IFPRI Project Abstract

Controlling Rheology via Boundary Conditions in Dense Granular Flows

Karen E. Daniels
Dept. of Physics, North Carolina State University
Project Start Date: 1 August 2018
Abstract Date: 11 May 2022

Project Objective:

In prior work, we have previously shown that nonlocal rheologies successfully model granular flows across different packing densities, particle sizes and shapes, and shear rates. This success is provided by the measurement and specification of a set of three material properties for a particular set of particles. However, the results of the model depend on *a priori* knowledge of the flow profile, limiting its predictive power. Here we aim to (Q4) understand how changing the roughness and/or compliance of a boundary changes the resulting flow. This will ultimately allow us to determine a set of general principles (Q5) which allow us to correctly set the boundary conditions in nonlinear rheological models, and test their generality in other geometries (Q6).

Approach:

As done during our work for the first funding period, we perform our experiments in an existing quasi-2D annular shear cell with a rotating inner wall and stationary outer wall. Both walls are laser-cut and are interchangeable to modify roughness, as shown at right. In each experiment, we aim to measure both the velocity (shear) profile and the bulk rheology (shear and normal stresses) in order to characterize the dependence of the boundary conditions on both the particle-shape and boundary-characteristics.

In the past year, we have added a second apparatus that allows us to test the same wall, but in faster flows. This hopper is the same one used by Bob Behringer (IFPRI report FRR 56-06).
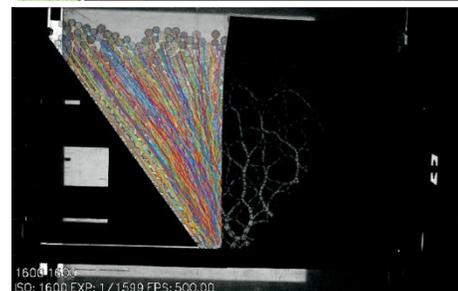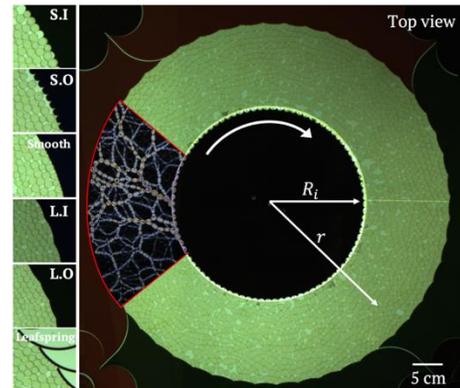
Recent Results:

During 2021-2022, graduate student Farnaz Fazelpour has continued conducting experiments and data analysis using a variety of particle and boundary conditions, improving our ability to make quantitative measurements of the stress field, and its spatial and temporal fluctuations. A key finding is that decreased wall slip, which is highly dependent on wall roughness, is associated with an excess of force fluctuations (MAE, in the plot at right).
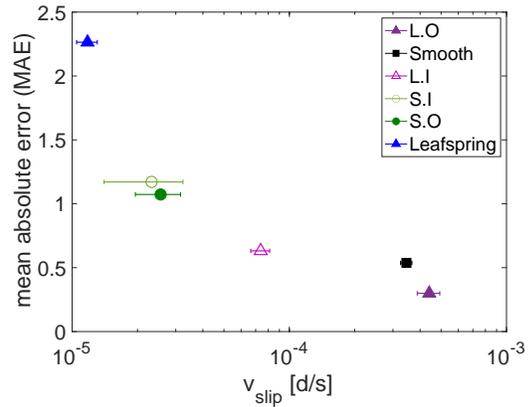
An undergraduate student, Emily Cecchini, has refurbished a hopper (from Bob Behringer's lab) and made flow measurements using the same boundary shapes as done for the annulus. These runs are at much higher inertial number, and data is collected using a high-speed camera. While the hopper datasets do not provide full stress fields, we are nonetheless observing similar trends as shown above.

Next Steps:

In the remaining few months, we will submit a paper on boundary-condition results from the annular experiment; this work is nearly complete. This summer, we will complete our analysis of the hopper dataset, and present those results in our Final Report. In addition, through a 3-month IFPRI-funded collaboration with Prabhu Nott, we will undertake a comparison between our rheological data and his nonlocal model that accounts for dilatancy under shear.

More broadly, this project has shown that nonlocal models can successfully, quantitatively model granular flows. In order to achieve *predictive* capabilities, a better understanding of how to quantitatively set model parameters and boundary conditions (slip) from either first-principles or simple-experiments still remains.

IFPRI Project Abstract

A multiscale study of powder reconstitution phenomena

Management : Claire GAIANI, Jennifer BURGAIN & Jeremy PETIT
PhD : Tristan FOURNAISE – **Post Doc :** Arnaud PAUL
University of Lorraine, LIBio (Laboratoire d'Ingénierie des Biomolécules), France
Project Start Date: January 1, 2019
Abstract Date: May 03, 2022

Project Objective:

Organic and biologically-derived materials in powder form are involved in the manufacturing of many products available in the industry (e.g., cosmetics, food, pharmaceutics). Their reconstitution is of upmost importance for the industry considering that most powdered ingredients are dissolved or infused before use. Therefore, deeper mechanistic understanding and global approaches are needed regarding the great variety of powders industrially available. Also, fundamental understanding enabling improvement of the reconstitution of these powders is still lacking with a focus on the particle surface.

Approach:

This three-year project (plus one due to COVID) approach aims at:
(1) Developing a reconstituability index to draw a predictive criterion for the classification of unknown industrial powders according to their reconstitution behavior from the knowledge of their physical and chemical characteristics.
(2) Bringing new knowledge in the reconstituability of industrial powders with a focus on the particle surface.

Recent Results (year#3):

A semi-empirical model able to describe the kinetics of reconstitution by particle size monitoring was developed and made it possible to better understand the impact of the physicochemical properties of the powders on the reconstitution steps. Finally, a predictive model of the reconstitution time of the powders under the reference conditions used in the experiments adapted during the first year was developed using the most influential physicochemical properties.

Next Steps (year#4):

Due to COVID lockdown, a 4th year was financed by IFPRI. The PhD student (Tristan Fournaise) will defend his thesis in July 2022 and a post doc was recruited in order to focus the year 4 on the following parts:
(1) Development of a method to discriminate powder components depending of their location within particle
(2) Deciphering the segregation of components in particles in order to correlate reconstitution properties and surface composition.
(3) Identify molecules segregation and links with reconstitution in various powders studied in year 1

IFPRI Project Abstract

Spray characterization at industrially relevant conditions

Nasser Ashgriz
University of Toronto, Department of Mechanical and Industrial Engineering
Project Start Date: October 2018
Abstract Date: 20 May, 2022

Project Objective:

Spray drying is a process used in the production of powdered products such as pharmaceuticals. Pilot scale experiments are used to determine the optimal operating conditions of the sprays and correlations are used to extrapolate to the production scale. Current correlations lack validation across a wide range of nozzle sizes and for fluids relevant to the pharmaceutical industry. This study aims to develop these correlations for a wide range of nozzle sizes and fluid viscosities by focusing on understanding and mechanistically modelling the near-nozzle breakup behaviour.

