Single Drop Drying at High Temperatures

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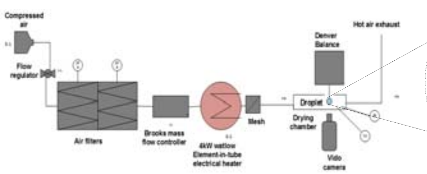
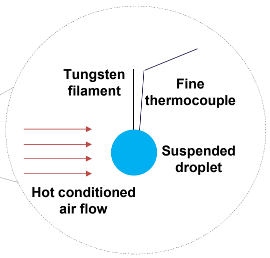
This project seeks to develop experimental and modelling methods that enable dried particle structure, properties and drying rate to be predicted based on droplet drying history. The project will focus on effects driven by boiling and look to develop material independent models which capture behaviors 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. A key goal is the development of a regime map which links material properties and drying conditions to morphology.

Particle property variability in a spray dried product makes structure mapping challenging: new rigs to control size and temperature experience.

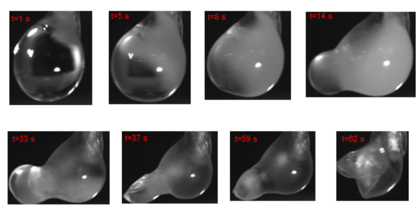
Filament rig

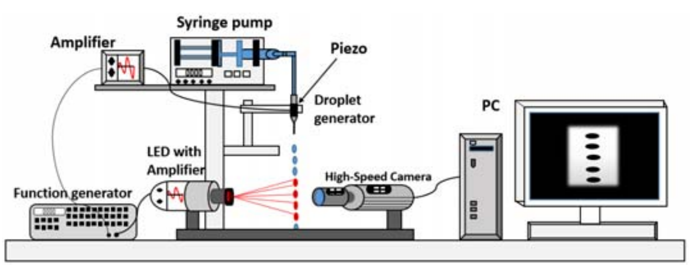


Process flow diagram of a developed single droplet drying rig

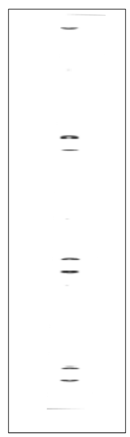


HPMC - T_{air} = 190° C , 5 %, d_{init} = 1.4 mm
inflation and rupture



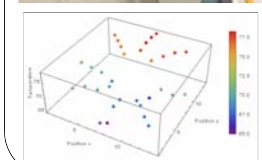


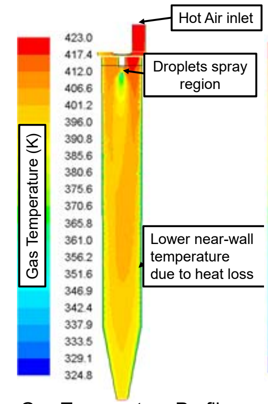
Monodispersed droplet generator - HPMC droplets generated. This will be used in the drop tube.



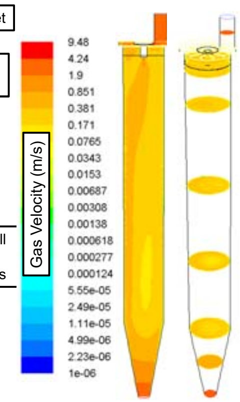
5% HPMC in water at 3 ml/min,
650 Hz, 150 um i.d. nozzle

Dryer/drop tube performance - Inlet air distribution mapped; air and temperature distribution modelled





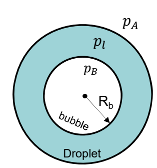
Gas Temperature Profile



Gas Velocity Profile

Modelling

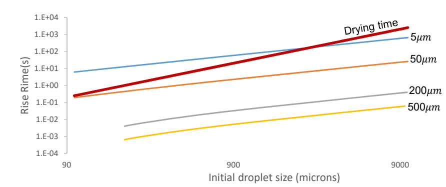
Bubble expansion within a droplet



- **Aim**: to formulate the bubble expansion rate as a function of internal bubble pressure and air pressure.
- **Approach**: Rayleigh-Plesset method.

Symmetrical approach?

Comparison of bubble rise and drying times



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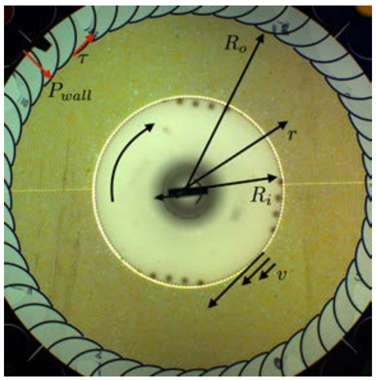
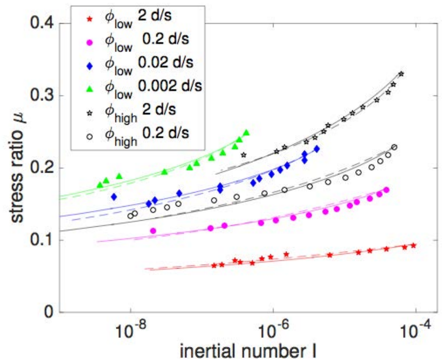
Nonlocal Rheology of Intermediate Granular Flows: Particle Shape and Size Distributions

Zhu Tang, Ted Brzinski, Michael Shearer, Karen Daniels
Dept. of Physics, North Carolina State University

Our research aims are to connect grain-scale parameters to macroscale behaviors within a sheared granular material, through the use of nonlocal rheologies. 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? (Q3) Can the fluctuations in local forces (via force chains breaking/forming) account for the non-local contribution to the fluidity? We will perform experiments in a quasi-2D annular shear cell, in which tracking the locations and velocities of individual particles is possible. This will allow for direct tests of competing nonlocal theories which so far have seen little experimental validation.

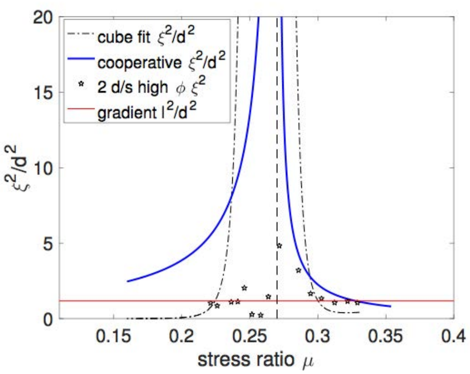


Top view of apparatus, with leaf springs around outer confining wall. The inner disk rotates, shearing the granular material. The amount of deformation of the leaf springs measures the confining pressure on the granular material [1]. The shear stress is measured via the torque required to rotate the inner disk, as well as by calibrated measurements of the deformation of the outer leaf springs.



Rheological measurements from six different experiments (symbols), compared to modelling via the cooperative model [2] and the gradient model [3]. A single set of parameters (for each model) successfully provides agreement for all six runs.

$\mu(r)$ = shear stress / pressure
I = dimensionless shear rate



Measured length scale (symbols, dash-dotted line) compared to predictions from the cooperative model [2] and the gradient model [3]. The nonlocal length scale ξ diverges at the yield stress μ_s, which is a feature of only the cooperative model.

Next Steps

- collect data on how different particle shapes (or other properties) effect the model parameters (Q2) and the choice of model [2,3].
- determine what generalization about the material properties is possible, and which shape/size properties are the most important to characterize in a simplified description.
- implement the tracking of the internal stress field, through the use of photoelastic particles, to more directly test the origins of nonlocality (Q3).

References:

[1] Z Tang, TA Brzinski, and KE Daniels. *Powders & Grains 2017* http://arxiv.org/abs/1704.08295
[2] K Kamrin and G Koval. *Physical Review Letters.* 108, 178301 (2012)
[3] M Bouzid, M Trulsson, P Claudin, E Clément, B Andreotti. *Physical Review Letters.* 111, 238301 (2013)

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Creating tuneable agglomerates via 3D printing

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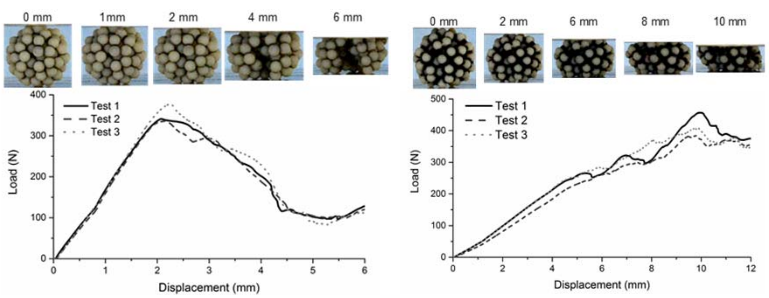
Monash University & Leeds University

It is essential to produce test agglomerates with controlled properties to replicate the complex agglomerate structure, and validate simulation models. In this research, agglomerates with defined and tuneable properties were produced using 3D printing technology. Quasi-static and impact breakage tests were conducted to vary strain rate under controlled conditions. Macroscopic agglomerate breakage experiments were compared with DEM simulation results to validate the newly developed Timoshenko Beam Bond Model (TBBM) contact model. The results shown that DEM simulation was an excellent match for the cubic tetrahedral agglomerate structure. However, DEM underestimated compressive loads for spherical random agglomerates that can be attributed to TBBM bond geometry definition differences and non-linear polymer behavior.

Agglomerate breakage experiment

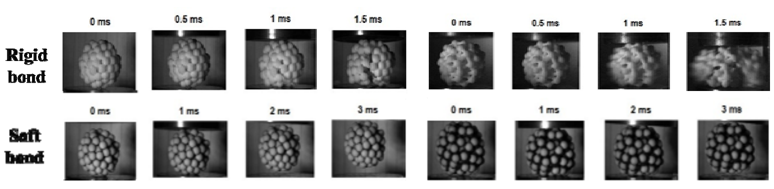
Quasi-static compression tests

- Dense structured agglomerate (ε=44%) shows meridian crack.
- Loose structure (ε=57%) is crushed progressively via ductile failure.



Impact tests

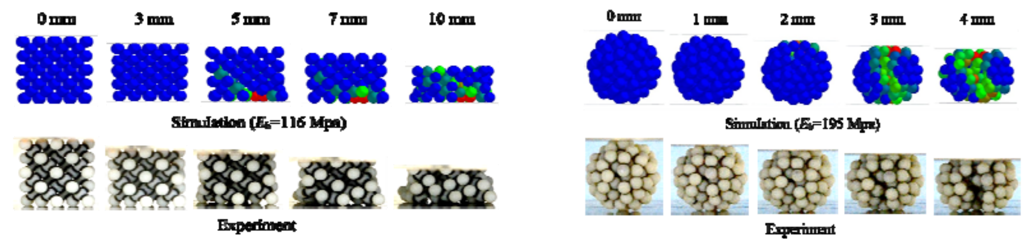
- The rigid material bonds show brittle breakage characteristics, the dense structure shows meridian crack while the loose structure shows oblique crack.
- The agglomerates with soft bonds show rebound behavior.



(a) Dense structure (a) Loose structure

Comparison of agglomerate breakage

- DEM simulation & experiments show similar breakage patterns.
- For the cubic tetrahedral structured agglomerate, the bonds fail along slip planes.
- The spherical random structured agglomerate fails through the meridian plane.

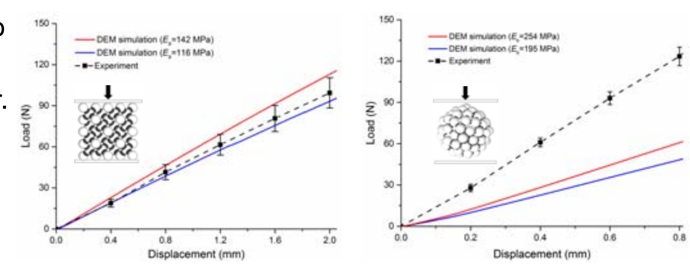


(a) Cubic tetrahedral structure (b) Spherical random structure

Comparison of load-displacement curves

- DEM simulation predictions are within the range of experimental results of cubic tetrahedral structure, but underestimate the compressive loads of spherical random structure.
- The underestimation of compressive loads are attributed to different bond geometry definitions and non-linear behavior of polymer.

(a) TBBM (b) Reality



(a) Cubic tetrahedral structure (b) Spherical random structure

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Flowability Assessment of Weakly Consolidated Powders

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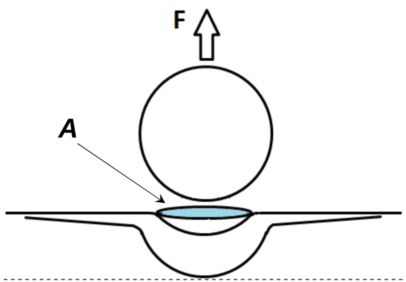
[1]Department of Chemical and Process Engineering, University of Surrey, [2]Institute of Particle Science & Engineering, University of Leeds

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. The ball indentation method is applied to establish powder flow behaviour at low stresses. Indentation measurements are compared to shear cell measurements at the same major principal stress, for a range of moderate stresses, in order to 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.



Ball Indentation

$$H = F_{max}/A \qquad A = \pi\left(2Rh - h^2\right)$$

$$H = C \cdot \sigma_c \qquad h_d = h/R$$

H = hardness
F_{max} = maximum force
A = projected area of impression
R = indenter radius
h = penetration depth
C = constraint factor
σ_c = unconfined yield stress
h_d = dimensionless penetration depth

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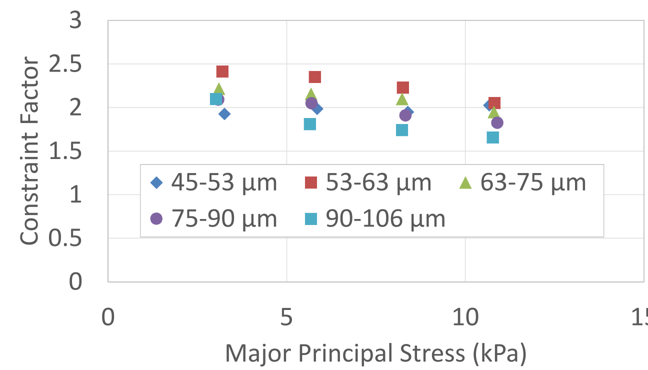
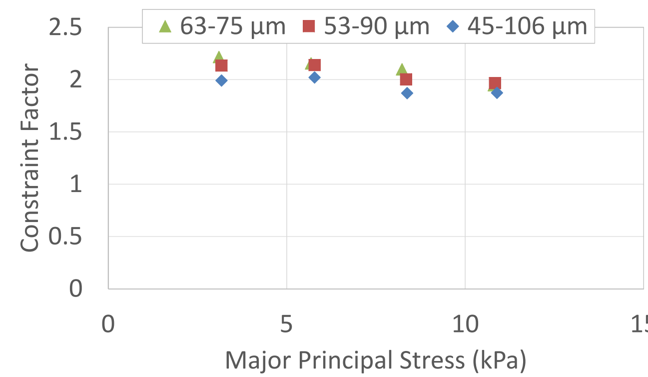
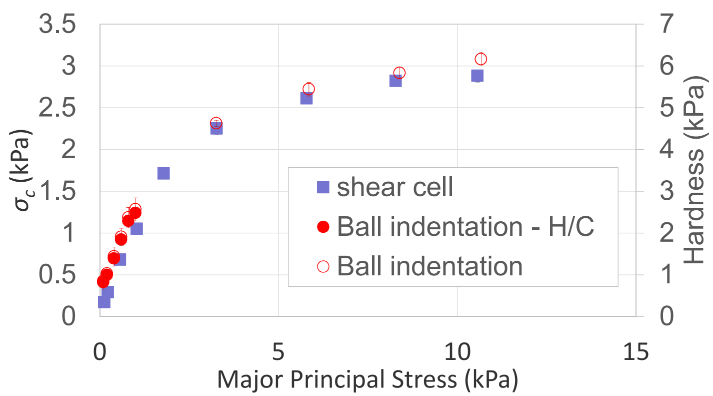
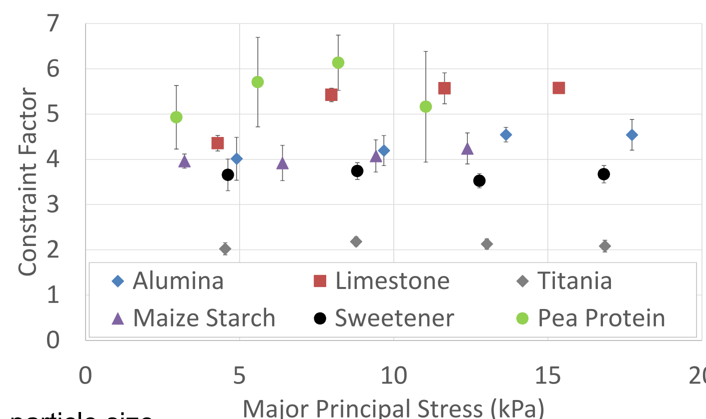
Glass Beads







Other powders



- Constraint factor exhibits a slight reduction with an increase in particle size
- Constraint factor is almost independent of size distribution
- Constraint factor is independent of major principal stress for all materials, except for pea protein and limestone
- Constraint factor ranges from 1.5 – 6 for the tested materials
- For 45-53 µm silanised glass beads the shear cell provides a reproducible yield locus at 0.1 kPa
- The trend of σ_c matches that inferred from indentation, with the latter yielding a larger value

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Powder Structure Control

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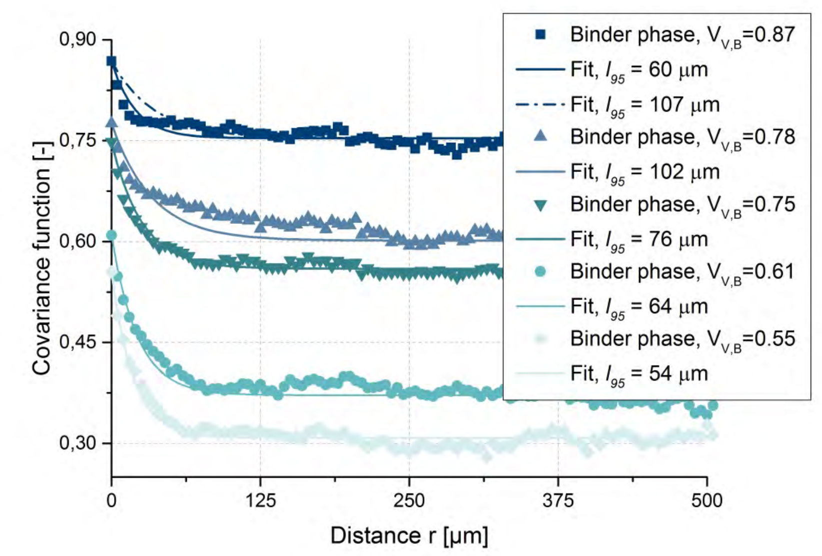
The fundamental approach of the project is based on defining a set of numerical descriptors of powder structure and understanding how they relate to relevant powder properties, e.g. dissolution, dispersing or granule strength. This requires developing an understanding of the mathematical meaning of structure descriptors and their relation to physical meaning, i.e. connectivity, interface prediction, correlation length and voidage. Additionally the best descriptors for a given purpose, e.g. either prediction/modeling of product characteristics or control of structure generation shall be given. The basic components are primary particles, binder or liquid components and intra particle porosity. The quantification of granule structure shall allow setting up process maps and provide input for simulations such as DEM. The descriptors are derived from image analysis procedures, e.g. morphological sieves, point sampling or line scanning techniques.

