IFPRI Project Abstract

Single Droplet Drying at High Temperatures

Prof Andrew Bayly; Ph.D Students: Wael Ebrahim, Tien Nguyen
Institute of Particle Science and Engineering, School of Chemical & Process Engineering, University of Leeds
Project Start Date: April 2016
Date: 1st June 2018

Project Objective:

This project seeks to develop experimental and modelling methods that enable dried particle structure, properties and drying rate to be predicted based on droplet drying history. The project will focus on effects driven by boiling and look to develop material independent models which capture behaviors of industrial interest. In particular it will look to address key limitations in current understanding: *1>* the impact of a non-isothermal drying history on particle structure and consequently drying rate; *2>* improved measurements of material properties under the non-equilibrium conditions experienced during drying; *3>* extension of models to include the mechanics of structure formation.

Approach:

Initially, material classes and materials will be identified. Novel experimental rigs and methods will be developed to allow unsteady drying behaviour to be investigated and material properties to be measured under relevant conditions. Established models will be extended to include better material models and mechanical deformation. We will bring this together into a regime map(s) which links material properties, particle size, initial moisture content and drying history to morphology.

Recent Results:

Experimental development for regime mapping: Single droplet filament rig developed, upgrades to imaging, geometry, control, droplet handling and analysis. Drop tube now in place. Mass measurement technique successfully evaluated.

Mapping: Detailed investigation of sucrose, HPMC, sodium silicate. HPMCs with different viscosity and different solvent investigated. Behaviours linked to phase change and material properties

Material properties: Evaluation of rheologies, diffusivities. Atomic Force Microscope investigated for solid material measurement.

Modelling: Bubble expansion models, in non-drying systems in 1-D, 2D/3-D using finite volume/VOF method (Fluent) and finite element/ALE method (Comsol)

Next Steps:

Regime mapping: Linking filament rig measurement to 1-D drying model to develop regime map, prediction of puffing onset. **Drop tube:** compare to filament rig, understand size effects. **Mass measurement:** drying rate once deformation has started and particle density. **Modelling:** explore non-viscous material models, couple bubble expansion and drying models. **Material properties**: back calculate via single drop experiment; collaboration with Schutyser, Wageningen and Reid, Bristol.

Nonlocal Rheology of Intermediate Granular Flows: Particle Shape and Size Distributions

Karen Daniels
Dept. of Physics, North Carolina State University, USA
Project Start Date: 10 August 2015
Abstract Date: 29 May 2018

Project Objective:

Our research aims are to connect grain-scale parameters to macroscale behaviors within a sheared granular material, through the use of a nonlocal rheology. We aim to answer 3 questions: (Q1) What do the flow field and fluidity field look like for real experiments? (Q2) How do the empirically-measured material properties and cooperativity length vary as we change particle shape and size and distributions in a controlled way? Do these properties help distinguish the competing models? (Q3) Can the fluctuations in local forces (via force chains breaking/forming) account for the non-local contribution to the fluidity?

Approach:

We perform experiments in a quasi-2D annular shear cell [1], in which tracking the locations and velocities of individual particles is possible. This allows for direct tests of two competing nonlocal theories [2,3] which so far have seen little experimental validation. The present work focuses on testing these models using 4 different sets of laser-cut particles. We seek to uncover whether (Q2) the nonlocal rheology approach works for non-circular particles, and what principles underlie the dependence of the fit parameters on particle properties. Since the apparatus at NC State has not been able to reach the intermediate flow regime, we have added an IFPRI-funded collaboration with Nathalie Vriend, to perform and analyze experiments in a chute flow. This aspect of the project is described separately, and will lead to advances in our understanding of (Q3).
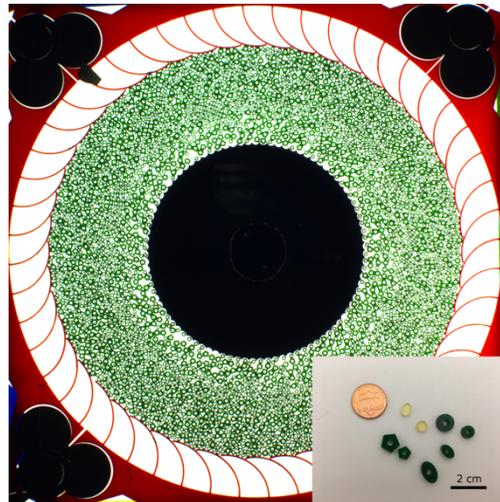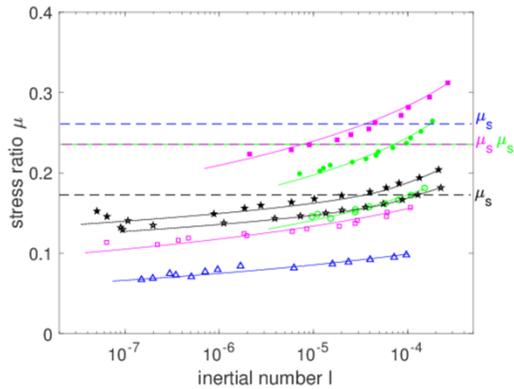

Figure 1: Top view of apparatus, with leaf springs around outer confining wall. The inner disk rotates, shearing the granular material. New laser-cut particles are shown as an inset.

Recent Results:

Since the last annual meeting, we have conducted experiments on the new, laser-cut particles and determined that they (as also seen in the results last year) can successfully be modeled using nonlocal rheology. Again, we find that it is possible to conduct experiments under one set of flow conditions, and then use the same 3 fit parameters to model the same particles until new flow conditions (here, higher pressure and packing fraction). We have begun to identify which model parameters are associated with which particle properties. We find that, unlike for simulations of circular particles [4], there is not a universal formula for a particle-scale measurement of the particle fluidity.

Figure 2: Success of nonlocal rheology [2] in describing flow of acrylic circles (green circles), ellipses (magenta squares), pentagons (black stars), and original photoelastic circles/ellipses (blue triangles) at both higher (solid symbols) and lower (open symbols) pressures.

	Vishay circles/ellipses	acrylic ellipses	acrylic circles	acrylic pentagons
μ_s	0.26 ± 0.02	0.24 ± 0.02	0.24 ± 0.02	0.17 ± 0.01
b	1.1 ± 0.3	1.1 ± 0.5	1.1 ± 0.6	1.1 ± 0.6
A	0.402 ± 0.003	0.231 ± 0.003	0.280 ± 0.003	0.101 ± 0.001

Figure 3: Nonlocal fit parameters determined for each set of particles. We find that the yield stress ratio μ_s (determined by slow-shear experiments) is shape-dependent for same material, the local parameter b is insensitive to material and shape, and the nonlocal parameter A is sensitive to both material and shape.

