

Batch Granulation Control Using a Simplified Population Balance and Nonlinear Model Predictive Control

C. E. Long, J. A. Gantt, and E. P. Gatzke

Department of Chemical Engineering, University of South Carolina, Columbia, SC 29208

Abstract—This paper presents a simple granulation model using geometrically spaced bins in order to accommodate changes in particle size spanning many orders of magnitude. Assuming a particle size distribution measurement is available online using emerging measurement techniques, optimal modifications to the process input values are calculated using a constrained nonlinear model predictive control formulation. It is assumed that additional binder can be added at any point in time and that the mixer speed can be modified online. Given variations in initial conditions and process parameters, the shrinking horizon nonlinear moving horizon controller simulation shows improved performance as compared to open loop trajectories.

I. INTRODUCTION

Granulation is the process of particle size enlargement through the use of a binder to produce a granular material with desired physical properties. Particle size enlargement techniques are used in every processing industry that handles particulate feeds, intermediates, or products [1]. If one wishes to eliminate dust, reduce caking / lump formation, control porosity of a product, improve flow properties for processing, or just to improve the product appearance, the process of granulation must be addressed in terms of modeling and control.

Due to the level of detail and complexity, population balance equations represent an efficient and accurate way of modeling the complicated factors associated with particle nucleation, growth, consolidation, and breakage occurring in granulation. Population balance models create a macroscopic view of rate processes affecting the process of granulation. This equation is analogous to a mass balance, which incorporates convective terms, accumulation terms, and kinetic expressions for each mechanism affecting the change of particle size. Some mechanisms include consolidation, layering, coalescence, and attrition. The most general form to the Population Balance Equation (PBE) is presented in Equation 1:

$$\frac{\partial n}{\partial t} + \nabla \cdot \bar{v}_e n + \nabla \cdot \bar{v}_i n + \dot{d} - \dot{b} = 0 \quad (1)$$

The particle distribution, n , may be described as a function of external coordinates (granule position) and internal coordinates (granule size, porosity, saturation, etc). The vector \bar{v}_e accounts for external particle velocity as particles

enter and exit the vessel, while vector \bar{v}_i is the velocity vector with respect to internal coordinates such as a growth rate due to layering or consolidation. Finally, \dot{b} and \dot{d} are birth and death terms resulting from agglomeration and breakage.

While a large number of papers addressing the modeling of particulate processes exists, the literature in the field of particulate process control is somewhat limited. Zhu et al. used a linear model predictive control strategy based of spatially discretized PBE models for the stabilization of oscillating yeast cultures [2]. Mantzaris and Daoutidis used nonlinear feedback control laws to control different moments of the cell mass distribution using PBE models [3].

The lack of adequate online particle size measurement techniques has previously limited the practice of feedback control approaches in particulate processing. Feedback implementations have employed indirect methods for particle sizing based on both moisture [4], [5] and impeller torque measurements [6], [5]. From these readily available system characteristics, particle size can be inferred. However, due to the emerging technology in obtaining online particle size measurements, feedback control based on direct measurements in the granulation industry is becoming more and more feasible. Such measurement techniques include laser-based online chord length estimates as well as high-speed CCD cameras with online image processing [7].

Given that particle size distribution measurements are available online, this paper presents a Nonlinear Model Predictive Control (NMPC) formulation [8] for the control of a high-shear batch granulation system. This nonlinear method requires the solution to a nonconvex optimization problem at each time step. A variety of solution approaches have been utilized in the NMPC context. Local gradient-based search techniques have been implemented, but optimality is highly dependent on the choice of an initial guess. Rigorous deterministic methods can guarantee global optimality to the nonconvex problem, however typically prove to be computationally demanding [9], [10]. Alternatively, parametric methods can be used in which the solution space is partitioned and characteristic problems are solved offline [11]. The appropriate solutions can then be referred to by the controller. This can be suboptimal as the exact

problem is not necessarily solved at each point. In this work, the optimization problems are both nonconvex and non-smooth as a result of the PBE model, making gradient-based methods unsuitable. Therefore a stochastic search approach is implemented [12], [13]. Here, the solution space is randomly explored for the allotted time and the best solution is selected. Note that optimality cannot be guaranteed, however, parallelization strategies can be utilized to allow for further exploration of the solution space.

