As discussed in Chapter 6, the 2-D model predicted excessive dilation of the compacted powder upon release from rolls since the stress path upon unloading fells onto the Drucker-Prager shear failure line (which predicted dilation as part of its definition of the flow potential). The 2-D model consequently used the maximum relative density (under load) as the reference relative density. Thus, part of the disagreement of the experiment and model results is related to the expansion that occurs in the experimental results. The level of expansion of MCC was predicted to be approximately 2° based on the linear elastic analysis discussed in Chapter 5. MCC, however, expands significantly more as indicated by the approximately 4° release angles observed experimentally in Chapter 3. This additional expansion of MCC is also seen in die compaction (Figure 8.2) in which the relative density decreases by close to 10% for axial stresses comparable to
those observed in the experiments and models. Accounting for this excessive expansion brings the predicted results closer to the experimental results (Figure 8.1). The additional reason for the discrepancy is the non-linearity of the elastic unloading in low cohesion materials. It is often observed with materials such as MCC that the compacted powder rapidly expands in the final stages of unloading within a die (Cunningham, et al., 2001; Procopio, 2006).
Figure 8.1. Relative density of the 2-D FEM model and the experimental results. The 2-D model results - corrected for expansion of the MCC – are also provided.

Figure 8.2. Relative density of the MCC during die compaction at various maximum axial stress (under maximum loading and upon ejection).
Chapter 9. Conclusions and Future Work

9.1. Conclusions

The following conclusions may be drawn the series of experimental and modeling studies conducted on roller compaction of pharmaceutical powders:

i) Experimental studies of roller compaction:

- An instrumented roll was designed, constructed and used to study the effect of various process parameters and system response.
- Key experimental observations include:
  - In a roll force controlled system, the exit $RD$ correlates only with maximum roll pressure (over range of gap settings studied)
  - Increasing feed force increased roll gap for given roll force (floating roll design)
  - Relative density was independent of roll gap (over range studied)
  - Nip angle for the smooth roll with microcrystalline cellulose was approximately 8°
  - Neutral angle preceded centerline of rolls (smooth and textured rolls)
  - Maximum roll pressure was about 1° before centerline of rolls (smooth rolls) and near the centerline (textured roll)
  - Roll shear stress gradually builds on entrance side of roll, reverses at the neutral angle and reaches relatively high values at exit side, which
may contribute to defect formation, e.g., lamination of the compacted strips

- Significant variation in roll pressure was observed in both the rolling and roll width directions, which is related to oscillatory nature of screw feeder and to friction with the side seals

ii) Characterization of Powder Mixtures

- Developed a calibration technique using simple mechanical tests and principles of plasticity theory (normality).
- Friction coefficient was found to be variable as function of normal stress and/or relative density.
- Linear elastic, modified Drucker-Prager/cap constitutive model (with RD dependencies) and variable frictional coefficient were implemented and validated in die compaction finite element model based on local density variation.

iii) Modeling of Roller Compaction

- 1-Dimensional
  - corrected stress in roll width direction used in previous slab analysis
  - did not require assumption on neutral angle or position of maximum roll pressure
  - highlighted nip angle strongly dependent on roll friction and
- feed conditions (initial RD, entry angle and feed stress) significantly affect development of contact stress and densification

### 2-Dimensional

- demonstrated significant variation in stress, strain and velocity in the thickness direction, which could not be examined previously in 1-D models
- provided a closer prediction to experimental than 1-D
- nip angle is strongly affected by roll friction and approximately independent of feed stress, roll gap, initial density and entry angle
- Entry angle, feed stress, roll friction, initial density, low pressure constitutive behavior significantly affect pre-densification in feeder and slip regions, which in turn significantly affected the achievable densification
- Importance of oscillatory feed stress and variable roll friction were demonstrated
- Different combination of parameters (e.g., entry angle and roll friction) may result in similar development of contact stresses and densification due to similar effects on pre-densification in feeder and slip region
- For given material, various process conditions result in a common relative density vs maximum roll pressure curve

