A REGIME MAP APPROACH FOR PREDICTING THE AGGLOMERATION OF FINE WET POWDERS AFTER DRYING UNDER VARYING LEVELS OF SHEAR

IFPRI Research Project Proposal: Drying Wet Powders with Shear to Prevent Agglomerate Formation

Dr. Heather Emady, Associate Professor of Chemical Engineering, Arizona State University

BACKGROUND

Wet powder drying is a unit operation used in a variety of industries, yet the many variables involved in this process make it very difficult to predict the dried powder product attributes. A particular concern for drying of fine powders is the tendency for agglomerate formation and also attrition, which are highly dependent upon the level of shear in the dryer. The extents of both agglomeration and attrition can be characterized by the change in particle size and size distribution between the initial powder and the dried powder product.

Important dried powder product attributes include particle size and size distribution, particle shape, and bulk density. Formulation properties (e.g., particle size and size distribution, particle shape, particle density, bulk and tapped density) and process variables (e.g., liquid content, temperature, shear rate, time) can all influence the product attributes.

A regime map can be a useful tool to guide formulation choices and process operation, and is an improvement upon just performing many experiments to test variables, and is a positive step toward predictive model development.[1] Regime maps for wet granulation (the process of adding a liquid binder to a powder bed, usually with some level of agitation, for particle size enlargement) were established over 20 years ago, with separate regime maps for nucleation[2] and growth.[3,4] More recently, we expanded this regime map framework for single drop granule formation in static powder beds (see **Figure 1**).[5] Two distinct mechanisms and corresponding granule shape regimes were discovered, based upon the powder bed porosity and the granular Bond number. Spreading occurred with granular Bond numbers <65,000 (roughly corresponding to surface mean particle sizes >30 μm) for all bed porosities below the minimum fluidization porosity and formed flat granules, while Tunneling occurred with granular Bond numbers >65,000 (roughly corresponding to surface mean particle sizes <30 μm) for all bed porosities and formed round granules. These two regimes in particle size, from coarser particles that naturally form a smooth, packed bed of individual particles, down to finer particles that form a fluffy powder bed comprised of dry agglomerates, are expected to be critical components in the development of a drying regime map for this proposed work.
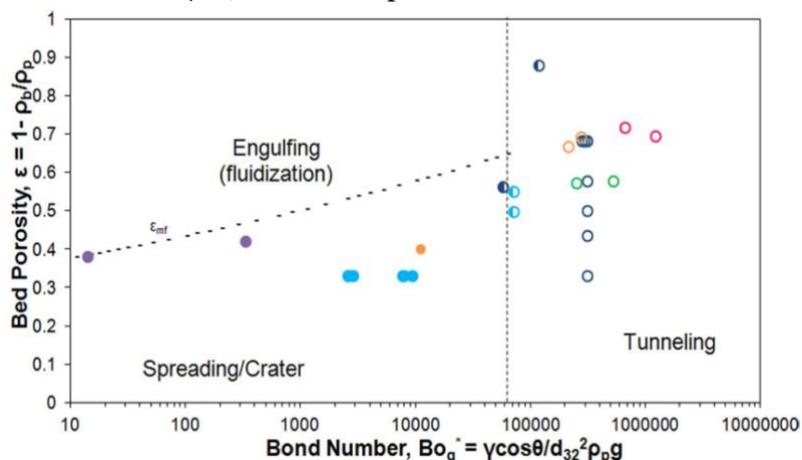

Figure 1. Granule formation mechanism regime map.[5]

PROPOSED RESEARCH PLAN

The overall goal of this research is to develop a particle drying regime map that determines the conditions under which agglomerate formation will occur in drying processes with varying shear rates. Toward this end, the following research objectives will be explored:

(**RO 1**) Raw Material Characterization and Preparation of Wet Particle Assemblies;
(**RO 2**) Tray Drying of Wet Particles [*no shear*];

(**RO 3**) Rotary Drying of Wet Particles [*low-medium shear*];
(**RO 4**) Jet Milling of Wet Particles [*high shear*];
(**RO 5**) Dried Product Characterization; and
(**RO 6**) Regime Map Development.

Details of each objective are provided in the subsequent sections, and a proposed timeline for carrying out the work for all objectives is included at the end of the proposal, in **Table 3**. More details are given for **RO 3** to demonstrate the capability of our in-house rotary drum dryer system, while the drying tests for **RO 2** and **RO 4** use standard commercial equipment.