II. MODELING BACKGROUND

Population balance equations present many challenges for optimization and control of particulate processes. The first challenge is finding a suitable model for the process at hand. The PBE for granulation is a partial integro-differential equation. Solutions to this equation are not trivial. Few analytical solutions exist, and those that are available are confined to special cases [1], [14].

For most control applications, numerical solutions to the population balance equation are necessary. Many discretized solution techniques have been proposed (Gelbard and Seinfeld, 1980 [15], Hounslow et al., 1988 [16], Marchal et al., 1988 [17], Litster et al., 1995 [1], Vanni, 1999 [18], etc.). These numerical solution techniques typically separate particles into discrete size classes resulting in a series of ordinary differential equations, which can be solved using different solution techniques. A geometric discretization as opposed to an arithmetic discretization reduces the number of discrete size classes required using the z th root of the 2 series in terms of particle volume:

$$v_j = 2^{1/z} v_{j-1} \quad (2)$$

where v_j is the volume of the largest granule in the j th interval and z is an integer greater than 1. Using the geometric discretization, one may convert the general population balance from Equation 1 into a series of ordinary differential equations:

$$\begin{aligned} \frac{dN_i}{dt} = & N_{in_i} - N_{e_i} + \left(\frac{dN_i}{dt}\right)_{growth} \\ & + \left(\frac{dN_i}{dt}\right)_{aggregation} + \left(\frac{dN_i}{dt}\right)_{break} \end{aligned} \quad (3)$$

If the process is batch, $N_{in} = N_e = 0$. One may generally assume that growth due to layering, consolidation, and attrition are assumed negligible compared to the change in particle size due to aggregation and breakage.

Vanni [19] compared numerical solution methods for modeling aggregation as well as breakup in particulate processes. Vanni notes that Litster's numerical solution performed well, giving "accurate predictions even with a small number of nodes" [19]. It should also be noted that while other methods proved to be more robust (such as the approach by Gelbard), for complexity concerns which may arise during the controller formulation, the most simple numerical solution was chosen in this model. Litster et al. gives the discretized terms for coalescence as:

$$\begin{aligned} \left(\frac{dN_i}{dt}\right)_{agg} = & \sum_{j=1}^{i-S-1} \beta_{i-1,j} N_{i-1} N_j \frac{2^{(j-i+1)/q}}{2^{1/q}-1} \\ & + \sum_{k=2}^q \sum_{j=i-S-k+1}^{i-S-k} \beta_{i-k,j} N_{i-k} N_j \frac{2^{(j-i+1)/q} - 1 + 2^{-(k-1)/q}}{2^{1/q}-1} \\ & + \frac{1}{2} \beta_{j-q,i-q} N_{i-q}^2 \\ & + \sum_{k=2}^q \sum_{j=i-S-k+1}^{i-S-k+1} \beta_{i-k+1,j} N_{i-k+1} N_j \frac{1 - 2^{\frac{(j-i+1)}{q}} - 2^{\frac{-(k-1)}{q}}}{2^{1/q}-1} \\ & - \sum_{j=1}^{i-S} \beta_{i,j} N_i N_j \frac{2^{(j-i)/q}}{2^{1/q}-1} - \sum_{j=i-S+1}^{\infty} \beta_{i,j} N_i N_j \end{aligned} \quad (4)$$

where q is an integer greater than or equal to 1, N_i is the number of particles in the i th size interval, and $\beta_{i,j}$ is the coalescence rate constant, or kernel. The coalescence kernel will be discussed in more detail later in this paper. Computation time is decreased as q approaches 1. In this example, $q = 1$ is used. For this special case, two of the summations disappear reducing to the simpler equation used by Hounslow et al. [16].

