IFPRI Project #130 Renewal Proposal

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

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1. Background

Building upon the significant progress achieved for the original project, 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. Advancements made towards the deliverables of the original project proposal are summarized below. Specifically, deliverables D1-D9 have been completed, model integration of D10 has been covered in published papers [1, 2], and D11-12 are ongoing. Deliverable D10 is completed by incorporating a dynamic model [3] accounting for host and guest properties as well as process intensities for dry coating effectiveness and allowed us to compare performance of four different flow additives while also considering the original Chen model [4] and guest-host suitability considerations summarized in [5]. Thus all originally proposed models are now well developed, and they account for the effect of flow aid type and amount, host powder particle size distribution, its natural roughness, as well as provide insight as to which flow additive would work well. Having said that, we have realized that implementation of these models by industry members would be challenging without further efforts to develop for user-friendly computer-based integration of all the models. Further, the already developed models have been shown to capture the effect of flow aid very well, including for the powders that are not spherical, provided the dispersion of the flow aid on the host surface is very good. Nonetheless, it was found that industry relevant mixing devices do not achieve good dispersion of the flow aid and there are issues related to non-uniform dispersion and formation of aggregates of the flow aids on the host particle surface that are not captured in the current models. Such issues as well as industry user-friendly package development will be addressed in the renewal proposal.

Overall, the original project is well on track and the final report will also include a preliminary version of the integrated model accounting for the effect of flow aid type and amount, host powder particle size distribution, and characteristic mean roughness of the host powders, applicable for the cases where the dispersion of the flow aid on the host surface is very good. For the renewal proposal, the focus will be on advancing the models and associated experimental investigation to provide a more comprehensive mechanistic understanding of powder behavior where the addition and coating of the flow aid is done using industrially relevant mixing devices that have differing process intensities. The renewal project will also develop user-friendly computerized package for industry usage.

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 [5].
- Tested and validated a deterministic multi-asperity model explaining the effects of silica amount and its uniform distribution on flow enhancements [6]
- Tested the models for determining the amount and type of guest particles [4, 6]
- Introduced the granular Bond number concept for cohesion nondimensionalization and prediction of bulk powder properties [6].

- Showcased enhancements in flow, packing, agglomeration, and dissolution resulting from silica dry coating [2]
- 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
- Developed a model to account for the entire powder sample particle size distribution (PSD) using the *size class dependent* Bond number approach to better predict the PSD effect on bulk properties
- Proposed initial 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 [7]

Year 3 (2023-24) Current and Planned Activities:

- Utilized multiple models in an integrated fashion to explain the nuances in silica performance on cohesion reduction considering silica size and its intrinsic agglomeration tendency [7]
- 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 offered to the IFPRI membership along with a current version of the integrated model accounting for the effect of flow aid type and amount, host powder size distribution, and their characteristic roughness, provided the dispersion of the flow aid on the host surface is very good.

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, 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: (i) Flow aides other than silicas, e.g., nano-sized alumina, titania, and carbon black. (ii) Developing guidelines for the effect of humidity during/on the coating process by examining the different coated product characteristics as a function of humidity during sample conditioning under low, medium, and high humidity levels. The main goal is to generate guidelines as to how well these need to be controlled during the coating process. (iii) Accounting for the effect of key parameters of the processing device such as the processing intensity. (iv) 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. In addition, we will be integrating all the models as a packaged modeling/simulation tool with user-friendly interface for easier industry implementation. We will offer workshops each year to demonstrate the capability of this simulation package which will be continuously expanded in scope and additional improvements based on the member input. Such a simulation package would also offer the capability to perform sensitivity analysis by varying key parameters to generate practical guidance for the members.

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 assumption of "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.

$$\text{Roughness} = \alpha\ (\text{Particle Size})^{0.32} \qquad (1)$$

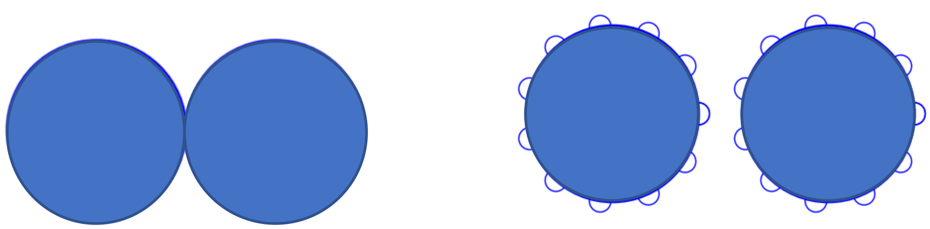

Figure 1. Smooth vs Macro Rough Interactions

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

To date, equation (2) of Chen's [4] 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). The 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 [8].
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 [9, 10], emphasizing the orientations that are more likely and have higher cohesive forces.
    - Validate the model through empirical data combining size and shape descriptions, along with employing simulations as needed [3].

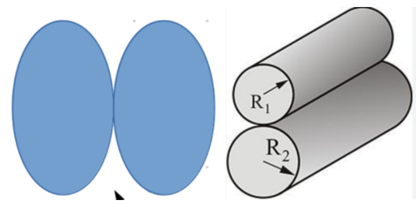

Figure 2. Model interactions using ellipses and cylinders to account for particle shape

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 renewal, we will extend the models to account for the variations in size coupled with non-spherical shape, captured through aspect ratio. The other possible extension of the model is by incorporating an average value of surface roughness in the size-class dependent Bond number model. While assessment of aspect ratios is becoming available through 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 that are easier to implement for IFPRI members. Keeping that in mind, both the shape and surface roughness would be incorporated under the assumption that they are invariant with respect to particle sizes in a powder sample of interest.