Approach:

1. Develop benchmark data set for near-nozzle spray characteristics for lab and production nozzle scales with high viscosity and polymeric fluids (Phase 1)
2. Determine correlations and models for nozzle scaling and for ligament formation and breakup (Phase 2)

Recent Results:

Phase 1: A benchmark database of droplet size distributions and near nozzle images has been generated for pressure-swirl and two fluid nozzles of varying scales for water (low-viscosity), glycerin (high-viscosity), and CMC (polymeric) solutions. The database is presently being packaged for online distribution.

Phase 2: Modelling of the spray behaviour was focused on two nozzle designs:
Pressure-swirl: A perforation-based atomization model was proposed for atomization in pressure-swirl nozzle based on images at near nozzle region. The model predicted two kinds of ligament with different characteristic diameter. Droplet distributions downstream showed bimodal distributions, with each mode related to one type of ligament. A semi-empirical correlation was developed by fitting a mixture of log-normal distribution to the bimodal volume distribution. The major mode corresponding to large span-wise ligament takes up 95% of total volume and outweighs the minor mode and the parameters in the distribution were correlated with physical parameters. A series of experiments on fan spray nozzles had been done to further study the relationship between operating conditions and ligament sizes. Mass ratio between two types of ligaments, ligament sizes at the breakup positions were measured using image processing techniques. Ongoing work aims to understand the physics between these parameters and operating conditions.

Twin-fluid: The dynamics of air-assisted atomization were studied in the form of droplet breakup, resulting in a model that describes how the initial deformation is related to the mean size of the resulting ligaments (link), while a variety of mechanisms leads to the distribution of droplet sizes (link). The droplet breakup model was applied in the context of a twin-fluid nozzle, providing a good prediction of the droplet size distribution for a wide range of operating conditions and nozzle scales. High viscosity slows the deformation, leading to larger sizes overall, and slows the breakup, leading to long-lived ligaments. For polymeric fluids with high extensional viscosity, only the breakup is slowed, leading to a network of thin, stringy ligaments. Ongoing work aims to include such effects in the model.

Next Steps:

We have proposed to extend this project by further studying the atomization of complex fluids, such as solid suspensions and fluids with significant elongational viscosity, to further develop models and correlations for the droplet size distribution from such fluids.

IFPRI Project Abstract

Dynamic and structural investigation of capillary suspensions

Jens Allard and Erin Koos
KU Leuven
Project Start Date: 25 September 2018
Abstract Date: 13 May 2022

Project Objective:

Structure and function and inherently related. Using a model system for dense and capillary suspensions, we will investigate the microstructural changes during interesting rheological transitions, e.g. yield, using confocal microscopy. Capillary suspensions, particles suspended in a bulk fluid with a small amount of immiscible secondary fluid added to form a percolating network of capillary bridges between the particles are of particular interest as they show a wide range of rheological behavior.

Approach:

Changes to the microstructure of a suspension of fluorescently labelled silica particles are investigated using a confocal microscope. Using particle detection and semi-local measures, we can compare the structure of the samples with their bulk rheological response.

Recent Results:

This year, we finalized the work on particle roughness. By varying the roughness, we could switch between bridges and films (wetting between asperities) with increasing roughness. By increasing the secondary fluid content, we could match G' in the LVE, but there is still a dependence of the roughness on G'', the flow point, and on the structure (as captured using the coordination number and clustering coefficient). We repeated several experiments, particularly the medium amplitude oscillatory shear (MAOS) measurements, to ensure repeatability and accurate interpretation of the data. This work appears in *Colloids and Surfaces A* (DOI: 10.1016/j.colsurfa.2022.129224) and is freely available on arXiv (https://arxiv.org/abs/2203.07779).

The focus, however, this year has been on the confocal-rheometer setup. This setup is now operational and preliminary experiments have been conducted. These experiments show the solid body rotation present in capillary suspension networks where flocs move as single objects. This is more pronounced for rough particles; for the smooth particles, particle/bridge rotation and particle rearrangements happen before the G'' peak whereas rearrangements for the rough particles only occur after the G'' peak and usually occur in the form of rearrangements of small clusters rather than individual particles.

Next Steps:

The rheo-confocal setup will now give us more information on the dynamic changes to the network. The initial experiments conducted for the smooth and rough particles will be continued where the particle movement and structure will be quantified, focusing on mechanism of yielding. Some additional improvements should be made and we will then implement better detection of the fluid bridges to look at the bridge size distribution. We have also started experiments with nanoparticles in the bridges.

A Holistic Approach for the Model-based Control of Crystal Size, Shape and Purity in Integrated Continuous Crystallization – Wet Milling – Classification Systems

Rojan Parvaresh, Wei-Lee Wu, Zoltan K. Nagy
Purdue University, Davidson School of Chemical Engineering, West Lafayette

Specific aims: "*To develop and provide the first proof-of-concept simulation and experimental demonstration of a real-time implementation of a full population balance model based nonlinear predictive control approach for batch and continuous crystallization processes integrated with wet milling and classification/recycle system, in order to achieve desired crystal size and shape distribution. The effects of impurity/additive mixtures on the size, shape and crystal purity distribution will be incorporated in the model.*"

Crystallizer – Wet Mill – Classifier

Classification system design ($S = \frac{e^{EX}-1}{e^{EX}+e^{E}-2}$)