(RO 1) Raw Material Characterization and Preparation of Wet Particle Assemblies:

The selection of materials will be done with the input of IFPRI members and will cover the two important particle size regimes (e.g., <30 μm and 30-100 μm). In addition to particle size, thorough raw material characterization is essential for elucidating the powder properties that will be relevant for regime maps. The techniques available in our powder characterization lab and at the ASU imaging facility, outlined in **Table 1**, will be utilized for this purpose.

Table 1. Equipment and corresponding measurements used for material characterization.

| Equipment	Measurement
Malvern Morphologi G3 Automated Particle Characterization System with Automated Sample Dispersion Unit	Particle size and shape, and their distributions
AccuPyc II 1340 Automatic Gas Pycnometer	Particle density
SOTAX Tap Density Tester	Bulk and tapped density
Bruker MultiMode 8 Atomic Force Microscope (AFM)	Particle surface topography and particle adhesion
Zeiss Auriga FIB-SEM	Particle surface topography

The wet particle assemblies will be prepared by mixing the powder with the desired amount of liquid and kneading until the mixture appears homogeneous. The mixtures will be prepared immediately before each test in order to minimize evaporation.

(RO 2) Tray Drying of Wet Particles [no shear]:

Oven tray drying will be performed in order to have a control drying case without shear. We have a Fisher Scientific 60L Gravity Oven in our lab that is capable of operating at temperatures up to 250°C. In these experiments, the weight of the drying particles will be monitored at regular intervals throughout the drying process. The weight loss in the system will be equal to the quantity of water lost from the wet particles upon drying. This mass balance can be used to determine the drying kinetics and heat transfer rate within the wet particles. Since the particles are tray dried without any particulate motion within the system, the shear rate will be zero for these experiments.

(RO 3) Rotary Drying of Wet Particles [low-medium shear]:

Rotary drying will be performed in order to test the low-medium shear regimes. We will use our in-house rotary drum experimental setup that is designed for conductive and convective granular heat transfer, with particle temperature quantification via a thermal IR camera. Rotary drum systems have been used to explore mixing of fine particles, as characterized by the axial dispersion coefficient,[6,7] but little experimental work on the heating and drying of this class of materials in rotary drums is available in the literature.
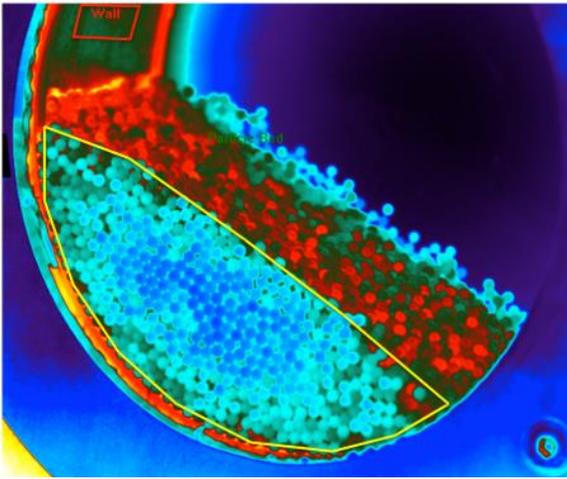
Figure 2. Sample IR camera image, with region of interest selection for the drum wall (red) and the particle bed (yellow).

Details of our experimental rotary drum setup and thermal IR camera are presented in Adepu et al.[8] The setup consists of a stainless-steel cylinder (ID = 15.24 cm, OD = 16.51 cm, L = 7.62 cm) that is closed on each end by 6 mm thick windows. One end of the drum is closed by a sapphire window (D = 15.24 cm) that provides optical and thermal access to the interior particle bed, and the other side of the drum is closed by a quartz window (D = 16.51 cm) that provides optical access to the particle bed. Both windows are held in place using 1.27 cm thick titanium rings that are specifically chosen due to the low conductivity, thereby preventing direct contact of the drum walls with the rollers used for rotating the drum and reducing heat loss through any contact. The sapphire window is held using a 14.478 cm ID, 27.94 cm OD titanium ring, and the quartz glass is held using a 15.24 cm ID, 27.94 cm OD titanium ring. The drum rotates on the two 27.94 cm OD titanium wheels using rollers with variable rotational speed. Three heat guns are placed with equal spacing, at 120° apart, around the drum, to provide effective uniform heating. An infrared radiation (IR) camera (FLIR A6701SC) is used to capture the temperature profile through the sapphire window (see **Figure 2**). Therefore, the sapphire window is specifically chosen to give a high transmittance to IR light, with a transmission range from 0.17 μm to 5.5 μm.

