STELLINGEN

behorend bij het proefschrift

Control and Dynamic Modelling of Industrial Suspension Crystallizers

R.A. EEK

1. Het valideren van modellen voor het dynamische gedrag van deeltjesgrootteverdelingen in deeltjesprocessen met behulp van experimentele data, kan slechts dan succesvol zijn als het procesmodel een nauwkeurig model voor de toegepaste deeltjesensor bevat.


3. De claim van Randolph, dat een succesvolle besturing van de kristalgrootteverdeling in een continu kristallisatieproces mogelijk is door te meten in het kristalgrootte domein van 17 tot 32 micrometer, is niet algemeen geldig en zal bij veel processen ongewenst of zelfs instabiel gedrag veroorzaken (United States Patent, No. 5.124.265, June 23, 1992).

4. Stabiliteit van het dynamische gedrag van kristallisatieprocessen dient met behulp van procesregeling te worden verkregen in plaats van met wijzigingen aan het procesontwerp.

5. Bij het ontwerp van regelsystemen voor de deeltjesgrootteverdeling in een kristallisatieproces is kennis over gevoeligheid van de sensor als functie van de kristalgrootte een belangrijke ontwerpparameter.

6. Ook het regelen van niet-technische systemen door middel van terugkoppeling heeft een redelijke kans op succes, alleen als er voor het ontwerp van de regelaar gebruik wordt gemaakt van een voldoende betrouwbare beschrijving van het onderhavige systeem.

7. Ten onrechte denken velen dat de strijd tegen vergankelijkheid kan worden beslecht met fotografische reproductie.

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10. De beste stellingen zijn van hout (Ikea gids, 1995).
Control and Dynamic Modelling

of

Industrial Suspension Crystallizers
Control and Dynamic Modelling of Industrial Suspension Crystallizers

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Preface

This Ph.D. thesis reports the results of 5 years of research carried out as a part of the UNIAK crystallization research project.

The contributions of many people to the development of this thesis are greatly acknowledged. First of all I would like to thank my supervisors Okko Bosgra and Sjoerd Dijkstra for their support, encouragement, and the freedom they allowed me.

I would like to thank the project leader Brian Scarlett for managing the UNIAK project in a stimulating way. I also like to thank Gerda van Rosmalen for the valuable insight she provided on crystallization topics.

My UNIAK colleagues Arthur Boxman, Hans Gerla, Camiel Heffels, Herman Kramer, and Ruairi O’ Meadhra, thanks for the enthusiastic support, the stimulating discussions, and the friendship. Also I wish to thank my predecessor Sjoerd de Wolf. He provided me with important insights into crystallization, and plant operation, enabling me a quick start in the project. He also provided me with the right vocabulary.

The main results in this thesis are based upon a set of approximately 40 pilot plant experiments, each lasting 2 days in average. This experimental work could not have been realized without the commitment and the enthusiastic and unconditional support of 10 students who worked in the project during their graduation work. They assisted with pilot plant experiments, also during long and weary night shifts, and during unaccustomed hours for students, such as weekends, early mornings, and hot summer days, where the temperature around the test rick could exceed 35 °C. I am greatly indebted to Ben de Boer, Taco Boerstra, Jaap Both, Arjan Hoogenboezem, Mirjam Korstanje, Wouter Kuiper, Albert Pouw, Binnert Prins, Sven Rusticus, and Jan Herman Spanjersberg. I also wish to acknowledge Harm Mosterd who joint the project as a post graduate student for half a year.

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Summary

Industrial crystallization from suspension is still a troublesome unit operation. The crystal size distribution (CSD) of the product crystals of a continuously operated crystallizer may show badly dampened or even oscillatory behaviour. In addition, the (CSD) of both batch wise and continuously operated crystallizers may deviate significantly from the desired specifications. These fluctuations and offsets cause solid-liquid separators in the process downstream to perform poorly, and diminish several end-use properties, such as the solubility of the crystals. Furthermore, fluctuations in the CSD cause fluctuations in the growth rate of crystals, which may lower their purity, for instance due to the increase of liquid inclusions at higher growth rates.

This thesis concerns the design of control systems for the CSD, with the objective of enhancing the product quality and the overall economic performance of industrial suspension crystallizers. A main requirement is that the control system should appeal to the requirements of the process industry.

As a starting point, a first principles modeling procedure is applied. This leads to a parametrized nonlinear population balance model that describes the dynamics of the crystal population as a function of crystal birth, crystal growth, attrition, and the classified removal of both fine and product particles from the crystallizer. Several numerical solution schemes are presented, to simulate the model, to analyze the process properties, and to derive low order models for controller design.

The first principles model contains several empirical parameters with large uncertainties. An essential step, therefore, is to obtain good estimates for these parameters, and to assess the quality of the model based on measured plant data. Relevant plant input-output data have been measured from a semi-industrial pilot crystallizer, equipped with effective process actuators, and on-line sensors for the CSD dynamics. On-line measurement of CSD dynamics is achieved with a forward light scattering technique. In addition, a simple particle counter is used that outputs a measure related to the number of fine particles. Special attention is paid to sampling, and to the interpretation of measured detector data.

Model verification is done in three steps. In the first step, some model assumptions are evaluated based on published data, and results of simple experiments. In the second step the model is verified, using process start-up experiments that are often readily available in historical databases. Start-up data have been measured from the
pilot plant for both classified and unclassified removal of product crystals at different process conditions.

In the third step, system identification experiments are conducted. In this case the process dynamics are persistently excited with random test signals added to the nominal process inputs. It is found that these experiments have to be conducted in closed loop to prevent the oscillatory dynamics from overruling the actuator excitation. The closed-loop input-output data are used to verify the first principles model, and to estimate low order experimental input-output models directly. A special effort is made to prevent the estimated input-output model from getting biased due to the correlation of noise between the closed-loop input-output data. Both the verified first principles model, and the input-output model are considered for controller design.

With forward light scattering, the control relevant quantities cannot be directly inferred from the process measurements. Therefore, a dynamic estimator is developed, that reconstructs control relevant process variables, using the process model, from noisy input-output measurements. The dynamic state estimator is shown to perform better than a simple static estimator that employs an estimated inverse of the sensor model.

Before designing a controller, the controllability of the process is studied. The major objective in this step has been to study both the degrees of freedom of the process, and the process input-output configuration that yields a controllable plant at steady-state and dynamic process conditions. Properly locating the sensor in the crystal size domain, is found to be an important factor. For the pilot crystallizer, operating with ammonium sulphate, measuring fine particles with a size smaller than 30 microns introduces a right half plane zero in the transfer function that strongly hampers the achievable control performance. In principle, the location of this zero cannot be influenced with the controller, but merely by relocating the sensor. Good closed-loop behaviour is obtained with a particle counter that counts particles in the range of 40 to 100 microns.

Some guidelines are given for the design of stabilizing controllers for industrial crystallizers. Control schemes are restricted to fines removal based control schemes using a scalar measure for the fines density, and a simple SISO PI-controller. It is discussed how these controllers can be applied for different crystallizer types that may exist in industrial practice. Experiments have been conducted that show the validity of the designed simple controller.

The last subject addressed, is the design of a model based controller. Model predictive control based on state-space models is used. This controller can handle process input-output contraints explicitly, and includes a state estimator as well. With simulations and pilot plant experiments, it is shown that good closed-loop behaviour can be established with this controller. However, the results suggest that model based control only slightly improves the closed-loop behaviour of the process in comparison to simple multiloop PI controllers, also when constraints are present.
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Chapter 1

Introduction

The work covered in this thesis is motivated from operational problems existing in industrial crystallization practice. Results on model identification and parameter estimation are reported. Robust control schemes are designed and verified experimentally.

1.1 Motivation and background

Crystallization from a solution or melt is one of the oldest industrial separation and purification processes. With crystallization a multi-component liquid solution or melt is treated to form a crystalline solid phase, which has a composition different from that of the liquid. In comparison to distillation, crystallization has the main advantage that it can produce products with very high purity, at a low level of energy consumption. Further it operates at moderate process conditions, which is attractive for a wide range of products, and in particular for temperature labile substances.

The economic importance of crystallization is enormous [131]. It is applied in continuous operation on a large scale, for the bulk production of inorganic materials like potassium chloride (a fertilizer), sodium chloride, and ammonium sulphate (a fertilizer), and organic materials like adipic acid (a raw material for nylon), para-xylene (a raw material for polyester), and pentaerythritol (used for coatings). On a small scale crystallization is often applied batch wise to obtain high purity fine chemicals or pharmaceuticals, such as aspartame (a sweetener), L-serine (an amino acid), and L-ascorbine (vitamin C).

The main driving force in crystallization is the supersaturation of the liquid phase or the mother liquor, which determines the rate at which the dissolved component to be crystallized, is transferred to the solid phase by growth of existing crystals and by birth or nucleation of new crystals. Supersaturation can be created by evaporation of solvent, cooling, or precipitation [174, 130]. Mostly, one refers to solution or suspension crystallization when mass transfer is the dominant effect in the process, whenever heat transfer is the dominant factor the term melt crystallization is used [235, p.
151]. Melt crystallization will thus give rise to much higher growth rates than solution crystallization.

The produced crystal product is a wet product slurry containing solid crystals, dissolved crystal material and solvent. The quality of the crystallized product is mainly determined by the shape, the purity, and the spectrum of sizes of the crystals in suspension. The latter is denoted as the particle or crystal size distribution (CSD). The fraction of impurities, which can be present in the crystal lattice as for instance, solid inclusions, or entrained mother liquor, determines the purity of the product crystals. Generally, the level of impurities should not exceed a prescribed value to prevent undesired by-products in further processing steps, for which the crystallized material serves as a raw material.

For many applications the CSD of the product crystals is of primary importance. Quality requirements of customers are often stated in terms of the extent of dust forming, the ability to flow and the dissolution rate of particles. These parameters are directly relate to the CSD. In addition, the CSD determines the operability of wash columns, and the performance of other downstream solid-liquid separation devices like thickeners, centrifuges, and dryers. Also the storage in bulk and the bulk transportation is strongly influenced by the CSD, and for some substances by the morphology as well. For instance, the transportability of dried crystals is lowered due to caking problems, which occur whenever the small crystal (fines) density is too high, or when the density of mother liquor inclusions is high. More examples on the influence of the CSD on end-use properties of slurry and bulk crystals are described by Randolph [197, p. 3].

Despite its wide spread use and the long history of experience that exists in process industry, the operation of crystallization processes still poses difficulties. Undesired values of the described product specifications — the purity, the morphology, and the size distribution — will occur during startup and shutdown of a plant, due to instabilities, due to cycling behaviour of the process itself, and due to process disturbances or load and setpoint changes. The latter may be due to changing marketplace conditions. A non exhaustive list of frequent operational problems is:

1. Deposition of crystal solids on the crystallizer internals. This phenomenon, denoted as fouling, scaling or incrustation, lowers the yield as a part of the production will be lost on the crystallizer internals, and necessitates for periodic shutdown and successive cleaning of the crystallizer. Fouling will be lowered by the erosive action of the circulating crystals in suspension and promoted by the supersaturation level of the mother liquor [244, 246]

2. Process startup and shutdown. Most (continuous) crystallizers have to be shutdown frequently to remove salt incrustated on the crystallizer internals, or for other maintenance of cleaning purposes. The process time between startup and shutdown can vary from a few days to several months, depending on the process
conditions and the substance system that is crystallized. During the dynamic startup and shutdown phases, the CSD of particles will show significant offsets.

3. Accumulation of undesired components (sludge). In most industrial plants the clear mother liquor is recycled to the crystallizer after separation from the solids fraction. This causes accumulation of foreign soluble material, which may cause transients in the nucleation and growth rates. This phenomenon necessitates for periodic or continuous draining of clear mother liquor.

4. Alternating feed composition. These may change the growth and nucleation kinetics and thereby the CSD, the purity and the morphology of crystals.

5. Inappropriate seed size distribution. This problem is often encountered in batch crystallizers, where a classified population of seeds is used to initiate crystallization. A bad initial seed distribution will cause the final distribution to be off specifications as well.

6. Failure of the power supply system. This will mainly decreases the yield and disturb the size distribution as well.

7. Failure of the product removal flow due to blockage of the product removal system. Also this problem will cause upsets of the CSD.

8. Temperature changes. These can be caused by failure of the vacuum system and seasonal or day/night offsets. Temperature changes can affect both the yield and the kinetics of crystallization.

9. Rinse water injection. Rinse water is injected to remove salt that is incrustated on the crystallizer internals or to dissolve blocked lines, or centrifuges. Injection of rinse water will create a (local) undersaturation in which particles will dissolve (partially). This may cause significant upsets of the CSD and the yield.

In addition, a major problem with many continuous industrial crystallization processes is the occurrence of slow CSD oscillations. This phenomenon is caused by an intrinsic physical feedback mechanism, which has first been studied by Randolph et al., [193]. With a cycling CSD periodically an excessive number density of fine crystals will be present in the product, which causes problems with solid-liquid separation in the process downstream.

The above problems cause significant production losses and lower the yield. With the increase of material and energy costs, the increase of environmental and product quality requirements evolving over the last 20 years, and the growing competition between large chemical companies, industrial and academic researchers are paying an increased attention to the operational problems arising from industrial crystallization processes. In part, these problems may be solved by improvement of the design, and the use of other equipment materials that prevent for instance incrustation. Most
problems, however, are caused by unpredictable and often temporal changes in the environmental conditions. For these, controllers are needed that adapt the process conditions in a systematic way to prevent violation of the economic and safety constraints. Besides, especially feedback controllers are needed to suppress cycling or open-loop unstable behaviour of the process itself.

Modelling and control of crystallization processes has been treated in the literature extensively. The basic challenges stem from the distributed nature of the process, the limited controllability and observability of the CSD, and the relative large uncertainty of the models related to crystallization kinetics. Important progress is achieved by Hulburt and Katz [113] who introduced the population balance concept. After introduction in the field of crystallization by Randolph and Larson [196], this concept is a widely accepted tool for modelling of crystallization processes.

So called Mixed Suspension Mixed Product Removal (MSMPR) crystallizers are studied by most authors. For this system the first order partial differential equation, which governs the population balance, is approximated by its first moments, resulting in a low order set of ordinary differential equations, which can be simulated easily. During the seventies and eighties many parameter estimation studies are reported that aim at the estimation of empirical parameters related to nucleation and growth, within the MSMPR model framework. In Randolph [194] the process dynamics, are studied as a function of the empirical parameters in the nucleation law. Despite some successful applications these MSMPR models still do not provide much insight in the process physics and they have limited capabilities in describing more complicated process configurations, such as classified fines and product removal and length dependent growth phenomena.

More detailed process models are reported, among others, by De Wolf [49] and Tsuruoka [233], Rawlings and co-workers [204], and Jager [120]. In [120] a compartment model is used to describe the spatial distribution of supersaturation in the vessel. For a review on modelling of crystallization processes the reader is referred to [201]. In [233, 49], a finite difference scheme is applied to lump the population balance model. The latter scheme has more flexibility and is capable to describe classified removal systems and length dependent growth phenomena. However, the resulting models have a high order which is not attractive for control. Therefore, De Wolf and Van Den Hof [50] proposed the use of system identification to derive directly low order models that suit the requirements for controller design. Rawlings and co-workers, use orthogonal polynomials to transform the population balance to a finite set of ordinary differential equations. They present an approach for the estimating parameter confidence intervals on the basis of experimental data from batch experiments.

A shortcoming of most studies — and in particular published work on the dynamics of continuous crystallization processes — is that a profound verification of the derived models based on experimental process data is absent. This is mainly due to the absence of reliable and robust on-line sensors and sufficiently robust experimental facilities.
1.1 Motivation and background

Regarding control studies, much attention has been paid to the problem of CSD control. The work of Randolph and co-workers [14, 198, 195] represents the first stone in this field. Their work shows that both stability and disturbance rejection (in a continuous KCL crystallizer), can be achieved with simple output feedback controllers, which manipulate the fines destruction rate, based on information on the fines or nuclei number density. Recently a patent has been issued on this scheme to Randolph [192]. Rohani [211] describes a similar control scheme, however with a sensor that measures the concentration of fines in the fines destruction loop. This scheme has been evaluated experimentally with a 20 litres potash crystallizer. In numerous articles alternative control schemes, including optimal control solutions for batch crystallizers, and multivariable control schemes are proposed. Details on these can be found in [49, 201] and the references therein.

Thorough experimental evaluation of stabilizing controllers has not been reported. This is due to a lack of sufficiently reliable and robust equipment as well. In addition, due to the lack of good models, the crucial step of controllability assessment is often neglected or absent in the literature. With controllability assessment the achievable control performance is studied in relation to inherent limitations in a plant’s design, including the sensors. Further no contributions are found addressing basic questions such as, what is the best sensor from an economic and operational point of view, what crystal size range should we measure, and what model accuracy and complexity do we need to design a controller based on a dynamic process model.

Modelling and control of purity and morphology gained less attention than CSD control. In the first instance, both purity and morphology of crystals have a large importance for crystallization of pharmaceuticals and fine chemicals as often strict customer requirements exist with respect to these product properties. For bulk crystallization, however, purity and morphology can be important as well. In Klug [132] it is described that even small amounts of impurities may have a dramatic effect on crystal growth, nucleation, and morphology, and by these mechanisms also on the aforementioned bulk properties. In addition, the uptake of impurity will be promoted when the growth rate of crystals increases. The main challenges that currently exist in this field are prediction of the particle morphology, also when impurities are present, and to describe the impurity of crystals as a function of the process operating conditions. In addition, to predict the influence of impurities on the rate of nucleation and growth of crystals poses a large research challenge.

To sum up, the crystallization literature until now has provided many often theoretical modelling and control studies, whereas a profound experimental evaluation of process models and related controllers, is lacking. Mainly due to the absence of good models, several important questions related to controllability and operability of crystallization processes are not answered. In addition, the control schemes, developed in the literature so far, do still not have a sufficient appeal for application in process industry. This reflects into today’s practice in process industry, where the most pop-
ular crystallizer control strategy is still: "hold everything constant" [191]. Compared to progress in the field of process control in distillation, as reported among others by [225], it is concluded that the control of crystallization processes is way behind.

1.2 The UNIAK project

In 1984 a new project, named UNIAK has been initiated. The main topic of this research project is the design and experimental evaluation of control systems, including sensors and actuators, which should be applicable on industrial crystallizers. In the first phase of this project an emphasis has been put on theoretical and experimental modelling and the design of on-line measurement systems. The results of this phase are reported in the thesis of Jager [117], De Wolf [49] and Boxman [24].

The main achievements of this first phase are: a well-documented approach for experimental identification and physical modelling, concepts for product classification, and an automated sampling and measurement system that enables on-line measurement of crystal size distributions based on a forward light scattering technique. Operational problems with the experimental equipment, however, did put a strong limitation on the results obtained in this first phase. Plant operation was disturbed repeatedly by blockage of tubes, and failures of the CSD sampling system.

A second phase of the UNIAK project was started in 1990. It aimed at continuation and generalization of the results and ideas developed during UNIAK-I, with a strong emphasis on experimental evaluation of models and controllers. At the start of this project a thorough revision of the experimental equipment has been carried out. The flow sheet of the plant has been improved, and an improved sampling and slurry dilution system has been installed. To improve the on-line interpretation of CSD sensor data and the detection of sensor faults, better software for the CSD sensor has been implemented. In addition, a new process monitoring and control system has been set up, which has improved early detection of process faults. Besides tight control, of secondary process quantities, such as the slurry temperature and the slurry removal flows, could be achieved with this system.

1.3 Problem Statement

The current state of the art on modelling and control of crystallization processes shows that a thorough insight in the physical mechanisms is still limited, and that no control systems are available that have an appeal for direct industrial use. These deficiencies are mainly due to a lack of robust and cheap on-line measurement systems for the particle size distribution and the absence of robust experimental equipment, which can be operated over a sufficiently long time span.

Therefore the objective of the work described in this thesis is,
to design and to evaluate experimentally, robust control schemes, including actuators and sensors, that enhance the stationary product quality and the yield, stabilize the process dynamics, and reject disturbances.

A major requirement is that the results should appeal to the requirements of the process industry. Obviously, industry will require the controlled process to have a better economic performance than the open-loop process. Smith [226], added to this that controllers should be:

- Flexible, that is, the controller should be able to handle constraints in the process inputs and outputs, and to handle sensor or actuator failure in a way that disturbances of the process output quantities are minimized.

- Quickly implementable. Generally it is most desirable that the structure of the designed controller allows for direct implementation in a standard DCS system.

- Robust, that is, the performance and stability of the closed-loop systems should be guaranteed for errors in the process models and the presence of constraints in both process inputs and outputs.

- Friendly and predictable to operations personal, that is operators should feel ownership.

These requirements served as an important guideline in this work.

1.4 Approach

A key step in the quest to the above problem is the development of a rigorous model. Rigorous modelling based on the first principles of crystallization is chosen as it provides (transferable) insight in the dominant physical mechanisms and can be used for controller design as well. Besides, this approach is in line with a current trend in process industry, where one strives to develop process models, that serve a variety of process related applications such as process design, operator training, controller design, and fault detection.

To overcome the deficiencies of most literature contributions, much attention has to be paid to the experimental verification of models and controllers. As a main tool for this, a 970 litres evaporative draft-tube baffled (DTB) crystallizer will be used. We realize that this plant cannot, mirror the "average" industrial crystallizer. The industrial practice shows an abundance of crystallizer designs. for example, Oslo, DTB, forced circulation type crystallizers; substance systems ranging from fine pharmaceuticals to rough bulk products; scales ranging from 1 litre to 500 cubic metres; operation principles, for example batch, continuous, multistage (multiple effect), isothermal, cooling and evaporative [16, 17, 153, 166]. Therefore, we only pretend to develop validated schemes for the specific situation of a continuous evaporative isothermal suspension
crystallizer, in the first instance, and present them as a good basis for application to other crystallization process configurations.

The data from the crystallizer will be used in a first step to verify the model and to estimate the parameters. At this stage it will be important to assess the uncertainties in the parameters, as these enable to evaluate the robustness of designed controllers. In addition, robustness analysis will indicate what parameters have to be estimated accurately for control applications.

In this thesis control of the CSD dynamics will be given the highest priority. For on-line measurement of the CSD dynamics a system for on-line measurement is required. As most CSD measurement techniques are indirect, also reconstruction schemes are needed, which estimate or infer control relevant CSD properties from the raw sensor data. With respect to this, an important role is attributed to dynamic observers. With observers, the major process constraints in the form of a dynamic model are combined with actual process input-output information to estimate values for process quantities that cannot be measured or inferred directly. The observer may be incorporated in the control loop or be used for process monitoring or fault diagnosis purposes [75]. Besides, this technique is off interest for on-line estimation of the supersaturation level. Then using these reconstructed supersaturation values in a control loop will enable indirect control of the purity.

When a model is available, a control structure has to be selected. Here tools are needed to assess what plant configuration gives the best controllability, that is, the best ability to achieve the desired control performance. The results from this step may have implications for process design, as the sizing of certain parts of the equipment or flow sheet changes may drastically affect the plant's controllability [221, 167]. Note as well that most industrial crystallizers are not equipped with the sensing and actuation devices applied in this work. Adding these or similar systems will imply large investments, which will only gain the managements approval when the economic benefits are indisputable. Therefore, to study static and dynamic controllability aspects is of major importance.

In reaction to the industrial requirements, attention will be paid to the derivation of heuristic rules that enable easy and quick implementable controllers for which no elaborate modelling and controller design procedure is required. More advanced control schemes are required as well, as these are pertinent to applications where significant interaction between process inputs and outputs is present, and where tight control should be achieved in the presence of constraints in the process input and output variables.

An overview of the approach followed in this thesis is given in Fig. 1.1. Notice that two important feedback loops are present in this figure.

The first loop relates process control to the design stage. When the results of controllability analysis suggests that control performance is hampered by intrinsic limitations of the plant itself (e.g., a too narrow physical range of an actuator), the
plant design or the flow sheet has to be changed.

The second loop reiterates the modelling and identification steps, whenever control performance appears to be insufficient. Generally, in a first modelling and control step, insufficient information can be gained from the process as it performs open-loop in a broad nonlinear range, while the dynamics may even be dominated by unstable or cycling behaviour. The process dynamics observed during identification thus will strongly differ from process conditions met during control. Therefore, a controller designed on the basis of this model will have a limited performance, and should be seen merely as an attempt to establish the (experimental) process conditions, e.g. to stabilize the dynamics or suppress cycling behaviour, that allow for proper identification of the plant behaviour. A controller designed in a second step based on the closed-loop
re-identified model, is therefore expected to give better closed-loop performance.

1.5 Outline of thesis

The core of this thesis is subdivided into nine chapters. The structure is roughly in line with the scheme given in Fig. 1.1. The first part (chapter 2 to 7) deals with process analysis via process and sensor modelling, parameter estimation and system identification. The second part (chapter 7 and 10) is devoted to the selection of control structures and the design of controllers based on the model knowledge and the process insight gained from the first part.

Chapter 2: Process modelling and simulation

A dynamic model based on the first principles of crystallization—including sensors and actuators—form the basics for the design and analysis steps. The model equations are presented in this chapter and the underlying assumptions and several limitations are discussed.

The derived model will be used to study several process properties related to stability, observability, and controllability. Therefore, in the second part of this chapter different lumping, simulation, linearization and model reduction tools are introduced. To judge the accuracy of the numerical schemes, a simple analytic solution of a linearized population balance model in the frequency domain is used. A discussion is given on the merits and limitations of the selected numerical solution schemes.

Chapter 3: Process description

This chapter summarizes the experimental facilities, including a pilot crystallizer, a process monitoring, and control system, and a set of actuators and sensors used throughout the work to evaluate the model and the feedback controllers. A discussion will be given on the limitations of the equipment.

Chapter 4: Data interpretation

Information on the CSD is inferred from a sensor based on forward light scattering, that includes a multiple array detector. An important characteristic of this technique, and many comparable techniques as well, is that the CSD cannot be inferred directly from the raw sensor readings. Besides, the different sensor output signals will be mutually dependent, which is not desirable for control. Therefore, in this chapter some existing reconstruction schemes are reviewed and some new possibilities are outlined to reconstruct uncorrelated control relevant parameters from the raw data. In addition, other signal preparation steps that had to be performed before the use of the data are discussed.
Chapter 5: Model verification

Several model assumptions will be evaluated in this chapter based on experimental process data. In addition process data are used to estimate parameters related to empirical relations for hydrodynamics and crystallization kinetics. For this purpose experimental data obtained from the free-run startup response of the pilot crystallizer are measured. The experimental data are used in a nonlinear optimization scheme to obtain estimates of their values and corresponding variances. A discussion is given on the quality of the results in relation to the data and the model structure that are chosen.

In the final two sections of this chapter the model accuracy for both batch and for continuous operation using product classification is verified.

Chapter 6: System identification

This chapter elaborates further on model verification. However, here we aim specifically at the derivation of models for control purposes only. A series of experiments is described where the plant dynamics are controlled with a simple SISO feedback controller to preserve stability. The resulting data are used to estimate empirical parameters in the nonlinear process model, and to identify low order linear input-output models which can be used for control directly. Some specific problems related to closed-loop identification are outlined. The validity of the nonlinear process model is judged by comparing time and frequency domain responses of the linearized and reduced versions of this model with directly estimated linear models.

Chapter 7: Observer design

For on-line measurement of the CSD and supersaturation in crystallizers different sensors have been developed, which however still have practical limitations. With the nonlinear model obtained so far, a nonlinear observer will be devised to reconstruct control relevant properties from raw sensor data, and known process input parameter values. Experimental results, presented in previous chapters, will illustrate the effectiveness of the observer.

Chapter 8: Controllability assessment

A crucial step prior to the design of a controller is controllability assessment. In this chapter controllability assessment will boil down to the selection of a control structure, i.e a set of actuators and controllers, that will give the desired closed-loop behaviour. Both steady-state and dynamic controllability will be discussed.

A discussion will be given on the different techniques that are available to judge the controllability of a plant and how controllability is affected by different inherent plant characteristics.
In this chapter we will also consider controllability assessment in relation to design. The selection of optimal design parameters for classification systems for fine and large particles—which are considered as the main CSD actuators—are discussed.

**Chapter 9: Simple control solutions**

The aim of this chapter is to present a cheap and easy implementable control scheme, which can be applied in industry without elaborate model knowledge. Using the process model some limitations and properties of simple feedback controllers will be analyzed. The role of the sensor on the closed-loop performance is analyzed and the robustness of this controller for a set of uncertain model parameters is evaluated. From this, some rules of thumb and recommendations are given for design and implementation of these controllers on industrial crystallizers, in case no rigorous process models are available.

The controller performance is evaluated experimentally. Closed-loop behaviour of the system is compared with open-loop behaviour to judge the ability of the controller to improve the startup and to reject process disturbances. Also, the ability of the controller to follow setpoint changes is studied.

**Chapter 10: Model based control**

Currently, Model based Predictive Control (MPC) is seen as the best available option for advanced control of multivariable processes. In this chapter some basic features of MPC and its relation to LQG control will be discussed. A design procedure on the basis of the first principles model is outlined, and several robustness properties will be analyzed. In addition, the merits of advanced control are studied by comparison with simple multi-loop PI controllers.

Experimental results from continuous pilot plant experiments will be demonstrated. In addition, one pilot plant run will be reported where an identified input-output model serves as internal model in the MPC controller. Finally, control of batch crystallizers is reviewed shortly and some perspectives for batch crystallizer control using MPC are outlined.

**Chapter 11: Conclusions and discussion**

At the end of the thesis the main conclusions will be summarized and some open areas relevant for future work will be touched upon.
Chapter 2

Dynamic modelling and simulation of suspension crystallizers

In this chapter a model for the dynamics of a single stage suspension crystallizer is presented. This model will serve a basis for process analysis and the design of controllers. In the second section of this chapter several numerical schemes are outlined that will be used in the sequel for simulation and analysis purposes.

2.1 Introduction

Accurate modelling of the behaviour of an industrial crystallizer requires knowledge of several both macro and micro scale phenomena. Microscale modelling efforts focus for instance on the growth rate, the mechanical strength, the purity, and the shape of crystals in relation to the substance and the (local) supersaturation and impurity level. Both Klug [132] and Myerson [169] give overviews of recent advances in this field. A more complete and fundamental treatment of microscale crystal growth, nucleation in relation to morphology is given in the recently issued handbook on crystal growth [114].

Macroscopic modelling efforts focus on the CSD dynamics as a function of effective crystal birth (nucleation), crystal growth, and breakage or abrasion (attrition) phenomena [197]. These phenomena will depend on the physical properties of the substance system, the process operating conditions, and hydrodynamic phenomena, which depend on the geometry and the scale of the reactor. In this field macroscopic population balance models are widely applied. These models were introduced in the field of particulate technology by Hulburt and Katz [113]. Randolph and Larson [196] first applied the population balance concept to a crystallization process.

Currently microscopic and macroscopic models are treated separately. As mentioned in [201] a major challenge for future research is to merge both models into rigorous process models that may be used to solve a variety of macroscopic control
and design related problems, using the best available knowledge from the microscopic phenomena.

The aim of this chapter is to develop a macroscopic model for the dynamics of a crystallization process. A separate model for the CSD sensor will be included. The purpose of the complete model is twofold: it should contribute to the knowledge on the relevant physical mechanisms that determine the crystallizer dynamics, which is relevant for controllability assessment and process design. Secondly, the model should serve as a basis for CSD controller design.

