# International Fine Particle Research Institute
# Renewal Proposal
# Modeling defect formation during powder compaction with application to tableting failure
# ARR-108-01

Ken Kamrin and William Zunker
Massachusetts Institute of Technology

December 20, 2023

# 1 Executive Summary

The focus of this project is to understand the physical mechanisms that lead to defect formation – pitting, cracking, and delamination – during pharmaceutical tableting. One hypothesis among IFPRI members is that trapped interstitial air leads to high pore pressures that tend to fracture adhered particle interfaces after removal of the confining pressure. Another is that residual stress from the tableting procedure fractures the tablet upon punch release. The project objective is to explore this problem through coupled numerical methods including: i) continuum mixture models and ii) the discrete element method (DEM) coupled with a fluid solver. The primary barrier to using these methods is that fact that the behavior of cohesive powders is not well understood, with neither a generally accepted constitutive relation nor contact model in existence.

To address this, the project so far has emphasized developing a reliable cohesive powder contact model for usage in DEM. This is the natural progression, since a powder DEM model will be indispensable in determining a constitutive relation for continuum simulations. In particular, we have concentrated on creating a mechanically-derived contact model for adhesive elastic-perfectly plastic particles that can potentially model an array of powder types.

In year one of the project, the majority of the theoretical framework for the contact model was developed, exploiting a subtle trick from mechanics called the "Method of Dimensionality Reduction," which allowed us to capture significant particle plasticity of the grains within a contact model framework, as well as represent rigorously adhesive particle contact. Still a number of issues remained. The JKR-type adhesion of the contact model needed to be validated once significant plastic deformation had occurred and the scheme to respect plastic incompressibility required an overhaul. The contact model was limited to simple symmetric loadings of a single particle, necessitating adaptation to many-particle interactions. Additionally, the model, initially coded in Matlab, needed implementation in an established software like LIGGGHTS or LAMMPS, with a reliable fluid-solid coupling strategy.

In year two, the theoretical framework was completed by validating the adhesive model within the fully-plastic regime and correcting the plastic incompressibility scheme. The completed contact model was published in the premier solid mechanics journal, *Journal of Mechanics and Physics of Solids*, as a two part series [1, 2]. Efforts have extended to modeling many-particle interactions, a necessary step to allow simulation of full-scale industrial applications. Implementation into LIGGGHTS is underway and preliminary simulations show promise in replicating compaction simulator data. In the upcoming phase, we are working directly with Sandia National Laboratories to create a LAMMPS implementation. In addition to greater computational efficiency and formal support for the model, LAMMPS provides the advantage of allowing immediate coupling with a multi-particle collision dynamics fluid solver, capable of simulating a compressible gas phase.

In summary, the project has made substantial progress on the front of creating a reliable powder DEM model and we are now poised at an exciting place where beginning to understand defect formation during pharmaceutical tableting is tangible. Because the model is mechanically-derived, it can also be trusted to assist in the future development of a continuum constitutive relation. Finally, because of the planned open-source nature of the implementation into LAMMPS, a familiar software to most IFPRI members, the powder DEM model can be used for industrial applications extending beyond tableting.

Moving forward, there is still much to do. In order to provide IFPRI with a complete simulation of capping over a survey of possible input conditions (e.g. with air, without air, and various levels of grain cohesion) we need to complete the large-scale DEM capability and introduce the air phase. This is the focus of much current work on our side, in collaboration with Sandia. See Section 3 for details.

After that, we propose to perform a completely continuum-level treatment of this problem based on two-phase mixture theory. This tool will have many applications beyond tableting, such as situations where grain numbers exceed billions. See Section 4. The Kamrin group has been simulating grain-fluid mixtures

as full continua since 2019 and has developed multiple numerical tools to solve this problem. However, these tools require a generalizable and accurate constitutive relation for the solid phase, here the powder bulk. Without this, any such simulation would rightfully qualify as just "Colorful Fluid Dynamics"! For this, reason the vast majority of our work so far has been to produce a DEM model that can be used as our digital twin to obtain a shear- and volumetric- plasticity continuum model for the powder bulk, whose material parameters are knowable as functions of the grain properties. The connection back to the grain properties is key — powders are exponentially more diverse than hard cohesionless grains, and thus the modeling parameters for two different powder specimens cannot be expected to be at all similar. Thus, the relation back to the grain details is key to avoid a massive fitting exercise as the model is applied to different powders.

Using the powder DEM model as a digital twin, we will develop and calibrate a soil-mechanics-based model with an added damage variable to represent generic powder bulks. We will then implement the model numerically as part of an air-continuum mixture model using the two-phase Material Point Method (MPM). By comparing tableting simulations at the continuum level to those of the digital twin including the air phase, and then directly to experimental tableting failures, we can evaluate the capabilities of the two-phase mixture framework for this set of applications.

# 2 What we have done already

## 2.1 Focusing the project objective

The objective of this project is ultimately to develop an understanding of the underlying physical mechanisms that lead to defect formation during powder compaction. Of chief interest to the members of IFPRI is defect formation – pitting, cracking, and delamination – during pharmaceutical tableting. A popular hypothesis shared amongst IFPRI members is that trapped interstitial air leads to high pore pressures that tend to fracture adhered particle interfaces once the confining pressure of the punch is removed post compaction. Coupled numerical methods involving a fluid and solid phase provide a viable avenue for testing this hypothesis and uncovering other important defect inducing mechanisms. At present, there are two primary numerical methods for the problem at hand: i) continuum mixture models and ii) the discrete element method (DEM) coupled with a fluid solver. Although the general infrastructure of these techniques is robust and well-developed, neither are currently equipped to understand defect formation during powder compaction. This is because the behavior of cohesive powders is still not well understood. This fact was discussed openly at the panel of the 11th IFPRI Workshop - Modeling of Powder Flow held prior to the 2023 IFPRI AGM, and **obtaining a reliable DEM model for powder grains was identified as a solution route for this difficult problem.**

For continuum mixture models, the issue becomes that there is no generally accepted constitutive relation for cohesive powders that can be calibrated robustly and easily to different powder grains. Without it, meaningful simulations cannot be conducted, even if the other aspects of the model exist such as how to model the compressible gas phase. Therefore, constructing a physically justified constitutive relation for cohesive powders is a necessity, an endeavor that will require guidance from highly detailed experiments (often unavailable for granular materials) or simulations. Previous major advancements in granular constitutive relation development have been heavily aided by the stiff-particle DEM due to its ability to provide high fidelity full-field information; multiple constitutive relations have been deduced based primarily on DEM data, such as the $\mu(I)$ model and its nonlocal extensions [3, 4, 5]. Given the intimate guiding role that DEM plays in the development of granular constitutive relations, and the difficulty of experimentally creating tuneable powders, it is essential to first establish a reliable powder DEM model, otherwise we are simply guessing constitutive forms that might not be physically justified.