### 3-Dimensional

- Presence of side seal friction resulted in reduction in maximum roll pressure at the near stationary seals was observed
Non-uniform feeding resulted in non-uniform development of maximum roll pressures

Three-dimensional effects can be effectively examined with finite element model

iv) Experimental Results vs Model Predictions

- Model effectively predicted trends for variation in roll pressures with varying feed force and position of nip and neutral angles
- Model predicted trends for roll gap, roll force and feed force and relationship of maximum roll pressure and densification
- Due to deficiencies in accounting for the expansion upon release, model over-estimates roll pressure required to achieve final densification

9.2. Recommendations for Future Research

The following recommendations are made for future work:

i) Experimental

- More accurate calibration of constitutive model in the low to medium triaxial stress states is required to improve predictions, e.g., use of axisymmetric triaxial testing, and of the frictional model at relevant roll pressure and relative densities of the material;
Measurements of feed stress and relative density in feed region are essential as more accurate inputs for the initial and boundary conditions

ii) **Modeling**

- To further improve the confidence in the present models, a more simplistic experimental set up of roller compaction should be considered, e.g., use a controlled piston feeder that induce more uniform stress across the roll width and roll direction. Verification of the roll force, roll contact stress and resulting densification will provide greater confidence to extent the model into more complicated feed designs. As demonstrated in this research, the various combinations of geometrical and material factors can results in similar behavior, e.g., roll pressure profiles. To objectively verify any model, accurate inputs across the range of relevant process conditions must be considered.

- A more accurate frictional model may need to be developed to understand the design of the roll surface and its frictional and geometrical features with reference to the mechanical behavior of the deforming powder. A simplified model, e.g., plate-geometry, may help understand the salient features of the design, friction and material properties. More experimental investigation on the location of the nip and relative velocities of the powder and roll will help elucidate the friction at the roll.
Further development of the 3-D model based on representative experimental data on feed conditions (screw feeder) and side seal friction will advance understanding of process and design.

Improved understanding of defects is needed through combination of improved material model that accounts for stress history, anisotropic effects and low triaxial behavior (stress and deformation).
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Appendix A Calibration of modified Drucker-Prager/Cap

A.1. Stress invariants

The stress state at a material point is unique. Its representations with respect to two different coordinate systems are connected through the classic rotation transformation for 2nd order tensors

\[ \sigma'_{ij} = \sum_{k=1,3} \sum_{l=1,3} R_{ki} \sigma_{kl} R_{lj} \]  

(A.1)

where \( R_{ij} \) is the rotation matrix between the two coordinate systems. Invariants are mathematical functions of the stress components that are independent of the coordinate system orientation and are invariant under a rotation transformation such as in (A.1). There are only three independent invariants for a second rank tensor such as stress or strain and their increments (Spencer, 1971). The most common invariants used in compaction modeling are the hydrostatic pressure and the Mises stress defined in equation (4.5).

A third independent invariant exists, but for simplicity is not considered in compaction models. Experimental measurements of its effect indicate that either the effect is small or within the experimental error (Mosbah et al., 1997).

A.2. Determination of modified Drucker-Prager/cap parameters

The modified Drucker-Prager/cap model is composed of two regions – the Drucker-Prager failure line and the elliptical cap. The intercept in \( p-q \) space, i.e., the cohesion, and the slope, i.e., the angle of internal friction, can be determined from two
points on the failure line. These data can be provided by a number of tests (see Figure 4.2.1(a). Here we suggest diametrical compression and simple compression because of the simplicity of their geometry. Diametrical compression is a common mechanical test to determine the tensile strength of brittle materials. A circular disc is loaded diametrically until failure and tensile stresses develop along the transverse to loading direction. The stress state at the center of the disk where fracture occurs (according to the Hertz solution) is given by:

\[
\begin{bmatrix}
\sigma_{xx} & \sigma_{xy} & \sigma_{xz} \\
\sigma_{xy} & \sigma_{yy} & \sigma_{yz} \\
\sigma_{xz} & \sigma_{yz} & \sigma_{zz}
\end{bmatrix}
\begin{bmatrix}
\sigma_T \\
0 \\
0
\end{bmatrix}
\begin{bmatrix}
0 \\
-3\sigma_T \\
0
\end{bmatrix}
\]