In the following an elaborate discussion is given on the model assumptions and the empirical relations used. The derivation of a set of simplified equations is presented for the UNIAK pilot crystallizer. The main model assumptions for both the general model, and the pilot crystallizer model are summarized in Appendix C. In the second section of this chapter numerical schemes are presented that are used to analyze and simulate the process behaviour governed by the model.

2.2 Process modelling

The system boundary of the process that will be considered, and the set of process input and output variables with some major process parameters, are indicated in Fig. 2.1. It is assumed that the fines are totally dissolved by heating ($P_{ex}$). For cooling crystallizers the vapour flow $Q_v = 0$, and the heat input ($P_{in}$) will attain a negative sign. When product is removed without classification ($Q_{pf} = Q_p$), the mean slurry residence time, often denoted as the product retention time, defined as:

$$r_p \triangleq \frac{V}{Q_p}, \quad (2.1)$$
is considered as a parameter. As CSD actuators the crystal size classification systems for fine and product crystal removal, and the heat input or cooling rate, are considered.

2.2.1 Characterization of crystals

The geometry of individual crystals is characterized by their size, their morphology and their habit. The latter two terms describe the appearance of faceted crystals and the shape given by the relative length to width of the crystal faces [197], respectively. Fig. 2.2 (after [197]) shows the difference between morphology and habit. Here it is assumed that the morphology and habit of crystals is described by a rectangle with dimension $x_l \cdot x_w \cdot x_l$. The (average) aspect ratio $\alpha$ for this shape is defined by: $\alpha = x_l/x_w$, and can be estimated from visual inspection of the particles. In the model the crystal size $x$ is defined to be the size $x_l$. The corresponding crystal shape factor, defined by the ratio of the particle volume $v$ and the cube of a particle with size $x$: $k_v = v/x^3$, is then given by $k_v = 1/\alpha^2$. This factor is used in the mass balances to correct for a possible noncubic shape of the particles.

The distribution of crystal sizes is characterised by the population density function $n(x,t)$, defined by:

$$n(x,t) \triangleq \lim_{\Delta x \to 0} \frac{\Delta N(x,t)}{\Delta x}, \quad (2.2)$$

with $N(x,t)$ the cumulative number function describing the number of crystals per unit of volume, with a size equal to or lower than $x$.

As discussed below, a sensor model is developed for rectangular-shaped particles with the same aspect ratio. With respect to particle size and shape a consistent basis is thus created for the validation of the model with experimental data as further discussed in Chapter 5. We will show in Section 4.3 that significant errors are introduced when an incorrect assumption on the particle shape is made.
With respect to the chemical composition it is assumed that a single pure component is present in the solid phase. Hence the presence of impurities, for instance due to liquid inclusions, is neglected.

2.2.2 Modelling size classification

The size classification subsystem for the fines and product removal loops is schematically depicted in Fig. 2.3. An input flow $Q_0$ containing crystals with a population density $n_0$ is separated into two different flows $Q_1$ and $Q_2$ each containing crystals with a population density of $n_1$ and $n_2$ respectively. The flow $Q_2$ is removed from the process, and the flow $Q_1$ is returned to the crystallizer.

To model size classification in the fines and product removal system the following assumptions are made: the saturation condition of the liquid containing the crystals to be separated is unaltered, that is, the system is isothermal, and no growth or dissolution of crystals occurs; no breakage or attrition of particles occurs; and dynamics or holdup in the separator is neglected.

The general separator model, constrained by the given assumptions, is described by two mass balances, for example, a solid and a liquid phase balance:

$$Q_2(t) \left[ 1 - \epsilon_2(t) \right] = Q_0(t) \left[ 1 - \epsilon_0(t) \right] - Q_1(t) \left[ 1 - \epsilon_1(t) \right], \quad (2.3)$$

$$Q_2(t)\epsilon_2(t) = Q_0(t)\epsilon_0(t) - Q_1(t)\epsilon_1(t), \quad (2.4)$$

and the number balances for respectively the returned and the removed flow:

$$\left[ 1 - h(x, t) \right] Q_0(t)n_0(x, t) = Q_1(t)n_1(x, t) \quad (2.5)$$

$$h(x, t)Q_0(t)n_0(x, t) = Q_2(t)n_2(x, t) \quad (2.6)$$

The function $h(x, t)$ is the removal efficiency function that describes the probability that a particle with size $x$ does not survive the fines trap and $\epsilon$ is a void fraction, which
is calculated from:

$$\epsilon\star(t) = 1 - k_v \int_0^\infty n_\star(x,t) x^3 dx = 1 - k_v m_3\star(t), \quad (2.7)$$

with $m_3\star$ the third moment of the population density $n_\star$ in a certain flow ($\star$).

**Fines classification**

The fines removal system includes a fines classifier and a fines dissolver. Fines classification can be achieved in an annular zone, based on differences in settling velocities of the crystals. According to Eqn. 2.6, the fines distribution $n_f(x,t)$ is described as:

$$n_f(x,t) = \frac{Q_{ff}(t)}{Q_f(t)} h_f(x,t) n(x,t) \quad (2.8)$$

with $h_f(x,t)$ the fines removal efficiency, $Q_f(t)$ the fines removal flow, and $Q_{ff}(t)$ the unclassified flow that enters the bottom section of the annular zone.

The factor $Q_{ff}(t)/Q_f(t)$ is a flow split factor denoted here as $\gamma_f$. Assuming that only particles and no liquid returns from the annular zone back to the main crystallizer volume, the flow split factor can be calculated from: $\gamma_f(t) = \epsilon_f(t)/\epsilon(t)$, with $\epsilon$ and $\epsilon_f$ the void fractions of the fines slurry flow and the slurry in the crystallizer, respectively. In the literature this factor is often taken to be equal to one, which is a good approximation when the void fraction in the crystallizer is high. However, for industrial plants, which mostly operate at 10-30% solids, $\gamma_f$ will be significantly larger than 1, that is approximately between 1.1 and 1.4.

Several empirical relations are published for the function $h_f(x,t)$. Different authors [45, 134, 197] discuss idealized functions, which describe constant classification behaviour. Randolph [197] presents the so-called $R - Z$ model, which is a combined model to describe both classified fines and product removal. In this model the function $h(x)$ is presented as the ratio of the removal rate in the middle crystal size range $(x_f \leq x \leq x_p)$, where $h(x) = 1$, to the removal rate in the fines region $(x < x_f)$, where $h(x) = R$, and the product classification region $(x > x_p)$, where $h(x) = z$, respectively.

Experimental results [135, 45] make clear that ideal classification of fines will not occur. Cut-size calculations may deviate from measured values because of the asphericity of crystals and hindered settling or flotation due to air bubbles occurring in the fines trap. The non ideal sharpness of the actual efficiency curve may result from hydrodynamic effects in the classifying section. Furthermore, classifying conditions will change with varying fines removal flows. The following nonlinear two-parameter function is proposed to describe the fines removal efficiency:

$$h_f(x,t) = \frac{1}{1 + \left(\frac{x}{x_c(t)}\right)^{p/2}}, \quad (2.9)$$

with $x_c(t)$ the cut-size, defined as the crystal length where the probability that a particle does not survive the fines trap is 50%. Because the solid concentration in the
Fig. 2.4: Examples of classification functions \( h_f \) and \( h_p \), and the growth function \( G_x \).

The fines loop is low (< 1%), Stokes’ law is applied, to derive a relation for the cut-size [3, 228]. This gives:

\[
x_c = \sqrt{\frac{18 \mu v}{(\rho_s - \rho_l) g}}
\]

(2.10)

where \( v \) is the average liquid velocity, \( \mu \) the absolute viscosity of the mother liquor, and \( \rho_s \) and \( \rho_l \) the density of the solid and liquid phase, respectively. Substitution of \( Q_f = v A \), where \( A \) is the cross-sectional area of the fines classifier gives:

\[
x_c(t) = \sqrt{\frac{p_f}{A(\rho_s - \rho_l) g}}
\]

(2.11)

The constant factor \( p_f \) is introduced to correct for errors due to the asphericity of the particles, hindered settling effects, the existence of a flow profile and errors in the value for the absolute viscosity as obtained for instance from a handbook. As depicted in Fig. 2.4, with Eqn. 2.9 a S-shaped curve is obtained with a point of inflection given by the cut-size and a degree of imperfection of the classifier given by the parameter \( p_f \).

The separated fines are dissolved by heating of the fines slurry flow. To model fines dissolution all fines are assumed to dissolve. With this assumption two mass balances and a heat balance over the fines section are derived. The corresponding equations are summarized in Appendix A.
2.2 Process modelling

Product classification

Product classification can be established with mechanical separators, for example, vibrating screens; gravitational separators, for example, hydraulic classifiers; and centrifugal separators, for example, hydrocyclones. For these systems the product size distribution \( n_p \) can be calculated, analogously to the fines removal function, from:

\[
n_p(x, t) = \frac{Q_{pf}(t)}{Q_p(t)} h_p(x, t) n(x, t),
\]

(2.12)

where \( Q_p \) is the product flow, \( Q_{pf} \) the unclassified flow from the crystallizer and \( n(x, t) \) the population density of crystals in the crystallizer. The corresponding product flow split factor is now defined by:

\[
\frac{Q_{pf}(t)}{Q_p(t)} = \gamma_p = \frac{\epsilon_r(t) - \epsilon_p(t)}{\epsilon_r(t) - \epsilon_{pf}(t)},
\]

(2.13)

with \( \epsilon_r \), \( \epsilon_p \), and \( \epsilon_{pf} \) the void fractions of the return flow, the product flow, and the classifier feed flow, respectively. These void fractions are calculated from Eqn. 2.7. The following general model for classification is used to describe product classification:

\[
h_p(x) = \frac{p_{p3} + (1 - 2p_{p3}) \left( \frac{x}{p_{p1}} \right)^{p_{p2}}}{1 + (1 - 2p_{p3}) \left( \frac{x}{p_{p1}} \right)^{p_{p2}}}.\]

(2.14)

Three empirical parameters are used: the cut-size \( p_{p1} \), the imperfection \( p_{p2} \), and an offset \( p_{p3} \). The latter parameter defines the void fraction, leaving the crystallizer with the separated product. This factor is also called the dead-flux [228] We assume that \( 0 < p_{p3} < 0.5 \), to obtain a positive and monotonically increasing function. An example of this product classification function is given in Fig. 2.4. Notice, that for certain devices more complicated separation efficiency functions, like for instance non-monotonically increasing functions, may be more feasible. More detailed ad-hoc models for solid-liquid separators are provided by Svarovsky [228], and for the separators used in the UNIAK project by Gerla [86]. The total set of empirical parameters, introduced so far to describe fines and product classification, is denoted as:

\[
\theta_C = \{p_{f1}, p_{f2}, \{p_{p1}, p_{p2}, p_{p3}\}\}.
\]

(2.15)

A relation for the product flow

For classified removal of product slurry, the product flow \( Q_p \) is usually not directly manipulable, but depends on the classification function, the feed flow, and the void fraction of the feed flow. Two cases are considered:

1. **Constant dead-flux.** Assuming \( p_{p3} \) to be constant will imply that a constant fraction of the mother liquor void fraction is transferred to the product outlet.
For this case the product flow can be written as the sum of the void flow and the solids flow:

$$Q_p = Q_p \epsilon_p + Q_p(1 - \epsilon_p)$$

(2.16)

The void flow is calculated from:

$$Q_p \epsilon_p = Q_p \frac{Q_{pf}}{Q_p} \left[ 1 - k_v \int_0^\infty nx^3 dx \right]_{p_{p3}}$$

(2.17)

The solids flow is given by:

$$Q_p(1 - \epsilon_p) = Q_p k_v \frac{Q_{pf}}{Q_p} \int_0^\infty h_p nx^3 dx$$

(2.18)

Substitution of Eqn. 2.17 and Eqn. 2.18 into Eqn. 2.16 gives the following expression for the product flow:

$$Q_p = Q_{pf} \left[ [1 - k_v \int_0^\infty nx^3 dx]_{p_{p3}} + k_v \int_0^\infty h_p nx^3 dx \right]$$

$$= Q_{pf}(t) \left[ [1 - k_v m_3(t)]_{p_{p3}} + k_v \int_0^\infty h_p nx^3 dx \right]$$

(2.19)

2. Constant void fraction in product. For some classification systems (e.g., a vibrating screen classifier) it is reasonable to assume that the product void fraction is constant. For these systems the dead-flux $p_{p3}$ which is used in Eqn. 2.16 will not be constant. Substitution of Eqn. 2.18 into Eqn. 2.17 gives the following implicit equation

$$\int_0^\infty \frac{h_p}{p_{p3}} nx^3 dx = \frac{(1 - \epsilon_p)}{k_v \epsilon_p}$$

(2.20)

which relates the offset $p_{p3}$ to the known values for the void fraction of slurry in the crystallizer $\epsilon$ and the constant product void fraction $\epsilon_p$. The latter is often calculated from the measured slurry density using

$$\epsilon_p = \frac{\rho_{sl} - \rho_c}{\rho_l - \rho_c},$$

(2.21)

with $\rho_l$ the mother liquor density and $\rho_c$ the crystal density. Substitution of Eqn. 2.14 into Eqn. 2.20 results in an implicit equation for $p_{p3}$, which can be solved numerically. We will apply this in Chapter 5.

2.2.3 Modelling crystallization phenomena

Before modelling of crystallization phenomena (like, crystal nucleation, growth and abrasion), an assumption has to be made on hydrodynamics and mixing occurring in the process.
2.2 Process modelling

Hydrodynamics

In the first instance it is assumed that the crystallizer volume is ideally mixed. Two phenomena do violate this assumption:

1. The existence of supersaturation profiles.

2. Spatial segregation of larger particles due to settling.

Calculations done by Jager [120] show that a significant supersaturation profile exists along the draft-tube of an evaporative DTB crystallizer. For instance in the UNIAK pilot crystallizer (cf. Fig. 3.1), the supersaturation in a liquid volume that travels along the draft-tube will reach a peak value right after the boiling zone and the bottom value (which may be an undersaturated solution) is present right after the volume has passed the internal heat exchanger. The existence of a supersaturation profile will have two important implications: the growth rate of the particles will not be constant and fines may dissolve in the locally undersaturated solution. Notice that well-growing and well-soluble substances (i.e., Class II systems; see also Section 2.2.5) are probably more strongly affected by supersaturation differences.

Jager et al., [120] use a so called compartment model (see e.g., [15, 30]) to study the effect of supersaturation profiles. Their simulation results show that for larger scale equipment the cycle time increases while the dampening decreases. In addition, they show that without internal fines dissolution (due to local undersaturation) no oscillations are observed at all. They conclude from this that the introduction of a supersaturation profile significantly affects the predicted CSD dynamics, especially at a larger scale of operation. Recent results, presented in [65, 175], do not support this idea. In [175] it is found that the cycle period is equal for a 20 and a 970 litres DTB crystallizer. Furthermore, experimental results presented in Chapter 5 of this thesis suggest that differences in cycle times are mainly explained by differences in the mean slurry residence times. The dampening of oscillations is found to depend strongly on the fines removal rate.

Therefore it seems more appropriate to assume the supersaturation profile to introduce a rapid dynamic phenomenon, especially for the growth rate of crystals, which does not relate to the macroscopic CSD dynamics observed in the product CSD. Consequently, this phenomenon may be neglected, without loss of accuracy by assuming average supersaturation levels causing an average growth rate.

Spatial segregation of particles, however, will have a significant effect. In [86, 92, 95, 120], some preliminary results of a mixing study in a 1 m³ DTB crystallizer, equipped with a marine type impeller are reported. In [95, 120] the circulation speed of slurry at 300 rpm impeller speed is estimated to be 350 mm/s. Experimental results [135] show that the settling velocity of ammonium sulphate crystals is significant and decreases for larger void fractions. A velocity of 7.5 mm/s was found for crystals with a size of 400 μm and a void fraction of 80%. Clearly, settling will be larger when the density difference between the solid and liquid phase becomes larger.
Particle settling has two major consequences. A first consequence is that larger crystals will probably not reach the boiling zone, and consequently have retarded growth rates. We include this phenomenon as a length dependent growth rate function given by Eqn. 2.26, which is also used to describe attrition.

A second consequence is that the measured slurry density and the CSD will depend on the sensor location. In particular, for DTB-type crystallizers spatial segregation causes the most significant difference between the CSD in the inner and outer draft-tube, as is illustrated with Fig. 2.5.

Accurate modelling of slurry transport is difficult as highly complicated particle/liquid interactions occur, which will depend on the liquid and solid substance properties, the tube or reactor geometries, the particle geometry and the particle size. Oroskar [178] gives a review of correlations for the critical velocity of particles in tubes. The critical velocity (denoted as \( v_c \)) is the minimum velocity of a flow in which the solids form a bed; Alternatively, it denotes the velocity at which the particle settling velocity equals the upward velocity due to the drag force exerted on its surface by the liquid.

A simple equation that relates the distributions in the inner and outer draft-tube of a DTB crystallizer can be obtained from the particle number balance. If a stationary distribution of particles is assumed this gives:

\[
    n_{in} = n_{out} \frac{v_l - v_c}{v_l + v_c}
\]

with \( v_l \), the liquid flow in the inner and outer draft-tube and \( n_{in} \) and \( n_{out} \), the populations in the inner and outer draft-tube respectively. The liquid velocity \( v_l \) will mainly
depend on the impeller speed.

Notice that Eqn. 2.22 predicts the distribution measured in the outer draft-tube to have a lower number of large particles than present in average in the vessel. Another point to note is that due to spatial segregation changes in the impeller speed will directly affect the measured crystal size distribution. In Section 5.2.3 some experimental data UNIAK pilot crystallizer is shown that confirms this observation. Because spatial segregation is difficult to quantify accurately, it is not incorporated explicitly in the crystallizer model. Consequently, especially the size dependent growth function discussed below may get biased.

**Secondary nucleation**

The mechanisms of nucleation or birth of new crystals is generally classified into primary and secondary nucleation. Secondary nucleation is defined as "nucleation that takes place only because of the prior presence of crystals of the material being crystallized" [23]. Primary nucleation denotes the formation of a solid phase from a clear mother liquid, without the catalic effect of other particles. This type of nucleation is further subdivided into homogeneous and heterogeneous nucleation. The latter is assumed to start on foreign substrate particles, for example dust particles. In the former case the onset of nucleation is assumed to occur due to clustering of molecules at a highly supersaturated solution lacking a foreign substrate. In many papers evidence is found for the fact that primary nucleation will not occur in dense slurries at moderate growth conditions [83]. Therefore, secondary nucleation is assumed to be the dominant nucleation mechanism. Further it is assumed that nucleation takes place at a crystal size $x_0$, which denotes the left bound of the crystal size domain.

Secondary nucleation indicates that nuclei will originate only because of the presence of other crystals. These secondary nuclei are assumed to originate from two different breeding mechanisms [40]: mechanical and surface breeding. Mechanical breeding is a surface abrasion effect that can even occur in undersaturated solutions. Surface breeding is an attrition phenomenon that strongly depends on the supersaturation, because it depends on the surface structure of the crystals, that is a result of supersaturation driven growth. Results presented by Garside and Larson [84] confirm the presence of two sources of nuclei. Experiments with potash alum crystals revealed that most of the nuclei were in the size range 1-10 $\mu$m, and could be produced in supersaturated as well as undersaturated solutions. Additionally, fragments in the range up to 50 $\mu$m were formed only under conditions of growth, and the number of these fragments was proportional to the supersaturation level. In the framework of Daudey et al., [40] these observations are classified as mechanical and surface breeding, respectively. Detailed models for surface breeding can be found in [240]. Daudey [40] further distinguishes three subsequent steps in both breeding mechanisms: a formation, a removal and a survival/outgrowth step. In [40, 240] models are formulated for these steps.
Well-defined models are developed for the mechanical breeding mechanisms. Ottens [179] presents a framework which describes particle formation rates as a function of collision energies. In the papers of Pohlisch and Mersmann [155, 185, 186] detailed models are presented for the generation of small particles within a mechanistic framework. Garside [83], reviews simple empirical power laws that relate the stirrer speed or pump rotation rate to the secondary nucleation rate. In Fig. 2.6 an overview is given of the different nucleation and breeding mechanisms.

Published empirical models, that describe secondary nucleation in a suspension crystallizer, are mostly power law models [197, 83, 155, 239]. These models describe both mechanical and surface breeding phenomena by relating the effective nucleation rate to the growth rate and the saturation condition of the mother liquor, on the one hand, and the presence of other particles, which are subject to particle-particle and particle-impeller interactions, on the other hand. An extensive overview of these secondary nucleation models is given by [241]. In [176] a model is presented that explicitly relates the secondary nucleation rate to the rate of attrition.

In this work the following power law model is used.

\[ B(t) = p_3 \left[ \int_{p_4}^{\infty} n(x, t)x^{p_5}dx \right]^{p_1} \Delta C(t)^{p_2}, \]  

(2.23)

with \( p_1, \ldots, p_5 \) empirical constants. The factor \( p_3 \) describes the combined effect of stirrer speed, pump rotation rate, slurry temperature, crystallizer geometry, and so on. This model differs from most standard power law models present in the literature in that a lower bound of integration (parametrized with \( p_4 \)) is assumed. The idea of using this lower bound was published first by Ottens [179].

With this lower bound the contribution of small crystals to secondary nucleation is neglected. This assumption is valid as small crystals are exposed to insignificant hydrodynamic and mechanical forces as compared to large crystals and therefore do not contribute to the aforementioned breeding mechanisms. The numerical value will
depend on the density difference between the crystal and the solution and the flow conditions in the near vicinity of the crystal. The images depicted in Fig. 3.11 further support this assumption in that small crystals are sharply edged and ideal shaped, while larger crystals are strongly rounded due to mechanical breeding induced by collisions. From simulation studies (cf. Chapter 5) we found that CSD cycles can be described with this model, without the large physical meaningless values for the powers, which are needed to describe cycles with the standard power law model [197].

**Crystal growth kinetics**

The mechanism of crystal growth requires two successive steps: a diffusion step, which is followed by a surface integration step of the solute crystal material. Occasionally it is assumed that a growth rate distribution function exists for crystals of the same size [49]. This phenomenon, is called growth rate dispersion (GRD). Reviews of published work on GRD are provided in [83, 260]. The effect of GRD on the CSD will be most pronounced between smaller particles ($x < 100 \mu m$). However, accurate on-line measurement of smaller particles in a crystallizer is difficult because of the lack of sufficiently accurate sensors. Therefore, in first instance GRD is omitted also to prevent overparametrization of the model.

The (macroscopic) kinetic growth rate, as a result of diffusion and surface integration of molecules, is often assumed to depend on the supersaturation. Here a standard power law model, as proposed among others by [70, 130, 197], is used to describe the kinetic growth rate as a function of the supersaturation:

$$G_k(t) = p_0 \Delta C(t)^p.$$  \hspace{1cm} (2.24)

Alternatively a relative measure of supersaturation is often used, for example $\Delta C/C_s$. In this case the constant factor is denoted as $p_0^*$, with $p_0^* = p_0 \times C_s$. The unit of this constant is [m/s]. Numerical values for $p_0^*$ and $p_r$, as a function of the temperature, are given for different substances by: [129]. Published data reviewed by Daudy [39, p. 165] show that for ammonium sulphate $p_0^*$ has a value in the range of $1 \times 10^{-3}$ to $2.5 \times 10^{-4}$. In Section 2.2.5 the influence of this parameter on the static and dynamic process behaviour is studied.

For an MSMPR crystallizer the stationary CSD will be described by a straight line in the semi-logarithmic population density plot [197]. In practice, a CSD will decrease after a certain size [185, 121]. This phenomenon is denoted as growth retardation. The retardation of the crystal growth rate can be the result of: attrition or length dependent growth and spatial segregation of large particles, as already discussed. Attrition of crystals is an abrasion or surface erosion effect that describes the reduction in size of crystals due to collisions among crystals or crystals and the crystallizer hardware. The attrition rate is in principle related to the rate of nucleation. In [121, 185, 155], different models for the attrition of large crystals are presented. Here the attrition rate is modeled as a length dependent growth function. To prevent overparameterization
of the model, the same function is also used to describe the retardation of growth caused by spatial segregation of larger particles. Notice that a direct coupling between secondary nucleation and attrition could therefore not be made in the model.

The effective growth rate is assumed to be the product of the kinetic growth rate and the length dependent growth function:

$$G_e(x,t) = G_k(t)G_x(x).$$  \hspace{1cm} (2.25)

The empirical model for this length dependent growth function is chosen to be:

$$G_x(x) = 1 - \frac{x^p \alpha + x^p \alpha}{x^p \alpha + x^p \alpha},$$  \hspace{1cm} (2.26)

which is an S-shaped curve, with $x_\alpha$ as the point of inflection and $x_e$ as the maximum simulated crystal size, both parametrized by the unknown parameters $p_\alpha$ and $p_10$, respectively. The sharpness of this curve is parametrized by $p_\alpha$. Fig. 2.4 describes a typical shape of the length dependent growth curve.

The total set of kinetic parameters, introduced in the model so far to describe effective growth and nucleation, is denoted as: $\theta_K = \{p_1, \ldots, p_10\}$.

2.2.4 Initial state

The initial state of the crystallizer considered here is the initial CSD and the initial saturation level of the mother liquor (Eqns. 2.29 and 2.32). The initial CSD can originate from a distribution of seed crystals or results from heterogeneous primary nucleation in a crystal-free supersaturated liquid.

A Rosin-Rammler [197] distribution is chosen for the initial CSD of the crystallizer. This distribution fitted well to the experimental data (cf. Section 5.3), however, simulation results indicate that other distributions like log-normal or inverse gamma distributions give comparable results. The Rosin-Rammler distribution contains three parameters: $\theta_I = \{p_{i1}, p_{i2}, p_{i3}\}$ related to the location, the spread, and the height of the initial CSD:

$$n_s(x) = p_{i1} p_{i2} p_{i3}(x^{p_{i3} - 1}) \exp(-p_{i1}x^{p_{i2}}).$$  \hspace{1cm} (2.27)

The initial supersaturation is given by $\Delta C_s$. This value is often difficult to measure. However, the supersaturation decay due to growth of particles is a rapid dynamic phenomenon. Thus, errors in this value are not of great concern, if one is interested in the slower CSD dynamics only. In most CSD simulations we have taken $\Delta C_s$ equal to zero.

2.2.5 Balance equations

The empirical relations are embedded in a set of balances to obtain a parametrized model for the crystallizer dynamics. A population balance is used to describe the
2.2 Process modelling

dynamics of the CSD. A heat balance with a set of mass balances, for respectively the solution, the solvent and the solids concentration in the slurry, describes the dynamics of the slurry properties. For the mass balances, a crystal mass, a salt mass and a total mass balance are used. A detailed derivation of these equations is reported by De Wolf [49]. The balance equations used here are provided in Appendix A for the class of well-mixed, non isothermal, non constant volume, batch as well as continuous, and evaporative as well as cooling crystallizers. In addition to the assumptions described in the previous sections, it is assumed that: the stirrer speed is constant; the vapour flow \( Q_v \) contains no solute or crystalline material; and nucleation takes place at a negligible small size \( x_0 \), so \( \Psi_1 = 0 \) (Eqn. A.3). Further the heat consumption needed for crystallization is taken as:

\[
P_{\text{crys}} = k_{\text{crys}} \Psi_2
\]

(2.28)

where \( k_{\text{crys}} \) is the heat of crystallization, and \( \Psi_2 \) is the solids mass production rate due to growth, as described by Eqn. A.4. Furthermore, it is assumed that only one pure component is present in the crystal lattice.

The model equations can easily be adapted to describe a cascade of crystallizers, crystallizers equipped with a clear liquor advance, or for instance double draw-off crystallizers [197].

The reduction of the general model equations (provided in Appendix A) to a set of simplified equations is presented for the continuous UNIAK DTB-type crystallizer introduced in Chapter 3.

For this crystallizer the following additional assumptions are made: the crystal volume is constant; the temperature is constant; the heat consumption for crystallization \( P_{\text{crys}} \), the heat loss \( P_{\text{loss}} \) and the heat supplied with pumps and impellers \( P_{\text{imp}} \) are negligible compared to the heat consumed for solvent evaporation; the feed flow \( Q_f \) has a constant temperature and is free of crystals; The main model assumptions for both the general model and the pilot crystallizer model are summarized in Appendix C.

**Population balance**

With the above mentioned assumptions the population balance Eqn. A.1 is written as:

\[
\frac{\partial n(x,t)}{\partial t} + \frac{\partial G_e(x,t)n(x,t)}{\partial x} + \frac{Q_f(t)}{V} \gamma_f(t)h_f(x,t)n(x,t) + \frac{Q_{pf}(t)}{V} h_p(x,t)n(x,t) = 0,
\]

(2.29)

with the boundary condition:

\[
n(x = x_0,t) = B(t)/G_e(x_0,t)
\]

(2.30)

and the parametrized initial condition: \( n_s(x,t = t_0,\theta_f) \). Note that the flow split factor \( \gamma_f \) is introduced here, because \( Q_f(t) \) instead of the unclassified slurry flow \( Q_{ff} \) (Eqn. 2.8) is used.
An equation for the supersaturation

Several internal variables from the balances presented in Appendix A can be eliminated. First a concentration balance is obtained by substitution of the crystal mass balance (Eqn. A.5) in the salt balance (Eqn. A.6). An equation for the supersaturation is derived by substitution of the supersaturation:

$$\Delta C(t) = C(t) - C_s,$$

into Eqn. A.19. Results described in [29, p. 261] show that for non isothermal systems (e.g., cooling batch crystallizers), the use an absolute measure of supersaturation may lead to false conclusions. For these systems a relative measure of supersaturation (e.g., $\Delta C/C_s$) should preferably be used. Here the absolute concentration is used. The transformation to relative supersaturation is straightforward.

Substitution of the crystal mass balance (Eqn. A.5) in the total mass balance gives an expression for the input flow $Q_i$. An expression for the vapour flow $Q_v$ is obtained by substitution of Eqns. A.11 and A.9, in the heat balance (Eqn. A.8). Substitution of the expressions for $Q_i$ and $Q_v$ in the supersaturation balance then results in the following equation for the supersaturation:

$$\frac{d\Delta C(t)}{dt} = \frac{1}{V \varepsilon(t)} \left[ -Q_p(t) - Q_f(t)[1 - \varepsilon_f(t)] + \frac{\Psi_2(t)}{\rho_c} \right] \Delta C(t)$$

$$+ k_{c1}Q_{pf}(t) + k_{c2}Q_f(t)[1 - \varepsilon_f(t)] + k_{c3}Q_f(t) + k_{c4}P_{tot}(t) + k_{c5} \frac{\Psi_2(t)}{\rho_c}$$

(2.32)

in which $k_{c1}, k_{c2}, k_{c3}$ and $k_{c4}$ are constants determined by the operation conditions, the geometry of the crystallizer and the physical properties of the substance system. The variable $\Psi_2(t)$, which describes the solids mass production rate due to growth, is given by Eqn. A.4. Note that for classified product removal Eqn. 2.19 or Eqn. 2.20 should be substituted for the product flow $Q_p$.

The Class II assumption

Most industrial evaporative and cooling-type crystallizers operate on relatively well soluble substances like KCl, NaCl, or (NH₄)₂SO₄ [154]. Sparingly soluble substances like CaCO₃ or BaSO₄ are mostly crystallized by precipitation, which involves a reaction mechanism that is outside the scope of this work. The main characteristic of well soluble substances is that the supersaturation decay due to growth is fast as compared to that of sparingly soluble substances. For these so-called Class II systems [197] the supersaturation balance can be transformed to a simple algebraic constraint for the crystal growth rate. The Class II model is derived in Appendix A and further evaluated in Section 5.2.2.
2.2.6 Sensor modelling

The population balance model is extended with a sensor model to enable validation of the model with measured sensor data, as we will do in Chapter 5 and 6. At first a model will be derived for a sensor based on forward light scattering, which is used as the main sensing technique in this work (cf. Section 3.3.3). Secondly, a simple empirical model for a particle counter will be presented (cf. Section 3.3.4).