For simplicity it is assumed that each breakage event produces two equally sized daughter particles. Therefore, the discretized term for breakage can be presented as [1]:

$$\left(\frac{dN_i}{dt}\right)_{break} = 2\Xi_{i+1} N_{i+1} - \Xi_i N_i \quad (5)$$

Ξ_i is the breakage kernel for particle i . In granulation processes, larger granules are more likely to break due to shearing forces. Due to this fact, breakage kernels for granulation processes are typically in the power-law form [1]:

$$\Xi_i = \Xi_0 N_i^y \quad (6)$$

where Ξ_0 is the size independent breakage rate constant and y is an integer.

A. The Coalescence Kernel

The coalescence kernel determines the rate at which particles of size i and size j coalesce. This is a key parameter for not only modeling, but also for control because the kernel must be able to take into account all the factors affecting agglomeration. Most coalescence kernels presented in literature are empirical in nature (Kapur and Fuerstenau, 1969 [20], Sastry and Fuerstenau, 1975 [21], etc.). These kernels rely on empirical constants which must be fit to laboratory data. Therefore, a kernel based on physical, non-empirical data would be beneficial. Furthermore, a kernel is needed that can provide several degrees of freedom, allowing for application in model based control methods. Both of these factors are addressed using the kernel proposed by Adetayo and Ennis (1998) which is consistent with granulation regime analysis [22]:

$$\beta(u, v) = \begin{cases} k_0, w < w^* \\ 0, w > w^* \end{cases} \quad \text{where } w = \frac{(uv)^a}{(u+v)^b} \quad (7)$$

here, w^* is the critical average granule volume. Successful collisions only occur when the average granule volume is less than the critical average granular volume. From Stokes regime analysis for non-deformable systems, $a = b = 1$ and w^* can be calculated from:

$$w = \frac{uv}{u+v} = w^* = \frac{\pi}{6} \left(\frac{16\mu}{\rho u_0} St^* \right)^3 \quad (8)$$

where μ is the liquid binder viscosity, ρ is the granule density, u_0 is the impact velocity, and St^* is the critical Stokes number which is shown in Eq 9.

$$St^* = (1 + 1/e) \ln(h_0/h_a) \quad (9)$$

e is the coefficient of restitution, h_a is the height of the surface asperities, and h_0 is the height of the surface binder layer. For granule growth in the non-inertial regime where $St \ll St^*$, the kernel simplifies to the size independent kernel. Growth only occurs in the non-inertial regimen and coalescence ends when the viscous Stokes number, St is larger than the critical value, St^* .

This kernel also provides an insight into how growth is influenced by dynamic granular quantities such as impact speed and granular liquid saturation. The granular velocity upon impact, u_0 can be assumed to be proportional to the impeller speed in a high shear mixer or the rotational speed in a low shear drum. This quantity can be altered during the granulation process. Analysis of Equations 7 and 8 show that as the impact velocity increases, the critical granule volume decreases threefold, which leads one to believe that as the kinetic to viscous force ratio increases the rate of coalescence would decrease. This value, u_0 , will be a primary process input in the model predictive control scheme as one of the few degrees of freedom in the process.

The second process input is expressed as in the ratio of the binder layer thickness to the surface asperity height, $h_r = h_0/h_a$. The assumption of a constant height of the surface asperities is often accurate as the size of the constituent particles do not change. Therefore, this ratio which determines the critical Stokes number is primarily a function of the binder layer height. Binder first appears on the surface of a granule when the pore saturation of a granule surpasses a critical value at which point the granule reaches the capillary state of pore saturation according to Equation 10:

$$h_0 = \begin{cases} \frac{D(v-\varepsilon s^*)}{6} & v > \varepsilon s^* \\ 0 & v < \varepsilon s^* \end{cases} \quad (10)$$

where D the granule diameter, v is the granule liquid volume fraction, ε is the granule porosity, and s^* is the critical granule pore saturation. If it is assumed that the granules are at the capillary state of saturation, $v > \varepsilon s^*$, then the binder layer height is proportional to the volume of binder in the process. Assuming that binder is equally dispersed throughout each granule and given that the porosity is defined as $\varepsilon = (l+a)/(l+s+a)$ and that the liquid

volume fraction is $v = l/(l+s+a)$ the granule volume can be rewritten as a function of the liquid binder height by:

$$l = \frac{as^* + 6h_0(s+a)/D}{1-s^* - 6h_0/D} \quad (11)$$

where l is the granule liquid volume, s is the granule solids volume and a is the volume of air. In this work the critical pore saturation is assumed to be 0.85 and the air fraction is assumed to be 0.10.