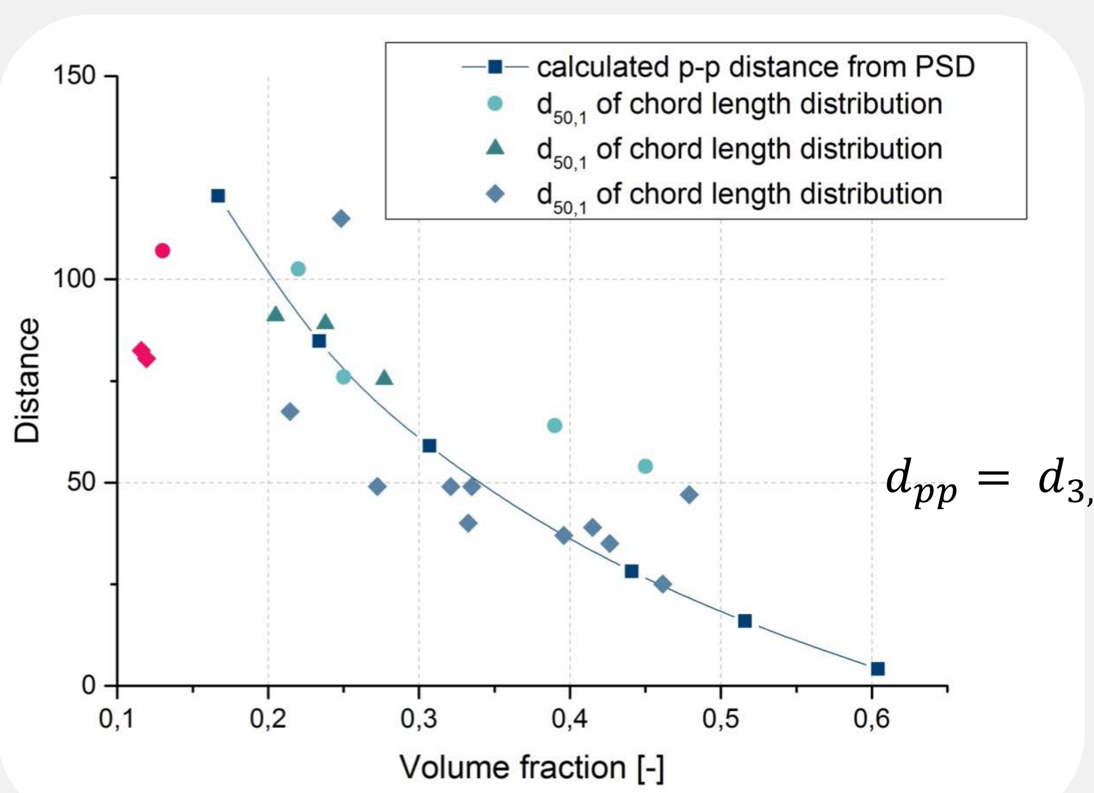
Structure description

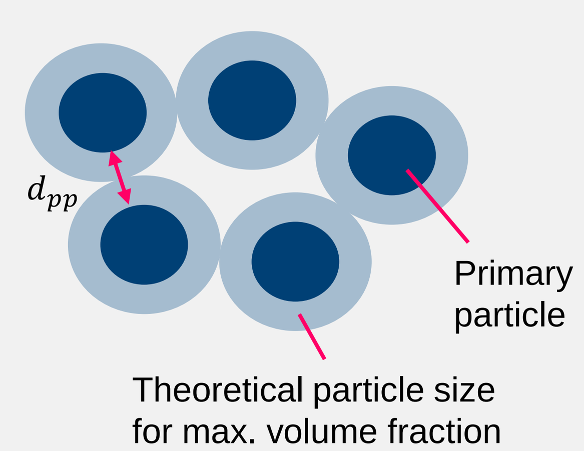
Structure → described by length distribution

- Amount (V_V)
- Size ($d_{4,3}$)
- Distance distribution (CLD)

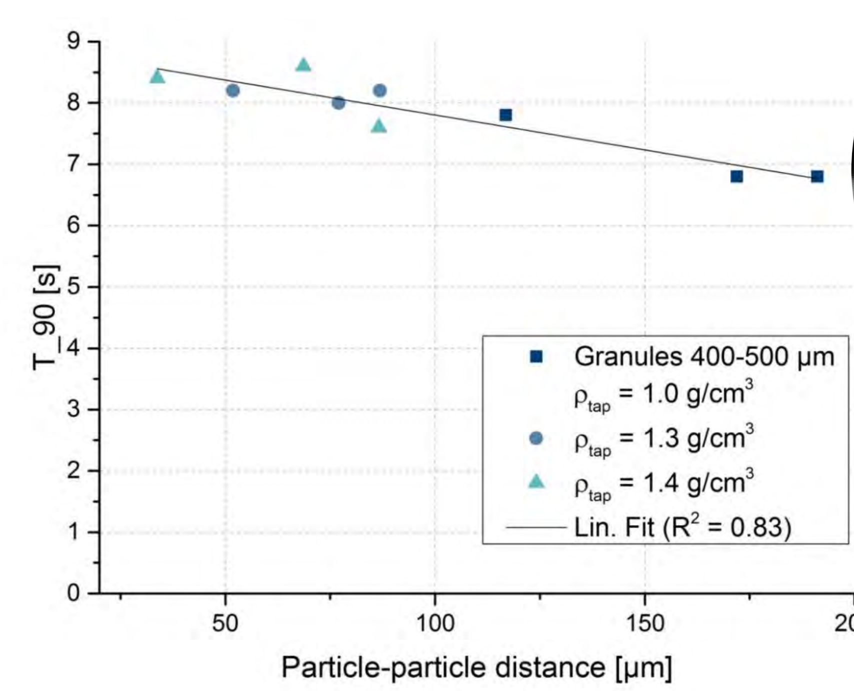


Prediction of structure measures: particle-particle distance

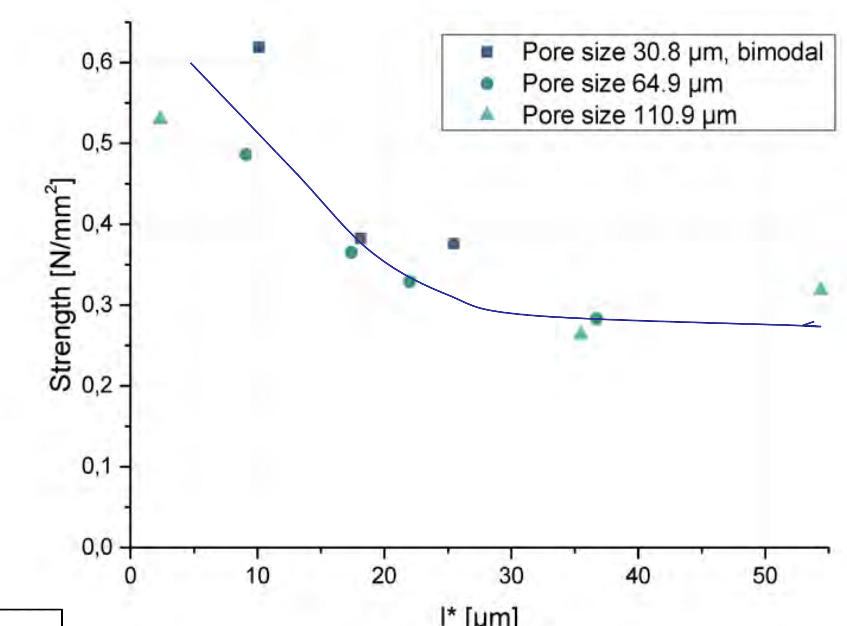




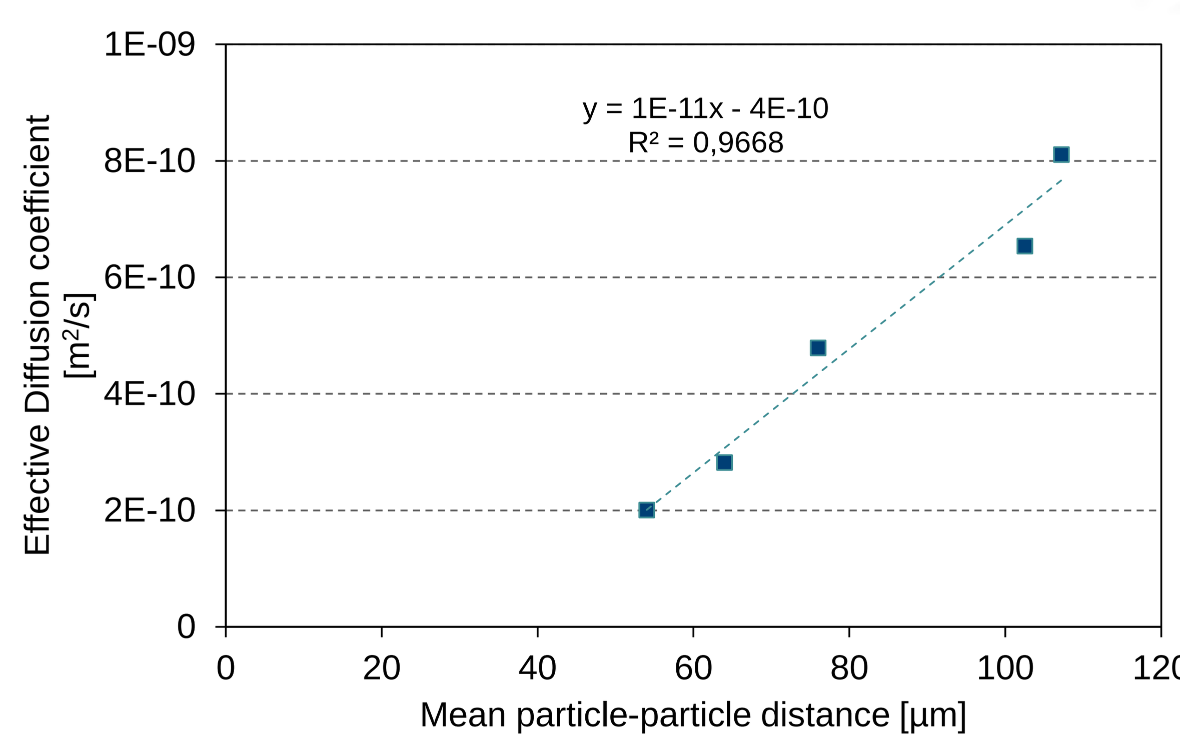
$$d_{pp} = d_{3,0} \cdot \left(\sqrt[3]{\frac{\phi_{max}}{\phi_c \cdot \rho_{tap}/\rho_{solid}}} - 1 \right)$$