Development of a reliable powder DEM model requires creation of tractable and accurate contact

models. Currently, there exists no comprehensive contact model for cohesive powder particles suitable in large deformation, an issue that was identified as a critical knowledge gap during the Modeling of Powder Flow workshop. A very detailed justification of this claim is given in the introduction of our paper [1], which was recently accepted for publication and posted online. Further personal discussions with company representatives from both Merck and Vertex, emphasized the lack of reliable powder DEM models and the issue this presents in simulating the tableting process. For example, Vertex informed us that they cannot predict the correct ratio of axial (punch) stress to radial (die) stress within a compaction simulator using a DEM reconstruction, and that even predicting this without the presence of air would be a major advancement for them. Another observation gained from discussion with IFPRI members is that there is significantly more familiarity and comfort with the DEM method and its widely-used software packages (e.g. LIGGGHTS and LAMMPS) over continuum methods. Additionally, defect formation during tableting, the primary industrial problem of interest, is exceptionally well-suited to DEM given the number of particles involved is between 10,000-50,000. In fact, DEM provides a number of desirable advantages over continuum methods for this problem including: resolution of particle-by-particle behavior that leads to nucleation of defects, natural capability to model inhomogeneous porosity distribution, and flexibility to use fluid solvers such as the lattice Boltzmann method that resolve flow around each particle, stopping the need to approximate a homogenized drag law under extremely low porosity if a homogenized fluid solver were used instead.

Given the above discussion, we first focused our effort to the development of a mechanically-derived contact model for powders, with focus on adhesive elastic-perfectly plastic particles. This model is given directly in terms clear mechanical properties of the grains themselves, such as surface energy, yield stress, and stiffness. With the theory of the contact model developed and validated, we have turned to implementing the model into LIGGGHTS. Once completed, we will work with Sandia National Laboratories to implement the model into LAMMPS, which has built-in capabilities to couple with fluid solvers. This will make the powder DEM model openly available to all IFPRI members on two different familiar platforms and will enable simulation of industrial applications beyond tableting. With significant effort placed into the development of a mechanically-derived contact model it can also then be trusted to assist in the future development of a continuum constitutive relation. We foresee this workflow providing the greatest long-term utility to all members of IFPRI, well beyond that of the original proposal objectives.

In the first year of the project, we developed a significant portion of the theoretical basis for a mechanically-derived contact model able to capture the response of interacting adhesive elastic-perfectly plastic spherical particles under a variety of simple symmetric loading's. When describing the model we will speak in present tense since all the general elements developed in year one persist in the current version. The model is built upon the method of dimensionality reduction (MDR) which allows the problem of a 3D axisymmetric contact to be mapped to a corresponding problem of a 1D rigid indenter penetrating a bed of independent Hookean springs. Because the MDR is valid for purely elastic contact, we account for plasticity by continuously varying the 1D indenter profile subject to a constraint on the contact pressure. In this sense, we view the problem as a sequence of elastic contacts defined by an evolving (or continually blunting) reference configuration. With the correct reference configuration defined at all instants, unloading falls out naturally, and simply requires lifting the 1D indenter out of the springs and tracking the force. By accounting for the incompressible nature of this plastic deformation through allowing the free surface to grow, the contact model is able to capture multi-neighbor dependent effects such as increased force and formation of new contacts. JKR type adhesion is recovered seamlessly within the MDR contact model by simply allowing the springs to 'stick' to the 1D indenter's surface. Contact under high confinement, the bulk elastic regime, is treated with a superimposed force that captures the rapid stiffening in the force displacement curve. Because of the mechanics-focused formulation of the contact model, only a few physical inputs describing the interacting particles are needed: particle radius, Young's modulus, Poisson ratio, yield stress, and effective surface energy.

Despite the range of features developed in the first year there were a variety of issues with the contact model that needed to be addressed:

- A JKR-type adhesion was included in the model, however it was only validated within the elastic small deformation regime. Additional simulations and work were required to determine the validity of the adhesive model after plastic deformation had occurred. This then becomes a highly nontrivial problem because the profile shape is no longer spherical, but rather blunted and can be continuously changing.
- The initial scheme used to grow the free surface to respect the incompressiblity of plastic flow turned out to be incorrect and required a complete overhaul.
- Only simple symmetric loadings of a single particle and rigid walls were considered. Adaptation of the contact model to many-particle interactions was necessary.
- The contact model was coded within Matlab, to allow greater accessibility and performance, implementation within an established software such as LIGGGHTS or LAMMPS was desired.
- A reliable way to couple a high fidelity fluid solver to the DEM implementation of the contact model needed to be formulated.

A highlight from year two is that the contact model has been published in *Journal of Mechanics and Physics of Solids*, as a two part series. Part I [1], focuses on a multi-neighbor dependent contact model for adhesive elastic-plastic particles built upon the method of reduction that is valid up until high confinement known as the bulk elastic regime. In Part II [2], the contact model is completed by proposing a superimposable treatment for the bulk elastic regime which is characterized by a rapid stiffening in the force-displacement curve as interstitial pore spaces vanish.

In the following sections we will discuss our efforts to address the bulleted action items from above. Some of the relevant information is contained within the sections of the papers of [1] or [2]. To reference this information I will use the following convention: "[1], Section 2" means look in Section 2 of the paper [1].

### 2.1.1 Adhesion after plastic deformation

The additional effort to understand adhesion after plastic deformation has occurred culminating in the addition of [1], Section 8.3.2 and [1], Appendix F.

### 2.1.2 Summary of findings

Adhesive contact behavior is directly affected by the relaxed profiles of the contacting bodies. After plastic deformation has occurred (i.e. the fully-plastic regime) this introduces complexity because the relaxed profile is non-spherical and continuously changing as more plastic deformation accumulates, a point that is apparent in Fig. F.17 of [1], Appendix F.1 which shows a number of relaxed profiles after loading into the fully-plastic regime. This effect is responsible for the fact that pressing particles together with more force results in higher adhesion between them. Interestingly, we found that after sufficient loading into the fully-plastic regime (something that would be common during powder compaction) the profiles dramatically changed in nature from being purely convex throughout to concave near the point of first contact. Despite the property of changing curvature significantly altering the adhesive behavior, we found that the adhesive MDR contact model – which assumes a purely convex profile throughout – is still able to reliably predict the strength of the adhered contact for a wide range of conditions. To validate the adhesive MDR contact

model we compared the critical pull-off force for both FEM profiles (the ground truth) and MDR contact model as shown in Fig. 13(c) and (d) of [1]. Generally good agreement between the two was observed, however specific criteria needed to be satisfied: one related to the size of the adhered contact and one for small scale yielding conditions. Detailed development of the two criteria is in [1], Section 8.3.2 and [1], Appendix F.2.

Compared to the reviewed contact models in [1], Section 1 ours is the only one to have a JKR type adhesion valid well into the fully-plastic regime — a state that is common during powder compaction and responsible for the easily observed fact that particles stick more the harder they are pressed together. This effort in validating the adhesion across this entire regime allows confidence in the fact that physical results are being predicted.

#### 2.1.3 Scheme for free surface growth

In our contact model we respect the incompressible nature of plastic deformation by defining a state variable known as the apparent radius; it grows to ensure that volume change due to plastic deformation is not lost. Geometrically, it is the radius of the sphere that most closely matches the deformed particle's free surface, that is, the part of the surface not in contact. Defining the apparent radius helps account for multi-neighbor-dependent effects such as formation of new contacts caused primarily by free surface movement (i.e. not relative displacement of particles) and increased force at existing ones. A visualization of the initial and apparent radius for a particle is shown in Fig. 6 of [1].

