Control of Particle Formation Processes
Eric Otto * Jessica Behrens ** Robert Dürr ** Stefan Palis ***
Achim Kienle ***
* Otto-von-Guericke-University Magdeburg, D-39106 Magdeburg
** Max-Planck-Institute for Dynamics of Complex Technical Systems,
D-39106 Magdeburg
*** Moscow Power Engineering Institute, Moscow, Russia

Abstract: Discrepancy-based control (DBC) represents an elegant direct control design concept
for infinite-dimensional systems, which is frequently used to control particle formation processes,
an important class of operations in chemical engineering. So far, DBC has been applied to
continuous layering growth and agglomeration, which have been accounted for by partial
differential equations. In this contribution we develop control laws for infinite systems of ordinary
differential equations, inspired by DBC. The results are visualized by the application to a
continuous agglomeration process involving additional particle breakage. We show how a two-
dimensional controller is able to control the industrially relevant Sauter mean diameter of the
product particles, resulting in improved performance of the closed-loop system compared to a
single control loop.

Keywords: Population Balance Modeling, Particle Formation, Agglomeration, Breakage,
Discrepancy-based control

1. INTRODUCTION

Particle formation processes such as granulation, polymer-
ization, and crystallization are widespread unit operations
in industrial processes, producing versatile goods, such
as pharmaceuticals, fertilizers, synthetics, food powders
and many more. Those formation processes have the same
mechanistic subprocesses in common, such as nucleation,
growth, agglomeration (or aggregation, coagulation) and
breakage (or fragmentation) and can therefore be treated
similarly from an engineering perspective. The resulting
particle collectives are characterized by distributions of
relevant particle properties such as size and shape as
well as composition for polymers, porosity for granules
and crystallinity for crystals. The production of particles
with desired product properties is a common goal of the
processes, since these properties define the quality of the
manufactured product, which also explains the high re-
search interest in this direction (Litster and Ennis, 2013;
Crowley et al., 2000; Ramkrishna and Mahoney, 2002).

A powerful tool to ensure the desired product quality on
the one hand and to optimize the production process on
the other hand is model-based process control. In order to
implement advanced control strategies, suitable dynamical
process models are required. An established framework is
population balance modelling (Ramkrishna, 2000), where
the process dynamics are usually expressed by nonlinear
partial differential equations (PDEs) or less often by high-
dimensional systems of ordinary differential equations
(ODEs). The formulation as PDEs makes control tasks
challenging, since there are no standard feedback control
methods available for such nonlinear infinite-dimensional
problems. Nevertheless, a variety of approaches is pre-
sented in the literature. Since this contribution focuses
on the particle size as one of the most important particle
properties, the following overview will be restricted to
time of control of particle size distributions. However, most
of the methods can be applied to any distributed particle
property.

The aim of this contribution is to design a control law for
an infinite system of ODEs, inspired by the discrepancy-
based control approach and to apply it to a particle
formation process. An extended and discrete version of
the population balance example from Otto et al. (2021) is
used as an example process. Finally, it is shown that the
control methodology can be used to control an industrially
relevant particle property.

The remainder of this paper is structured as follows: In the
second section, the population balance equation (PBE) for
the agglomeration and breakage process is presented and
the control design procedure is introduced briefly. In the
third section, the control laws are derived and verified by
simulation. The last section summarizes the results and
gives an overview of further possible research directions.

2. METHODS

In this section a simple process model, including particle
agglomeration and breakage, is presented. Furthermore,
the control design procedure based on the discrepancy-
based control approach is reviewed briefly.

2.1 Process Model

As an example for a particle process a continuous ag-
lomeration process is chosen. It is an industrial process
commonly used to increase the size of particles. During
an agglomeration process, which is often performed in a
The agglomeration term is given by
\[ A_i(t) = \frac{1}{2} \sum_{j=1}^{i-1} a_{i,j} n_j(t) n_{i-j}(t) - n_i(t) \sum_{j=1}^{\infty} a_{i,j} n_j(t) \] (4)
where \( a_{i,j} \) is the agglomeration kernel which represents the rate of agglomeration for particle pairs with volume \( v_i \) and \( v_j \). For more information regarding the agglomeration term the readers are referred to Narni et al. (2012).