Next Steps:

This project is in its final year before renewal. In order to make nonlocal rheology easy to use in the largest variety of conditions, two things are required. First, to better-understand (rather than measure directly) the appropriate boundary conditions to allow for prediction without first conducting experiments. Second, the key open question surrounding the particle-shape would be conduct systematic variations in particle properties in order to provide predictions for each parameters. Finally, we have uncovered some interesting observations relating force chain fluctuations to the location of μ_s in the experiment; relating that quantity of the physics of phase transitions (i.e. as a susceptibility) would be interesting.

References:

[1] Tang, Brzinski, Shearer, Daniels. "Nonlocal rheology of dense granular flow in annular shear experiments." *Soft Matter*. 14, 3040-3048 (2018).
[2] Kamrin & Koval. "Nonlocal Constitutive Relation for Steady Granular Flow." *Physical Review Letters*. 108, 178301 (2012)
[3] Bouzid, Trulsson, Claudin, Clément, Andreotti, "Nonlocal rheology of granular flows across yield conditions." *Physical Review Letters*. 111, 238301 (2013)
[4] Zhang & Kamrin. "Microscopic Description of the Granular Fluidity Field in Nonlocal Flow Modeling." *Physical Review Letters*. 118: 058001 (2017)

IFPRI Project Abstract

Non-local effects in intermediate flows

Nathalie Vriend and Karen Daniels
DAMTP, University of Cambridge, UK
Dept. of Physics, North Carolina State University, USA
Project Start Date: 01/11/2017
Abstract Date: 25/05/2018

Project Objective:

The aim of this collaboration is to perform tests of two nonlocal rheologies using a faster flow (intermediate flow: $I \sim 0.25$) than is available in the annular Couette cell at NC State. The lab at Cambridge has an existing chute-flow apparatus and imaging system, which has been slightly adapted for this research, and is able to reach these higher inertial numbers.

Approach:

Identical custom-made 2D photoelastic particles allow for the direct measurement of the contact forces on each particle, combined with particle tracking analysis for dynamical properties. Contact-force measurements can be time-averaged and coarse-grained to provide the continuum stress fields at the boundaries of the materials to test the rheologies. We obtain continuum fields, as a function of depth, for the velocity, and normal and shear stresses, and use this to test the nonlocal rheology against $\mu(I)$ data.

Recent Results:

From the continuum fields, we are able to calculate the stress ratio, granular fluidity, fluctuations in light intensity and decorrelation times, which can be analyzed to reveal the underlying physical behavior. We found a strong correlation (1) between stress ratio μ and force chain fluctuations, and (2) between the inverse of the fluidity g and the decorrelation time τ. Interestingly, the latter correlation shows a clear transition in slope at the yield ratio μ_s.

Next Steps:

This one-year project has already produced very interesting and novel experimental results, which will allow us to continue the work of validating nonlocal rheologies. The next steps are to interpret the different theories with our data to provide a quantitative comparison.

IFPRI Project Abstract

Prediction of the Effect of Solvents and Impurities/
Additives on Crystal Shape and Growth Kinetics

Michael F. Doherty
University of California Santa Barbara
Project Start Date: 1 September, 2017
Abstract Date: 31 May, 2018

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, ammonium acetate and a variety of drug substances, all grown from solution.

Approach:

Our approach is to leverage six years of research & development building our crystal design software tool called ADDICT. Two major lessons emerged from the last six years of R&D; (1) the growth models needed to be extended to cover non-centrosymmetric growth units (i.,e., real molecules), and (2) that the architecture of ADDICT needed to be completely re-invented in order to cope with cocrystals, solvates and organic salts.

Recent Results:

We have just completed a total re-write of ADDICT tool to make it applicable to a much wider selection of growth units (all developed in Carl Tilbury's doctoral dissertation, 2017) and crystal forms (e.g., cocrystals, solvates, etc.).

Next Steps:

We will spend the next year testing ADDICT for a wide variety of complex crystalline forms, and for crystals that exhibit supersaturation-dependent morphologies. We will then start to extend the models in the "growth engine" to encompass new types of materials, such as solvates, cocrystals and organic salts. We will seek IFPRI's advice for the best order to approach these classes of materials.

IFPRI Project Abstract

Mixing Rules for Powder Mixing

Indresan Govender[1,2], Suren Moodley[1], Marcelle Pillay[1], Malcom Rengasamy[1]
[1]Chemical Engineering, University of KwaZulu-Natal, Glenwood, Durban, South Africa, 4041
[2]Chemical Engineering, University of Cape Town, Rondebosch, Cape Town, South Africa, 7701
Project Start Date: 02 February 2018
Abstract Date: 31 May 2018

Project Objectives:

1. Develop fundamental scaling rules for different mixing mechanism.
2. Determine relative importance of each mixing mechanism.
3. Determine influence of material properties on these mechanisms.

Approach:

To evaluate mixing rules in rotating drum mixers using:
1. in-situ measurements via Positron Emission Particle Tracking (PEPT)
2. numerical simulation via the Discrete Element Method (DEM) with the granular suspension modelled via explicit integration of a lubrication force into the DEM framework.
3. Continuum modelling with PEPT and DEM data as inputs to the continuum equations.

The project will be conducted in three phases with first phase (in-situ measurements using PEPT) reported herein.

Recent Results:

Positron Emission Particle Tracking (PEPT) was used to measure the 3D trajectory of a binary mixture (3mm and 5mm diameter plastic beads) within a laboratory rotating drum fitted with lifter bars. The experimental matrix spanned four fill fractions and seven drum rotation rates across the cascading and cataracting Froude regime. Matlab code was developed to evaluate mixing proxies: shear rate, energy fluctuation, probability distribution, and Péclet number. The initial results clearly show radial segregation by size consistent with the Kinetic Stress Theory (KST) at low Froude numbers; however, *exactly* the opposite behaviour to KST is observed for Froude numbers consistent with cascading and cataracting.

Next Steps:

The next year (phase 2) will focus on a detailed analysis of the large set of PEPT data already gathered. The PEPT numerical schemes will be necessarily extended to include a full field analysis of the mixing proxies. The numerical modelling via DEM will also begin in phase 2.