This rotary drum system was designed for conduction heat transfer via the drum walls. However, to incorporate convection heat transfer, the setup has been modified to introduce forced convection into the drum system. For the conduction setup, one side of the drum is closed using a quartz window and another side is covered using a sapphire window. For convection, besides the sapphire window that is used for IR imaging, a quartz window with holes to pump hot air into the drum is used. The quartz window used features a central inlet hole for hot air to be forced into the drum for internal heating. It also has four smaller holes around the edge to let air outside, in order to release the pressure from inside the drum. The internal heat gun is attached to an air duct that connects with pipe fittings to a temperature sensor and the air inlet port. This heat gun serves to insert a hot air stream into the drum to heat the particle bed inside the drum via forced convection. The rotary drum experimental setup, showing the placement of all four heat guns, is provided in **Figure 3**.

The FLIR A6701SC infrared camera is a non-contact device that detects and transforms infrared radiation (heat) into a visual image through digital video outputs. A lens, a heat sensor, processing circuitry, and a mechanical housing comprise the IR camera's components. The lens focuses infrared radiation on the sensor. This perceived energy could be carefully quantified and recorded – as low as 0.1ºC – as a picture or video, which can then be evaluated for additional calculations.[9] The IR camera records the temperature evolution as a video, coupled with the temperature scale for the region of interest, while the particles are heated/dried inside the rotating drum. The rotating drum's outer drum wall temperature can be measured with a thermocouple, and the internal drum wall temperature can be measured with the IR camera. As the heating advances, the temperatures on the exterior and interior drum walls will equalize. The IR camera captures the front and top faces of the particles inside the rotating drum, which will be utilized to calculate the temperature profile. Since the rotary drum is short (7.62 cm in length) and cylindrical, it is reasonable to assume that the cross-sectional area and temperature profile remain consistent along its length. Thus, the heat transfer rate in the rotary drum can be measured.

We will control the shear rate inside of the drum via the rotation rate, which can operate from 1-15 rpm (if the IFPRI members would like to see higher shear in the rotary drum than is achievable from 15 rpm, then we could modify the system by replacing the motor). Particle image velocimetry (PIV) is a

noninvasive method that employs the idea of particle displacement over a defined tiny finite separation period, which offers a quantifiable, immediate velocity vector field that can be used to determine shear rates.[10] As the IR camera records the temperature evolution as well as the particle motion inside the rotary drum, it can be used to apply PIV and hence calculate the shear rates.

(a)　　　　　　　　　　　　　　　　　　　　　　　　　(b)
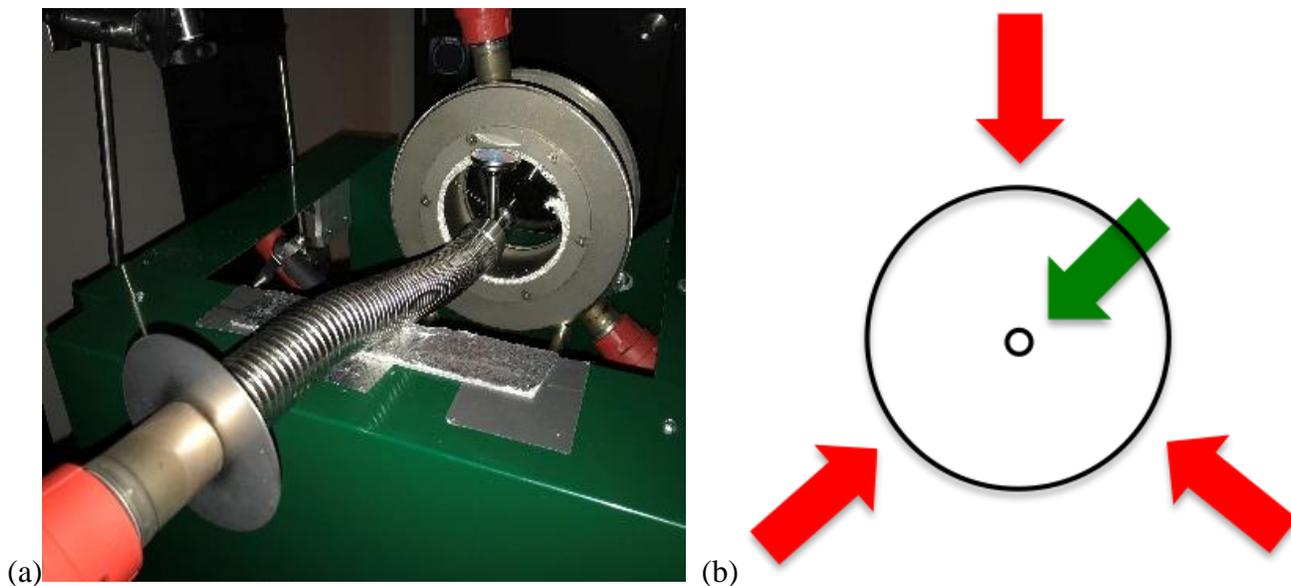
Figure 3. Rotary drum experimental setup. (a) Rear view of empty drum with air inlet duct and quartz window. The opposite end of the drum is enclosed with a sapphire window, where the thermal IR camera is positioned in order to obtain the particle temperature profile within the drum. (b) Schematic of the heat addition to the rotary drum, with the red arrows indicating external heat guns, and the green arrow indicating an internal heat gun.