III. CONTROLLER FORMULATION

Recent works have focused on the actual control of granulation systems [23], [24], [25], [26]. of these, Pottmann et al. [25] and Gatzke et al [27] implemented a Model Predictive Control (MPC) approach.

MPC approaches rely on the ability to use current process data and a process model to predict how the system to be controlled will evolve over some future window of time. A review of MPC technology is presented by Morari and Lee [28] and by Henson in [8]. An appropriate constrained optimization problem is formulated at each time step using this information to determine the best control action to maintain ideal system operation.

Specifically, the MPC is formulated to choose a sequence of input moves over the move horizon (m) that minimizes some cost function. This cost function typically quantifies the difference between the model predicted evolution of the system and the desired setpoints over some prediction horizon (p). Typically, a 2-norm is used in the MPC objective function. The traditional objective function can be written as:

$$\Phi = \sum_{i=1}^p e(i)^T \Gamma_e(i) e(i) + \sum_{j=0}^{m-1} \Delta u(j)^T \Gamma_u(j) \Delta u(j) \quad (12)$$

where $e(i)$ is the difference between a state or output and its setpoint at a particular time given by:

$$e(i) = y(i) - y_{sp}(i) \quad (13)$$

where $y(i)$ is a modeled output value at a particular time step and $y_{sp}(i)$ is the corresponding setpoint value. The change in input position, $\Delta u(j)$ is a velocity term quantifying the difference in input positions for consecutive moves for each input channel.

$$\Delta u(j) = |u(j) - u(j-1)| \quad (14)$$

Both $\Gamma_e(i)$ and $\Gamma_u(j)$ are diagonal weighting matrices used to define the relative importance of each term.

For this particular system, 2 process inputs (u), 14 process states (x), and 2 process outputs (y) are considered. The first process input is the particle velocity (u_o) inside the granulator, which is assumed to be proportional to the mixing speed. The second process input is the binder layer thickness ratio (h_r), which is assumed to be proportional to the amount of binder that has been added to the system. The fourteen states define the number or fraction of particles in

each of the fourteen geometrically spaced volumetric bins of particles. While the current particle size distribution is assumed to be measured at each sampling time, the mean particle size (μ) and the particle size variance (σ^2) will be the target for control in this study.

The nonlinear granulation system can be run in batch mode for the purposes of particle enlargement. This leads to a batch implementation of the MPC which has been examined previously [29], [30], [31]. For this work, the nominal PBE system is acting as both the system to be controlled and the model. A shrinking horizon approach is used in which the controller projects state trajectories into the future from the current time step to the final batch time. This is opposed to looking over a window of a fixed length for the duration of the run. The objective function is then modified to:

$$\Phi = \sum_{i=t}^{t_{final}} e(i)^T \Gamma_e(i) e(i) + \sum_{j=0}^{m-1} \Delta u(j)^T \Gamma_u(j) \Delta u(j) \quad (15)$$

where t is the the current time and t_{final} is the duration of the batch. The resulting optimization problem to be solved at each time step is:

$$\min_{u(1) \dots u(m)} \Phi \quad (16)$$

and is subject to actuator constraints, $u_{min} \leq u(i) \leq u_{max}$.

IV. SOLUTION METHOD

The use of a nonlinear model to project state trajectories over the prediction horizon results in a Nonlinear Program (NLP) to be solved at each time step. The resulting NLP is inherently nonconvex and non-smooth, making deterministic gradient-based approaches to finding the global optimum difficult. Gradient-based approaches can still be utilized effectively in many cases, provided that an appropriate initial guess can be made. For this particular system, the hybrid nature of the coalescence kernel which has coalescence occurring only under some critical particle size results in both non-smoothness and flat regions in the objective function. Therefore, gradient based approaches can have difficulties in converging to a reasonable minima.

