

Predictive Framework for Multicomponent Powder Compact Strength

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The goal of this project is to develop a *predictive micromechanical framework* for the strength of *multicomponent powder compacts*. This model will be based on original theoretical and numerical approaches and validated by experiments. The project is of a fundamental and innovative nature in terms of both methodology and scientific goals but will have the potential to provide a framework for application to real materials. In the following, we describe the context and general objectives, the main ingredients of the proposed approach, the methods and materials, and successive steps in its implementation.

Context

Room temperature (“cold”) compaction of multicomponent powders is a commonly used operation to produce monolithic compacts with a prescribed shape and a multiplicity of physical or performance characteristics, such as effective diffusivity (ceramic membranes), pore size (catalysts), disintegration rate (pharmaceutical or detergent tablets), and burn rate (propellants). In all cases, the compact must have sufficient *tensile strength* to survive subsequent processing, packaging, transport, and use. The desired functionality of these compacts is commonly achieved by the addition of specific “functional components”, and this often leads to highly complex, multicomponent formulations. Most models proposed in the literature are not based on the real characteristics of the microstructure. In the absence of a suitable model for compaction of multicomponent mixtures, industrial practitioners are forced to resort to empiricism, making mixtures and measuring compact strength, which can lead to expensive or incomplete exploration of the formulation space and increased risk in development.

The objective of this project is *to develop a predictive framework for strength development in multicomponent powder mixtures undergoing cold compaction* at compaction stresses large enough to cause significant *plastic deformation* of particles. The model will be informed by and verified with experiments. More specifically, we are interested in semiquantitative prediction of strength as a function of composition, ingredient particle size distribution, and compaction pressure for a blend of *at least 3 components*, which requires limited mechanical characterization of the individual components. This model will be based on *micromechanics of compaction*, involving the texture of the compact (composition, contact network, contact area distribution between components, pore size distribution, force transmission...), its evolution with applied pressure, and mechanisms of crack nucleation and propagation under loading. The project will, therefore, seek to identify, in the first place, the *lowest-order texture variables* that control the strength development during compaction and the onset of collapse under external loading. It will require simulations of deformable particles, extensive parametric study, analytical modeling, and comparison with experiments.

Simulation of deformable particles

For a micromechanical approach, a *particle dynamics method* is needed to simulate the compaction process and generate extensive datasets at the particle scale for the characterization of the

microstructure. Since we are interested in particles undergoing significant plastic deformation, particle dynamics simulations based only on the Discrete Element Method (DEM) are not appropriate (Radjai and Dubois, 2012). We will, therefore, rely on the Material Point Method (MPM) for modeling elasto-plastic deformable particles. We have developed a new technique that combines implicit or explicit formulations of MPM for the simulation of deformable individual particles with the Contact Dynamics (CD) method for the treatment of cohesive-frictional contacts between particles (*MPM-CD approach*) (Nezamabadi et al., 2015, 2017, 2019, 2022).

Although the Finite Element Method (FEM) has been successfully applied to a broad range of solid mechanics problems, body-fixed FEM meshes can be difficult and time-consuming to generate for complex objects, and mesh distortion associated with large deformations compromises solution accuracy, ultimately requiring re-meshing. In contrast, the MPM avoids mesh distortion and never maintains a body-fixed mesh. To deal with the frictional contacts between deformable particles, a multi-mesh mapping algorithm based on the CD method is used to implement contact laws such as the Coulomb friction law. This algorithm allows us to compute the contact forces (normal, frictional, and cohesive forces) and the contact area, and to define different frictional and short- or long-range cohesive interactions (van der Waals, capillary, or electrostatic) between material points in the contact zone. Linear and nonlinear elasto-plastic laws can be used for particle behavior.

A major advantage of the MPM-CD approach is that the interactions are independent of particle shape. For this reason, the use of arbitrary particle shapes does not involve additional computational cost. It is noteworthy that the computational cost of MPM-CD simulations is high due to the large number of degrees of freedom associated with each particle. For this reason, in our investigations of the microstructure during compaction and compression tests up to failure, we will be able to simulate only packings of relatively small numbers of particles (several thousands). To avoid wall effects and further increase the number of particles, we will use three-periodic boundary conditions with a parallelized version of our new code (Nezamabadi et al., 2019). This will significantly increase computational efficiency and the overall homogeneity of the simulated samples, leading therefore to statistically meaningful data for the characterization of the microstructure.