(RO 4) Jet Milling of Wet Particles [high shear]:

Jet milling will be performed in order to test the high shear regime. We have a Fluid Energy Model 00 Jet-O-Mizer Mill in our lab, which can process 0.75-15 g/min of material. We wanted to include a high shear case in our experimental plan, but our control and measurement of the temperatures and shear rates for this mill are not as sophisticated as those we are able do with the rotary drum in **RO 2**. The operating variables for this mill include the supply air flow rate, the grinding nozzle pressure, and the pusher nozzle pressure. We will manipulate these variables in an attempt to vary the shear rate, although accurately quantifying this may be challenging. We will attempt to measure the heat transfer rates using the mass balance approach presented in **RO 1**, but we acknowledge that there will likely be some loss of material within this milling system. Thus, this research objective will be more qualitative than quantitative, but should nonetheless provide some useful insights into the dried product resulting from a high shear system.

(RO 5) Dried Product Characterization:

The dried powder will be characterized using the same techniques outlined in **RO 1**. A key attribute of the dried powder product is the extent of agglomeration. The product particle size, shape, and their distributions will be measured using the Malvern Morphologi G3. These product size and shape characteristics will then be compared with those of the original powder, as obtained in **RO 1**, to quantify the extent of agglomeration. The surface properties of the dried powder are also critical, and will be characterized using AFM and SEM and compared with the measurements of the original powder, as obtained in **RO 1**, to quantify the change in surface properties.

(RO 6) Regime Map Development:

The regime map will incorporate fundamental properties of the particles (e.g., particle size and shape), as well as process conditions (e.g., moisture content and shear rate), into dimensionless groups. **Table 2** provides a summary of all of the variables that will be tested, and the results will be obtained from **RO 1-5**, which will be used for derivation of the dimensionless groups and development of the regime map that will predict agglomerate formation upon drying under shear. From the literature in the areas of unsaturated porous media[11] and the general dynamics of wet particle systems,[12,13] we found some relevant quantities that incorporate these variables, to be used as a basis for the regime map development. These quantities include the van der Waals binding energy, capillary force or pressure, degree of saturation, Weber number, Bond number, and capillary number.

Table 2. Tested variables for inclusion in regime map.

