IFPRI Project #130 Renewal Proposal

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

Rajesh N. Davé
New Jersey Center for Engineered Particulates
New Jersey Institute of Technology, Newark, NJ, dave@njit.edu

1. Background

Building upon the significant progress achieved in the initial project period, this renewal proposal aims to address critical gaps and extend our understanding of powder flowability and cohesion in the context of adding flow aid, and flow aid-coated cohesive powders. Significant advancements made are summarized below. Going forward, the focus will be on advancing the models and associated experimental investigation to provide a more comprehensive mechanistic understanding of powder behavior.

Year 1 (2021-22) Key Advances (ARR-107-01):
- Reviewed van der Waals force-based particle-contact models for both smooth and rough particles
- Experimental investigation of model-guided enhancements in flow and packing of dozens of fine cohesive powders due to dry coating with silica. Generated a database of industry-relevant materials and their bulk properties (1).
- Tested and validated a deterministic multi-asperity model explaining the effects of silica amount and its uniform distribution on flow enhancements (2)
- Tested the models for determining the amount and type of guest particles (2, 3)
- Introduced the granular Bond number concept for cohesion nondimensionalization and prediction of bulk powder properties (2).
- Showcased enhancements in flow, packing, agglomeration, and dissolution resulting from silica dry coating (4)
- Identified mixing synergy as a key factor for blend flowability improvement where only one of the components is dry coated
- Key findings were covered in two published journal articles

Year 2 (2022-23) Key Advances (ARR-107-02):
- Advanced understanding of selecting flow aids (silica) type and amount, detailed in a published paper
- A model accounting for intrinsic macro-roughness of particles in cohesion reduction post dry coating was proposed and will be further developed and validated
- Accounted for the entire powder sample particle size distribution (PSD) using *size class dependent* Bond number approach to better predict the PSD effect on bulk properties
- Proposed guidelines for mechanistically determining the best component of a blend to select for dry coating
- Examined the performance of different industry relevant silicas on property enhancements of fine APIs, blends, and tablets (5)
- Utilized multiple models to explain the nuances in silica performance on cohesion reduction considering silica size and its intrinsic agglomeration tendency (5)
- Key findings were covered in four published journal articles

Year 3 (2023-24) Planned Activities:

- Model development will continue to account for bulk property enhancements for as received macro rough cohesive powders upon dry coating with nano-sized flow aides
- Further testing and validation of the model accounting for the powder PSD will be carried out
- Ongoing work will examine the applicability of industry relevant, potentially scalable approaches to dry coating, assessing the dry coating performance from a batch device such as LabRAM against using low-intensity batch-mode V-Blender, as well as potentially continuous, higher intensity COMIL while varying the processing parameters
- Key findings during this project period will be covered in two or more journal articles
- A workshop on cohesion reduction through the coating of flow aides will be developed and offered to the IFPRI membership

2. Addressing Key Gaps

Significant progress towards developing mechanistic understanding of cohesion reduction and consequent flow enhancement due to flow additives included the determination of the suitability of the flow additives, estimation of the ideal amount of flow additives, estimating reduced cohesion due to coating with flow additives via mechanistic contact models, albeit under certain simplifying assumptions, and linking particle-scale measures, nondimensionalized using Granular Bond number with bulk scale properties. Several critical gaps remain, which may be broadly classified into three categories: (1) Limitations due to the assumptions related to the host particles, such as having spherical shape, scale of roughness invariant to the PSD, and roughness approximated as mono-sized spherical asperities. (2) Limitations due to the assumptions related to the guest (flow aid) particles, such as uniform coating, and their non-aggregated state, i.e., *monolayer* coating. (3) Addressing industry relevant topics such as accounting for the effect of key parameters of the processing device, examination of the dynamic effects such as the transfer of silica from one constituent to another in a formulation, and the stability and/or transfer of coated flow aids during downstream processing. It is noted that the models developed and tested during the first three years perform remarkably well in predicting the enhancements due to dry coating with various types and amounts of silica for a variety of fine, cohesive industrial powders. However, the above-mentioned advancements are necessary for furthering the industry relevance of our work. That way, the models would be able to cover the broader set of materials and processes used in the industry and help provide guidelines for avoiding potential pitfalls and failures due to inadequate levels of enhancements.