In year one of the project, we proposed a method for calculating this apparent radius, however we realized that it was incorrect and have entirely updated it. The apparent radius update scheme now has a differential form allowing the apparent radius to be explicitly updated provided a number of basic kinematic quantities are known. One notable improvement that was made during the reconstruction, is that it now calculates the elastic volume change. This means the contact model has an accurate prediction of the current particle volume at all instants along with the forces, plastic displacement, contact areas, and average pressures at each contact.

## 3 What we are doing now and in the near future

### 3.1 Implementing many-particle interactions

In the development of the contact model, only simple symmetric loading conditions of a single elastic-plastic particle with rigid walls was considered. Rigid walls were used because of their simpler geometric properties and equivalence to two identical contacting particles. The loadings investigated included the same three as in [6]: uniaxial compression, die compaction, and triaxial compaction as shown in Fig. 1 of [2]. Although they provided a tractable, yet diverse, set of contacts upon which to build the contact model they are not representative of the myriad of loading conditions seen in powder compaction. Most problematic, is that in the powder compaction setting, all particles may be of slightly different radii, making usage of a rigid flat less accurate. Thus, we need to develop a way to extend the MDR contact model, built only with consideration of rigid flat contacts, to the case of contact between particles of varying radii.

After consideration, our current approach is to continue with the idea of rigid flats as shown in Fig. 1. Instead of a complicated partitioning of the displacement between particle $i$ and $j$, we simply imagine placing a rigid flat at the midpoint of the displacement. Once the rigid flats are in place for each of the contacts on particle $i$ we can then reduce it back to the previous picture of an isolated particle being squished between rigid flats. To calculate the force between particles $i$ and $j$, the force on either side is calculated then it is averaged. In this way, the particles can begin with different radii and grow to different radii with causing issues.

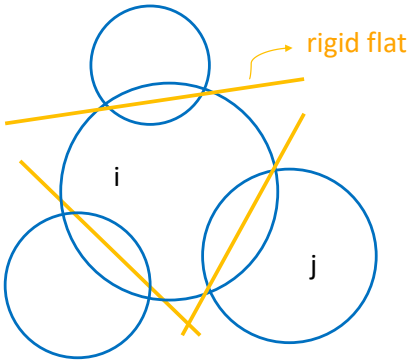


Figure 1: Many-particle, multi-powder-species contact implementation. Example: Four particles contacting each other, with two labeled as $i$ and $j$. Imaginary rigid flats are placed at the midpoint of the overlap.

Refinement to this approach might be needed, such as a more rigorous rigid flat placements that enforce identical areas, but for now this seems to be producing reasonable results.

### 3.2 LIGGGHTS and LAMMPS implementation

Implementation of the MDR contact model into LIGGGHTS and LAMMPS will enable simulation of industrially relevant processes such as tableting. Implementation into LIGGGHTS is mostly complete. A number of simple test cases checking the consistency between the LIGGGHTS implementation and the original Matlab version have been conducted. This process required us to ensure that particle contact with primitive walls, imported stl meshes, and other particles were all correct; each of which is handled separately in LIGGGHTS. During these small test cases we discovered that large deformation DEM has a unique issue where overlap between two particles or a wall and particle can be detected despite another particle physically separating them. This remains an unresolved issue that we will solve moving forward.

In moving to more realistic many-particle simulations, we introduced damping and a tangential contact model alongside the normal MDR contact model. Our current many-particle benchmark test is a tableting simulation, intended to mimic the compaction simulator shown in Fig. 2. Our experimental data is for Avicel PH102 and was provided by Vertex. The data contains position measurements of the upper and lower punch; the force on the upper and lower punch; and the radial stress measured by the upper, middle, and lower pressure sensors. From this, and the geometric measurements of the instruments, a number of plots can be created as shown in Fig. 4: (a) axial strain versus time, (b) axial stress ($\sigma_{zz}$) versus axial strain, (c) radial stress ($\sigma_{rr}$) versus axial strain, and (d) $\sigma_{zz}/\sigma_{rr}$ versus axial strain.

A corresponding LIGGGHTS simulation replicating the compaction simulator was created as shown in Fig. 3(a) and (b). The geometric and material properties of all particles was homogeneous. The intial radius of the particles was 500 microns leading to a total of 250 particles. The Young's modulus, yield stress, and Poisson's ratio were set to 0.2 [GPa], 0.5 [MPa], and 0.3, respectively. The Young's modulus and Poisson's ratio were inferred from reported literature values [7, 8], whereas the yield stress (due to lack of reported values) was fitted to a reasonable value. Adhesion was turned off.

During the simulation, which took approximately five minutes to run in serial, the axial strain, axial stress, and radial stress were monitored, allowing them to be overlaid on the compaction simulator data as shown in Fig. 4(b)-(d). The results showcase the nice predictive capabilities of the contact model. For both the axial and radial stress there is reasonable agreement throughout all of forward loading. Unloading is not yet implemented. We highlight the fact that after an axial strain of 0.3, both the radial and axial stress correctly stiffen, a trend caused by the superimposed bulk elastic response being triggered due to

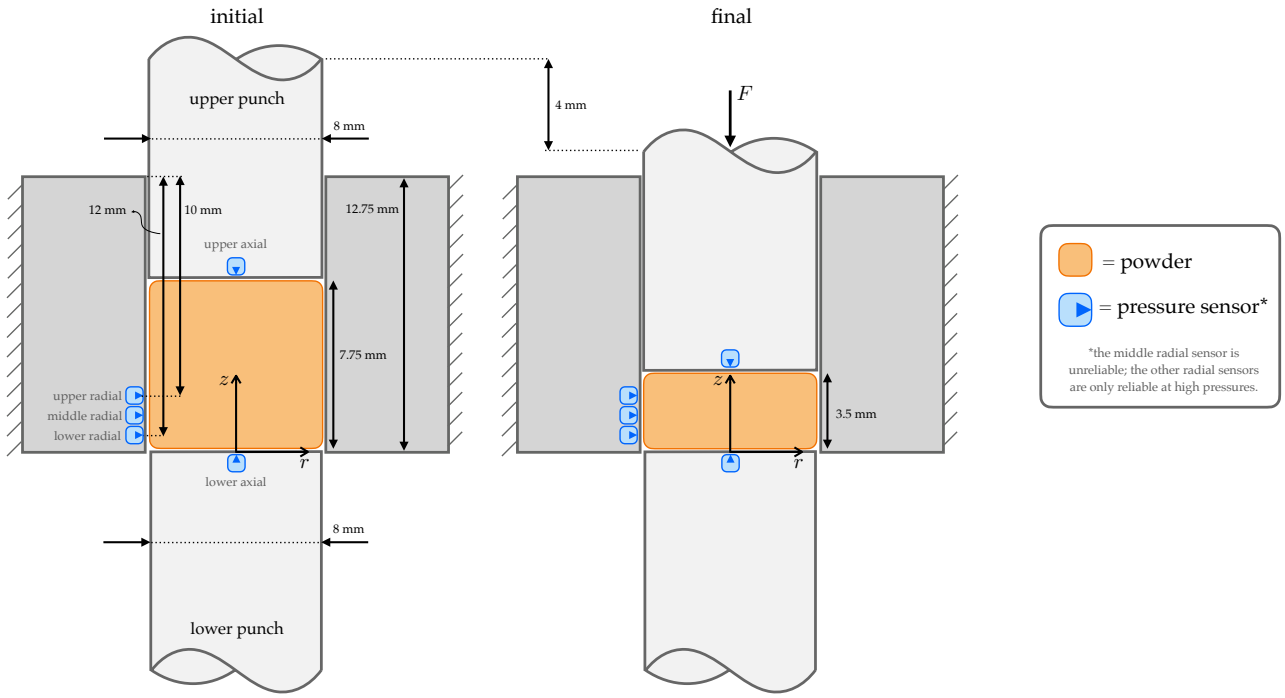


Figure 2: Compaction simulator setup.