Modelling forward light scattering

Fig. 3.8 indicates schematically the operating principle of this method. The scattering of an incident laser beam by a population of differently sized particles, flowing through an optical cell, can be described using Fraunhofer diffraction theory [24, 99]. The resulting models are denoted as diffraction models. One of the major assumptions underlying Fraunhofer diffraction theory is that light does not pass through the particles itself. In addition, a difficulty in applying the theory is that it is difficult to account for multiple scattering which might occur when the solids concentration is too high [106]. However, as discussed in Section 3.3.2, this effect is reduced sufficiently with a slurry diluter. Therefore, we do not consider it further.

A complete theory for describing light scattering by particles is called the Mie theory. For small scatter angles and small refractive indices also the so called anomalous diffraction theory can be applied [242]. Both theories do account for light travelling through the particles, which is most significant when the ratio of the solid to medium (liquid) refractive indices, (denoted as the relative refractive index) is close to unity.

The relative refractive index for ammonium sulphate crystals is 1.07. Heffels [102] shows that a significant amount of light may travel through particles for this value. A model using Mie can, however, only be evaluated numerically assuming spherical particles, due to the large computational burden involved in the evaluation of nonspherical, for example, rectangular-shaped crystals. The same author indicates that the errors involved in neglecting this phenomenon are much smaller than the errors involved in the wrong shape assumption, for example, spheres versus rectangles. Therefore, it is decided to use a light scattering model based on Fraunhofer diffraction theory using information on the particle shape. For an elaborate discussion on this topic we refer to [102]. In Section 5.3.5 we will show that the error caused by neglecting light travelling through particles may be (partially) responsible for a difference between measured and simulated diffraction data.

The diffraction model is developed for rectangular-shaped crystals with an average aspect ratio $\alpha$, assuming that only their rectangular shapes are projected on the detector plane. The Fraunhofer model describes the light energy $e$ at a location P $(\theta, \phi)$ in the polar plane scattered by a particles with normalized size $\lambda$ and aspect ratio $\alpha$ as [99, 102]:

Fig. 2.7: Scattering of light by a rectangular aperture (after [99]).

\[
e(\lambda, \theta, \phi, \alpha) = \frac{\left[1 - \cos(2\lambda \theta \cos(\phi))\right] \left[1 - \cos(\alpha 2\lambda \theta \sin(\phi))\right]}{4\theta^3 \cos(\phi)^2 \sin(\phi)^2}
\]  

(2.33)

See also Fig. 2.7 for an explanation of the variables involved. This model has been evaluated for the specific case of a Malvern particle sizer, which employs a multi ring detector to record the scattered light (cf. Section 3.3.3). The total light energy \(l_i\) observed on a single detector ring \(i\) with respective inner and outer radii \(r_i\) and \(r_{i+1}\), due to scattering of particles with a size distribution \(n\) in the size range \([x_i, x_{i+1}]\), is obtained by integration of Section 2.33 for one specific value of \(\alpha\):

\[
l_i = \int_{\lambda=x_j}^{\lambda=x_{j+1}} \int_{\theta=\theta_i}^{\theta_{i+1}} \int_{\phi=0}^{2\pi} e(\lambda, \theta, \phi, \alpha)n(\lambda) d\theta d\phi d\lambda
\]  

(2.34)

Note that \(R \gg R \sin \theta\), thus it reasonable to assume that \(\psi_i = r_i\), with \(r_i\) the inner radius of the \(i\)th detector ring, and \(R\) the length of the beam. With this relation, the so called diffraction matrix is computed that maps a discrete (simulated) CSD onto a finite set of energy values measured from the detector elements:

\[
L = G(\alpha)n,
\]  

(2.35)

with \(L\) the vector containing the light energy values. Integration over the three variables \(\{\lambda, \theta, \phi\}\) is performed with the NAG routine D01FCF [150]. Details on this and an elaborate discussion on several other assumptions made in this model are given in [102].

Note that in Chapter 5 the diffraction matrix is used in the simulation model to calculate the sensor output, directly from simulated distributions. Herewith, the numerically ill conditioned inversion of the sensor model to obtain measured data in the same format as the simulated data, is avoided.
2.2.7 Modelling the particle counter

The second CSD sensor employed in this work is a fine particle counter. This sensor comprises a settler and an optical counter, (see also Section 3.3.4). A model has been developed for the combined system of settler and counter. The probability that a fine particle in the crystallizer is detected by the counter can be described by both a separation efficiency, and an optical detection probability. A physical model describing the efficiency with which particles are detected as a function of their size (denoted as detection efficiency) is, however, difficult to develop. Therefore, an overall empirical detection efficiency function \( K_S(x) \) is introduced. This function describes the combined probability that a particle is removed from the crystallizer volume using a simple gravitational settler, and detected in the optical cell by blocking a sufficiently large part of the incident laser beam. When noise sources are absent, the counted number of fines \( N_f \) is modelled as the weighted zeroth moment of the population density function:

\[
N_f(t) = \int_0^\infty K_S(x)n(x,t)dx. \tag{2.36}
\]

The detection efficiency function is approximated with a standard log-normal distribution:

\[
K_S(x) = \frac{p_{s3}}{\sqrt{2\pi}\ln(p_{s2})} \exp\left(-\frac{\ln(x/p_{s1})}{\sqrt{2}\ln(p_{s2})}\right) \frac{1}{x}. \tag{2.37}
\]

The parameter \( p_{s1} \) denotes the location, and \( p_{s2} \) the spread of the distribution. In the sequel the term location denotes the crystal size where the detection efficiency curve reaches a maximum. The parameters \( p_{s1} \) and \( p_{s2} \) are estimated from experimental data. This will be discussed further in Chapter 5.

2.3 Development of analysis tools

This section first outlines the numerical tools needed for simulation and analysis of the crystallizer model. Secondly, a programming environment is presented, in which all tools have been implemented. This software comprises several numerical approximation schemes, which will be discussed in detail in the subsequent sections.

2.3.1 Basic functions and tasks

The model derived in the previous section is a nonlinear first order hyperbolic partial differential equation, which can be written as:

\[
\frac{\partial z(x,t)}{\partial t} = f_1(z,x,t,\theta) \frac{\partial z(x,t)}{\partial x} + f_2(z,x,t,\theta)z(x,t) \tag{2.38}
\]

and the boundary condition:

\[
z(x_0,t) = f_3(z,x,t,\theta) \tag{2.39}
\]
where $\theta$ denotes the set of empirical parameters, comprising uncertain parameters for crystallization kinetics $\theta_K$, the initial condition $\theta_I$, the classification devices $\theta_C$, and the sensor $\theta_S$:

$$\theta = \{\theta_K, \theta_I, \theta_C, \theta_S\}$$  \hspace{1cm} (2.40)

Several software tools are required that fulfill the following functions:

- Estimation of the empirical parameters $\theta$ on the basis of measured process data.
- Provide understanding on the process dynamic behaviour.
- Assess controllability at both dynamic and steady-state conditions.
- Serve as an observer that extracts relevant information from the raw process data when they become available from the on-line sensor (see e.g., Chapter 7).
- Design and evaluation of robust controllers.

Analytic solutions for the model can be derived for the linearized population balance in the frequency domain, for some specific process configurations. These will be employed later in this section. Generally, however, to arrive at a time domain solution of a partial differential equation, numerical schemes are required that first approximate this equation with a finite set of ordinary differential equations. In a second step these can be solved with a standard numerical integration scheme. In addition, linearization of the nonlinear model at a certain stationary condition is necessary, to apply standard control design methods. Finally, the linearized and lumped population balance model will have a dynamic order that is too high for control design applications. Therefore, model order reduction schemes are desired as well.

In conclusion, to realize the basic software functions discussed above the following tasks have to be performed:

**Lumping**
Approximation of the partial differential equation by a finite set of ordinary differential equations.

**Nonlinear simulation**
Simulation of the nonlinear equations obtained after lumping.

**Parameter estimation**
Numerical estimation of empirical parameters, on the basis of experimental data, using a numerical search technique.

**Stationary solution**
Estimation of stationary conditions for a given set of process input parameters.
2.3 Development of analysis tools

Linearization
The derivation of a linearized model at a certain stationary condition.

Model reduction
Reduction of high order linear models to low order models that still give a sufficient approximation of the control relevant dynamics.

User interface
The display of results using a graphical user interface.

We will focus onto the software package first, and subsequently discuss the numerical schemes. Parameter estimation is further discussed in Chapter 5.

2.3.2 Software implementation

The simulation of distributed parameter systems often involves the simultaneous solution of large sets of differential equations. Here a custom made program is developed in Fortran to perform the nonlinear simulation task. Use is made of some basic modules from the DSS/2 package [184]. The Fortran model is linked directly to Matlab to achieve a simulation environment, in which all graphic, design and analysis tools are readily available. The implemented numerical tools will be discussed in more detail below. Appendix E summarizes the main software tools used.

Alternative tools are standard packages like Matlab Simulink[152], MatrixX System-Build[116], or Speedup[7]. The latter has the advantage that it is most well known in process industry and has a good interface to property databases. A new tool called GPROMS [12] is being developed, which offers promising perspectives for the simulation of partial differential equations.

2.3.3 Analytic solutions

An analytic solution of a simplified population balance model in the Laplace domain is used to analyze the accuracy of the numerical schemes. For this the following simple transport equation given by Eqn. G.1 is used that describes the CSD dynamics in a finite size domain \( 0 < x \leq x_e \). The growth rate is assumed to be constant and the removal efficiency of product particles is described by the time independent function \( h(x) \). Fines removal is assumed to be absent. The left boundary condition is formulated as an independent disturbance input (cf. Eqn. G.2). The product removal flow \( u_1(t) \) is considered as an input parameter. The stationary solution for this model is given by Eqn. G.3. A sensor is assumed that detects particles with an efficiency \( k(x) \). With this efficiency function a scalar sensor output is described as a function of the population density according to:

\[
y(t) = \int_{x=0}^{x_e} k(x)n(x, t)dx
\]  
(2.41)
with \( x_e \) the largest crystal size accounted for in the model. In the sequel \( k(x) \) will be denoted as the detection efficiency of the sensor.

In Appendix G this model is linearized, and transformed to the Laplace domain using the work of Franke [76]. In the following the \( \Delta \)-symbol, introduced in the linearized model, is omitted for notational convenience. The transformed model presented in Appendix G is simplified here for the case that all particles are removed from the process continuously with the same efficiency, that is: \( h(x) = 1 \) for \( 0 < x < x_e \). Further two different sensor configurations are assumed.

1. The sensor detects only monosized particles with a size \( x = x_k \) with probability \( 1: k(x) = \delta(x - x_k) \).

2. The sensor counts all particles in the same size range with an uniform probability: \( k(x) = 1 \) for \( 0 < x < x_e \).

These two cases are summarized with the following transfer function model:

\[
\begin{bmatrix}
y_1 \\
y_2
\end{bmatrix} = \begin{bmatrix}
G_1 & G_2 \\
G_3 & G_4
\end{bmatrix}
\begin{bmatrix}
u_1 \\
u_2
\end{bmatrix}
\] (2.42)

with \( u_1 \) the removal rate, \( u_2 \) the boundary condition, \( y_1 \) the monosize sensor output, and \( y_2 \) the output of the sensor with an uniform particle detection efficiency. In the following, explicit formulas for these transfer functions are given.

**Monosized detection**

With \( k(x) = \delta(x - x_k) \), and \( h(x) = 1 \) for \( 0 < x < x_e \), the distributed transfer functions Eqn. G.14 and Eqn. G.15 are written as:

\[
G_1(s) = G_{x1}(x_k, s) = \frac{-\tilde{u}_2}{s} \exp\left( -\frac{\tilde{u}_1}{g} x_k \right) \left[ 1 - \exp\left( -\frac{s}{g} x_k \right) \right]
\] (2.43)

\[
G_2(s) = G_{x2}(x_k, s) = \tilde{u}_2 \exp\left( -\frac{\tilde{u}_1}{g} x_k \right) \exp\left( -\frac{s}{g} x_k \right)
\] (2.44)

The Bode plot for \( G_1 \) is plotted in Fig. 2.10. This plot shows that for specific frequencies of the input signal \( u_1 \) the output \( y_1 \) remains zero. These frequencies correspond with so called transmission zeros [110]. These are computed easily by substituting \( s = \delta + j\omega \) in Eqn. 2.44. Straightforward algebraic manipulation then gives for the real and the imaginary part of \( G_2 \):

\[
\Re\{G_1\} = \exp(\delta\tau) - \cos(\omega\tau) + \frac{\omega}{\delta} \sin(\omega\tau)
\]

\[
\Im\{G_1\} = -\exp(\delta\tau) + \cos(\omega\tau) + \frac{\delta}{\omega} \sin(\omega\tau)
\]
where \( \tau = x_k / g \). At the zeros both the imaginary and the real part will be zero. This will be the case for:

\[
\omega = 2i\pi g / x_k \quad i = \pm 1, \pm 2, \ldots,
\]

\[
\delta = 0
\]  

(2.45)

Hence, if the removal rate is varied in time according to: \( u_1 = \sin(2\pi g / x_k t) \) (or at multiples of this frequency) the number density \( n(x_k) \) measured with the sensor at \( x_k \) remains zero. Clearly, disturbances in the distribution caused over the first half period of the sinusoidal disturbance at \( u_1 \) are then compensated exactly by the second half of the sinusoid, when they have grown to a size \( x_k \).

The solution for \( G_2 \) reveals a very simple delay equation. For this case the relation between the input and output signal is given by:

\[
y_1(t) = n(x_k, t) = u_2(t - \tau) \exp(-\tau x_k), \quad \text{with, } \tau = \frac{x_k}{g}
\]  

(2.46)

This relation demonstrates that, without compensating control actions, disturbances at the input \( u_2 \) are transferred to the output \( n(x_k, t) \) with a delay \( \tau \), and reduced by amplitude with a factor \( \exp \left( -\frac{\tau}{g} x_k \right) \). Clearly, the delay time is proportional to the location of the sensor \( x_k \), and inversely proportional to the growth rate.

**Uniform detection and uniform removal**

For this case it is assumed that \( k(x) = 1 \) for \( 0 < x < x_e \) and \( h(x) = 1 \), with \( 0 < x < x_e \). Now Eqn. G.14 and G.15 can be simplified to:

\[
G_3(s) = \frac{u_2 g}{\bar{u}_1 s} \left[ \exp \left( -\frac{\bar{u}_1}{g} x_e \right) \left( 1 - \frac{\bar{u}_1}{\bar{u}_1 + s} \exp \left( -\frac{s}{g} x_e \right) \right) - \frac{s}{\bar{u}_1 + s} \right]
\]  

(2.47)

\[
G_4(s) = \frac{g \bar{u}_2}{s + \bar{u}_1} \left[ 1 - \exp \left( -\frac{s}{g} x_e \right) \exp \left( -\frac{\bar{u}_1}{g} x_e \right) \right]
\]  

(2.48)

Choosing physically attainable values for the parameters (i.e., \( u_1 = 1, \ u_2 = 0.200 \times 10^{-3} \ l/s, \ g = 5.0 \times 10^{-8} \ \text{m/s}, \ x_e = 1000 \times 10^{-6} \ \text{\mu m} \)), the first order term, in the last term of Eqn. 2.47 and the first term of Eqn. 2.48, dominates the input output behaviour as depicted in the Nyquist curve in Fig. 2.8. If, however, the growth rate is increased to for example \( 1.5 \times 10^{-7} \ \text{m/s} \), which is fairly large, the Nyquist curve, as depicted in Fig. 2.8, attains the shape of an extended spring in the high frequency range. Clearly, two basic dynamic elements are present in the system: a first order behaviour describing a well-mixed vessel continuous reactor, and a delay, which is determined by the time
crystals need to reach a certain size $x$. The combination of these elements typically leads to the extended spring like curves in the frequency domain as the one depicted in Fig. 2.8 (cf. Section 9.3.)

2.3.4 Finite difference approximation

For lumping second and fourth order finite difference schemes are considered. In different text books [245, 217], these schemes are discussed. Application to crystallization processes is described, among others by [233, 51]. Alternative lumping methods are the moment transformation, which is introduced by [113], or methods in which the population density function is approximated as a linear combination of chosen basis functions. Details on the latter class of methods are given by [201] and the references therein.

We found that finite difference schemes are most attractive as they do not pose any restrictions on the complexity of the equation to be solved. In addition, these schemes are now available in some software packages [184, 12]. A disadvantage might be that a large number of equations is generally needed to obtain a sufficiently accurate approximation.

With the second order scheme the first derivative to the $i$th spatial variable (see
Eqn. 2.38) is approximated with:

\[
\left. \frac{df(x)}{dx} \right|_{x=x_{i}} = \frac{1}{2\Delta x} \left[ -f_{i-1} + f_{i+1} \right] + O(\Delta x^2)
\]

\[
\left. \frac{df(x)}{dx} \right|_{x=x_{k}} = \frac{1}{2\Delta x} \left[ f_{k-2} - 4f_{k-1} + 3f_{k} \right] + O(\Delta x^2)
\]

(2.49)

The fourth order scheme gives a higher order approximation:

\[
\left. \frac{df(x)}{dx} \right|_{x=x_{0}} = \frac{1}{12\Delta x} \left[ -25f_{0} + 48f_{1} - 36f_{2} + 16f_{3} - 3f_{4} \right] + O(\Delta x^4)
\]

\[
\left. \frac{df(x)}{dx} \right|_{x=x_{1}} = \frac{1}{12\Delta x} \left[ -3f_{0} - 10f_{1} + 18f_{2} - 6f_{3} + f_{4} \right] + O(\Delta x^4)
\]

\[\vdots\]

\[
\left. \frac{df(x)}{dx} \right|_{x=x_{i}} = \frac{1}{12\Delta x} \left[ f_{i-2} - 8f_{i-1} + 8f_{i+1} - f_{i+2} \right] + O(\Delta x^4)
\]

\[\vdots\]

\[
\left. \frac{df(x)}{dx} \right|_{x=x_{k-1}} = \frac{1}{12\Delta x} \left[ -f_{k-4} + 6f_{k-3} - 18f_{k-2} + 10f_{k-1} + 3f_{k} \right] + O(\Delta x^4)
\]

\[
\left. \frac{df(x)}{dx} \right|_{x=x_{k}} = \frac{1}{12\Delta x} \left[ 3f_{k-4} - 16f_{k-3} + 36f_{k-2} - 48f_{k-1} + 25f_{k} \right] + O(\Delta x^4)
\]

(2.50)

The constant coefficients are derived from standard Lagrange interpolation formula's for three and five points, respectively [1].

For the simple linear distributed parameter process described in Section 2.3.3, discretisation onto k lumping elements \(n_1, n_2, \ldots, n_k\), and application of the second order
scheme, results in the following state equation:

$$
\begin{bmatrix}
\dot{n}_1 \\
\dot{n}_2 \\
\vdots \\
\dot{n}_k \\
\end{bmatrix} =
\begin{bmatrix}
b & -a & \cdots & 0 \\
a & b & -a & \ddots \\
a & b & \ddots & \ddots \\
0 & \cdots & a & b & -a
\end{bmatrix}
\begin{bmatrix}
n_1 \\
n_2 \\
\vdots \\
n_k \\
\end{bmatrix} +
\begin{bmatrix}
1 & a \\
0 & 1 & 0 \\
\vdots & \vdots & \vdots \\
0 & \cdots & 1 & 0
\end{bmatrix}
\begin{bmatrix}
u_1 \\
u_2 \\
\vdots \\
u_k \\
\end{bmatrix}
$$

(2.51)

with $a = g/2\Delta x$, and $b = V/\bar{u}_2$. The state matrix is a simple tridiagonal matrix. The eigenvalues $\lambda_i, \quad i = 1, \ldots, k$ of this matrix are given by [55, page 179]:

$$
\lambda_i = b - 2a \cos \frac{i\pi}{k+1} = \frac{V}{\bar{u}_2} - \frac{g}{\Delta x} \cos \frac{i\pi}{k+1}, \quad i = 1, \ldots, k,
$$

(2.52)

where $j$ denotes the imaginary unit value. The eigenvalues are plotted in Fig. 2.9. Note that the real part is determined by the mean residence time (as defined with Eqn. 2.1) and the imaginary part is determined by both the growth rate and the grid spacings $\Delta x$.

Another point to note is that the second order and fourth order schemes have large imaginary valued eigenvalues, that points at badly dampened modes. This artefact is due to the finite difference scheme, rather than an inherent property of the process. For instance a lower accurate solution obtained with the first-order scheme [245, 49]:

$$
\frac{df(x)}{dx} \bigg|_{x = x_i} = \frac{1}{\Delta x} \left[ -f_i + f_{i+1} \right] + O(\Delta x^1)
$$

(2.53)

would give only real valued eigenvalues at $\lambda_i = V/\bar{u}_2, \quad i = 1, \ldots, k$. Thus, provided that the distribution is sufficient smooth, the high order scheme will not cause problems. On the other hand, if large values for $df/dx$ occur the badly dampened modes are excited and cause high frequent oscillations in the solution. From simulations it is found that the latter problem does not occur.

Finally, note that Eqn. 2.51 employs a first order approximation of the derivative at the right boundary, with accuracy $O(\Delta x^1)$. This lower accuracy affects the whole solution. To avoid this De Wolf [49] proposed to use a non-central second order accurate scheme at the boundary, which uses three points.

$$
\frac{df(x)}{dx} \bigg|_{x = x_k} = \frac{1}{2\Delta x} \left[ f_{k-2} - 4f_{k-1} + 3f_k \right] + O(\Delta x^2)
$$

(2.54)

In Fig. 2.9 the eigenvalues of the corresponding state matrix are given. Note that, significant faster dynamics are introduced, which will require smaller simulation time
Fig. 2.9: Location of eigenvalues of state matrix using three different finite difference schemes.

steps and thereby a larger simulation time. In the same figure also the eigenvalues of the state matrix are given, using the fourth order scheme, with a fifth order boundary condition and the same discretisation grid. The fourth order scheme will provide a more accurate solution, however, at the expense of a large simulation time.

Errors involved in Lumping

Using the analytic solutions, described in Section 2.3.3, the accuracy of the lumping schemes is studied. For control, frequencies up to 1/100 Hz are considered to be important. In Fig. 2.10 the Bode plot of the four analytical transfer functions summarized with Eqn. 2.42, are given in the frequency range of $10^{-4}$ to $10^{-2}$ Hz. For the model parameters the values $u_1 = 0.20 \times 10^{-3}$ l/s, $u_2 = 1$, $g = 15 \times 10^{-8}$ m/s, $x_e = 1005 \times 10^{-6}$ µm) are chosen. In the same plot the transfer functions of the lumped models, using second and fourth order lumping schemes, are given. The grid spacing was chosen 15 µm. As shown both schemes give a small error ($\pm 10\%$) in describing the phase shift of $G_4$ at frequencies larger than 1/500 Hz. The fourth order scheme accurately approximates the other transfer functions over the whole frequency range. The second order scheme only has a significant deviation for frequencies larger than 1/500 Hz in $G_2$. From this it is concluded that the largest errors in the finite difference approximation occur in describing the growth or convection term.
Further studies on finite difference approximations to hyperbolic partial differential equations, conducted by Vichnevetsky [249, 248], show that the significant errors in describing convection at higher frequencies are due to a numerical dispersion effect. This effect causes high frequent "down-wind" oscillations when sharp gradients are present in the solution. Simulations conducted by De Wolf [49, p. 81] further illustrate this effect. From the simple simulation example given here we may conclude that this phenomenon only plays at frequencies outside the control relevant range.

This comparison shows the good ability of the finite difference scheme to describe the process dynamics. The results described in [49, p. 75] show that the finite difference approximation using second order and fourth schemes causes errors smaller than 5% in the steady-state solution, when the grid-spacing is lower than 25 μm. Based on this most simulations described here have been performed with a second order scheme using a grid spacing of 15 μm.

2.3.5 Integration

The program modules that were used in the simulator are described in Appendix G. The module INTEG (cf. Fig. F.1) is adopted from the DSS/2 package [184] without modifications. It calls 15 explicit Runge-Kutta integrators for initial-value ordinary differential equations. All integrators include error estimation formulas on the basis of which the integration step size is adjusted [32]. Generally the accuracy of the error estimation formulas, used to adjust the integration step size, increases down the list (from 1 to 15) at the expense of additional computational effort.

The integrators are all explicit and therefore have limited stability intervals. Consequently, they should not be applied to so called stiff systems [189], that is, sets of differential equations containing a mixture of very fast and very slow dynamics.

INTEG integrates an nth-order system of initial-value ordinary differential equations of the form:

\[
\begin{align*}
\frac{dz_1}{dt} &= f_1(z_1, z_2, \ldots, z_n, t) \\
\frac{dz_2}{dt} &= f_2(z_1, z_2, \ldots, z_n, t) \\
&\quad \vdots \\
\frac{dz_n}{dt} &= f_n(z_1, z_2, \ldots, z_n, t)
\end{align*}
\]

subject to the initial conditions

\[
\left[ z_1(t_0), z_2(t_0), \ldots, z_n(t_0) \right]^T
\]  

(2.55)
Fig. 2.10: Bode plots for analytic (solid) and lumped models using a second order (dashed) and fourth order (dotted) schemes with a grid spacing of 15 μm.
The program flow of the first Runge-Kutta algorithm is as follows: set initial integration interval to one-eighth of the print interval\(^1\): \( h = t_p/8 \). Evaluate the derivatives in a second step at the base point \( t_0, z_0 \), using (Fig. F.1):

\[
\frac{dz}{dt} = f(z_0, t_0)
\]

(2.56)

and implement the forward Euler algorithm:

\[
z = z_0 + f_0 \times h.
\]

(2.57)

Estimate the truncation error for each dependent variable, using

\[
e = \left( f(t_0) - f(t_0 + h) \right) \frac{h}{2}.
\]

(2.58)

Now test if the integration interval is at the allowable minimum. If not, test if the estimated truncation error exceeds the allowable error. If the chosen error criterion bound has been violated, then reduce the integration interval and repeat the integration. If the error criterion bound has not been violated, then apply the truncation error correction: \( z = z + e \), repeat the procedure until \( t = t_0 + t_p \).

**Errors involved in integration**

The errors involved in both integration and lumping are studied using both the second order and fourth order lumped versions of the simple model described in Section 2.3.3. The accuracy is tested in the frequency domain by applying input sinusoid to the input \( u_2 \) and recording the corresponding output signal \( y_2 \). The phase and magnitude calculated from these signals are compared to the Bode plots described in Fig. 2.10. From this it is found that the solution in time, calculated with a simple Euler scheme with truncation error correction does not introduce significant additional errors in the solution, provided that the minimum integration step size is not too large. In addition, this scheme is found to be most fast as compared to higher order routines. The integration step size can be chosen a priori on the basis of the eigenvalues of the state matrix depicted in Fig. 2.9. Notice that increasing the order of the finite difference scheme requires a smaller step size. In addition, Eqn. 2.52 shows that the integration step size should be changed proportional to the grid-spacing.

2.3.6 Stationary solution of model

This section shows how a steady-state solution of the model presented in the previous section can be derived. This steady-state solution will be used to derive a linearized

\(^1\)The print interval is the interval at which output is generated. Usually it is chosen equal to the process sample time.
model, which describes the process dynamics in a neighbourhood of this solution. Linear models are used for analysis and design of model-based controllers. Besides, the steady-state model solution as a function of process operation conditions is important for design related purposes, for example, to assess the best stationary operation condition (see e.g., Chapter 8).

The stationary solution of the population balance model Eqn. G.1 should obey:

\[ \bar{n}(x) = \frac{\bar{B}}{G_e(x)} \exp \left( - \int_{z=x_0}^{x} \frac{1}{G_e(z)} \frac{\dot{Q}_h \dot{h}_p(z) + \dot{Q}_f \dot{h}_f(z)}{V} \, dz \right) . \] (2.59)

This equation is an implicit equation as the stationary values for \( G_e \) and \( B \) depend on the solution: \( \bar{n}(x) \).

This solution is therefore approximated with a numerical scheme as follows:

1. Choose initial value for \( G_e \) and \( B \).

2. Calculate the right hand side of Eqn. 2.59.

3. Use the resulting solution to calculate new values for \( G_e \) and \( B \).

4. Calculate the error distance between the new and old values of \( G_e \) and \( B \) with the following quadratic criterion:

\[ J_{err} = \left[ \frac{G_e^{i+1}}{G_e^{i}} - 1 \right] \left[ \frac{B^{i+1}}{B^i} - 1 \right] . \] (2.60)

5. With a standard Newton-Raphson technique the optimum is estimated.

6. If \( J_{err} \) is larger than a certain value repeat this procedure otherwise a stationary solution is found.

A disadvantage of this approach is, however, that the final solution of the theoretical equation does not exactly match the stationary solution of the lumped model as the finite difference scheme introduces an error. These errors, can cause significant upsets in the simulation.

Therefore, the aforementioned procedure is applied to the following lumped sta-
tionary population balance model. Using the second order finite difference scheme.

\[
\begin{bmatrix}
Q_1^- & -G_2^- & \cdots & 0 \\
G_1^- & Q_2^- & -G_3^- \\
G_2^- & Q_3^- & -G_4^- \\
\vdots & \ddots & \ddots & \ddots & \vdots \\
G_{n-3}^- & Q_{n-2}^- & -G_{n-1}^- \\
G_{n-2}^- & Q_{n-1}^- & -G_n^- \\
0 & \cdots & G_{n-2}^- & -4G_{n-1}^- & 3G_n^- + Q_n^- \\
\end{bmatrix}
\begin{bmatrix}
n_s(1) \\
n_s(2) \\
\vdots \\
n_s(n) \\
\end{bmatrix}
= 
\begin{bmatrix}
b \\
0 \\
\vdots \\
0 \\
\end{bmatrix}
\]

or

\[A \ast n_s = b \] (2.62)

With \(G_i^-\) and \(Q_i^-\) as a short notation for the terms:

\[
G_i^- = \frac{G_e(x_i, t)}{2\Delta x}
\]

\[
Q_i^- = \frac{Q_p(t)h_p(x_i, t) + Q_f(t)h_f(x_i, t)}{V}
\]

A solution to Eqn. 2.62 is obtained by inverting matrix \(A\) in Eqn. 2.62. This matrix is a sparse matrix. Using a second order finite difference scheme only a single upper and lower diagonal are non-zero, and the right band side vector only has its first element as non zero. For this case, efficient routines (NAG [150], F01KBF and F01FBN) could be used for inversion. The code was implemented in Fortran. On a VAX3100 M76 workstation, the time to find a solution for a 100th order model was found to be lower than 1 second.

### 2.3.7 Linearisation

A linearized model is derived from the nonlinear model. The linearized model provides an approximation of the nonlinear model in a neighbourhood of the steady-state process behaviour. Linear models are needed primarily for design and analyses of model-based controllers in the frequency domain (cf. Chapter 10) and the design of state estimators (cf. Chapter 7).

Here a linearized model is derived analytically from the nonlinear model equations. The main motivation for this is that the state vector of the analytically linearized model still has its physical meaning. This is important for the design of a state estimators. Further the computational requirements are low and in-stable models can
be linearized as well. Hence, calculation of the pole locations as a function of certain process parameters may be used to study the stability of the process. A disadvantage might be that the approach is elaborate, and sensitive to programming errors. In addition, the order of the lumped linearized model is high, which requires model reduction as will be discussed below.