2.1. Extended mechanistic particle contact models

Our ongoing work has addressed the issue of smooth surface assumption for as-received (uncoated) cohesive particles to better understand adhesion estimation for uncoated and dry-coated particles. Since most industrial powders are naturally rough, ignoring the extent of roughness when selecting the amount and type of flow additives is not prudent. Even the "naturally rough" powders having 200 nm roughness owing to Massimilla and Donsi, referencing the fluidized bed cracking catalysts (FCC) particles, was found to be invalid in our work. Therefore, we have carefully examined this topic, identified the need to incorporate the rough surface within the contact models, and developed a preliminary understanding of when the macro roughness scale cannot be ignored. Interestingly, the scale where the roughness cannot be ignored was found to have a power law relationship, see Equation (1), between particle size and roughness size, thus emphasizing the need to assess the scale of powder roughness at various spatial scales of scrutiny.
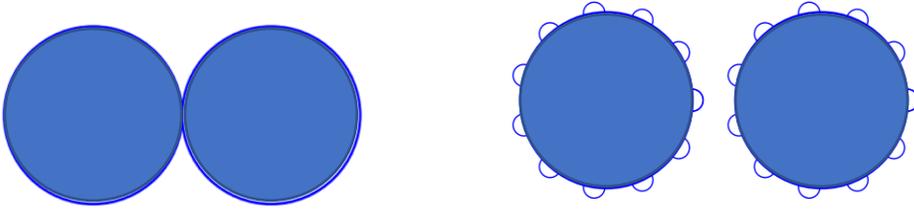
Roughness = α (Particle Size)$^{0.32}$
(1)

Figure 1. Smooth vs Macro Rough Interactions

$$F_{ad} = \frac{Ad}{8z_0^2} + \frac{AD}{24(2d+z_0)^2} \tag{2}$$

To date, equation (2) of Chen's (3) multi asperity contact model has been used to estimate the adhesion of dry coated powders. We have also used this equation to estimate the impact of macro roughness by estimating roughness as spherical asperities. However, the natural roughness would be better approximated as hemispherical asperities (Figure 1, right). Proposed work will address that.

In addition to extending the models to account for natural roughness, this proposal aims to improve the models by incorporating more realistic particle interactions, thus removing the ubiquitous assumption of spherical shaped host particles. The proposed work will combine extensions for macro roughness associated with particles as well as particle shape descriptor distributions in estimating the interaction of particles with varying shapes and surface roughness.

1. *Incorporate effect of host particle macro roughness*: We will extend the contact models to account for various typical shapes for asperities, including hemispherical. However, the challenge lies in the accurate measurement of roughness and its distribution. We propose to analyze SEM images or optical light interferometry by implementing machine learning-based Convolutional Neural Networks (CNNs) and, as needed, validating through AFM-based measures (6).

2. *Incorporate the effect of Host Particle Size and Shape*: To account for the variability in particle size and shape parameters within the material, which significantly impacts adhesion and cohesion forces, extended data collection that accounts for various particle descriptors will be done to help:

   - Develop van der Waals (vDW) interaction models to account for particle shape, captured through the "aspect ratio" as a descriptor of elongated particles. We will consider typical analytical vdW interactions, such as elliptical and cylindrical particles, see Figure 2. Thus, we will examine and incorporate a theoretical framework for vDW interactions between finite cylinders and ellipses contacting in different orientations (7, 8), emphasizing the orientations that are more likely and have higher cohesive forces.

   Figure 2. Model interactions using ellipses and cylinders to account for particle shape

   - Validate the model through empirical data combining size and shape descriptions, along with employing simulations as needed (9).

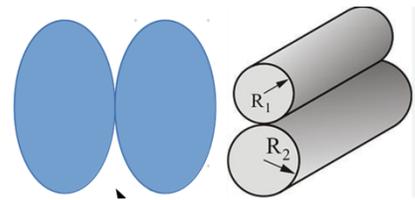3. *Extend the size class dependent Bond number model*: Ongoing work demonstrated that the impact of the entire powder sample PSD can be captured via what is called the size class dependent Bond number model (*a manuscript in preparation*). However, that work only considers all particles to be spherical within all size classes of a PSD. In the proposed work, our ambitious plan is to extend the models that account for both the variations in surface roughness and shape (captured through

aspect ratio) distributions instead of considering them invariant to the size classes. We realize that while assessment of aspect ratios is becoming available through using imaging-based particle analyzers (e.g., Sympatec QicPic and Malvern Morphologi), estimating roughness as a function of size may not be easy. Towards the latter goal, we will explore the applicability of the power law relationship, such as that in Equation 1, or other simplifying approaches. Thus, we propose the extension of the models to cover:

- Rough and Smooth Particle Interactions: To simulate real-world scenarios where particles with similar size though with different surface roughness's interact.
- Shape Variability: While size class dependent Bond number considers particles as smooth spherical particles, to expand the model beyond spherical particles, incorporating ellipses and potentially other shapes (cylinders) as model particle descriptors accounting for aspect ratio, roundedness, sphericity, and other shape descriptors.
- Explore computational methods to simulate interactions between mixed particle shapes.

4. *Comparison with ideal sphere assumption***:** The estimations from the extended models will be compared against currently used ideal sphere models to ascertain the utility of the current model as well as the need to employ advanced characterization and models.

2.2. Uneven coating of flow aids and their agglomeration

The models developed and used in the current project can assess the effect of the various levels of silica, such as sparse or dense yet monolayer coatings, such as those depicted in the left and middle cartoons, Figure 3. Unfortunately, mixing devices have uneven performance that depends on the device type, scale, operating parameters, and processing time. We have shown how the process intensity, as well as host and guest sizes, may influence coating due to the mixing process dynamics (10).   It is expected that even when the amount of silica used is less than 100 % SAC equivalent, the coating may lead to aggregation of silica

Figure 3. Cases of flow aid aggregation, affected by both amounts and device intensity

particles on the host surfaces. The proposed work will address such likely scenarios in industrial processes by developing advanced models for understanding and predicting the effect of the guest agglomerates, right cartoon, Figure 3.
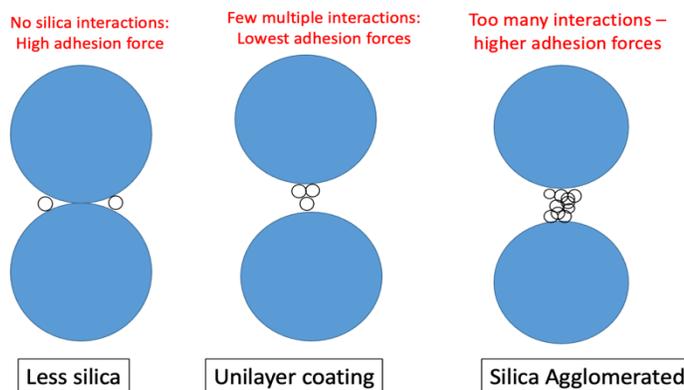
1. *Accounting for silica aggregation***:** A major challenge is to account for the interactions between the aggregated silica and the host particles when it is likely that the main contacts between two host particles are still through multiple guest-guest and a few guest-host contacts. Current literature has not addressed such issues, including considerations such as if an aggregate behaves as a single "porous" body and, if so, what the porosity is. The aggregation of silica is usually quantified using fractal dimensions, but those are generally valid for diffusion-limited aggregates (11, 12). Agglomerates that occur during mixing and coating are not expected to follow such trends. Our work will explore simplifying assumptions to account for such effects, including analytical approaches that account for all possible pairwise interactions between the host and guests contained within an aggregate.

2. *Bulk property improvements due to high or low-intensity mixing***:** We plan to employ low and high-intensity devices which are expected to lead to different types of silica aggregates on host particle surfaces. We will examine the influence of these differences on bulk property enhancements and use the outcomes to validate the models and developed methodology accounting for the effects of both low and high-intensity shear when mixed with flow aids.

2.3 Powder mixing devices, dynamic effects, and flow aid stability analysis

We have initiated the investigation to assess the effect of mixing device type on the outcomes of dry coating. The proposal further extends that work by investigating the effect of processing parameters as well as time for industry relevant devices such as a batch blender and a continuous device such as a conical mill (COMIL). For the COMIL, a few key parameters will be selected from the list; processing time, impeller speed, feed rate, screen (or sieve) size, partial blocking of the sieves, and number of passes. The outcomes will be compared with those obtained from the material sparing batch device such as the LabRAM.