Granule strength



Dissolution depends on pore size of dissolving material → Diffusion resistance

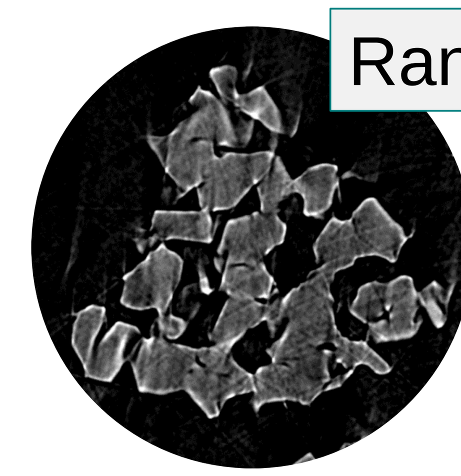


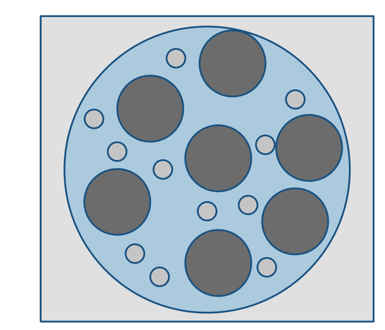
Types of structure

Random

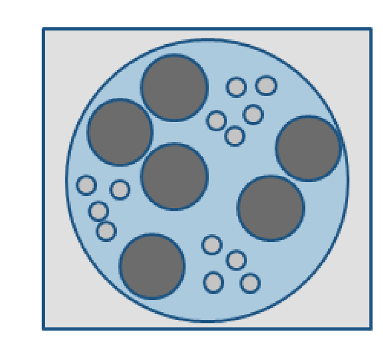
Random close

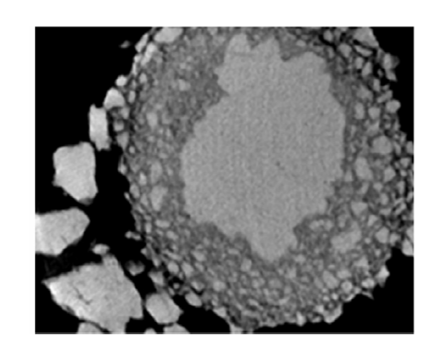
Random loose

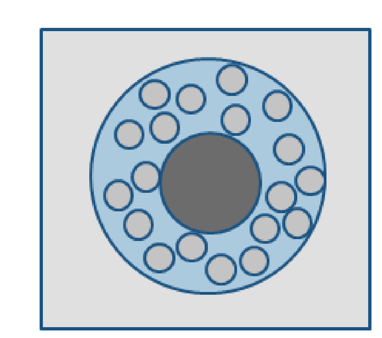




Clustered / Layered







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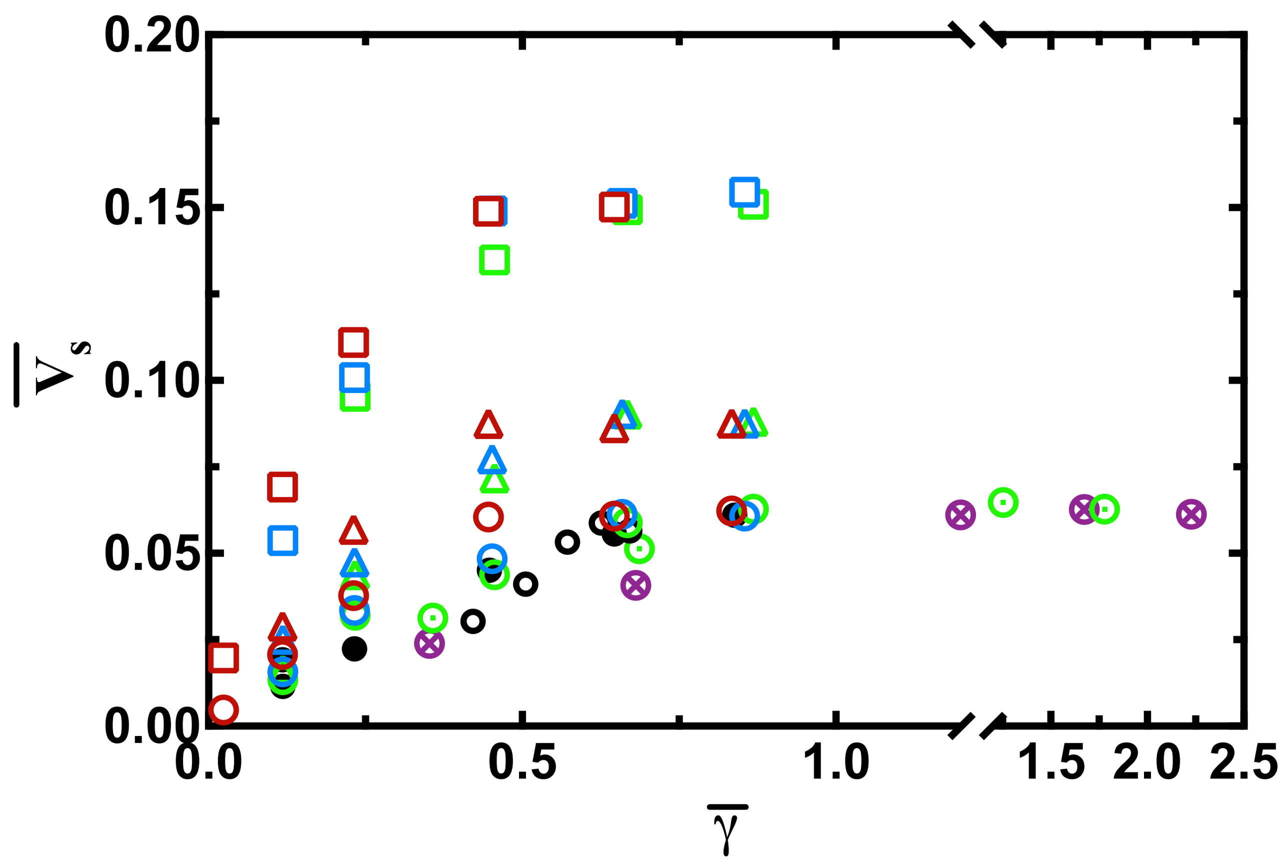
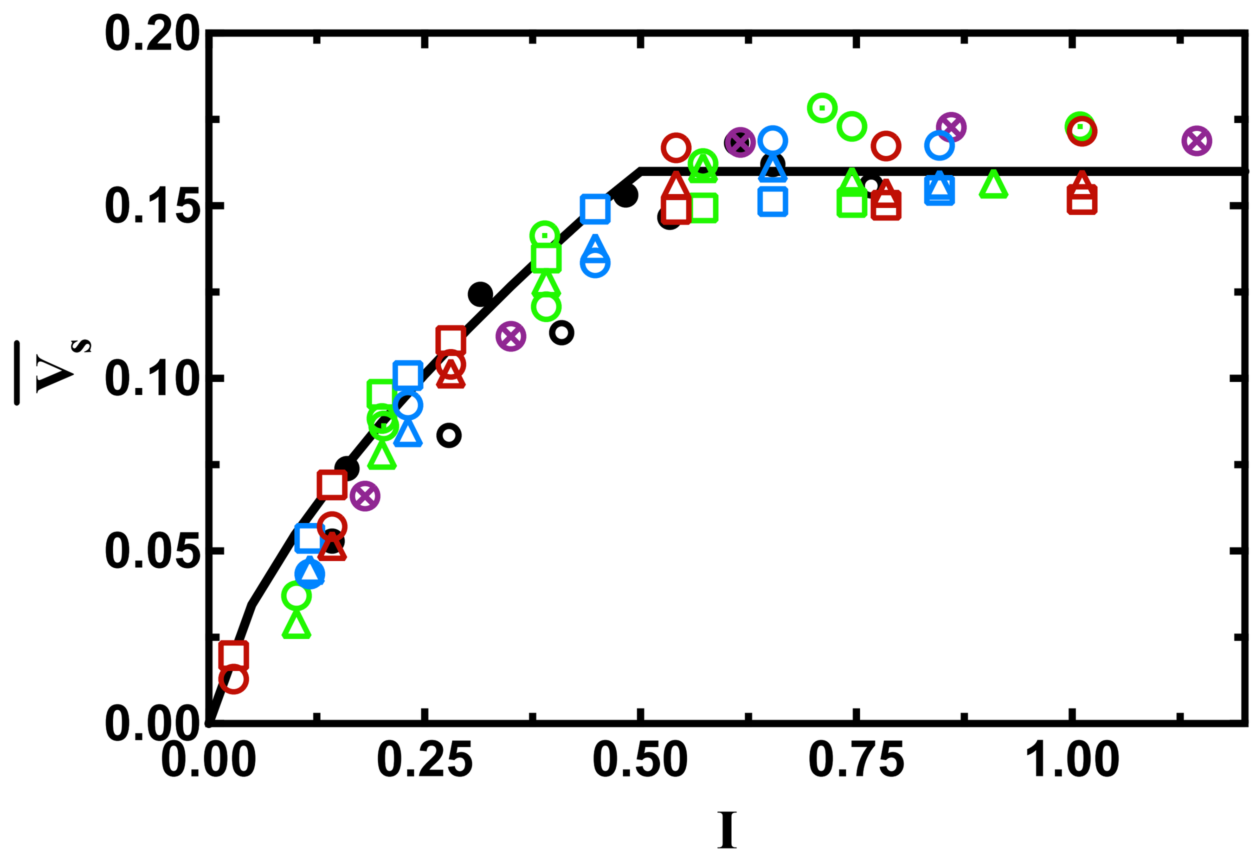
Quantitative Prediction of Segregation at Process Scale

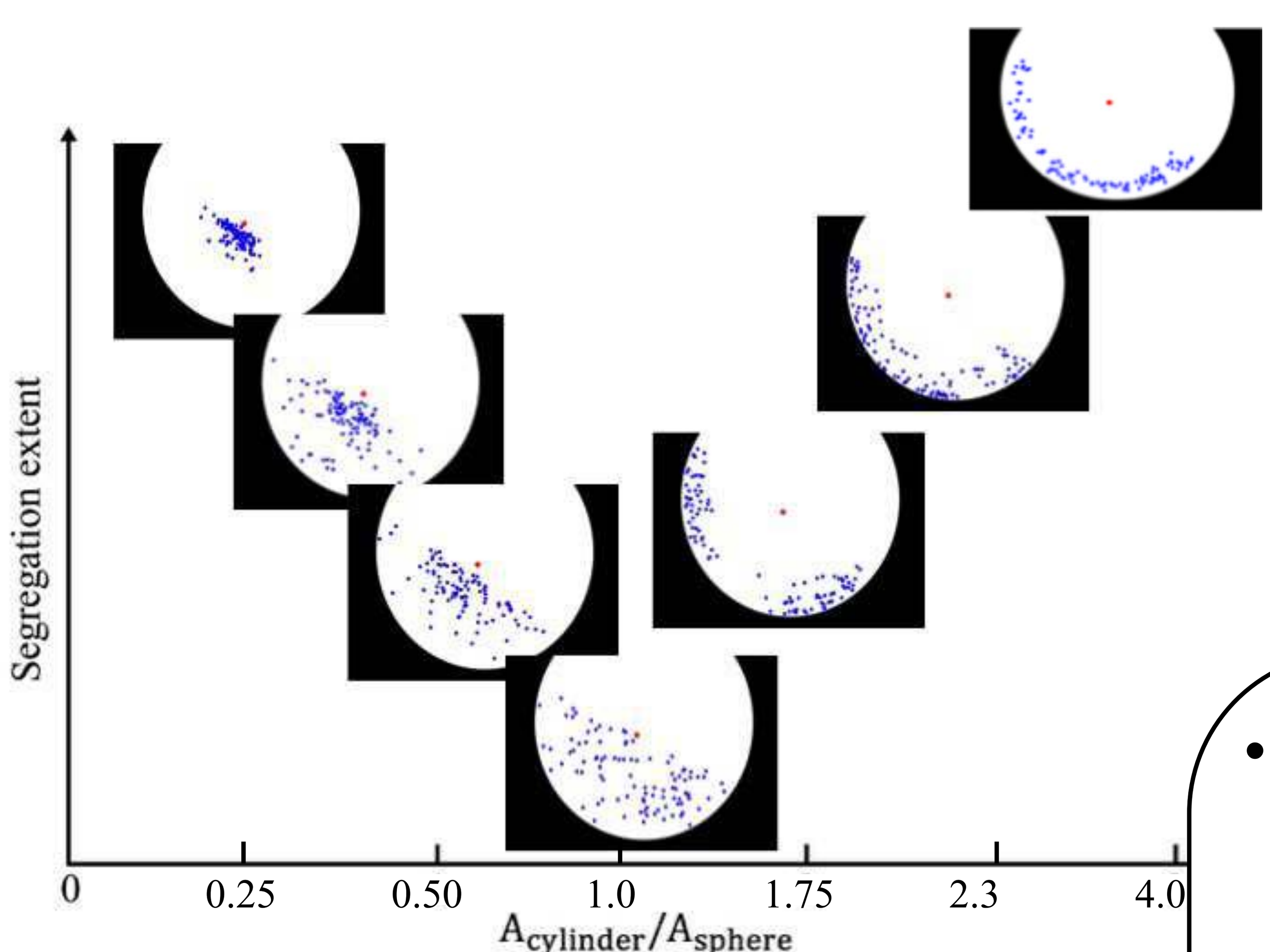
Joseph J. McCarthy
University of Pittsburgh

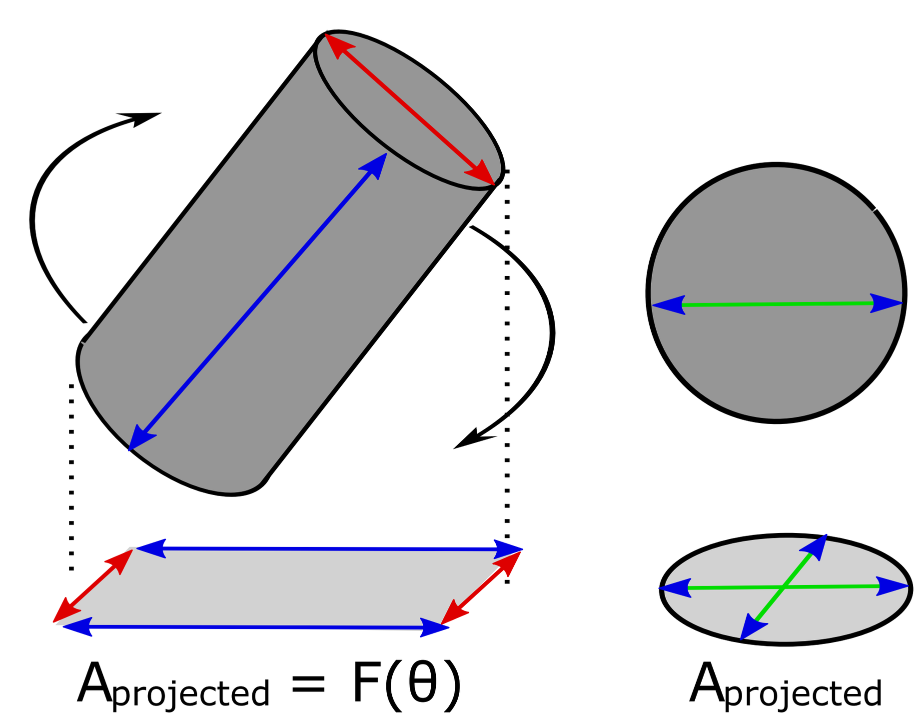
- Particle segregation is a problem for a wide range of industries
- Segregation rate models hold promise for scale-up via continuum-level analysis (via device-specific transport equations), **however**
    - Experimental validation of models is extremely difficult
    - Models often not built with scale-up in mind
- Competing time-scales via flow perturbation simplifies experimental validation by collapsing required measurements onto the “end state”
- Segregation models written expressly in dimensionless form enable more transparent scale-up
- Additionally, connection between segregation and rheology allows further advances through analogy





- Naïve scaling fails to capture density segregation results
- Novel rheology-inspired model collapses data for different densities, particle sizes, boundary conditions, gravity, shear rates, etc.





- Shape segregation experiments show that the “time/angle-averaged” projected area is a predictor of segregation tendency

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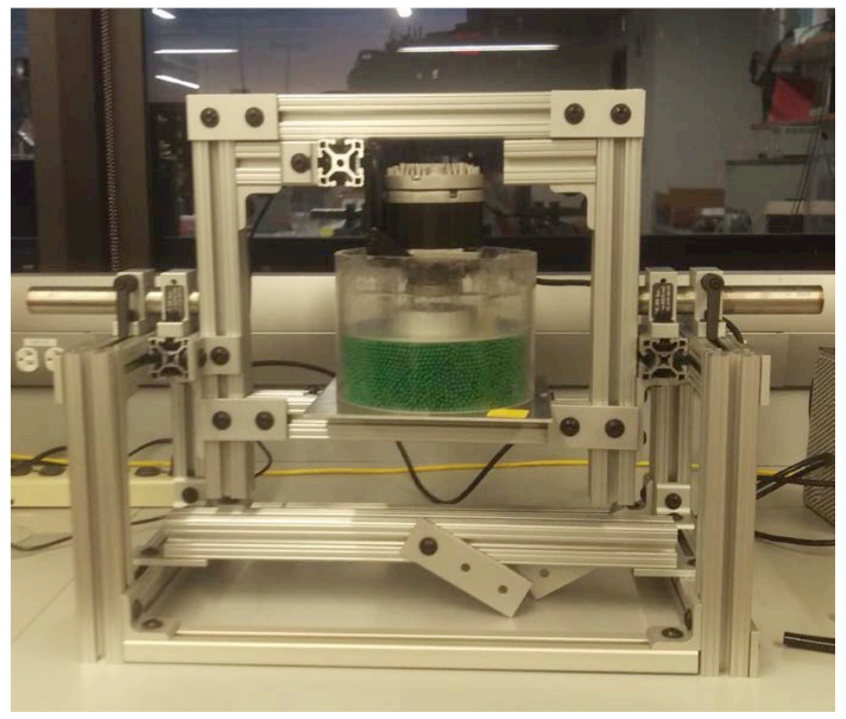
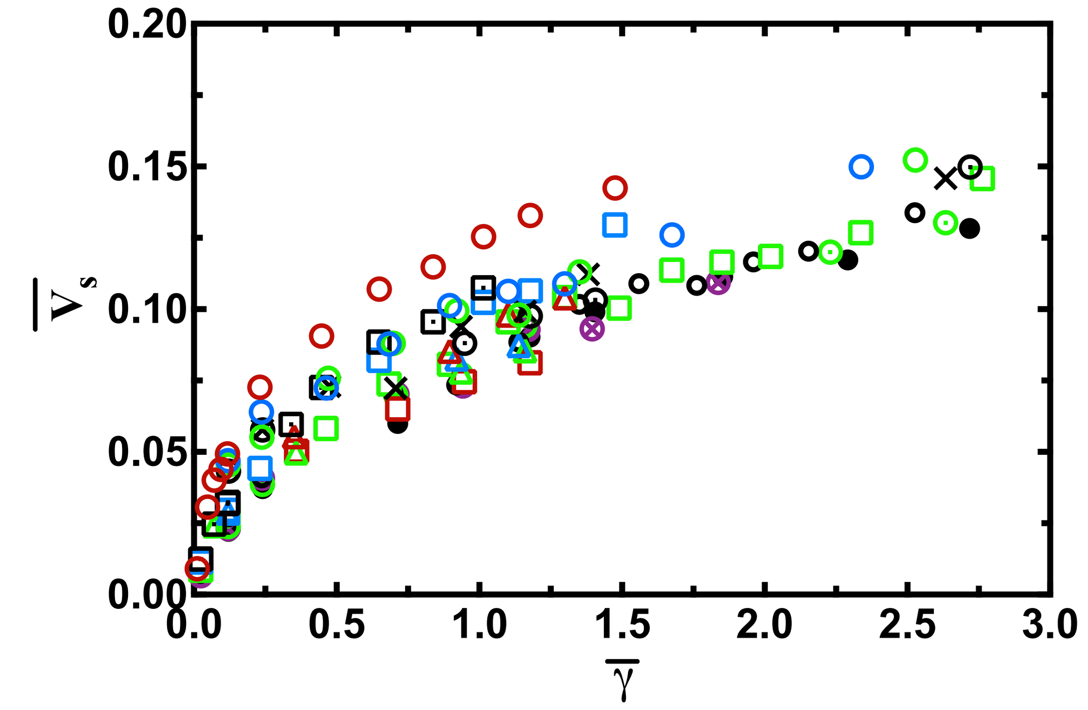
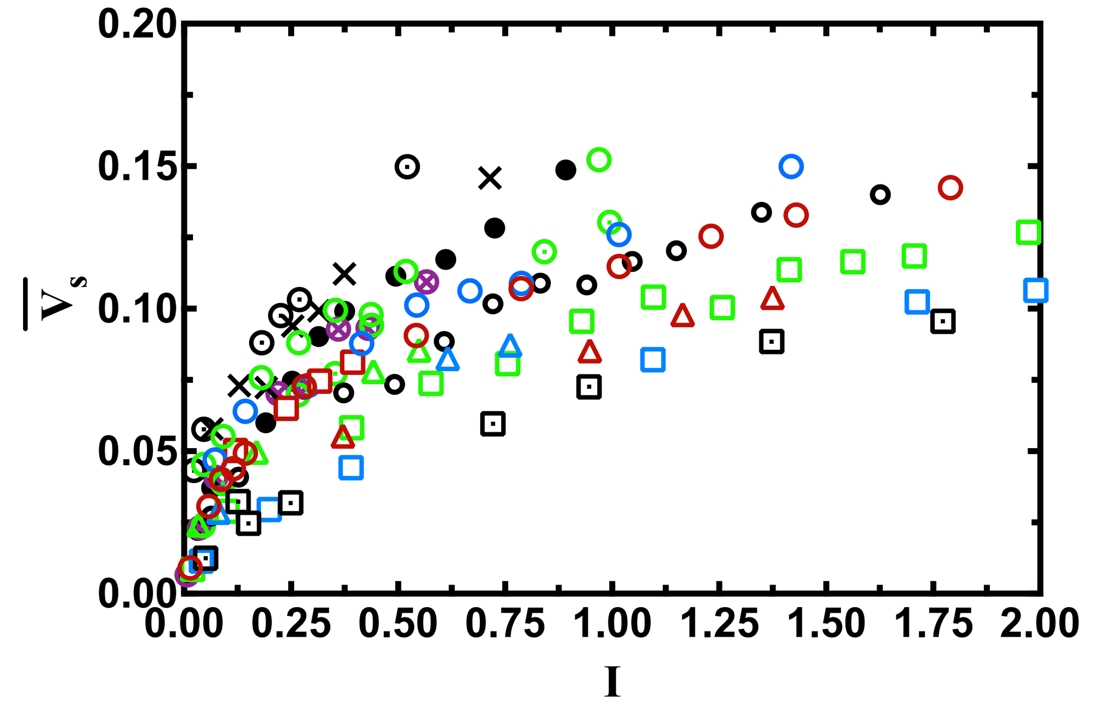
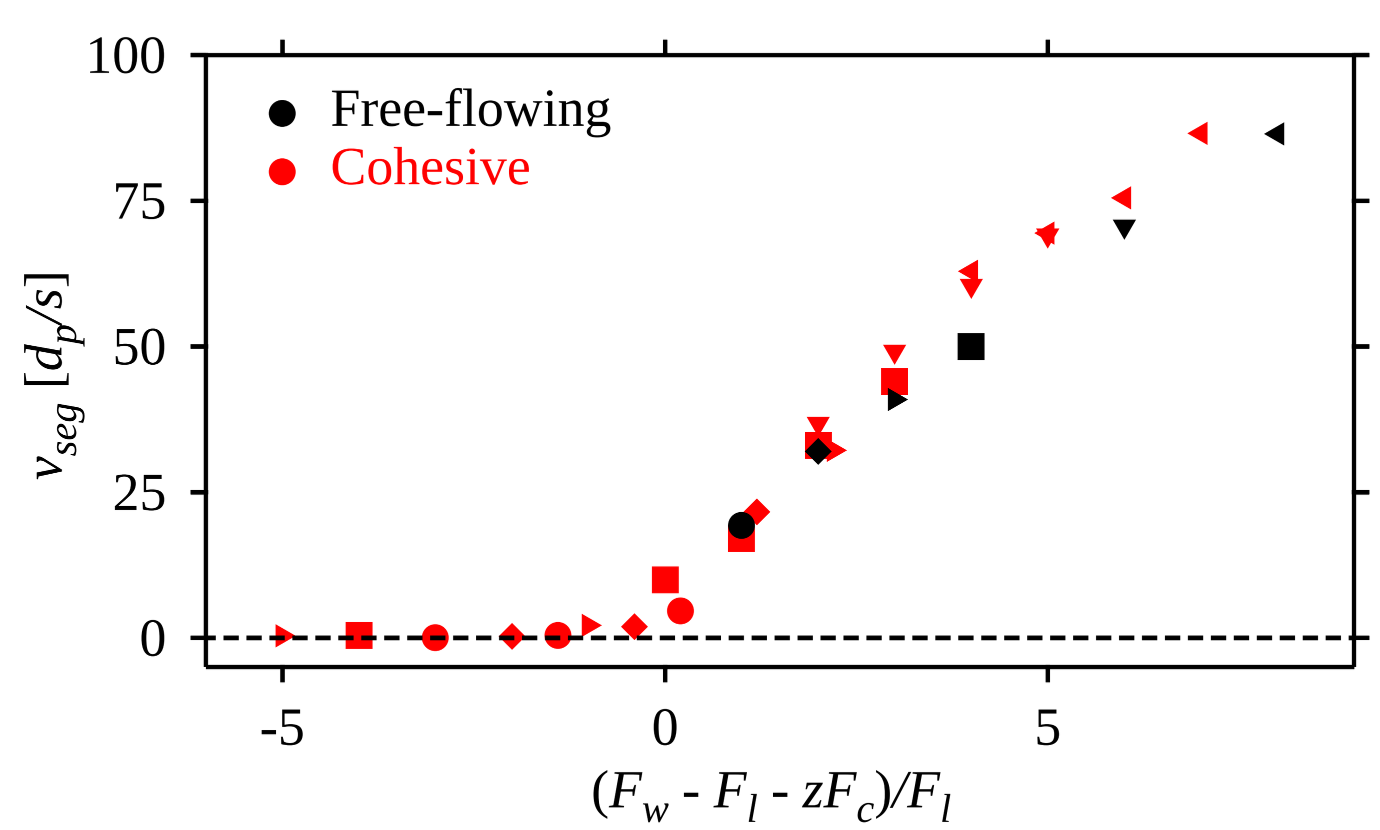