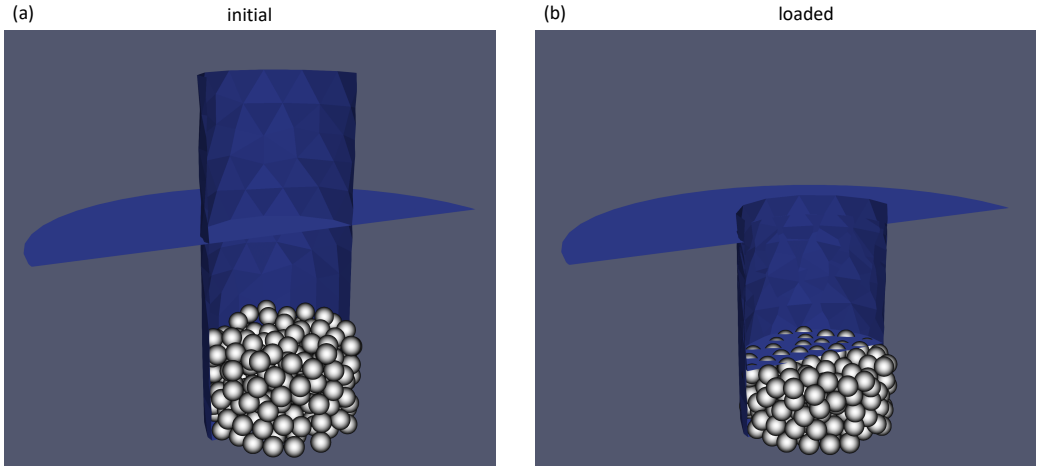


Figure 3: LIGGGHTS tableting simulation. (a) initial configuration and (b) loaded configuration.

high confinement.

At current, there are still some issues including selection of the time step[1] and the aforementioned issue of particles contacting despite being separated by another particle. Both of these are currently being worked on and may continue into the renewal period. Once corrected and coupled with the adhesion we anticipate even better agreement. We would like to once again emphasize that the current results were achieved with essentially no fitting and simply by taking reported literature values for grain properties — a major advantage over the other highly empirical existing contact models.

[1]Because we have a continually varying stiffness at the contact it is nontrivial to define the time step.

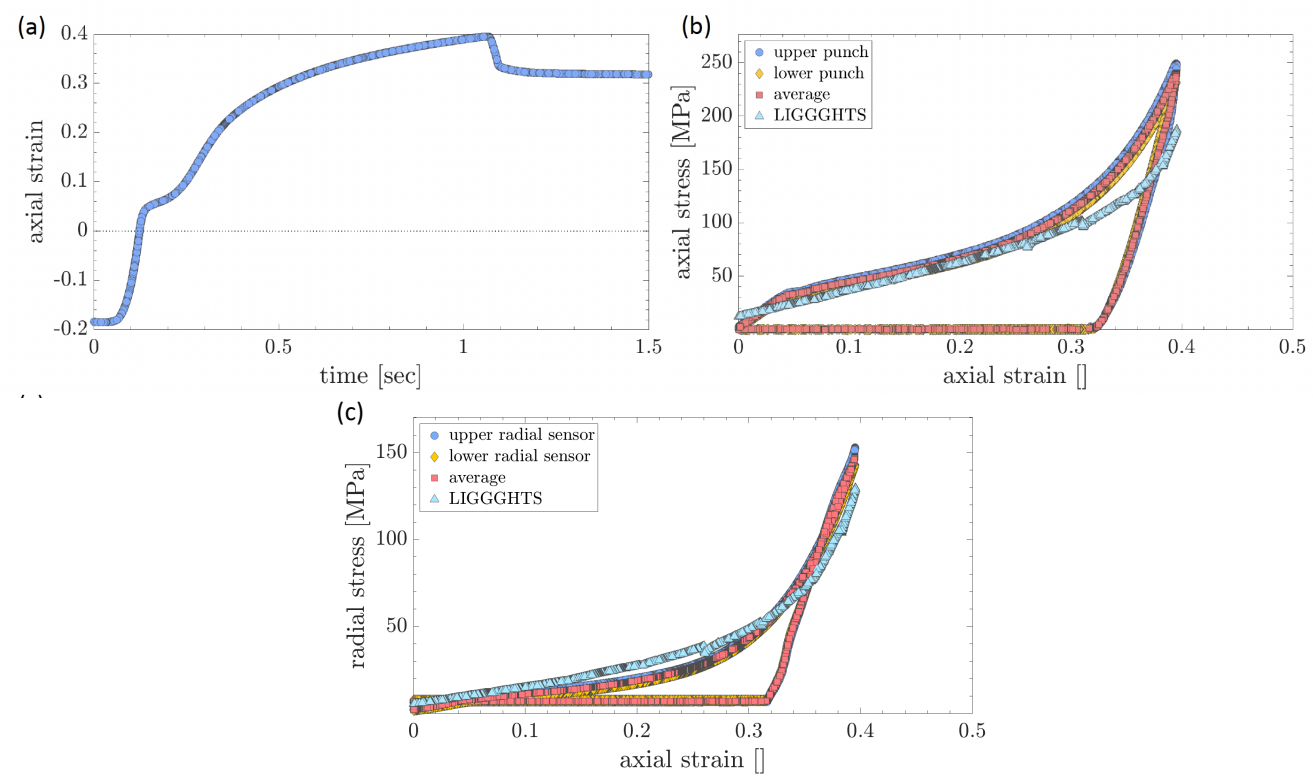


Figure 4: (a) Axial strain program for test. (b)-(c) Experimental powder compaction data (from Vertex; Avicel PH102 powder) compared against the LIGGGHTS simulation of our DEM model. DEM parameters taken from particle properties of Avicel PH102.

Once the LIGGGHTS implementation has been completed we will then work on a LAMMPS implementation. The LAMMPS implementation of the contact model will provide a number of advantages over the LIGGGHTS version. To start, we are in direct contact with the researchers at Sandia National Laboratories who maintain and develop the code. This should allow us to implement the code in a more computationally efficient way, whereas the LIGGGHTS version is a bit more 'hacked'. LAMMPS has historically been and will continue to be fully open-source, whereas LIGGGHTS has a paywall for some features. LAMMPS also has more consistent updates and improvements, in addition to superior documentation. In speaking with members of IFPRI, the majority of them have experience with either LIGGGHTS or LAMMPS, meaning there should be a low barrier to entry for running a LAMMPS simulation with the updated contact model. Finally, LAMMPS has a built in fluid solver readily available for coupling with any DEM simulation, a topic that will be discussed next.