3D PRINTED "PERFECT PARTICLES"

Karen Hapgood
Deakin University
Project Start Date: Jan 2018
Abstract Date: June 2018

Project Objective:

The 2nd part of this project aims to demonstrate the multiple ways that 3D printing can be used to advance our understanding of powder technology – including powder flow & segregation, agglomerate dissolution, agglomerate breakage. Each idea would be demonstrated up to "proof of principle" stage, and the published and made available (including files) to enable the entire particle technology community to implement the ideas more broadly.

Approach:

A range of particles with different properties will be 3D printed, varying particle shapes, sizes and/or densities, similar to what is done *in silico* in DEM simulations, and physical experiments conducted.

Recent Results:

The focus over the last 12 months has been on finalizing the breakage project, and why the quantitative compression simulations did not agree with the experimental results. We used FEM of doublets to establish the linear elastic region of the bond deformation, over which the TBBM model is valid. The non-linear behaviour observed experimentally is highly reproducible but not modelled by any model we know of. A new PhD student has started (but is focusing on binder jetting printing of powders) and a new postdoc has been hired (starting June 4th). Access to EDEM and ROCKY software at Deakin has been arranged and some initial non-spherical particles have been identified and printed.

Next Steps:

* Initiate basic DEM simulations and the effect of non-spherical particles, and conduct matching experiments to demonstrate the technique.
* Write review of 3D printing technology in particle applications (food, pharma, catalysis, consumer products).
* Write a review of the key research groups working on 3D printing internationally and their specific expertise.
* Begin on the other topics outlined – dissolution of test agglomerates, wetting into irregular porous media, use of 3D printed experimental "boundary conditions".
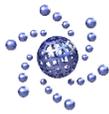
IFPRI Project Abstract

Flowability Assessment of Weakly Consolidated Powders

Colin Hare[1], Ali Hassanpour[2]
[1] Department of Chemical and Process Engineering, The University of Surrey, UK
[2] School of Chemical and Process Engineering, The University of Leeds, UK
Project Start Date: 01 October, 2017
Abstract Date: 01 June, 2018

Project Objective:

Measurement of unconfined yield strength of powders can be made with a variety of commercially available shear testing devices. Traditional flowability measurement devices have a number of short comings, e.g. reproducibility of unconfined yield strength is greatly reduced at low stresses, sometimes measurement is inconsistent with observed behaviour, or materials found to be cohesionless may still have practical differences. Generally the onset of flow is measured, which may not be a complete flow description. IFPRI seek to develop a theoretical understanding of flow of weakly consolidated and weakly cohesive powders.

Approach:

We adopt the ball indentation method for determining powder flowability by measuring the resistance of a powder bed when penetrated by a sphere which is quasi-statically introduced. It is necessary to determine the constraint factor of a powder in order to infer the yield stress from the indentation measurement. Indentation measurements are carried out on vertically consolidated beds and sheared beds (critically consolidated) at the same major principal stress as shear cell and uniaxial compression measurements, in order to compare flowability measurements and determine constraint factor. The influence of single particle properties on constraint factor is explored in order to be able to predict its value so that indentation alone can be used to infer the yield stress.

Recent Results:

In the first funded period the ball indentation method was applied using vertical consolidation to the same stress as the major principal stress in the shear cell, in order to determine constraint factor. This approach has now also been compared to measurements by uniaxial compression, where consolidation and failure stresses are in the same direction in both techniques. Additionally, ball indentation has been applied to beds which have been critically consolidated by pre-shearing in a shear cell, using DT51 titania (Crystal Global). Figure 1 shows the bed surface available for indentation after pre-shear at a range of stresses. At low stresses insufficient material is retained in the

shear head upon removal, as such the exposed surface has not been critically consolidated and the stress state is unknown. As pre-shear stress is increased more of the critically consolidated surface is available for indentation, and more successful indents are achievable. It is expected that more cohesive powders will enable measurements to be made by this method at lower stresses, however the titania tested here is very cohesive (ff_c = 1.1 at 1 kPa pre-shear normal stress).

σ_{pre}	0.5 kPa	1 kPa	2kPa	3 kPa	6 kPa
After lid removal					
Reliable indents	0/15	2/15	2/15	4/15	8/15

Figure 1. Bed surface available for indentation after pre-shearing in a shear cell

Constraint factor is determined by comparing indentation hardness measured after vertical consolidation with unconfined yield strength determined by (i) a shear cell (mixed method), (ii) uniaxial compression (vertical method) and additionally (iii) by comparing indentation hardness measured on a sheared bed with unconfined yield strength determined in the shear cell (critical method). Figure 2 shows the constraint factor determined through these methods. Constraint factor determined by the vertical method is almost independent of consolidation stress in the reliable range (> 3 kPa), whereas the mixed and vertical methods exhibit an increase in constraint factor with applied stress.
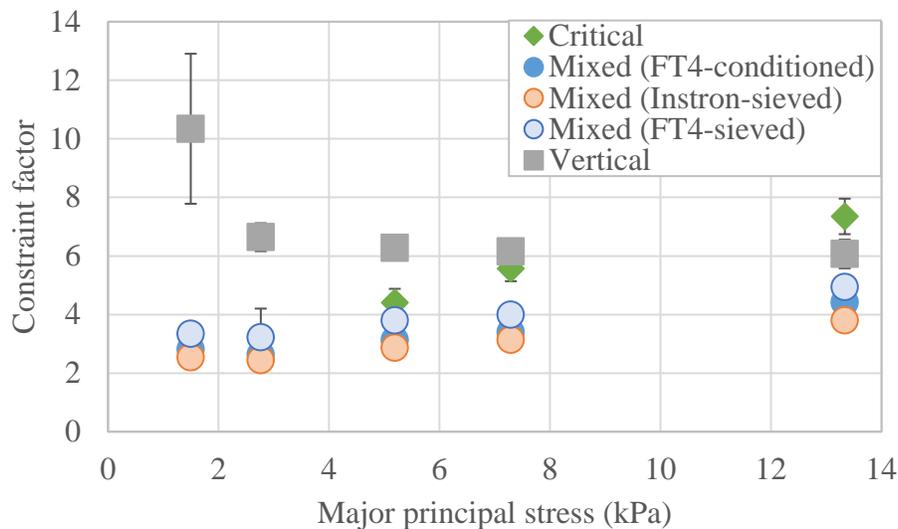

Figure 2. Constraint factor determined by the three methods

Next Steps:

The flowability of various grades of titania will be measured using the approaches applied so far, as well as operating the shear cell under constant volume control, with measurement of normal and shear forces. The most promising methods will be applied for other powders, such as alumina. A key aspect of the analysis will be to investigate the optimum approach to estimate the curvature of the yield locus. Additionally, the flow field around the ball indenter will be assessed by Synchrotron radiation at the Diamond Light Source (Oxford, UK).