For linearization of a nonlinear process model for the purpose of controller design, other approaches may be applied as well. De Wolf [49] presented a system identification procedure to arrive at low order linear models on the basis of simulated input-output data. This approach has given good results. It has the added advantage that it may be applied directly onto experimental process data as we will do in Chapter 6. A disadvantage might be that it is not directly applicable to unstable models and wrong answers may be obtained when the excitation signals are not chosen properly. Another approach to model linearization are numerical perturbation approaches, as exploited for instance in the CDS module of Speed-Up [7]). For high model orders these numerical methods are often ill conditioned, and the results will depend on the type of perturbations or excitation signals.

Linearization of the nonlinear distributed parameter model given by Eqn. 2.38 gives:

$$\frac{\partial \Delta z}{\partial t} + f_1(x) \frac{\partial \Delta z}{\partial x} + f_2(x) \Delta z + f_3(x) \Delta u = 0$$

(2.63)

and the boundary condition:

$$\Delta n(x_0, t) = f_4(x) \Delta z + f_5 \Delta u$$

(2.64)

The process state is given by: $z = \{n, \Delta C\}$, the process inputs are $u = \{Q_f, Q_p, P_t\}$. The functions $\{f_1(x), \ldots, f_5(x)\}$ are continuous size dependent time invariant functions. $f_3$ and $f_5$ are vector functions, which contain a number of elements equal to the number of input variables.

For two cases this equation is linearized analytically:

- Class I model
- Class II model

The equations are derived in Appendix A.

After lumping to a finite set of elements the linearized equations can be cast into a finite dimensional, linear, time invariant, continuous, state-space model, which is denoted as:

$$\dot{x} = Ax + Bu$$

(2.65)

$$y = Cx + Du$$

(2.66)

The system matrix $A$ has a sparse structure (see Appendix A), in which only a limited number of non-zero elements are present. Using structural information, both the disk capacity for file storage and the simulation speed could be enhanced drastically.
Fig. 2.11: Comparison of linear (dashed) and positive and inverse negative (solid) nonlinear responses of the discrete population density at: $x = 30, 150$ and $1005 \mu m$, on a step in the product removal rate, using a second order lumping scheme.

To enhance the numerical condition of this model an equivalence transformation \{A, B, C, D\} $\rightarrow$ \{\(T^{-1}AT, T^{-1}B, CT, D\}\) is performed, where the state vector of the model is normalized by subdividing through the steady-state solution: $T = \text{diag}(z_0^{-1})$.

In Appendix G linear relationships of some important nonlinear sensor characteristics or nonlinearities involved in the computation of output quantities, are given.

Results model linearization

The linear and nonlinear step responses are compared to judge the nonlinearity of the system. For this purpose the linear and the nonlinear model were simulated with identical integration schemes. It was found that nonlinearities are most strong when step changes to the product removal flow are made. Using the Class I model, with parameter values derived in Chapter 5, the linear and nonlinear models are compared. 10% step changes are applied, and as output the population density at a crystal size of $x = 30, 150$ and $1005 \mu m$ is taken. In Fig. 2.11, the response to a positive and
inverse negative nonlinear step responses are shown with the linear model response. The results reveal that the nonlinearities are mild. Thus linearized models provide a good description to the process dynamics, whenever the process is operated close to its stationary behaviour.

2.3.8 Model reduction

The derived analytically linearized models have an order that is to high for control design purposes. As an alternative to system identification a balanced model reduction is applied [158].

The key solution to balanced model reduction is to find a balancing transformation [158]. Assuming a linear time invariant state-space model \( \{A, B, C\} \), a transformation \( \{A, B, C\} \rightarrow \{T^{-1}AT, T^{-1}B, CT\} \) is a balancing transformation, if:

\[
T^{-1}PT^{-T} = T^TQT = \Sigma, \tag{2.67}
\]

where \( \Sigma \) is a diagonal matrix, containing the so-called Hankel singular values in decreasing order, and \( P \) and \( Q \) represent the controllability Gramian and observability Gramian of the system \( \{A, B, C\} \), given by:

\[
P = \int_0^\infty \exp(At)BB^T\exp(A^Tt)\, dt \tag{2.68}
\]

\[
Q = \int_0^\infty \exp(A^Tt)C^TC\exp(A^Tt)\, dt \tag{2.69}
\]

A reduced order model \( \{A_R, B_R, C_R\} \) is devised by taking the first \( n_r \) columns of \( T \) denoted as \( R \) and the first \( n_r \) rows of \( T^{-1} \) denoted as \( L \). The reduced model with order \( n_r \) is then given by \( \{A_R, B_R, C_R\} = \{LAR, LB, CR\} \).

Several methods can be employed to solve this problem. Here, a frequency weighted balanced reduction method is used, which employs a singular value decomposition of the observability and controllability Gramians. With this method the fit of the reduced model can be favoured in the frequency range, which is relevant for control. A Matlab toolbox developed by Wortelboer is used [254].

**Results model reduction**

It is found that 4th and 5th order models give good approximations of the process input-output behaviour. In Fig. 2.12 a comparison is made between the magnitude plots in the frequency domain of a reduced 5th order and the full 100th order three input, three output linear model. The inputs and outputs of this model are described in Section 6.2 (see Table 6.1). The parameter values used in the first principles model are described in Chapter 5. The computation time was 20 seconds on a VAX3100 M76 Workstation.

\[ \text{This toolbox can be obtained by anonymous ftp [254]} \]
Fig. 2.12: Comparison of magnitude plots in the frequency domain for linear 100th order (solid), and reduced 5th order model (dashed) with $i \rightarrow j$ denotes the transfer relationship of the $i$th input to the $j$th output.

2.4 Conclusions

In this chapter a macroscopic population balance model is described. The aim has been to present a basic set of model equations that form a good basis for modelling of industrial crystallizers.

The model includes empirical relations for the effective crystal growth and the effective birth rate of crystals. Empirical relations for the removal efficiency of fines and product classification systems are given. In addition, an explicit model for the sensor is formulated, mainly to enable proper validation of the model on the basis of measured process data. The latter also enables to account in a correct way for the effect of particle shape on simulated sensor data. It is shown that the assumption on the well-mixedness introduces an error. This is mainly due to settling of particles and the presence of supersaturation profiles.
2.4 Conclusions

For simulation and analysis purposes numerical solution schemes are outlined. Using an analytical solution to a simple population balance it is shown that lumping a population balance using a finite difference scheme gives good approximations to the CSD dynamics. The largest errors occur in simulating convection due to growth. For simulation, a second order accurate scheme is chosen with a maximum grid spacing of 15 \( \mu \text{m} \).

Reduction of the lumped model can be achieved using linearized models with a frequency weighted balanced reduction method. It is found that the response of a linear model is close to the nonlinear model when the system is operated close to its stationary behaviour. As the latter condition is fulfilled with an effective controller, the design of this controller on the basis of a linear model is expected to give good results.
Chapter 3

Experimental equipment, actuators and sensors

This chapter describes the experimental equipment that is used for experimental evaluation of the dynamic model introduced in the previous chapter. A description will be given of the 970 litre pilot crystallizer, including some important parts of the peripheral equipment and the process monitoring and control system. Finally, the sensors used for on-line CSD measurement are described.

3.1 The pilot crystallizer

3.1.1 General lay out

The pilot crystallizer is an evaporative draft-tube baffled crystallizer, which can be operated in a batch as well as a continuous mode of operation. A schematic drawing of the crystallizer is provided in Fig. 3.1. The total height of the system is 10 meters. The volume of the main crystallizer volume is estimated to be 970 litres. All piping and vessels are made of stainless steel (316). All tubes and vessels are insulated on the outside to prevent cold spots that may serve as a growth surface for crystal material and thereby cause incrustation [246, 244]. DSM technically pure ammonium sulphate using municipal water as a solvent is used as the model material.

Feed is supplied to the crystallizer from the feed preparation tank, by an air driven membrane pump (DEPA). The feed preparation tank is fed from an overflow line connected to the top section of a settler, which is located in the recycle vessel. This vessel recycles product slurry, clear mother liquor used for CSD measurement and condensed water vapour, its temperature is kept constant at 50 °C. The temperature in the feed preparation tank is raised up a further 5 °C to ensure a crystal-free feed. Insoluble dust and other impurities are removed by filters. Two condensers are used to extract vapour. An air driven membrane pump is used to recycle condensed water vapour to the recycle vessel. This water recycle closes the material balance of the
3.1 The pilot crystallizer

**Fig. 3.1:** Flow sheet of pilot crystallizer and environment.

Overall system, which is important to prevent accumulation of impurities which are present in the solvent (i.e., municipal water).

Vacuum in the system is maintained at approximately 80 mbar. Inert gases are removed with a liquid ring pump (SIHI), which is placed after the valves in the vapour line. Heat is supplied at two locations: with an internal tube heat exchanger located in the draft-tube and with an external plate type heat exchanger located in the fines loop. Heating is done using hot water with a temperature of approximately 85 °C. The slurry in the crystallizer is pumped along the draft-tube with a 485 mm marine type impeller rotating at an average speed of 315 rpm. The impeller is located right below the internal heat exchanger. The average slurry circulation time is approximately 15 seconds [120].

A rinse water system is present, with inputs at different locations in the process. Rinse water is used to remove scaling, which causes blockage of the tubes and also to clean the system after experiments. The rinse water flow is measured and its value is stored in the database to use it as known disturbance input to the system if necessary. During normal process operation, rinse water to remove scaling had to be injected approximately once every 6 hours.

All slurry transportation lines are kept as short as possible and sharp bends which
may cause additional wear of crystals are avoided. Slurry transportation is achieved with Mohno™ pumps. These pumps give low abrasion to the crystals, and have a reasonable constant yield. A seal water system is present which serves a cooling and cleaning flow to the double mechanical seals implemented in the Mohno pumps. With these seals inert leakage into the system is prevented effectively.

3.1.2 Fines removal

A fines removal and dissolution system is installed to enhance the steady-state product CSD and to serve as an actuator for CSD control. Fines are removed selectively from the crystallizer volume using an annular zone around the vessel. The outer diameter of the annulus is 1.2 m and the inner diameter is 0.7 m, the cross sectional area is 0.746 m² and the total volume is roughly 600 litres. Fines are removed at 6 different locations at equidistant radial positions at the top of the annular zone. In Fig. 3.10 a
schematic drawing of the crystallizer vessel and the fines removal system is given. The removed fines stream is collected in a small collection vessel (10 litres) and pumped through a plate type heat exchanger. Here the fines slurry flow is raised by 10 °C to dissolve the crystals. The fines removal rate can be varied in the range of 0.4 to 3.5 l/s. To ensure total dissolution of fines a mixed vessel with a content of 50 litres is placed after the heat exchanger before the dissolved fines are returned to the crystallizer.

3.1.3 Product removal

Two lines for product withdrawal are present in the crystallizer: one for unclassified product removal and one for classified product removal. Classified product removal is generally applied to obtain a coarser product. Both withdrawal locations are in the bottom section of the crystallizer. When product classification is applied the latter pump serves as feed pump for the CSD measurements only, while all discharged slurry except a small measurement flow is returned to the crystallizer. Entrance classification at the inlet of these lines in the crystallizer is minimized by choosing a tube diameter in the crystallizer that gives a liquid velocity in the tube that is approximately equal to the velocity of the slurry at this location in the crystallizer. Because the removal flows are varied, for different experimental reasons, this condition is not fulfilled in all experiments. The unclassified removal flow rate can be varied in the range of 100 to 400 ml/s. The feed for the product classification unit can be operated in the range of 0.5 to 2.0 l/s. In the product discharge loop the slurry density is measured with an E&H massflow/density sensor (M-point, DQ 600).

Because product classification is performed at atmospheric conditions, the recycle flow of small particles is driven by the pressure difference between the crystallizer and the atmospheric pressure. Hereby additional attrition or wear of crystals could be minimized. The recycled slurry is collected in a small vessel with a volume of 20 litres, before returned to the crystallizer. This vessel enables CSD sampling and has sufficient residence time to allow air bubbles to separate from the slurry flow.

3.1.4 Principles of classified product removal

For product classification three different types of classifiers are studied:

- gravitational settlers
- mechanical separators
- centrifugal separators

For all three principles, separation units are implemented on the UNIAK crystallizer. A more thorough description and a discussion on these devices is given by Gerla [86].
Fig. 3.3: Schematic drawing of vibrating screen.

Fig. 3.4: The Algaier vibrating screen classifier.
3.1 The pilot crystallizer

Gravitational settling

Gravitational settling is performed with a Stokes hydrosizer [232]. In this system a bed of large crystals is formed that is washed from the bottom section with clear mother liquor, for which we used a part of the dissolved fines flow. Unclassified slurry is injected in the top section. The overflow, containing small crystals and wash liquid are returned to the crystallizer, while the product crystals form a fluidized bed. The height of this bed is controlled using a PI controller that acts on a valve in the bottom section of the settler, on the basis of a measured pressure inferred from a silometer (E&H Silometer FMC 425). The main disadvantages of this system are the bad observability of the conditions at which separation takes place, the large settling volume needed to achieve good separation, and the difficulty to control the parameters that influence the separation efficiency, for example, the bed height.

The success of gravitational settling with this unit or others (e.g. elutriation legs [17]) will strongly depend on the scale, the material properties like the viscosity of the mother liquor, the density of the solid and liquid phase, and the availability of wash flows.

Mechanical separator

Mechanical separation is achieved with a vibrating screen classifier. A schematic drawing is provided in Fig. 3.4 and Fig. 3.3 depicts the Algaier vibrating screen classifier, which is installed onto the UNIAK pilot crystallizer.

Vibration of the screen deck enhances the separation efficiency and enables transportation of slurry over the screen deck. The product leaving the screen is a viscous cake which does not disperse. The void fraction is rather constant at 55%, which is close to the void fraction of an ideal packed bed [214].

The main design parameters considered in this work related to this separator are the aperture size of the screen and the feed flow from the crystallizer to the screen. Deviations from ideal separation are mainly caused by small crystals that adhere with liquid to the surface of large crystals. Improved separation may be expected when a clear mother liquor flow flow (e.g., dissolved fines) is used, to wash these small particles from the product crystal surface.

Strong advantage of a screen are its small size, the mild process conditions at which separation takes place, the sharp and large separation efficiency curve that can be achieved, and the low slurry transportation costs. Besides, in contrast to the other separators discussed here, good separation can be achieved when the difference between the solid and liquid phase density is small. However, mechanical excitation of the crystal surface by the vibrating screen deck can be a point of concern as it can break or erode the crystals, causing an increase of small crystals recycled to the crystallizer. Further the screen apertures may get obstructed by wedged particles that have a size slightly larger than the screen apertures. Despite the presence of a large
amount of elastic bouncing balls that hit the screen frequently at the bottom side, this problem occurred during the experiments. The blockage started as a circular spot right at the location where slurry is dumped on the screen and the radius of the spot grew slowly. After approximately 12 hours the non-blocked surface becomes so small to assure proper separation, and rinsing with hot water was necessary. However, already a small amount of water was sufficient to free the aperture for hours again. For further details we refer to [86].

**Centrifugal separators**

Centrifugal separation is achieved with hydrocyclones. The use of hydrocyclones for control of crystal size distribution is reported by [86, 119, 187, 219]. A major disadvantage of the application of hydrocyclones are the large hydrodynamic forces imposed on the crystals to be separated, which can cause breakage or erosion. The efficiency of separation is also strongly limited by material properties such as viscosity of the mother liquor and the density difference between crystals and the mother liquor. For this reason only a limited number of substances can be separated with a hydrocyclone. Hydrocyclones are not considered further in this work.

**3.1.5 Local PI control loops**

PI control loops are present in the process to keep several process conditions at a prescribed set-point value: the slurry temperature is controlled at 50°C with an accuracy of 0.1°C with a master-slave controller in which the master temperature controller acts on the slave pressure controller. The pressure is controlled using butterfly valves (Keystone™) located in the vapour line. Constant control of volume in the crystallizer is based on a level measurement. The level is calculated from the difference between the pressure in the vapour zone and the pressure at approximately 60 cm below the boiling zone. The latter measurement is equipped with a small water purge of 1 ml/s to prevent blockage of the measurement probe. The level controller acts on the feed flow to the crystallizer to control the level and thereby the volume, with an accuracy of approximately ±10 litres during stationary operation. The level measurement is biased when density changes occur, however, the density fluctuates at a maximum between 1250 and 1350 [kg/m^3], which corresponds to insignificant level differences.

The fines, product and mother liquor flow rates are controlled on the basis of measured flow rates obtained from E&H magnetic inductive flow indicators (Picomag™ DMI 6530). Control of the pumps is achieved with (Hitachi™ VWS) frequency controllers at a rate of 0.2 Hz. The speed of response of the controlled pumps is approximately 30 seconds.

The temperature of the dissolved fines is controlled by manipulating a hot water flow rate with an air driven valve (Kammer™) located in the hot water loop for the external fines heat exchanger. The total heat input is controlled with an air driven
3.2 Process control system

General lay out

The complete measurement and control system is described in Fig. 3.5. The majority of process control and monitoring tasks related to the basic process variables (e.g., temperature, pressure and flow rates), is performed with a commercial software package called FIX DMACS™ [115] running on a 486-PC. The FIX-DMACS package is coupled with the I3852 process hardware interface with a special purpose software interface. In the I3852 interface analogue input, analogue output, pulse counting, digital input and digital output tasks are performed. Digital IO is directly coupled to PLC blocks that handle all logic control actions on valves and pumps. Analogue input and output signals are all based on the IEEE 4-20 mA standard.

The PLC also triggers the PC's on which on-line software for the CSD sensors runs. The raw CSD data are processed locally on these PC's and are successively transferred to FIX DMACS which serves as a host. For transfer of these signals a special purpose C-driver is developed, which receives CSD sensor data via a RS232 serial port connection and directly stores this data in the FIX DMACS database, or sends it to the VAX workstations. Details can be found in [44].

The FIX DMACS process monitoring and control system is coupled with a network of four VAX 3100 m76 workstations, using an Ethernet connection. The VAX workstations are used primarily for simulation and controller design purposes and for analysis of experimental data. For the model based control experiments, reported in Chapter 10, a VAX station has been used on-line to calculate the optimal process inputs on the basis of the measured CSD and the slurry density information. Exchange of process input-output data is performed with ASCII structured data files, for which Fortran communication software has been developed.

Process monitoring and control

With FIX-DMACS all PI-control and logical control tasks are performed. Process information in the form of reports, displays, archive data, and alarm messages are displayed. A graphical display is designed which comprises a main screen displaying all important process parameters and sub-screens which display information on one specific control loop.
A necessity for successful experimenting is the presence of sensitive alarms to alert the operator when process faults occur. Several visual alarms have been implemented in the graphic interface. As a criterion for fault detection mostly a maximum and minimum value for the process values are used. For detection of faults occurring in the slurry transportation lines, the following criterion is used:

\[
y_{\text{sensor}}/u_{\text{actuator}} < x_{\text{lim}},
\]

where \(u_{\text{actuator}}\) is the output of the PI controller and \(y_{\text{sensor}}\) is the measured slurry flow. Choosing the limit \(x_{\text{lim}}\) sufficiently tight, the onset of tube blockage could be detected and thereby the occurrence of major process faults could be prevented. Both a visual and an acoustic alarm have been used to alert the operator for these faults.

For monitoring of the signal from the on-line particle counter, that is introduced below, the signal trend is displayed together with upper and lower warning and alarm limits. The latter values are adapted on the basis of the signal variance. Use is made of a statistical process control module (SPC) present in the FIX DMACS process monitoring and control system [115].
3.3 CSD sensors

CSD sensors are generally classified into off-line, on-line and in-line techniques. Off-line techniques, like sieve analysis, are the most elaborate and often introduce a large time delay between the instance of sampling and the time at which the measurement results are available. Because these characteristics are highly undesired for control, we do not discuss off-line techniques further. On-line techniques still involve a sampling step, however, the time between sampling and actual measurement is substantially reduced. Finally, with in-line techniques the measurement is performed in the system itself. For this case sampling is not needed and actual process values are directly available.

3.3.1 Overview of on and in-line sensing techniques

Robust on- and in-line CSD sensing is a difficult problem. Currently more than 20 suppliers exist world-wide who offer different systems for on-line measurement of solid or liquid particle size distributions in gasses or liquids. These systems may be broadly subdivided into the following basic physical principles:

1. acoustic, for example, acoustic spectroscopy [210],

2. optical, for example, forward and backward light scattering and imaging [24, 139, 172],

3. electrical, for example, the Coulter Counter™ [3].

The commercially available sensors strongly differ in their ability to deal with different process and environmental conditions and in particular with respect to their maintenance requirements. Up to date reviews of on-line and in-line CSD sensing techniques are not available. For an outline of several basic issues related to sampling and sensors the reader is referred to [3, 105, 216]. In Hobbel et al., [107] a comparison between forward scattering and back scattering is made. In addition to size measurement, also shape measurement becomes increasingly important. A new method for on-line shape sensing has recently been proposed by a hef [101].

Within the UNIAK project different CSD sensors have been tested. Some test results with the OPUS sensor [229], which is based on ultrasonic spectrometry, are available [21]. Results on the use of on-line imaging in the UNIAK crystallizer are reported by [100, 175]. A single particle optical sensor provided by [172] has been tested. With this sensor a high resolution could be obtained. The major drawbacks, however, are that a small aperture in the measurement chamber causes frequent blockage and that dilution of the slurry prior to measurement is a necessity.

Most sensing techniques have the major drawback that they are indirect, that is, models for the sensor are needed to infer the CSD properties from the sensor readings. Often a so called deconvolution step is applied, where the sensor model is inverted to
estimate the size distribution directly on the basis of raw sensor data ([24, 37, 36]). More sophisticated schemes use a dynamic model, which is implemented as an observer or state estimator, to estimate the distribution properties from sensor data. These are discussed further in Chapter 7.

For the derivation of sensor models, assumptions have to be made on particle shape, particle orientation, and other physical properties such as the refractive index (when using forward light scattering). Modelling errors may cause significant errors in the estimated CSD properties. For instance in Chapter 4 it will be shown that over- or under-estimating the aspect ratio of rectangular-shaped particles may cause significant errors in the estimated size distribution.

These modelling errors are circumvented with direct image analysis techniques. Using a charged coupled device (CCD) camera, images are recorded on video. After digitising, image analysis techniques are applied to quantify the size distribution and the shape of the particles directly. However, a disadvantage of this direct technique is that a considerable number of samples and thereby computation time is needed. In [25, 100] the use of forward light scattering, in combination with image analysis, is therefore proposed. Analysis of recorded images then provides information on for instance the particle shape, which is used in a deconvolution step to accurately estimate the CSD from the recorded scatter pattern.

In this work, extensive use is made of both a Malvern™ particle sizer and a ready-made optical single particle counter. In addition, an on-line CCD camera is used merely to monitor the particle shape and the condition of the mother liquor. Also some experiments are conducted using a Lasentec™ in-process particle characterization monitor. In the following these four devices are further discussed. First the slurry dilution unit will be described.

### 3.3.2 On-line slurry diluter

A large problem in using forward light scattering, is the possible occurrence of multiple scattering [106, 242] when the solids concentration in the measurement chamber is too high. To account for this phenomenon accurate models are needed which are, however, not available. Therefore an on-line diluter has been designed, which lowers the particle concentration on-line to less than 2.5%.

With this unit continuous dilution of slurry is performed. A schematic drawing is provided in Fig. 3.6 and a picture of the experimental set-up is provided in Fig. 3.7. The clear mother liquor is transported continuously with a separate transportation line (see Fig. 3.1), from the recycle vessel through the optical cell of the CSD measurement unit and returned to the recycle vessel. The flow is kept constant at approximately 400 ml/s. Before entering the CSD measurement section the flow is delayed in a vessel to free entrapped air bubbles (Fig. 3.1). A slurry density indicator, equal to the one used in the product removal loop, is implemented to check the saturation conditions of the mother liquor. Slurry is injected from the product discharge line into
3.3 CSD SENSORS

Fig. 3.6: Schematic drawing of continuous slurry diluter.

the saturated crystal-free mother liquor flow, with a Jabsco™ impeller pump. This pump has a variable speed to control the dilution ratio. The actual dilution ratio is calculated from the dilution liquor flow rate and the injection flow rate, both measured with magnetic inductive flow indicators. All slurries and mother liquor in the CSD measurement unit are transported using PFA™ tubing. This material is chosen for its smooth surface layer, which lowers scaling of salt and for the wide range of pressures and temperatures it can handle.

The total time needed for one measurement is 1 minute. Each CSD measurement starts with just saturated clear mother liquor in the loop to record the background pattern from the Malvern detector. After 20 seconds the slurry injection pump is switched on, and after 3 seconds the diffraction patterns is recorded during approximately 10 seconds. After this the injection pump is switched off. The remaining time before the next measurement is used for data handling and transfer of data to the host computer.

Due to its simplicity, the continuous diluter proved to be very robust, causing negligible problems. With the frequent background measurement the quality of the data improves strongly, while monitoring of the recorded backgrounds provides the operator directly with information on the quality of the dilution flow (e.g., presence of air bubbles or dust particles) and the alignment of the sensor. More advanced batch-wise and semi-batchwise slurry diluters, which can be useful for special cases, are discussed by [86, 118].

3.3.3 Forward light scattering

A Malvern 2600e particle sizer is used, which has 31 concentrical rings to record the diffraction patterns. A lens with a focal distance of 1000 mm is used to record the light scattered from the product crystal size distributions, and a 300 mm lens for the fines
Fig. 3.7: Picture of continuous slurry diluter, comprising a Jabsco impeller pump and a flow indicator; in the background the Malvern particle sizer and the optical cell.
distributions. Fig. 3.8 explains the working principle of Forward light scattering (see [24, 148] for further details). The measurement locations are indicated in Fig. 3.1. During each measurement a batch of 1000 sweeps (detector readings) is obtained with a read-out delay of approximately 10 ms. The average and standard deviations for this batch are calculated and stored locally and transferred to the host computer. The background measurement, which is averaged from 1000 sweeps, is subtracted. Data acquisition from the Malvern detector, and several monitoring and data processing tasks are performed with a PC software package called ROMA [24].

The optical cells used for the Malvern particle sizer and the on-line camera are identical. The width of the optical window is 15 mm and the path length\(^1\) is 3 mm.

### 3.3.4 Particle counter

A particle counter in combination with a simple gravitational settler has been developed in this project, to get information on the number of particles present in a certain size region in the fines domain of the crystal population. This sensor is applied to explore what the cheapest and simplest option is for particle sensing for the purpose of control. Commercially available optical particle counters are described by [143, 180]. These, however, have the disadvantage that a distribution is measured over different channels. An one-channel measurement is in many cases sufficient for control of a CSD, in which only a limited number of degrees of freedom are present (See also Chapter 8).

In the literature different contributions describe results obtained with commercially available sensors. In [200] the use of the Microtac\(^\text{TM}\)

---

\(^1\)the distance between the windows
indexMicrotac for on-line use in the fines loop of a KCL crystallizer is described. In [71, 72] experiences with the Lasentec™ in-process particle characterization monitor for batch and continuous CSD control are described.

A schematic drawing of the particle counter is given in Fig. 3.9. In Fig. 3.10, the location of the settler and the counter system are indicated. The settler has a diameter of 60 mm. A constant flow of 15 ml/s is withdrawn, resulting in an average upward velocity of 5.3 mm/s. The optical cell has a relatively large dimension to prevent blockage. The inner distance between the optical windows is 2 mm and the diameter of the window is 10 mm. A branch pipe is added to free air bubbles present in the flow prior to measurement. The slurry is transported with a Netsch Mohno pump which is slight over-dimensioned to lower particle abrasion.

Particles passing through the optical cell induce a pulse shaped signal in the output of the photo transistor, which is transformed by a comparator circuit that transforms the raw signal to a standard 0-5 Volt TTL block pulse signal. A particle is counted when the level of obscuration of the incident beam is sufficient to arouse a voltage difference in the photo diode, that exceeds a threshold of 38 mV. The TTL block pulse signal is counted by a 5 channel HP44715A counter totalizer (max. 200 kHz), which is present in the HP3852 data acquisition system.

A strong advantage of particle counting in comparison with laser light obscuration is the low sensitivity for background noise originating from dust particles. Note that the sensitivity for CSD changes is determined by the geometry and separation flow from the separator, on the one hand, and the characteristics of the optical cell and the optical sensor, on the other hand.

A single sensor has been applied here. For robust on-line industrial application, however, operation with two or more sensors is advised with back flushing capabil-
Fig. 3.10: *Geometry of crystallizer vessel with locations of gravitational settler and product discharge.*

ities; if one aperture becomes plugged or blocked by particles, measurement can be transferred to one of the other apertures, while the first is back-flushed.

### 3.3.5 In-line CSD sensing

A new sensor called Lasentec in-process particle monitor has recently been introduced, which can be applied in-line. The basic principle of this sensor is back-scattering of laser light on the particle surface. Some experiments have been conducted with the Lasentec probe inserted in the bottom location of the 970 litre crystallier. From the results it is concluded that for ammonium sulphate the Malvern gives better results than the Lasentec monitor. This may be due to the fact that, the refractive index of ammonium sulphate particles is close to 1. Consequently, emitted laser light will mainly travel through the particle, instead of being scattered back. Therefore we do not recommend the sensor for systems with a relative refractive index which is close to 1. Experiences in applying this sensor to other materials are reported among others by [20, 71, 72, 103].
During operation, using ammonium sulphate we also found that the onset of primary nucleation from a clear liquid is detected very fast with the lasentec, merely by observing the total number of counts. In addition, note that in-line techniques—despite the important merit that sampling is not need—have the disadvantage that the conditions at which measurement occurs are not directly observable. For instance a slowly fouling probe window maybe wrongly ascribed to a real process trend by operators.

3.3.6 On-line CCD camera

For monitoring purposes an on-line digital CCD camera [90] is used. The camera is in the same line as the Malvern particle sizer and the optical cell has the same geometry as the one used for the Malvern. The recorded images are displayed on a monitor, which is placed in the operator room. In addition, the images are recorded with a video recorder and grabbed with a computer for further analysis. More details on this sensor and the image analyzer are provided by [100].

During the experiments described in this thesis the camera has been used to monitor the evolution of the CSD, to judge the condition of the mother liquor (e.g., presence of air bubbles) used for dilution, and to enable direct visual inspection of the particle shape. Shape information is used to develop a diffraction matrix as discussed in Chapter 2. Examples of typical images inferred from the pilot crystallizer are given in Fig. 3.11. These images are recorded with the camera, on-line, during unseeded startup and successive outgrowth of crystals. Similar startup response are described in Fig. 5.6. With the sequence depicted here the most important phenomena are directly visualized. At 1 minute after the onset of (primary) nucleation the system is filled with an abundance of small particles with a size lower than 50 μm. Due to the large supersaturation these particles rapidly agglomerate to larger particles with a size of approximately 250 μm, which are visible at 2 and 3 minutes. These particles slowly develop in time due to normal growth and reach a size of approximately 500 μm at 3 hours after startup. At that time 2 to 3 mean residence times have elapsed thus the majority of the first particles have been removed by the product discharge. Due to this, a gradual increase of supersaturation has occurred. In addition, large particles erode due to collisions. Due to the large level of supersaturation most of these particles survive and thereby create a new population of small particles, which are clearly visible in between 3 to 5 hours after startup. With these new particles a new cycle of outgrowth-erosion-nucleation starts. Meanwhile some very large particles are still visible (see the last image). Note that these large particles are strongly rounded by the erosive action due to collisions.