### 3.3 Coupled fluid-solid simulations

As noted in Section 2.1, one of the primary industrial problems of interest to IFPRI members is defect formation during pharmaceutical tableting. One hypothesis among the members is that the trapped interstitial air plays a key role in initiating defects. Coupled numerical methods involving a fluid and solid are therefore essential, with a DEM–high fidelity fluid solver coupling as the most attractive option. Initially, we thought that the LIGGGHTS-OpenFOAM (DEM-CFD) coupling might provide the necessary toolbox to investigate the problem, however we have come to realize that a compressible gas phase does not come with the open-source release and would require significant coding to implement. Through our

discussion with the folks at Sandia, we were informed that the LAMMPS software is equipped with a fluid solver that can be coupled to a DEM simulation. The specific fluid solver is known as multi-particle collision dynamics (MPCD), a simulation technique for complex fluids which fully incorporates thermal fluctuations and hydrodynamic interactions [9]. Importantly, like the lattice Boltzmann method, it is able to resolve the fluid flow around the grains, halting the need to develop a grain-scale-accurate homogenized drag law under low porosity conditions, a task that is foreseeable to be difficult.

To demonstrate the current capabilities of the DEM-MPCD method in LAMMPS a simplified tableting simulation within a box was conducted as shown in Fig. 5. There are 600,000 MPCD particles representing air (small blue dots) and 500 powder particles (red spheres). The powder particles first settle in the air, then the entire system is compressed with a non-visible moving wall. The simulation runtime was less than 1 hour on a single processor, but the implementation is fully MPI parallel so much larger systems could be simulated if needed.

Although a Hookean contact model with friction was used, the simulation could be run with any contact model implemented in LAMMPS. Therefore, once our powder contact model is coded within LAMMPS and validated, the coupled DEM-MPCD can be used immediately allowing the problem of defect formation during tableting to be investigated with small-scale precision. Thus one of the main goals of the renewal period will be to implement our powder model into the LAMMPS DEM-MPCD framework and perform a vast parameter study to discern if and when trapped air is causing tableting failure. By comparing these results to tests with no air, we can isolate the role other suggested tablet failure causes, such as "popping" from residual stress after plasticity.

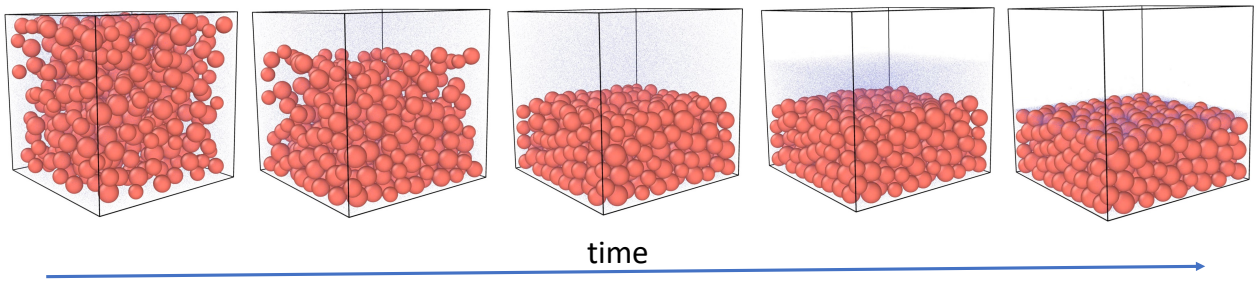


Figure 5: Coupled discrete element method (solid, red spheres) and multi-particle collision dynamics (fluid, blue dots) compaction simulation run in LIGGGHTS.

# 4 What we will do next

## 4.1 Fully Continuum Mixture theory

### 4.1.1 Background and existing expertise in the group

In mixture modeling, both fluid and solid phases are taken as two seperate continua living simultaneously on the same computational domain of interest, which is much larger than the individual particles but is still small compared to the size of the process equipment (Anderson & Jackson [10]). PI Kamrin's group has developed a general, thermodynamically consistent fluid-sediment mixture model and corresponding constitutive theory [11]. A complete numerical toolbox for implementing the system was also developed therein and was successfully able to model a range of fluid-particle interaction scenarios. We plan to use this basic framework for our problem. It is to be noted that the previous work did not consider cohesive granular media, the modeling of very large plastic volume changes, or using air as the fluid phase. Hence, suitable

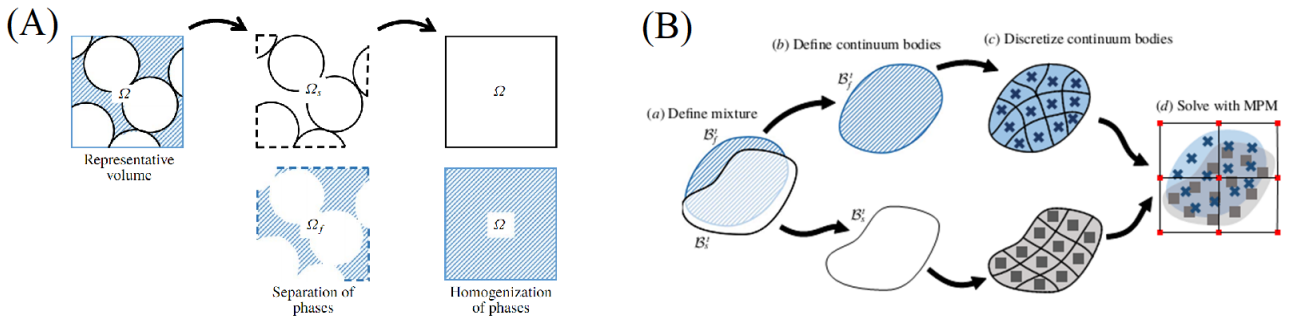


Figure 6: Two-phase continuum mixture simulation. (A) Pictorial description of the representative volume, the decomposition of the domain into fluid and solid volumes, and the homogenization of the two phases. (B) Solving mixture problems using the material point method (MPM). (a) Define the mixture and initial configuration including densities, porosities, stresses. (b) Define the solid and fluid phase continuum bodies. (c) Break the continuum bodies into piecewise-defined blocks of material represented by discrete material points. (d) Solve the equations of motion for the mixture on a background grid according to the material point method algorithm

modifications will be required in the constitutive theory as well as in the implementation to use it for our problem. Once completed, this tool **will be the first of its kind** in the powder compaction industry, i.e. a fully coupled mixture theory implementation, capable of modeling deformation of the elastic-plastic powder including cohesion and damage responses, all while in the presence of and interacting with a compressible air flow within. This could offer a significant computation time speed-up over DEM-MPCD.