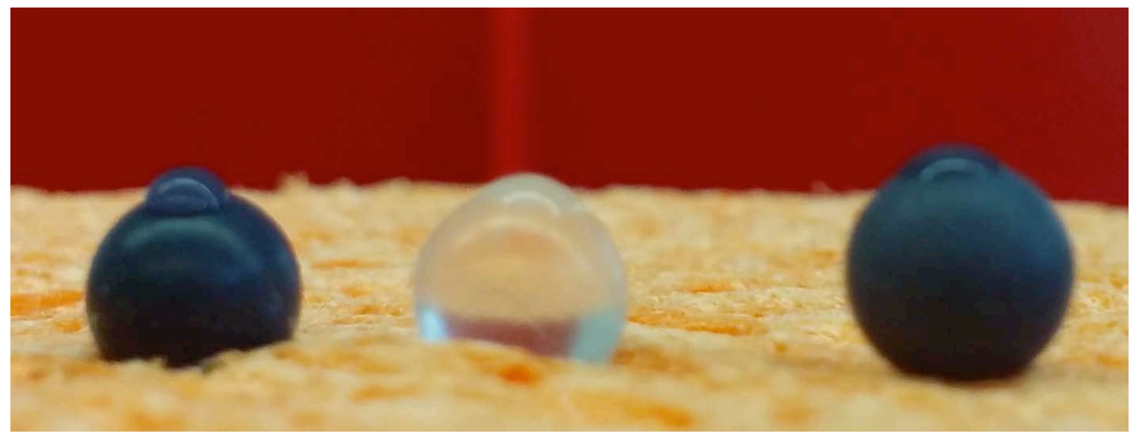
- New experiment will allow segregation studies in “perturbation mode” or simple shear
- Size segregation is more complex than density (but progress is close!)





- Simple cohesive model predicts that density segregation is “predictably” impacted by cohesion
- New surface treatment will allow carefully controlled experiments



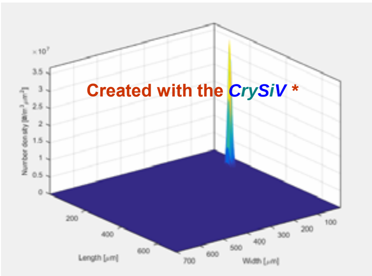


A Holistic Approach for the Model-based Control of Crystal Size, Shape and Purity in Integrated Batch and Continuous Crystallization - Wet Milling Systems

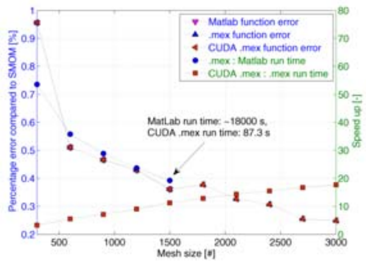
Botond Szilagyi, Kanjakha Pal, Zoltan K. Nagy
Purdue University, Davidson School, of Chemical Engineering, West Lafayette, US

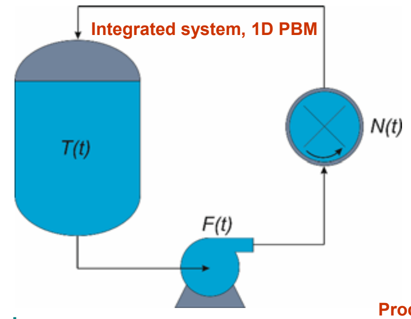
1. Integrated crystallizer-wet mill models for batch and continuous operation with 1D and 2D PBM, solved with and without GPU acceleration
2. FBRM and PVM soft sensors applying backward and forward transformation for 1D and 2D crystals
3. 2D PBM development for the system configurations with and without impurity model
4. Model Discrimination Framework and Iterative Model-based Experimental Design (IMED)
5. Development of the estimation and 1D and 2D PBM and MOO based NMPC and validation via simulation for the batch and the continuous systems

2D CSD evolution



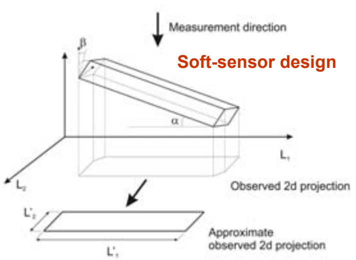
Implementation efficiency – the CrySiV function*





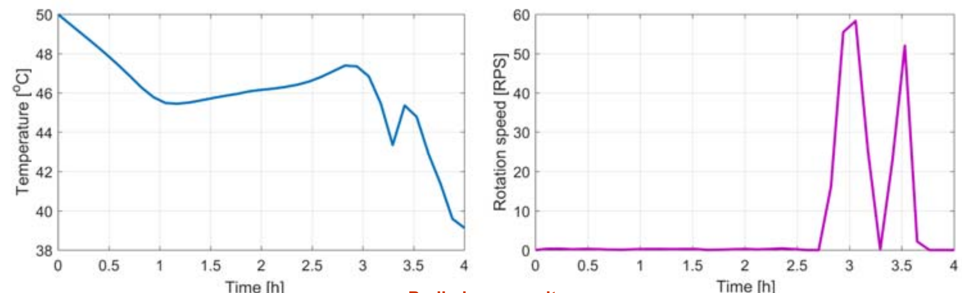
Numerical details

Accuracy	CFL	Mesh size	Compute platform
High	CFL < 0.3	N > 1000	CPU + GPU
Moderate	CFL > 0.3 CFL < 0.5	N > 500 N < 1000	CPU + GPU; CPU only
Low	CFL > 0.5	N < 500	CPU only

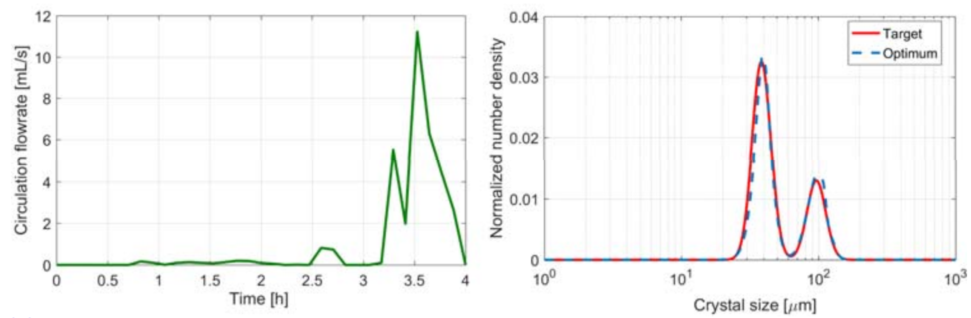


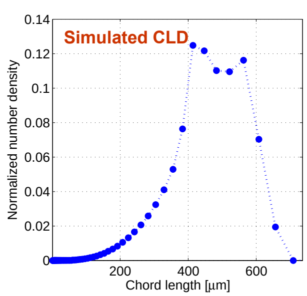
- Soft sensor: based on random projection of pre-determined crystals (crystal dimensions used in the FVM solution)
- Mapping ***all*** possible chord lengths and aspect ratios of ***all*** possible projections (***off-line calculation***)
- ***Weighted summing*** of individual crystal's CLDs and ARDs to generate the CLD and ARD of the crystals population (***on-line calculation***)

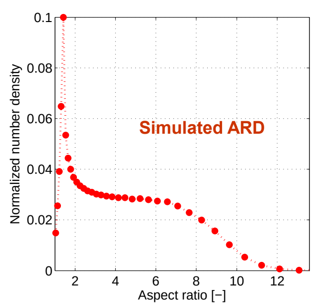
Process optimization

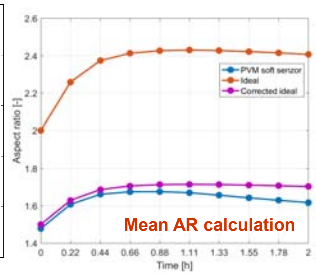


Preliminary results









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* crysiv.github.io

CrySiV 1.2
simulating crystallization easier

Milling and material grindability: modelling, measurement and mill fingerprinting

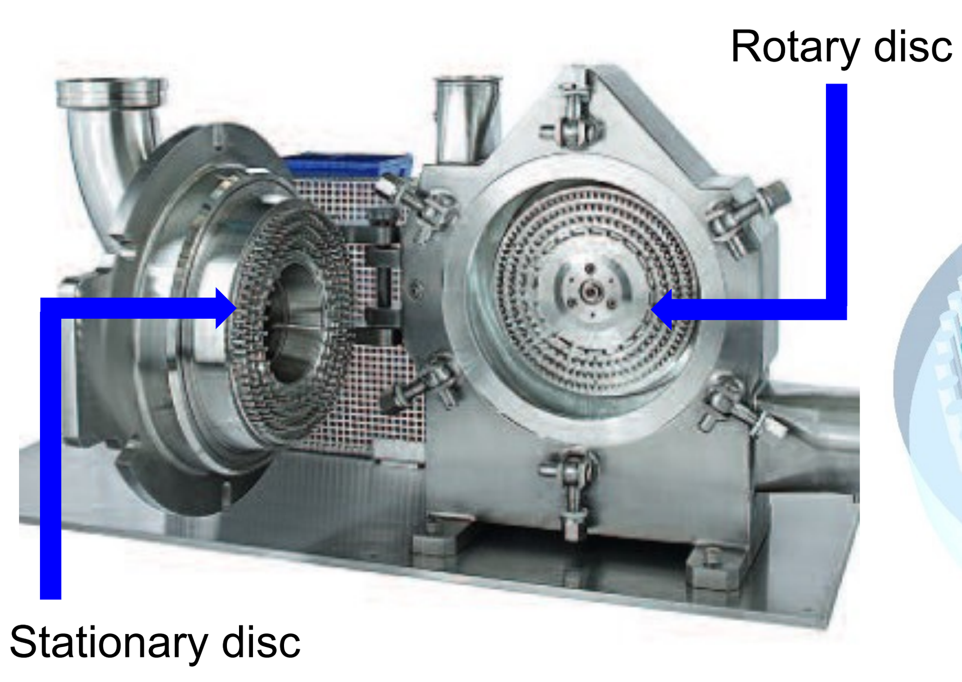
J.Y. Ooi, X.Z. Chen, L.G. Wang, J.F. Chen*, J. Sun
University of Edinburgh, Scotland, UK ; *Queen's University Belfast, UK

Milling is a common industrial operation for particle size reduction that is highly inefficient. The goal of this project is to develop a generic methodology to link material grindability with particle dynamics in a mill, involving:

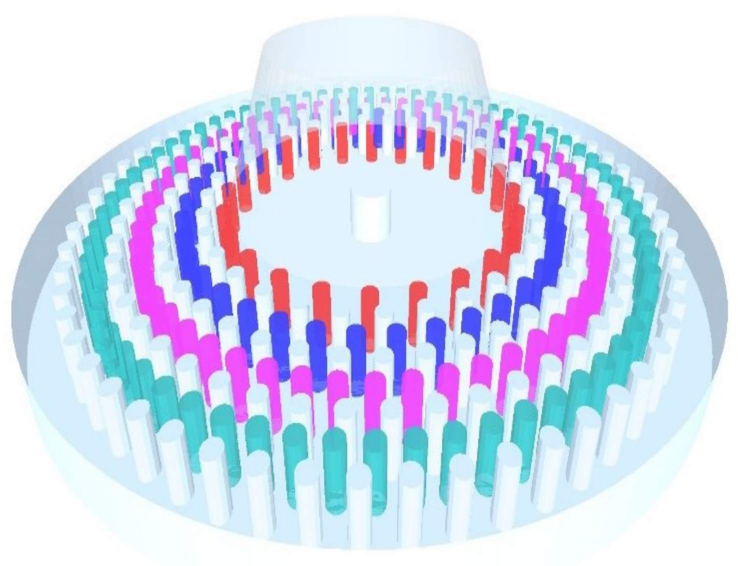
- Computational modelling and experiments to characterise the milling function
- Developing grindability measures to characterise the comminution behaviour of particulates
- Hierarchical verification and validation leading to robust evaluation of milling performance