A major novel outcome of the ongoing investigation was that a powder formulation or a blend, where the constituents are cohesive, need not require dry coating of the entire powder mix or even the majority of the constituents. The proposed work will further explore this phenomenon of significant industry relevance because it can greatly simplify the task of flow enhancements and related processing. For example, one may need to dry coat as low as 5 or 10 wt% of a blend to achieve good flowability of the entire blend due to the mixing induced silica transfer and associated synergy (4). That reduces the burden of processing a large quantity of materials and handling of silica. We will investigate the impact of blending device parameters and blend constituents on the flowability of powder blends.

Last, we plan to examine the stability of the flow aid coating during the downstream processing to ascertain if the flow is enhanced after dry coating is sustained through the process train. A simple experiment is proposed to mimic such effects by subjecting the coated powders to a small-scale fluidized bed. We will examine the changes in the coated material, including the potential loss of flow aids through SEM and mass balance analysis by using a host material that can be easily dissolved in water and filtering the suspension to quantify the remaining flow aids after prolonged fluidization.

1. *The effect of processing parameters and time for industry relevant devices*: We plan to further investigate the effect of low and high-intensity mixing devices on the flow aid coating performance. A conventional batch mixer and a COMIL will be investigated. We also plan to use the LabRAM as a material-sparing device by operating it at various vibrational intensities to examine if such an approach may be a substitute for mimicking different types of devices. The advantage of the latter approach is the ease at which LabRAM type device could be modeled via continuum or discrete models.
2. *Dynamic effects of the mixing process on the blends of coated and uncoated components***:** This task will help examine the phenomenon of synergistic transfer of flow aids during the blending of dry coated components with uncoated powders. The investigation may include variables such as proportion of the dry coated constituent, blending time, intensity, and device design on blend flowability, bulk density, and agglomeration. We will examine the applicability of the developed contact mechanics models to capture such synergistic behavior due to the dynamics of silica transfer when a coated component is blended with uncoated ones using different devices (4).
3. *Flow aid stability analysis***:** The stability of flow aid, specifically silica, on the host surface of a selected powder, will be studied using a fluidized bed to mimic various excitations caused during industrial processing. We will use SEM imaging to analyze the surface morphology of blends pre- and post-fluidization, qualitatively assessing the extent of silica adherence and coating stability. In addition, mass balance analysis will be performed to identify the extent of the loss of flow aids.

The overall effect on the flow behavior after prolonged fluidization will be determined by assessing any extent of property deterioration. The coated powder samples will be obtained using low and high-intensity processing devices.

3. Proposed Tasks and Timeline

The proposed work will naturally lead to enhanced or new models, briefly outlined below.

Model A. *Van der Waals (vDW) interaction model to account for hemispherical asperities***:** The existing multi-asperity model, which considers spherical asperities that are evenly distributed, represented by a normalizing parameter called percentage surface area coverage (SAC), will be extended to account for hemispherical asperities. The model will be tested using roughness data of several different industry relevant fine powders.

Model B. *Model to account for particle shape through aspect ratio***:** The vDW-based modeling of interactions between ellipses and cylinders will be considered, and a lumped-parameter model will be developed that could essentially capture the shape influence via a single parameter such as the aspect ratio. Such a model will be tested by deriving the equivalent Bond number against available bulk flowability data.

Model C. *Account for the distributions of particle size, shape, and roughness*: This will be an extension of the *size class dependent* Bond number model accounting for size, shape, and roughness distributions. The model will be first tested via sensitivity analysis using simulated data, followed by testing against realistic data. It is expected that obtaining accurate distributions of both the shape and roughness would be challenging. Therefore, the overall goal is to establish such model-based methodology, knowing well that all the required data may be unavailable and perhaps not necessary. The performance of the model will be tested against the *size class dependent* Bond number model that only accounts for the PSD and not shape or roughness.

Model D. *Accounting for the effect of flow aid or silica aggregation on the host surface on cohesion reduction*: The presence of agglomerated silica or guest particles is expected, and any such occurrences would reduce the effectiveness of the flow aid in reducing the cohesion of the host particles. We will develop simplified models that account for such effects for typical aggregates, which may range from a few particles to dozens of particles.

3.1. Major Research Tasks

Task 1. Develop a mechanistic model accounting for more realistically shaped surface asperities that lead to macro-scale surface roughness.

Task 2. Develop a mechanistic model to account for the particle shape effect through simplified shape estimates such as ellipses and cylinders and using aspect ratio as a lumped parameter.