IFPRI Project Abstract

Quantitative Prediction of Segregation at Process Scale

Joseph J. McCarthy
University of Pittsburgh
Project Start Date: 1 September 2015
Abstract Date: 11 June 2018

Project Objective:

This project is aimed at identifying critical material and process parameters that control the extent of powder segregation, such that we might develop quantitative models that predict segregation. These predictive models should be valid at full process scale and are to be validated against appropriate experiments.

Approach:

Segregation rate models hold promise for scale-up via continuum-level analysis using device-specific transport equations; however, experimental validation of dynamic models is extremely difficult and typical segregation models are not inherently built with scale-up in mind. Our approach overcomes the experimental limitation by exploiting a novel framework for segregation testing based on establishing an "equilibrium" between mixing and segregation. With regard to the scale-up limitation, we have built – and continue to build – segregation models that are written expressly in dimensionless form with an aim at both connecting segregation modeling to flow rheology as well as enable more transparent scale-up. We are using our experimental segregation validation framework both to benchmark existing segregation rate models as well as to test our new and developing models.

Recent Results:

Our most recent successes include experimental validation of our novel density-based segregation model which was the first of its kind to establish a direct analytical connection between to granular rheology. Moreover, we have extended and computationally tested segregation models based on size differences and cohesive density-driven systems. Finally, a new model for shape segregation has been proposed and extensively tested experimentally.

Next Steps:

In our second phase of IFPRI funding we anticipate completing our modeling efforts for size, shape, and cohesive segregation. In addition, we expect to continue additional experimental validation of these models. Finally, we plan to incorporate our newly developed models into transport equations to enable large-scale validation against process-scale data. Our next year of effort will focus on additional experimental validation as well as transport equation testing.

IFPRI Project Abstract

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

Zoltan K. Nagy
Purdue University, Davidson School of Chemical Engineering, West Lafayette, US
Project Start Date: 10/1/2016
Abstract Date: 06 01, 2018

Project Objective:

The main objective of this project is the design of novel, integrated crystallization systems, which are able to produce wider variety of crystalline materials, in the context of crystal size and shape distribution as well as purity. The attainable region of crystal size distribution (CSD) is widened by the application of recirculation stream and by integrating wet mill for batch, and multiple MSMPR units and downstream wet mill with recirculation stream(s) for the continuous operation. Growth rate modifiers are also considered for crystal size, shape and purity control.

Approach:

Model based approaches are applied, namely the population balance (PB) framework. 1D and 2D PBMs will be developed for the batch and continuous integrated systems, involving the high resolution finite volume method (HR-FVM) for the solution of generated model-equations. In the HR-FVM implementation advanced computing infrastructures, such as parallel calculations on low-cost and high performance GPU accelerators, are involved, which will also provide generic numerical platforms for the aforementioned systems. Novel, iterative model-based experimental design (IMED) will be used for optimal estimation of the unknown model parameters.

Recent Results:

The improved geometrical model based FBRM and PVM soft sensor performance was evaluated by comparing the simulated CLD of a known sphere-like crystals population with the measured CLD of the same crystals. The agreement was acceptable, which enables the application of the proposed transformation for model based control approaches, which also benefits from the auto-correcting feature of the feedback information.

A nonlinear model predictive control system (NMPC) has been developed for the batch cooling crystallization of ascorbic acid. As preliminary work, the secondary nucleation and growth kinetics was estimated based on CLD and concentration data of batch experiments, which was then transferred to the NMPC system. The control system employed high

fidelity full PBM solution through the CrySiV Matlab function, which enabled the temperature profile optimization and the kinetic parameter re-adjustment based on the available experimental data for adaptive feature in a three minutes sampling time. The NMPC produced larger crystals with less nucleation than the corresponding linear cooling operation.
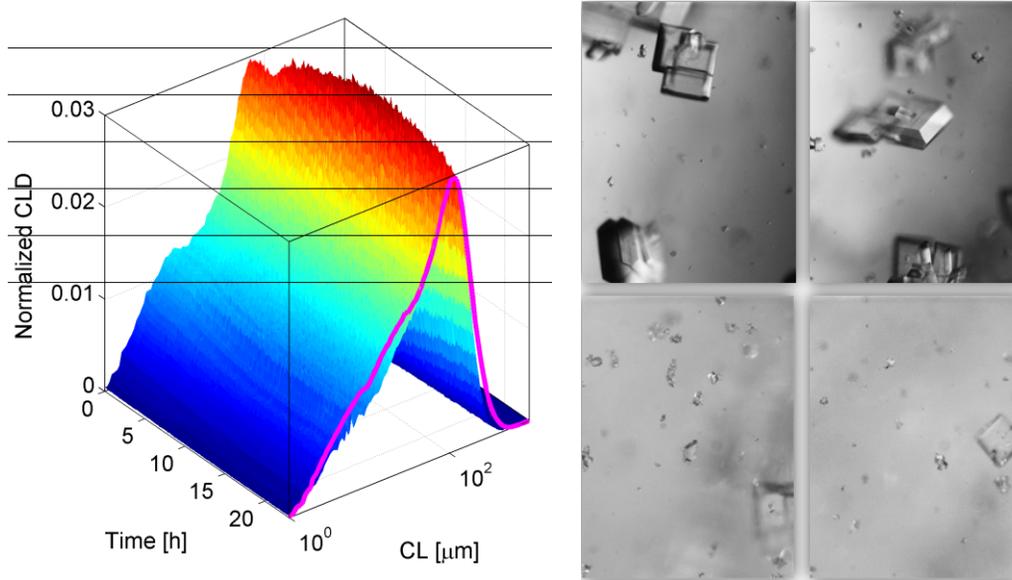

Figure 1. The CLD evolution of the NMPC batch and the crystals produced by the NMPC (top) and linear cooling (bottom)