The rate of breakage for a particle with volume \( v_i \) is given by
\[ B_i = \sum_{j=1}^{\infty} b_{i,j} s_j n_j - s_i n_i. \] (5)
Here, \( s_j \) denotes the selection rate, which describes the rate at which a particle of volume \( v_j \) breaks into smaller fragments. The breakage kernel \( b_{i,j} \) denotes the probability that a particle of volume \( v_j \) breaks into a particle of volume \( v_i \). Since breakage is assumed to be binary and volume-conserving, the second particle has volume \( v_{j-i} \). Since it is desirable to only withdraw particles from the process that exceed a specified volume, the particle outlet is assumed to be classifying with a separation function \( T_i(t) = T(v_i) \). Here, we choose \( T \) to be a cumulative Gaussian function with mean value \( \mu_s \) and variance \( \sigma_s \). The outlet term is then given by
\[ O_i(t) = K(t) T_i n_i \] (6)
with the withdrawal rate \( K(t) \). It is assumed that \( K \) is a plant parameter that can be manipulated freely.

It is worth mentioning that the presented process model represents the discrete version of the continuous one proposed in (Otto et al., 2021).

2.2 Control Design Method

Discrepancy-based control is a stability-based control approach for PDEs (Movtsesan, 1960; Sirazetdinov, 1967). It relies on a stability notion where the “distance” between the state of a process and its equilibrium is not measured using metrics but a generalized distance measure, the so-called discrepancy. In the finite dimensional case, the discrepancy can be interpreted simply as system output and can be stabilized (asymptotically) using control Lyapunov functions. The distribution is then (asymptotically) stable if the zero-dynamics are (asymptotically) stable.

In Palis and Kienle (2012); Geyyer et al. (2017) and Otto et al. (2021), moments of particle size distributions have been shown to be useful control variables. The \( k \)-th moment of a particle size distribution is defined as
\[ \mu_k(t) = \int_0^\infty x^k n(t, x) \, dx \] (7)
for continuous variables \( x \). If \( x \) is the particle volume \( v \), the zeroth moment is proportional to the total number of particles \( N_{t,0} \), the first moment is proportional to the total particle volume \( V_{t,0} \) and the moment with \( k = 2/3 \) is proportional to the total particle surface \( A_{t,0} \). In Otto et al. (2021), for example, the discrepancy
\[ \rho(n, t) = (\mu_{0,k} - \mu_0(t))^2 \] (8)
is utilized, where \( \mu_{0,k} \) is the desired steady state value of the zeroth moment. In order to apply the DBC-concept to
the discrete PBE formulation, we simply use the discrete moments
\[ \mu_k(t) = \sum_{i=1}^{\infty} x_i^k n_i(t) \]  
(9)
as system outputs and control variables. Then the control design effort is reduced to finding suitable Lyapunov functions and, if possible, showing stability of the zero-dynamics.

3. RESULTS

In the following, a two-dimensional control law for the PBE (Eqn. 2) is derived. Afterwards the control law is tested in a simulation scenario and discussed in terms of dynamics and convergence.

3.1 Derivation of the Control Law

Particle size distributions are often represented by mean values. In particular the Sauter mean diameter \( d_{32} \) is frequently used in industrial applications. It represents the mean diameter of a collection of particles of different sizes that is equal to the diameter of equisized spherical objects forming a collection with same total volume and same total surface. For the discrete particle size distribution, it can be defined as

\[ d_{32}(t) = \frac{6V_{\text{tot}}}{A_{\text{tot}}} = 6^{5/3} \pi^{2/3} \frac{\mu_1(t)}{\mu_{32}(t)} = 6^{5/3} \pi^{2/3} \frac{\sum_{i=1}^{\infty} v_i n_i(t)}{\sum_{i=1}^{\infty} v_i^2 n_i(t)} \]  
(10)

and therefore represents the ratio of the first moment \( \mu_1 \) to the 2/3th moment \( \mu_{32} \). Controlling the Sauter mean diameter as a measure of product quality therefore requires controlling both moments and a two-dimensional control law. It is derived using two commonly accessible parameters as manipulated variables: the withdrawal rate \( K(t) \) is used to control \( \mu_1 \) and the feed rate \( f(t) \) to control \( \mu_{32} \). It is intuitively clear that the total particle surface is highly sensitive to the rate at which particles with low diameter and high relative surface are fed to the process. Likewise, the total particle volume is more sensitive to the rate at which large particles are withdrawn, since a fixed number of large particles has a higher volume than a fixed number of smaller particles. The control errors are therefore defined by

\[ e_{2/3}(t) = \sum_{i=1}^{\infty} v_i^2 (n_{d,i}(t) - n_i(t)) \]  
(11)

\[ e_1(t) = \sum_{i=1}^{\infty} v_i (n_{d,i}(t) - n_i(t)) \]  
(12)

where \( n_{d,i} \) denotes the desired number distribution, which is assumed to be constant with respect to time. The following control design procedure consists of two steps: At first, a controller for \( \mu_{32} \) is derived. Afterwards, the resulting closed-loop system is used to calculate the controller for \( \mu_1 \). The final control structure is shown schematically in Fig. 2.