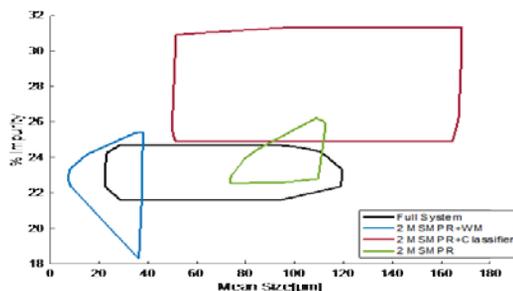$L_{crit} = 70\ \mu m$

Eff = 0.5
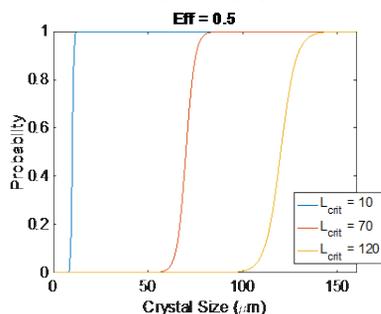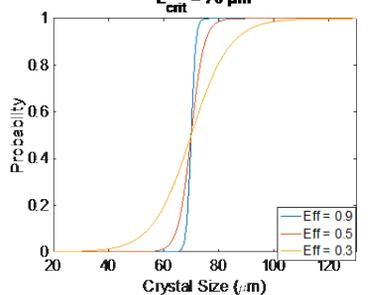
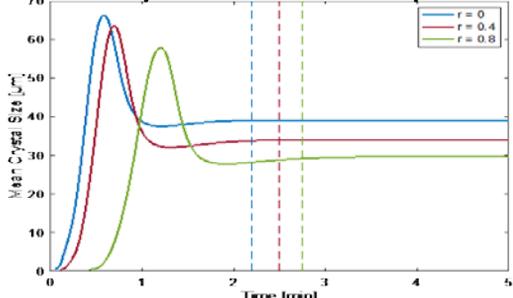### Impurity model development
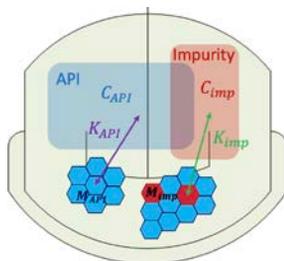
α : partition coefficient
K : relates solid phase (M) to liquid phase (C)

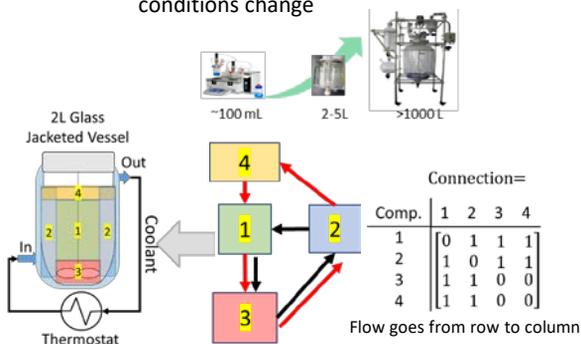### Attainable region for an integrated system

Dynamic model – Startup

Application – Scale-up

- Batch crystallization scale-up issues:
  - Crystallizer geometry, scale, and operating conditions change

~100 mL   2-5 L   >1000 L

2L Glass Jacketed Vessel

Connection=

Comp.	1	2	3	4
1	0	1	1	1
2	1	0	1	1
3	1	1	0	0
4	1	1	0	0

Flow goes from row to column

- Unseeded batch thermocycling temperature profile
  - Heat transfer delayed in compartment 1 (bulk phase) and compartment 3 (impeller)
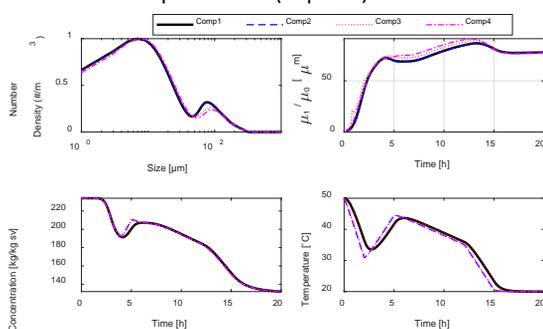
Future Plans

- Experimental validation
- Crystallizer network applications
  - Implement flow velocities from CFD simulations
  - Incorporating attrition at impeller
- 2D simulation of the integrated system
- Model based quality by control (QbC)
  - Developing a moving horizon estimation (MHE) based nonlinear model predictive control using the full 1D and 2D PBMs for size, shape and inferential purity control for both process systems.
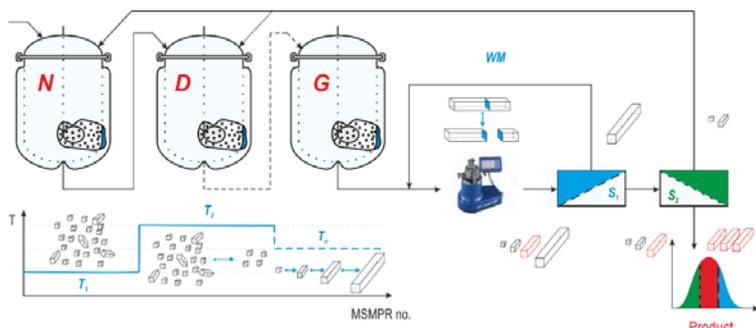
Absract: Discrete Element Method Round Robin

Kit Windows-Yule, Leonard Nicuşan and Jonathan Seville

May 2022

The overarching goal of the IFPRI Round Robin project is to establish a "best practice" for the calibration of discrete element method (DEM) simulations, which at present represents perhaps the most significant bottleneck in an otherwise powerful and easily-adoptable technique.

In previous work, we have quantitatively assessed the effectiveness of discrete element method (DEM) calibration methods utilised by 8 industrial DEM practitioners for a number of differing experimental geometries, particulate media, and combinations thereof. The accuracy of the methods was assessed by comparing the outputs of simulations performed following the procedures of the 8 participants with detailed experimental data produced using Positron Emission Particle Tracking (PEPT), a technique which allows the dynamics of particulate systems to be imaged, in three dimensions, with sub-millimetre spatial resolution and sub-millisecond temporal resolution.

Strikingly, of all the participants surveyed, no two institutions adopted the same practices, highlighting the need for a more standardised approach and best practice. Our results show that while most contemporary calibration methods are able to successfully capture the dynamics of simple, free-flowing, spherical particles under low-shear conditions, the vast majority of procedures tested were unable to correctly reproduce the behaviours of smaller, more cohesive particles, or higher-shear environments. For the latter case, though qualitative agreement and visual similarity between simulated and experimental systems could be observed, deeper and more quantitative analysis using PEPT revealed significant disparities.

A number of methodologies were able to successfully capture the dynamics of aspherical, highly-angular particles, but no advantage was observed in the implementation of complex and computationally-intensive geometric models over the simpler and more efficient rolling-friction method for the materials and systems explored.

Of the calibration methods examined, the most effective – indeed the only one to consistently reproduce the experimentally-measured dynamics of the cohesive systems tested – involved the combination of both static and dynamic powder characterisation tests, suggesting this to be the best practice for multi-parameter DEM calibration.

Based on the outcomes of 'Round 1' of the Round Robin, a number of questions were raised by both the team conducting the Round Robin, our industrial partners, and the wider IFPRI community. Our current, ongoing work aims to address these questions. The foci of the current project are as follows:

1. A deeper exploration of the ability of companies' texhniques to correctly calibrate the *elastic*, as opposed to frictional, properties of particles.

2. An investigation of the ability of DEM calibration methodologies to capture the *stresses* within systems, as opposed to only their dynamics.

3. Investigation of the ability of DEM models to quantitatively simulate experiments involving smaller ($d \sim 100\mu$m) particles than initially explored, as well as to successfully model highly elongated particles.

4. The development of DEM 'digital twins' of a wide variety of commonly-used powder characterisation tools, to be freely distributed to IFPRI members with an interest therein.