	Variable
Formulation	Particle size and size distribution
	Particle shape
	Tapped and bulk density
	Root mean square of roughness
	Hamaker constant or force/work of adhesion
Process	Temperature
	Shear rate
	Mass of material
	Moisture content
Product	Particle size and size distribution
	Particle shape
	Tapped and bulk density
	Root mean square of roughness
	Hamaker constant or force/work of adhesion

CRITICAL UNKNOWNS

The primary uncertainties in the proposed work are: (1) the quantification of the shear rate, and (2) determination of the dimensionless groups to be used in the resulting regime map. Shear rate is variable across various equipment designs, but we strive to develop a single dimensionless group that quantifies shear rate across a variety of process equipment. For example, in the rotary drying process, there will be a balance between the shear of drum rotation versus the shear of air flow into the drum. We will not have an understanding of what this balance looks like until we actually perform the experiments. Additionally, we do not know the dominating variables in each of these drying processes before we do the experiments (although we now have a mechanistic basis upon which to start), so our experimental results will dictate the formation of the dimensionless groups for the regime map. Despite these unknowns, we have experience using this framework of characterizing the feed material, performing experiments looking at the effects of varying process variables, and characterizing the product material; then, we combine all of these components to determine the dominating dimensionless groups and corresponding regime map.

INTEGRATION INTO EXISTING RESEARCH PROGRAM

The proposed research combines our lab's two main research thrusts, namely single drop granulation and granular heat transfer. The regime map framework comes from the granulation project, which is currently funded by the NSF CAREER award. However, we do not currently have external funding for our granular heat transfer project, and the proposed work can leverage our existing infrastructure for this project, namely the rotary drum with the thermal IR camera.

OPPORTUNITIES FOR IFPRI MEMBER SUPPORT

The proposed research would benefit from the supply of appropriate particulate materials from IFPRI members. The two categories of particles needed are as follows: particles with surface mean sizes <30 μm, and those between 30-100 μm. Beyond these size requirements, we would be open to any type of particulate materials from relevant industries.

In addition to the supply of test materials, we would appreciate the input of IFPRI members for industrially relevant drying conditions (e.g., liquid saturation levels, temperature, drum rotation rate, etc.) to test in our setups.

PROPOSED TIMELINE

The proposed research will take place in Dr. Heather Emady's laboratory at ASU from August 1, 2022 – July 31, 2025, and the details are provided in **Table 3**.

Table 3. Timeline of research activities for the proposed work.

	Year 1	Year 2	Year 3
RO 1: Raw Material Characterization and Preparation of Wet Particle Assemblies			
Select materials of the following surface mean particle sizes: (1) <30 μm binary, and (2) 30-100 μm	▓		
Characterize materials: particle size and size distribution, particle shape, particle density, bulk and tapped density, surface topography and adhesion	▓	▓	
Prepare wet particle assemblies for each drying test	▓	▓	▓
RO 2: Tray Drying of Wet Particles [*no shear*]			
Perform oven tray drying experiments and test for the effects of: (1) temperature, (2) mass of material, and (3) moisture content	▓		
RO 3: Rotary Drying of Wet Particles [*low-medium shear*]			
Perform rotary drying experiments and test for the effects of: (1) temperature, (2) mass of material, (3) moisture content, and (4) shear rate		▓	
RO 4: Jet Milling of Wet Particles [*high shear*]			
Perform jet milling experiments and test for the effects of: (1) mass feed rate, (2) moisture content, and (3) air pressure/shear rate			▓
RO 5: Dried Product Characterization			
Measure dried powder product properties, including size, shape, and their distributions, in order to quantify the extent of agglomeration; also measure surface topography and adhesion		▓	▓
RO 6: Regime Map Development			
Develop regime maps that predict agglomerate formation after drying based on dimensionless groups involving the formulation properties and the process variables			▓

RESPONSES TO IFPRI REVIEWER COMMENTS

Overall, the members feel that your proposal meets the project brief and believe that development of a regime map for agglomerate breakage in drying will be useful. They are concerned, however, about your vagueness about the controlling dimensionless groups and worry that your approach is too empirical. You note that the Bond number is very large in these systems (in the pendular regime), indicating that they are capillary-dominated. You may find it useful to draw from the unsaturated porous media literature to develop a mechanistic basis for your regime map.

We understand the concerns in using an empirical approach for developing the regime map, and we appreciate the reviewers directing us toward the unsaturated porous media literature. However, from our experience, a mechanistic basis is usually too ambitious a goal for powder systems. Our approach in gaining an understanding of the controlling variables of the material and process will help us better understand the mechanisms, but we will not necessarily end up with a useful mechanistic description. As an example, our past work looked at granule formation mechanisms, which can be determined from a regime map of dimensionless groups that include formulation properties and operating parameters (see **Figure 1**). We did attempt to provide a mechanistic description of the granule formation mechanisms via a force balance approach, but due to the inhomogeneous packing tendencies of fine powders, a single ideal model was unable to accurately predict the formation mechanism behavior, as compared to the empirical regime map.[14] Nonetheless, we have now discovered some useful existing mechanistic quantities in the literature (thank you again to the reviewers for pointing us in this direction) that will be relevant to our system and will give us a more mechanistic basis to start for our dimensional analysis, and have now included the discussion of these under **RO 6**.