Fig. 2. Two-dimensional control scheme.

Now the following Lyapunov-functional is used to derive the first stabilizing controller:

\[ V_{2/3} = \frac{1}{2} e_{2/3}^2. \]  
(13)

Calculating its time derivative yields

\[ \dot{V}_{2/3} = -e_{2/3} \left( f(t) w + \sum_{i=1}^{\infty} v_i^2 (A_i + B_i - K T n_i) \right) \]  
(14)

By choosing

\[ f(t) = \frac{1}{w} \left( c_{2/3} e_{2/3} - \sum_{i=1}^{\infty} v_i^2 (A_i + B_i) \right) \]  
(15)

the time derivative of the Lyapunov-functional results in

\[ \dot{V}_{2/3} = -2c_{2/3} V_{2/3}, \]  
(16)

which shows that the controller exponentially stabilizes \( \mu_{32} \). The positive constant \( c_{2/3} \) is a tuning parameter for the convergence speed of control loop. Now the second control law for \( \mu_1 \) is derived using the following Lyapunov-functional

\[ V = \frac{1}{2} (e_{2/3}^2 + e_1^2). \]  
(17)

Calculating the time derivative of \( V \) and introducing equations (2) and (15) yields

\[ \dot{V} = -e_{2/3} e_{2/3} - e_1 \sum_{i=1}^{\infty} v_i \left( \frac{n_i}{w} \right) \left( c_{2/3} e_{2/3} - \sum_{j=1}^{\infty} v_j^2 (A_j + B_j) \right) + \sum_{i=1}^{\infty} v_i \left( -T_i n_i + \frac{n_i}{w} \sum_{j=1}^{\infty} v_j^2 T_j n_j \right) \]  
(18)

The choice

\[ K = \left[ \sum_{i=1}^{\infty} v_i \left( -T_i n_i + \frac{n_i}{w} \sum_{j=1}^{\infty} v_j^2 T_j n_j \right) \right]^{-1} \]  
(19)

results in

\[ \dot{V} = -c_{2/3} e_{2/3}^2 - c_1 e_1^2 \]  
(20)

and therefore asymptotic stability of the closed-loop system. The positive parameter \( c_1 \) is used again as tuning constant.
The presented control law derivation results in a decoupled control system, which is shown by the final control error dynamics

$$\dot{e}_{2/3} = -c_2/e_{2/3}$$ \hspace{1cm} (21)
$$\dot{e}_1 = -c_1 e_1,$$ \hspace{1cm} (22)

which are obtained by differentiating Equ. (12) and (11), and introducing equations (2), (15) and (19). In addition, the derivation is independent of the agglomeration and breakage kernel and can therefore be applied to a wide spectrum of particle population balance models.

3.2 Simulation

The control law is now applied to the process described in section 2.1. Before the simulation results are presented, the rate constants as well as the feed distribution and the separation function for the example process are given.

The normalized primary particle distribution is chosen as a decaying exponential function:

$$\tilde{n}_{i,t} = \frac{\exp(-v_i)}{\sum_{i=1}^{\infty} \exp(-v_i)}, \quad i \in \mathbb{N}.$$ \hspace{1cm} (23)

For the agglomeration kernel $a_{i,j}$, the discrete formulation of the well-known Kapur-kernel (Kapur and Fuerstenau, 1969)

$$a_{i,j} = a(v_i, v_j) = a_0 \left(\frac{v_i + v_j}{v_i v_j}\right)^{a_1}, \quad (i, j) \in \mathbb{N}^2$$ \hspace{1cm} (24)

with the agglomeration efficiency $a_0$ and two empirical parameters $a_1$ and $a_2$ is chosen.