(A.2)

where \( \sigma_T = \frac{2P}{\pi D t} \)  

(A.3)

Using the definitions in equation (4.5), we obtain:

\( p = \frac{2}{3}\sigma_T \), and \( q = \sqrt{13}\sigma_T \)  

(A.4)

The stress state in simple compression is given by:

\[
\begin{bmatrix}
\sigma_{xx} \\
\sigma_{yy} \\
\sigma_{zz}
\end{bmatrix}
\begin{bmatrix}
-\sigma_c \\
0 \\
0
\end{bmatrix}
\begin{bmatrix}
0 \\
0 \\
0
\end{bmatrix}
\]

(A.5)

where \( \sigma_c = \frac{4P}{\pi D^2} \)  

(A.6)

where \( P \) and \( D \) are the failure load and diameter of the cylindrical compact, respectively. Using the definitions in equation (4.5), we obtain:

\( p = \frac{1}{3}\sigma_c \) and \( q = \sigma_c \)  

(A.7)
Cohesion, \(d\), and the slope representing the internal angle of friction, \(\beta\) can be determined by solving the system of equations formed by substituting equations (A.4) and (A.7) in equation (4.10):

\[
d = \frac{\sigma_c \sigma_T (\sqrt{13} - 2)}{\sigma_c - 2 \sigma_T} \quad \text{and} \quad \tan(\beta) = \frac{3(\sigma_c - d)}{\sigma_c}
\] (A.8)

To define the elliptical cap region as expressed in equation (4.11) with \(\alpha = 0\), the material parameters of \(p_a\) and \(R\) are needed. These parameters can be determined based in information derived from a die compaction test and the use of the yield function and plastic flow potential. At low friction at the powder/tooling interface (shear stress components of the stress tensor are zero) and assuming the die is perfectly rigid (no radial displacement), the stress tensor for a cylindrical compact in \((r, \theta, z)\)-coordinates are:

\[
\sigma_{ij} = \begin{pmatrix} \sigma_{rr} & \sigma_{r\theta} & \sigma_{rz} \\ \sigma_{r\theta} & \sigma_{\theta\theta} & \sigma_{\theta z} \\ \sigma_{rz} & \sigma_{\theta z} & \sigma_{zz} \end{pmatrix} = \begin{pmatrix} \sigma_r & 0 & 0 \\ 0 & \sigma_\theta & 0 \\ 0 & 0 & \sigma_z \end{pmatrix}
\] (A.9)

Considering the angular and radial components of the stress tensor are equal, i.e., \(\sigma_r = \sigma_\theta\), the hydrostatic pressure, \(p\), and Mises effective stress, \(q\), are given by:

\[
p = -\frac{1}{3} (\sigma_r + \sigma_r + \sigma_z) = \frac{1}{3} (2 \sigma_r + \sigma_z)
\] (A.10)

\[
q = |\sigma_z - \sigma_r|
\] (A.11)

Given that in compaction \(\sigma_z < \sigma_r < 0\), equation (A.11) can be written as

\[
q = \sigma_r - \sigma_z
\] (A.12)

The presence of a non-zero shear stress component \(\tau_{rz} = \mu \sigma_{rr}\) due to wall friction introduces a small error in the calculation of \(q\). This percentage error is proportional to
\[
\left( \sqrt{1 + 3 \mu^2 / \left(1 - \frac{\sigma_z}{\sigma_r}\right)^2} - 1 \right). 
\]
Using our frictional data for the wall lubricated die (Figure 4.3.4) and the corresponding values of axial and radial stresses, we can observe that it is less than 1% in the whole range of relative densities for the lubricated die.