Also, you are unspecific about how you will measure heat transfer rates (including temperature profiles) and shear rates, A more detailed description about how you plan to do this would be useful.

We apologize for neglecting to include this information in our original proposal. We have now added details about the temperature and shear rate measurements for each of our three types of experiments, under **RO 2-4**, at the end of each of these sections.

Your characterization plan doesn't include surface characterization. This seems to be a significant omission, as the surface properties of the particles are likely to dominate adhesion at the very end of drying.

We agree that surface characterization is critical to the success of this project, and thus we have now included that as part of our proposed work. We have added information about the equipment we will use for surface characterization in **Table 1**, as well as some text about this characterization under **RO 5**. We have also now included surface characterization with our regime map development in **Table 2**. Further details about how we will characterize the particle surfaces using this equipment are provided here.

The particle-particle adhesion in a powder is due to the following forces: van der Waals, electrostatic, and capillary. The contact area between the particles heavily influences these forces, and can be influenced by the size of asperities on the surface.[15,16] Atomic force microscopy (AFM) has been commonly used to acquire information on the nature of particle-particle and particle-surface interactions, as well as surface topography.[17] The use of the colloidal probe method, where a particle is mounted onto the tip of a cantilever of an AFM, allows for the measurement of the adhesion of the particle to other particles or surfaces.[15,17–19]

Scanning electron microscopy (SEM) has also often been used to characterize the surface features of particles. The SEM stereoscopy technique has been used to obtain data of the three-dimensional particle surface for analysis of the topography.[20]

An AFM and an SEM will be used to characterize the surface topography of the particles. The images and the root mean square (RMS) of the roughness will be reported to give insight into the area of interaction for the adhesion of the particles. The colloidal probe method will be used to determine the adhesion of the particle-particle system. The SEM will be used to confirm and determine how the particle is mounted on the cantilever tip. The adhesion of the particle-particle system will be quantified from the force-distance curves as one or more of the most-used values in literature: force of adhesion, work of adhesion, and/or a Hamaker constant (assuming only van der Waals forces are present).[21]

REFERENCES

(1)   Kayrak-Talay, D.; Dale, S.; Wassgren, C.; Litster, J. Quality by Design for Wet Granulation in Pharmaceutical Processing: Assessing Models for a Priori Design and Scaling. *Powder Technology* **2013**, *240*, 7–18. https://doi.org/10.1016/j.powtec.2012.07.013.

(2)   Hapgood, K.; Litster, J. D.; Smith, R. Nucleation Regime Map for Liquid Bound Granules. *AIChE Journal* **2003**, *49* (2), 350–361.

(3)   Iveson, S. M.; Litster, J. D. Growth Regime Map for Liquid-Bound Granules. *AIChE Journal* **1998**, *44* (7), 1510–1518.

(4)   Iveson, S. M.; Wauters, P. A. L.; Forrest, S.; Litster, J. D.; Meesters, G. M. H.; Scarlett, B. Growth Regime Map for Liquid-Bound Granules: Further Development and Experimental Validation. *Powder Technology* **2001**, *117* (1–2), 83–97.

(5)   Emady, H. N.; Kayrak-Talay, D.; Litster, J. D. A Regime Map for Granule Formation by Drop Impact on Powder Beds. *AIChE Journal* **2013**, *59* (1), 96–107.

(6)   Koynov, S.; Wang, Y.; Redere, A.; Amin, P.; Emady, H. N.; Muzzio, F. J.; Glasser, B. J. Measurement of the Axial Dispersion Coefficient of Powders in a Rotating Cylinder: Dependence on Bulk Flow Properties. *Powder Technology* **2016**, *292*. https://doi.org/10.1016/j.powtec.2016.01.039.

(7)   Paredes, I. J.; Yohannes, B.; Emady, H. N.; Muzzio, F. J.; Maglio, A.; Borghard, W. G.; Glasser, B. J.; Cuitiño, A. M. Measurement of the Residence Time Distribution of a Cohesive Powder in a Flighted Rotary Kiln. *Chemical Engineering Science* **2018**, *191*, 56–66. https://doi.org/10.1016/j.ces.2018.06.044.

