Population balance modelling and $H_\infty$—controller design for a crystallization process

Ulrich Vollmer$^{a,*}$, Jörg Raisch$^{a,b}$

$^a$Max-Planck-Institut Dynamik komplexer technischer Systeme, 39106 Magdeburg, Germany
$^b$Lehrstuhl für Systemtheorie technischer Prozesse, Otto-von-Guericke-Universität Magdeburg, 39106 Magdeburg, Germany

Abstract

In this paper we consider the robustly stabilizing control of a 1000l draft tube baffled crystallizer. When operated at high fines dissolution rates, the crystallizer exhibits sustained oscillations. A detailed population balance model for the process can be found in the literature. Based on this detailed model we develop a simpler population balance model. This, in turn, permits the derivation of an irrational transfer function from manipulated to measured variable. An $H_\infty$ mixed sensitivity minimization problem is formulated and solved using an infinite-dimensional version of $H_\infty$ theory. Two different controllers are designed and compared in simulation studies.

© 2002 Elsevier Science Ltd. All rights reserved.

Keywords: Crystallization; Population balance; Process control; Infinite-dimensional system; Robust control

1. Introduction

Continuous crystallization from solution is a purification and separation process that is well established in the chemical industry. It is applied in large scale for the production of solids from solutions.

In an industrial crystallizer, crystals of different sizes are present at the same time. In many cases, the crystal size distribution (CSD) determines the product quality, since many physical properties of the crystalline product are closely related to the CSD. In addition, the efficiency of downstream processing such as filtering and drying is heavily influenced by the CSD. Furthermore, the dynamics of a crystallization process can only be captured adequately if the CSD is taken into account. Therefore, the CSD is the most important process variable. It changes in time due to several physical processes taking place in a continuous crystallizer such as nucleation, growth, breakage or attrition and removal of crystals.

The so-called population balance equations form an appropriate framework for the dynamic modelling of crystallization processes, see e.g. Ramkrishna (2000). Population balance models are capable of describing the dynamic behaviour of distribution functions with one or more internal coordinates, e.g. crystal size. Typically, population balance models consist of partial differential and integro-differential equations.

In industrial crystallizers, fines dissolution is commonly used to improve CSD properties, namely to shift the size distribution in the direction of larger particles. However, it is well known that continuous crystallizers can exhibit oscillatory behaviour, especially when operated at high fines dissolution rates, see e.g. Randolph and Larson (1988). Hence, improved CSD properties are being paid for by a deterioration of the dynamic process behaviour. In many cases, it is impossible to choose the fines dissolution rate as high as it would be necessary to achieve the desired CSD quality, because the process shows sustained oscillations and therefore the desired steady state is never attained. Stabilizing feedback control appears to be a promising tool to counteract the destabilizing effect of fines dissolution. By using feedback control, crystallizers can be operated at higher fines dissolution rates and thus superior product quality can be obtained.

Control of oscillatory crystallization processes is an area of active research, see Rawlings, Miller, and Witkowski (1993) for a review. Model-based controller design for crystallization processes is a challenging task, because the process models are of high complexity. Population balance models are of distributed nature and they are usually
In the literature, several different ways to deal with the model complexity problem have been reported. For the tuning of PID-type controllers no explicit plant model is needed at all, see e.g. Randolph, Beckmann, and Kraljevich (1977), Redman, Rohani, and Strathdee (1995). A finite-dimensional model obtained by system identification is used as a basis for controller design in Eek (1995) and Rohani, Haeri, and Wood (1999). Thus, no first principles model is needed in this approach. In Eek, Pouw, and Bosgra (1995), an infinite-dimensional population balance model is used as a starting point. The distributed model is first linearized and then lumped using the method of lines. Thus, a finite-dimensional approximation is obtained on which the subsequent LQG controller design is based. In Chiu and Christofides (1999a, b) the infinite-dimensional system is approximated using a combination of the method of weighted residuals and the concept of approximate inertial manifold. An input–output linearizing controller is designed based on the nonlinear, finite-dimensional approximation. Semino and Ray (1995) proved approximate controllability of a population balance model for a continuous crystallization process.

In this article, the idea of “late lumping” is pursued: controller design is based on the linearized distributed parameter model. This results in a controller that is itself infinite-dimensional. For implementation, the controller is approximated by a finite-dimensional transfer function. Thus, the system’s distributed nature is preserved as long as possible in the controller design process. The same approach has been successfully applied to an idealized crystallization model in Vollmer and Raisch (2001). This article is organized as follows. In Section 2, the draft tube baffled (DTB) crystallizer under consideration is briefly described. For this plant, a detailed population balance model was developed in Mitrovic (2001). Using this model as a starting point, a simpler population balance model is derived which, on the one hand, preserves the main effects of the original model. It takes into account size-dependent growth, attrition due to crystal-stirrer collisions, and fines dissolution, and it explicitly accounts for the effect of the annular settling zone. On the other hand, it is formulated such that it admits the derivation of an irrational transfer function that forms the basis for controller design. To obtain this transfer function the model is linearized and Laplace-transformed. The overall crystal mass (i.e. the third moment of the distribution) is considered the measured variable. The controller acts on the crystallizer by manipulating the fines dissolution rate. In Section 3, a robust performance problem is formulated based on this transfer function, taking into account multiplicative model uncertainty. The corresponding infinite-dimensional $H_{\infty}$ mixed sensitivity problem is solved using an $H_{\infty}$ synthesis method by Özay (1993); Foias, Özay, and Tannenbaum (1996). Finally, in Section 4, the effectiveness of the controllers is demonstrated in simulation studies.

2. Description of the process and model

2.1. The process

The process studied in this work is the evaporative crystallization of ammonium sulphate in a DTB crystallizer. A sketch of a crystallizer of this type is shown in Fig. 1. Crystal-free saturated solution is fed to the vessel at the bottom. The solution is supersaturated by evaporation of solvent. Due to supersaturation, crystals emerge from solution and existing crystals grow. The suspension is mixed by an impeller. Product is continuously withdrawn from the crystallizer. The crystallization vessel is surrounded by an annular settling zone. At the top of this zone suspension is withdrawn. Therefore, inside the annular zone there is a flow from the bottom to top. However, due to gravitation, only small particles arrive at the top. These fines are fed to a heat exchanger where they are dissolved. The clear solution is returned to the bottom of the crystallizer.