A brief overview of this approach is described next and a detailed discussion about the possible ways of modifying the existing constitutive model framework for our needs follows this overview. As stated earlier, in this approach the solid and fluid phases coexist in the complete domain (see Figure 6(A)). Even though both phases live in the complete domain, the effective densities, $\overline{\rho}_f$ and $\overline{\rho}_s$, and phase velocities, $\overline{v}_f$ and $\overline{v}_s$ of the fluid and solid phases are evolved in such a way that the conservation of mass and momentum in the continuum correspond to conservation of mass and momentum in each phase of the real mixture. The overall stress in the continuum can be represented as:

$$\boldsymbol{\sigma} = \tilde{\boldsymbol{\sigma}}_s + \boldsymbol{\tau}_f - p_f \mathbf{1} \tag{4.1}$$

where, $\tilde{\boldsymbol{\sigma}}_s$ represents the stress from particle contacts (typically called the *effective stress*), $\boldsymbol{\tau}_f$ represents the homogenized stress tensor in the fluid phase, and $p_f$ represents the pore pressure. Individual solid and fluid phase momentum balances are implemented as follows:

$$\bar{\rho}_f \frac{D^f \boldsymbol{v}_f}{Dt} = \nabla \cdot \boldsymbol{\tau}_f - n \nabla p_f + \bar{\rho}_f \boldsymbol{g} + \boldsymbol{f_d} \tag{4.2}$$

$$\bar{\rho}_s \frac{D^s \boldsymbol{v}_s}{Dt} = \nabla \cdot \tilde{\boldsymbol{\sigma}}_s - (1-n) \nabla p_f + \bar{\rho}_s \boldsymbol{g} - \boldsymbol{f_d} \tag{4.3}$$

where, $n$ represents the porosity ($= 1 - \phi_s$) and $\boldsymbol{f}_d$ represents the interphase drag force. In our current implementation, $\boldsymbol{f}_d$ representing Darcy-like drag is given as:

$$\boldsymbol{f}_d = \frac{18\phi(1-\phi)\eta_0}{d^2} \hat{F}(\phi, Re)(\boldsymbol{v}_s - \boldsymbol{v}_f)\,. \tag{4.4}$$

The form of $\hat{F}(\phi, Re)$ can be chosen at will, ranging from a simple Stokes form to the Carman-Kozeny relation and its offshoots like Van der hof (2005) relations. (See Baumgarten and Kamrin [11] for more details).

The theory also takes care of the fact that in the absence of a solid phase, the behavior of the mixture is identical to that of a pure fluid and in absence of fluid it reduces to the behavior of the solid phase in vacuum. The numerical implementation in [11] was carried out using a material point method (MPM) framework capable of solving all of the governing equations simultaneously on the computational domain. In MPM, each phase of is discretized as a set of material point tracers that carry their full continuum state. These tracers, representing a chunk of their corresponding material about their position, are then 'cast' onto a background simulation grid where all the governing equations are solved. The nodal representation of the mixture velocities and acceleration are projected back to material tracers to update the mixture state (as stored on the two sets of material points). A brief pictorial representation of the explicit time-marching algorithm used by them for time integration of the problem is shown in Figure 6(B). More details about alternative implementations of TFM can be obtained from Zhou et al[12].

An example of our method in action can be found in Figure 7, where it was used to simulate the collapse of two piles of grains submerged in an aquarium. The model used was calibrated to match glass beads in water using from data in [13], which studied a different geometry. The match-up between experiments and simulations shown in Fig 7(c) showcases the ability of this approach to attain quantitative predictive accuracy.

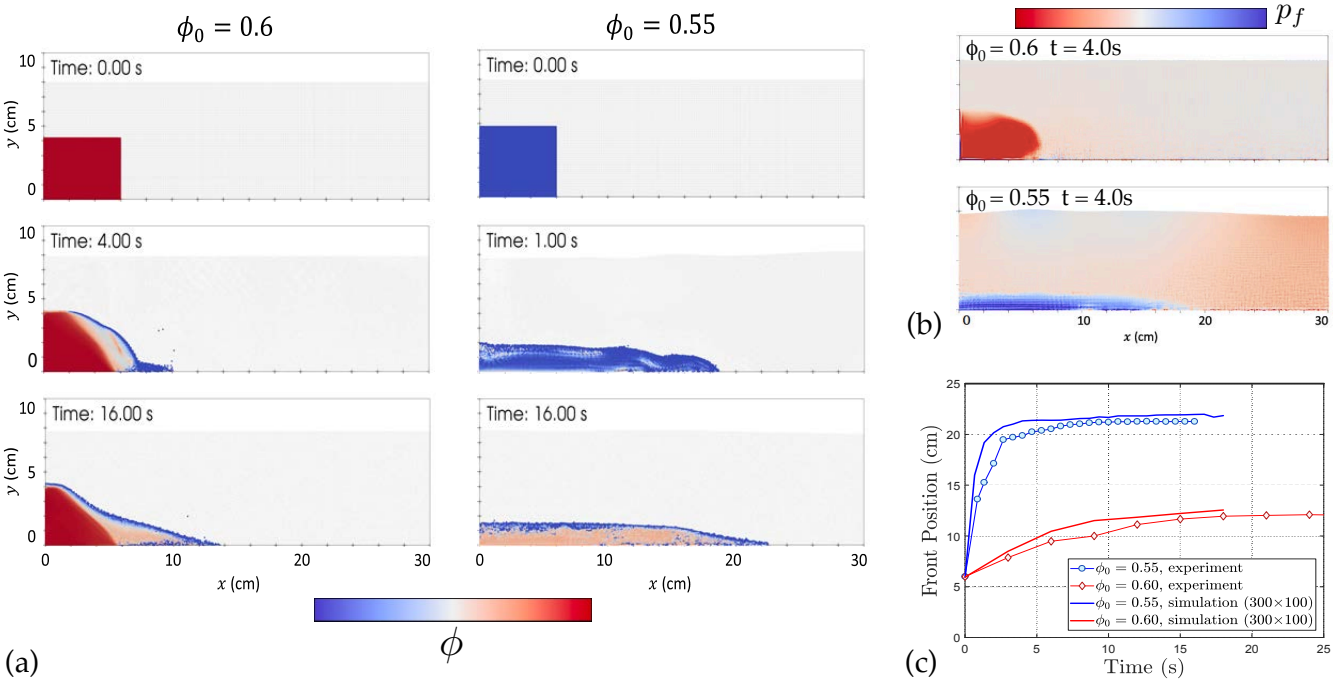


Figure 7: Grain-fluid mixture continuum simulations by PI Kamrin's group using two-phase MPM [11]. (a) The collapse of two columns of simple grains submerged in a water tank. The two tests differ only by the initial packing fraction ($\phi_0$). Note the much slower run-out of the packing that starts more tightly packed. (b) Fluid phase pressure field for each case at $t$ =4s. During collapse, initially tight packing shows pore tension relative to surroundings, unlike initially loose case. (c) Comparison of results to runout experiments [14] of submerged glass beads.

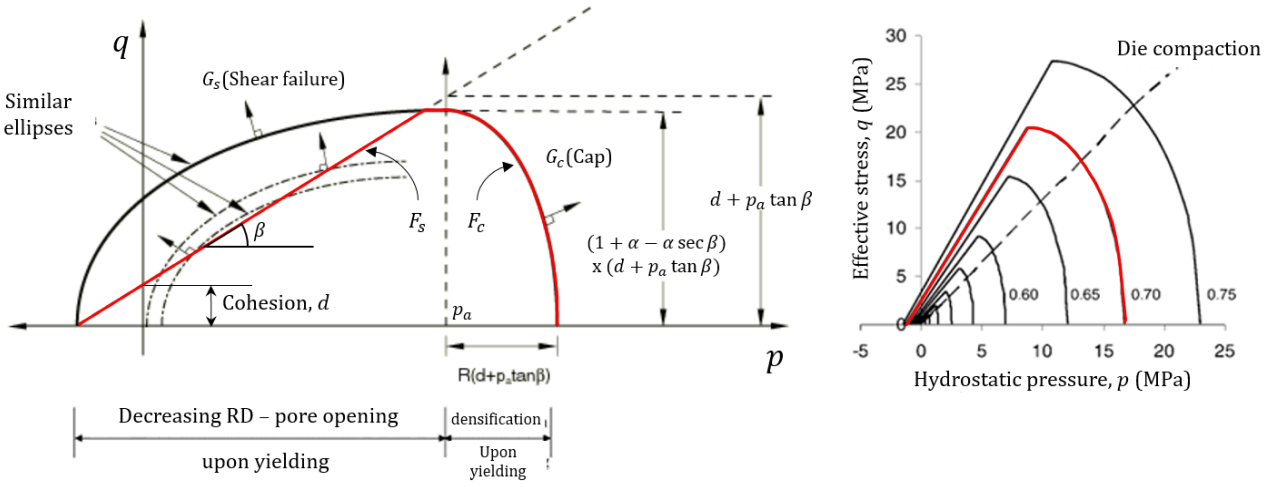


Figure 8: DPC model: (a) Model parameters for a given relative density (RD), (b) Family of DPC yielding curves for a range of relative density values (using compaction data for MCC). The dotted line represents the typical evolution of stress in a close die experiment.