Generation of samples

Independently of experimental validation presented below, we will conduct a detailed parametric study using MPM-CD simulations with various combinations of particle characteristics. For each combination, particles are initially placed inside the simulation cell. Subsequently, an *isotropic quasi-static incremental compaction* is applied. The particles initially jam in a dense configuration due to rearrangements and then begin to deform beyond the jammed state with decreasing porosity. At regular intervals of compaction, the sample is replicated and allowed to relax by removing the confining pressure. The sample will expand elastically and stabilize in a new equilibrium state with permanent plastic deformations. This procedure makes it possible to generate unconfined samples of decreasing porosity for various combinations of particle characteristics. An example of a deformed sample is shown in Fig. 1.

Each sample will be subjected to *simple compression* until failure. Under fully periodic boundary conditions, the tensile strength can be evaluated by subjecting the samples to simple extension (applied in one direction while keeping unconfined conditions in the other two directions). Otherwise, simple compression also allows for the evaluation of tensile strength from the largest stress reached before failure. Two different regimes must be distinguished:

- The regime where the cohesive strength due to surface energy is lower than the elastic and plastic moduli of the particles. In this scenario, failure occurs due to crack propagation across

the particles. Consequently, this process results in particle fragmentation, and the overall tensile strength of the compact is governed by particle strength.

- The regime where the cohesive strength is greater than the elastic and plastic moduli of the particles. In this scenario, the applied extension primarily results in deformation localized in the contact areas, and failure occurs through crack nucleation and propagation across the contacts, while the particles themselves do not break.

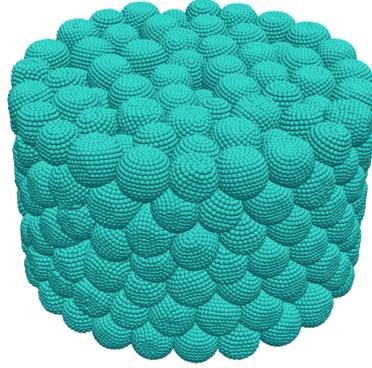


Fig. 1: Compact of 400 deformable particles using the MPM-CD approach. The material points are represented by small spherical particles.

We are interested in the second regime, which requires low cohesive stress between particles that is sufficiently high to ensure the mechanical integrity of an unconfined compact. It is also noteworthy that, due to cohesive interfaces, the samples may involve self-stresses induced by compaction and subsequent removal of confinement. The residual elastic energy of the samples will be evaluated in each case, and its influence on the tensile strength will be quantified.

The order of magnitude of the mean cohesive strength is $\sigma_c = \gamma S$, where γ is *surface energy* and S is the *specific contact area* (contact area per unit volume). In a multicomponent compact, the mean cohesive stress can be evaluated from the surface energies γ_{ij} and specific contact areas S_{ij} between the components i and j . Hence,

$$\sigma_c = \sum_{(ij)} \gamma_{ij} S_{ij} P_{ij} \quad (1)$$

where P_{ij} is the number fraction of contacts between the components i and j . In the absence of segregation, i.e. for a homogeneous distribution of the components inside the compact, we have

$$P_{ij} = 2P_i P_j \quad \text{if } i \neq j \quad (2)$$

$$P_{ij} = P_i^2 \quad \text{if } i = j$$

where P_i is the proportion of particles of type i .

The tensile strength σ_t of the compact is expected to scale with the cohesive strength:

$$\sigma_t = \xi \sigma_c \quad (3)$$

where the pre-factor ξ is a *structural factor* depending on the microstructure. It is not, however, obvious whether ξ is fully independent of porosity or whether it depends on the second-order variables pertaining to the statistical distribution of contact areas or pore volumes inside the

compact. Furthermore, the evolution of tensile strength with the proportions P_i of the particle types may involve discontinuous changes due to the percolation of one component as its proportion increases. The tensile strength may also be controlled by the *weakest links*, i.e. the interface of lowest energy, rather than the average cohesive strength σ_c . This may depend on the proportion of the component that has lowest surface energy with other components.