The Laboratory of Process Equipment at Delft University of Technology operates a 1000 l pilot plant crystallizer of this type. Operating conditions and parameter values for this plant can be found in Meadhra (1995); Jager (1990); Neumann, Bermingham, Kramer, and van Rosmalen (1999). In experiments, Eek (1995) and Bermingham, Neumann, Verheijen, and Kramer (1999) have found that this crystallizer exhibits sustained oscillations when operated at high fines dissolution rates.

Mitrovic (2001) developed a detailed population balance model that is capable of reproducing the oscillatory behaviour observed in the experiments with the Delft pilot plant crystallizer. This model includes the growth rate expression developed by Mersmann, Angelhöfer, Gutwald, Sangl, and Wang (1992) accounting for integration and diffusion limited growth. Attrition due to crystal-stirrer collisions are modelled based on the results of Gahn and Mersmann (1999a), for details on the attrition model see

![Fig. 1. Sketch of a draft tube baffled crystallizer.](image)
also Motz (2000) or Gerstlauer, Mitrovic, Motz, and Gilles (2001), where a model with two particle properties is developed. The model further includes homogeneous and heterogeneous nucleation. Fines dissolution is modelled using a separate population and mole balance for the annular settling zone, assuming that no growth, nucleation or attrition occurs in the annular zone. There is one particularity associated with fines dissolution in this model. The reflux stream from the fines dissolution unit is returned to the crystallizer vessel close to the lower part of the annular zone. Thus, hot undersaturated solution enters a region where, due to the hydrodynamic conditions, mainly large crystals accumulate. It is argued that this reflux stream leads to a local under-saturation and therefore the dissolution of large crystals present in that region.

The intention when developing this model clearly was to gain a more thorough understanding of the crystallization process under consideration and to obtain a model that is scalable and predictive. To achieve this, the physical phenomena taking place on a microscopic level where modelled in detail and incorporated in the overall process model. Great effort has been taken to make the model as physically meaningful as possible.

With respect to the objectives of scalability, predictiveness and physical meaning this model is excellent. However, for further analytic investigations such as computation of steady states and testing of their stability or as a basis for controller design, this model is by far too complex. For the latter purpose, the model of Mitrovic (2001) is used as a starting point to obtain a simplified model that serves as a basis for controller design. Thus, physical accuracy is traded off against model simplicity to make the problem tractable.

2.2. Derivation of a simple population balance model

The objective of this section is to obtain a model that is simple enough to make the controller synthesis problem tractable and whose dynamic behaviour is still, in some sense, close to the detailed model in Mitrovic (2001) and therefore also to the real plant. Furthermore, to preserve an intuitive understanding of the problem, it is desirable to maintain as much of the original model structure as possible, in particular, to preserve the physical interpretation of system states and thus the system’s distributed nature.

With these requirements in mind, a population balance model is developed. In a first step, the dispersed phase in the crystallizer vessel is modelled by a population balance equation (PBE) accounting for growth, attrition, outflow and exchange with the annular settling zone

\[ \frac{\partial f(L, t)}{\partial t} = -V \frac{\partial}{\partial L} [G(L, \Delta c)f(L, t)] + F_{\text{attr}}(L, t) - F_{\text{out}}(L, t) - F_{\text{AZ,in}}(L, t) + F_{\text{AZ,out}}(L, t) \]  

with boundary condition

\[ f(L = L_0, t) = \frac{B(t)}{G_{\text{eff}}(L_0, \Delta c)} \]  

where \( f(L, t) \) is the volume-based number density function in [\# of crystals/ crystal length/volume of suspension]. \( L \) is the characteristic crystal length, \( \Delta c \) is the supersaturation of the solution, i.e. the difference between actual concentration and saturation concentration of the liquid, \( V \) is the total volume of suspension in the crystallizer, \( L_0 \) is the smallest possible crystal length, \( B \) is the nucleation rate and \( G_{\text{eff}} \) is the effective crystal growth rate. Note that the symbols \( \dot{F} \) do not denote time derivatives but represent the various number density fluxes. The individual expressions in (1) and (2) are treated in detail below.

The dispersed phase in the annular zone is modelled by a separate PBE with the assumption of Mitrovic (2001) that no nucleation, growth or attrition occurs. Thus, only exchange with the crystallizer and flux to the fines dissolution unit are considered:

\[ V_{\text{AZ}} \frac{\partial f_{\text{AZ}}(L, t)}{\partial t} = \dot{F}_{\text{AZ,in}}(L, t) - \dot{F}_{\text{AZ,out}}(L, t) - \dot{F}_{\text{FD,in}}(L, t). \]  

The volume-based number density in the annular zone is denoted by \( f_{\text{AZ}} \), the volume of the settling zone is \( V_{\text{AZ}} \). The flux from the crystallizer to the annular zone is

\[ \dot{F}_{\text{AZ,in}}(L, t) = V_{\text{AZ,in}} \dot{f}(L, t). \]

The flow in the opposite direction due to back mixing is

\[ \dot{F}_{\text{AZ,out}}(L, t) = V_{\text{AZ,back}} h_{\text{back}}(L) f_{\text{AZ}}(L, t), \]

where the classification function \( h_{\text{back}}(L) \) accounts for the fact that all but the very small crystals are returned to the crystallizer. Small crystals are withdrawn from the annular zone to the fines dissolution unit at the rate

\[ \dot{F}_{\text{FD,in}}(L, t) = V_{\text{AZ,out}} h_{\text{AZ,FD}}(L) f_{\text{AZ}}(L, t). \]  

The volumetric flow rates from the crystallizer to the settling zone and from there to the fines dissolution unit are denoted by \( V_{\text{AZ,in}} \) and \( V_{\text{AZ,out}} \), respectively. \( V_{\text{AZ,back}} \) is a fictitious flow rate accounting for the back mixing of crystals due to gravitational settling. The classification function \( h_{\text{AZ,FD}}(L) \), accounting for the classifying effect of the settling zone, is different from zero only for the very small crystals.