IFPRI Project Abstract

Adhesion of powders to metal surfaces during compaction

Csaba Sinka
University of Leicester, UK
Project Start Date: 1 October 2018. Renewal: AGM 2023
Abstract Date: 13 May 2022

Project Objective:

This project aims to develop a fundamental understanding and predictive relationships between a powder's physical and chemical properties and its propensity to adhere to metal surfaces of compaction tooling.

Approach:

Sticking hypotheses:

1. Temperature. Sticking can be understood as a coupled thermo-mechanical problem with two sources of heat: dissipative processes during compaction and friction between powder and tooling. The materials and surfaces evolve, e.g. phase transformations due to stress, strain rate, temperature. Currently the project is focused on temperature.
2. Humidity. Moisture uptake by hygroscopic materials leads to water acting as a binder at the interfaces, leading to sticking.
3. Particle breakage. Brittle materials or granules break during compaction, creating new surfaces. These new unlubricated surfaces give cohesion/strength to compact but also lead to sticking.

Recent Results:

The project has effectively started in August 2021 with PhD student Ahmad Ramahi and PhD student Vishal Shinde joined in February 2022. We aim to finish the project by the renewal date, AGM 2023. We hold monthly meetings with IFPRI members which are extremely informative.

WP1: Identification of test materials and characterisation procedures – complete.

WP2: Characterisation of single component materials and surfaces. PSD – Malvern 3000 (wet), AFM for surface energy and surface roughness. AFM Kelvin Probe (surface potential) developed as method to observe sticking.

WP4: Redesigned heated die and trilayer (metal disk-tablet-metal disk) system. Characterised Ibuprofen, Paracetamol and Mannitol.

Identified strain rate as a new potential parameter responsible for sticking.

Next Steps:

- Characterise: aspirin, maize starch, sorbitol, 2 lactose grades, MCC.
- Nano-indentation, DSC and surface composition (EDX – SEM) analysis.
- AFM with particle probe requires method development.
- Rotary friction system requires redesign.
- Establish predictive criteria for sticking propensity.

IFPRI Project Abstract

Model Assisted Design of Granular Products: Linking Process and Product Models for Wet Granulation

Rachel Smith
The University of Sheffield
Project Start Date: 1st October, 2019
Abstract Date: 25th May, 2022

Project Objective:

The objective of this project is to develop multi-dimensional process models of binder agglomeration that have as their output the appropriate particle properties (likely distributed) that are linked to associated product models to predict product performance. The project scope is constrained to high-shear and/or fluid bed granulation, however the choice of formulation(s) and modeling approach(es) are open.

Approach:

The aim of this research project is to develop a modeling framework for product performance driven process design which is deployable to industry, and demonstrate the potential of this strategy with two case studies: fluidised bed spray granulation and high shear granulation. This will involve the development and linking of population balance models for fluidised bed and high shear granulation with a mechanistically-based product model.

Recent Results:

The project started in October 2019. Since the previous annual meeting presentation, the following key progress has been made:

- The previously developed coupled single-particle model and population balance model for granule disintegration has been further developed and refined, to better incorporate essential physics, and to resolve issues with numerical stability of the model.
- Preliminary sensitivity analysis of the improved model has been performed, and the model is performing as expected in response to changes in granule size, porosity, and primary particle size and absorbance capacity.
- An experimental strategy for model validation has been developed in conjunction with the University of Strathclyde, to verify and validate the granule dispersion model.

Next Steps:

In the next year, focus will be placed on performing a global sensitivity analysis of the coupled single particle – PBM model, and on implementing the experimental strategy to verify and validate the models. The product model will then be linked with our existing high shear wet granulation model. Finally, we will commence the validation of the linked process-product model.

IFPRI Abstract- Annual meeting 2022
Simplified industrial formulations.
Design challenges

Jan Vermant and Lucio Isa,
Department of Materials, ETH Zurich

May 20, 2022

Goals

Our goals within the IFPRI project remain threefold

1. To explore how, moving away from model systems containing spherical colloids with near hard interactions, we can widen the range of rheological responses by changing the properties of the building blocks of the suspensions, so that even in simple formulations a wide range of behaviors can be "built in", i.e. obtaining formulation guidelines to do "more with less" or simplifying formulations from within.

2. To further develop a limited number of rheological and structural tools to interrogate the rheological response of the such dispersions, focusing on

   - Advanced rheological methods

   - High resolution confocal microscopy to probe structural development in situ during flow (4D imaging)

   - Local scale tribological measurements using AFM.

3. Apply these methods to simplified industrial dispersion by industrial partners and compare with the formulation guidelines obtained from (1).

Simplified industrial systems : bottom-up

Model systems with more complex topography : During the past year[1] we achieved one of the main results we were aiming for. A model thermoreversible gel system was designed, using a new synthesis approach for better controlling the grafting for octadecyl grafting to silica particles. This chemistry can be can be combined with different surface roughnesses to influence yielding behavior under shear conditions. For the same particle size, medium viscosity and surface chemistry, but different roughness, the elastic modulus and yield pont can now be varied independently. For the rough system, the elastic response regime (and hence the yield stress in rheological terms) can be increased by almost two orders of magnitudes, while the modulus remains the same. The rough systems are also less thixotropic and recover much faster. The

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[1] Work of graduate student Florence Mueller

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hypothesis is that interlocking of rough particles inhibits cluster densification, meaning the network structure can resist to shear. Work is ongoing to vary the roughness scale to get scaling relations, and micro-structural imaging and SAXS analysis of the microstructure.

> **Versatile particle toolbox**:The specific goal of having roughness and tunable attractions to control yield stresses and thixotropy by having control over the openness of the aggregate structures has been achieved. An original and practically useful result, which will be submitted for publication.

Advanced rheological measurements: The main advances made in this area over the last year were the final improvements on the setup, image processing routine validation and measurements on both shear thickening systems and thixotropic gels. The key result was that we have new results on tracking of individual particle motion[2], now looking at for rotation at least 3 points in space. Rough particles with only fluorescent asperities are used and have been succesfully studied. The role of microscopic contact mechanics is now made possible by comparing this with single particle AFM methods. For the gels work has focused on identifiying local pastic event in gels, for no not yet the rough ones[3].

> **Rheoconfocal imaging**: To elucidate the link between formulation, the microstructure and the rheology, rheo-confocal measurements are being performed. Last years' progress has focused on tracking particle rotation in stable systems and the local plastic events .

Simplified industrial systems: top down

The following samples have been identified as being relevant for and suggested by the members as systems relevant for a class of problems. They will be characterized by the rheological techniques mentioned above

1. Carbopol dispersions, relevant for thickening of consumer products (suggested by Unilever)

2. Latex suspensions, with $Ti0_2$ and $CaCO_3$ as model coating materials (suggested by Chemours)

3. milled paracetamol and a range of additives (Bentopharm (Clay) and Rhodopol 23 (Xantham gum))

4. Protein samples for understanding protein viscosity in bulk and understanding how processing conditions affects protein stability using interfacial tests (suggested by Merck)

Work has started on sample 1,3 and 4. Sample 2 has arrived. For the results on sample 1 a paper is being readied for publication

Concluding remarks

1. Concerning the bottom up increased complexity of model systems, the essential result has been achieved. Microscopic studies will now lead to a more precise predictive capacity.