An alternative is a stochastic optimization approach. Banga et al. [12] utilized this approach in the MPC context. This is achieved here by randomly searching the feasible solution space for an allotted amount of time and then returning the best solution. The limitation lies in the ability to adequately search the entire feasible region of the parameter space. It is noted that no guarantee can be made on local or global optimality of the resulting solution.

One interesting aspect of this particular problem is the possibility of the continually shrinking feasible solution space. Since the binder added to the system cannot physically be removed, and since there is an upper bound on the amount of binder in the system at a given time, the stochastic search must only focus on feasible solutions that

have the binder layer thickness at the current time step $h_r(i)$ between that of the previous time step and the maximum allowable by the actuator limits such that:

$$h_r(i-1) \leq h_r(i) \leq h_{r,max} \quad (17)$$

This implies that if the binder layer thickness reaches its maximum, then the controller no longer has this degree of freedom and no additional binder can be added to the system. It is also important to note that in implementing this stochastic solution approach, the solver must be forced to evaluate the cost of not moving the system in addition to randomly searching the solution space. From a practical viewpoint, this is necessary when the controller is already operating optimally, so that the controller always has the option of not changing the inputs. Relying on the purely random search might not provide the controller this opportunity and as a consequence, the controller may move the system away from the current position.

Considering a sampling rate of 1 sample / minute and choosing a small move horizon ($m = 1$) results in relatively few degrees of freedom. The stochastic approach can be implemented in a sequential fashion where a single processor enumerates as many possible input scenarios in the allotted time. The obvious drawback is in failing to adequately search the solution space, and therefore unnecessarily implementing a suboptimal solution.

For real-time application in cases of higher dimensionality in order to ensure the optimal solution, the sequential stochastic approach may not be adequate. Thus, a parallel approach can be taken. This control formulation lends itself well to parallelization as the particular optimization problem formulated can be distributed to numerous processors that can individually enumerate many possible solutions. Additionally, a multi-step approach to parallelization may be used to limit the search space after an initial stochastic search period.

V. CLOSED-LOOP RESULTS

Consider a large batch of particles with radii normally distributed about a mean (μ) of 0.45 mm. Negative particle radii are removed from consideration. These particles are placed in a set of 14 geometrically spaced bins by volume. According to the discrete time PBE model, it is assumed that all particles in a given bin are of equal volume. The resulting distribution effectively approximates the normally distributed set of particles of $\mu = 0.45$ mm. The ideal initial conditions for the fourteen bins are given as:

$$x(0) = \begin{bmatrix} 0.001 & 0.001 & 0.004 & 0.017 & 0.065 & 0.206 & 0.385 \\ 0.276 & 0.044 & 0.001 & 0.000 & 0.000 & 0.000 & 0.000 \end{bmatrix}$$

The bin sizes range from particle radii of 114 microns to 2.3 mm. The nominal operating specifications of the system are taken as a 50 minute batch carried out with both a constant mixing speed and binder level. The mixing speed is set such that the particle velocity is maintained at 5 m/s. The binder layer thickness ratio is set to 5. An

open-loop simulation under these conditions is performed and the resulting evolution of the distribution, mean particle size, and particle size variance will act as the ideal setpoint trajectory for all simulations.

To test the capabilities of the proposed controller, two closed-loop runs are carried out in which the initial distribution of particles has either a higher or lower mean than typically used in the nominal operating conditions. The initial conditions for the simulation starting with a higher than normal mean are:

$$x_H(0) = \begin{bmatrix} 0.000 & 0.001 & 0.002 & 0.008 & 0.039 & 0.149 & 0.370 \\ 0.351 & 0.077 & 0.003 & 0.000 & 0.000 & 0.000 & 0.000 \end{bmatrix}$$

The initial conditions for the simulation starting with a lowered mean are given by:

$$x_L(0) = \begin{bmatrix} 0.001 & 0.003 & 0.009 & 0.031 & 0.099 & 0.257 & 0.375 \\ 0.202 & 0.024 & 0.000 & 0.000 & 0.000 & 0.000 & 0.000 \end{bmatrix}$$