It is also important to distinguish between quasistatic and dynamic conditions. In quasi-static compression, crack nucleation is controlled by stress concentration due to pores whereas in dynamic conditions (such as impact) the outcome depends on the ratio of impact kinetic energy to the total cohesive energy $\sigma_c V$, where V is the volume of the compact. In this case, at low impact energy, the compact is only damaged without fracture as a result of the failure of the weakest contacts. The compact breaks at a critical ratio for which there is enough energy absorbed by the compact to dissociate a large enough number of contacts. When the compact does not break, the tensile strength decreases due to damage, so that the same compact will break for a lower impact energy compared to the initial compact. Although both quasistatic and dynamic conditions can be investigated, in both simulations and experiments we will focus in the first place on quasistatic loading for the evaluation of tensile strength (Orozco et al., 2019, 2020).

Parametric study

The parametric space, even for three different particle types, is quite large. Some examples of interesting combinations are as follows:

- We consider three different particle sizes mixed at various proportions. All other properties (surface energy, elastic and plastic moduli) of the three components will remain the same. The reference state is the case of equal mass fractions of the three particle sizes. Two independent size ratios will be systematically varied for equal mass fractions to quantify the trends. Then, the size ratios will be set to 1.5 and 3, while the mass fractions will be varied around the reference state. The idea here is to have at least one component with much smaller particle sizes.
- Different values of surface energy: The interface energies γ_{ij} between the components can have different values. We will consider particles of the same size but different combinations of the values of γ_{ij} . One particular case is when only one component sticks to the two other components or when its surface energy is much higher than that between the two other components. Hence, this component behaves either as the only binding phase or as the strongest binder. We will therefore investigate this particular limit for increasing mass fraction of the binding particles. We will also investigate the effect of the plastic modulus of this phase. For a low plastic modulus (as compared to the two other components) the particles of this phase will spread and partially fill the contact zones and pores between particles. The opposite limit case of interest is when one component sticks less to the two other components. We will investigate the effect of the proportion of this component on the overall tensile strength.
- Other combinations will be considered based on the results and analyses performed in the above cases. We will also simulate compaction and simple compression with calibrated values of the experimental materials, as discussed below.

In all simulations, we will analyze the microstructure and its evolution during compaction and during simple extension or compression tests as well as the tensile stress with focus on the following quantities:

- Distribution of contact areas
- Connectivity (coordination number)

- Distribution of pore volumes
- Stress transmission (distribution of stress components across different phases and interphases)
- Failure modes (initiation and propagation)
- Tensile stress at failure
- Variability

The variability of tensile strength can be evaluated by performing independent compaction tests with random initial positions of the particles. Given the high computational costs of such statistical studies, they be carried out for a limited number of cases. The distribution is expected to follow a *Weibull distribution* with parameters that will be determined in each case (Blanc et al., 2021). The variability study will also allow us to obtain an estimation of the intervals of confidence.

Predictive model

The datasets generated by MPM-CD simulations and analyzed along the lines described previously will lead to correlations between material parameters, component proportions, and applied pressure, on the one hand, and variables pertaining to the microstructure, tensile strength, and porosity, on the other hand. To develop an analytical model from these correlations, we proceed in the first place by looking for a *general scaling* or partial scaling to reduce the parametric space. Then, we focus on the values of the parameter ξ defined by equation (3) as a function of microstructural variables. If ξ is weakly dependent on all or some parameters, we may develop model in which statistical mixing of material parameters (including cohesive stresses or surface energies between components) determines the tensile strength. Otherwise, the *deviations from this mean field prediction will guide the development of a refined model* of the behavior. Another important aspect concerns comparisons with some of the existing classical models although the scope of the present project goes well beyond what can be found in the literature.