At this point a model has been obtained that comprises two PBEs, one for the crystallizer (1) and one for the settling zone (3). The separation of fines is linked to the settling zone as it is in the real process. In order to simplify the model for the annular zone, from here on the separation of fines and the dynamics of the annular zone are treated completely separately. For the purpose of modelling the dynamics of the annular zone, the content of crystals in the flow to the fines dissolution unit is neglected, i.e. \( h_{\text{AZ,FD}}(L) = 0 \) and \( h_{\text{back}}(L) = 1 \). Of course, this is not true for very small crystals and therefore an error is associated with this assumption.
It is further assumed that the residence time in the annular zone is small compared to the time constants associated with crystal growth or attrition. Then, the settling zone can be regarded quasi-stationary and it follows from (3) that

$$F_{AZ, in} - F_{AZ, out} = 0$$

and therefore with (4) and (5)

$$f_{AZ}(L, t) = f(L, t).$$

With these simplifying assumptions the dynamics of the annular zone are captured adequately. But, of course, fines dissolution has been ignored. Therefore, fines dissolution is modelled independent of the settling zone:

$$\dot{F}_{FD, in} - \dot{F}_{FD, out} = 0$$

This term needs to be added to Eq. (1). The classification function

$$h_{FD}(L) = \left(1 - h(L - L_{FD}) \right)$$

accounts for the removal of small particles and the dissolution of large crystals due to the reflux of undersaturated solution, where $h(\cdot)$ is the unit step function, $L_{FD}$ is the fines dissolution cut size. Crystals larger than $L_{FD, large}$ are dissolved at a rate $k_{FD, large}$ times the fines dissolution rate due to the hot reflux stream. The classification function $h_{FD}(L)$ is plotted in Fig. 2.

Adding (1) and (3) using (8) and incorporating the fines dissolution term (9) yields the final version of the PBE:

$$(V + V_{AZ}) \frac{\partial f(L, t)}{\partial t} = -V \frac{\partial}{\partial L} \left[ G(L, \Delta c) f(L, t) \right] + F_{at}(L, t)$$

$$- \dot{F}_{out}(L, t) - \dot{F}_{FD}(L, t).$$

(11)

The liquid phase is modelled by a mole balance for the solute (substance A), yielding an integro-differential equation (IDE) for the solute concentration $c_{LA}$. The concentration in the settling zone can be considered to be quasi-stationary, with an argument analogous to the above. The IDE reads

$$\frac{dc_{LA}}{dt} = \frac{\dot{V}_{out}}{V + V_{AZ}} (c_{LA, in} - c_{LA})$$

$$- \frac{\dot{c}_{LA}}{\varepsilon} + \frac{\rho_{A} k_{F}}{\varepsilon M_{A} (V + V_{AZ})}$$

$$\times \left( \dot{V}_{FD} \int_{L_{0}}^{L_{\infty}} h_{FD}(L) f(L, t) L^{3} dL ight)$$

$$- 3V \int_{L_{0}}^{L_{\infty}} G(L, \Delta c) f(L, t) L^{2} dL.$$

(12)

with the volumetric shape factor $k_{F}$, the molar mass $M_{A}$ and density $\rho_{A}$ of substance A in crystalline form. This IDE accounts for in- and outflow, increase of concentration due to fines dissolution and the consumption of solute due to crystal growth. For the derivation of the individual terms the reader is referred to the standard literature, e.g. Randolph and Larson (1988). The volume fraction $\varepsilon$ is defined as

$$\varepsilon = \frac{V_{\text{liquid}}}{V} = 1 - k_{F} \int_{L_{0}}^{L_{\infty}} f(L, t) L^{3} dL.$$

(13)
2.3. Growth and attrition kinetics

In the following, the remaining terms appearing in the PBE (11) and the boundary condition (2) are explained in detail. All parameter values are listed in Table 1. They are obtained in three different ways: they can either be taken directly from the description of the physical system in Bermingham et al. (1999) or they are determined by curve fitting to approximate the kinetics used in the detailed model in Mitrovic (2001). The third way parameters are chosen is to match certain steady-state results of the detailed model. These steady-state values are taken from simulations of a model (see Mitrovic, 2001) that does not consider the dissolution of large particles due to the reflux of undersaturated solution from the fines dissolution unit as discussed in Section 2.1. This model is stable and so a steady state is achieved in simulations. Parameters are chosen such that the overall crystal mass, the supersaturation and the maximum crystal length of the simple model derived here are in agreement with the detailed model, where in both cases the dissolution of large particles is disregarded. Then, adding the dissolution of large crystals makes both models oscillate.

The growth rate $G$ is assumed to be of separated form and the dependence on crystal length $L$ to be piecewise linear:

$$G(L, \Delta c) = k_g \Delta c \cdot \left( \frac{L(1-h(L-L_{\text{growth}}))}{L_{\text{growth}}} + h(L-L_{\text{growth}}) \right).$$

The values for $L_{\text{growth}}$ and $h$ are determined by curve fitting with the growth law of Mersmann et al. (1992); the parameter $k_g$ is chosen such that the overall crystal mass (i.e. the third moment of the distribution) during steady state in the simplified model matches with simulations of Mitrovic (2001). A plot of the original and approximated growth rates is shown in Fig. 3.

A detailed model for attrition due to crystal-stirrer collisions based on the results of Gahn and Mersmann (1999a), as used in Motz (2000); Mitrovic (2001); Gerstlauer et al. (2001) comprises three terms: a sink term $F_{\text{attr}}^+$ due to the destruction of large crystals that collide with the stirrer, a source term $F_{\text{attr}}^+$ accounting for the generation of crystals that are slightly smaller than the original crystal and a source term $F_{\text{attr},1}^+$ describing the generation of a distribution of a large number of small fragments, see Fig. 4.

For the use in this study, this detailed attrition model is approximated by a simpler one. The source–sink combination $F_{\text{attr},2}^+ - F_{\text{attr}}^-$ is replaced by a negative growth rate

$$G_{\text{attr}}(L) = k_g, \text{attr}(L + 3(L - L_{\text{attr}})h(L - L_{\text{attr}})).$$

$G_{\text{attr}}(L)$ is a continuous, piecewise linear function with a low slope for $L < L_{\text{attr}}$. For crystals that are larger than $L_{\text{attr}}$ the negative growth becomes much faster, see Fig. 5. This shape of $G_{\text{attr}}(L)$ qualitatively captures the fact that large particles are abraded at a much higher rate than small ones. The length $L_{\text{attr}}$ where the corner occurs is chosen somewhat arbitrarily in the middle of the possible size range. The growth coefficient $k_g, \text{attr}$ is determined such that the effective steady-state growth rate $G_{\text{eff}}(L, \Delta c_{\text{eff}}) = G(L, \Delta c_{\text{eff}}) + G_{\text{attr}}(L)$.
Fig. 5. Negative growth rate due to attrition (left), effective growth rate at steady state (right).