The granulator is first charged with a set of particles approximating a normal distribution with $\mu = 0.48 \text{ mm}$, which is slightly higher than the typical initial distribution with $\mu = 0.45 \text{ mm}$. The controller is tuned to have $m = 1$ and to only penalize the error associated with the particle mean. The choice to only penalize the particle mean is made due to the “gas-brake” nature of the control problem. Both inputs generally have the same effect on the outputs of the system. There are two methods by which to facilitate particle enlargement: either more binder can be added or the mixing speed can be reduced. Similarly, for particle size reduction, either the mixing speed can be increased or binder level in the system could be reduced. Note that binder level reduction is assumed impossible. With both process inputs working to drive the process in a single “direction”, controlling two outputs (both the mean and variance) could prove difficult with effectively one single input direction. Penalizing errors in both the variance and mean would allow the process to be run with a trade-off between tracking either the mean or variance without offset. Again, for purposes of this example, only the error associated with the mean is penalized. Note that no penalty is placed on input movements or changes in the input values.

Closed-loop output trajectories and the corresponding input trajectories are shown in Figure 1. Here it is shown that the controller is able to force the system into providing a batch of particles with the desired mean. The mean particle size at the completion of the batch was 0.873 mm as compared to the target of 0.864 mm . However, this is done with some sacrifice to increased particle variation. The resulting particle variance was 0.088 while the target was 0.045 . It should be noted that the process inputs reached constraints for portions of the run. At the end of the run, the binder level was increased to its upper limit. The only degree of freedom remaining was the mixing speed. At this point, the controller becomes more aggressive as the prediction horizon is continually shrinking, leaving the remaining error of great importance as the system has only a few remaining

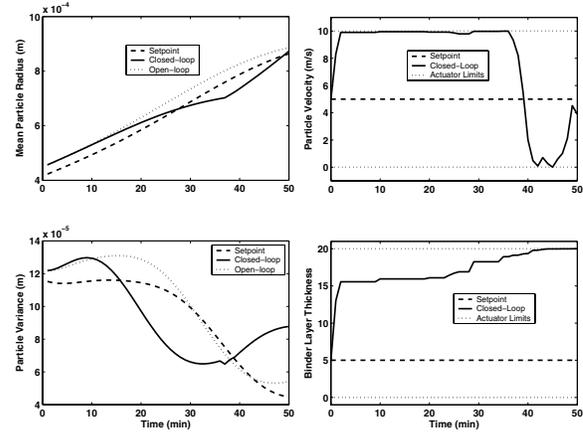


Fig. 1. Closed-loop results of the NMPC batch granulation system determined using the stochastic solution method for the case with an initial distribution that has a larger mean particle size as compared to the ideal batch run.

input moves to drive the error down. Snapshots of the distributions are provided in ten minute intervals in Figure 3. From this, it can be seen that the particle mean trajectory is being closely followed, however the particle size variance is changing significantly.

For the second example, the granulator is charged with a set of particles approximating a normal distribution with $\mu = 0.43 \text{ mm}$, which is slightly lower than the typical initial distribution with $\mu = 0.45 \text{ mm}$. The controller is again tuned to have $m = 1$ and to only penalize the error associated with the particle mean. Again the controller is able to push the system to the desired target mean. Despite

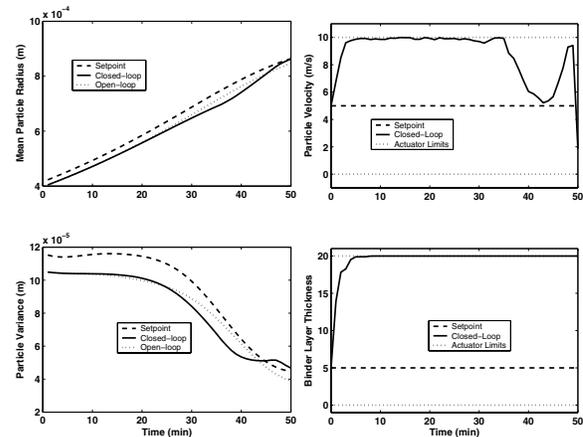


Fig. 2. Closed-loop results of the NMPC batch granulation system determined using the stochastic solution method for the case with an initial distribution that has a smaller mean particle size as compared to the ideal batch run.

starting at a different initial distribution, the closed-loop batch run provided a particle mean of 0.863 mm compared to the target of 0.864 mm , and did so without sacrificing the variation in particle size as seen in Figures 2. This run

resulted in a particle size variance of 0.047 while the target was 0.045. Again, snapshots of the particle size distribution in ten minute intervals are provided in Figure 3.