Experimental validation

Experimental compaction experiments will be performed on powder pharmaceutical excipients (*alpha-lactose monohydrate, microcrystalline cellulose, sodium chloride and native corn starch*). These materials are chosen for their known plastic or close-to plastic deformation mechanism under compaction strain. Before compaction, a multi-scale physico-chemical characterization of the particulate materials on the different characterization platforms available at ICGM (Institut Charles Gehardt Montpellier) (powder rheology, particle size and morphology as well as molecular and microstructural properties) will be performed. The objective of this characterization is to fully identify and classify the critical properties that can influence the particulate material behaviors during the compaction. The materials will then be compacted with an instrumented press coupled with an exhaustive characterization of the produced compacts. The press is a unique R&D single punch tablet press (Stylone Evolution, Medelpharm, France) (Boudina et al., 2022), that benefits from very versatile volume reduction and a displacement of both punches at high speed which allows to reproduce production cycles of all the major industrial presses and analysis of these production cycles. This methodology has proven useful in understanding particle bed behavior for pharmaceutical tablets.

Experimental compaction will consist in compacting the standard reference materials of homogeneous particulate composition followed by mixtures in different proportions. This first step will allow to calibrate the press and identify what compression profiles can and should be applied to the different materials. We will use these reference materials to produce bi-ter-quarter-nary

mixtures. The instrumented press will load the material according to different size and shapes of compacts, compression speeds, pressures and times. Thus, by varying materials, compaction kinetics and force, the obtained compression cycle stress-strain profile can be used to identify the deformation behavior of the particulate bed during the different compaction phases and how this is affected according to the different modes of material loadings. We will evaluate the compact properties such as tensile strength, solid fraction/porosity and elastic recovery.

We will use the values of parameters to conduct MPM-CD simulations and compare the results with the experimental data. The comparison will mainly focus on the tensile strength predicted by the model extracted from the simulation data and general scaling arguments. We may find poor agreement, in which case we will have to examine possible model form errors and input parameters. In other words, the model extracted from the simulation data may not be suitable for the range of parameters for which it has been designed. To reduce such a risk, the calibration and validation step must be performed in parallel with simulations, and the range of values of simulation parameters (elastic and plastic moduli, surface energies, particle sizes, component mass proportions) must be chosen to include the material parameters of the experimental powders. The experimental data, numerical results and model predictions will also be compared with classical empirical methods, such as Heckel analysis and Kawakita equation.

Implementation

The project will be implemented within a 3-year program.

During the first year, we will focus on the experiments with three components and calibration of MPM-CD simulations using experimental measurements. The goal in this step is to obtain all the necessary elements for validation although the validation itself may require further experiments and simulations. In particular, we seek to compare not only the macroscopic quantities such as tensile strength but also the evolution of porosity during compaction. We will also need to develop the appropriate tools for the analysis of the microstructure of simulated samples.

The second year will be devoted to broad parametric study using exclusively numerical simulations and analyzing the microstructure and correlations between variables pertaining to the microstructure and compact-scale variables such as stresses and porosity. We will use these correlations to examine the relation between tensile strength and the mean cohesive strength defined from statistical mixture of different component characteristics. Comparisons with existing models and ideas in the literature will also be made.

The conclusions of the first two years will guide further simulations and experiments to be carried out during the third year of the project for both refining the model and validating the simulations. The model and simulations data will also be compared with independent data provided by IFPRI partners. We believe that this 3-year program will lead to a solid foundation for the elaboration of a predictive model. The extensions and scopes of the project for further developments are broad and will be defined at the end of the third year.

Collaborations

This project will benefit from local collaboration with **Tahmer Sharkawi**, a professor at Institut Charles Gerhardt Montpellier (ICGM) more specifically for experimental validation. A PhD student will be hired to work on this research. Further collaborations will be established with interested members of IFPRI. In particular, due to the broad scope of this project, the numerical simulations and values of system parameters can be adjusted and adapted to existing datasets provided by members. However, we have locally access to the appropriate materials for validation through our

collaboration with ICGM. We are also currently involved in a project with CEA (French Atomic Energy Commission) on the tensile strength of agglomerates of mixtures of UO₂ and PuO₂ powders. The insights provided by DEM simulations used in the project with CEA are relevant to the current project.

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