(Fig. 5) becomes zero for the maximum crystal length at steady state obtained in simulations by Mitrovic (2001). The negative growth rate \( G_{\text{attr}}(L) \) determines the attrition term in (11) to be

\[
\dot{F}_{\text{attr}}(L,t) = -V \frac{\partial [G_{\text{attr}}(L) f(L,t)]}{\partial L}.
\]

The source term accounting for the production of small attrition fragments is modelled as a boundary condition for the PBE. Other forms of nucleation (primary nucleation) are negligible compared to the formation of attrition fragments as long as a sufficient amount of crystals are present in the process. Hence, secondary nucleation due to attrition is the only source of nuclei included in this model. Of course, this model is therefore not capable of describing the startup of the process in a situation where, initially, the crystallizer is clear of particles. All attrition fragments are assumed to emerge at the same length \( L_0 \). Since there are no crystals smaller than that, \( L_0 \) is the left boundary of the possible size range. The production rate of attrition fragments according to Mitrovic (2001) can be approximated by a term proportional to the fourth moment of the distribution (see Fig. 6).

Hence, the nucleation rate is

\[
B(t) = k_b \int_{L_0}^{L_{\infty}} L^4 f(L,t) \, dL.
\]

The nucleation coefficient \( k_b \) is determined such that the steady-state concentration of the simplified model matches with the value in the detailed model by Mitrovic (2001).

The process under consideration is an evaporative crystallizer. However, the production of supersaturation by evaporation of solvent is not modelled explicitly, here. Instead, the value of the inlet flow rate is decreased by the amount of the vapour outflow and the feed concentration is increased accordingly, such that the inlet mole flux of solute is maintained.

This concludes the derivation of the population balance model. In Fig. 7 the mass median crystal size \( L_{50} \) from a simulation of this model is plotted together with a measurement result for the Delft crystallizer by Bermingham et al. (1999) using the same operating conditions. The transient behaviour in the beginning is different due to different initial size distributions (the initial condition is not given in the cited reference). However, it can be seen that frequency and amplitude of the sustained oscillations in simulation and experiment match quite well.
3. Controller design

3.1. Derivation of plant transfer function

In contrast to a detailed model including the growth and attrition kinetics according to Mersmann et al. (1992) and Gahn and Mersmann (1999a, b), the steady-state solution for the simplified model presented here can be obtained analytically from the model equations by setting time derivatives equal to 0. The stationary size distribution

\[ f_{ss}(L) = Af_{ss,n}(L) = A \exp \left( -\int_{L_0}^{L} \frac{V_{out} + h_{FD}(\xi)}{V_{FD,\text{out}}/V + \bar{c}G_{\text{eff}}(\xi, \Delta c_{ss})} \, d\xi \right) \]

is the analytic solution of the ordinary differential equation (ODE) resulting from (11). The constant \( A \) would normally be determined by the boundary condition. However, in this case (2) yields

\[ A = k_b \frac{\int_{L_0}^{L} L^4 \, dL}{G(L_0, \Delta c_{ss}) + G_{\text{ate}}(L_0)}. \]

The constant \( A \) cancels and therefore it is not possible to determine it from the boundary condition. Instead, the stationary supersaturation \( \Delta c_{ss} \) can be obtained from this equation. This, in consequence, means that the steady-state supersaturation is independent of the inlet concentration: if the inlet concentration is increased, the steady-state concentration does not change, but the overall amount of crystals increases. The missing constant \( A \) can be obtained from the mole balance for the liquid phase (12) with time derivative set equal to 0.

To serve as a basis for controller synthesis, the model (11), (2), (12) is linearized with respect to the desired steady state (18). Via Laplace transformation an ODE with independent variable \( L \) is obtained from the linearized PBE (11). This ODE can be solved analytically. With this solution and the linearized and Laplace transformed IDE (12), a SISO transfer function \( P(s) \) from the manipulated variable (fines dissolution rate \( V_{FD}(s) \)) to the measured variable (third moment of the distribution \( m_3(s) \)) is obtained

\[ \Delta m_3(s) = P(s) \Delta V_{FD}(s) \]

\[ = \frac{Q P_1(s)}{Q P_2(s)} \Delta V_{FD}(s), \]

where \( \Delta m_3(s) \) and \( \Delta V_{FD}(s) \) denote the deviations from the respective steady-state values. For details on the computations required for this derivation see Appendix A. Both, numerator and denominator of this transfer function are so-called quasi-polynomials

\[ Q P_1 = \sum_k P_k(s) e^{-\xi_0 s}, \quad i = 1, 2 \]

with polynomials \( P_k(s) \). Quasi-polynomials have infinitely many zeros. Hence, the transfer function \( P(s) \) has infinitely many poles and zeros, which reflects the fact that the system is infinite-dimensional. A bode plot of the plant transfer function \( P(s) \) is shown in Fig. 8.

3.2. Control problem formulation

The reason for designing a controller for the crystallization process considered here, is to stabilize the unstable steady state. Furthermore, it is desirable to minimize the influence of disturbances \( v \), e.g. a nonconstant feed concentration or temperature changes on the output \( y \). In this study the overall crystal mass was chosen as the measured system output. Since the controller design is based on a simplified model, the controller needs to be robust with respect to plant model mismatch. Therefore, the controller is deliberately designed to guarantee stability and disturbance attenuation not only for the nominal plant model \( P(s) \) but also for a set of transfer functions containing \( P(s) \):

\[ \mathcal{P}_\text{m} = \{ (1 + A_m(s))P(s) : |A_m(j\omega)| < |W_m(j\omega)|, \]

\[ P(s) \text{ and } (1 + A_m(s))P(s) \text{ have the same number of right half plane poles} \}, \]

where \( W_m(s) \) is a frequency dependent error bound. This is referred to as “multiplicative model uncertainty” (Fig. 9).

![Fig. 8. Bode plot of plant transfer function P(s).](image)

![Fig. 9. System with multiplicative uncertainty.](image)
Often, plant models are quite accurate at low frequencies but are rather uncertain in the high-frequency range. In particular, several parameters for the model derived in Section 2.2 were adapted to meet requirements at steady state, i.e. for \( \omega \to 0 \). In addition, the annular settling zone was considered quasi-stationary, neglecting fast dynamics. Therefore, the model is expected to be “good” for low frequencies but rather inaccurate in the high-frequency range.