### 4.1.2 Planned approach

The primary difference between this implementation and the one required for our powder compaction study will be in the constitutive equations. While the implementation used in Baumgarten and Kamrin [11] models the solid phase as non-cohesive hard-grain granular media, the problem of powder compaction requires the solid phase to undergo large plastic deformations and large changes in effective grain density (from 30-40% to 78% packing fraction) under high applied pressures. As the compaction pressure is released, the pressurized, trapped air can act on the now-dense solid structure, and might cause internal failure of the material. We will introduce an evolving damage variable to measure the extent of internal breakage, which we believe is key to predicting the tablet's final integrity. Hence, to capture both compaction and damage, we propose to modify the Ducker-Prager Cap (DPC) model by coupling it to this damage state variable, which serves to scale down the solid cohesive strength. We base the solid continuum model on DPC due to its previous success at representing tablet excipients. A brief description of the modified DCP with a general outline of the proposed model is provided below. **Importantly, we intend to use our DEM capability as a *digital twin* to help us determine parameters for this model and how they relate to grain properties.** These will include appropriate homogenization tests involving uniform deformations applied to an air-grain sample in a periodic box.

### 4.1.3 Model for powder compaction and its limitations

The DPC model is a modified form of the conventional DP model used for modeling the non-linear stress response of granular media undergoing plastic shear and volume changes [15]. The behavior of the compacting powder is assumed to be isotropic and the elastic response is taken as linear. The model's yield criterion, which relates effective shear stress $q$ and pressure $p$ for a given value of relative density (RD) of powder, is shown in figure 8 (A). The model parameters $d$ (cohesion), $\beta$ (internal friction angle), $R$ (cap excentricity parameter) and $p_b$ (hydrostatic yield stress) must be fit as a function of RD. The exact values of the same for microcrystalline cellulose (MCC) can be obtained from [15].

#### 4.1.4 Plastic response of modified DPC model

A pictorial representation of the modified DPC model [16] is shown in Figure 8. At low hydrostatic pressures (the region of $p < p_a$ in Figure 8), the model is a shear failure model, represented by yield surface $F_s$ as indicated. Similar to many other granular models, this model also assumes a linear dependence of the shear strength on the confining pressure in this regime. The yield surface at high hydro-static pressures ($p > p_a$) is represented by a 'cap' denoted $F_c$. It can also be seen from Figure 8 that the material has far less shear strength when in tension ($p < 0$). The flow potential ($G$) whose normal direction indicates the 'direction' of plastic flow upon yielding is also shown in the figure. The flow potentials in shear failure and in the cap region are represented as $G_s$ and $G_c$ respectively. The flow is associative in cap region ($G_c = F_c$) but is non-associative in shear region ($G_s \neq F_s$). Description of the complete material behavior at all relative densities (RD) requires a family of yield surfaces and flow potentials to be obtained experimentally. The exact calibrated values for MCC can be obtained from Cunningham [15].

#### 4.1.5 Challenges and planned study

For the use of this model in our work, we must integrate a damage variable. The current form of the model does not have the capability of accounting for the development of cracks/damage in the formed material, which is a major focus of this project. To model the development of non-healing cracks due to the flow of air between them, a non-reversible damage model is planned to be incorporated. We plan to incorporate damage on the lines of the famed Johnson–Holmquist-2 (JH-2) model, often used in ceramic damage modeling [17, 18, 19]. Here the material softens with increasing damage (captured using a damage parameter, $D$) and loses all its cohesive strength at $D = 1$. The rate of damage increase is expected to be dependent on the current cohesive strength of the material, the rate of volumetric expansion, the current relative density, and the current stress. Some additional assumptions are also planned to be taken: (i) The damage is unidirectional i.e. it can only increase and cannot decrease, (ii) damaged material can not be repaired, and (iii) damage occurs only during plastic expansion i.e. $\Delta\epsilon_v^p > 0$. A representative relation for the damage and its effect on the state of the material can be given as follows.

Define the damage, $D$, to be 0 for fresh, undamaged material. The damage level then increases by the following rule

$$\Delta D = \frac{\langle \Delta\epsilon_v^p \rangle}{\epsilon_f} \qquad \epsilon_f = \hat{f}(d, \boldsymbol{\sigma}, RD, D)$$

where $\langle \cdot \rangle$ represents the Macauley bracket. The damage in the material is expected to affect the load-carrying capacity of material and hence, the cohesion $d$, which appears in the DPC model. Thus, $d = \hat{d}(RD, D)$. The constitutive relation of the solid phase will primarily affect the form of $\tilde{\boldsymbol{\sigma}}_s$ in equation 1-3. In addition to the solid phase constitutive equations, we will also modify the coupling equations for solid-fluid interaction to compensate for the change. The coupling equations use an explicit formulation for the Darcy drag. Thus, any required modification due to porosity change in the solid phase of the mixture can be applied directly.

It is to be noted that the real-life capping and laminating surfaces for tablets are never smooth. The irregularities are expected to be a result of stochasticity in the properties of the real granular media at continuum level and a result of finite grain size at a granular level (if irregularities characteristic lengths are on order of the grain diameter). Even a small variation of properties like powder strength and density variation in the granular volume is expected to play a crucial role in defining the final capping surface. We expect adding variation can provide potential damage nucleation sites in the bulk and thus bias the capping surface. The conventional powder characterizing experiments like ones done for modified DPC above, consider only the mean response/properties of the powder blends. We believe consideration of such variation in the compaction process will be the key to capturing cracking. We plan to add a random, zero-mean, low-variance density variation in the initial definition of the material domain. This distribution can either

be decided based on known data or on information from detailed DEM simulations. We expect that the zero mean density inhomogeneity will help to capture cracking in two ways. First, the inhomogeneity will result in variation of the material strength in the compact. These zones will act as stress concentration and nucleation sites for plastic volume changes during the die compaction. Secondly, the inhomogeneity of powder packing fraction will also introduce an inhomogeneous air pressure distribution in the compact. These zones grow the damage variable when the die is lifted, reducing the local cohesive strength. As the stress continues to concentrate around these damage fronts, the process propagates outward, emulating cracking.