In Chapter 5 we further discuss these phenomena. In [133] the use of the CCD camera is described specifically for monitoring of the agglomeration phase. In using the CCD camera Ó Meadhra [175], found a strong dependence of particle shape on the chromium level in the system.
Fig. 3.11: Twelve images observed with the on-line CCD camera, during startup and successive outgrowth of crystals, at different time instants ranging from 0 min. to 8 hr.
Chapter 4

Interpretation of dynamic CSD sensor data

In the previous chapter forward light scattering and a simple particle counter are introduced, and some implementation issues for the on-line use of these sensors have been addressed. In this chapter problems are addressed that arise from the fact that these sensors are used to measure a parameter that is both temporarily and spatially varying.

4.1 Introduction

Forward light scattering and particle number counting are the main sensing techniques, applied in this work for both model verification and control. Before applying the sensors some basic questions are addressed.

A first question that arises is what information is actually needed for model verification and control. For control, in the first instance, the information needed is determined by the control objective, for example a process parameter to be optimized. Secondly, for control the answer to this question relates strongly to the concept of input-output controllability [221]. Input-output controllability denotes the ability of a plant to achieve acceptable control performance, that is, to keep the process outputs within specified bounds or displacements from their setpoints.

As shown in Chapter 8, controllability is an inherent plant property, that can be influenced only by changing the plant, for example, re-sizing the equipment, relocating the actuators, and changing or relocating the sensor. Hence, if an existing plant has inherent limitations to influence a certain (sub-)set of output quantities, it is no use (for control) to install a sensor that measures these quantities. Furthermore if controllability analysis, on the basis of a model, reveals that only one parameter in the process is controllable, whereas the sensor outputs five parameters, only one output should be selected — or a combination of the five outputs— that gives favourable dynamic behaviour, and is "close" to the process parameter that can be influenced. So for control purposes, the information that is needed depends on the parameter that should be
optimized with the controller. A plant model is needed to study how the selection and location of a sensor affects the input-output controllability. In Section 8.6 this study will be elaborated further. In this chapter we merely focus on the information that can be gained from a CSD sensor.

For model verification the answer to the above question is different. Loosely speaking, process information is needed on the phenomena for which we have introduced uncertain model parameters. Thus, accurate process data are needed to estimate model parameters which have a large uncertainty. Because our crystallizer model contains uncertain parameters that affect the CSD in both the small and large crystal size ranges, information is needed over a relative broad size range. In addition, because only a limited number of degrees of freedom are present in the CSD (cf. Chapter 8), the range of the sensor is much more important than its resolution.

A second problem is that most CSD sensors are indirect, i.e. the output of the sensor does not provide information that is stated in terms of a CSD. Consequently, it is important to know what information is outputted by the sensor and how the CSD or related quantities can be inferred from the raw sensor data on the basis of a model. Different authors discuss so called deconvolution or inversion methods as a solution to the latter problem. Basically, these methods rely on an inverse of the sensor model to estimate the process parameter of interest from the raw sensor output data. Problems will arise when the sensor model is ill conditioned, for example, when large fluctuations in the parameter(s) of interest cause small and correlated fluctuations in the measured sensor outputs. In addition, the presence of modelling errors may cause significant errors in the estimated parameters as well.

Generally sensors for forward light scattering are ill conditioned. Consequently, the estimation of CSD information directly from noisy diffraction data may give poor results. Within a control context the concept of observability is closely related to the invertibility of the sensor model. Qualitatively a process equipped with a sensor that has a limited invertibility is denoted as badly observable. In Chapter 7 we will further elaborate on the observability of the process.

A third problem is that changing conditions in the process or the sampling system may affect the measurements. For the specific case of forward light scattering the particle volume concentration, which is determined by the magma density of the product slurry and the dilution ratio, will be strongly fluctuating mainly due to a non constant dilution ratio. Therefore, it will be important to known whether these fluctuations influence the measurements and whether the measured data can be corrected for it.

In summary, the application of a sensor for the size distribution of crystals in a crystallization process—and to a large class of other particulate processes as well—brings up the following questions:

1. What information is needed for a certain application.

2. What PSD information is outputted by the sensor, and how can the desired
information, such as the CSD or related quantities, be inferred from the sensor output.

3. How do several varying operational conditions, such as the concentration of solids in the optical cell, affect the measurements.

An exhaustive treatment of the posed questions is not aimed at in this chapter. We will merely address the last two problems in view of the modelling and control applications presented in the sequel. For this both sensor data and sensor models will be used. The first problem requires a verified process model and will be treated within a control context in Chapter 8 and Chapter 9.

First the influence of the volume concentration of solids and the particle shape on the data from the forward light scattering sensor is studied. On the basis of this we will outline how the disturbing effect of concentration fluctuations can be removed from the data. Secondly the sensitivity of the particle counter is estimated on the basis of additional Malvern measurements that were made in the counter loop. Applying principal component analysis to the experimental data from the Malvern particle sizer, we will show to what fluctuations in the CSD this sensor is most sensitive. Finally we will show how specific quantities can be estimated from the CSD data and we will shortly discuss the relevance of direct inversion for model verification and control.

4.2 Effect of solids concentration on CSD measurements

Knowledge on the relation between solids concentration in an optical cell and CSD information obtained from forward laser light diffraction on these particles is important. It determines the amount of dilution needed to obtain CSD measurements, that are not corrupted by multiple scattering. In addition, experience in operating the pilot crystallizer showed that often transients are present in the dilution ratio due to a varying yield of the slurry injection pump or due to a varying dilution liquid flow (cf. Fig. 3.6). Only in the last set of experiments the flow indicators were added to the slurry diluter to determine the dilution ratio. Therefore the concentration fluctuations are seen as an unknown disturbing factor for which we seek an adequate correction here.

From two sets of experimental results, the effect of particle concentration has been studied. For this the slurry flow to be diluted and the dilution liquor flow have been measured according to the set-up described in Section 3.3.2. In a first experiment the concentration has been changed over a broad range during a short period of time, by deliberately changing the slurry flow. During this experiment the CSD in the crystallizer remained constant. A second experiment has been performed, where the particle concentration was measured over a long period of time, while the CSD fluctuated strongly.
The solids concentration in \([m^3/m^3]\) of the diluted slurry is calculated from:

\[
c = 1 - \varepsilon = \gamma_d \frac{\rho_l - M_t}{\rho_l - \rho_c}
\]  
(4.1)

where \(M_t\) is the measured from an E&H M-point\textsuperscript{TM} density indicator, which was located in the product slurry flow, and \(\gamma_d\) is the slurry dilution ratio calculated by subdividing the mother liquor flow by the slurry injection flow. The mother liquor density \(\rho_l\) and the crystal density \(\rho_c\) are calculated from standard formulas presented in Appendix D.

The results of the first experiment are given in Figs. 4.1 and 4.2. In Fig. 4.1 the measured obscuration is given as a function of the solids concentration in the optical cell, which has been varied from 0.5 to 5 percent, by changing the non-diluted slurry flow rate.

The obscuration versus concentration data are described using the well known Beer-Lambert law:

\[
obsc = 1 - \exp(-l \int_0^\infty A_p(x)n(x)Q_{ext}(x)dx)
\]  
(4.2)

where \(l\) is the width\textsuperscript{1} of the optical cell, and \(Q_{ext}\) is the extinction coefficient. \(A_p\) is the cross-sectional area of particles (e.g., for spheres: \(A_p = \frac{\pi}{4}x^2\)). Assuming Fraunhofer diffraction is the dominant diffraction mechanism, \(Q_{ext}\) is given by \(Q_{ext} = 2\), and the exponent will be proportional to the total projected surface area of crystals.

Furthermore, if we assume that the distribution did not change the exponent in Eqn. 4.2 is taken proportional to the solids concentration: \(Kc\). Which reduces this equation to:

\[
obsc = 1 - \exp(-Kc)
\]  
(4.3)

Notice that for low values of the concentration \(c\), this relation is approximated well with the first (linear) term of the Taylor series expansion of Eqn. 4.3 at \(c = 0\): \(Obsc \approx 1 + Kc\).

Estimation of the constant factor from the experimental data using Eqn. 4.3 resulted in \(K = 16.94\). The corresponding fit is given by the solid line in Fig. 4.1. As can be seen this line reasonably fits the trend in the measured data, however, some small deviations are present. These might indicate that the distribution has changed during the experiment. This might be due to changing entrance classification conditions at the location where the varying flow to be diluted is extracted from the main slurry flow (see Fig. 3.6). However, also anomalous diffraction or multiple scattering might be responsible for the observed deviations. Unfortunately on the basis of the presented results it is not possible to discriminate between these error sources.

In Fig. 4.2 also the median crystal size (Eqn. 4.20) and the quartile ratio (Eqn. 4.21) are plotted as a function of the concentration. These values are estimated from the scattered light energy, which has been recorded from the detector rings during the same experiment, by inverting the diffraction model as described in Section 4.6.2. These

\textsuperscript{1}also denoted as \textit{path length}.
plots also indicate that the CSD has changed. However, for solids concentrations below 2 percent, these deviations are in the noise. Consequently, for the described experimental setup it is decided to dilute the slurry until the obscuration is in the range of 20 to 30 percent (i.e., 1.5 to 2 volume percent solids).

The relation between concentration and obscuration for a non-constant CSD has been studied from the results of a system identification experiment denoted as RUN56, which is described in Chapter 6. During this experiment the CSD is excited by deliberately manipulating the process inputs with frequency rich test signals. In Fig. 4.3, the scatter plot of the concentration versus obscuration is given, with the Beer-Lambert law fitted on the results given in Fig. 4.1. Notice that the concentration varied over a smaller range than in the former experiment. In addition, these variations have not been deliberate, but due to both a changing magma density in the process and to disturbances in the dilution ratio as well.

The main axis in the cloud of points in Fig. 4.3 is due concentration variations. The deviations that are present in the direction orthogonal to this axis are mainly explained by the changing distribution, that affects the obscuration according to Eqn. 4.2. Miller [156] uses this additional information for control of a batch crystallizer.

In Fig. 4.4, also the total scattered energy calculated from

\[
y_T = \sum_{i=1}^{31} y_i, \tag{4.4}
\]
4.2 Effect of solids concentration on CSD measurements

Fig. 4.2: Effect of solids concentration on reconstructed median and quartile ratio, fitted with a second order polynomial.

Fig. 4.3: Scatter plot of solids concentration versus obscuration for non constant CSD, with fitted Beer-Lambert law.
is plotted versus the obscuration. As can be seen from this plot the total scattered light energy, after scaling, approximately equals the obscuration signal.

The presented results show that changes in the magma density as well as disturbances in the dilution ratio, cause variations in both the obscuration and the scattered light measured on the detector rings. These variations are approximately proportional to the fluctuations in the solids concentration in the optical cell. On the basis of this it is decided to eliminate the corrupting effect of the varying dilution ratio simply by subdividing each independent ring signal by the total scattered light.

\[
y_i = y_i / y_T, \quad i = 1, \ldots, 31
\]

The total scattered \( y_T \) instead of the obscuration signal is used, because the noise contribution on the former signal is found to be significantly lower. The merits of this normalization step are illustrated in Fig. 4.5. In the upper plot raw data, measured from the fifth ring of the Malvern detector during RUN60 (see Chapter 10), is given. In the lower plot the same signal after applying the normalization step is depicted. As can be seen the signal to noise ratio is improved and the strange transients in the raw data, which are mainly due to transients in the dilution ratio, are removed effectively.

However, due to the normalization step some information is lost. If the distribution does not change (cf. Fig. 4.1), merely concentration information is lost. But actually there will be no loss of information, because the concentration information is already obtained directly (i.e., without needing the uncertain dilution step), from the independent density indicator. When the distribution changes (Fig. 4.3) the obscuration mainly correlates with the concentration. However, for this case also a dependency on the total projected surface area, or second moment of the CSD is present, as described by Eqn. 4.2. Consequently, normalization will eliminate this information from the Malvern data. Because the concentration fluctuations are measured with the independent density sensor, merely the relative second moment of the distribution \( (m_2/m_3) \) is eliminated from the measurements.

Obviously, the same normalization step is implemented in the simulation model to prevent the introduction of additional model errors.
4.3 Effect of the particle shape on CSD measurements

For a comprehensive discussion on the role of particle shape in forward light scattering we refer to [101, 100]. Here we will shortly address how uncertainties in the aspect ratio, which is a main parameter in our sensor model Eqn. 2.33, may bias estimated CSD quantities.

Impurities may drastically affect the aspect ratio. Experimental results presented by [175] show that over a period of 1 year in which the ammonium sulphate substance system was not changed the aspect ratio slowly changed from 1.2 to 2.5. This change was due to an increase of Chromium, which originates from the equipment material\(^2\). In addition, it is observed that individual crystals may have a different shape as well due to attrition (see also CCD images depicted in Fig. 3.11).

To study the influence of these shape changes on the measured diffraction data and on the estimated CSD properties, a set of 10 Fraunhofer matrices have been calculated. Eqn. 2.33 has been evaluated for 10 different values: \(\alpha = \{1.2, 1.4, 1.6, \ldots, 3.0\}\). Assuming a stationary CSD for an MSMPR crystallizer (i.e., the plot of \(\ln(n) - x\) describes a straight line), we calculated the corresponding light energy values on the 31 Malvern detector rings. The results are given in the upper plot of Fig. 4.6. The arrow indicates the direction of the effect of an increasing aspect ratio. In a next step the 10 light energy patterns were used to re-estimate the crystal size distribution by inverting the Fraunhofer matrix that was calculated for the aspect ratio \(\alpha = 2.0\). The results are depicted in the lower plot of Fig. 4.6. Finally, also the volume based median crystal

\(^2\)This is stainless steel 316, containing 18 percent chromium
Fig. 4.6: Effect of particle shape on CSD measurements. Upper plot normalized energy patterns calculated from exponential distribution fraunhofer matrices for 10 different shape factors; lower plot reconstructed distributions for scatter matrix with shape factor $\alpha = 1.2$.

size was calculated from these reconstructed distributions: the median crystals sizes varied in the range of 809 $\mu$m to 763 $\mu$m is found, whereas the original distribution had a median of 783 $\mu$m.

Clearly, these results show that the particle shape has a significant effect on the measured light energy and the reconstructed CSD. Careful modelling of the CSD sensor is therefore a necessity when light scattering data are used to to validate models or to estimate model parameters related to CSD dynamics.

In most simulations a value of 1.4 for $\alpha$ has been used. When fitting the process model to the measured data (see Chapter 5), deviations in the shape caused, for instance, by attrition or impurities may thus cause biased parameter estimates.

### 4.4 Identification of the counter model

The counter model is described in Section 2.2.7. Estimation of the unknown parameters $\theta_S = \{p_{s1}, p_{s2}, p_{s3}\}$ in the model (Eqn. 2.37) is performed on the basis of measurements on the population density of fines $n_f(x,t)$, that flows through the optical cell of the counter. For this purpose an additional Malvern particle sizer has been installed in the counter loop.
Table 4.1: Nominal parameter values and uncertainty bounds.

<table>
<thead>
<tr>
<th>parameter</th>
<th>nominal value</th>
<th>lower bound</th>
<th>upper bound</th>
</tr>
</thead>
<tbody>
<tr>
<td>$p_{s1}$</td>
<td>120 [μm]</td>
<td>85 [μm]</td>
<td>155 [μm]</td>
</tr>
<tr>
<td>$p_{s2}$</td>
<td>1.8</td>
<td>1.4</td>
<td>2.1</td>
</tr>
<tr>
<td>$p_{s3}$</td>
<td>1.0</td>
<td>0.7</td>
<td>1.3</td>
</tr>
</tbody>
</table>

Assuming a perfect counter, the particle detection efficiency Eqn. 2.37 is approximated with $K_S(x) = n_f(x,t)/n(x,t)$. Using this relation, $p_{s1}$ and $p_{s3}$ could be estimated from the Malvern measurements. However, these measurements provide reliable information on the relative distribution only, and a density indicator was not present in the loop. Consequently, the absolute value for $p_{s3}$ could not be estimated. Therefore it is used as a scaling factor for the total counted number of fines. In Chapter 9 this factor is used to fit the simulation data on the model data. The normalized (i.e., $p_{s3} = 1$) estimated detection efficiency curve is depicted in Fig. 4.7. The estimated values for $p_{s1}$ and $p_{s2}$ are given in Table 4.1.

Proper identification of the detection curve $K_S(x)$ was difficult. To describe $K_S(x)$ with a log-normal curve is expected to be a rough approximation. Furthermore, good sampling conditions in the counter loop were difficult to establish: the low sample flow rate (20 ml/s), might have cooling of the slurry which causes additional nucleation. Besides, plugging of the slurry transportation line occurred frequently. Therefore the estimated parameter values are uncertain. A (conservative) guess is made of the uncertainty region where the true parameter value will lie. The numerical values are given in Table 4.1: for $p_{s3}$ only a relative uncertainty is given. In Fig. 4.7 the dotted area indicates where the true detection curve will be located. In Section 9.3 these values are used to evaluate whether a designed closed-loop controller is robust in the face of these parameter uncertainties.

4.5 Analysis of the Malvern particle sizer using experimental data.

Using the data which is normalized according to Eqn. 4.5 we seek an answer to the question: what information on the CSD dynamics is present in the normalized sensor data. The 31 signals from the Malvern are likely to be correlated, due to the principle of forward light scattering. An important tool to find the number of (uncorrelated) components in data that is both spatially and temporally varying, is the principal component analysis (PCA). We will apply PCA to find the principal components in the Malvern data. Successively the sensor model is used to analyze how the principal components relate to the CSD. First, however we will shortly explain PCA. For a comprehensive treatment, we refer to Jolliffe [122].
Fig. 4.7: Estimated detection efficiency of the fine particle counter with uncertainty range.

4.5.1 Principal component analysis

The basic idea with principal component analysis is to describe a set of data in terms of a reduced set of data, which retains as much as possible of the variations (energy) present in the original set. The principal components are computed easily from an eigenvalue-eigenvector decomposition. In mathematical terms, the principle component vector \( z \) that contains the principal components, \( z_k, k = 1, \ldots, p \) of the correlated elements of the original vector \( x \) (e.g., a sample from a multiple array sensor), is obtained from a linear transformation:

\[
z = A^T x
\]  

where \( A \) is an orthogonal matrix whose \( k \)th column, denoted as \( a_k \), is equal to the \( k \)th eigenvector of the covariance matrix \( \Sigma \) of \( x \). The latter given by:

\[
\Sigma = E\{(x - \bar{x})(x - \bar{x})^T\}
\]  

where \( E \) denotes the expected value. Furthermore, the following relation will hold [122]:

\[
\Sigma A = A \Gamma
\]  

where \( \Gamma \) is the diagonal matrix whose \( k \)th diagonal element is \( \lambda_k \), which is the \( k \)th eigenvalue of \( \Sigma \). In addition the eigenvalues directly represent the variance of the principle components (see [122] for a proof):

\[
\lambda_k = var(a_k^T x) = var(z_k)
\]

So if the eigenvectors and eigenvalues are sorted in decreasing order, the best linear combination of elements in \( x \), representing the largest signal variance (energy), is represented by the eigenvector \( a_1 \), and the corresponding eigenvalue \( \lambda_1 \) then represents the signal energy in this specific signal direction. Clearly, the least significant signal direction is represented by the eigenvector \( a_p \) and the eigenvalue \( \lambda_p \). A reduced representation of the original vector \( x \) is obtained easily by omitting the least significant signal directions, which will mainly represent a noise contribution to the signal.
The derivation of this result is standard and can be found in many text-books [122, 77]. In some text-books the decomposition method is also known as the discrete version of the Karhunen-Loève expansion [77]. An illustrative application of PCA to a spatio-temporally varying chemical process is reported by [136].

A simple example may illustrate some merits and characteristics of PCA, when applied to a spatio and temporally varying process. Consider a data set consisting of \( p \) sinusoids \( x_i \) with amplitude 1: which are shifted in phase according to:

\[
x_i(t) = \sin\left(\frac{2\pi}{T} t + \frac{2\pi i}{p}\right) \quad i = 0, \ldots, p - 1
\]

and recorded over \( t = [0T] \). The variance of each original signal is equal to 0.5, thus the total variance of all signals is 0.5\( p \). If an eigenvalue decomposition of the covariance matrix \( \Sigma \) of the original data is conducted, two non-zero eigenvalues are found each with a value of 2.5\( p \). Thus each principal component sums the energy of \( p/2 \) original signals. If a white noise with a variance of 0.01 is added to the original signal, then still two dominant eigenvalues are found. However, now also \( p - 2 \) eigenvalues are found which fluctuate randomly around a mean value of: 0.01, whereas the noise variance on the first two principal components is still 0.01 percent. So applying PCA to this data set all the energy of the deterministic part of the original set is condensed on two uncorrelated signals, whereas the remaining \( p - 2 \) components are white noise sequences with variance 0.01. Clearly, adding a sinusoid to the original data set with the same amplitude but a higher frequency, one additional eigenvalue would be found with the same magnitude.

Notice that PCA has no filtering capabilities: if it is applied to a set of uncorrelated signals, which are corrupted with a white noise, the number of principal components is equal to the number of original signals, and the noise contribution is the same as well. This exercise shows that PCA applied to a spatio and temporarily varying process, projects correlated process information onto a reduced set of uncorrelated components.

**Finding the number of elements**

A major problem in applying PCA to multivariate data is to find a value for the number of components. Most rules for this are ad-hoc rules of thumb, which may work good in practical situations. The most simple method is to select enough components to explain a large part of the signal energy. According to Eqn. 4.9, this may be done on the basis of the eigenvalues directly. Figures between 80 and 90 percent are often used. Alternatively the eigenvalues may be plotted. Then the location in the plot where large eigenvalues cease and small eigenvalues begin is taken as an indicator for a good value for \( m \). The eigenvalue plot is also known as the scree diagram. Often, also the so called logarithmic eigenvalues (LEV) diagram is used, where \( \log \lambda \) rather than
\( \lambda \) is plotted. In Jolliffe [122] an overview is given of all possible criteria to select the number of components, including methods with a more sound statistical foundation.

Most PCA applications reported in the literature elaborate on mutually uncorrelated sets of correlated variables. The sensor data used here differs from this in that it has both a temporal and spatial component, which are both correlated. Hence we can cross validate the result by estimating the original vector from the principal components. Because the columns of \( A \) are orthogonal this can be done easily with: \( \tilde{x} = Az \) and the residuals can be calculated from:

\[
e = x - \tilde{x} = x - Az
\]  

(4.11)

Visual inspection of the residuals may then be used to inspect whether these the errors are white. More formal whiteness tests may be applied as well (see [144]).

**Scaling**

The structure of the principal components will depend on the numerical values of the original signals. For instance, these may depend on the applied units of measure. To prevent large values to dominate the results or a small value to be rejected, scaling is important prior to the calculation of the principal components. A common approach is to transform \( x \) according to \( x = x/\sigma_{jj}^{1/2} \). Instead of the covariance matrix \( \Sigma \), the correlation matrix \( R \) is then used for eigenvalue calculation. The correlation matrix relates to \( \Sigma \) according to:

\[
\Sigma = \Gamma R \Gamma
\]  

(4.12)

with \( \Gamma \) a diagonal matrix with the variances: \( \{\sigma_1, \ldots, \sigma_p\} \) on its diagonal.

Alternatively a weighting factor \( w_{jj} \) may be applied, however it will be difficult to find a non subjective justification for such a choice.

**4.5.2 Application of PCA to dynamic Malvern sensor data**

PCA is applied to 10 data sets of historical process data which have been measured from the UNIAK pilot crystallizer over 4 years with different sampling systems and different process conditions. An example of one raw data set (RUN56) is given in Fig. 4.8. Notice that this set of data is of particular interest as the CSD has been excited strongly with uncorrelated frequency rich test signals which are added to the process inputs simultaneously (see Chapter 6). Another point to note is that the large correlation visible in this data further stresses the importance of applying PCA.

The first three eigenvectors \( \{a_1, a_2, a_3\} \) corresponding to the first three eigenvalues for each set are presented in Fig. 4.9. In Fig. 4.10 the log eigenvalue plots of all estimated correlation functions are given.

These results show that in all data sets the same principal signal directions are present. The directions are most reproducible for the first eigenvector; down the list the coherence between the eigenvectors vanishes. The last eigenvectors do no show
4.5 Analysis of the Malvern particle sizer using experimental data.

Fig. 4.8: Normalized Malvern detector data (RUN56).

any coherence. The eigenvalue plots reveal that there is not a sharp changeover in
the eigenvalue trend, which demarcates a changeover from the desired signal variance
—that is, due to process variations—and the noise variance. As expected the runs
with limited process excitation (cf. Fig. 5.6) have a steeper eigenvalue plot than the
runs with much excitation (cf. Fig. 4.8). However, for all sets the first 6 principal
components grasp already more than 99 percent of the total energy of the original
signal.

Fig. 4.11 shows the first, fourth and seventh principal components that are esti-
mated from the data set in Fig. 4.10. As is predicted by the eigenvalues already,
the variance in the first principal component is the largest and the variance decreases
rapidly down the sequence of principal components. Furthermore we see that signal
to noise ratio is best for the first PC and already extremely bad for the seventh PC.
Notice also that the variance of the first signal is close to 10, which indicates, loosely
speaking, that it contains a contribution of at least 10 correlated signals in the original
set.

Also residuals have been calculated using Eqn. 4.11. Visual inspection revealed that
no significant correlations were present in the residuals after the 20th PC’s. Below this
value still information, or correlations were visible, however, between the 20th and 7th
PC the noise contribution is found to be dominant.

From a control point of view the first principal component is most attractive as it is
the parameter that is most sensitive to variations in the process. Also the second and
third principal component may be useful to serve as output parameters in a control
loop. For control, the principal components have the added advantage that they are mutually uncorrelated. Thus, no interaction will be present in the system, which may strongly facilitate the controller design. Note, however, that it will also be important to analyze the dynamic input-output characteristics of the plant, that uses these signals as an output parameter. The first principal component as a process output parameter to be controlled is denoted in the sequel as: $y_{r1}$.

The principal components estimated so far are used in the following section to show where the largest observable variations in the CSD dynamics occur.

### 4.5.3 Relation between principal components and the CSD

How the PC relate to the CSD is shown directly with:

$$a_{frh,i} = a_i^T H$$  \hspace{1cm} (4.13)

where $H$ is the sensor matrix and $a_{frh,i}$ shows how the $ith$ component relates to the CSD, i.e $z_i = a_{frh,i}n$. For the eigenvectors depicted in Fig. 4.9 the corresponding vectors in the crystal size domain ($a_{frh,i}$) are shown in Fig. 4.12.
Fig. 4.10: Log eigenvalue plot of eigenvalues corresponding to all 31 principal components in 10 different experimental data sets from the pilot crystallizer.

As can be seen from this plot the first PC mainly correlates with the fines in the system, which are present in the region of 50 to 300 μm. The second and higher PC's correlate with larger particles as well. As can be seen, the sensitivity to variations is negligible above 1200 μm. This shows that when we use the sensor data for model identification it is difficult to obtain good estimates for attrition, which is dominant above 1000 μm. These curves also show that the very small particles (i.e., x < 50 μm), are also not measurable.

4.6 Estimation of CSD properties by inversion of the Fraunhofer matrix

The principal components represent the CSD variations that are best measurable. In addition to this information it may be useful to know how well a certain — user chosen — CSD property can be reconstructed from sensor data. First the estimation of a scalar CSD property from raw sensor data is addressed. Thereafter, the estimation of whole size distributions is discussed.

4.6.1 Estimation of a scalar CSD property

Estimation of the mean crystal size from raw sensor data is discussed to illustrate how well a scalar property or the CSD can be reconstructed. The (mass based) mean
Fig. 4.11: Principal components $z_1$, $z_4$ and $z_7$, calculated for the data set presented in Fig. 4.8 (RUN56).

crystal size is calculated from the third and fourth moment of the population density:

$$X_{43} = \frac{\int_0^\infty n(x)x^4dx}{\int_0^\infty n(x)x^3dx} = \frac{m_4}{m_3} \quad (4.14)$$

When only small perturbations around the stationary distribution are assumed, this relation can be linearized, resulting in:

$$\Delta X_{43} = \frac{m_{4s}}{m_{3s}} \Delta m_4 - \frac{m_{4s}}{m_{3s}^2} \Delta m_3 \quad (4.15)$$

Lumping gives:

$$\Delta X_{43} = T_{43} \Delta n \quad (4.16)$$

with $T_{43}$ a column vector containing the elements $[a_1, a_2, \ldots, a_n]$ given by:

$$a_i = \left[ \frac{m_{4s}}{m_{3s}} x_i^4 - \frac{m_{4s}}{m_{3s}^2} x_i^3 \right] \Delta x \quad i = 1, \ldots, n \quad (4.17)$$

With the sensor model $H$, and the relation $y = H \Delta n$, the transformation of the sensor output $y$ to the mean crystal size can be written as:

$$\Delta X_{43} = T^T y = T_{43} H^{-\dagger} y \quad (4.18)$$
4.6 Estimation of CSD properties by inversion of the Fraunhofer matrix

Fig. 4.12: Major directions in the crystal size domain related to the first three principal components.

Fig. 4.13: Comparison of vectors $T_r^T H$ with $\Delta X_{43}$ for different values of the number of components $n_r$.

where $T$ is the vector that maps the measured sensor output onto $\Delta X_{43}$, is the vector that is searched for.

The pseudo inverse $H^{-\dagger}$ is approximated from the singular value decomposition $H = U^T \Sigma^{-1} V$ [89], where the matrices $U$ and $V$ contain the $n_y$ right, and left singular vectors of the sensor model $H$, respectively, and $\Sigma$ contains the corresponding singular values. As the matrix $H$ is ill conditioned (i.e., contains both large and very small singular values), only the $n_r$ singular vectors are used in the inversion: $T_r = T_{43} U_r \Sigma_r^{-1} V_r^T$, with $n_r < n_y$. If a sufficient number of components $n_r$ are taken into account, then $T_r^T H$ is expected to closely approximate the vector $\Delta X_{43}$. In Fig. 4.13 these vectors $T_r^T H$ are compared with the vector $\Delta X_{43}$ for different values of the number of components $n_r$.

Using simulation data we can now evaluate how well $T_r y$ approximates the real trend of $\Delta X_{43}$. The results are given in Fig. 4.14. First the case without noise is studied. As can be seen for $n_r < 20$ a significant bias from the trend of $X_{43}$ is observed. So this would suggest to take into account more than 20 components. However, in the same figure the case with noise on the output signals is considered. As can be seen above 20 components a large noise contribution is introduced in the reconstructed signal. This is mainly due to large numerical values in the vector $T_r$, which are caused
by taking into account small singular values in the inversion of the sensor model. Consequently, for reconstruction of a certain parameter from sensor data, a trade-off should be found between bias and noise. Clearly for the given example a number of 20 components would be reasonable.

4.6.2 Estimation of whole size distributions

Many authors elaborate on numerical deconvolution schemes to estimate the whole size distribution on the basis of the measured sensor output [24, 37, 36, 253].

A common approach to realize this is to invert the sensor model $H$ directly. For this the standard quadratic solution can be used:

$$\hat{n} = (H^T H)^{-1} H^T y$$

As shown by the results in the previous subsection, such an approach leads to a highly erratic solution that is due to the ill-conditioned sensor model. Different modifications to this basic solution have been published. These mainly attempt to dampen the solution and to constrain the solution to a physically feasible range. A review of these techniques is given by Boxman [24].

