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# Coalescence model for induction growth behavior in high shear granulation

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ABSTRACT

Complex wet granulation behavior is often dependent on process parameters and formulation properties simultaneously. Mathematical models of granulation need to account for the complex physics if they are to be predictive. In this paper, a physically-based coalescence model is evaluated for use in the three-dimensional volume-based framework. The Liu coalescence kernel [20] is calculated for the three-dimensional system as a function of (1) the solid, liquid and gas volumes that form the granule, (2) an empirical correlation for granule mechanical properties as they depend on the granule composition and strain rate, and (3) the process conditions. A complex but well characterized industrial formulation is used as a case study. For this formulation, the granule strength varies over an order of magnitude for a range of conditions that correspond to actual behavior during granulation as granules densify due to consolidation. The kernel predicts that most collisions will result in rebound early in the granulation. As granules densify they become stronger, but also surface wet, resulting in Type I coalescence for many collisions once the average granule porosity drops below a critical value of 0.37. A kernel sub-model that incorporates changes in granule mechanical properties as a function of composition can then describe classic induction time behavior. The model is compared to an experimental study in a 50 L horizontal axis high shear granulator. Measurements of the size, porosity and morphology are used to identify rate process mechanisms over the course of the wet granulation process. The proposed coalescence model successfully describes the experimentally observed induction behavior as a function of composition, based on comparison to the kernel regime map for this formulation. The experimental value of critical porosity corresponding to the observed induction time, and continued growth of surface wet granules well past 1 mm in size also match the micro-scale behavior predicted by the kernel. This comparison shows that the physically-based kernel can be used to predict complex granulation behavior such as induction growth.



## 1. Introduction

Wet granulation is a subset of size enlargement processes that is widely used to change the properties of bulk powders such as strength, size, flowability, and attrition resistance for either a final product or a process intermediate. Applications of wet granulation are spread across a wide range of industries, including pharmaceutical, detergent, fertilizer, and food processing. While granulation processes are widely used, control and design of the process are often based on extensive empirical work rather than a first principles model. Changes in formulation properties require extensive pilot scale measurements for adaptation to the production system, and even so, scale-up rules may not remain consistent as formulations change [1]. Macro-scale approaches to granulation have been productive in determining the effect on product attributes of both formulation properties [2,3] and process parameters [3–5]. More recently, physically-based sub-models have been developed that attempt to describe granulation rate processes at the micro-scale. These models have the promise of becoming more widely adopted in practice in granulator design and scaling [6]. To this end, research has been directed at the development of fundamental models that are based on physical properties and process parameters and predict the behavior of granulation rate processes.

The three major rate processes used to describe the change of granule properties during granulation are wetting and nucleation, consolidation and growth, and attrition and breakage [4,7]. Granules are initially formed from primary powder coming in contact with liquid binder. Primary particles can layer around these nuclei granules sticking to any exposed binder on the granule surface. As liquid is distributed in a granulator and more granules are present, it becomes more likely they could impact the granulator walls or other granules. These impacts can cause consolidation of loosely bound granules that have macro-voids filled with gas [8]. During similar collisions, coalescence occurs

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when two granules stick together to form a new larger granule. Granules can break if impact stresses are large enough [4,9]. Several reviews in the literature give a good summary of the state of the art of quantitative understanding of the three classes of granulation rate processes [4,7,10,11]. While these rate processes govern the product distribution of a granulator, a robust, general and predictive model for granulation based on formulation properties has yet to be fully developed and validated [12,13].

The population balance model (PBM) has been used extensively in particulate systems in order to track distributions of particle properties and their changes throughout a process [10]. The general form of the population balance equation and several of its applications have been developed and discussed thoroughly [14]. While one-dimensional models of granulation typically track granule size, granulation rate processes are dependent on liquid content and granule strength among other factors [15]. Therefore, to model these processes using physically-based sub-models, it is necessary to keep track of more than one property of the granule. Verkoeijen, Meesters, and Scarlett developed a three-dimensional population balance model for granulation that tracked the changes in solid, liquid and gas volumes ($v_s$, $v_l$, $v_g$) within the granule population [16]. The coalescence kernel $\beta$ for a 3D PBM can be separated into parts: a collision efficiency term $\beta_{eff}$ (the size dependent part) and a collision rate term $\beta_o$ [17,18].

$$\beta\left(v_s, v_l, v_g, v_s', v_1', v_g', t\right) = \beta_o\, \beta_{eff}\left(v_s, v_l, v_g, v_s', v_1', v_g', t\right). \quad (1)$$

Many forms of this equation exist in literature. Typically an empirical form of the equation defines a functional form for the collision efficiency and the collision rate is used to fit experimental data. Examples of empirical kernels are the equipartition kinetic energy kernel (EKE) and the size dependent kernels of several forms [18–21]. While effective in many circumstances these coalescence sub-models do not take the physical properties of the formulation, or process parameters directly into account. This requires experimental fitting for changes in these operating conditions.