#### 4.1.6 Support from IFPRI members

Aside from aformentioned data sharing from Vertex, data from Cunningham [15] is planned to be reinterpreted for the development of this model but no information about the damage of the material in this data is known. In the absence of such data, experimental data from IFPRI members will of great help for us. Similarly, expert insight from IFPRI members about the stochasticity of properties in the powder blends will be of great value to us. Once the formulation is complete, we will also need exact powder compaction geometry dimensions, and tool and powder properties along with experimental results to check the accuracy and robustness of our model. In this regard, we also hope that DEM modeling of cohesive/non-cohesive grains (which takes particle size effects into account and thus local variations) will be of great use (for improving mixture model formulation). More details about the same are provided in the later sections.

## 5 Detailed outline of the proposed work for the renewal period

The first year will be primarily dedicated towards development of the DEM-MPCD tool and using it to map out correlations between grain properties and the consequent strength and damage modes in tableting simulations. Next will come the development of continuum damage models for a cohesive powder bulk. This model for the powder phase will then be combined with the gas phase to obtain an implementation of an air-powder mixture model. DEM-MPCD data will assist in calibrating the mixture theory. The third year is planned to be the final phase of the project, which will mainly focus on cross-verification of our implementations with actual experimental data in real-life tablet compaction geometries, and iterating the methodology accordingly. A few more enhancements are also planned for the final year and will be implemented based on the availability of time. A detailed outline of our three-year project plan is given in Figure 9.

## References


[1] W. Zunker and K. Kamrin, "A mechanically-derived contact model for adhesive elastic-perfectly plastic particles, part i: Utilizing the method of dimensionality reduction," *Journal of the Mechanics and Physics of Solids*, p. 105492, 2023.

[2] W. Zunker and K. Kamrin, "A mechanically-derived contact model for adhesive elastic-perfectly plastic particles, part ii: Contact under high compaction—modeling a bulk elastic response," *Journal of the Mechanics and Physics of Solids*, p. 105493, 2023.

[3] F. Da Cruz, S. Emam, M. Prochnow, J.-N. Roux, and F. Chevoir, "Rheophysics of dense granular materials: Discrete simulation of plane shear flows," *Physical Review E*, vol. 72, no. 2, p. 021309, 2005.

| SN | Activity | Year 1 | | Year 2 | | Year 3 | |
|---|---|---|---|---|---|---|---|
| | | H1 | H2 | H1 | H2 | H1 | H2 |
| 1 | Integrate powder DEM model into LAMMPS and LIGGGHTS packages. | | | | | | |
| 2 | Integrate air into powder DEM model using MPCD capability in LAMMPS. | | | | | | |
| 3 | Conduct parameter study of tableting with and without air using DEM-MPCD simulations. Compare to experiments from IFPRI collaborators. Compose failure mode chart. | | | | | | |
| 4 | Develop damage-modified DPC continuum model and qualitative verification using single-phase MPM implementation. | | | | | | |
| 5 | Calibrate damage-modified DPC model without air using powder DEM modeling as a digital twin. Extract mapping from grain property to continuum property. | | | | | | |
| 6 | Perform theory modification of existing mixture model for the incorporation of updated damage-modified DPC model. | | | | | | |
| 7 | Implement and calibrate modified mixture theory in two-phase MPM setup. | | | | | | |
| 8 | Perform qualitative and quantitative comparison between mixture theory and DEM-MPCD results in tableting tests. Modify former based on the results from latter. | | | | | | |
| 9 | Final calibration of two-phase continuum model. Conduct final analysis of two-phase continuum model ability to capture tableting failure in air-powder mixtures. | | | | | | |

Figure 9: Research plan for three years. Items 1 and 2 are already underway.


[4] K. Kamrin and G. Koval, "Nonlocal constitutive relation for steady granular flow," *Physical review letters*, vol. 108, no. 17, p. 178301, 2012.

[5] S. Chialvo, J. Sun, and S. Sundaresan, "Bridging the rheology of granular flows in three regimes," *Physical review E*, vol. 85, no. 2, p. 021305, 2012.

[6] B. Harthong, J.-F. Jérier, P. Dorémus, D. Imbault, and F.-V. Donzé, "Modeling of high-density compaction of granular materials by the discrete element method," *International Journal of Solids and Structures*, vol. 46, no. 18-19, pp. 3357–3364, 2009.

[7] R. Roberts and R. Rowe, "The young's modulus of pharmaceutical materials," *International journal of pharmaceutics*, vol. 37, no. 1-2, pp. 15–18, 1987.

[8] A.-S. Persson and G. Frenning, "An experimental evaluation of discrete element simulations of confined powder compression using an extended truncated-sphere model," *Powder Technology*, vol. 284, pp. 257–264, 2015.

[9] A. Malevanets and R. Kapral, "Mesoscopic model for solvent dynamics," *The Journal of chemical physics*, vol. 110, no. 17, pp. 8605–8613, 1999.

[10] T. B. Anderson and R. Jackson, "Fluid mechanical description of fluidized beds. equations of motion," *Industrial & Engineering Chemistry Fundamentals*, vol. 6, no. 4, pp. 527–539, 1967.

[11] A. S. Baumgarten and K. Kamrin, "A general constitutive model for dense, fine-particle suspensions validated in many geometries," *Proceedings of the National Academy of Sciences*, vol. 116, no. 42, pp. 20828–20836, 2019.

[12] Z. Zhou, S. Kuang, K. Chu, and A. Yu, "Discrete particle simulation of particle–fluid flow: model formulations and their applicability," *Journal of Fluid Mechanics*, vol. 661, pp. 482–510, 2010.

[13] M. Pailha and O. Pouliquen, "A two-phase flow description of the initiation of underwater granular avalanches," *Journal of Fluid Mechanics*, vol. 633, pp. 115–135, 2009.

[14] L. Rondon, O. Pouliquen, and P. Aussillous, "Granular collapse in a fluid: role of the initial volume fraction," *Physics of Fluids*, vol. 23, no. 7, p. 073301, 2011.

[15] J. Cunningham, I. Sinka, and A. Zavaliangos, "Analysis of tablet compaction. i. characterization of mechanical behavior of powder and powder/tooling friction," *Journal of pharmaceutical sciences*, vol. 93, no. 8, pp. 2022–2039, 2004.

[16] L. Han, J. Elliott, A. Bentham, A. Mills, G. Amidon, and B. Hancock, "A modified drucker-prager cap model for die compaction simulation of pharmaceutical powders," *International Journal of Solids and Structures*, vol. 45, no. 10, pp. 3088–3106, 2008.

[17] D. S. Cronin, K. Bui, C. Kaufmann, G. McIntosh, T. Berstad, and D. Cronin, "Implementation and validation of the johnson-holmquist ceramic material model in ls-dyna," in *Proc. 4th Eur. LS-DYNA Users Conf*, vol. 1, pp. 47–60, 2003.

[18] S. Gharehdash, L. M. Shen, Y. X. Gan, and E. Flores-Johnson, "Numerical investigation on fracturing of rock under blast using coupled finite element method and smoothed particle hydrodynamics," in *Applied Mechanics and Materials*, vol. 846, pp. 102–107, Trans Tech Publ, 2016.

[19] J. Wang, Y. Yin, and C. Luo, "Johnson–holmquist-ii (jh-2) constitutive model for rock materials: Parameter determination and application in tunnel smooth blasting," *Applied Sciences*, vol. 8, no. 9, p. 1675, 2018.