This is taken into consideration by choosing \( |W_m(j\omega)| \) to be small at low values of \( \omega \) and large for high frequencies. The influence of the disturbance \( v \) on the output \( y \) is described by the sensitivity transfer function

\[
S_\omega(s) = \frac{1}{1 + P(s)C(s)}.
\]

Mathematically, the desire of attenuating the effect of disturbances is expressed in terms of the \( H_\infty \)-norm:

\[
||S_\omega(s)W_\omega(s)||_\infty \leq 1 \quad \forall P(s) \in \mathcal{P}_m.
\]  

(22)

\( W_\omega(s) \) is a frequency dependent weighting function that determines the desired degree of disturbance attenuation. Usually, disturbances acting on the plant are varying relatively slowly. Therefore, disturbance attenuation is desired to be good in the low-frequency range. Hence, \( |W_\omega(s)| \) is chosen to be large at low frequencies.

A controller \( C(s) \) solves the robust performance problem (22) if

\[
\gamma(C) = \left\| \begin{bmatrix} W_\omega(s)S(s) \\ W_m(s)T(s) \end{bmatrix} \right\|_\infty \leq \frac{1}{\sqrt{2}},
\]  

(23)

where

\[
S(s) = \frac{1}{1 + P(s)C(s)},
\]

\[
T(s) = \frac{P(s)C(s)}{1 + P(s)C(s)}
\]

are the nominal sensitivity and complementary sensitivity transfer functions. By minimizing the performance index \( \gamma \), a controller is obtained, which yields the highest possible degree of disturbance attenuation and robustness with respect to the frequency weighting chosen. The problem of minimizing \( \gamma \) in (23) is called the \( H_\infty \) mixed sensitivity minimization problem, which is well known in the linear, time-invariant, finite-dimensional case, see e.g. Zhou, Doyle, and Glover (1996). The underlying ideas of \( H_\infty \) control theory such as the above performance and robustness considerations are valid for linear, time-invariant systems, for the finite- as well as the infinite-dimensional case. However, in the finite-dimensional case, the solution of the \( H_\infty \) minimization problem, i.e. the actual computation of a controller \( C(s) \), heavily relies on state space methods. These techniques cannot be applied to the infinite-dimensional case because the system state cannot be represented by a finite vector in \( \mathbb{R}^n \) but involves a continuous function, namely the number density function \( f(L, t) \). An extension of state space methods to the infinite-dimensional case was developed in van Keulen (1993). However, this approach requires solving operator Riccati equations and is therefore not applicable easily. Instead, an alternative frequency domain approach was chosen in this study as illustrated in the following section.

3.3. Controller synthesis theory

The mixed sensitivity problem for infinite-dimensional SISO plants was solved by Özbay (1993), Foias et al. (1996) using operator theoretic methods in the frequency domain. It has been shown that this problem can be reduced to an eigenvalue–eigenvector problem for a Hankel + Toeplitz type operator. If certain assumptions hold, the solution to the problem can be derived from a finite number of linear equations. Results from the above references that are necessary for the actual design procedure are briefly summarized in the following, as also presented in Vollmer and Raisch (2001).

The synthesis method is applicable if the following assumptions are met:

1. The plant transfer function can be decomposed as

\[
P(s) = \frac{M_\omega(s)N_1(s)N_2(s)}{M_d(s)},
\]  

(24)

where \( M_d \in \mathbb{H}_\infty \) is rational inner, \( M_m \in \mathbb{H}_\infty \) is inner and possibly irrational, \( N_1 \in \mathbb{H}_\infty \) is outer, possibly irrational and furthermore \( N_1^{-1} \in \mathbb{H}_\infty \) and finally \( N_2 \in \mathbb{H}_\infty \) is rational outer.

A transfer function is in \( \mathbb{H}_\infty \) if and only if it is stable and proper. An \( \mathbb{H}_\infty \) function \( M \) is called inner if \( |M(j\omega)| = 1 \). All pass transfer functions and pure delays are inner. An \( \mathbb{H}_\infty \) function \( N \) is called outer if it has no zeros in the open right half plane.

This means in particular that (i) the plant transfer function \( P(s) \) has only finitely many unstable poles (the zeros of \( M_d(s) \)) and (ii) for \( \omega \to \infty \), the Bode magnitude plot of \( P(s) \) has a constant roll off rate:

\[
\frac{\log |P(j\omega)|}{\log \omega} = \text{const}.
\]

(22)

2. The weighting functions \( W_m(s) \), \( W_d(s) \), \( W_m^{-1}(s) \) are rational. Furthermore, \( (W_m(s)N_2(s))^{-1} \in \mathbb{H}_\infty \). This implies that if \( P(s) \) is strictly proper, \( W_m(s) \) has to be improper.

To obtain the factorization (24) of the plant model, the right half plane poles and zeros of \( P(s) \) have to be computed, i.e. the right half plane zeros of the quasi-polynomials \( QP_1(s) \) and \( QP_2(s) \) (20) have to be determined. This problem can be solved numerically by using the Nelder–Mead simplex search algorithm of Lagarias, Reeds, and Wright (1998). Appropriate starting values for the search can, e.g., be provided based on the inspection of a three-dimensional magnitude plot of the questionable quasi-polynomial over the complex plane.
If the above assumptions are met a minimizing controller can be computed from (i) the inner and outer factors of the plant transfer function \( P(s) \), see Eq. (24), (ii) two finite-dimensional expressions \( E_I(s) \), \( F_I(s) \) obtained from the weighting functions \( W_d(s) \), \( W_m(s) \) and (iii) a rational function \( L(s) \) that satisfies a set of interpolation equations. An optimal controller is given by

\[
C_{\text{opt}}(s) = \frac{E_{\text{opt}}(s)M_d(s)F_{\text{opt}}(s)L(s)}{N_1(s)N_2(s)(1 + M_n(s)F_{\text{opt}}(s)L(s))}, \tag{25}
\]

with

\[
E_I(s) = \frac{W_d(-s)W_d(s)}{\gamma^2} - 1,
\]

\[
F_I(s) = H_I(s) \prod_{k=1}^{n_I} \frac{s + \eta_k}{s - \eta_k},
\]

where \( \eta_1 \ldots \eta_{n_I} \) are the poles of \( W_d(s) \), and \( H_I(s) \) is the stable, minimum-phase transfer function determined by the spectral decomposition

\[
H_I(s)H_I(-s) = \left(1 - \frac{W_d(s)W_d(-s)}{\gamma^2} - 1\right) \times \left(\frac{W_m(s)W_m(-s)}{\gamma^2} - 1\right)^{-1}.
\]