The simultaneous crystal size and shape control of high aspect ratio crystals was analyzed in a batch crystallizer-external wet mill system through numerical simulations. The model assumed nucleation, growth and dissolution mechanisms in the crystallizer, whereas nucleation, growth, dissolution, attrition and fragmentation were considered in the external wet-mill. The optimization revealed that the attainable 2D CSD region of the integrated system is significantly broader in the case of integrated system because of the more degrees of freedom provided by the recirculation stream and the existence of two systems, which can be decoupled from each-other by turning the pump off.
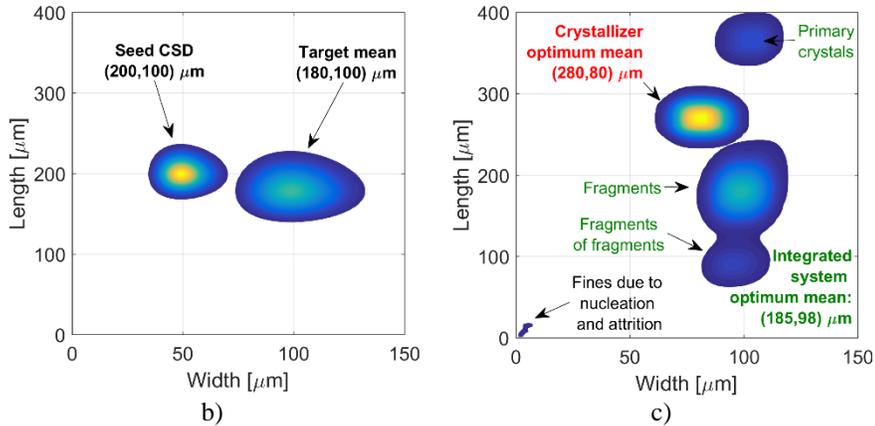

a)
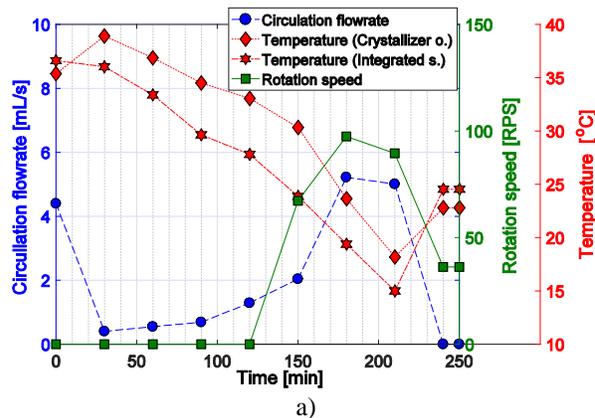
Figure 2. a) Optimal profiles for the integrated system and for the crystallizer-only configuration; b) initial and target CSD; c) CSD realized by the crystallizer-only configuration and by the integrated system

---

Next Steps:

In the next year parameter estimation of a representative 2D crystallization system based on in-situ measurable experimental data will be carried out. Using the calibrated process model, a model based simultaneous size and shape control will be implemented based on high fidelity full PBM and PVM, FBRM and concentration feedback information.

The integrated crystallization-wet mill systems (1D and 2D) will be extended to continuous case with multiple MSMPRs for nucleation, growth and dissolution.

The extension of the CrySiV GUI, as well as its generic PBM solver function counterpart will be continued.
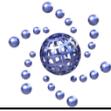
IFPRI Project Abstract

Milling and Material Grindability

Jin Y. Ooi, Lige Wang, Xizhong Chen, Jin Sun, Jian-Fei Chen*

University of Edinburgh, U.K.; * Queen's University Belfast, U.K.

Project Start Date: 07 January 2013

Abstract Date: 30 May 2018

Project Objective:

This project aims to develop a robust methodology to characterize the grindability of particulate products in milling operations. This involves developing grindability measures to characterize the comminution of particulates. The measures, when coupled with milling function through computational modelling, will be used to predict milling processes.

Approach:

1. Explore the population balance model (PBM) in comminution process and conduct PBM simulation to predict the product size distribution under varying operational conditions.

2. Conduct multiscale analysis of DEM simulation at particle scale and further coupled with PBM at process scale to inform the milling behavior in an impact pin mill;

3. Develop the material function: characterize particle breakage subjected to the dominant loading events identified within a milling operation.

Recent Results:

1. Edinburgh bonded particle model was calibrated through experimental single particle impact test and used to predict the breakage pattern under high impact velocities.

2. Coarse-grained model was developed for analysis of breakage mechanisms under impact

3. DEM-PBM coupling model was used to predict the effect of high feed rates on the product size distribution of impact pin mill

Open questions:

1. We propose that material grindability should be "measured" in the context of prevailing particle dynamics in a specific mill – is this reasonable proposition?

2. Will the DEM-PBM coupling model work for a variety of milling predictions?

3. Can we better utilize DEM simulations to inform the predictions of other mills with increasing complexities?
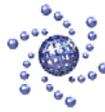
IFPRI Project Abstract

Principle Investigator(s): Mashia M. Foghi, Erik Löwer,T. Leißner, Urs A. Peuker
Affiliation(s): TU Bergakademie Freiberg
Project Start Date: 01.10.2017
Abstract Date: 31.05.2018

Project Objective:

1. Improvement of Dead-End filtration process based on microscale laboratory and macroscale image analysing approaches.
2. Needle-like,cubed-like and spherical shaped particles are used through the filtration process to compare the shape factor effect on the filter cake structure and affecting parameters like as; porosity, permeability, surface area, wettability
3. Investigating the impact of surface chemistry (wetting / DLVO-effects) on the filter cake structure

Approach:

The aim is the estimation of the filtration parameters directly form the information available on the particles like particle shape (distribution), particle size distribution, particle interaction (distribution). On the basis of both macroscopic lab-scale work with a nutsch filter as well as microscopic work using in-situ experiments applying µ-CT (computer tomography) a joint approach is developed to connect particle properties with pore properties and filtration results. The insight given by the experiments will also guide the way what kind of filtration model has to be applied and further developed to describe de-saturation processes.