The parameters \(R\) and \(p_a\) can be obtained by solving the following system of two equations

(a) the yield condition

\[
F_c = \sqrt{\left( \frac{1}{3} \sigma_z + \frac{2}{3} \sigma_r - p_a \right)^2 + R^2 \left( \sigma_z - \sigma_r \right)^2 + R(d + p_a \tan \beta) = 0} 
\] (A.13)

and (b) assuming that permanent change in radial dimension is zero:

\[
d \varepsilon_r^{pl} = d \lambda \cdot \left( \frac{\partial G_c}{\partial \sigma_r} \right) = 0 
\] (A.14)

or equivalently

\[
\frac{\partial G_c}{\partial \sigma_r} = \frac{\partial G_c}{\partial q} \frac{\partial q}{\partial \sigma_r} + \frac{\partial G_c}{\partial p} \frac{\partial p}{\partial \sigma_r} = 2R^2 q - \frac{4}{3} \left( p - p_a \right) = 0 
\] (A.15)

For a given stress state, equations (A.13) and (A.15) can be used to solve for \(R\) and \(p_a\).

\[
p_a = \frac{3q - 4d \tan(\beta) + \sqrt{9q^2 - 24d \tan(\beta)q - 24 \tan(\beta)^2 pq - 16 \tan(\beta)^2 q^2}}{4 \tan(\beta)^2} 
\] (A.16)

\[
R = \sqrt{\frac{2}{3q} \left( p - p_a \right)} 
\] (A.17)

Equation (A.14) is an approximation. The solution of the full problem that includes the elastic deformation is also possible. In that case

\[
d \varepsilon_r = d \varepsilon_r^e + d \varepsilon_r^{pl} = \frac{(1 - \nu)d \sigma_r}{E} - \frac{\nu d \sigma_z}{E} + d \lambda \cdot \left( \frac{\partial G_c}{\partial \sigma_r} \right) = 0 
\] (A.18)
\[
d\varepsilon_z = d\varepsilon_z^e + d\varepsilon_z^{pl} = \frac{d\sigma_z}{E} - \frac{2\nu d\sigma_r}{E} + d\lambda \cdot \left( \frac{\partial G_c}{\partial \sigma_z} \right) \tag{A.19}
\]

Equations (A.18) and (A.19) and \( F_c = 0 \) can be solved for \( \lambda, p_a, \) and \( R \).

### A.2. Determination of the Elastic Parameters

The elastic parameters can be estimated from the unloading data of die compaction studies in which the axial and radial stresses and axial strain are measured. In addition, the following assumptions are made:

1. the die wall perfectly rigid;
2. friction is ignored and no porosity variation exists in the specimen; and
3. compact can be represented by a linear elastic, isotropic material.

Equation (4.8) can be then written in cylindrical coordinates as:

\[
d\varepsilon_z^e = \frac{d\sigma_z}{E} - \frac{\nu}{E} (d\sigma_r + d\sigma_{\theta\theta}) \tag{A.20}
\]

\[
d\varepsilon_r^e = \frac{d\sigma_r}{E} - \frac{\nu}{E} (d\sigma_r + d\sigma_z) \tag{A.21}
\]

Given that the radius of the specimen remains constant during unloading and that
\[
d\sigma_r = d\sigma_{\theta\theta} \text{ due to equilibrium},
\]

\[
\nu = \frac{d\sigma_r}{d\sigma_r + d\sigma_z} = \frac{\frac{d\sigma_r}{d\varepsilon_r^e}}{\frac{d\sigma_z}{d\varepsilon_z^e}} \frac{d\varepsilon_r^e}{d\varepsilon_z^e}, \quad E = \frac{d\sigma_z}{d\varepsilon_z^e} - 2\nu \frac{d\sigma_r}{d\varepsilon_z^e} \tag{A.22}
\]
Appendix B  Determination of Coefficient of Friction using Differential Slice Method

Consider a cylindrical compact of diameter $D$ and height $H$ and an elemental slice $dz$ situated at distance $z$ from the bottom of the compact, as presented in Figure B.1.