The main attractiveness of deconvolution is that it can be applied on-line easily, because the computational burden is low and not much a priori knowledge on the underlying process physics is needed [24, 63]. Besides, the estimated quantities are
4.7 Conclusions

directly physically interpretable which may be valuable for process operators or engineers.

An important alternative to deconvolution is to use a state estimator. With state estimation a dynamic model is used to estimate the crystal size distribution from sensor data. The importance of this scheme in comparison to deconvolution is that it takes into account the correlation in time of the data as well. We will elaborate further on the design and evaluation of state estimators in Chapter 7.

No incentive is found for integrating a deconvolution step in a control loop, because the control relevant information can often be obtained directly from the raw sensor output as is described in the previous sections and as is done in the control chapters at the end of this thesis. Also for model verification it is better not the use results from deconvolution as the deconvolution algorithm introduces additional errors in the data. In addition, the sample covariance matrices, which are used to penalizes errors subject to large random fluctuations in a model fit criterion (cf. Eqn. 5.1), are obscured by the deconvolution step.

In this work deconvolution has been applied for monitoring purposes. Using the ROMA software [24], CSD properties are estimated from the raw sensor data. The inversion algorithm employed in this package is based on a weighted least squares method with an additive non-negativity constraint on the estimated distribution. From the deconvolution step a relative volume distribution on 20 size classes, denoted as \( q(j) \), \( i = 1, \ldots, 20 \), is obtained on a logarithmic grid with 23 % resolution, that is, the width of successive logarithmic classes increased with a factor 1.23. The size range, in most results reported in this work, is chosen from 13.35 \( \mu \text{m} \) to 1333.5 \( \mu \text{m} \).

From these results, volume distribution characterizing quantities are calculated. Robust parameters are derived from the quartiles of the size distribution, defined by:

\[
\int_{x=0}^{X_k} n_v(x) \, dx \triangleq \frac{k}{100}, \quad \text{with } k = 25, 50, 75 \text{ and } 100.
\]  
(4.20)

The median (\( X_{50} \)), is a measure for the location of the CSD. The logarithmic interquartile range is used as a measure for the spread:

\[
qr \triangleq \ln \frac{X_{75}}{X_{25}}.
\]  
(4.21)

As shown above the linearized versions of these quantities may be estimated directly from the raw sensor data as well.

4.7 Conclusions

In this chapter we have shown that interpretation of the data from on-line sensors is not trivial.
We have shown that fluctuations in the volumetric solids concentration may corrupt sensor data. Normalization of the data using the total scattered light energy is applied to correct for this. If the distribution remains constant this correction merely eliminates concentration information from the data. If the distribution changes information on the total projected surface area of crystals is lost by applying this normalization.

It is found that errors in the assumption on the shape of the particles may significantly affect the simulated output of Incorrect shape changes do affect the Malvern data.

From a principal component analysis it is found that only a limited number of independent degrees of freedom can be grasped from the output. In terms of signal variance a set of 6 principal components always describes more than 99 percent of the variance in the original data. The first component is found to be a signal with a low signal to noise ratio which explains already 85 percent of the energy that is present in the 31 outputs of the Malvern. It is found that these components are mainly sensitive to CSD changes in the range 50 to 1000 μm outside this range the Malvern has a low sensitivity.

For estimation of physical quantities from diffraction data a trade-off should be found between sensitivity to noise and bias. Singular value decomposition of the Fraunhofer matrix is found to be a valuable tool for achieving this.

Deconvolution applied for estimation of crystal size distributions is concluded to be a valuable tool for process monitoring and (rapid) presentation of data in terms of understandable physical quantities. However no incentive is found for integrating a deconvolution step in a control loop as it is merely a mathematical trick which does not add new information, but transforms information which can be achieved from the raw sensor data directly. For model verification the same argument applies.
Chapter 5

Model verification and parameter estimation

In this chapter several hypothesis underlying the first principles model described in Chapter 2 will be verified. For this, mainly data from the pilot plant described in Chapter 3 is used.

5.1 Introduction

Model verification is concerned with the question whether the quality of a model is sufficient for its intended use. The answer to this question will strongly depend on the intended use. If for instance a model is used to study design issues by means of time series analysis, a fairly accurate description of the static and dynamic model behaviour over a broad nonlinear range of operation conditions is required. On the other hand, for the design of a stabilizing control some model errors, especially in the lower frequencies, will not pose major difficulties.

Some important concepts related to model uncertainty, are introduced in the field of robust control [146]. Here a model—as we have introduced so far—is often referred to as a nominal model and a deviation of this model (e.g., due to uncertain parameters) is called a model perturbation. The robust control design method aims at the design of a controller which has a satisfactory performance for both the nominal model and the complete set of perturbed models. Consequently, model errors may not pose problems as long as an analysis of the robustness of a designed controller reveals that the stability or performance requirements are guaranteed not only for the model, but also for the set of perturbed models. Clearly, the derivation of reliable uncertainty models is a major difficulty for the applicability of this approach. Currently, these robustness concepts are also introduced in related application areas like controllability assessment, process design [222], and observer design [76].

In addition to the knowledge on model uncertainties, it is also important to have knowledge (i.e., models) of disturbances and how these affect the process dynamics.

The intended use of our model is both process analysis and controller design. Hence,
we aim at the derivation of an accurate model that can be applied for different purposes. This also reflects the trend in current industrial practice, where one strives to build one rigorous model including both macroscopic and microscopic phenomena for a specific process or a class of processes, to serve a broad spectrum of control, analysis and design related applications.

The process model presented in Chapter 2 comprises a large set of unknown empirical parameters. In addition, several hypothesis are introduced. In Chapter 4 we evaluated the model hypothesis already for the sensor models. Here we will first evaluate some major hypothesis, using experimental data from some specific experiments. This will give some sense for the limitations of the model. Obviously, a thorough (experimental) evaluation of all model assumptions is laborious and does not fit within the framework of this thesis. For this we refer to the work of the other UNIAK members [86, 100, 175].

In the next step a procedure is outlined for the estimation of experimental model parameters from dynamic free-run startup experiments. With these free run experiments, no CSD controller is active and no deliberate excitation of the plant dynamics is enforced. Therefore this method may be attractive in industrial practice. The resulting data are expected to bear information on several kinetic crystallization phenomena. The estimation of kinetic parameters from these data are discussed and we will point at some limitations that exist in the model and the proposed parameter estimation procedure. In addition, estimates will be provided for the parameter errors. Later, these estimates will be used to judge the robustness of the designed controllers.

Finally the accuracy of the optimized model is judged from two additional experimental runs conducted at different experimental conditions. In the first set the process is operated with the vibrating screen classifier as a product classification device. In the second set a short series of batch experiments is conducted. For both cases the model responses are compared to the measured experimental data, and conclusions are drawn with respect to the model validity.

In Chapter 6, which is closely linked to this chapter, we will focus specifically on the derivation of models for control purposes. Then deliberate excitation of the process inputs and noise inputs will be applied. The process input-output data thus obtained will be used for verification of the first principles model and the estimation of input-output models directly from process input-output data.

### 5.2 Evaluation of model hypothesis

Most model assumptions need no further discussion as they are related to tightly controlled conditions in the experimental equipment, e.g. a constant volume, a constant temperature and a constant fines temperature. In this section the following phenomena and model assumptions are studied:
5.2 Evaluation of model hypothesis

1. The factors that do influence the onset of crystallization right after unseeded startup and the variability of the initial distributions.

2. The difference between the Class I and Class II model.

3. To what extend internal classification in the crystallizer vessel (see discussion Section 2.2.3) occurs.

5.2.1 Evaluation of the onset of crystallization at unseeded startup

In industrial practice crystallizers are frequently restarted, after a cleaning or maintenance stop, with an initial charge in which no or just a sparse amount of crystals (seeds) are present. The absence of crystal surface, necessary for the consumption of the supersaturation by growth, will give rise to large supersaturation levels. The development of an initial CSD is therefore expected to be dominated by primary heterogenous nucleation (see also Fig. 2.6) and growth of primary nuclei in the supersaturated mother liquor. We will denote the result of this startup process as the initial CSD. During further outgrowth of this initial CSD primary nucleation is assumed to be absent. The reproducibility of this initial CSD is and the factors that might influence its shape are explored using experimental data.

Results of unseeded startup experiments

The startup CSD response of the pilot crystallizer has been measured, over a period of 10 minutes, as a function of the total heat input. Fines removal was absent during these runs. Four different experiments have been conducted, where the pilot crystallizer is initially charged with a crystal-free liquid. During each experiment the development of the initial CSD has been observed for different values of the heat input. Besides each experiment has been done triple to judge the reproducibility.

As the initial CSD is developed in a short time period, a rapid sampling technique has been applied. For this the Malvern particle sizer has been installed directly in the product line and all removed slurry was measured without dilution. A CCD camera has been used to monitor the crystals in the product line.

The scattered light has been recorded at 31 detector elements by the Malvern 2600 particle sizer using a 300 mm collimating lens. At each sample instant a batch of 10 detector readings (sweeps) has been taken with a sample period between two sweeps of approximately 10 ms. These sweeps were averaged and the standard deviation \( \sigma_i, i = 1, \ldots, m \) for all the \( m \) rings was calculated. The sample time between averaged batches of sweeps was 5 seconds. Deconvolution has been applied as described in Section 4.6.2 to obtain a relative volume distribution on a logarithmic grid with 20 size classes. From the distribution a median and quartile ratio are calculated.

Table 5.1 summarizes the different values for the heat input. The other experimental conditions are summarized in Appendix D. In Fig. 5.1 the measured startup
Table 5.1: Values for total heat input for agglomeration experiments.

<table>
<thead>
<tr>
<th>experiment</th>
<th>EXP1</th>
<th>EXP2</th>
<th>EXP3</th>
<th>EXP4</th>
</tr>
</thead>
<tbody>
<tr>
<td>heat input [KW]</td>
<td>40</td>
<td>80</td>
<td>120</td>
<td>160</td>
</tr>
</tbody>
</table>

responses of the obscuration, the median and the quartile ratio are depicted over 10 minutes in time for different values of the heat input. In Fig. 5.2 the development of the reconstructed relative volume distributions are given corresponding to a heat input of 160 KW and a heat input of 40 KW respectively.

Discussion of unseeded startup experiments

The results reveal a strong dependence of the CSD response on the evaporation rate. For large values for the heat input $P \geq 80$ KW the obscuration and the CSD dynamics show fast dynamics. After the first 2.5 minutes, the obscuration reaches a peak value of approximately 75%, while the median and the spread remain at a low value. The increase of the obscuration is mainly due to strong primary nucleation, driven by the high value of supersaturation. Surprisingly, between 2.5 and 3.5 minutes the obscuration decreases, while the median increases sharply from 40 to 250 μm, within 30 seconds, and the spread only peaks shortly. The latter points at the existence of a bi-modal shaped CSD. Notice that due to the large values of the obscuration the values for the median crystal size are biased due to multiple scattering.

Both the decrease of the obscuration and the rapid increase of the median crystal size, point at the occurrence of a short temporal agglomeration phase, in which particles of approximately 20 to 50 μm are "glued" by the supersaturated liquid to form a cluster with a size of approximately 250 μm. With low evaporation rates only a relative slow change of the CSD shape is detected, induced by normal diffusion controlled outgrowth of the initial distribution. The results suggest a good reproducibility of the CSD from an unseeded start-up, provided that the heat input is not altered.

It is common to use the term agglomeration instead of aggregation whenever particle clusters are formed, that are tightly cemented and do not rupture. The agglomeration rate is determined by the number density of small particles and strongly promoted by the level of supersaturation. For large values of the heat input agglomeration can be present due to both the large number density of small nuclei caused by excessive primary nucleation and the high level of supersaturation. Agglomeration is undesired as it lowers the mechanical properties of particles and due to liquid inclusions, it may lower the purity as well.

The common approach to model aggregation phenomenon, in the absence of rupture, is to include a death and birth term in the population balance (see Eqn. 2.29). Detailed models for aggregation are presented by different authors [28, 41, 42, 97, 96, \ldots]
Fig. 5.1: Trends of obscuration, median and quartile ratio, during the first 10 minutes after startup, at four different values for the heat input: 160 KW (upper left), 120 KW (upper right), 80 KW (lower left), 40 KW (lower right).
Fig. 5.2: Trend of relative light intensity distribution at startup, for 160 KW heat input (left) and 40 KW heat input (right)

Examples of a bi-modal shaped CSD, which may exist due to agglomeration are given by [13, 28]. For modelling attempts of similar experiments as the one reported here, we refer to [176].

The results give rise to the following practical rule: operate unseeded crystallizers with a low value for the heat input at start-up. Further the variability of the results, for one value of the heat input is small. Therefore its is reasonable to describe the initial CSD with a deterministic distribution as is proposed in Section 2.2.4.

5.2.2 Comparison of Class I and Class II model

The difference between the Class I and Class II models is studied using the simulation model. This difference is found to be most pronounced for variations in the heat input to the system. Therefore a step change in the heat input is simulated and the response to the supersaturation and the population density at 150μm is studied. Model parameter values presented in Section 5.3 are used. The power \( p_7 \) (cf. Eqn. 2.24) is taken equal to one, which corresponds to published reasonable [129] In Fig. 5.3, the responses of the Class II and Class I model are given. For the latter, different values for the parameter \( p_6^* \) are used. In Fig. 5.4 also the location of the poles of the linearized Class I models are given. In Table 5.2 the values for the relative supersaturation at steady-state conditions for different values for \( p_6^* \) are given.

The results show that \( p_6^* \) affects the speed of response. The effect of \( p_6^* \) is traced back to the location of one pole in the linearized model. The location of this pole is found to be proportional to the value of \( p_6^* \). From the results it is concluded that for \( p_6^* < 2.0 \times 10^{-6} \) the Class I model should be used as the dynamic response differs significantly from the Class II model. For \( p_6^* > 2.0 \times 10^{-6} \), the response of the supersaturation is fast and close to the response of the Class II model. Values of \( p_6^* \) presented by [130, 40] show that most systems can be described using Class II models. Only a
Table 5.2: Steady-state values of relative supersaturation for different values of $p_0^*$. 

<table>
<thead>
<tr>
<th>$p_0^*$ [m/s]</th>
<th>0.57·10^{-6}</th>
<th>1.14·10^{-6}</th>
<th>2.35·10^{-6}</th>
<th>5.71·10^{-6}</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\Delta C/C_s$ [%]</td>
<td>7.3</td>
<td>3.9</td>
<td>1.6</td>
<td>0.8</td>
</tr>
</tbody>
</table>

Fig. 5.3: Response of rel. supersaturation, and $n(x = 150 \mu m)$, for a 10% step change in the total heat input for different values of $p_0^*$. 

limited class of substances do require the use of a Class I model.

5.2.3 Experimental evaluation of spatial segregation effects

On the basis of the discussion given in Section 2.2.3 a short experiment is conducted to study spatial segregation in the pilot crystallizer. In this experiment the magma density $M\gamma$ and the CSD have been measured in the product outlet as a function of the impeller speed. The impeller speed has been varied in the range of 260 to 370 rpm. After each change of the impeller speed the responses to the median crystal size and the magma density the product slurry have been measured during 10 minutes. The total duration of the experiment has been three hours, thus the rotational speed has been varied 18 times. In Fig. 5.5 the 18 values of the median and the concentration of solids, calculated from the magma density according to Eqn. 4.1, are depicted. These values are averaged values of the samples acquired over 10 minutes.

A significant effect is observed: the solids concentration is disturbed for roughly 8% and the median for 5%, for a change in impeller speed of 15% around its nominal value of 315 rpm. Notice that the mass balance requires that the solids concentration
Fig. 5.4: Location of poles of linearized Class I models for different values of $p_6^*$. 

should return to its original value on the longer terms, because the heat input was kept constant. Results presented in [175] show that the long term dynamic response time to impeller variations is slow, that is 6 to 8 hours. For a well-mixed vessel the response time of the solids concentration is determined by the mean residence time of slurry [49], thus with spatial segregation this time will generally be larger and in the order of magnitude of the CSD response dynamics (cf. [175]).

In conclusion spatial segregation has two important implications:

1. The distribution present at the sampling or product withdrawal location generally differs from the average distribution that is present in the crystallizer.

2. The dynamics of the magma density in the crystallizer (and the mass production rate) is significantly correlated with the CSD dynamics. As was shown by De Wolf [49], the magma density in a well stirrer vessel should obey a simple first order differential equation. Consequently higher order dynamics in the response of the magma density to for instance a step change in the heat input is indicative for significant spatial segregation.

In the experiments reported below the impeller speed is kept constant at 315 rpm. Besides, the start-up trend of the magma density is found to be well described with a first order response (see Fig. 5.7). Therefore, spatial segregation will mainly affect the length dependent growth curve Eqn. 2.26, that is estimated from the experimental data.
Fig. 5.5: Effect of stirrer speed on volume fraction of solids (left) and the median crystal size (right) both measured in product removal line.

5.3 Estimation of parameters on the basis of startup responses

The process model described in Chapter 2 is a parametrized model that includes models for the process actuators given by the separator model equations: Eqn. 2.9 and 2.14, as well as the CSD dynamics given by: Eqn. 2.23, 2.24, 2.26, A.22, 2.29, and 2.30, and the CSD sensor, which is described by Eqn. 2.35. The total set of unknown model parameters in this model is summarized with Eqn. 2.40. For estimation of these parameters experimental startup data from the pilot crystallizer are used. The information content of process startup data is considered to be large because the CSD varies strongly. Besides, plant startup data are often readily available in historical databases, because plants have to be shut down frequently for cleaning or maintenance purposes.

The classification parameters $\theta_C$ are estimated in a first step. In the next step the optimal fines classification functions are incorporated in the population balance model and the remaining set of parameters $\{\theta_1, \theta_K\}$ are estimated. First the parameter estimation procedure is outlined and the experimental set-up is described.

5.3.1 Parameter estimation

The kinetic parameters are estimated from raw light scattering data, directly. The following general criterion is used:

$$J(\theta) = \sum_{j=1}^{k} \sum_{i=1}^{m} \frac{[L_i(j) - \hat{L}_i(\theta, j)]^2}{\sigma_i(j)},$$

with $m$ the number of detector rings and $k$ the total number of samples. The term $\hat{L}(\theta, i, j)$ is the light energy at ring $i$ and time instant $j$, predicted by the model, as
a function of the parameter vector $\theta$. $L_i(j)$ is an average energy vector calculated from a large ($> 100$) number of detector readings (sweeps). The variance of the fluctuations around the average, $\sigma_i$, $i = 1, \ldots, 31$, is used as a weighting factor for the quadratic errors. The general quadratic criterion uses the full covariance matrix of the sweeps to weight the errors, however, only the diagonal elements of this matrix were available from the data acquisition system that is used. Inspection of the weighted errors revealed that all errors contribute to the total error with the same order of magnitude. Both measured and predicted light energy vectors used in the criterion are normalized for reasons discussed in Section 4.2.

Parameter estimation is applied to find the minimizing argument $\hat{\theta}$ for this criterion. The nonlinear gradient-based optimization module E04UCF provided in the NAG library can be used to solve this problem.

A rough estimate of the uncertainty of the estimated parameters is obtained by calculating the standard deviation of the parameters obtained for the different data sets. More advanced methods like Monte Carlo studies or methods based on residual evaluation [11, 201] to assess parameter confidence intervals are not applied here because the model residual is not a Gaussian white noise and because Monte Carlo studies ask for an unacceptable computation time.

### 5.3.2 Experimental set-up

A series of experiments is conducted with the pilot crystallizer described in Chapter 3. CSD measurements are performed using the Malvern particle sizer at sample locations 1. and 2. (see Fig. 3.1), where unclassified product and classified fine crystals are measured, respectively. The scattered light is recorded at 31 detector elements by two Malvern 2600 Particle Sizers™ with two different collimating lenses. A 300mm lens is mounted to observe the fine crystals at sample location 2. and a 1000 mm lens to observe the crystals at sample location 1 (see Fig. 3.1).

The sampling frequency is kept at a rate of one sample per 2 minutes and the duration of each experiment was 40 hours. For dilution the dilution unit as described in [117] was used. Outliers caused by blockage of the measurement system are removed by an automatic peak-shave procedure.

During the experiments product is removed unclassified, that is, $h_p = 1$ and $Q_p = Q_{pf}$. The responses are measured for three different constant values of the retention time of product, for three fines removal rates, and for three values for the total heat input. Other constant controlled process parameters are summarized in Appendix D. In total 7 different startup experiments were carried out. For the first experiment referred to as RUN6 the constant input parameters, kept unaltered during the experiment, are tabulated in Table 5.3. This first experiment is succeeded by six other experiments where only one input variable at the time is varied compared to RUN6.
5.3 Estimation of parameters on the basis of startup responses

<table>
<thead>
<tr>
<th>Exp. No.</th>
<th>variable</th>
<th>value</th>
</tr>
</thead>
<tbody>
<tr>
<td>RUN6</td>
<td>fines flow</td>
<td>1.0 litre/s</td>
</tr>
<tr>
<td></td>
<td>total heat input</td>
<td>120 KW</td>
</tr>
<tr>
<td></td>
<td>product flow</td>
<td>0.22 litre/s</td>
</tr>
<tr>
<td>RUN7</td>
<td>fines flow</td>
<td>2.2 litre/s</td>
</tr>
<tr>
<td>RUN14</td>
<td>fines flow</td>
<td>3.4 litre/s</td>
</tr>
<tr>
<td>RUN13</td>
<td>product flow</td>
<td>0.15 litre/s</td>
</tr>
<tr>
<td>RUN12</td>
<td>product flow</td>
<td>0.39 litre/s</td>
</tr>
<tr>
<td>RUN9</td>
<td>total heat input</td>
<td>90 KW</td>
</tr>
<tr>
<td>RUN11</td>
<td>total heat input</td>
<td>150 KW</td>
</tr>
</tbody>
</table>

5.3.3 Experimental data startup experiments

Fig. 5.6 shows the dynamic responses of the median crystal size $X_{50}$ and the quartile ratio $qr$ on the initial CSD of the crystallizer for three different fines removal flows: 1.0, 2.2 and 3.4 litres/s, (RUN6, RUN7 and RUN14) respectively. The initial CSD results from primary nucleation and possible agglomeration occurring in a supersaturated liquid during the startup phase, as shown in Section 5.2.1. In these plots also the so-called mother liquor retention time given by: $r_f = V/Q_f$, is printed. In Fig. 5.7 the mass production rate in kg/s, corresponding to the startup with 1.0 l/s fines removal is given. As expected, this response closely resembles a first order like response, with a time constant of 75 minutes, that is determined by the mean residence time of product defined by Eqn. 2.1. In Fig. 5.8 also the obscuration in the fines loop is given. These figures reveal that fines removal promotes cycling and even induces a limit cycle in the CSD. The results for RUN14 show that the quartile ratio peaks at the time where the median crystal size collapses, at the same time the obscuration in the fines loop reaches a maximum value as well. This indicates the existence of a temporal bi-modal shaped distribution in which both large crystals, and an abundance of newly born small crystals are present.

Fig. 5.9 shows the dynamic responses of $X_{50}$ and $q_r$ for three different product removal flows: 0.39, 0.22, and 0.15 litres/s, (RUN12, RUN6 and RUN13), respectively. It can be seen that the time period of oscillations is approximately proportional to the retention time of product $r_p$. This can be explained by the fact that increasing the product flow will mainly decrease the total crystal surface area available for growth, and therefore accelerates the kinetic growth rate of the crystals, which increases the speed of response subsequently.
**Fig. 5.6:** Experimental time response of median crystal size (left) and quartile ratio (right), at startup with a fines flow of 1.0, 2.2 and 3.4 l/s (RUN6, RUN7 and RUN14), respectively.

**Fig. 5.7:** Experimental response of mass production rate corresponding to RUN6.

**Fig. 5.8:** Experimental time response of obscuration measured in fines overflow loop (RUN14).
5.3 Estimation of parameters on the basis of startup responses

Fig. 5.9: Experimental time response of median crystal size (left) and quartile ratio (right), at startup with a product removal flow of 0.39, 0.215 and .15 l/s (RUN12 RUN6 and RUN13) respectively.

These results also indicate that the larger oscillation time periods observed in industry (e.g. 20 hours) may be directly attributed to the larger mean residence times (as defined with Eqn. 2.1) at which industrial crystallizers are normally operated (e.g., 2 to 3 hours). As described in Section 2.2.3, this weakens the statement of Jager [120], who ascribes the observed differences in the cycle time periods to the local dissolution of fines which is due to the existence of supersaturation profiles.

Fig. 5.10 shows the influence of three different values for the total heat input: 90, 120 and 150 kW. (RUN9, RUN6 and RUN11) respectively. The measured trend of RUN9 is disturbed, approximately 18 hours after startup, by a rinsewater flow. These plots show that the influence of a varying heat input on the startup CSD dynamics is insignificant. Variations in the heat input will, however, change the crystal mass production rate at stationary conditions, proportionally.

During RUN6, RUN7 and RUN14, fines distributions where measured at sample location 2. The relative volume distribution of fines corresponding to a specific fines flow varied only slightly in time. Therefore average fines distributions were calculated for the three different fines flows. Fig. 5.11 demonstrates how the cut-size of the fines classifier shifts to larger values for larger fines flows. In Fig. 5.12 and 5.13 the raw light energy distributions as observed at sample location 1, during RUN6 and RUN14 are depicted respectively. For the experiments RUN6, ..., RUN11 these raw energy data recorded directly from the Malvern sensor are used to estimate the model parameters $\theta$. 
Fig. 5.10: Experimental time response of median crystal size (left) and quartile ratio (right), at startup with a total heat-input of 90, 120 and 150 KW, (RUN9, RUN6 and RUN11) respectively.

Fig. 5.11: Experimental relative volume based CSD of fines for fines removal flows of 1.0, 2.2 and 3.4 l/s, (RUN6, RUN7 and RUN14), respectively.
5.3 Estimation of parameters on the basis of startup responses

![Graph](image)

**Fig. 5.12:** Experimental time response of the scaled light energy measured after startup over 30 hours on the first 20 Malvern detector rings, for a fines removal flow of 1.0 l/s (RUN6).

![Graph](image)

**Fig. 5.13:** Experimental time response of the scaled light energy measured after startup over 30 hours on the first 20 Malvern detector rings, for a fines removal flow of 3.4 l/s (RUN14).
5.3.4 Estimation of the fines classification function

The fines classification function was estimated from data obtained at both sample locations 1 and 2. As delays and dynamics are assumed to be negligible, the estimation of this function is a static nonlinear function estimation problem, where the nonlinear function $h$ has to be estimated from raw light energy data $L$. We solved this problem by formulating it as a prediction problem. First we estimate the product size distribution by inverting the model Eqn. 2.35, after which the light energy observed on the detector located at sample location 2 is predicted by the following equation:

$$
\hat{L}_f = H_f \gamma_f \text{diag}(\hat{h}(\theta_C)) \hat{n}_p,
$$

where $\hat{n}_p$ represents the estimated product size distribution obtained by inverting the model Eqn. 2.35. $H_f$ is the sensor model that is calculated for the fine particles. $\text{diag}(\hat{h}(\theta_C))$ is the discretized diagonal removal matrix parametrized with the fines separation parameters $\theta_C = \{p_{f_1}, p_{f_2}\}$. Because the different void fractions did not alter significantly, the flow split factor was approximated by $\gamma_f = 1.12$. The average aspect ratio $\alpha$ was estimated to be 1.4. Now the quadratic criterion Eqn. 5.1 and a numerical optimization routine are used to minimize the difference between the predicted and measured values for $L_f$ as a function of the parameters $\theta_C$.

This scheme is used to estimate the parameter values $p_{f_1}$ and $p_{f_2}$ of the fines classification function given by Eqn. 2.9 and 2.11, based on data sets obtained during three different runs: RUN6, RUN7, and RUN14. The estimated parameter values are listed in Table 5.4, with their average value and the corresponding standard deviation.
Table 5.4: Estimated fines classification parameters.

<table>
<thead>
<tr>
<th>Exp. No.</th>
<th>( \hat{p}_{f2} )</th>
<th>( \hat{p}_{f1} [10^{-5}] )</th>
<th>( x_c [\mu m] )</th>
</tr>
</thead>
<tbody>
<tr>
<td>RUN6</td>
<td>4.68</td>
<td>0.232</td>
<td>48.2</td>
</tr>
<tr>
<td>RUN7</td>
<td>4.17</td>
<td>0.252</td>
<td>74.5</td>
</tr>
<tr>
<td>RUN14</td>
<td>4.21</td>
<td>0.223</td>
<td>87.1</td>
</tr>
<tr>
<td>average</td>
<td>4.35</td>
<td>0.236</td>
<td></td>
</tr>
<tr>
<td>st. dev.</td>
<td>0.23</td>
<td>0.012</td>
<td></td>
</tr>
</tbody>
</table>

(st. dev.). In the last column of Table 5.4 the estimated cut sizes are given. They were calculated from Eqn. 2.11. The corresponding estimated removal efficiency curves are plotted in Fig. 5.14. The estimated fines removal efficiency curves show the expected shift of the removal efficiency curve to the range of larger crystal sizes for larger fines removal flows.

5.3.5 Estimation of the kinetic parameters and the initial state

Prior to the estimation of parameters, a parameter sensitivity analysis is made by perturbing parameters from the set \( \theta_K \), and by inspection of the resulting perturbation of the criterion (Eqn. 5.1). The parameters \( p_2, p_6, p_7 \) and \( p_{10} \) were denoted as insensitive parameters and were given the following fixed values: \( p_2 = 0.0, p_6 = 10^{-8}, p_7 = 1.0 \). The largest crystal size \( p_{10} \), considered in the model is chosen 1850 \( \mu m \). As a rule of thumb the latter value can be chosen equal to the crystal size where the absolute value of the population density becomes smaller than one. An important result of this preliminary study is that the influence of supersaturation on the secondary nucleation rate (Eqn. 2.23) is small. The remaining set of parameters to be estimated is given by: \( \{p_1, p_3, p_4, p_5, p_8, p_9, p_{11}, p_{12}, p_{13}\} \).

Before estimating empirical parameters from data it is essential to scale the model equations properly. For example without scaling, the sensitivity of the nucleation rate Eqn. 2.23 to changes in the parameter \( p_1 \) may be \( 10 \cdot 10^6 \) larger than the sensitivity to changes in \( p_2 \). Consequently, \( p_1 \) will be well-identifiable and \( p_2 \) not.

To achieve a parameter sensitivity that is in the same order of magnitude, Eqn. 2.23 is scaled by the stationary solution of the population balance according to

\[
B(t) = p_5 \left( \frac{\int_{p_1}^{\infty} n(x,t)x^{p_6}dx}{\int_{p_4}^{\infty} n_s(x,t)x^{p_5}dx} \right)^{p_1} \left( \frac{\Delta C(t)}{\Delta C_s(t)} \right)^{p_2}, \quad (5.3)
\]

where \( n_s \) and \( \Delta C_s \) denote the stationary population density and the stationary super-
The value for $p^*_3$ is obtained easily from $p_3$ using:

$$p^*_3 = p_3 \left( \int_{p_4}^{\infty} n_s(x,t)x^{p_5} \, dx \right)^{p_1} (\Delta C_s(t))^{p_2}.$$  \hspace{1cm} (5.4)

An advantage of this specific scaling is that $p^*_3$ now solely affects the steady-state value of the nucleation rate and the remaining parameters in this equation \{p_1, p_2, p_4, p_5\} solely affect the nucleation rate when the process is non steady-state.