\( L(s) = \frac{L(s)}{L(s)} \) is a \((n_I + k-1)\)th-order, rational transfer function satisfying the following interpolation conditions:

\[
0 = L_1(\beta_1) + M_a(\beta_1)F_I(\beta_1)L_2(\beta_1), \tag{26a}
\]

\[
0 = L_1(\alpha_k) + M_a(\alpha_k)F_I(\alpha_k)L_2(\alpha_k), \tag{26b}
\]

\[
0 = L_2(-\beta_1) + M_a(\beta_1)F_I(\beta_1)L_1(-\beta_1), \tag{26c}
\]

\[
0 = L_2(-\alpha_k) + M_a(\alpha_k)F_I(\alpha_k)L_1(-\alpha_k), \tag{26d}
\]

where \( \beta_1, \ldots, \beta_{n_I} \) are the right half plane zeros of \( E_I(s) \) and \( \alpha_1, \ldots, \alpha_{k} \) are the unstable poles of the plant \( P(s) \). This means that the right half plane zeros of the \( M_d(s) \) term in the numerator of \( C_{\text{opt}} \) are cancelled by the \((1 + M_a(s)F_{\text{opt}}L(s))\) expression within the controller and therefore do not cancel the unstable poles of the plant. The largest value for \( \gamma \) such that (26a)–(26d) has a nontrivial solution is the minimal performance cost \( \gamma_{\text{opt}} \). Lower and upper bounds for \( \gamma_{\text{opt}} \) can be easily computed. Eq. (26a)–(26d) is a set of linear equations with the coefficients of \( L_1(s) \) and \( L_2(s) \) as unknown variables. It can be written in matrix form: \( 0 = A_{\gamma} \cdot \lambda \) with vector \( \lambda \) containing the unknown coefficients. A nontrivial solution is obtained if and only if matrix \( A_{\gamma} \) is singular. Therefore, \( \gamma_{\text{opt}} \) is found by searching the largest value of \( \gamma \) such that the minimum singular value of \( A_{\gamma} \) is 0. Once the optimal performance cost \( \gamma_{\text{opt}} \) is known, it is straightforward to determine the corresponding controller \( C_{\text{opt}} \) from Eq. (25).

The expression for \( C_{\text{opt}} \) involves the possibly irrational transfer functions \( M_n \) and \( N_1 \). Therefore, the optimal controller itself is irrational. For practical implementation, it needs to be approximated by a rational transfer function. This is achieved using an approximation technique based on Fourier transform and balanced model reduction developed by Gu, Khargonekar, and Lee (1989). In this approach, basis functions of the form \( E_I(s) = (s - s_1)/(s + s_1)^h \) are chosen. A Fourier series expansion for the irrational transfer function \( C_{\text{opt}}(s) \) is computed using these basis functions. This usually yields a very high-order approximation. In a second step the order is reduced via balanced realization truncation. The approximation obtained from the Fourier series can be realized in a special state space form for which the computations required for the order reduction are especially easy to perform. Convergence of this approximation in the \( H_\infty \) sense is established in Gu et al. (1989) under certain mild conditions. In particular, it can be applied to stable transfer functions. If the controller \( C_{\text{opt}}(s) \) is unstable, the individual stable factors in (25) need to be approximated.

The procedure described in this section is readily implementable on a computer. In fact, a Matlab implementation is available Özbay, 1998 for the computation of \( C_{\text{opt}}, \; \gamma_{\text{opt}} \) if \( P(s) \) is already decomposed according to (24). Also, factorization of the plant and controller approximation can be implemented e.g., in Matlab.

3.4. Controller design for the crystallizer

For the factorization of the transfer function \( P(s) \) in (20), its right half plane zeros and poles have to be determined. In Fig. 10 a sector of the complex plane is shown with the poles and zeros of \( P(s) \) lying in this region. There is one unstable pair of poles at \( s_{1,2} = (5.207 \times 10^{-2} \pm 1.010i) \mathbf{j} / \mathbf{h} \) and two pairs of zeros in the right half plane at \( z_{1,2} = (0.360 \pm 2.767 \times \mathbf{i}) \mathbf{j} / \mathbf{h} \) and \( z_{3,4} = (1.650 \pm 1.771i) \mathbf{j} / \mathbf{h} \). With this result the factorization of \( P(s) \) according to (24) is as follows:

\[
M_a(s) = \prod_{i=1}^{4} \frac{s - z_i}{s + z_i}, \tag{27}
\]

\[
M_d(s) = \prod_{i=1}^{2} \frac{s - z_i}{s + z_i}. \tag{28}
\]
Fig. 11. Weighting functions $W_d(s)$ for Controller 1 (solid), $W_m(s)$ (dashed), $W_d(s)$ for Controller 2 (dotted).

\[
N_1(s) = \frac{P(s)M_d(s)}{M_0(s)N_2(s)}, \quad N_2(s) = \frac{60}{s + 6 \times 10^{-2}}.
\]  

As the plant $P(s)$ has a constant roll-off rate of 20 dB/decade for large frequencies and $N_1(s)$ needs to be invertible in $H_\infty$ (i.e. it has to be proper but not strictly proper, see assumptions in Section 3.3), $N_2(s)$ has to be some rational stable minimum-phase transfer function of relative degree one.