Experimental:
- ✓ **Material:** $Al2O3$ *and* $CaSiO_3$ particles with different properties are used as solids fraction; the mother liquid is an aqueous salt solution.
- ✓ **Extracting cake structure characteristics:** Porosity: Calculated based on the filter cake structure after drying, Digital image processing and quantitative date exploitation with key figures of the 3D cake-images
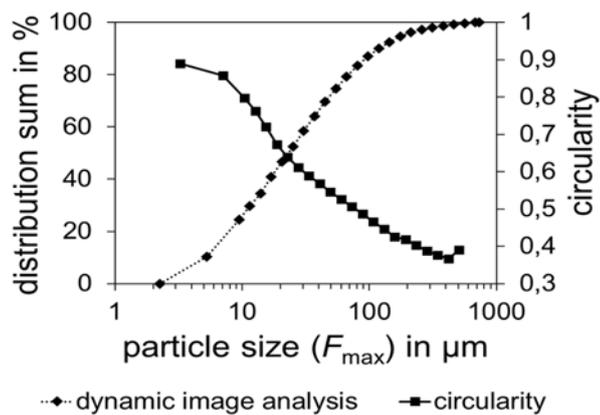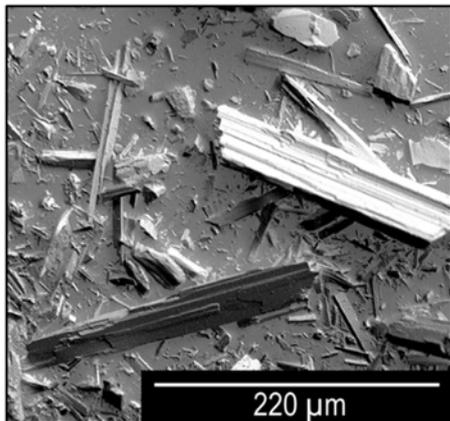
Recent Results:

- ✓ Down-scale of filtration cell for in-situ tomography measurements
- ✓ Three phase identification (gas, liquid, solid) inside the CT by contrast enhancement for future investigation of de-saturation experiments
- ✓ Characterization of particle properties of model materials

Compact particles
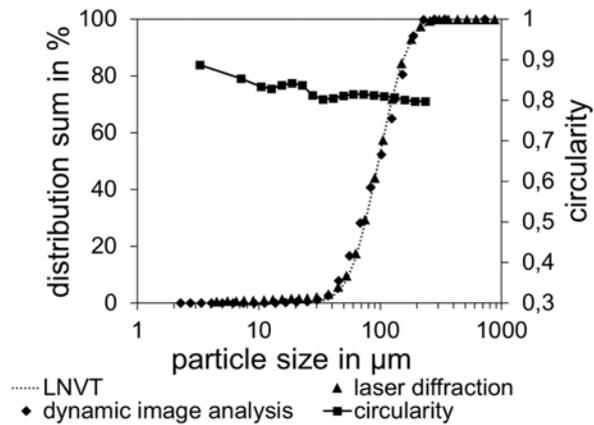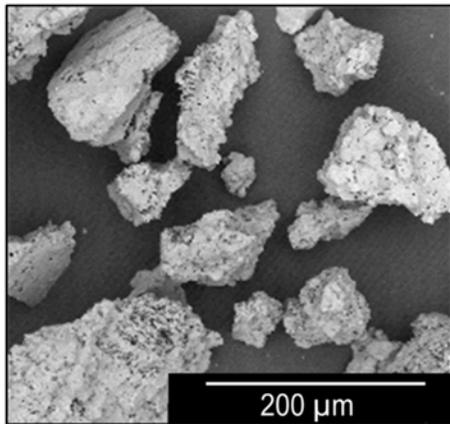
needle-shaped particles

Next Steps:

- Quantitative determination of structural parameters of filter cakes (number of contact points, tortuosity, pore-size distribution, …),
  -1- application of commercial software tools
  -2- development / application customized software tools
- Variation of particle properties
  -1- wetting properties of compact particles
  -2- shape of particles (aspect ratio)
- Phenomenological filtration experiments focused on de-saturation

IFPRI Project Abstract

On the long-term stability of colloidal gels (2)

Wilson Poon
The University of Edinburgh
Project Start Date: 1 October 2016
Abstract Date: 04 June 2018

Project Objective:

A new paradigm for colloidal gel collapse emerged from our first project: dense "debris" accumulates at and then falls from the top of the gel, simultaneously solvent bubbles rise from the bulk, both rupturing the gel. This, together with hydrodynamic back flows, leads to rapid collapse. In the current project, we aim to elucidate the origins of the falling debris and rising solvent bubbles, explore the existence of different collapse mechanisms in other regions of parameter space, and study in detail in the role of hydrodynamics in all processes relevant to triggering gel collapse. We also aim to extend our study into systems with two sizes of particles to move closer to applications. Such binary systems can model both polydispersity and the use of small-particle gels to keep bigger particles in suspension.

Approach:

We use a combination of experiments with well-characterized model systems and simulations. This approach allows quite direct comparison between modelling and experiments. Simulations permit the isolation of different physical effects, enabling us to relate their respective contributions to experimental observations.

Recent Results:

Experimentally, we find that gel-air meniscus curvature of *either sign* leads to unbalanced tangential forces breaking the gel structure into debris that triggers collapse. In simulations, we discover that hydrodynamics clearly distinguish two regimes of concentration: below $\phi = 0.1$, there is no pre-existing percolating cluster and backflow leads to accelerating collapse; above this concentration, gels sediment slowly. Finally, we have established a model system for studying large repulsive particles suspended in a gel of smaller attractive particles; we found a non-trivial effect of the former on the rejuvenation of the latter.

Next Steps:

We believe that our binary model system shows significant potential for qualitatively new insights and we will focus on probing it further using both experiments and simulations. Further work on quasi-monodisperse gels will focus on simulations to explore wider regions of parameter space.

IFPRI Project Abstract

Self-Assembled Monolayers as Nucleating Surfaces to Study Early Formation Pathways of Crystal Polymorphs

Ulrich Wiesner

[Materials Science and Engineering, Cornell University]
Project Start Date: [15 September, 2015]
Abstract Date: [31 May, 2018]

Project Objective:

IFPRI is interested in understanding and controlling crystallinity, polymorphism, and particle morphology in the early formation stages of crystals. High level objective of this project is therefore to identify appropriate model systems to study, adapt, and apply characterization techniques to describe early particle formation states, and to collect data that is relevant for the development of molecular dynamics simulations or other models of assembly of molecules into nascent crystals. Focus should be on identifying possible early non-classical crystallization pathways, including amorphous states and oriented attachment of clusters to growing particles.

Approach:

Self-assembled monolayers (SAMs) will be used to identify model systems for studies of the relationship between surface chemistry, solution conditions, and nucleation and growth of particular crystal polymorphs. Interesting early particle/crystal formation stages will be identified using synchrotron-based *in-situ* x-ray experiments. In final stages of this project cryo-(scanning) electron microscopy will be used to study the morphology of such early particles/crystals.