Impact pin mill UPZ100

- Impact milling tests conducted at four feed rates and rotary speeds

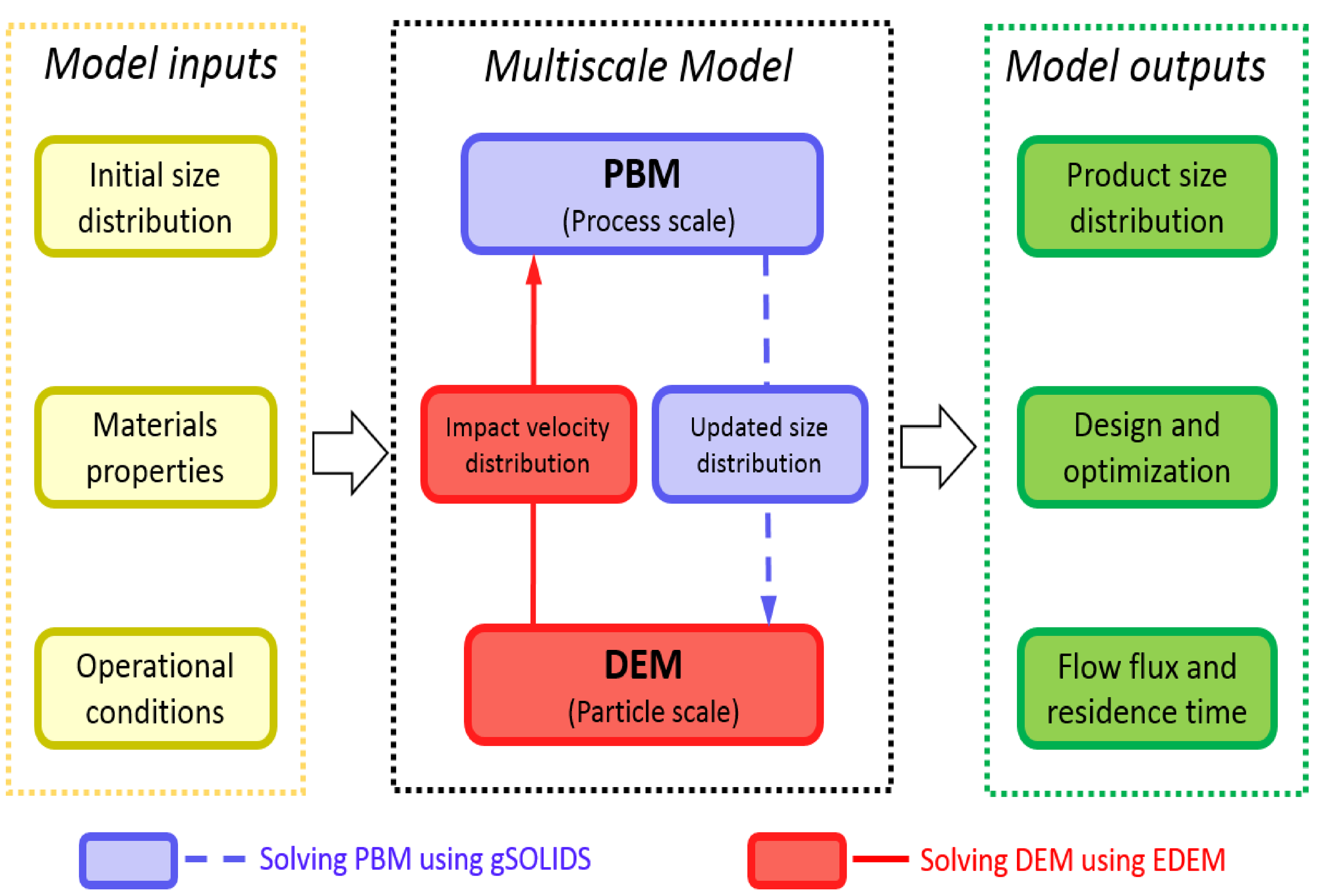


Impact pin mill-UPZ100
Courtesy from Hosokawa Ltd .UK



Real-geometry DEM simulation layout (rotary discs coloured)

DEM-PBM coupling framework



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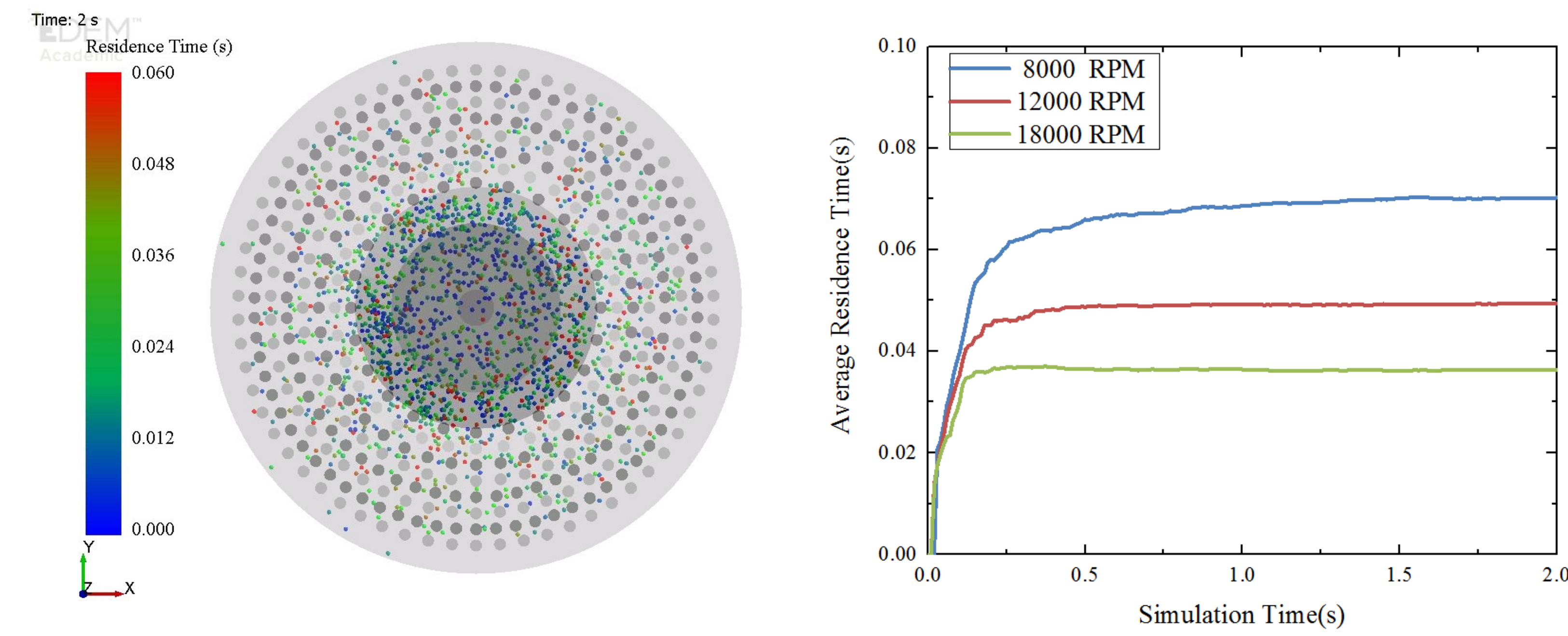
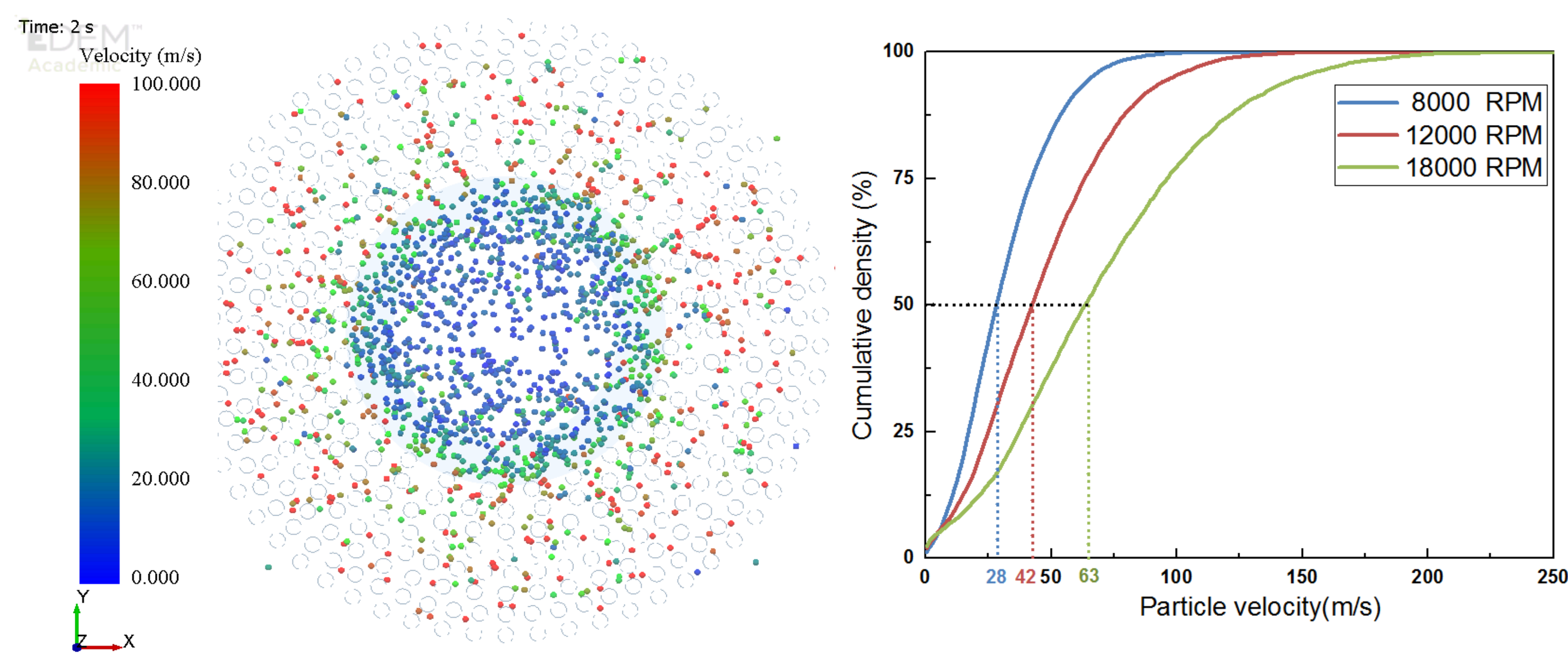
Particle dynamics from DEM simulation

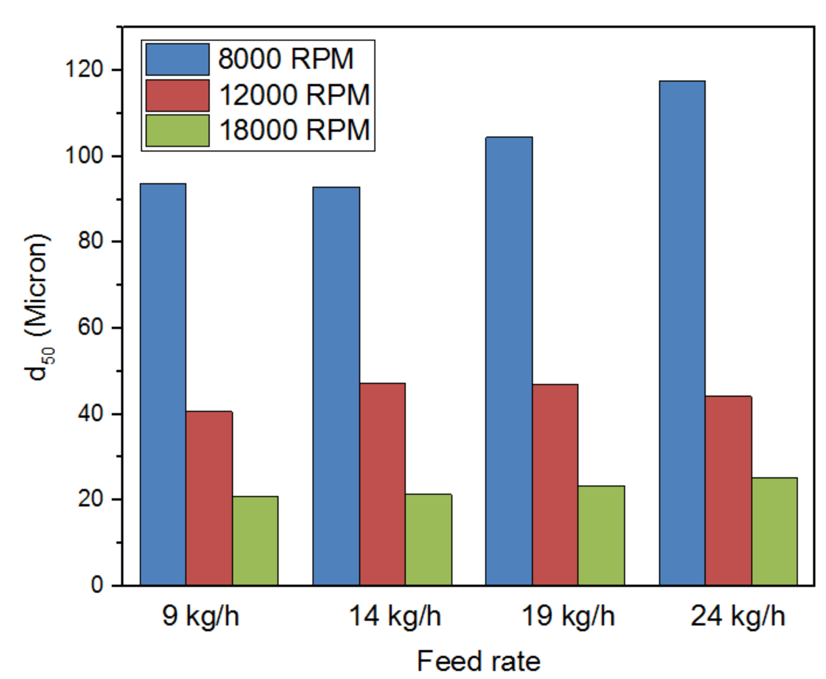


- Average particle residence time decreases with increasing rotary speed

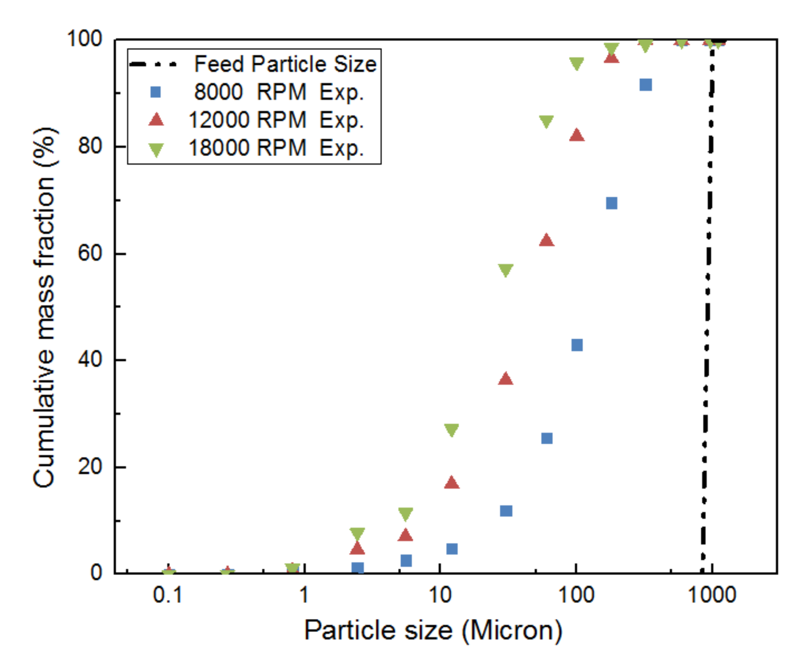


- Average particle velocity increases with increasing rotary speed

Impact milling tests



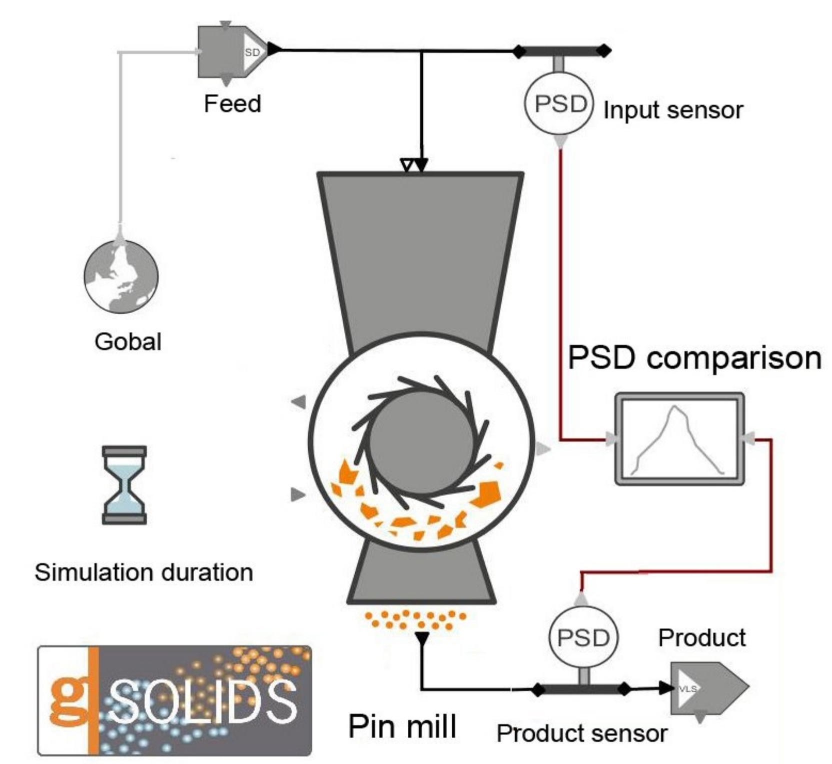
Median size d_{50} of milled alumina under varying rotary speed and feed rate



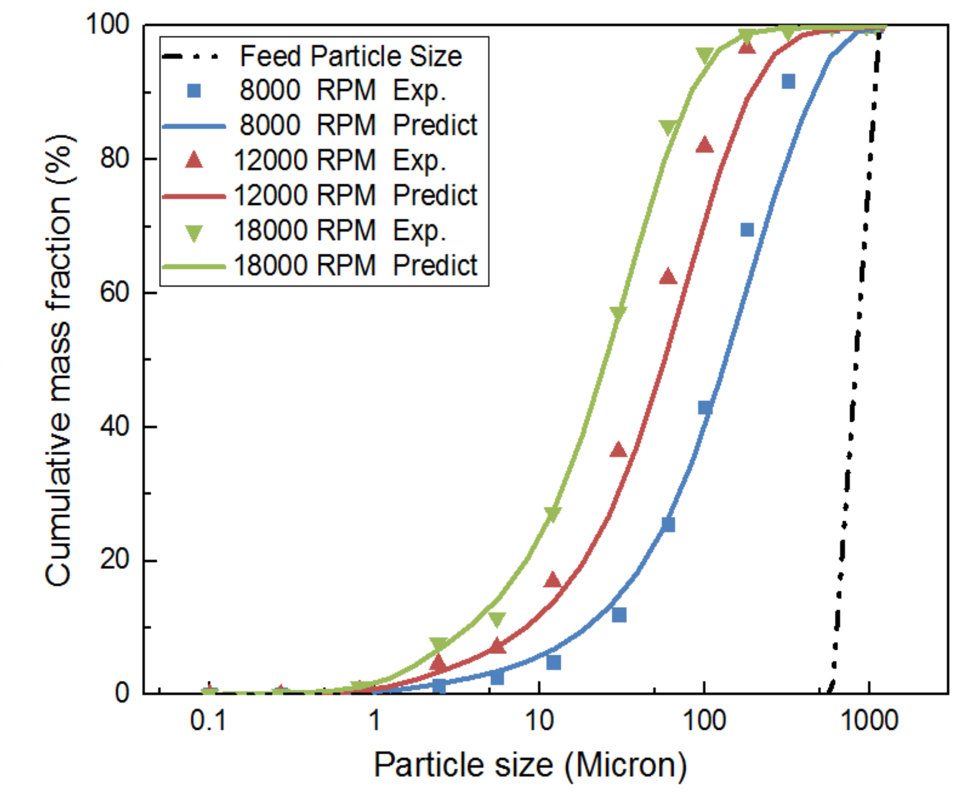
PSD of milled alumina under varying rotary speed at 24 kg/h