In the following, two different controllers are designed. The weighting functions for the first controller $C_1(s)$ are chosen as follows:

\[
W_d(s) = \frac{3(s + 60)^3}{500(500s + 60)^2s},
\]

\[
W_m(s) = \frac{(40s + 60)^2}{120(s + 60)}.
\]  

This choice reflects the desire of good disturbance rejection at low frequencies and of robustness with respect to model uncertainty in the high-frequency range. A magnitude plot of both weighting functions is given in Fig. 11. The weight $W_d(s)$ for the sensitivity function contains an integrating term, to ensure perfect setpoint tracking at steady state. Note that $W_m(s)$ needs to be improper such that $(W_m(s)N_2(s))^{-1} \in H_\infty$, see assumptions in Section 3.3. The optimal performance level that can be achieved is $\gamma_{\text{opt}} = 0.9520$. For the second controller the same $W_m(s)$ was chosen. The weighting function for the sensitivity was changed to

\[
W_d(s) = \frac{3(s + 6)}{50s}
\]  

in order to increase the frequency range in which performance is supposed to be good. This results in a higher controller bandwidth. Relative to $W_d(s)$, the weighting function $W_m(s)$ is lower in this design. Hence a decrease of robustness is expected. The optimal performance level in this design is $\gamma_{\text{opt}} = 1.2125$. A bode magnitude plot of the optimal controller $C_{\text{opt}}(s)$ along with a finite-dimensional approximation is shown in Fig. 12 for the first design and in Fig. 13 for the second case. Nyquist plots of the open loop transfer function $1 + P(s)C(s)$ are shown in Figs. 14 and 15. As the open loop system has two unstable poles, the Nyquist plot has to encircle the origin once to make the closed loop stable. It is obvious that the distance from the origin in the first design is bigger than in the second one. Consequently,
phase and gain margins are larger, which again reflects the superior robustness of the first design.

4. Closed loop simulation studies

Two different scenarios have been chosen to demonstrate the effectiveness of the controllers and to demonstrate their differences. All simulations are done with the nonlinear population balance model as derived in Section 2.2. The controllers have been designed on basis of the linearized model, so they are only expected to work “close” to the steady state. The first simulation starts at steady state. The system is disturbed by increasing the supersaturation in the feed by 1% for 1 h. Results with the first controller are shown in Figs. 16 and 17. The controller successfully stabilizes the plant. Oscillations are eliminated completely. The system approaches the steady state after approximately 20 h. The peak amplitude of the manipulated variable (fines dissolution rate, see Fig. 17) is about 10% of the steady-state value, i.e. the manipulated variable moves quite moderately. The results with the 2nd controller (see Figs. 18, 19) are similar. However, it reacts faster and brings the system close to steady state sooner than the first controller. The peak values of the manipulated variable are somewhat larger than in the first case.

As expected, the performance of the second controller is slightly superior to the first one. Robustness was not an issue since the system is relatively close to its steady state during the entire simulation time.

In a second simulation experiment, initially the system is far from its steady state. The initial condition is chosen such that the overall crystal mass in the crystallizer is the same as in the steady state. However, the size distribution is different. It is chosen such that the crystal mass is distributed uniformly between 400 μm and 1 mm. A plot of the initial and the steady-state mass density function is shown in Fig. 20. Being thus far from the steady state, the linearized model is not a very good approximation of the nonlinear model any more. It is therefore expected that the first controller, which is more robust than the second one, will behave superior in this case. Indeed, the second controller fails to stabilize the system.
starting from this initial state. The crystallizer exhibits oscillations with even higher amplitude than without control. In contrast, the controller that was designed to tolerate higher levels of model uncertainty is still successful. Simulation results are shown in Figs. 21–23. The third moment in Fig. 21 attains its steady state after approximately 32 h. As can be seen in Fig. 22 the maximum fines dissolution flow rate is still in a tolerable range. The peak value is around 40% above the steady state. In Fig. 23 the mass density is plotted versus time and crystal length. It can be seen that the initial uniform mass density function leads to a high peak in the mass density function after about 3 h. This high amount of big crystals leads to the production of a large number of attrition fragments that grow and abrade themselves after a couple of hours. This is exactly the mechanism that leads to oscillations in the open loop case. However, by manipulating the fines dissolution rate appropriately the controller dampens the oscillations and finally the steady state is reached.

5. Conclusions

Controller synthesis for the 1000l continuous DTB crystallizer at Delft University of Technology was considered. The starting point was a detailed population balance model for this plant taken from the literature. A less detailed population balance model was derived that, on the one hand, was simple enough to permit the derivation of a transfer function. On the other hand, this model was still accurate enough to preserve the main features of the original model and the real process. In particular, it was able to reproduce frequency and amplitude of the sustained oscillations observed in experiments quite well.

A $H_\infty$ robust performance problem was formulated reflecting the desire of disturbance rejection at low frequencies and robustness with respect to large multiplicative model uncertainty at high frequencies. This lead to a mixed sensitivity minimization problem that was solved using recent results from $H_\infty$ theory for infinite-dimensional systems. The controllers obtained by this design methodology are in the form of nonrational transfer functions. For practical implementation, these controllers were approximated by rational, i.e. finite-dimensional, transfer functions.

In the last section, closed loop simulation studies of the nonlinear population balance model controlled by two different finite-dimensional controllers were presented to demonstrate the effectiveness of the proposed method. It was shown that by the choice of weighting functions in the design procedure a trade off is possible between performance and robustness properties of the controller. If the weighting functions are properly chosen, the resulting controller does not only stabilize the linearized model for which it was designed but also the underlying nonlinear model.

Thus, it was demonstrated that infinite-dimensional $H_\infty$ theory is an appropriate framework to solve the problem
of stabilizing oscillatory crystallization processes. It has
been shown that, using feedback control, continuous crys-
tallizers can be operated at higher fines dissolution rates
than in the open loop case such that superior CSD prop-
cies can be achieved. The choice of fines dissolution
rate as the manipulated variable and overall crystal mass
as the measurement is a realistic setup that could be
implemented in an experiment. Of course, other combi-
nations of measured and manipulated variables can be
implemented in an experiment. Of course, other combi-
nations of measured and manipulated variables can be
used with the design procedure presented in this arti-
cle. For future investigations it is planned to apply con-
trollers designed on the basis of the relatively simple
model presented in this article to the detailed model
that served as a starting point for this work. There is
strong evidence that with a robust design, controllers can
be obtained that are able to cope with this plant model
mismatch.