2. Top down simplified industrial systems have been identified and are being characterized, a first successful example of the approach and new modeling insights have been achieved.

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[2]Work of graduate student Vincent Niggel
[3]Work of graduate student Pierre Lehericy

IFPRI Project Abstract

A Systems Engineering Approach to Dry-Milling with Grinding Aid Additives

Principle Investigator(s): Arno Kwade, Sandra Breitung-Faes
PhD-Student: Anderson Chagas
Affiliation(s): Institute of Particle Technology (iPAT) – TU Braunschweig
Project Start Date: 01.09.2019
Abstract Date: 16.05.2022

Project Objective:

1. Develop a system engineering approach for optimizing and scaling industrial dry fine grinding processes.
2. Obtain numerical relations about the effect of grinding aid additives and humidity on material behaviour, process aspects and energy requirements.

Material and Methods:

✓ **Material:** Calcinated alumina from Almatis Inc. and Calcium Carbonate from the IMERYS Minéraux Belgique SA.

✓ **Grinding aid additives:** Diethylene glycol (DEG) and Heptanoic acid (HepAc).

❖ **Test equipment (for this report):** Two dynamical air classifiers, the 100MZR in laboratory scale and the ATP-100 in industrial scale, both from Hosokawa Alpine.

❖ **Approach:** The classifiers were operated in passage mode, with samples collect from the feed material and both coarse and fine products outlet for particle size distribution and mass flow rate measurements. Tromp curves were determined for all tests.

Recent Results:

The first two years of this project focused on mapping which process aspects of batch and continuous ball milling are affected by grinding aid additives and how to implement those impacts into already established population balance models. The current funding period focused then, on the air classification stage and on closed circuit milling, mainly in terms of impacts by the use of additives. This document will deal only with air classification since closed circuit trails are still being conducted.

In order to allow the comparison between two scales of equipment, this study used a non-dimensional ratio between classifiers air volume flow and the wheel tip speed times the wheel open area, called here as process parameter ratio. Figure 1 presents the corrected cut size for the 100MZR as a function the process parameter ratio for different material additives. It is evident that the process cut size is not directly affected by the use of grinding aids. This indicates that, in terms of particle size, plant product quality is determined mainly by the classification stage. Figure 1 also presents the bypass of fine particles to the course

outlet. Interestingly, this process inefficiency was only reduced by the use of grinding aids for mid-range values of the process parameters ratio. The following can be concluded: for coarser cut sizes, the classifier don't require additives for an efficient operation and negligible recirculation of fines are observed. As the cut size reduces, the presence of additive become advantageous to maintain a low bypass. However, for very small cut sizes, near the minimum size achievable by the equipment, the process will be inefficient, with or without additive.

Figure 1 compares also the cutsize and bypass variation with process parameters for the two equipment scales. These results confirm the conclusion mentioned before. Additionally, it is also observed that the intensity of the additive contribution is also related to the equipment design. A take home message is that, additive can improve plant efficiency in certain operation ranges, but the combination of a well-designed classifier allied with additives is the best choice.
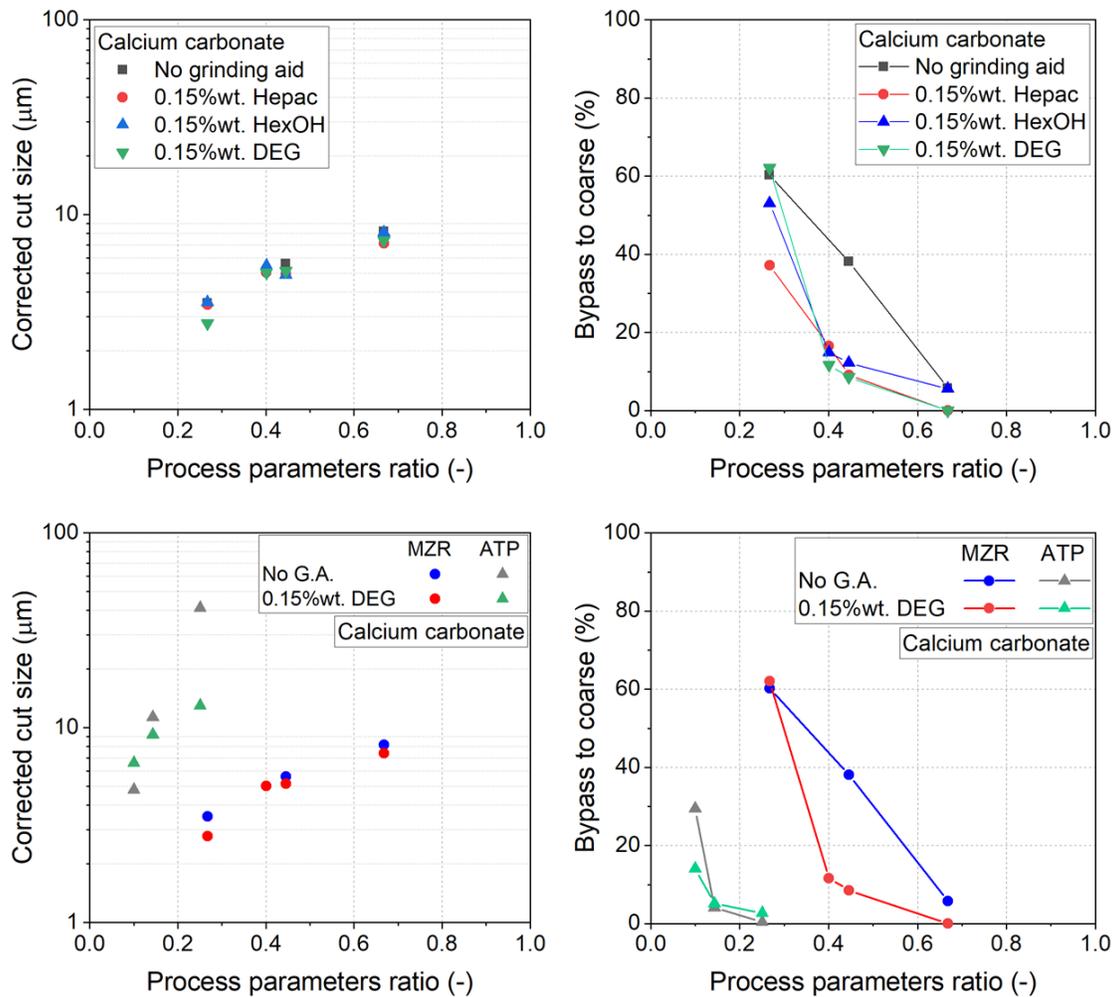

Figure 2: Variation of corrected cut size and bypass of fine particles to coarse outlet with process parameters and different material formulations for the 100MZR (Up) and between scales (below).