PBM to predict PSD in milling

- Material dependent parameters are estimated from 12000 RPM and then used to predict other cases
- Coupled with the impact velocity extracted from DEM, gSOLIDS shows good agreement with experimental tests



PBM simulation software: gSOLIDS



Predictive PSD of 8000 and 18000 RPM based on 12000 RPM parameter fitting

Key observations

- Multiscale framework of DEM-PBM coupling to predict PSD in the pin mill has been established
- DEM provides the particle dynamics and stress events from particle scale
- PBM coupled with DEM provide further insights into the predictive capacity of milling

Next steps

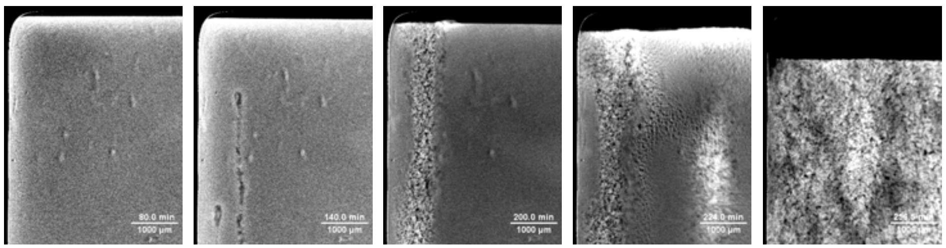
- Utilise DEM to obtain material dependent parameters in PBM functions.
- Extend DEM-PBM model with data exchange (two-way coupling)
- Potential to link with the multiscale particle modelling project funded by the National Formulation Centre (UK).

Gravity and Hydrodynamics join forces to destroy colloidal gels

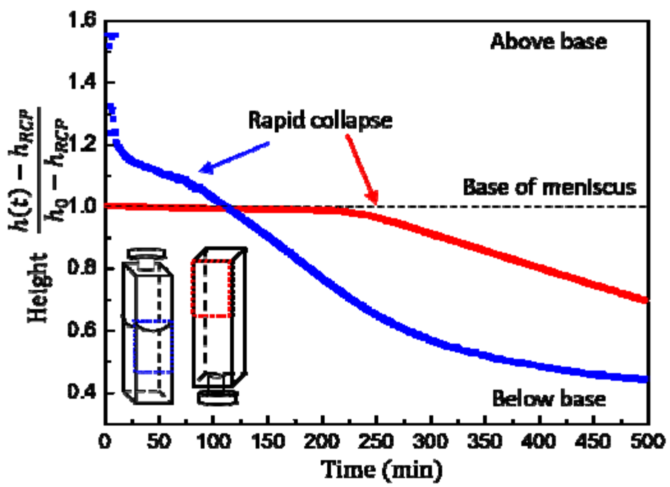
Xuemao Zhou, Joost de Graaf, Michiel Hermes, and Wilson Poon
School of Physics & Astronomy, The University of Edinburgh, United Kingdom

Our project is aimed at fundamental understanding of the long-term stability of colloidal gels. We previously identified the meniscus as a region of importance to gel stability, since a dense layer of colloids forms there, which falls through the gel and triggers rapid collapse [1]. This year, we have experimentally demonstrated that by eliminating the meniscus – thereby removing the dense layer – the collapse can be postponed considerably. Rapid collapse still occurs, showing that debris falling through the gel is only one possible collapse-triggering process. Turning to simulations, we have used the lattice-Boltzmann (LB) method to study the role of fluid flow. We find a strong dependence of the onset of collapse on the gravitational Péclet number, Pe_g. This reveals hydrodynamic backflow as another process leading to rapid collapse. However, we can only simulate a small part of the experimental system. We therefore plan to develop a phase-field theory inspired by our experiments and simulations that is able to describe the entire system.

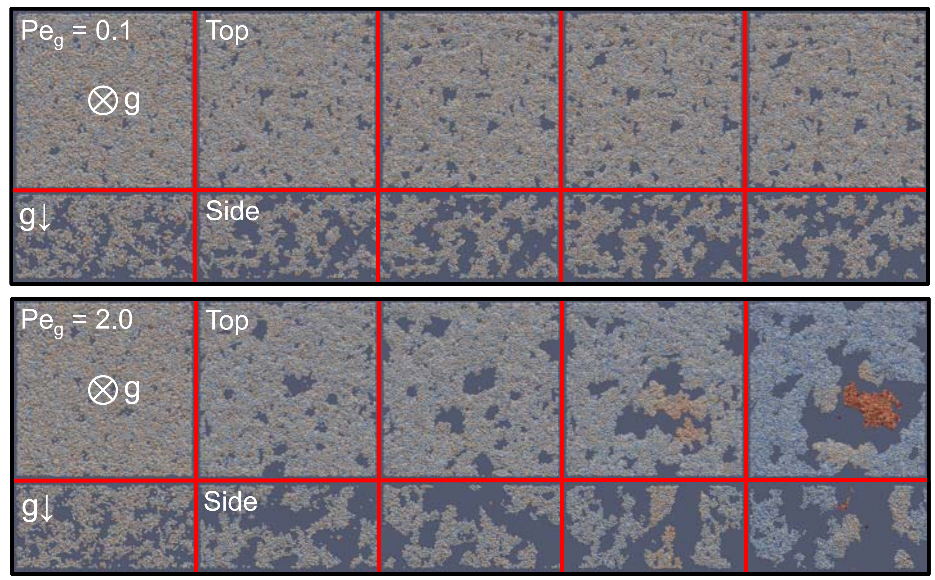
Rapid collapse can be postponed by removing the meniscus.



(above) Side view of the collapse in a system without a meniscus, the 4th snapshot marks the onset of rapid collapse, due to droplets rising through the gel, destroying it. (below) Position of the gel top with respect to the base of the meniscus for a partially filled (blue) and totally filled system (red, see above).



LB simulations evidence the importance of Pe_g on the collapse of the gel. The greater Pe_g, the faster the fluid backflow shears the gel to pieces.



A bulk piece of colloidal gel, $\phi = 0.1$, subjected to gravity, with low and high Pe_g. The time increases from left to right in steps of $50t_B$ (where t_B is the single-particle Brownian time). Red particles move up, blue ones down. The width of the domain is 1/20th of the experimental snapshots shown on the left.

$$\mathrm{Pe}_g = \frac{4\pi}{3} g \Delta\rho k_B T a^4$$

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[1] R. Harich et al., Gravitational collapse of depletion-induced colloidal gels, *Soft Matter* **12** (2016) 4300-4308

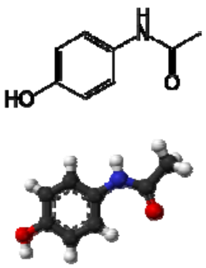
Self-Assembled Monolayers as Nucleating Surfaces to Study Early Formation Pathways of Crystal Polymorphs

Zihao Zhang, Katherine P. Barteau, Lara A. Estroff, & Uli Wiesner
Cornell University

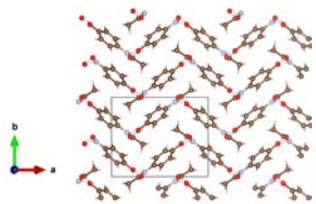
The understanding and control of crystallographic polymorphism and crystal habit of organic compounds is technologically important to a number of industries. Our goal is to use self-assembled monolayers (SAMs) as well-defined surfaces to study the relationship between surface chemistry, the nucleation event, and the final polymorph selection.

Investigated Drug: Acetaminophen (ACM)

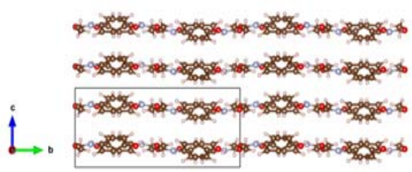
- 2 common polymorphs – orthorhombic easier to mill: (002) cleavage planes



Monoclinic (Form I)



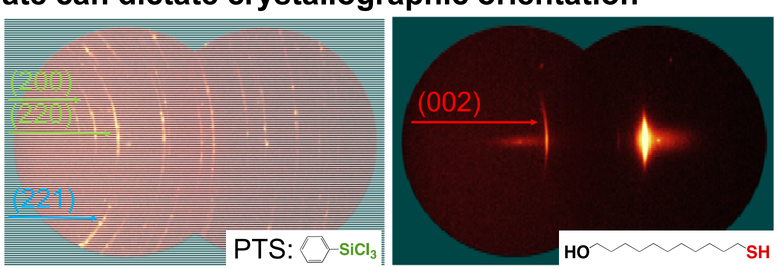
Orthorhombic (Form II)



Both solvent and substrate work together to control polymorph

Substrate / Solvent	$-CH_3$ on gold (hydrophobic)			-OH on gold (hydrophilic)		
	N	Form I	Form II	N	Form I	Form II
DI water	**9**	**93%**	7%	**17**	**100%**	0
DI water/ dioxane 20:80	**11**	9%	**91%**	**10**	0	**100%**
1,4-dioxane	**11**	**100%**	0	**10**	20%	**80%**
Ethanol	**20**	**80%**	20%	**11**	9%	**91%**

Substrate can dictate crystallographic orientation



Form II is crystallized on both hydrophobic and hydrophilic surfaces from the same solvent system, but the observed orientation of the crystal is vastly different.

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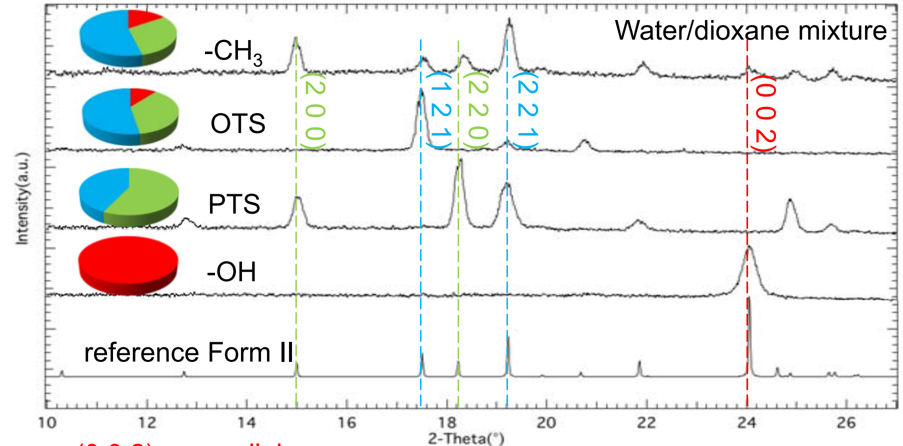
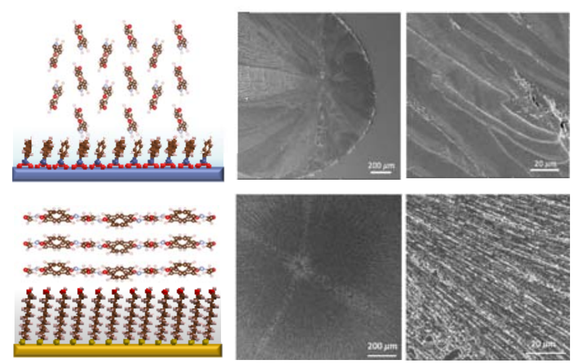
Substrate can dictate crystallographic orientation





Hydrophobic (PTS) surfaces: Form II
- (002) cleavage planes **perpendicular**
- Crystallites **large and smooth**.

Hydrophilic (-OH) surfaces: Form II
- (002) cleavage planes **parallel**
- Crystallites **small and rough**.

Results Summary

- Both solvent and SAMs surface chemistry act in concert to control polymorph selection
- The SAM surface chemistry further influences the polymorph crystal orientation
- Future directions: in-situ x-ray scattering of blade-coated solutions to observe early nucleation and growth processes

Die Filling of Aerated Powders

Chuan-Yu (Charley) Wu and Joesry F. El Hebieshy
Department of Chemical and Process Engineering, University of Surrey, Guildford, GU2 7XH, UK. C.Y.Wu@surrey.ac.uk

The primary goal of this renewal project is to understand the die filling behaviour of aerated powder blends. We will harvest the achievements on the current IFPRI project and our previous research outcome in this area to tackle this challenging problem. The objectives of this renewal project are
1) to explore fine powder mixtures during die filling processes encountered in real manufacturing process;
2) to identify the critical material attributes and critical process during die filling of aerated powders; and
3) to develop a design space for fine powder mixtures to achieve controlled/specified properties during die filling (such as mass variation, content uniformity, mass flow rate).

- A rotary die filling system (Fig.1) was constructed and used for exploring die filling behavior of a wide range of materials. The die filling behaviour was also compared to that of linear die filling. It was found that the filling efficiency is significant improved with rotary die filling (Fig.2).

- In addition to size induced die filling reported in the last AGM, density-induced segregation in the shoe and in the die was explore. It is shown that significant segregation can take place during die filling.

- In previous project, it was noted that there was a dramatic air pressure change in the die during die filling. Further mechanistic analysis was performed that the air pressure change in the die can be well predicted using a transient die filling model (Figure 3)

- The dependency of die filling performance on powder flowabiloty was also explored. It was found that there is a strong correlation between die filling performance and the flow index measure using Flodex (Figure 4).

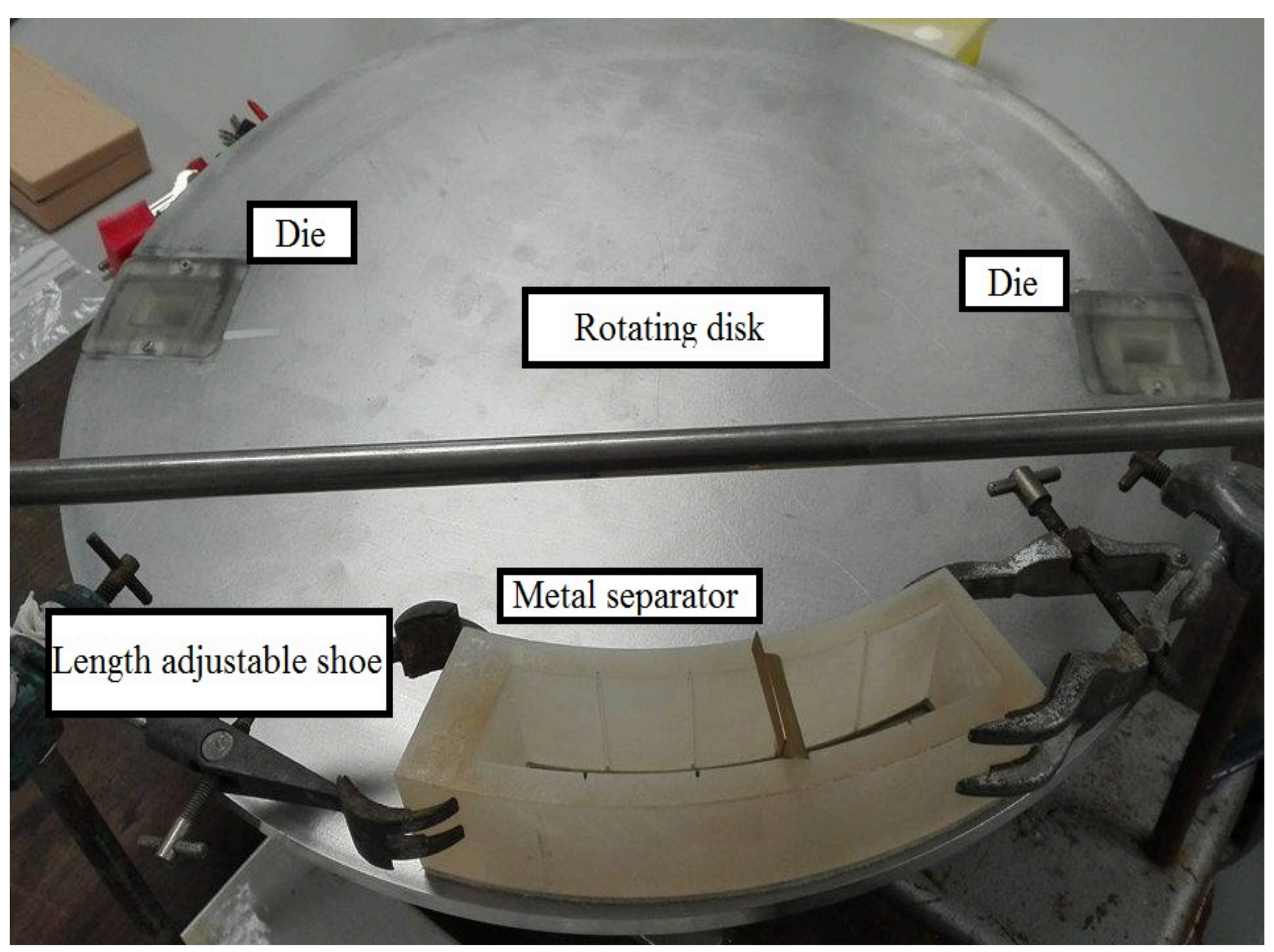


Figure 1 The rotary die filling with filling speed of 0-1500 mm/s.