Neglecting the weight of the powder, the force balance equation is:

$$\frac{\pi D^2}{4} d\sigma_z = \tau_{rz} \pi D dz$$  \hspace{1cm} (B.1)

This is strictly true only if it is assumed that the vertical stresses are uniform in any horizontal cross-section of the material. The radial stress at the given height $z$ can be written as:

$$\sigma_{rr} = K \sigma_z$$  \hspace{1cm} (B.2)

where $K$ is generally referred to as the Janssen constant, radial pressure transmission coefficient or radial to axial stress ratio. The shear stress due to friction acting on the elemental slice can be expressed according to Coulomb’s law of friction:

$$\tau_{rz} = \mu \sigma_{rr}$$  \hspace{1cm} (B.3)

Combining equations (B.2) and (B.3), the equilibrium equation becomes:

$$\frac{d\sigma_z}{\sigma_z} = \frac{4K\mu}{D} dz$$  \hspace{1cm} (B.4)

If it is assumed that the product $K\mu$ is independent of position $z$, Equation (B.4) can be integrated using the appropriate boundary conditions:

$$\ln \frac{\sigma_z}{\sigma_b} = \frac{4K\mu}{D} z \quad \text{or} \quad \ln \frac{\sigma_r}{\sigma_b} = \frac{4K\mu}{D} H$$  \hspace{1cm} (B.5)
Equation (B.5) implies an exponential distribution of the axial stress at a given height in the compact:

\[ \sigma_z = \sigma_B \exp\left(\frac{4K\mu}{D}z\right) \]  

(B.6)

or

\[ \sigma_z = \sigma_T \left(1 - \frac{z}{H}\right)^{\frac{z}{H}} \]  

(B.7)

The average axial stress in the compact can be obtained by integrating over the height:

\[ \sigma_{z(average)} = \frac{1}{H} \int_0^H \sigma_z \, dz = \frac{\sigma_T - \sigma_B}{\ln \frac{\sigma_T}{\sigma_B}} \]  

(B.8)

This manipulation is standard and is presented widely in the literature. The radial stress in the die at the sensor level \( \sigma_{rz} \) is measured directly. Therefore the radial to axial stress ratio at this point can be determined as:

\[ K_z = \frac{\sigma_{rz}}{\sigma_{zz}} \]  

(B.9)

Solving equations (B.6), (B.8) and (B.9) we obtain an estimate of the friction coefficient:

\[ \mu = \frac{D}{4H} \frac{\sigma_B}{\sigma_{rr}} \left(\frac{\sigma_T}{\sigma_B}\right)^{\frac{z}{H}} \ln \frac{\sigma_T}{\sigma_B} \]  

(B.10)

Ideally the die compaction experiments used for model calibration must be performed in a double action press, while the friction measurements need to be performed in a single action press. Practically speaking single action press experiments are adequate as long as the friction coefficient is low, as the inhomogeneity of the specimen is kept to a
minimum. To appreciate the level of variation present in a tablet used in our experiments, we note that $K \approx 0.5$, $m = 0.1$ then for $H/D \approx 0.3$ the lower stress is 6% less than the top stress. Although this difference is adequate to evaluate friction, it is small enough to allow the approximation of the stress state with average values of $p$ and $q$. For relative densities larger than $RD > 0.6$ the difference between top and bottom punch forces is less than 6% and decreases with increasing relative density.