Next Steps:

Conclusion of closed circuit trail where the main variables are plant mass feed rate, classifier cut size and additive type. It will be observed mainly plant product size, powder recirculation rate and energy consumption on each unit.

Flowsheet simulation experiments conducted based on population mass balancing. The project finding, in the point of view of additives impacts, will be used to adapt current models from literature. Additionally, it will be proposed a standard material characterization procedure, in the sense of obtaining the parameters required for process simulation.

IFPRI Project Abstract

Modeling of screw feeder performance

Prabhu Nott
Indian Institute of Science
Project Start Date: 21 October 2019
Abstract Date: 12 May 2022

Project Objective:

The main objective of the project is to develop a theoretical understanding and predictive model for screw feeder performance. The model should predict the dependence of feeder performance on powder properties and feeder geometry, and consider complicating factors such as flow variations at the feeder inlet. The model predictions should be tested against experiments on diverse powders.

Approach:

A continuum mechanical model for slow granular flow will be applied to screw feeders. An experimental setup will be constructed to measure the mean flow rate, and the detailed velocity and stress fields to test the predictions of the model. DEM simulations will be used to validate and refine the theoretical studies and guide experimental measurements.

Recent Results:

- We have used a non-local rheological model for slow granular flow, recently developed by us, to study flow in a single screw feeder. When the screw is frictionless and gravity is absent, the model recovers the solution of our earlier, simpler, model. The predictions of the of the wall stress are in good qualitative agreement with experimental data.
- Our experimental feeder setup was used to determine the feed rate of cohesionless glass beads; as predicted by the theory, we find an optimum value of pitch to diameter ratio for maximum feed rate. Our video imaging experiments indicate that the velocity field has a significant rotary component. We have also studied the flow of cohesive powders and find that their throughput is significantly higher than that of cohesionless powders, but fluctuations in the inlet flow are observed.
- We have conducted DEM simulations that couple the inlet hopper with the screw feeder. For short feeders, the fill height decreases with distance from the inlet, but long feeders remain nearly fully filled till close to the exit.

Next Steps:

We will extend our modeling efforts to study the flow when the screw and shaft surfaces are frictional, when there is partial fill, and to study axial variation of perturbations to understand flow variability and control. We plan further experiments and DEM simulations on inclined feeders, and feeders with a decreasing cross section area to promote even flow.

IFPRI Project Abstract

Delivery of a Practical Tool for Predicting the Effect of Solvents and Growth Inhibitors on Crystal Morphology

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Project Start Date: 1 September, 2017
Abstract Date: 13 May, 2022

Project Objective:

The goal of this research is to develop a practical engineering tool for predicting the relative growth rates (growth kinetics) and morphology of solution-grown faceted crystals, including the effects of solvent, and impurities/additives. The methodology will be tested on a variety of systems, including: paracetamol, olanzapine, adipic acid, naphthalene, and a variety of drug substances, all grown from solution.

Approach:

Our approach is to leverage a decade of research & development building our crystal design software tool called ADDICT in order to develop (fast) mechanistic models of crystal growth validated by experiments, molecular simulations and KMC simulations. KMC simulations are utilized for investigating impurity-mediated and noncentrosymmetric crystal growth in conjunction with novel kink density models. Solvent effects are incorporated into the modeling scheme via the use of COSMO-SAC software that we have created ourselves based on literature information.
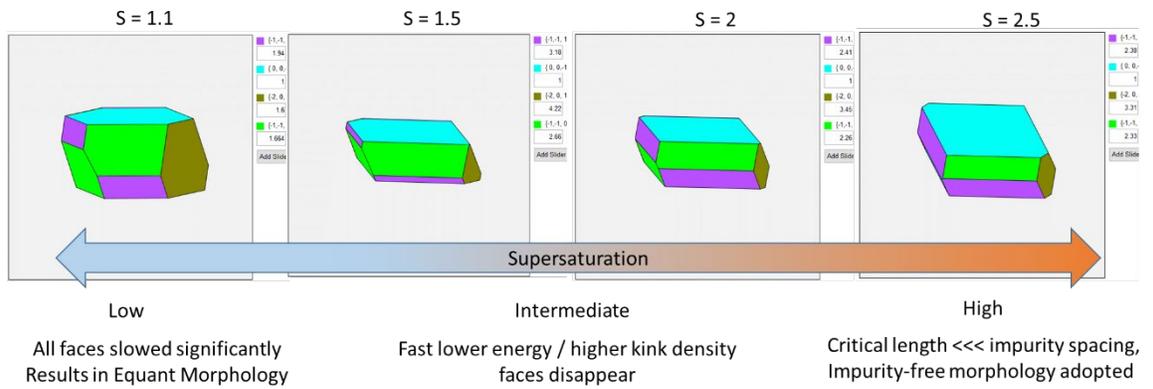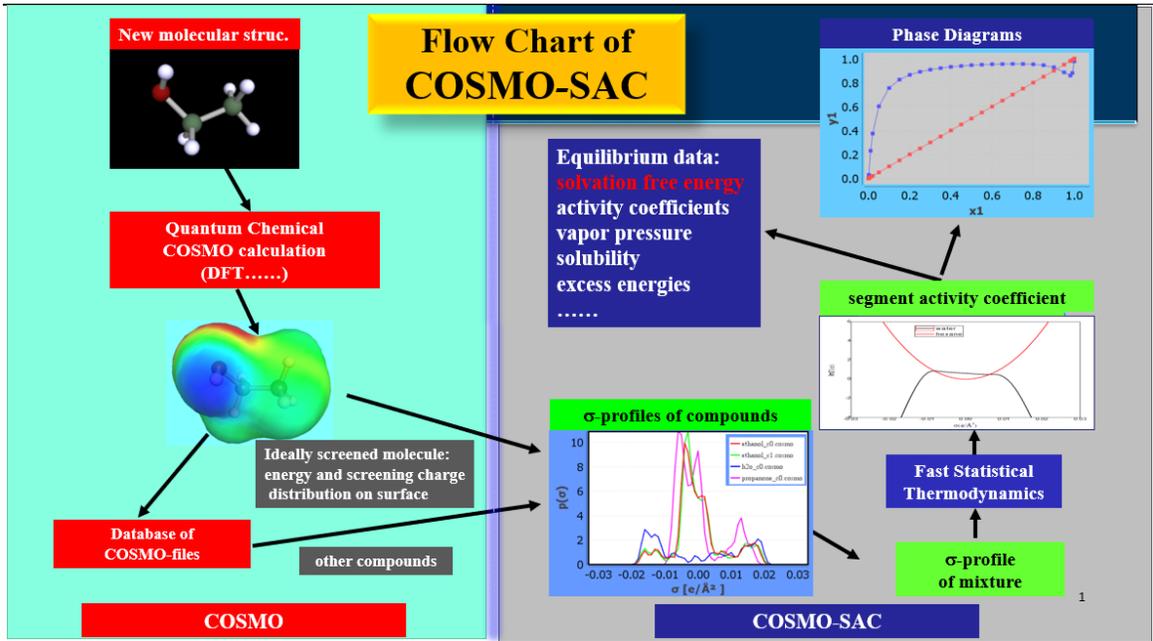
Recent Results:

We have validated and extended our Kinetic Monte Carlo simulations to handle more complex conditions such as impurity-mediated and noncentrosymmetric crystal growth systems. We have completed development of our own version of the COSMO-SAC method and investigated the use of different forcefields as well as improved kink density models to improve morphology predictions. We have also demonstrated the potential of using such inputs to visualize the modified crystal morphology in real time via ADDICT.