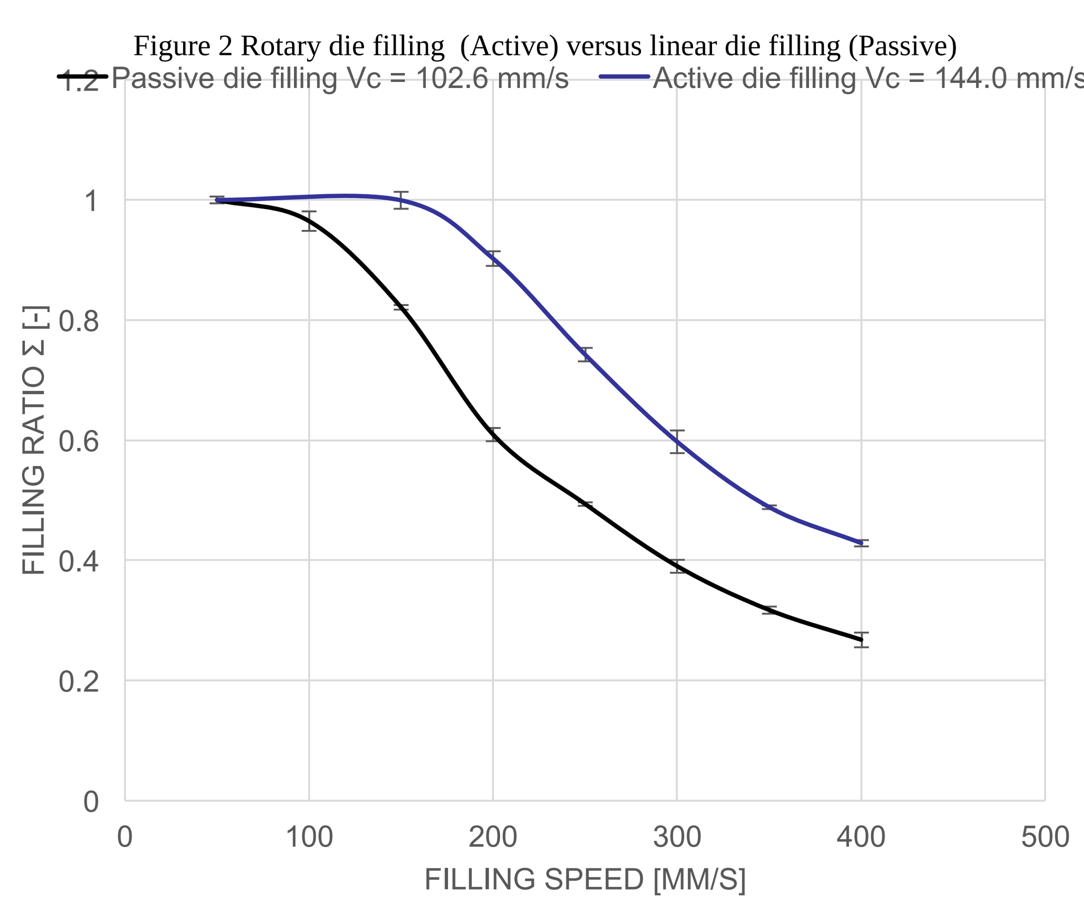


Figure 2 Rotary die filling (Active) versus linear die filling (Passive)

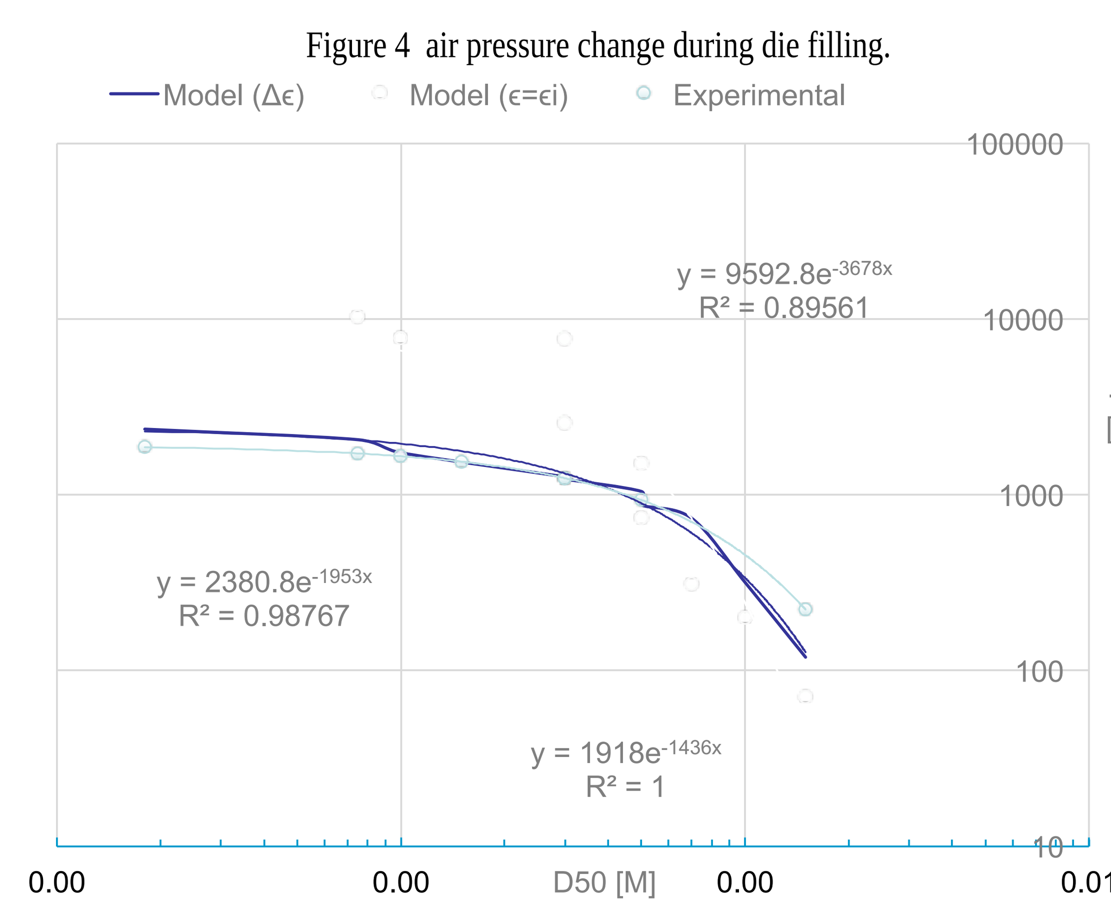


Figure 3 air pressure change during die filling.

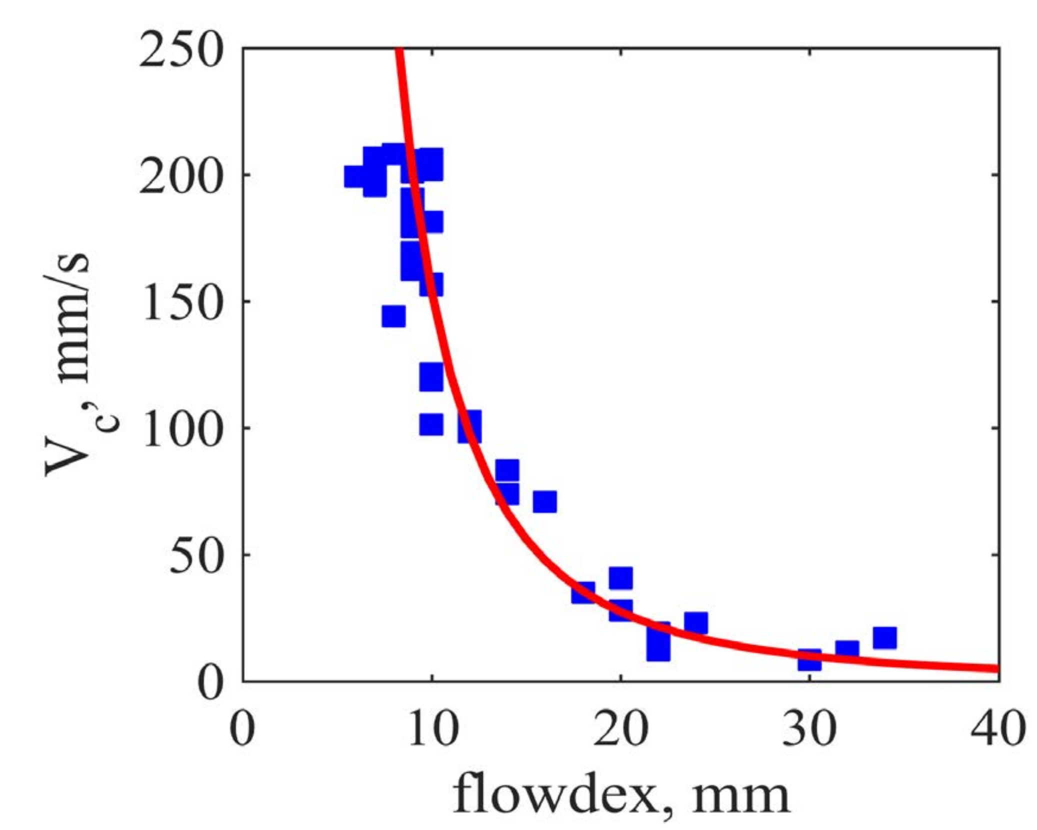


Figure 4 Critical filling speed versus flow index.

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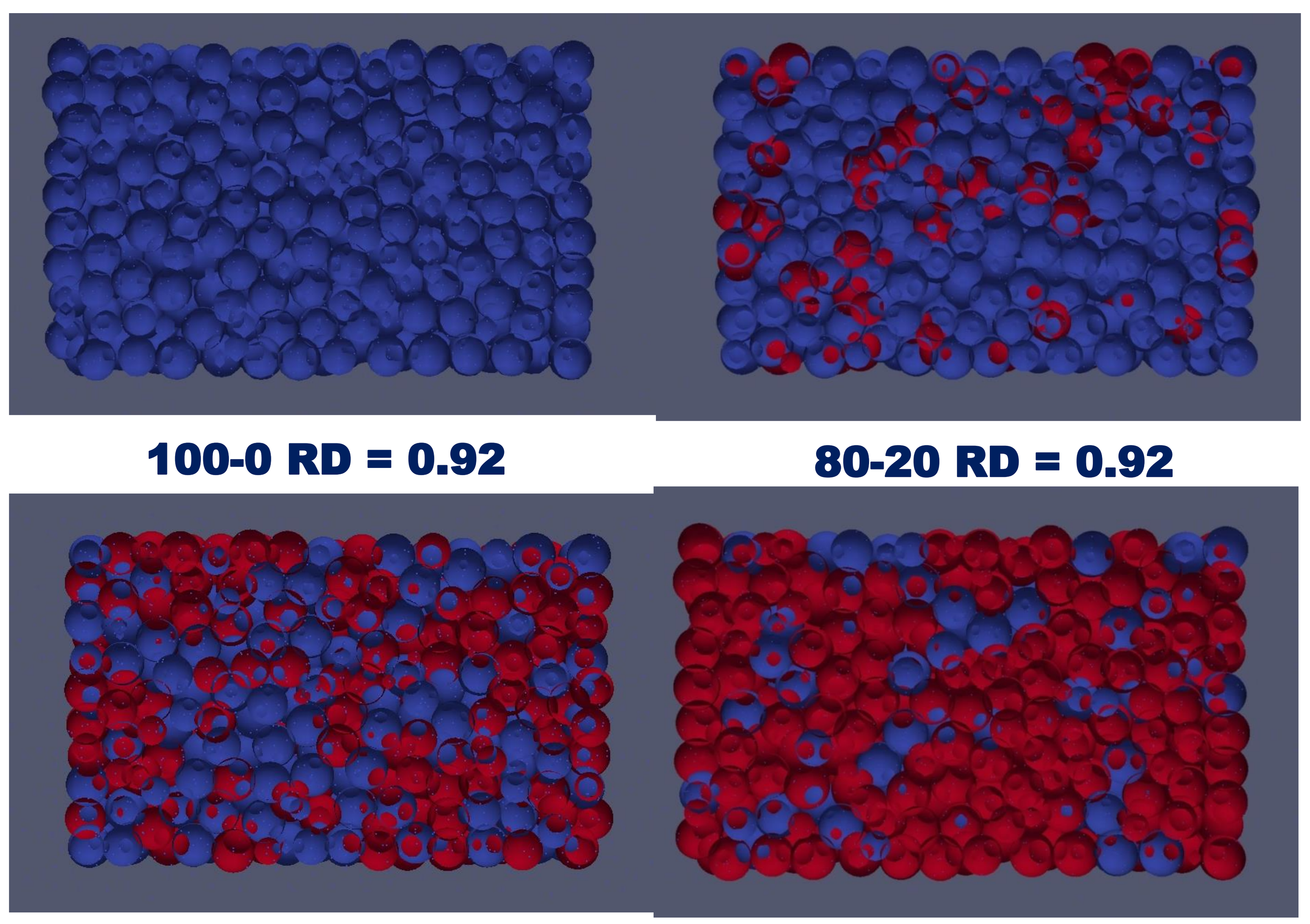
MULTI-SCALE MODELING OF COMPACTION WITH EMPHASIS ON POWDER MIXTURES

Antonios Zavaliangos
Department of Materials Science and Engineering, Drexel University, Philadelphia, PA19104, USA

OBJECTIVES
- Understanding the rules that determine the compaction and properties of multicomponent formulations

APPROACH
- Develop a discrete element framework for the analysis of compaction and mechanical properties of multicomponent mixtures
- Identify geometric and material parameters that control the behavior of such mixtures
- Validate the analysis

Discrete Element Model for High Density



Extraction of material parameters from data

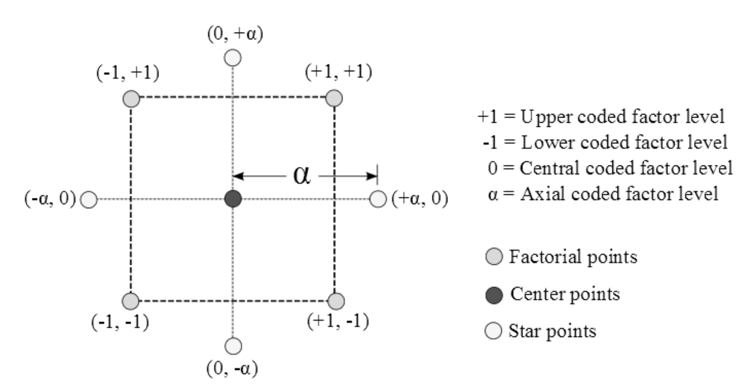
1) x_m, *m=1.. M* model parameters that needs to be estimated,

2) $y_n(x_m)$, *n=1..N* model response variables of interest

3) $y_n^*, n = 1..N$ experimental observations to match.