At first the parameter estimation results revealed a significant bias between the model and measured values of the large detector rings numbered $i = 18, \ldots, 31$, as depicted in the upper plot of Fig. 5.15. From light scattering theory it is known that the smaller particles scatter relatively strongly on the larger detector rings. We therefore adopt the idea of adding a second distribution, representing slowly growing fines, to the distribution predicted by the model. This added distribution should account for a model deficit in the region of small particles. A simulation study demonstrated that the model mismatch can be compensated with a constant exponential distribution given by:

$$n_2(x) = k \exp(- \frac{Q_f + Q_p}{V G_2} x),$$  \hspace{1cm} (5.5)

with $G_2$ the constant growth rate of the population of slowly growing fines ($G_2 < G_k$), and $Q_f + Q_p$ the removal rate of small particles. The linear factor $k$ is calculated from a simple least squares fit on the error. In Fig. 5.15 it can be seen that the error is indeed strongly reduced and that the added distribution has strongly magnified the estimated CSD in the region of small crystals. The average growth rate of the added crystal population was estimated to be $0.45 \times 10^{-8}$ m/s, which is roughly ten times slower than the growth rate $G_k$ of the modelled crystal population. This result points at a large
Table 5.6: Parameters related to growth and nucleation kinetics.

<table>
<thead>
<tr>
<th>Exp. No.</th>
<th>$p_1$</th>
<th>$p_2 \cdot 10^8$</th>
<th>$p_3 \cdot 10^{-6}$</th>
<th>$p_4$</th>
<th>$p_5 \cdot 10^{-6}$</th>
<th>$p_6$</th>
</tr>
</thead>
<tbody>
<tr>
<td>RUN6</td>
<td>0.76</td>
<td>0.10</td>
<td>674.</td>
<td>2.76</td>
<td>1191.</td>
<td>5.97</td>
</tr>
<tr>
<td>RUN7</td>
<td>1.15</td>
<td>1.23</td>
<td>519.</td>
<td>2.06</td>
<td>1060.</td>
<td>6.92</td>
</tr>
<tr>
<td>RUN14</td>
<td>2.00</td>
<td>0.61</td>
<td>1002.</td>
<td>2.01</td>
<td>983.</td>
<td>6.00</td>
</tr>
<tr>
<td>RUN13</td>
<td>0.85</td>
<td>0.08</td>
<td>765.</td>
<td>3.18</td>
<td>868.</td>
<td>11.55</td>
</tr>
<tr>
<td>RUN12</td>
<td>0.55</td>
<td>0.09</td>
<td>666.</td>
<td>1.34</td>
<td>1210.</td>
<td>10.66</td>
</tr>
<tr>
<td>RUN9</td>
<td>1.49</td>
<td>0.07</td>
<td>610.</td>
<td>2.07</td>
<td>924.</td>
<td>8.10</td>
</tr>
<tr>
<td>RUN11</td>
<td>0.71</td>
<td>0.20</td>
<td>621.</td>
<td>1.64</td>
<td>805.</td>
<td>12.13</td>
</tr>
<tr>
<td>average</td>
<td>1.07</td>
<td>0.34</td>
<td>694.</td>
<td>2.15</td>
<td>1006.</td>
<td>8.76</td>
</tr>
<tr>
<td>st. dev.</td>
<td>0.48</td>
<td>0.40</td>
<td>144.</td>
<td>0.58</td>
<td>144.</td>
<td>2.45</td>
</tr>
</tbody>
</table>

Fig. 5.15: Measured, not corrected and corrected light energy distributions (upper) and the corresponding uncorrected and corrected simulated ln(\(n\))-x plots (lower).
population of small crystals with a very slow growth rate. Other data supporting this
are presented by Ó Meadhra [177].

Deriving quantitative information on growth rate dispersion from light scattering
data should, however, be done with care for two reasons: firstly, the recorded light
energy distributions are corrected for the background signals that are measured. As
described in [24] these corrections do not accurately account for the presence of particles
in the optical cell. Secondly, as discussed in Section 2.2.6, light will travel also through
the particles because the relative refractive index of ammonium sulphate is close to
unity. Results presented in [100, 27] indicate that due to this effect more energy on
the larger rings is present, than predicted by Fraunhofer model. Therefore, also this
phenomenon may (partially) explain the deviation we observed here.

Consequently, if more information is wanted on growth rate dispersion, more accu-
rate sensor models and probably more sensitive CSD sensors that focus specifically on
the region of small crystals, are needed. In the current approach the model describes
effective phenomena such as the effective secondary nucleation rate and a possible
model deficit in the region of small crystals is only corrected to prevent the estimated
effective parameters from getting biased.

In Table 5.5 the three parameter values for the initial distributions estimated for the
different data sets, with their average value and the corresponding standard deviation
(st. dev.) are given. In Table 5.6 the estimated values for the nucleation and growth
rate parameters are listed with their average and standard deviation. Corresponding
model fits are depicted in Fig. 5.16 for three different detector rings \( j = 5, 10, 20 \),
for RUN6 representing the best fit, and RUN14 representing the worst fit. All other
fits are comparable to the result obtained for RUN6. Also, the change in oscillation
time period for a different mean slurry residence time was predicted accurately by
the model. The larger model mismatch and biased parameter values corresponding
to the data from RUN14 are probably caused by aggregation or primary nucleation
during the strong nucleation burst that occurred when the median reaches a peak
value (Fig. 5.6). These phenomena are not included in the model, as this would result
in overparameterization.

5.3.6 Discussion on the results of parameter estimation

From Fig. 5.16 it is concluded that the model gives a reasonable fit of the CSD dy-
namics. The nucleation parameters provide the best fit if the nucleation rate is taken
proportional to the total surface area of particles with sizes larger than approximately
600 \( \mu \text{m} \). Such a dependence on the total surface area instead of the mass of the
CSD points at surface breeding. The low sensitivity of parameter \( p_2 \) (which was kept
constant), however, shows that the influence of supersaturation on the observed CSD
dynamics is low, which is not expected in case of surface breeding. The values found
for \( p_1 \) and \( p_3 \) are in correspondence with values provided in the literature [83] Some
parameter values, especially \( p_3 \), are uncertain. This can be caused by differences in
Fig. 5.16: Responses of signals from Malvern detector rings [5,10,20] simulated with model (dashed) and measured from process (solid) for RUN6 (left) and RUN14 (right).

Experimental conditions, like an alternating unknown impurity concentration, or a limited sensitivity of the sensor for the phenomena to be estimated. Better results are expected with a sensor that is more sensitive to large and small particles. Besides, as mentioned in the introduction to this chapter, for control purposes the model will be sufficiently accurate as long as closed-loop stability and performance are guaranteed in the face of the estimated parameter uncertainties. Results on robustness analysis are reported in Chapter 9 and Chapter 10).

In Fig. 5.17 the ln(n)-x plots corresponding to the model fit to RUN6 at three different time points $t = 3, 6, 40$ hours, are given. This plot illustrates how badly dampened or oscillatory CSD dynamics can be explained from a simple secondary nucleation mechanism in which only large particles play a significant role: particle waves come up in the region of small crystals and grow into the region of large crystals $x > 600 \, \mu m$ in a time period of approximately 3 hours, then a new wave of small particles or new nuclei is initiated from attrition fragments bred on the surface of larger crystals. At the moment these new nuclei are formed, the total surface area of the crystals is relatively low and the supersaturation will rise because of a lack of consumption of ammonium sulphate molecules. This will increase the growth rate providing a large portion of the newly formed fines with a chance to survive the fines trap. In the initial stage of their outgrowth the total surface area will increase and the growth rate will decrease again, which strongly reduces the chance that small
Fig. 5.17: Simulated \( \ln(n)-x \) plot corresponding to RUN6 at three times.

Particles will survive the fines trap. Notice that the survival change of a small particle is determined by the residence time of a particle in the fines removal size region. This residence time depends on both the removal rate of fines and the growth rate according to:

\[
r_f \propto \exp{-x_f/G\tau_f}
\]

(5.6)

Hence if we take the cutsize \( x_f = 100 \) \( \mu m \), and the mean residence time of fines \( \tau_f = 1000 \) s., then a decrease of the growth rate with 10% causes a decrease of the survival change with 20%. If \( \tau_f = 500 \) s. the same growth rate decrease, decreases the removal change with 37%.

This example explains why cycling increases when fines removal is increased. In Fig. 5.18 a schematic illustration (after [197]) of the main interactions present in a crystal population are summarized.

5.4 Evaluation of the model for product classification

From the previous section estimates have been obtained for the kinetic model parameters. In this section product classification is applied, first to explore the general benefits of this mode of operation, and secondly to use the experimental results to validate the predictive quality of the model at a significant different operation condition.

With product classification large product particles are removed selectively from the crystallizer volume, to obtain a coarser product. An additional physical loop is introduced, to recycle small particles and liquid that passed the vibrating screen...
classifier, to the main crystallizer volume. Visual inspection revealed that the slurry leaving the screen deck is a viscous cake which does not disperse. The solids fraction in this cake, which is measured by hand, was fairly constant in time at 55%.

5.4.1 Experimental results

A series of two experiments has been done using the vibrating screen classifier as a product classification device. An outline of the experimental setup is given in Chapter 3 and Fig. 3.3. The crystals leaving the deck are now considered as product crystals. Some equipment details are provided in Section 3.1.4. The experiments are referred to as RUN45 and RUN54. During both experiments the crystallizer is started without product classification in closed-loop with the SISO PI controller that is described in Section 9.1.1. After reaching sufficient stability of the CSD, the controller is switched off and the product classifier is initiated. Particle size analysis is done with the Malvern at sample locations 1 and 2. Also, the on-line particle counter has been used to monitor the fines density independent from the Malvern measurements. The maze width of the screen apertures was 800 μm for both experiments.

The experimental results are shown in Fig. 5.19 to 5.22. At A in the plots, the flow $Q_{pf}$ to the screen is switched on, at B, C, and D step changes in this feed flow are made, as described in the lower strip in these plots.

During the first experiment RUN45, the process is operated with a relative large value for the feed flow ($Q_{pf}$). At the end of RUN45 ($t=45.6$ hrs) the classifier is switched off, and operation is returned to normal operation without product classification. During RUN45 the process is operated with a fines flow of 0.40 l/s just before operation with the screen. After the start of classification the fines flow is kept constant at 0.75 l/s. Note that the difference between the median crystal size before and
Fig. 5.19: Experimental response of median crystal in the vessel (upper), magma density in the vessel, the product mass flow, and the feed flow to the screen (lower), for RUN45: At A the screen is switched on and at D the screen is switched off.
after classification, may be due to this. The mass flow leaving the screen denoted as $Q_M$ was measured off-line, simply by drawing samples manually from the product outlet over a certain period of time. Samples are only available after two hours after startup until D. The strong fluctuations in the signal trend between B and D are due to some inaccuracies in taking the sampling. In Fig. 5.20 it is further shown that the fines density, measured with the independent particle counter, drops significantly when using the screen. Obviously, this is a highly desirable effect.

In the second experiment, RUN54 the process is operated at lower values for $Q_{pf}$. During operation with the screen the fines flow is kept constant at 1.4 l/s. As shown by the results, a lower feed flow $Q_{pf}$, in conjunction with a larger fines flow, induces a limit cycle in both the CSD as well as the mass production and the magma density in the crystallizer. In the upper strip of Fig. 5.21 also the median size of crystals in the return line is measured. The difference with the median measured in the feed-line is small as the removed large particles represent a small relative volume. Also the trend of the fines density signal $y_{r1}$ depicted in Fig. 5.22, points at a decrease of the fines density during operation with a product classifier.

From these results it is concluded that product classification introduces additional interaction between the CSD dynamics and the magma density, which was absent with non-classified product removal (see e.g., Fig. 5.7). Increasing the feed flow, gives a response that dampens after approximately 3 cycles. Besides, both experiments show that step wise changes in the feed flow to the screen seem to have a limited effect on the CSD. However, proper judgement is difficult in this case, due to the strong cycling behaviour, which may dominate the step responses. Finally, it is found that, similar to the case without product classification, the time period of oscillations is roughly proportional to the inverse of the flow $Q_{pf}$.

### 5.4.2 Verification of the model with experimental data

The experimental results are compared with simulation results to judge the accuracy of the nonlinear process model. In Fig. 5.23 the simulation results of the model are
Fig. 5.21: Experimental response of median crystal in the vessel and the median in the recycle loop (upper); The magma density in the vessel; The produced mass flow and the feed flow to the screen (lower) corresponding to RUN54.
5.5 Evaluation of the model for Batch operation

Fig. 5.22: Response of fines density $y_{r1}$ corresponding to RUN54.

compared to the experimental results. To obtain a reasonable fit on the CSD dynamics, only the constant $p_3$ in the nucleation law (Eqn. 2.23) is lowered in comparison to the parameters of the non-classified product removal runs. As can be seen the model gives a reasonable description, however some significant errors are present. The oscillation time periods and the median crystal size are predicted reasonably, however the simulated magma density is much larger than the measured trend. Further we found that the simulation results are very sensitive to changes in the classification parameters present in Eqn. 2.14. As described in [86] it is difficult to obtain accurate values for these parameters.

The model/plant mismatch may be due to the spatial segregation effects in the crystallizer vessel, entrance classification, which varies when the removal flow is varied, or errors in the classification function.

In Chapter 8 the merits of product classification are further explored.

5.5 Evaluation of the model for Batch operation

5.5.1 Experimental results

So far the model has been evaluated for continuous process operation. A small series of batch experiments has been conducted as well, where the outgrowth of an initial distribution is measured at a constant heat input. The outcome of these experiments will be compared to the predictions of the model. The initial distribution is the result of an unseeded startup that is described in Section 5.2.1.

With the first experiment the process is operated with a fines flow of 1 l/s, and a constant heat input of 120 KW. In a second experiment the process is operated with a fines flow of 2.2 l/s. During the experiments a recycle flow of 200 ml/s is maintained to feed the CSD measurement system. A negligible small part of this flow is wasted for measurement purposes. A small feed-flow is maintained to compensate the volume reduction due to evaporation of water. CSD measurements are taken at a rate of one sample per two minutes.
Fig. 5.23: Comparison of experimental (solid) and model (dashed) responses of median, magma density in crystallizer, and product mass flow $Q_M$ for RUN45 (left) and RUN54 (right).

Fig. 5.24: Measured solids fraction in the crystallizer during batch operation.
5.5 Evaluation of the model for Batch operation

![Graphs showing evolution of energy and relative volume distributions over time](image.png)

**Fig. 5.25:** Evolution light energy vector observed from the Malvern particle sizer (left), and the reconstructed relative volume distribution (right) for a complete batch run of 3 hours. The direction of change in time is indicated with an arrow.

In Fig. 5.24 the startup response of the solids-fraction in the crystallizer is given over the first three hours after startup corresponding to the first experiment. As expected the solids fraction increases linearly with time.

Fig. 5.25 shows the evolution in time of the measured light energy on the 31 detector rings with the relative volume distribution reconstructed from these measurements. In Fig. 5.26 also the response of the median crystal size and the quartile ratio are given for two different fines flow. These observations indicate that a larger fines flow causes a large final median crystal.

### 5.5.2 Verification of the model with experimental data

The model described in Chapter 2 is fitted to the light energy data according to the method described in Section 5.3.5. As initial distribution a Rosin-Rammler distribution is used (cf. Eqn. 2.27). Fig. 5.27 shows the evolution of the relative volume distribution, and the absolute population density. A good fit of the model to the data was obtained easily, merely by varying the parameters related to the initial distribution (see e.g Eqn. 2.27). The fits described in Fig. 5.27 reveal that there is even no contribution of secondary nucleation during outgrowth of the initial CSD. Also the trend of the solids fraction was described accurately.

These results confirm that ammonium sulphate is a growth dominant system. For batch operation this implies that secondary nucleation may even be discarded from the model. Note however, that with larger values for the evaporation rate a contribution of secondary nucleation may be expected. Further fines removal affects the CSD only at the initial stage of the batch. When the nuclei have passed the fines trap no effect is obtained.
**Fig. 5.26:** Effect of fines removal on transient behaviour batch crystallizer, described by the median crystal size (left) and the quartile ratio (right), for 1 l/s (solid) and 2.2 l/s (dashed) fines removal.

**Fig. 5.27:** Relative volume distribution (upper) and absolute number distribution (lower) of optimized model for batch crystallizer; the arrow indicates the evolution in time of the distributions.
Therefore, non-growth dominant substance systems are expected to pose a larger challenge for control. Examples of these are described, among others, in [175, 231, 156, 157, 252]. In Section 10.4 a short overview is given of model based control concept for batch crystallizers.

5.6 Conclusions

In this chapter the dynamic process model presented in Chapter 2 has been evaluated using experimental data from the UNIAK pilot crystallizer.

Some preliminary experiments point at the occurrence of agglomeration during unseeded startup of the crystallizer. Further it is found that significant spatial segregation occurs, which is most visible in the measurements when the impeller speed is varied.

From a series of unseeded startup experiments it is found that, in case no product classification is applied, CSD dynamics can be influenced with fines and product removal. Varying the heat input merely effects the mass production rate. Increasing fines removal rates promote CSD cycling. Varying the mean slurry residence time gives a proportional change in the time period of oscillations.

Nonlinear parameter estimation is found to be a flexible method for obtaining estimates for the unknown model parameters. A quadratic criterion is used in which errors are calculated directly from raw sensor data and weighted with the estimated variance. This weighting is advantageous since it effectively penalizes less reliable data, subject to large random variations.

It is shown that the model reasonably describes the dynamics of the crystallizer for the set of startup experiments. The fairly simple secondary nucleation model, containing physically interpretable parameter values, describes oscillatory or badly dampened CSD dynamics. The estimated secondary nucleation model shows that the effective secondary nucleation rate is proportional to the total surface area of particles, with a size larger than approximately 600 μm. The results further indicate that the growth of larger particles is hampered, which is explained by both attrition and spatial segregation of larger particles and is described accurately by a simple S-shaped length dependent growth curve. The presence of spatial segregation of particles has been proven by varying the impeller speed. If the impeller speed is kept constant spatial segregation mainly affects the steady-state distribution in the large particle size range.

Some evidence is found for the existence of a large population of slowly growing fines, with a growth rate that is approximately 10 times lower than the normal growing crystals.

With product classification additional interaction is introduced between the CSD dynamics and the mass production rate. It is found that the number of fines in the system drops significantly when classification is applied. Evaluation of the model points at a significant plant/model mismatch. Especially the magma density in the crystal-
lizer, predicted by the model, is larger than the measured value. These errors may be due spatial segregation effects, entrance classification or errors in the classification function.

The model is found to predict the outcome of batch experiments accurately. The model response could be fitted to the experimental data, merely by adjusting the parameters related to the initial CSD. This result was expected as ammonium sulphate is a growth dominant system.

In the next chapter the validity of the model is further explored. However in contrast with this chapter, process control will be taken as the intended use of the model. For this specific purpose we will investigate how the model performs when the process is operated close to the conditions it will be operated at when a controller is active.
Chapter 6

Closed-loop system identification

6.1 Introduction

In the previous chapter, results from first principles modelling of continuous crystallizers and the estimation of empirical parameter values on the basis of startup experiments are presented. The experimental data sets were obtained from the free startup responses of the UNIAK pilot crystallizer. Startup responses are advantageous as they are readily available in historical databases, and give excitation of the process dynamics, without requiring additional external test signals. A main disadvantage, however, is that the resulting process response exhibits only the slow process dynamics within a wide (nonlinear) process output range. For feedback control purposes, knowledge on the faster dynamics near the bandwidth (see also Section 10.2.1) of the system is important, as closed-loop stability is strongly determined in this frequency range. Another disadvantage is that the data contain no information on the combined effect of simultaneous process input disturbances, which may be important for multivariable controller design.

Therefore, it is desirable to perform multivariable identification experiments where the control relevant process dynamics are deliberately excited with uncorrelated, frequency rich test signals [144], which are added to the different process inputs simultaneously.

The purpose of this chapter is to describe a procedure for the identification of low order dynamic models for the dynamics of a continuous crystallization process on the basis of experimental process input-output data. Results shown by De Wolf and Van den Hof [50] expose the applicability of this approach. The intended use of the resulting models is model predictive control (cf. Chapter 9). As both input-output models and first principles models have specific advantages, we will apply both and discuss their respective merits. In particular, a route will be outlined where a priori, —first principles— model knowledge is applied to obtain an initial estimate for the identification of input-output models.
We will show that identification data obtained from open-loop excitation of the pilot plant reveal that open-loop identification of continuous crystallization processes leads to poor results as the crystal size distribution (CSD) has a tendency to oscillate. Improved experimental conditions are achieved, using a simple single loop feedback controller. Identification of low order models, on the basis of closed-loop data, is studied using the nonlinear first principles (FP) model and linear multivariable input-output models. Further two closed-loop identification methods are applied, one of which is recently introduced to provide accurate approximate models [238]. Closed-loop identification and validation data are obtained from the evaporative pilot crystallizer. Both identified and physical models are validated in terms of time and frequency domain responses.

6.2 Open-loop system identification

The (open-loop) process, which will be considered for identification, has three inputs and three outputs. A schematic overview is given by Fig. 6.1. As inputs the fines flow $Q_f$, the product flow $Q_p$ and the total heat input $P_t$ are taken. As outputs, the number density of fines $y_{r-1}$, the mean crystal size $X_{43}$ and the magma density $M_t$ are chosen.

At first, the process dynamics are considered by applying excitation signals directly onto the process inputs, in an open-loop fashion. As test signals multi-sine signals where chosen for the fines removal rate $Q_f$, and the product removal rate $Q_p$, each containing 29 logarithmically spaced frequency components, with phase angles chosen according to [218]. For every frequency in the first signal the second signal has the first harmonic next to it to obtain uncorrelated signals. In Fig. 6.2 the designed input signals, are depicted.

A 45 hours lasting experiment, which will be referred to as RUN35, is performed with the pilot crystallizer. In Fig. 6.3, the first three signal modes are given, estimated according to Eqn. 4.6. These signals are found to represent already 97.1% of the total energy present in the 31 Malvern ring signals.

Visual inspection of the data directly reveals that the process output response is strongly dominated by slow cycling behaviour of the CSD with a time period of
Fig. 6.2: Sinusoidal excitation signals used for open-loop identification (RUN35) after t>6 hrs, for fines flow $Q_f$ (upper) and product flow $Q_p$.

Fig. 6.3: First major three orthogonal signal components, obtained with open-loop identification (RUN35); $y_{r1}$ (upper), $y_{r2}$ (middle), $y_{r3}$ (lower).
approximately 6 hours. As shown in Chapter 5 this behaviour is typical for open-loop operated crystallizers. In the presented data this behaviour is strongly visible, due to the non-stationary initial state of the crystallizer. The main problem with the cycling behaviour of the open-loop process output is that its amplitude dominates all effects on the output of input excitation in a realistic (linear) range. This means that the process actually is driven through an area where it is not possible anymore to model its behaviour accurately by a linear time-invariant model. This has been the main reason for designing the stabilizing controller in the following section. Identification of the process in closed-loop with this stabilizing controller is then expected to yield a more accurate model for the control relevant dynamics of the process.

6.3 Design of a stabilizing controller

Exercises with the nonlinear model revealed that stability of the CSD can easily be obtained by applying a single input single output (SISO) PI-control loop, which uses the weighted number of fines $y_{r1}$ as its input, and the fines removal rate $Q_f$ as its output. The resulting closed-loop control configuration is depicted in Fig. 6.4. Prior to the first closed-loop identification experiment, the ability of the controller to stabilize the startup response of the crystallizer, and to follow step wise setpoint changes has been evaluated experimentally. Fig. 6.5 shows the responses of the controlled variable $y_{r1}$, and the manipulated variable $Q_f$, on the initial state after process startup and on two setpoint changes at A and B to the controller setpoint.

Clearly, the system is stabilized within approximately 5 hours after startup. Around 17 hours after startup some unwanted disturbances have occurred in the process, which caused a gradually, decrease of the manipulated variable as a function of time. The controlled variable, however, remains constant. The results indicate that this simple closed-loop configuration effectively dampens open-loop cycling of the CSD. The closed-loop process considered for identification further, is summarized in Table 6.1
Fig. 6.5: Closed-loop responses to setpoint changes (dash-dotted) in $y_{r1s}$ (at A and B): $y_r$ (upper) and $Q_f$ (lower).

Table 6.1: Process inputs and outputs.

<table>
<thead>
<tr>
<th>randomly disturbed inputs</th>
<th>process outputs</th>
</tr>
</thead>
<tbody>
<tr>
<td>controller setpoint $(y_{r1s})$</td>
<td>number of fines $(y_r)$</td>
</tr>
<tr>
<td>product flow $(Q_p)$</td>
<td>mean crystal size $(X_{43})$</td>
</tr>
<tr>
<td>heat input $(P_i)$</td>
<td>magma density $(M_i)$</td>
</tr>
</tbody>
</table>

(cf. Fig. 6.4). In addition, the controller output $Q_f$ is measured to estimate the open-loop process transfer-functions, following a method that is described in the next section. The signal $y_{r1s}$ is the setpoint for the stabilizing controller, and $v_i(t)$ is a noise contribution.

6.4 Identification method

The system identification procedure includes the selection of an appropriate model set and a criterion of fit. On the basis of informative process input-output data, the best model within this set is then searched that minimizes the chosen criterion.
6.4.1 Model structures

Following Ljung [144] the system to be identified is denoted as

\[ y(t) = G_0(q)u(t) + v(t) \]

(6.1)

with \( u(t) \) and \( y(t) \) the process input and output, \( G_0 \) a linear time-invariant system represented by its pulse transfer function in the shift operator \( q \), and \( v(t) \) a stationary stochastic process represented by \( v(t) = H_0(q)e_0(t) \). Here \( H_0 \) is the stable and stably invertible noise model, and \( e_0 \) is a white noise process. A corresponding model of this process is represented by the collection of transfer functions \( (G(q, \theta), H(q, \theta)) \), parametrized by some unknown parameter \( \theta \). The corresponding one step ahead prediction error is given by:

\[ \varepsilon(t, \theta) = H(q, \theta)^{-1}[y(t) - G(q, \theta)u(t)]. \]

(6.2)

with \( \{(G(q, \theta), H(q, \theta)), \ \theta \in \Theta\} \) and the parameter set \( \Theta \subset \mathbb{R}^q \). The parametrization of the set of models considered can be done in several ways, among which the popular ARX parametrization represented by \( G(q, \theta) = A(q, \theta)^{-1}B(q, \theta) \) and \( H(q, \theta) = A(q, \theta)^{-1} \) with \( A \) and \( B \) polynomials in the shift operator \( q^{-1} \), leading to the linear regression form:

\[ \varepsilon(t, \theta) = A(q, \theta)y(t) - B(q, \theta)u(t). \]

(6.3)

An alternative way of parametrizing the models is the Output Error (OE) form in which the noise model \( H(q) = I \) is not parametrized. The input/output model can be represented by a quotient of two polynomials as above, but can for example also be parametrized in a FIR (finite impulse response) form:

\[ G(q, \theta) = \sum_{k=1}^{n_b} g_k q^{-k} \]

(6.4)

where the coefficients \( \{g_k\}_{k=1,...,n_b} \) are collected in a parameter vector, or in terms of the state-space model:

\[ x(t + 1) = A(\theta)x(t) + B(\theta)u(t) \]

(6.5)

\[ y(t) = C(\theta)x(t) + D(\theta)u(t). \]

(6.6)

In the latter form the transfer function \( G(z) \) is given by \( D + C(zI - A)^{-1}B \). To obtain an unique representation of the transfer function within the set of models, the state-space model set is often parametrized in an observable canonical (or pseudo-canonical) form, (cf. e.g., Gevers [87]).

Both ARX and OE-FIR models have the advantage that the resulting prediction error is linear in the unknown parameters, leading to a linear regression identification
6.4 Identification method

problem. For a least squares identification criterion,

\[ \hat{\theta}_N = \arg \min_{\theta \in \Theta} \frac{1}{N} \sum_{t=1}^{N} \varepsilon^T(t, \theta) \varepsilon(t, \theta) \]  

(6.7)

this implies that the estimated parameter can be analytically determined, avoiding numerical nonlinear optimization procedures.

Output error model structures in either FIR or state-space form have the advantage that the input/output part \( G(q) \) of the model can be consistently identified even if the noise model part \( H(q) \) is miss-specified, [144].

However for FIR models the number of parameters to be estimated is generally large, especially when the underlying system dynamics are moderately damped, and when the sampling rate is high in relation to the dominant process dynamics. Consequently, the variance of the estimated model parameters will be large as well. A state-space model parametrization does not suffer from this problem, however, the output error to be minimized is a nonlinear function of the model parameters and the resulting parameter estimate can therefore only be obtained from iterative numerical optimization procedures. Especially for high order multivariable processes and large data sets the computational burden can be severe, besides possible convergence problems may occur.

As an alternative, recently renewed interest has risen for the use of more generalized (orthogonal) basis functions in a FIR-type model structure:

\[ G(z) = \sum_{k=1}^{n_b} b_k f_k(z) \]  

(6.8)

where \( \{f_k(z)\}_{k=1,\ldots,\infty} \) reflect orthonormal basis functions as, for example, Laguerre functions (cf. e.g., Wahlberg, [251], and Finn et al., [73]) but also more generalized functions generated by freely chosen all-pass transfer functions (Heuberger et al., [104]). In this latter approach a prior knowledge of the process dynamics can be used to flexibly choose the basis functions so as to increase the speed of convergence of the series expansion (Eqn. 6.8). In this way the number of parameters to be estimated can be kept small, while retaining a high accuracy of the identified model [237].

The model structure in Eqn. 6.8 will be denoted as ORTFIR. In this chapter the choice of appropriate basis functions will be based on both the linearized and reduced first principles model and estimated ARX models. For all estimated models the nonlinear OE approach in state-space form was applied in a final step, using the ORTFIR estimated model as initial estimate.

6.4.2 Closed-loop MIMO identification

A main and well known problem related to the identification of a process in a closed-loop configuration is that the noise on the outputs is correlated with the process inputs
due to the presence of a feedback loop. This correlation generally results in biased estimates for the model, where the bias distribution depends on the characteristics of the noise. Two possible approaches to this problem are employed in this chapter:

- The direct identification (DI) method. This method fully ignores correlation between inputs and outputs and identifies directly on the basis of closed-loop process input and output data [230].

- The two step (TS) method as proposed by Van den Hof and Schrama [238]. With this approach the usual problems of direct closed-loop identification are circumvented, by subdividing the model identification step into two successive open-loop identification steps.

Application of the direct approach is straightforward. The TS approach is shortly explained. We refer to [238] for further reading.

The two step approach

In a closed-loop setting, we assume the process input to be determined according to:

$$u(t) = r(t) - C(q)y(t) \quad (6.9)$$

with $r(t)$ an external reference signal, uncorrelated to the noise disturbance $v(t)$, and $C(q)$ the linear controller. Using the input sensitivity function

$$S_0(q) = [I + C(q)G_0(q)]^{-1} \quad (6.10)$$

and Eqn. 6.1, Eqn. 6.9 is rewritten to:

$$u(t) = S_0(q)r(t) - C(q)S_0(q)v(t). \quad (6.11)$$

In the first step, using measurement data of $r(t)$ and $u(t)$ one identifies the transfer function $S_0$. This is an open-loop type of problem as $r$ and $v$ are uncorrelated. Next the estimate $S(q, \hat{\beta})$ is used to reconstruct a noise-free input signal:

$$\hat{u}_r(t) = S(q, \hat{\beta})r(t) \quad (6.12)$$

that is used in the second step of the procedure when applying a model structure:

$$\epsilon(t, \theta) = y(t) - G(q, \theta)\hat{u}_r(t). \quad (6.13)$$

It can be shown that, provided the first step in the procedure is performed sufficiently accurate, this method can provide a consistent estimate of the plant dynamics in the second step.