VI. CONCLUSIONS

A simple population balance equation is presented to describe a simple granulation process for particle size enlargement. The hybrid nature of the PBE system proved to cause gradient-based batch model predictive control approaches to struggle as a result of both flat and non-smooth objective functions. A stochastic approach was taken in order to solve the resulting NLP. The stochastic implementation of the batch NMPC was then able to track the mean particle size in fixed length batch runs in which the initial distribution was not necessarily equivalent to the ideal batch run.

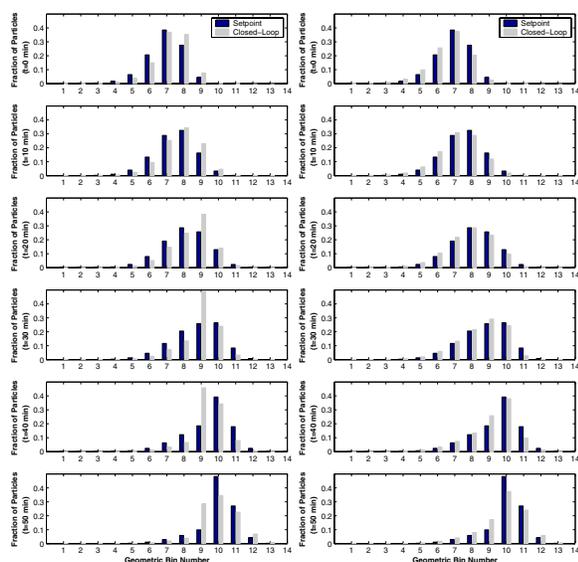


Fig. 3. Snapshots of the particles size distributions in ten minute intervals in the run in which the initial distribution has a higher than typical particle mean (left) and for the run in which the initial distribution has a lower than typical mean (right).

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REFERENCES

- [1] J. Litster and B. Ennis, *The Science and Engineering of Granulation Processes*. Kluwer Academic Publishers, 2004.
- [2] G. Zhu, A. Zamamiri, M. Henson, and M. Hjortso, "Model predictive control of continuous yeast bioreactors using cell population balance models," *Chemical Engineering Science*, vol. 55, pp. 6155–6167, 2000.
- [3] N. Mantazaris and P. Daoutidis, "Cell population balance modeling and control in continuous bioreactors," *Journal of Process Control*, vol. 14, pp. 775–784, 2004.
- [4] T. Schaefer and O. Worts, "Control of Fluidized Bed Granulation V: Factors Affecting Granule Growth," *Arch. Pharm. Chem. Sci.*, no. 6, pp. 1–13, 1978.
- [5] A. Goldszal and J. Bousquet, "Wet Agglomeration Of Powders: From Physics Toward Process Optimization," *Powder Technology*, vol. 117, pp. 221–231, 2001.