Task 3. Employ enhanced particle characterization methods to obtain the full set of particle scape properties and their distributions for using in the new models.

Task 4. Develop and extend mechanistic prediction accounting for particle size, roughness, and shape distributions through the extension of the *size class dependent* Bond number approach.

Task 5: Develop reduced order models to account for the effect of flow aid or silica aggregation on the host through typical aggregate size and porosity that occur when low or high-intensity coating/mixing devices are used. Assess the extent of reduced performance due to silica aggregation experimentally and validate the model(s).

Task 6: Carry out the investigation to assess the effect of mixing device type on the outcomes of dry coating. Low-intensity mixer such as a conventional batch blender, and a continuous higher-intensity device, such as COMIL will be used. Benchmarking will be done using a material sparing, LabRAM vibratory mixer.

Task 7: Phenomenon of synergistic transfer of flow aids during the blending of dry coated components with uncoated powders will be investigated. Variables for the study will include as blending time, intensity, and device design. The effect on blend flowability, bulk density, and agglomeration will be assessed. The applicability of the developed contact mechanics models to capture such synergistic behavior will be tested.

Task 8. Perform stability analysis of coated flow aid or silica through prolonged fluidized bed experiments.

3.2. Project Timeline

Deliverables	Year 1				Year 2				Year 3			
	Q1-Y1	Q2-Y1	Q3-Y1	Q4-Y1	Q1-Y2	Q2-Y2	Q3-Y2	Q4-Y3	Q1-Y3	Q2-Y3	Q3-Y3	Q4-Y4
Task 1	■	■	■	■								
Task 2	■	■	■	■	■	■						
Task 3	■	■	■	■	■	■	■					
Task 4			■	■	■	■	■	■				
Task 5			■	■	■	■	■	■	■	■		
Task 6			■	■	■	■	■	■				
Task 7					■	■	■	■	■	■	■	■
Task 8					■	■	■	■	■	■	■	■

References

1.   Davé R, Kim S, Kunnath K, Tripathi S. A concise treatise on model-based enhancements of cohesive powder properties via dry particle coating. Advanced Powder Technology. 2022;33(11):103836.

2.   Kunnath KT, Tripathi S, Kim SS, Chen L, Zheng K, Davé RN. Selection of Silica Type and Amount for Flowability Enhancements via Dry Coating: Contact Mechanics Based Predictive Approach. Pharmaceutical Research. 2023.

3.   Chen Y, Yang J, Dave RN, Pfeffer R. Fluidization of coated group C powders. AIChE Journal. 2008;54(1):104-121.

4.   Kim SS, Castillo C, Sayedahmed M, Davé RN. Reduced Fine API Agglomeration After Dry Coating for Enhanced Blend Uniformity and Processability of Low Drug Loaded Blends. Pharmaceutical Research. 2022;39(12):3155-3174.

5.   Kim SS, Seetahal A, Kossor C, Davé RN. On predicting the performance of different silicas on key property enhancements of fine APIs, blends, and tablets. Powder Technology. 2024;432:119104.

6.   LaMarche CQ, Leadley S, Liu P, Kellogg KM, Hrenya CM. Method of quantifying surface roughness for accurate adhesive force predictions. Chemical Engineering Science. 2017;158:140-153.

7.   Gan JQ, Zhou ZY, Yu AB. Interparticle force analysis on the packing of fine ellipsoids. Powder Technology. 2017;320:610-624.

8.   Yang RY, Zou RP, Yu AB. Computer simulation of the packing of fine particles. Physical Review E. 2000;62(3):3900-3908.

9.   Deng XL, Davé RN. Dynamic simulation of particle packing influenced by size, aspect ratio and surface energy. Granular Matter. 2013;15(4):401-415.

10.   Zheng K, Kunnath K, Ling Z, Chen L, Davé RN. Influence of guest and host particle sizes on dry coating effectiveness: When not to use high mixing intensity. Powder Technology. 2020;366:150-163.

11.   Deng X, Huang Z, Wang W, Davé RN. Investigation of nanoparticle agglomerates properties using Monte Carlo simulations. Advanced Powder Technology. 2016;27(5):1971-1979.

12.   Deng X, Davé RN. Breakage of fractal agglomerates. Chemical Engineering Science. 2017;161:117-126.