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 identify 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, not to mention the amount of flow aids in relation to the host material. We have shown how the process intensity, the relative amount of flow aid in terms of SAC%, as well as host and guest sizes, may influence coating due to the mixing process dynamics [11]. The model in (10) predicts that even when the amount of silica used is less than 100 % SAC equivalent, the coating may lead to aggregation of silica 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 [12, 13]. Agglomerates that occur during mixing and coating are not expected to follow such trends. While the detailed modeling to predict the structure and porosity of the agglomerates would be unnecessary and out of scope, our work will explore simplifying assumptions to account for such effects to assist parametric understanding of the effect of silica aggregation at different assumed porosity values. Our approach may require using analytical methods that account for all possible pairwise interactions between the host and guests contained within an aggregate.

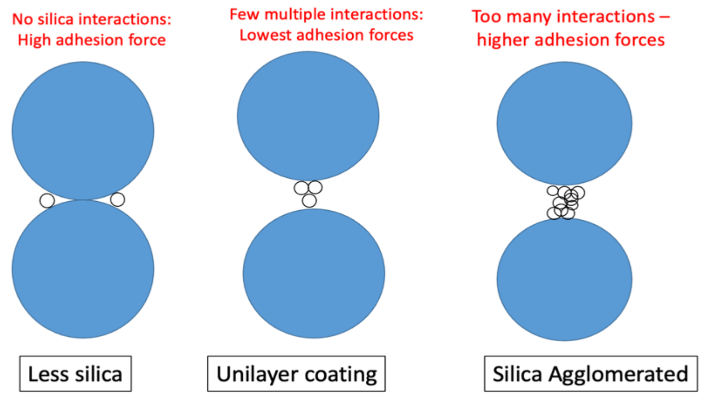

Figure 3. Cases of flow aid aggregation, affected by both amounts and device intensity

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 develop methodology accounting for the effects of both low and high-intensity shear when used to coat flow aids.

2.3 Industry relevant topics: Mixing device type/intensity, flow aids other than silica, effect of humidity, 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 operated at different processing intensities, presumably, lower LabRAM intensities may mimic other low intensity mixing devices.

As a *side project*, we plan to evaluate a novel strategy where only a portion of the powder sample may need to be *dry coated* using a high intensity device. In our recent work, we discovered 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 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 10 wt% of a blend to achieve good flowability of the entire blend due to the mixing induced silica transfer and associated synergy [2]. That reduces the burden of processing a large quantity of materials and handling of silica.

Amongst other industry relevant topics, a few examples of flow aids other than silica will be investigated and the guidelines related to the need to control humidity during the coating/mixing process will be developed. 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 [2].
3. *Flow aids other than silica*: Our group has examined several flow aids and nanomaterials in the past other than silica and generally found that many of those are effective in promoting flow after coating the host particles with those nano-sized particles. Examples of the materials tested include alumina, titania, iron oxide, and even carbon black. It was observed that their performance depends on our ability to effectively coat them and their size, which was later confirmed in our models [4, 14]. In the renewal proposal, we will test two or three such materials after input from the IFPRI members and analyze the results based on our previously known models.
4. *Effect of humidity*: The ambient conditions such as the relative humidity are likely to affect the flow aid coating process. We will do a limited exploration using a host-guest system selected in consultation with IFPRI members. The dry coating experiments will be conducted in our LabRAM system. Prior to dry coating experiment, the powder samples will be conditioned at low, medium, and high humidity levels in the processing jar. The coating effectiveness in terms of resulting powder characteristics will help understand the influence of humidity and adsorbed water molecules on the host powders.

5. *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 distributions along with non-spherical shape, and a characteristic value of host particle surface roughness. The model will be first tested via sensitivity analysis using simulated data, followed by testing against realistic data. 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 (Model A) accounting for more realistically shaped surface asperities that lead to macro-scale surface roughness.

Task 2. Develop a mechanistic model (Model B) 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 shape properties and their distributions for use in the new models.

Task 4. Develop and extend mechanistic prediction (Model C) 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 (Model D) 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: Develop integrated model(s) packaged as a computer-based tool with user-friendly interface for easier industry use and implementation. The capabilities of such simulation package will be expanded each year incorporating model advances and the members input.

Task 7: 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 8: The 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 9: Assess the use of flow aids other than silica and hence ascertain that the main mechanism of flow enhancement is governed by the altered surface roughness due to the coating of the flow aid on to host/carrier particles.

Task 10: Examine the effect of humidity on coating process effectiveness and subsequently, the effect of humidity on tribo-charging tendency of the coated particles.

Task 11: Perform stability analysis of coated flow aid or silica through prolonged fluidized bed experiments.

3.2. Project Timeline

Tasks	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					■	■	■	■	■	■	■	■
Task 9						■	■	■	■	■	■	■
Task 10						■	■	■	■	■	■	■
Task 11						■	■	■	■	■	■	■

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