Approximation of the numerical model with a response surface Using a composite central design methodology

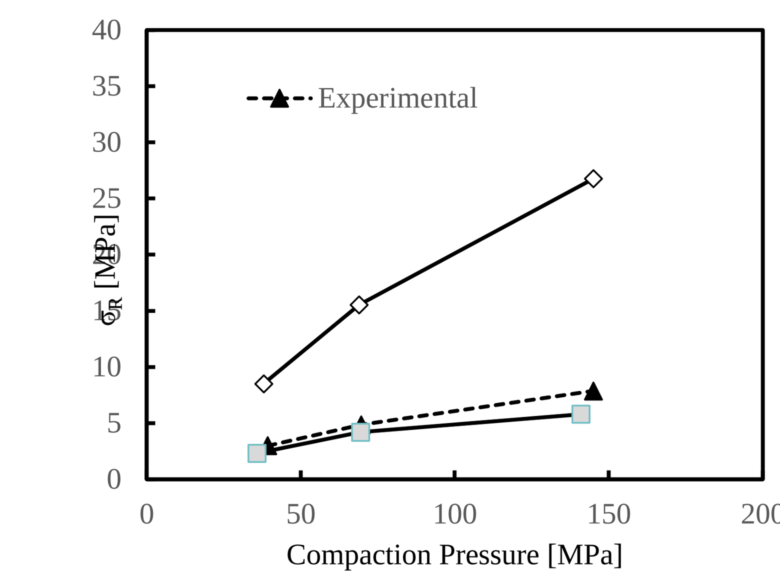
$$y_n = \beta_0 + \sum_{i=1}^{m}\beta_i x_i + \sum_{i=1}^{m}\beta_i^2 x_i^2 + \sum_{i<j}\sum \beta_{ij} x_i x_j$$

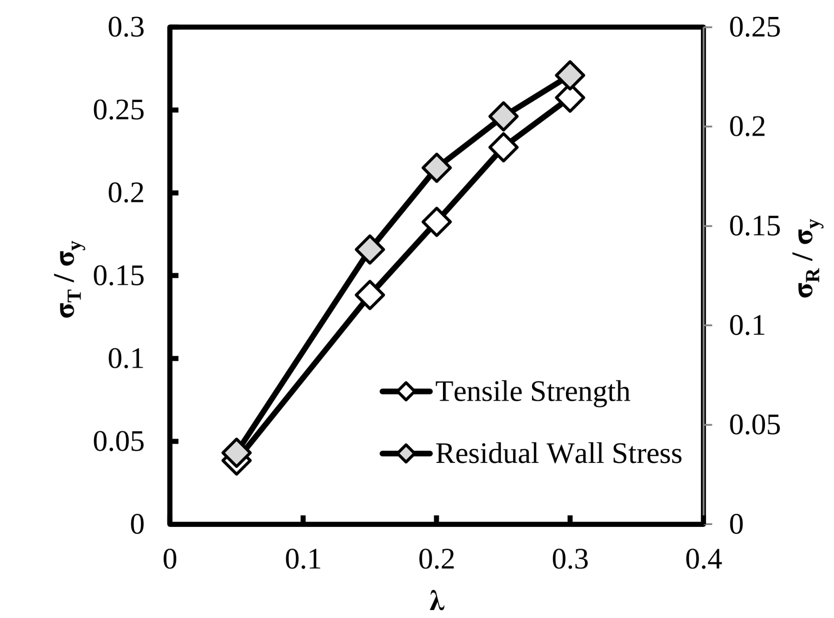


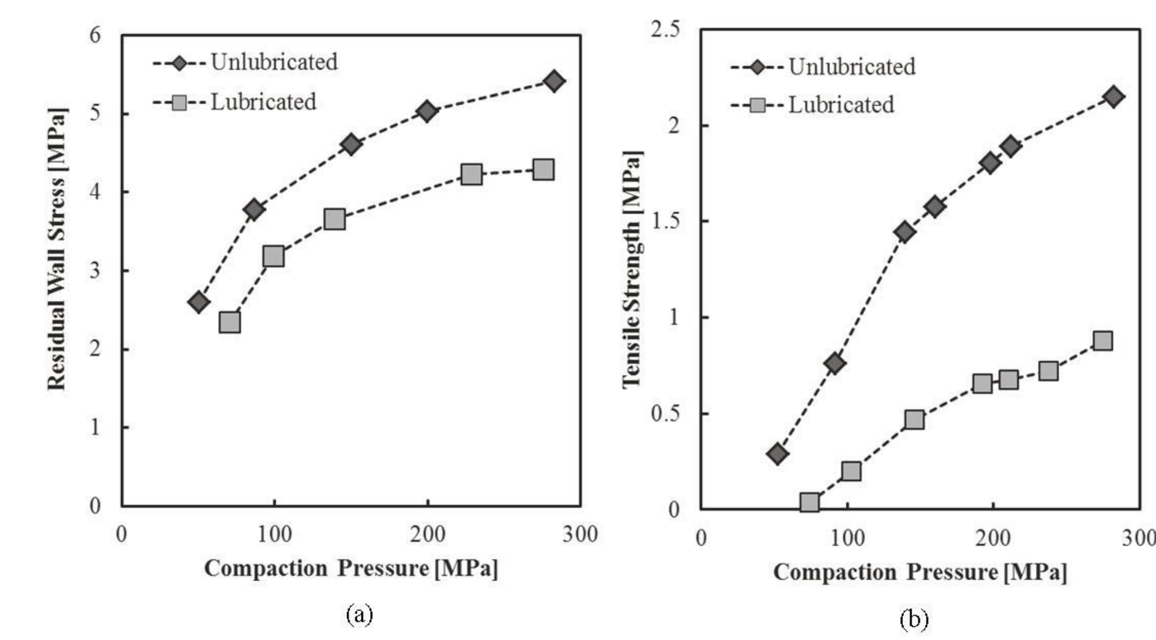
With the response surface determined (β's are now known) the model parameters can be extracted with an optimization method

Derringer, G., *Simultaneous optimization of several response variables.* J. Qual. Tech.

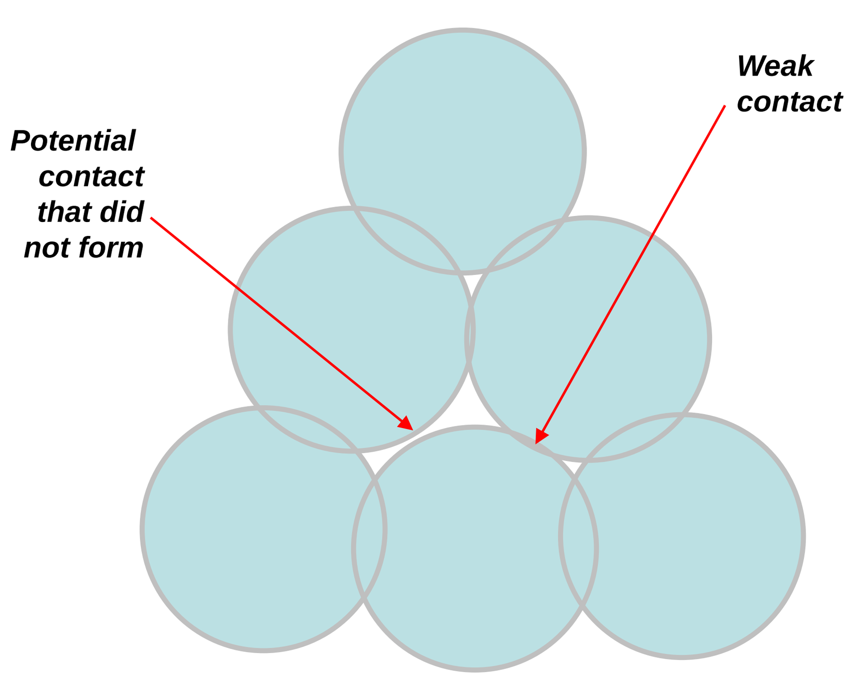
Predictions for single material

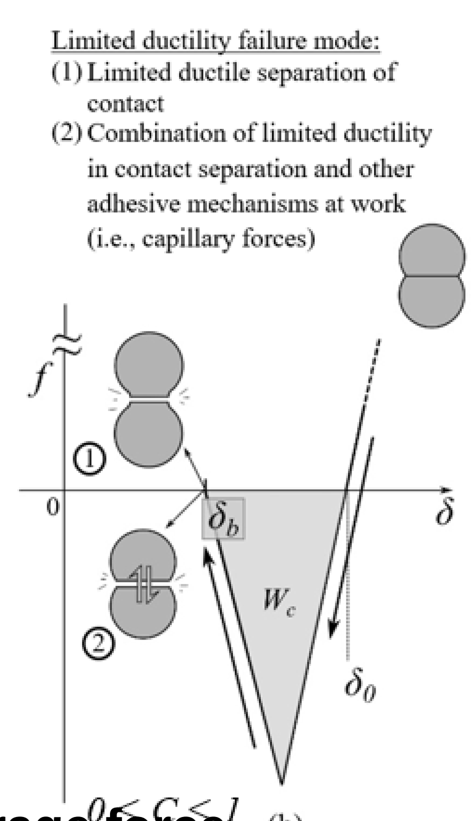






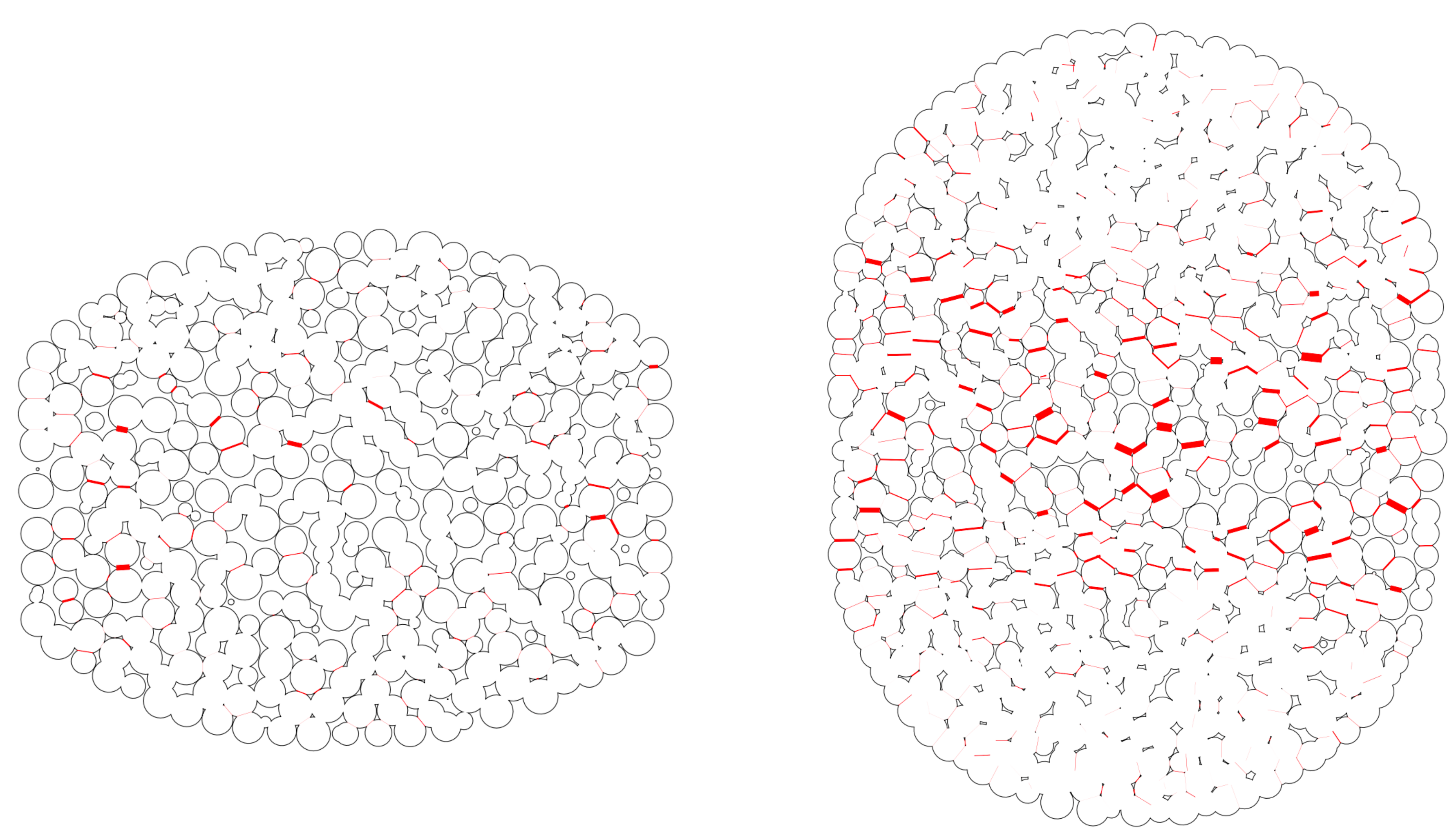
Understanding the strength of compacts





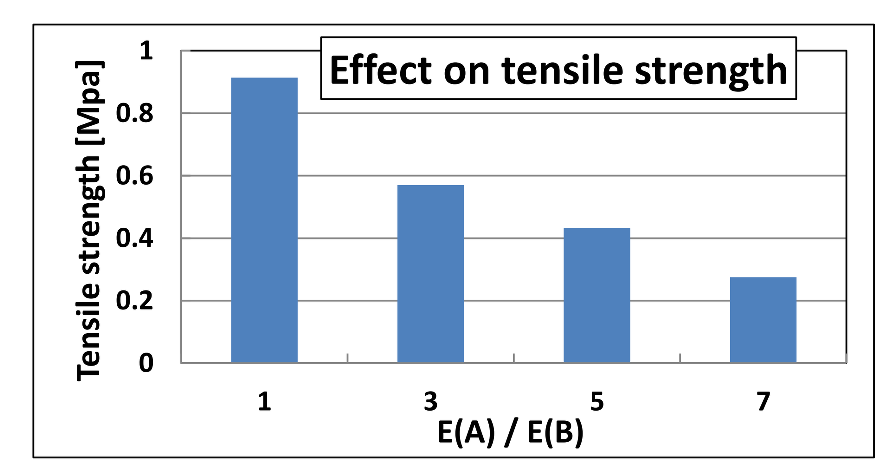
- **Contacts that did not form**
- **Weak contacts pressed at F << average force**
- **Broken contacts (during unloading)**

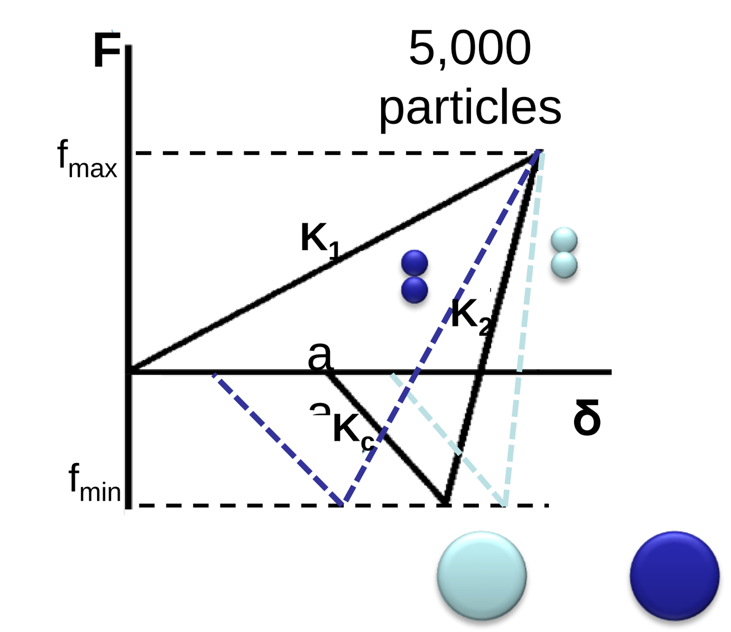
Damage Visualization

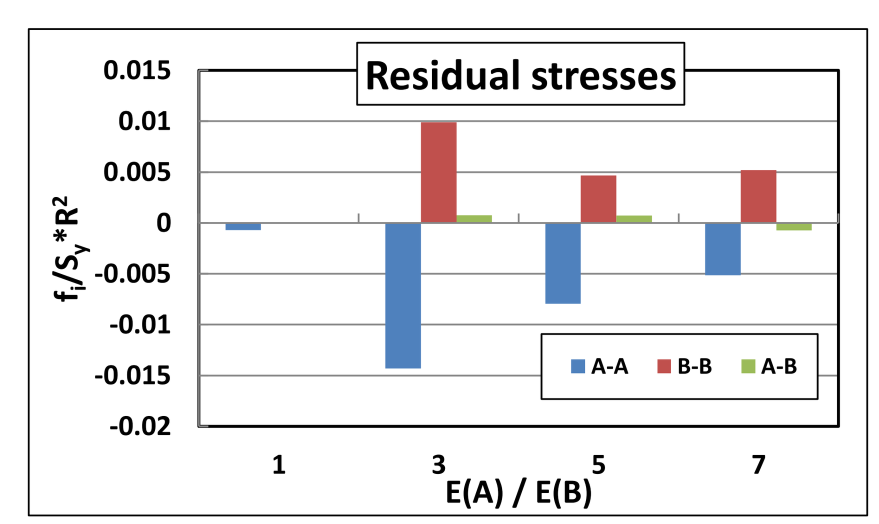


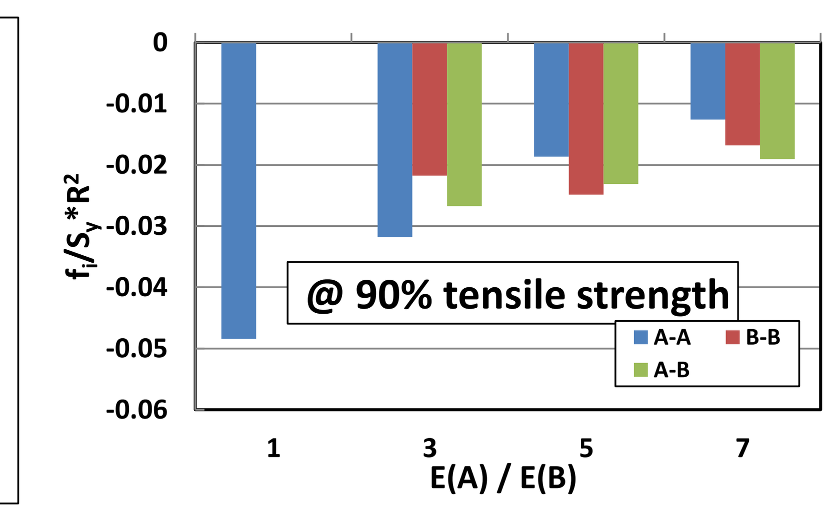
Residual stress: an important factor in strength

A+B mixture
A=B except in elastic modulus









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