Recent Results:

Working with the model compound acetaminophen (ACM) and self-assembled monolayers (SAMs) as model surfaces, in previous years we have established that (i) both solvent and substrate work together to control crystal polymorph, and that (ii) on hydrophobic SAMs, pure solvent systems such as ethanol, water, and 1,4-dioxane yield the thermodynamically stable, monoclinic polymorph form I, while mixtures of water and 1,4-dioxane produce the less favored orthorhombic form II.

In the third year of this project, we have studied the early formation stages of these form I and II crystallization events by means of time-resolved *in situ* wide-angle x-ray scattering (WAXS) at Cornell's High Energy Synchrotron Source (CHESS). Using seeded crystallization events of form II crystals and moving the x-ray beam vertically through

the sample we verified that crystallization originates at the substrate-solution interface. Studying spontaneous crystallization events of form I we identified unusual shifts along scattering vector, q, of isolated scattering peaks at the earliest time points of crystal nucleation and growth (see Figure 1) pointing to the possible existence of structural transformations at these early stages.
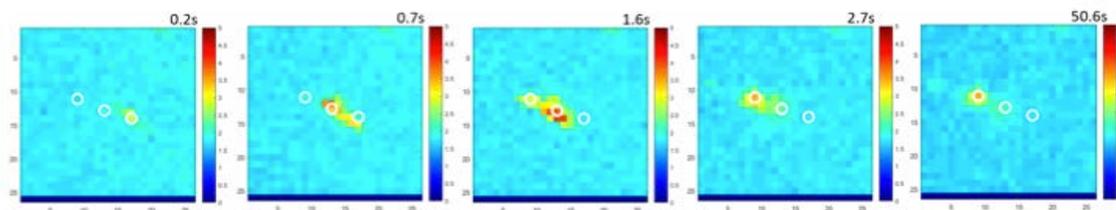

Figure 1. Early time evolution of in situ synchrotron x-ray diffraction peaks for ACM form I crystallization in water on a hydrophobic phenyltrichloro-silane (PTS) based SAM on silicon. Within less than three seconds of the original nucleation of the crystal (times indicated in upper right corner of each frame) the peak moves between three distinct positions (highlighted by white circles) along the scattering vector, q, before it attains its final position (see reference frame after 50 sec. on far right).

Next Steps:

First results of earliest formation stages of the crystallization of ACM on SAMs as revealed by *in situ* synchrotron x-ray experiments in the third year of the project are very exciting and warrant further in-depth studies of these phenomena. If further funded, we are also considering expanding the studies from simple droplet evaporation studies to blade coating experiments as well as to other model compounds.

Die filling of aerated powders

Prof. Chuan-Yu (Charley) Wu, Anastasiya Zakhvatayeva
Department of Chemical and Process Engineering, University of Surrey, Guildford, GU2 7XH, UK
16 October 2017
Abstract Date: 04 May 2018

Project Objective:

1. To investigate gravity die filling of common pharmaceutical excipients, using linear and rotary die filling devices.
2. To investigate suction die filling of common pharmaceutical excipients, suing linear die filling device.
3. To compare the die filling mechanism in suction and gravity filling and investigate potential correlations between powder properties and the die filling efficiency

Approach:

An experimental investigation of the following aspects is carried out:
1. Gravity die filling on the linear and rotary systems: die filling experiments are carried out systematically using seven common pharmaceutical powders. The die filling efficiency is compared with a set of powder properties, like particle size distribution, cohesion, flowability.
2. Suction die filling on the linear system: the device is upgraded with a vertical drive, connected to a piston, positioned inside the die. The downward movement of the piston creates a suction effect that aids the powder flow inside the die. The assembled system mimics therefore the process occurring inside the filling shoe of an industrial tableting machine. The study of suction die filling is carried out systematically, along with a full factorial DOE approach. The results are compared with the gravity filling.

Recent Results:

A rotary die filling system was used for the first part of the experiment (Fig.1). Its performance was compared with that of the linear die filling device. It was shown that the efficiency of the rotary die filling is higher than that of the linear one (Fig.2).

An extensive characterization of powder properties was performed and the results of die filling were analyzed in terms of such powder characteristics as particle size distribution (Fig.3), flow index, cohesion, flow function, air permeability.

The linear die filling device, equipped with pneumatic horizontal drive, was complemented with a vertical electromechanical drive (Fig.4) so that a fully controlled suction die filling system is developed. Special dies were manufactured, equipped with a removable piston (Fig.5).

The efficiency of suction filling at different suction speeds was compared with the gravity filling process. It was shown that the die filling efficiency is dramtically improved with suction filling and the variation of fill ratio with the shoe speed is linear rather than exponential (Fig.6).

Fig. 1. Rotary die filling system, top view.
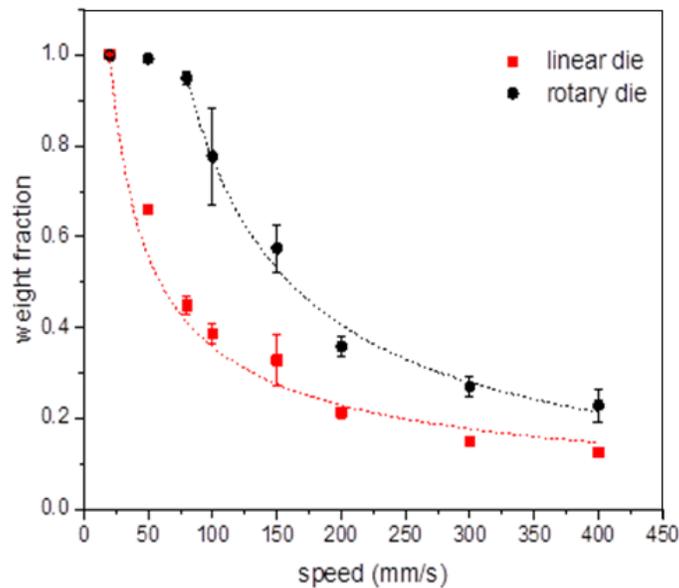
Fig. 2: Comparison of die filling efficiency of mannitol in linear (red line) and rotary (black line) systems.
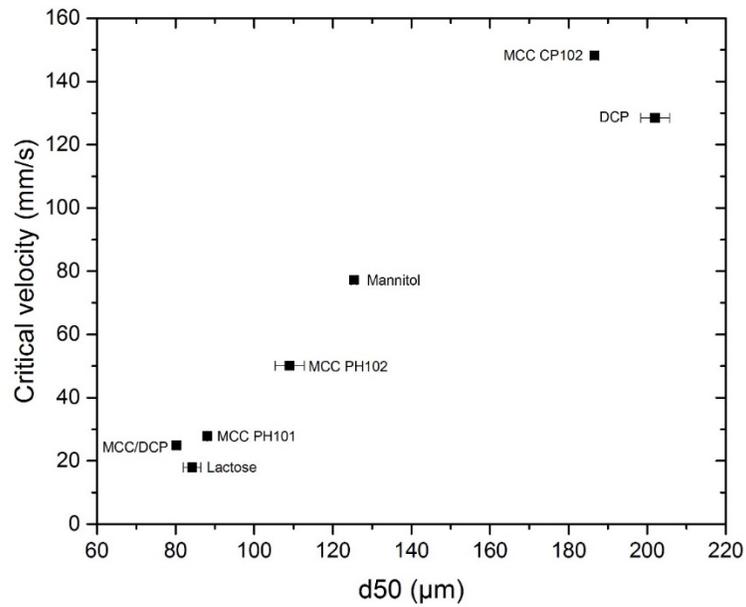