Next Steps:

The project will continue to investigate more complex systems (i.e. noncentrosymmetric growth units) and validate novel model developments via KMC simulations. With the help of COSMO-SAC and additional automated forcefields such as CLP, we will look to predict the solvation free energy of solid solutes to enable morphology predictions of mixed solvents and inform antisolvent crystallization. These advances will be adopted within ADDICT to examine the spectrum of crystal morphologies for a range of system growth conditions.

Flow Chart of COSMO-SAC

New molecular struc.

Quantum Chemical COSMO calculation (DFT……)

Ideally screened molecule: energy and screening charge distribution on surface

Database of COSMO-files

other compounds

COSMO

Equilibrium data:
solvation free energy
activity coefficients
vapor pressure
solubility
excess energies
……

Phase Diagrams

σ-profiles of compounds

segment activity coefficient

Fast Statistical Thermodynamics

σ-profile of mixture

COSMO-SAC

S = 1.1   S = 1.5   S = 2   S = 2.5

Supersaturation

Low   Intermediate   High

All faces slowed significantly   Fast lower energy / higher kink density   Critical length <<< impurity spacing,
Results in Equant Morphology   faces disappear   Impurity-free morphology adopted

IFPRI Project Abstract

Spray Drying Kinetics and Morphology

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Lodz University of Technology
Project Start Date: [30 September, 2021]
Abstract Date: [12 June, 2022]

Project Objective:

The aim of the project is to investigate the influence of rheological properties of spray solutions on morphological properties of particles during spray drying. The developed mathematical relationships are also to take into account the kinetics of drying the particles inside the drying chamber.

Approach:

The goals of the project will be achieved through the following steps:
- detailed rheological tests of water solutions of the selected substance will be performed in a wide range of concentrations and temperatures;
- tests of the co-current spray drying process of the tested solutions (for selected process conditions) will be carried out in order to obtain powders for morphological tests;
- the drop drying kinetics will be carried out in a specially designed and constructed column, in which the monodisperse chain of drops will be dried by the free fall method;
- relationships between rheological properties of solutions, drying kinetics and process parameters, and particle morphology will be determined using neural networks.

Recent Results:

The task of determining the rheological properties of the selected material, which is maltodextrin DE12, has been completed.

Viscosity measurements:
Rheological measurements were made with the Physica MR 301 rotational rheometer manufactured by Anton Paar. The measurements were carried out in the temperature range of 20-90 °C using a dedicated system of CC27-SN32556 coaxial cylinders used for rheological tests of low-viscosity media. The flow and the viscosity curves were determined in the range of shear rate from 1 to 500 s^{-1}.
On the basis of the flow curves made in the ordered range of temperatures and concentrations, the tested solutions showed Newtonian properties, and the value of dynamic viscosity was determined as the average value in the considered range of shear rate.

For all tested concentrations, a typical negative temperature dependence of the change in viscosity was observed, which was described by the two-parameter Andrade exponential model:

$$\eta = A \cdot \exp\left(\frac{B}{T}\right) \tag{1}$$

where:

η -dynamic viscosity   [mPa · s]
A -pre-exponential parameter   [mPa · s]
B -temperature parameter   [K]
T -temperature   [K]
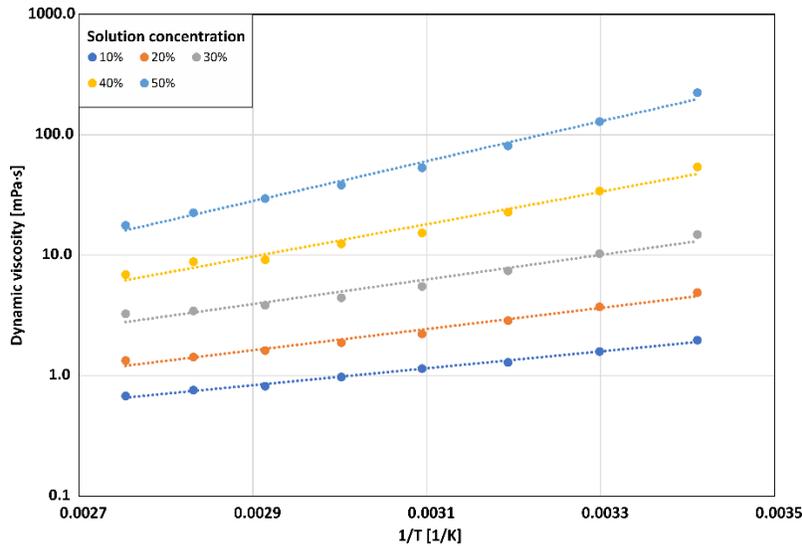
Dependence of viscosity on reciprocal of temperature for aqueous maltodextrin solutions.
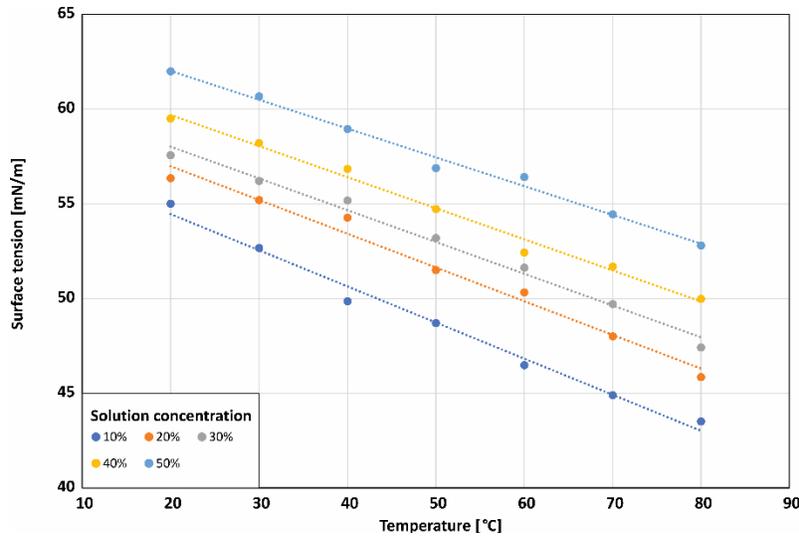
In the tested range of concentrations (10-50 %) and temperatures (20-90 °C), the change in viscosity can be described by equation (1), in which the value of coefficients A and B can be determined on the basis of equations (2) and (3), in which the weight concentration of mlatodextrin ($c_{s,0}$) is the independent variable:

$$A = -0.0002 \cdot c_{s,0} + 0.009 \tag{2}$$
$$B = 54.702 \cdot c_{s,0} + 939.46 \tag{3}$$

Measurement of surface tension:
Surface tension measurements were made with the K100 KRÜSS tensiometer using the ring method (Du Noüy) in the temperature range of 20-80 °C.