Regarding particle breakage the selection function and breakage kernel are chosen as

$$s_j = s_0 v_j^{2/3}, \quad j \in \mathbb{N}$$ \hspace{1cm} (25)

and

$$b_{i,j} = \delta_{i,1} + \delta_{i,j-1}, \quad (i, j) \in \mathbb{N}^2$$ \hspace{1cm} (26)

respectively. Here, $\delta_{i,j}$ denotes the Kronecker delta. Equ. (26) reflects the fact that a particle of volume $v_j$ breaks into two fragments of volumes $v_i$ and $v_{j-1}$.

The system of ODEs given by Equ. (2) is solved using the ODE-solver ode15s within the MATLAB environment. For the simulation, $N = 300$ particle volumes are assumed. In the following a shift of the operating point is simulated both in open- and closed-loop operation. In the first case, the nominal withdrawal rate $K_{\text{nom}}$ is increased to $10K_{\text{nom}}$ at $t = 1$ while $f_{\text{nom}}$ stays constant. In the latter case, the inputs are determined by the controller and limited by $f_{\text{max}}$ and $K_{\text{max}}$ from above and by zero from below in order to establish typical practical constraints for such a control scenario. The initial particle size distribution for both scenarios is given by the steady state distribution for $f_{\text{nom}}$ and $K_{\text{nom}}$. The tuning parameters $c_1$ and $c_2/3$ were determined iteratively. The complete set of simulation parameters is given in Tab. 1. Note that all parameters and the process time are unitless, since the simulation example does not represent a specific process plant.

![Fig. 3. Time evolution of the first moment of the open-loop (black dashed line) and closed-loop system (blue solid line) for an operating point change](image)

![Fig. 4. Time evolution of the 2/3th moment of the open-loop (black dashed line) and closed-loop system (blue solid line) for an operating point change](image)

| Parameter | Value |
|-----------|-------|
| $f_{\text{nom}}$ | $1 \cdot 10^7$ |
| $f_{\text{max}}$ | $2 \cdot f_{\text{nom}}$ |
| $\beta_0$ | $1 \cdot 10^{-5}$ |
| $\alpha_0$ | $9 \cdot 10^{-8}$ |
| $\alpha_1$ | $0.1$ |
| $c_2/3$ | $1 \cdot 10^1$ |
| $v_0$ | $1$ |
| $K_{\text{nom}}$ | $2$ |
| $K_{\text{max}}$ | $15 \cdot K_{\text{nom}}$ |
| $\sigma_s$ | $30$ |
| $c_1$ | $1 \cdot 10^5$ |

Note that, while the moments are already in steady state at around $t = 1.5$, the manipulated variables, especially $K$, are not. This is due to the fact that while stabilizing the outputs, the control law does not guarantee stability of the distribution with respect to a norm. It follows, among other things, that $K$ and $n$ still change while the moments are already in the steady state. From a mathematical...
In order to further investigate this, the volume weighted $L^2$ norm of the error distribution

$$L^2_{2,w} = \sum_{i=1}^{\infty} (n_i(n_d,i - n_i))^2$$

is computed and presented in Fig. 6. It can be seen that the closed-loop process converges also with respect to this norm, i.e. the zero-dynamics of the closed-loop system are asymptotically stable for this example, however the speed of convergence is smaller for the distribution than for the moments. Finally, the $d_{32}$ is analyzed as it was our motivation to introduce a two dimensional control law. In Fig. 7, the closed-loop is compared to the open-loop scenario. Clearly, the Sauter mean diameter converges faster with control. In order to show that the introduction of a second control loop improves the performance, the evolution of $d_{32}$ where only the $\mu_{2/3}$-control-loop is closed is presented. The two-dimensional controller achieves better results than the one-dimensional controller with respect to convergence speed.

4. CONCLUSION

This contribution was concerned with control of particle formation processes that are modelled mathematically by infinite-dimensional systems of ODEs. The control strategy was inspired by discrepancy-based control and is based on the choice of moments as control variables. It has been shown that an industrially relevant particle property can be controlled by introducing a two-dimensional control law. Furthermore, numerical simulation results show that the particle size distribution is stabilized and that the time of convergence is improved significantly compared to the open-loop case.

Future research should be concerned with further analysis of the zero-dynamics of the system and the practical implementation of the proposed control algorithm at an actual plant. This includes investigation of controller performance under model-plant mismatch. Furthermore, the control approach can be generalized to process models which account for more than one particle properties, e.g. particle porosity.

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