Fig. 3: Correlation between the particle size (d50) and die filling efficiency (fill ratio) for different powders.
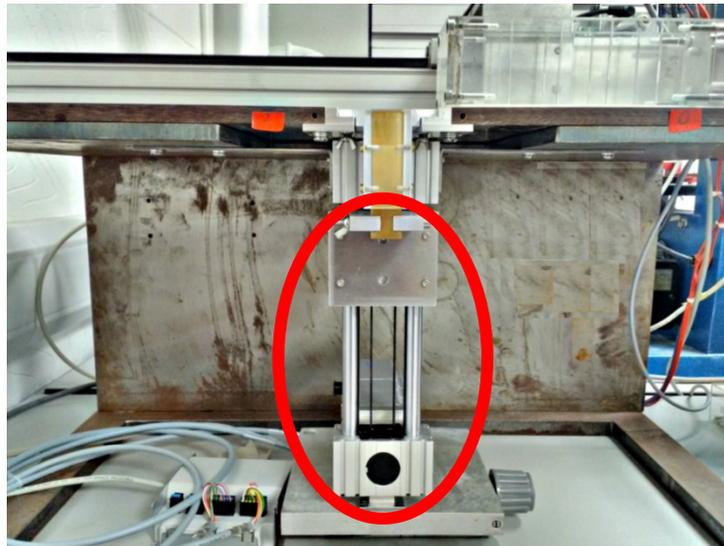

Fig. 4. The linear die filling device with the vertical electromechanical drive (circled in red)
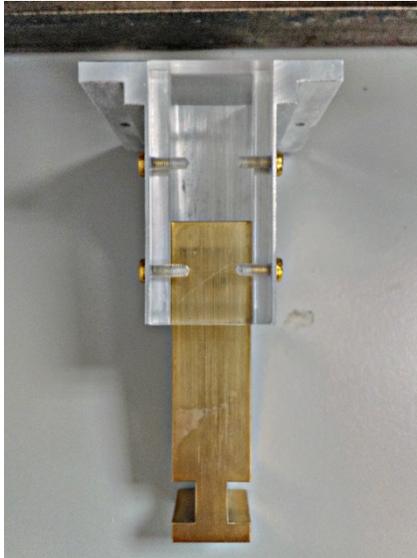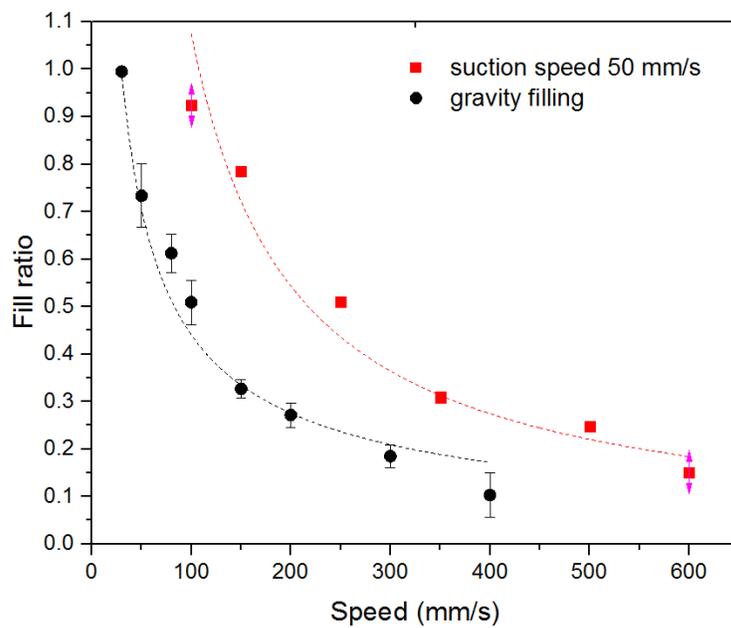
Fig. 5. Square die with a removable piston

Fig. 6. Comparison of die filling efficiency of MCC PH102 on a linear die filling system: gravity fill (in black) and suction fill (in red).

Next Steps:

Further investigation on the topic is needed, specifically:

- Study of suction die filling of blends: investigation of blend uniformity under different suctions speeds.
- Die filling with a paddle die filling system: study of forced feeding mechanism.

IFPRI Project Abstract

Multiscale Modeling of Compaction with Emphasis on Powder Mixtures

Antonios Zavaliangos
Department of Materials Sci. & Eng ., Drexel University, Philadelphia, PA19104, USA
Project Start Date: [2012]
Abstract Date: [3 June, 2018]

Project Objective:

This project focuses understanding the compaction and strength of powder mixtures. Multicomponent formulations are pervasive across diverse industries and understanding aspects of their behavior through modeling with minimum experimentation is highly beneficial, because the experimental matrix that addresses all potential combinations of variables is rather daunting.

Approach:

Modeling of multi-component systems using discrete element method (DEM) is a natural way to approach this complex problem, because the experimental matrix that address all potential combinations of variables is rather daunting. Modeling validated with experiments is the methodology that will provide us with insight into the cause of the various effects and can delineate the relative importance of multiple, coexisting factors. However, major work is required in single material DEM to form the basis of the current approach.

Recent Results:

We continued the efforts on (a) appropriate definitions for damage so that we can quantify propensity for failure, (b) visualization of damage, (c) explored the predictions of the model to the problem of triaxial unloading and its effect on strength as well as the problem of powder sticking. (d) improved our procedure for model calibration, (e) provided insight in the behavior of NaCl+Starch mixtures.