Appendix A. Computations for the derivation of plant
transfer function

As mentioned in Section 3.1 the PBE (11), the bound-
ary condition (2) and the mole balance (12) are linearized
with respect to the desired steady state. This leads—
after some restructuring—to the following linear process
model:

Population balance

\[
(V + V_{AZ}) \frac{\partial \Delta f(L,t)}{\partial t} + \frac{\partial G(L,t)}{\partial L} \frac{\Delta f(L,t)}{\Delta c_{L,A}} = \frac{\partial f_{ss}(L)}{\partial L} \Delta c_{L,A}(t) \\
- \left( \frac{\partial G(L,t)}{\partial L} \right)_{ss} \Delta c_{L,A}(t) \\
- \left( \frac{\partial g_{eff}(L,t)}{\partial L} \right)_{ss} \Delta f_{FD}(L,t) \Delta f_{FD,ss} \\
+ \Delta f(L,t) = \frac{\Delta B(t)}{G_{eff}(L_0, \Delta c_{ss})} - \frac{B_{ss}}{G_{eff}(L_0, \Delta c_{ss})^2} \\
\times \frac{\partial G(L_0, \Delta c)}{\partial c_{L,A}}_{ss} \Delta c_{L,A}(t). \\
(A.2)
\]

Mole balance

\[
G_{attr}(L) \partial f(L,t) \partial L \\
= \frac{h_{FD}(L,t) f_{ss}(L)}{V_{G_{eff}(\xi, \Delta c_{ss})}} \\
+ \frac{\partial g_{eff}(L,t)}{\partial L} \frac{\partial f_{ss}(L)}{\partial L} \Delta f_{FD}(L,t) \Delta f_{FD,ss} \\
- \frac{\partial g_{eff}(L,t)}{\partial L} \frac{\partial f_{ss}(L)}{\partial L} \Delta f_{FD}(L,t) \Delta f_{FD,ss} \\
+ \Delta f_{FD}(L,t) \Delta f_{FD,ss} \\
+ \frac{h_{FD}(L,t) f_{ss}(L)}{V_{G_{eff}(\xi, \Delta c_{ss})}} \\
\times \Delta c_{L,A}(s) d\xi. \\
(A.5)
\]

where \( s \) is the complex Laplace variable and

\[
A(L,s) = \int_{L_0}^{L} \frac{g_{eff}(\xi, \Delta c_{ss})}{V_{G_{eff}(\xi, \Delta c_{ss})}} d\xi.
\]

\[
(A.6)
\]

With the piecewise linear expressions for the growth rates
(14) and (15) the integrals in the above equations can be
computed analytically. This is done with a computer alge-
bra system and leads to rather lengthy expressions that are
boundary condition

\[
\Delta f(L = L_0, t) \]

\[
= \frac{\Delta B(t)}{G_{eff}(L_0, \Delta c_{ss})} - \frac{B_{ss}}{G_{eff}(L_0, \Delta c_{ss})^2} \\
\times \frac{\partial G(L_0, \Delta c)}{\partial c_{L,A}}_{ss} \Delta c_{L,A}(t). \\
\]

The deviations of time-dependent functions from their
respective steady-state values are denoted by \( \Delta f(L,t) \),
\( \Delta c_{L,A}(t) \), \( \Delta V_{FD}(t) \) and \( \Delta c(t) \). The time derivative
of the volume fraction \( \Delta c(t) \) (13) can be computed as

\[
\frac{d \Delta c(t)}{dt} = -k_f \int_{L_0}^{L} \frac{\partial f(L,t)}{\partial t} dL \\
\]

with \( \partial f(L,t)/\partial t \) according to (A.1). This linear model is
then Laplace transformed. Thus, a first-order ordinary dif-
ferential equation with nonconstant parameters with inde-
pendent variable \( L \) is obtained from the linear PBE (A.1).
This equation can be solved analytically which yields the
following expression for the crystal size distribution:

\[
\Delta f(L,s) = \\
\frac{e^{A(L,s)} \left( \Delta B(s) \right)}{G_{eff}(L_0, \Delta c_{ss})} - \frac{B_{ss}}{G_{eff}(L_0, \Delta c_{ss})^2} \\
\times \frac{\partial G(L_0, \Delta c)}{\partial c_{L,A}}_{ss} \Delta c_{L,A}(s) \\
- \int_{L_0}^{L} \left( \frac{g_{eff}(\xi, \Delta c_{ss})}{V_{G_{eff}(\xi, \Delta c_{ss})}} \right) \\
\left( h_{FD}(\xi) f_{ss}(\xi) \Delta V_{FD}(\xi) \\
+ \frac{\partial g_{eff}(\xi, \Delta c_{ss})}{\partial c_{L,A}}_{ss} f_{ss}(\xi) \right) d\xi \\
\times \Delta c_{L,A}(s) d\xi. \\
(A.5)
\]
therefore omitted here. However, the solution for the crystal size distribution is of the form

\[ \Delta f(L,s) = \Phi_1(L,s) \Delta c_{L,A}(s) + \Phi_2(L,s) \Delta B(s) + \Phi_3(L,s) \Delta V_{FD}(s). \] (A.7)

The measured output is the third moment of the distribution, i.e.,

\[ \Delta m_3(s) = \int_{L_0}^{L_\infty} L^3 \Delta f(L,s) \, dL = \Psi_{3,1}(s) \Delta c_{L,A}(s) + \Psi_{3,2}(s) \Delta B(s) + \Psi_{3,3}(s) \Delta V_{FD}(s). \] (A.8)

The nucleation rate is proportional to the fourth moment of the size distribution (6)

\[ \Delta B(s) = k_b \int_{L_0}^{L_\infty} L^4 \Delta f(L,s) \, dL = k_b(\Psi_{4,1}(s) \Delta c_{L,A}(s) + \Psi_{4,2}(s) \Delta B(s) + \Psi_{4,3}(s) \Delta V_{FD}(s)), \] (A.9)

where the functions \( \Psi_{i,k}(s) \) are defined as

\[ \Psi_{i,k}(s) = \int_{L_0}^{L_\infty} L^k \Phi_i(L,s) \, dL, \quad i = 1 \ldots 3, \]

\[ k = 3, 4. \] (A.10)

Inserting the expression for the size distribution \( \Delta f(L,s) \) (A.7) in the Laplace transformed version of the linearized mole balance (A.3) and solving it for the concentration \( \Delta c_{L,A}(s) \) leads to an equation of the form

\[ \Delta c_{L,A}(s) = \Gamma_1(s) \Delta B(s) + \Gamma_2(s) \Delta V_{FD}(s). \] (A.11)

Finally, a transfer function from manipulated input \( \Delta V_{FD}(s) \) to measured output \( \Delta m_3(s) \) can be obtained from Eqs. (A.8), (A.9) and (A.11) by eliminating \( \Delta c_{L,A}(s) \) and \( \Delta B(s) \). This concludes the derivation of the plant transfer function \( P(s) \) in (20).

References