In comparison, a physically-based coalescence kernel uses a micro-scale model to evaluate the coalescence event. The model includes physical parameters that can relate directly to laboratory measurements, granule properties, or process conditions. The physically-based model evaluates the possible outcomes of a coalescence event and describes them in the form of the collision efficiency portion of the kernel with a fitting parameter to describe the collision frequency. There is a limited amount of work on physically-based coalescence kernels [22]. Cryer developed one of the first coalescence kernels from a physical basis for the fluid bed granulator. It uses a probability density function to describe collision velocities of granules [23]. Liu et al. analyzed the kinetic energy balance of two wet elastic plastic spheres colliding head-on [9]. Immanuel and Doyle III incorporated the dynamic changes of the Liu and Litster kernel through the use of the Smoluchowski formulation [24,25]. Other multidimensional population balance models have incorporated liquid content into their size dependent expression but should not be described as physically based as they are the multi-dimensional equivalent of size dependent kernels [26,27]. Examples of these kernels are provided in Table 1 with a description of parameters and other comments.

Physically-based kernels are developed in order to make granulation models more robust to changes in formulation properties and process parameters. The physical basis for the model also provides the possibility of describing more complex rate processes though a physical means rather than an empirical one. In many systems the granules resist deformation and consolidation takes place over long periods of time. At some critical moment many of the granules in the system reach a critically consolidated state and liquid begins to immerge to the surface. This mechanism linking consolidation and coalescence is called induction growth. This type of growth and densification of strong granules can occur in many types of granulators and is of significant interest in high shear granulation where impact energies are relatively large. The change in porosity of granules with respect to time in a granulator has been previously studied and typically this data fits an exponential decay curve to some minimum porosity $\varepsilon_{min}$ [4,8]. When granules consolidate sufficiently that they become surface wet, coalescence may occur. In order to physically model this process an understanding of the granule mechanical properties as a function of concentration and collision velocity should be incorporated into a physically-based coalescence model.

The overall goal for this work is to estimate granule mechanical properties as a function of composition and use this information in a physically-based coalescence kernel sub-model. This will be achieved by the following:

1) Measurement of the dynamic yield stress and elastic modulus of an industrial formulation as a function of porosity, saturation, and strain rate.
2) Using an empirical fit, to estimate the mechanical properties of a granule based on composition and collision velocity.

**Table 1**
A list of physically-based coalescence kernels and models.

| | |
|---|---|
| Physically-based kernel<br>$\beta = \beta_0 \int_{-\infty}^{St^*} f(\Phi, t) d\Phi$ with<br>$St^* = \propto_1 (1 + \exp t - \propto_2)$ | The probability density function $f(\Phi, t)$ incorporates a variance (uncertainty) into parameters like collision velocity. The critical $St^*$ value can vary with time and thus the empirical, physically-based kernel is defined by parameters $\beta_0$, $\propto_1$and $\propto_2$ [22]. |
| Granule scale collision mechanisms<br>$\beta = \begin{cases} \beta_1, \text{ type I coalescence} \\ \beta_2, \text{ type II coalescence} \\ 0, \text{ rebound} \end{cases}$ | This kernel is based on micro-scale collisions of two elastic–plastic, deformable granules with possible liquid layer. Type I is liquid layer dissipated coalescence, while Type II is coalescence with some deformation of the granule [9]. |
| Mechanistic Kernel based on Liu et al. energy balance<br>$\beta(p_1, p_2) = c_1 \frac{4\pi u_o (r_1 + r_2)^2}{W}$<br>with<br>$\frac{W(p_1, p_2)}{(r_1 + r_2)} = \max\left( \begin{array}{l} \int_{D=(r_1+r_2)}^{\infty} \frac{\exp\left(\frac{-\psi_f(p_1, p_2, D - r_1 - r_2)}{kT}\right)}{D^2} d(D), \\ \int_{D=(r_1+r_2)}^{\infty} \frac{\exp\left(\frac{-\psi_r(p_1, p_2, D - r_1 - r_2)}{kT}\right)}{D^2} d(D) \end{array} \right)$ | A mechanistic kernel from emulsion polymerization. Here the kinetic energy balance from Liu's work is applied through the Smoluchowski formulation to describe the system from a physical basis. W is the Fuch stability ratio as defined by the second relation. $\psi_f$ and $\psi_r$ are the net attractive potential based on Liu et al.'s static analysis. $D$ is the granule diameter, k and T are the Boltzmann constant and temperature, $u_o$ is the original impact velocity and $c_1$ is the collision rate fitting parameter [23]. |
| Multidimensional function — size dependent equivalent<br>$\beta = \beta_0 (x^3 + x'^3)$<br>$\left[ (c_x + c_{x'})^{\alpha} \left(100 - \frac{c_x + c_{x'}}{2}\right)^{\delta} \right]^{\alpha}$ | Here $\alpha$ and $\delta$ are fitted parameters and also have the effect that they define a preferential composition average for particles, where $c_x$ is the concentration of binder. $x$ is defined as size of the granule so that $x^3$ is the volume [25]. |
| Mechanistic kernel based on volume of liquid<br>$\beta = \beta_0 \frac{q_{li} - q_{lr\,i}}{4\pi \left(\frac{d_i}{2}\right)^2 \frac{(q_{si} + q_{li})}{v_i}} - \frac{q_{lj} - q_{lr\,j}}{4\pi \left(\frac{d_j}{2}\right)^2 \frac{(q_{sj} + q_{lj})}{v_j}}$ | Here $q_{lj} - q_{lr\,j}$ is defined as the volume of liquid on the surface of the granule, which requires the simplification that no air is inside the granule. A rate equation for consolidation was derived to relate back to these values instead of porosity. [26] |