(8)   Adepu, M.; Boepple, B.; Fox, B.; Emady, H. Experimental Investigation of Conduction Heat Transfer in a Rotary Drum Using Infrared Thermography. *Chemical Engineering Science* **2021**, *230*, 116145. https://doi.org/10.1016/j.ces.2020.116145.

(9)   FLIR Teledyne. How Do Thermal Cameras Work? https://www.flir.com/discover/rd-science/how-do-thermal-cameras-work/.

(10)   Hochareon, P.; Manning, K. B.; Fontaine, A. A.; Tarbell, J. M.; Deutsch, S. Wall Shear-Rate Estimation within the 50cc Penn State Artificial Heart Using Particle Image Velocimetry. *Journal of Biomechanical Engineering* **2004**, *126* (4), 430–437. https://doi.org/10.1115/1.1784477.

(11)   Xu, J.; Louge, M. Y. Statistical Mechanics of Unsaturated Porous Media. *Physical Review E - Statistical, Nonlinear, and Soft Matter Physics* **2015**, *92* (6). https://doi.org/10.1103/PhysRevE.92.062405.

(12)   Herminghaus, S. Dynamics of Wet Granular Matter. *Advances in Physics* **2005**, *54* (3), 221–261. https://doi.org/10.1080/00018730500167855.

(13)   Liu, P. Y.; Yang, R. Y.; Yu, A. B. Dynamics of Wet Particles in Rotating Drums: Effect of Liquid Surface Tension. *Physics of Fluids* **2011**, *23* (1). https://doi.org/10.1063/1.3543916.

(14)   Emady, H. N.; Kayrak-Talay, D.; Litster, J. D. Modeling the Granule Formation Mechanism from Single Drop Impact on a Powder Bed. *Journal of Colloid and Interface Science* **2013**, *393* (1). https://doi.org/10.1016/j.jcis.2012.10.038.

(15)   Jaiswal, R. P.; Kumar, G.; Kilroy, C. M.; Beaudoin, S. P. Modeling and Validation of the van Der Waals Force During the Adhesion of Nanoscale Objects to Rough Surfaces: A Detailed Description. *Langmuir* **2009**, *25* (18), 10612–10623. https://doi.org/10.1021/la804275m.

(16)   Bowen, W. R.; Lovitt, R. W.; Wright, C. J. Atomic Force Microscope Studies of Stainless Steel: Surface Morphology and Colloidal Particle Adhesion. *Journal of Materials Science* **2001**, *36*, 623–629.

(17)   Etzler, F. M.; Drelich, J. Atomic Force Microscopy for Characterization of Surfaces, Particles, and Their Interactions. In *Developments in Surface Contamination and Cleaning: Detection, Characterization, and Analysis of Contaminants*; Elsevier Inc., 2012; pp 307–331. https://doi.org/10.1016/B978-1-4377-7883-0.00006-7.

(18)   Vaziri Hassas, B.; Caliskan, H.; Guven, O.; Karakas, F.; Cinar, M.; Celik, M. S. Effect of Roughness and Shape Factor on Flotation Characteristics of Glass Beads. *Colloids and Surfaces A: Physicochemical and Engineering Aspects* **2016**, *492*, 88–99. https://doi.org/10.1016/j.colsurfa.2015.12.025.

(19)   Louey, M. D.; Mulvaney, P.; Stewart, P. J. Characterisation of Adhesional Properties of Lactose Carriers Using Atomic Force Microscopy. *Journal of Pharmaceutical and Biomedical Analysis* **2001**, *25*, 559–567.

(20)   Podsiadlo, P.; Stachowiak, G. W. Characterization of Surface Topography of Wear Particles by SEM Stereoscopy. *Wear* **1997**, *206*, 39–52.

(21)   Liu, Y.; Song, C.; Lv, G.; Chen, N.; Zhou, H.; Jing, X. Determination of the Attractive Force, Adhesive Force, Adhesion Energy and Hamaker Constant of Soot Particles Generated from a Premixed Methane/Oxygen Flame by AFM. *Applied Surface Science* **2018**, *433*, 450–457. https://doi.org/10.1016/j.apsusc.2017.10.030.