- [6] I. Talu, G. I. Tardos, and J. R. V. Ommen, "Use Of Stress Fluctuations To Monitor Wet Granulation Of Powders," *Powder Technology*, vol. 117, pp. 149–162, 2001.
- [7] S. Watano, T. Numa, K. Miyanami, and Y. Osako, "A Fuzzy Control System Of High Shear Granulation Using Image Processing," *Powder Technology*, vol. 115, pp. 124–130, 2001.
- [8] M. A. Henson, "Nonlinear Model Predictive Control: Current Status and Future Directions," *Computers and Chemical Engineering*, no. 23, pp. 187–202, 1998.
- [9] C. E. Long and E. P. Gatzke, "Globally Optimal Nonlinear Model Predictive Control," in *DYCOPS 7, the 7th International Symposium on Dynamics and Control of Process Systems*, (Boston, MA), 2004.
- [10] G. R. Srinivas and Y. Arkun, "A Global Solution to the Nonlinear Model Predictive Control Algorithm Using Polynomial ARX Models," *Comput. Chem. Eng.*, vol. 21, no. 4, pp. 431–439, 1997.
- [11] J. A. Rossiter and P. Grieder, "Using Interpolation to Improve Efficiency of Multiparametric Predictive Control," *Automatica*, vol. 41, pp. 637–643, 2005.
- [12] J. R. Banga, R. Irizarry-Rivera, and W. D. Seider, "Stochastic Optimization for Optimal Model-Predictive Control," *Computers and Chemical Engineering*, vol. 22, no. 4/5, pp. 603–612, 1998.
- [13] B. Kouvaritakis, M. Cannon, and V. Tsachouridis, "Recent Developments in Stochastic MPC and Sustainable Development," *Annual Reviews in Controls*, vol. 28, no. 1, pp. 23–35, 2004.
- [14] A. Randolph and M. Larson, *Theory of Particulate Processes, Volume 2*. Academic Press, 1991.
- [15] F. Gelbard and J. Seinfeld *Journal of Colloid and Interface Science*, vol. 68, no. 2, pp. 363–382, 1979.
- [16] M. J. Hounslow, R. Ryall, and V. Marshall *AIChE Journal*, vol. 34, p. 1821, 1988.
- [17] P. Marchal, R. David, J. Klien, and J. Villermaux *Chemical Engineering Science*, vol. 43, p. 59, 1988.
- [18] M. Vanni *AIChE Journal*, vol. 45, p. 916, 1999.
- [19] J. G. V. Antwerp and R. D. Braatz, "Model Predictive Control of Large Scale Processes," *Journal of Process Control*, vol. 10, pp. 1–8, 2000.
- [20] P. Kapur and D. Fuerstenau *I. and E. Proc. Des. and Dev.*, vol. 8, no. 1, p. 56, 1969.
- [21] K. Sastry and D. Fuerstenau *Int. J. Miner. Process*, vol. 2, p. 187, 1975.
- [22] A. Adetayo and B. Ennis, "A Unifying Approach to Modeling Coalescence Mechanisms," *AIChE Journal*, vol. 43, pp. 927–934, 1997.
- [23] M. Bardin, P. C. Knight, and J. P. K. Seville, "On Control of Particle Size Distribution in Granulation Using High-shear Mixers," *Powder Technology*, vol. 140, pp. 169–175, 2004.
- [24] F. Y. Wang and I. T. Cameron, "Review and Future Directions in the Modelling and Control of Continuous Drum Granulation," *Powder Technology*, vol. 124, pp. 238–253, 2002.
- [25] M. Pottmann, B. A. Ogunnaiké, A. A. Adetayo, and B. J. Ennis, "Model-based Control of a Granulation System," *Powder Technology*, vol. 108, 2000.
- [26] A. Faure, P. York, and R. C. Rowe, "Process Control and Scale-up of Pharmaceutical Wet Granulation Processes: A Review," *European Journal of Pharmaceutics and Biopharmaceutics*, vol. 52, pp. 269–277, 2001.
- [27] E. P. Gatzke and F. J. D. III, "Multi-Objective Control of a Granulation System," *Journal of Powder Technology*, vol. 121, no. 2, pp. 149–158, 2001.
- [28] M. Morari and J. H. Lee, "Model Predictive Control: Past, Present, and Future," *Computers and Chemical Engineering*, vol. 23, pp. 667–682, 1999.
- [29] K. S. Lee and J. H. Lee, "Model Predictive Control for Nonlinear Batch Processes with Asymptotically Perfect Tracking," *Chem. Eng. Educ.*, vol. 21, pp. S873–S879, May 1997.
- [30] L. S. Balasubramhanya and F. J. D. III, "Nonlinear Model-based Control of a Batch Reactive Distillation Column," *Journal of Process Control*, vol. 10, pp. 209–218, 2000.
- [31] D. C. M. Silva and N. M. C. Oliveira, "Optimization and Nonlinear Model Predictive Control of Batch Polymerization Systems," *Computers and Chemical Engineering*, no. 26, pp. 649–658, 2002.