Figure B.1. Analysis of instrumented die data
Appendix C  List of Symbols

\( a, \alpha \equiv \) angular position

\( a_{\text{entry}}, \alpha_{\text{entry}} \equiv \) entry or delivery angle

\( a_{\text{neutral}}, \alpha_{\text{neutral}} \equiv \) neutral angle

\( a_{\text{nip}}, \alpha_{\text{nip}} \equiv \) nip angle

\( a_{\text{release}}, \alpha_{\text{release}} \equiv \) release angle

\( A \equiv \) cross-sectional area; material parameter describing elliptical yield surface

\( B \equiv \) material parameter describing elliptical yield surface

\( D \equiv \) diameter

\( d \equiv \) cohesion

\( E \equiv \) Young’s modulus

\( F \equiv \) yield function

\( F_c \equiv \) force in uniaxial compression

\( F_c \equiv \) yield function in cap region

\( F_S \equiv \) yield function in Drucker-Prager shear region

\( F_B \equiv \) bottom punch force

\( F_T \equiv \) top punch force

\( G \equiv \) elastic shear modulus

\( G \equiv \) plastic flow potential

\( G_c \equiv \) plastic flow potential in cap region

\( G_S \equiv \) plastic flow potential in Drucker-Prager shear region

\( H \equiv \) height of specimen
\( H_0 \equiv \) initial height of specimen

\( H \equiv \) thickness (\( y \)-direction) of element

\( h_o \equiv \) half of gap at centerline of rolls

\( k_m \equiv \) state variable, \( m = 1, 2, 3, \ldots n \)

\( K \equiv \) elastic bulk modulus

\( L_{ijkl} \equiv \) elastic stiffness

\( m \equiv \) mass

\( P \equiv \) force

\( p \equiv \) hydrostatic pressure

\( p_u \equiv \) pressure evolution parameter

\( p_b \equiv \) hydrostatic yield pressure

\( p_r, p_{\text{roll}} \equiv \) roll pressure

\( q \equiv \) Mises equivalent or effective stress

\( R \equiv \) cap eccentricity parameter

\( R \equiv \) roll radius

\( RD \equiv \) relative density

\( RD_0 \equiv \) initial relative density

\( t \equiv \) thickness

\( t_r, t_{\text{roll}} \equiv \) roll shear stress

\( U_B \equiv \) bottom punch displacement

\( U_T \equiv \) top punch displacement

\( V \equiv \) volume
Z ≡ axial position

α ≡ cap transition parameter

β ≡ angle of internal friction

ε_{ij} ≡ total strain tensor

ε^e_{ij} ≡ elastic strain tensor

ε^d_{ij} ≡ plastic strain tensor

ε_{ii} ≡ normal strains (i = 1, 2, 3)

ε_{zz} ≡ axial strain

γ_{ij} ≡ shear strain

μ ≡ coefficient of powder/die friction or powder/roll

ν ≡ Poisson’s ratio

σ_{ax} ≡ axial stress

σ_c ≡ uniaxial compressive strength

σ_{ij} ≡ stress tensor

σ_{ii} ≡ normal stress components, i = 1, 2, 3

σ_B ≡ bottom punch stress

σ_T ≡ tensile strength for diametrical compression

σ_T ≡ upper punch stress

σ_{rad} ≡ radial stress

σ_{rr}^z ≡ radial stress at axial position z

ρ ≡ density

τ ≡ tangential stress
\( d\lambda \equiv \text{plastic strain multiplier} \)
Vita

John C. Cunningham

Education

B.S.  1986  Drexel University  Materials Engineering
M.S.  1989  Drexel University  Biomedical Engineering

(MS Thesis: *RF Plasma Modification of Surfaces for Improve Blood Biocompatibility*)

Employment

Johnson & Johnson Pharmaceutical R&D, Spring House, PA, 2005 to present

Merck Research Laboratories, Pharmaceutical R&D, West Point, PA  1989 – 2004

Academic and Professional Honors

Society of Biomaterials Student Scholarship, 1989
Ernest J. Calhoun Biomedical Engineering Graduate Fellowship, 1986-1989
Alpha Sigma Mu Materials Engineering Scholarship, 1985
Foundry Educational Foundation Scholarship, 1985
American Society of Metals A.W. Grosvenor Scholarship, 1985
Dean’s List 1985, 1986
Hans Dresler Foundation Scholarship, 1983

Publications/Proceedings (peer-reviewed)