Dependence of surface tension on temperature for aqueous maltodextrin solutions

For all tested concentrations, a typical negative temperature dependence of the change in surface tension was observed (Figure 3), which was described by a linear relationship (equation 4).

$$\sigma = c \cdot T + d \tag{4}$$

Summing up, in the tested range of concentrations (10-50 %) and temperatures (20-80 °C), the value of surface tension (mN/m) can be described by the equation (4), in which the value of the coefficients a and b can be determined on the basis of equations (5) and (6), in which the independent variable is the weight concentration of maltodextrin ($c_{s,0}$):

$$a = 0.0009 \cdot c_{s,0} - 0.1978 \tag{5}$$
$$b = 0.1601 \cdot c_{s,0} + 56.838 \tag{6}$$

Measurements of the surface tension were not carried out at the temperature of 90 °C due to the too high probability of solvent evaporation due to the large free surface of the liquid in a dedicated, factory measuring vessel. High temperature and large contact surface with air lead to local concentration of the solution, which could lead to measurement errors related to the change of concentration.

In addition, in the case of tests carried out at high temperatures over concentrated solutions (40 and 50 % by weight), the formation of a film layer on the surface was observed, the existence of which would additionally disturb the correctness of the measurement.

Next Steps:

In the next year of the project, the following will be performed:
- reconfiguration of the drying installation to a co-current system,
- spray drying tests in accordance with the design plan,
- the beginning of the construction of a column for measuring the drying kinetics of a single drop.

IFPRI Project Abstract

Selection of Flow Aids: Model-based Prediction of Flow Properties Enhancements

Principle Investigator: Dr. Rajesh Dave
Affiliation(s): New Jersey Institute of Technology, Newark, USA
Project Start Date: [01 September 2021]
Abstract Date: [15 May 2022]

Project Objective:

Mechanistic prediction of flow properties and its enhancements from particle scale measures, developing a collection of models and decision tools that can be used for flow aid selection while minimizing the extent of experiments needed.

Approach:

Develop particle contact models based on van der Waals (vdW) force approximations and tailor cohesion between micro-sized fine powders by coating them with nano additives. The emphasis is on predicting cohesion reduction through dry coating on reducing the most dominant cohesion forces. Towards that objective, several models are being developed in conjunction with the necessary characterization and test materials.

Recent Results:

Review of existing particle adhesion models was carried out and the deficiencies of those models were outlined. Several of those shortcomings were addressed, such as the particle shape, the assessment of the particle surface roughness and models to account for their effect, the influence of nano-silica agglomeration, etc. Dozens of industry relevant fine powders were considered and tested for key bulk properties before and after dry coating with hydrophobic and hydrophilic silica at various amounts, leading to different levels of theoretical surface area coverage (SAC). Flowability, packing and powder agglomeration were measured before and after dry coating. Applicability of multi-asperity contact model was tested, specific cases where the model need to be advanced were identified. Powder agglomeration effect on drug particles dissolution was assessed. It was found that dry coating significantly reduced powder agglomeration which led to enhanced dissolution even when hydrophobic silica was used. Powder agglomeration was identified as a material sparing screening indicator of powder flowability and processability.

Next Steps:

Work will continue in expending the models for guest-host compatibility, selection of guest amount, accounting for surface roughness and particle shape, and linking particle scale to bulk scale properties. Experimental investigation will include assessment of surface roughness, surface area and powder agglomeration, and building of bulk property database. Enhanced interactions with the IFPRI members and submission of manuscripts covering project advances are planned.

IFPRI Project Abstract

Air-induced defect formation during powder compaction

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Project Overview: This work focuses on developing new simulation capabilities to improve the manufacturing of pharmaceutical tablets using powder compaction processes. Specifically, we concern ourselves with the expulsion of air during the compaction, which is initially trapped in voids between powder particles and at the die-powder interface. A desire to increase throughput of tablet forming requires high speeds of compaction. High compaction speeds lead to turbulent flow and high air pressures within the voids between particles that can cause cracking or capping/lamination after ejection from the die. Defective tablets must then be thrown out leading to wasted material. The objective of this research is to use a mechanics-based approach to create models capable of predicting the conditions under which defect formation will occur during powder compaction. The research has two major branches. The first is to create a discrete model capable of capturing the compaction process. To accomplish this an improved contact model for powders undergoing high deformations is being developed. Coupling of the discrete element method with the improved contact models and computational fluid dynamics will then lead to high resolution numerical "experiments". These can then be used as a guide for the second branch of the research–developing a (computationally cheaper) continuum mixture model capable suitable for powder compaction. When complete, the two branches will provide a robust complementary toolkit for predicting defect formation during powder compaction that properly considers the coupled mechanics between the air and powder.

Current Research Thrust–Towards continuum accuracy at low computational cost: an extended Hertz contact model for elastic-plastic particles: We present a contact model able to capture the response of interacting elastic-plastic particles. The model adopts a constitutive form for the stress along the contact identical in construction to that of Hertz's law. The key difference is that it accounts for changing curvature of the contacting surface, an effect that arises due to plasticity causing permanent plastic deformation leading to blunting. Importantly, our model distinguishes itself from previous large deformation contact models in that it allows pointwise variation of the plastic displacement and hence curvature along the contact rather than assuming a new constant radius of curvature after plastic deformation, an assumption that leads to a non-physical stress profile. In the absence of plastic deformation, the choice of constitutive form naturally reduces to the celebrated Hertz's law.

To validate the model, we compare it to finite element simulations of elastic-plastic contact. These comparisons show that the proposed contact model is able to accurately capture the total force at the contact and the plastic deformation upon unloading. The end result is a powerful contact model that seamlessly extends Hertz's law into large deformation. Moreover, the contact model allows pointwise evaluation of stresses and plastic

displacement across the contact, counter to typical contact models which lack pointwise evolution of quantities. This property moves the contact model closer to the more robust solutions provided by the continuum mechanics of solids without sacrificing computational efficiency.