Application to a MIMO process is reported in Van der Klauw et al., [243].
6.5 Design of a closed-loop experiment

6.5.1 Selection of test signals

Experiment design is mainly concerned with proper selection of the test signal characteristics used for identification. Clearly, a trade-off exists between production and safety losses, and the amount of information that can be gained from an identification experiment. Test signals are required that give a sufficient excitation of the process to be identified, however for safety and economical reasons, the test signals should obey the strict requirements that exist in process industry for the maximum duration of the experiment, the test signal energy at the critical plant frequencies, and on the maximum signal amplitude.

As an alternative to multi-sine excitation (as applied in Fig. 6.2), random- and pseudo-random test signals can be applied, which switch (pseudo-) randomly between two fixed signal levels at discrete points in time. Maximum length binary noise (BN) sequences are often used for this purpose [10]. These signals can be generated easily with a shift register [43]. Tulleken [234] proposes the use of a stochastic distribution that determines the switching probability. This allows for manipulation of the frequency spectrum of the input signal. The corresponding signals are denoted as generalized binary noise signals (GBN). Choosing a switching probability between 0.5 < p < 1 will provide more excitation in the low frequency range, conversely, choosing 0 < p < 0.5 will increase the signal energy in the high frequency range.

In comparison to multi-sine wave excitation, conventional BN and GBN signals are much more accepted in processes industry. They have the advantage that the energy is roughly evenly distributed over the frequency range of interest, hence the maximum signal amplitude is lower. In addition, plant operators are most familiar with step wise or pulse wise signals.

Two closed-loop experiments for identification and successive model validation have been performed with the pilot crystallizer. We refer to these experiments as RUN55 and RUN56. All three inputs of the closed-loop process, e.g: \( y_{t,1s} \), \( Q_p \), and \( P_t \) (cf. Fig. 6.4) have been excited with the GBN signals described above. The real nominal signal values and their amplitudes are given in Table 6.2. From simulation tests it was found that the signal \( Q_f \) saturates easily. This problem is reduced by limiting the amplitude of the excitation signal. Moreover, this further legitimates the use BN signals instead of sinusoidal excitation, which were used for the open-loop experiments.

The basic intervals for the GBN signals are chosen 20 minutes. The switching probability was chosen \( p = 0.7 \), which implies more excitation in the low frequency range. The test signals designed for the second experiment (RUN56) and their spectrum are depicted in Fig. 6.6. The signals for RUN55 are approximately uncorrelated with these signals, while their frequency contents are comparable. For both experiments, a duration of 50 hours was chosen. This value roughly equals 8 times the dominant time constant in the system. Evaluation of the method with input-output data generated
Table 6.2: Nominal values and amplitudes of the GBN test signals.

<table>
<thead>
<tr>
<th>process input</th>
<th>nominal value</th>
<th>amplitude</th>
</tr>
</thead>
<tbody>
<tr>
<td>setpoint $y_{r1s}$</td>
<td>0.5</td>
<td>0.25</td>
</tr>
<tr>
<td>setpoint $y_{r1s}$</td>
<td>0.5</td>
<td>0.20</td>
</tr>
<tr>
<td>product flow $Q_p$</td>
<td>215</td>
<td>35</td>
</tr>
<tr>
<td>heat input $P_t$</td>
<td>120</td>
<td>30</td>
</tr>
</tbody>
</table>

![Images of time and frequency domain plots]

Fig. 6.6: Excitation signals applied during the RUN56; time domain (left), frequency domain (right)

with a low order linear model, derived from the simulation model, shows that the original model can be recovered.

6.5.2 Data preprocessing

Proper treatment of the data, prior to the estimation of parameters, is a necessity to obtain consistent identification results [144, 10]. The raw process data consist of measured values for the magma density recorded from the independent density sensor, with a sample time of 10 seconds, and the scattered light energy vectors, which are sampled with a frequency of 1 minute. The raw scattered light intensity vectors are averaged values from batches of 1000 sweeps each, which are recorded from the detector in approximately 5 seconds.
The following data preprocessing steps were applied to the raw data, (cf. [144, p. 386], [10, p. 164]):

1. All scattered light energy patterns were corrected for the signal background (cf. Chapter 4).

2. The recorded light diffraction patterns are normalized by subdividing each element by the total scattered light energy.

3. Signal outliers are removed by linear interpolation between good measurements that neighbour outliers.

4. The process outputs $y_{t-1}$ and $X_{43}$ are estimated from the normalized diffraction patterns, according to the method described above.

5. All output signals are detrended. Disturbances acting on the process may contribute to the process outputs as well. These contributions are often slow varying trends, which are not correlated with the inputs. Because the data set used for identification has a limited length, these trends may significantly affect the identification results. In this work we separated the trend from the signal by removing the best straight line fit from the data.

6. All input and detrended output signals are scaled and their mean values are subtracted. To illustrate the importance of this step, assume that both the mean crystal size in $\mu$m and the magma density in kg/m$^3$ are considered as outputs without scaling or subtraction of the mean. The error criterion used in the identification step will then be dominated by errors in the magma density as this signal has a numerical value which is a factor $10^7$ larger than the values for the mean. To obtain input-output signals with comparable numerical values, the mean values of all signals are subtracted and each signal is subdivided by its estimated standard deviation. The resulting signals are equal in terms of energy.

7. Decimation of the data. The original data are sampled to fast for control. By filtering the data high frequency disturbances, which are not relevant for control, are suppressed. The filtered data are resampled by picking each 1 sample of every 5 samples. Notice that filtering prior to resampling is also a necessity to avoid aliasing.

The set of 3 input and 3 output signals that is obtained after processing the data is described in Table 6.1. Each signal contained 600 data points in both data sets with a sampling interval of 5 minutes.

6.5.3 Model verification

A model verification step is included to judge whether the estimated models are sufficiently accurate. Model verification is done in the time and frequency domain on the
basis of the independent data set (RUN55). A scalar measure, denoted as the relative mean square (RMS) value is evaluated for assessing the quality of fit. This measure is calculated from:

$$\text{RMS}_i = \sqrt{\frac{\sum_{k=1}^{N} (y_i(k) - \hat{y}_i(k)) \times (y_i(k) - \hat{y}_i(k))}{\sum_{k=1}^{N} y_i(k)\hat{y}_i(k)}} \quad (6.14)$$

with \( \hat{y} \) the noise free simulated output and \( y \) the measured output.

In the frequency domain, models can be compared using their Bode amplitude and phase plots. In addition, the Bode plot may be compared to the so called empirical transfer-function estimate (ETF) [144], estimated from the input-output data directly. From a simulation exercise we found that due to the low number (600) of samples in our input-output data set, a large bias in the low frequency range exists. This bias can be reduced to a certain extent by applying a frequency window, however, at the expense of an increased variance in the estimate [144]. For this reason ETFE are not considered further.

### 6.6 Results and discussion

The excitation signals presented in the previous section were applied to identify the dynamics of the open-loop process, on the basis of closed-loop data. The data from RUN56 will be used for identification and of RUN55 for validation. Appendix E summarizes the main software tools used.

#### 6.6.1 Experimental data

The three output signals corresponding to RUN55 and RUN56 are depicted in Fig. 6.8, respectively. The corresponding controller output signals \( Q_f \) are given in Fig. 6.7. Comparable responses were found for the validation run. Note that the control signal in Fig. 6.7 is truncated at the bottom because the lower bound of the actuator range was reached. During the validation experiment (RUN55) a relatively large density of inert gas bubbles were observed in the solution, due to air leakage into the (vacuum) crystallizer. As these bubbles are observed as particles by the sensor, the observed process trends are likely to be corrupted by this non quantified disturbance.

#### 6.6.2 Results direct identification

Three state-space models in canonical observability form, with order 3, 4 and 5, were estimated directly on the basis of the input-output data from RUN56. Also an initial state and an offset were estimated, while the direct coupling matrix \( D \) in Eqn. 6.6 was kept zero, hence in total 24, 31, and 38 parameters were estimated, respectively, on the basis of 600 input-output samples. The RMS values of the estimated models,
Fig. 6.7: Measured control signal $Q_f$ for RUN55 (left) and RUN56 (right).

Fig. 6.8: Measured output signals used for identification for both RUN55 and RUN56.
Fig. 6.9: Identification results of fourth order model (solid) estimated with direct identification method on data from RUN56 (dashed); fines density $y_{r1}$ (upper), mean crystal size $X_{43}$ (middle) and magma density $M_t$ (lower).

for both the identification (denoted with "idf") and the validation (denoted as "val") experiment, are given in Table 6.3. The consistency of the model is checked by adding the (known) stabilizing feedback controller to the estimated open-loop model and simulate the output response successively on the basis of the three (GBN) test signals. The resulting RMS values (denoted as "cl") are also provided in Table 6.3.

Checking the RMS values reveals that the fourth order model gives the best fit. In Fig. 6.9 and Fig. 6.10 the fit of this model on both the identification and the validation data set are given, respectively. Note that the first portion of the simulated signal trend in Fig. 6.10 is biased as the initial model state was kept zero.

The results show that reasonable fits are obtained. However, in the validation data set some large deviations are present, in particular in between 24 and 32 hours. These deviations may be due to process disturbances, and the non-stationary initial state as well.

6.6.3 Results two step identification
Fig. 6.10: Validation results of fourth order model (solid), estimated with direct identification on data from RUN56 (dashed), with uncorrelated data from RUN55: fines density $y_{r1}$ (upper), mean crystal size $X_{43}$ (middle) and magma density $M_t$ (lower).

Application of the first step of the two step identification procedure resulted in a 6th order state-space model in canonical observability form, for the sensitivity model Eqn. 6.10. Note that all three inputs of the process (cf. Fig. 6.4) had to be used in this step as all three affect the input $Q_f$. The known value for the proportional action of the controller is used to determine the direct coupling matrix $D$, a priori. Also an initial state is estimated. With the sensitivity model a noise free output signal for the stabilizing controller is simulated. The measured and reconstructed output signal of the controller are given in Fig. 6.11. The RMS value corresponding to this fit is 0.25. This plots elucidates that the difference between the reconstructed and the measured controller output signal are small. This, however, does not necessarily imply that the result of the second step is equal to the result of the direct identification step. Bode plots of the estimated transfer functions are used to explore this difference further.

In the second step, again 3rd, 4th, and 5th order state-space models in canonical observability form were estimated in an open-loop fashion. However, for the controller output $Q_f$, the reconstructed controller output $\hat{u}_c$, which is obtained from the first
Fig. 6.11: Measured (dashed) and reconstructed process output signal $Q_f$ (solid) of stabilizing controller on the basis of 6th order state-space model.

Fig. 6.12: Identification results of fourth order model (solid) estimated in second step of the 2 step identification method on data from RUN55 (dashed), with uncorrelated data from RUN56; fines density $y_{r1}$ (upper), mean crystal size $X_{43}$ (middle) and magma density $M_l$ (lower).
Fig. 6.13: Validation results of fourth order model (solid) estimated with 2 steps identification method on data from RUN55 (dashed), with uncorrelated data from RUN56; fines density \( y_{t-1} \) (upper), mean crystal size \( X_{43} \) (middle) and magma density \( M_t \) (lower).

step, is used. An initial state and an offset were also estimated, while the matrix \( D \) was kept zero.

The RMS values of the residuals of the different models for both data sets are given in Table 6.3. Also for this model the estimated open-loop model is simulated in closed-loop on the basis of the excitation signals. The resulting RMS values are also provided in Table 6.3. Also for this case a fourth order model seems to provide the best estimate of the open-loop process dynamics. In Fig. 6.12 and Fig. 6.13, the fits on the identification and validation sets are given, respectively. At first hand, the RMS values and the time domain fits reveal only small differences between the DI and the TS models. However, if we compare Bode plots of these models, as given in Fig. 6.14, striking differences are observed, mainly in the region of higher frequencies. The deviations of the DI model from the TS model exist mainly due to the existence of a correlation in the noise between the input-output signals used in the DI approach. This result underlines the value of the proposed TS approach.
Fig. 6.14: Amplitude plots of fourth order models obtained from direct approach (solid), the two step approach (dashed), and the first principles model (dotted), with $i \to j$ denoting the transfer relationship of the $i$th input to the $j$th output (cf. Fig. 6.1).
Fig. 6.15: Fit of nonlinear process model (solid) on measured data of RUN56 (dashed); fines density $y_{r:1}$ (upper), mean crystal size $X_{43}$ (middle) and magma density $M_t$ (lower).

6.6.4 Validation of the nonlinear process model

One of the reasons for application of system identification was to obtain data for the evaluation of the nonlinear process model. The nonlinear process model was fitted on the experimental data. The results showed that the model error was not sensitive for the model parameters $p_1, \ldots, p_{10}$, except the parameter $p_3^5$, which describes the constant nucleation rate (cf. Eqns. e:bnscal and Eqn. 5.4). This parameter was adapted only slightly to improve the fit on the data. The fit of the optimized nonlinear model on the identification data are given in Fig. 6.15. The fits of the validation data were similar.

As can be concluded, the fit on the output $y_{r:1}$ is better than the fit on output $X_{43}$. The latter exhibits a less regular response on the input excitation than the real process trend. Probably a dispersion effect that smoothenes the particle waves when transported (by growth) over the crystal size domain is responsible for this. The dynamics of the magma density is described with approximately the same accuracy as the input-output model. On the basis of the identification data, the first principles model does not perform significantly worse or better than the identified model. However, on the
Table 6.3: RMS values for \(y_{r1}\) (RMS1), \(X_{43}\) (RMS2) and \(M_t\) (RMS3), of directly estimated models (DI), the two-step approach (TS) and the linearized first principles model (FP), on the basis of identification data (idf), identification data with the model in closed-loop (cl), and validation data (val).

<table>
<thead>
<tr>
<th></th>
<th>RMS1</th>
<th></th>
<th>RMS2</th>
<th></th>
<th>RMS3</th>
<th></th>
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</thead>
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<tr>
<td></td>
<td>DI</td>
<td>TS</td>
<td>FP</td>
<td>DI</td>
<td>TS</td>
<td>FP</td>
</tr>
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<td>third order model</td>
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</tr>
<tr>
<td>idf</td>
<td>0.52</td>
<td>0.54</td>
<td>1.08</td>
<td>0.59</td>
<td>0.56</td>
<td>1.00</td>
</tr>
<tr>
<td>cl</td>
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<td>0.55</td>
<td>1.22</td>
<td>0.78</td>
<td>0.92</td>
<td>1.11</td>
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<tr>
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<td>1.10</td>
<td>1.11</td>
<td>1.29</td>
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<tr>
<td>idf</td>
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<td>0.51</td>
<td>0.52</td>
<td>0.92</td>
</tr>
<tr>
<td>cl</td>
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<tr>
<td>val</td>
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<td>1.14</td>
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</tr>
<tr>
<td>cl</td>
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<td>1.22</td>
</tr>
<tr>
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<td>1.21</td>
<td>1.26</td>
<td>1.59</td>
<td>1.40</td>
</tr>
<tr>
<td>nonlinear model</td>
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<td></td>
</tr>
<tr>
<td>idf</td>
<td>0.65</td>
<td></td>
<td>1.17</td>
<td></td>
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</tr>
<tr>
<td>val</td>
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<td></td>
<td>1.08</td>
<td></td>
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<td>0.58</td>
</tr>
</tbody>
</table>

The nonlinear model is linearized and reduced successively according to the methods described in Section 2.3. Reduced linear models with order 3, 4 and 5, are thus obtained. The RMS values of the output errors of these models are provided in Table 6.3.

In Fig. 6.14 also the Bode plot of the fourth order model, obtained by linearizing and reducing the nonlinear model, is added as a dotted line. As can be concluded, the linearized first principles model also gives a reasonable fit on the data. The dynamics are well comparable, however, the static gain differs from the identified models. It may be expected that the physical model gives a better description of the low order dynamics in the system than the identified model. For control, however, a proper estimate of the static gain is less important as most controllers will have integral
action which tailors the process outputs to a zero static deviation.

As the first principles model also contains significantly less unknown parameters, less informative process data are needed, hence the method may suit the industrial requirements better. Whether it is better to use an empirical or a low-order linearized first principles model for an industrial plant should, however, be determined by other factors as well; for instance the intended use of the model, the cost of modelling, the availability of sensors, and the cost involved with elaborate identification experiments are important.

6.7 Conclusions

In this chapter we have shown that the first principles model provides a reasonable description to the process dynamic behaviour when the process is excited with frequency rich test signals, which are added to the process inputs simultaneously. In addition, it is found that low order linear MIMO input-output models, which are relevant for control, can be estimated from process input-output data directly.

To obtain input-output data for both model verification and identification purposes, we found that the process should be operated in a closed-loop to prevent a badly dampened cycling behaviour to dominate the process output response. With a simple SISO stabilizing feedback controller effective suppression of this cycling behaviour is achieved.

The presence of a feedback loop introduces a correlation between the process inputs and outputs, which posed a well known problem for the application of system identification. Direct identification of input-output models, on the basis of closed-loop process input-output data, shows that a significant bias in the high frequency range of the transfer-functions is introduced. This consistency problem could be avoided effectively with the two step identification procedure.

Validation of the estimated models with an independent data set, shows that the nonlinear first principles model gives the best fit. The difference with the low order linear input-output models is, however, small.
Chapter 7

Design and experimental evaluation of a state estimator

The development of a state estimator for a crystallization process, equipped with a sensor for the crystal size distribution (CSD), is discussed. The resulting state estimator will be evaluated with raw data from the pilot crystallizer. Sufficient robustness is demonstrated for sensor failure, initial state errors and process disturbances.

7.1 Introduction

A basic requirement for the implementation of a control system is the availability of accurate and reliable information on the relevant process dynamics. For on-line measurement of both the CSD and the supersaturation in crystallizers, different sensors have been developed which, however, still have severe shortcomings. On-line measurement of supersaturation is usually extremely difficult, as the relative level of supersaturation is often not more than 1 percent of the saturation concentration. A characteristic of most CSD sensors is that they are indirect, that is reconstruction scheme's are required to obtain an estimate of the CSD based on the raw output of the sensor. Solutions, often applied to the CSD estimation problem are direct inversion algorithms which estimate the CSD based on the estimated inverse of the sensor model. This inversion procedure is, however, ill conditioned and as a consequence sensitive to measurement noise.

Because the deconvolution does not take into account the correlation in time present in the data it can be referred to as a static estimator. An extension of the static estimator is a dynamic estimator that takes into account the evolution in time of the parameter to be estimated. A dynamic estimator can be constructed by using Kalman filtering techniques [128]. Basically, such an estimator is a dynamic model, which runs on a computer and operates sequentially on the raw measured process input and output data. The standard Kalman filter employs a linear state-space model, and is
also referred to as an optimal state estimator. The optimal estimator has the important property that provided a perfect linear model exists and certain conditions with respect to the process and the measurement noise are fulfilled, then the estimated process state has the lowest possible variance. Thus generally, when the dynamic estimator is well designed, the variance of the parameter to be estimated will be lower as compared to the static estimator approach.

The purpose of this chapter is to present a dynamic state estimator for a crystallization process, based on a first principles process model. This estimator should estimate the CSD and the supersaturation level for control purposes. Secondly, the output of the estimator should serve as a process monitor to facilitate the operator task in making decisions with respect to manual control actions.

Most dynamic estimator applications reported in the literature deal with space systems or navigation systems. The number of applications in the chemical process industry is steadily growing, notwithstanding the severe modelling limitations, and the limitations of on-line measurement systems. Some successful implementations in polymerization are reported among others by Dimitratos [53, 52], for distillation columns among others by Joseph et al., [126] and Lang [138], and in monitoring for hazardous conditions by Gilles and Schuler [88]. Applications of state estimation techniques in the field of crystallization are reported by Cooper and Clough [34, 35], Hashemi and Epstein [98], Myerson et al., [170] and Tsuruoka and Randolph [233]. Most of these applications, however, lack robust on-line process sensors and are often based on too simple (MSMPR) process models.

The 970 litre pilot crystallizer equipped with a robust CSD sensor was used to generate process data. The estimator is based on a lumped version of the nonlinear for the CSD dynamics (cf. Section 2.3.4). A detailed description of the process model and the estimation of unknown model parameters are given in Chapter 2 and Chapter 5, respectively. Step response analysis, based on a linearized version of the model around its steady-state, indicates that the crystallizer can be considered as a sufficiently linear process. For this reason we applied linear Kalman filter theory to estimate a constant Kalman gain matrix. The resulting filter is customized and evaluated on experimental data obtained from the pilot crystallizer. Several robustness features, like sensitivity of the estimator for sensor failure, and process disturbances, are outlined. The approach we follow is applicable to other crystallization processes, equipped with other sensors, provided that sufficiently accurate models for their behaviour are available.

### 7.2 Process model

For design of the estimator the model presented in the previous chapters is linearized according to the method described in Section 2.3.7 to achieve the high order continuous time state-space model given by Eqn. 2.66. Lumping of the continuous population density \( n \) onto 99 discrete equidistant crystal sizes, in the range of \([0\, 1500]\ \mu m\), allows
for a sufficiently accurate approximation of the dynamics to be described. Thus the linear state-space model, that is used for the filter design, has 100 states, e.g. 99 states for the discretized CSD n, and 1 for the supersaturation $\Delta C$.

### 7.3 Process observability

The state-space model $\{F, G, H\}$ enables the estimation of observability properties. An important tool to study observability properties of a process is the observability Gramian $Q$, which is defined as [158]:

$$Q = \int_0^\infty e^{Ft}H^THe^{F^*t}dt.$$  \hspace{1cm} (7.1)

If $Q$ is singular an unobservable initial condition will exist that will not contribute to the output signal energy. In addition, $Q$ will provide information about well or weakly observable states.

### 7.4 Kalman filter design

With Kalman filtering theory a state estimator can be devised, which takes into account the stochastic nature of process and measurement disturbances in a systematic way. The basic idea of a Kalman filter is depicted in Fig. 7.1. The error $\epsilon$ (also denoted as the innovation process) between the actual process output $y$ and the model output $\hat{y}$ is calculated and amplified with the so called Kalman gain $K_f$. This results in a correction signal which is used as an additional input to stably force the model in a direction such that the estimated state converges to the real process state.

For linear time-invariant state-space models the solution for the optimal filter gain is given by the solution of an algebraic Ricatti equation [4]. It is assumed in Kalman filter theory that the model, described by Eqn. 2.66, is disturbed with uncorrelated random process and sensor noise inputs:

$$\dot{z}(t) = Fz(t) + Gu + w(t),$$

$$y(t) = Hz(t) + v(t).$$ \hspace{1cm} (7.2)

The corresponding covariance matrices for the sensor noise $v(t)$ and the process noise $w(t)$ are given by:

$$E\{v^Tw\} = V,$$ \hspace{1cm} (7.3)

$$E\{w^Tw\} = W.$$ \hspace{1cm} (7.4)

Based on this model the filter gain matrix $K_f$ is given by:

$$K_f = P_fH^TV^{-1},$$ \hspace{1cm} (7.5)
Fig. 7.1: Schematic drawing of a continuous Kalman filter.

where $P_f$ satisfies an algebraic Riccati equation

$$
P_f F^T + FP_f - P_f H^T V^{-1} H P_f + W = 0. \tag{7.6}
$$

This solution indicates that an accurate process and sensor model, and good estimates for the covariance matrices for the different noise sources, play a decisive role in the filter design procedure. This can be seen as a disadvantage as it is often difficult or even impossible to obtain a sufficiently accurate model representation of the process and the corresponding noise sources. However, in many practical situations, it suffices to treat the noise covariance matrices as free design parameters that are adapted to achieve a satisfactory filter response based on an imperfect model. A designed filter can be denoted as optimal when the error $\epsilon$ is white with a zero mean.

7.5 Results and discussion

Experimental data

The results of two start-up experiments (described in Chapter 5), measured on the product output (sample location 1 in Fig. 3.1) of the pilot plant, at different process conditions, were used to customize and evaluate the designed state estimator. Figs. 5.12 and 5.13 show the raw light intensity patterns measured on the inner 20 detector rings
of the Malvern instrument, over 30 hours for a fines removal rate of 1.0 l/s and 3.4 l/s respectively. The measured signal trends have been corrected for outliers by linear interpolation. In Fig. 5.6, the corresponding experimental time response is given for the median crystal size $X_{50}$, which was obtained from volume based distributions calculated by direct inversion (cf. Eqn. 4.19). In this plot all outliers caused by tube blockage or other process disturbances are present.

As can be seen large fines flows induce limit cycles. The validity of the assumption that a stationary linear model may be used for the design may be questioned. In the first instance, a more suitable approach for this case is offered by extended Kalman filtering [4], which is a straightforward extension of the linear stationary Kalman filter. An extended Kalman filter solves a set of recursive filter equations at discrete time instances along a state trajectory that is followed by the process. The model matrices used in this approach can be obtained by repeatedly linearization of the nonlinear model at discrete time instants along the followed state trajectory. The resulting Kalman feedback gain will be time-varying, however, for small deviations from the stationary state the filter gain will approach the stationary filter gain.

The extended Kalman filter approach was tested on process data presented below, however, it did not lead to a significant improvement of the solution, it merely resulted in an excessive computational burden. Based on this, and the experimental results presented below, it is concluded that the use of a stationary feedback gain derived from the solution of a stationary Ricatti equation gives satisfactory and applicable results.

**Observability analysis.**

Inspection of the singular values of the observability Gramians, depicted in Fig. 7.2, reveal that only a limited number of modes ($< 6$) are well observable. The process should therefore be denoted as weakly observable. As a consequence of this, most of the 31 diffracted light energy values, which are outputted by the CSD sensor, will be correlated as they bear information on this limited set of process modes only.

An analysis of the correlation in the sensor output vector $y$ is made on the basis of the following singular value decomposition of the sensor model: $H = U_s \Sigma_s V_s^T$. The matrix $\Sigma_s$ contains the 31 singular values of $H$ on its diagonal. The matrices $U_s$ and $V_s$ are unitary matrices containing the right and left singular vectors, respectively. The left singular vectors, e.g. the columns of the matrix $U_s$, bear information on the correlation present in the output vector. The singular vectors corresponding to the largest singular values can be interpreted as the linear combinations of the 31 detector signals that bear information on the best observable modes of the CSD. The singular vectors corresponding to the smallest singular values will determine linear correlations that are mostly corrupted by signal noise.

To lower the sensitivity for noise, only the first $n_s$ left singular vectors corresponding to the largest singular values are taken into account. This leads to a reduced output
matrix $H_r$ given by: $U^T_{s_{r}}H_{r} = H$, with $U_{s_{r}}$ the matrix with left singular vectors corresponding to the largest singular values. Good results are obtained when only three modes ($n_{r} = 3$) are taken into account.

**Experimental evaluation of the filter**

The Kalman filter gain is calculated from the solution of the Ricatti Equation 7.6 for the model $\{F, G, H_r, V, W\}$. The matrix $W$ is estimated from noise present in the experimental data. A diagonal matrix is chosen for the process noise covariance matrix. A rough estimate for the diagonal values was obtained by first perturbing the physical parameters $\theta$ of the model randomly, within their estimated confidence bounds, and calculating the corresponding stationary crystal size distributions. From the resulting large set of stationary crystal size distributions, an estimate is obtained for the variance.

The optimal filter gain is calculated from Eqns. 7.5 and 7.6 In Fig. 7.3, the columns of the calculated optimal filter gain $K_f$ are given. For visualization purposes these vectors are scaled by subdividing each column element with the corresponding element of the stationary CSD. Note that the correction signal does not act on the last state element which is the supersaturation $\Delta C$. The calculated Kalman gain is implemented to the nonlinear model as depicted in Fig. 7.1. The final filter was customized by amplification of the process noise covariance matrix with a constant factor in the range
[0.5 to 2.0], while the measurement noise covariance matrix was kept constant. From tuning exercises it was found that a similar effect could be obtained by amplifying the designed filter gain slightly also in the range [0.5 to 2.0].

In Figs. 7.4.a and 7.4.b, the filtered and measured light intensity on the rings \{5,10\} of the detector are depicted for the cases of small and large fines removal flows, respectively. These results indicate that the filtering capabilities of the designed state estimator are good. The sensitivity for outliers is low and a significant noise level reduction is achieved without introduction of a significant bias.

The designed estimator has capabilities to reconstruct additional process information, which is relevant for operators or engineers. In Fig. 7.5.a, the reconstructed trends of the median crystal size $X_{50}$, the nucleation rate $B$, and the supersaturation $\Delta C$, corresponding to plant startup conditions with a low fines removal rate, are given. The median crystal size obtained from the direct inversion approach is added as a dotted line in the first strip of these plots.

In Fig. 7.5.b, the same trends are depicted, however, a large error in the initial state is introduced by selection of the steady-state CSD as the initial CSD. In addition it is assumed that sensor measurements are not available between 6 and 11 hours after startup. In this period the estimator generates an uncorrected open-loop estimate of the process state variables. It appears that the model still follows the signal trends reasonably well, however these unmodelled disturbances will always bias the estimated
7.5 Results and Discussion

Fig. 7.4: Filtered (solid) and measured (dotted) response of ring \{5,10\} after startup
a: (fines flow 1.0 l/s).  b: (fines flow 3.4 l/s).

Fig. 7.5: Reconstructed CSD values for first 14 hours compared to $X_{50}$ from inversion (dashed) for startup (fines flow 1.0 l/s)
a: With accurate nominal model.  b: With large error in initial CSD and sensor failure.
Fig. 7.6: Volume based CSD at 0.5, 2, 4 and 4.5 hrs. after startup (fines flow 1.0 l/s).

(a) Reconstructed with state estimator.
(b) Reconstructed with direct inversion.

In Fig. 7.6.a, the estimator reconstruction of a certain relative volume distribution, at four different time instants corresponding to the time points of the trends depicted in Fig. 7.5.a, are given. In Fig. 7.6.b, the same distributions obtained from direct inversion are depicted. It can be seen that the resolution of the estimated CSD is much larger than the resolution of the CSD obtained by direct inversion. This, however, does not necessarily imply that the accuracy is better. As the applied estimator is nonlinear and many numerical approximations are involved, a good comparison between state estimation and direct inversion is difficult to make.

The results further show that the error between the model and the measured output signals, as can be seen in Figs. 7.4.a and 7.4.b, is not an uncorrelated white noise signal. As no strong process disturbances occurred in the experiments, the reconstruction errors should be addressed to model errors or errors which are inherent to the applied design procedure, e.g. errors due to lumping of the population balance, linearization, or rough estimation of the covariance matrices.

7.6 Conclusions

With the application of a state estimator, dynamic process information stored implicitly in a dynamic model is combined with on-line process information to reconstruct quantities that cannot be measured directly. This approach has strong advantages compared to a direct sensor inversion approach, in which only a static sensor model is used, to interpret the measurements.

Good results are obtained with a nonlinear estimator which was designed from a locally linearized and stationary version of the process model:
• The variance of the reconstructed state may be expected to be lower, compared to the direct inversion technique. This claim can, however, not be proven and should be made with some reservation because, the system is nonlinear, the model is not exact and several numerical approximations are made, e.g. lumping of the population balance, linearization of the model and the assumption of a white process noise for which a diagonal structured noise covariance matrix exists.

• When process data are not available for a limited period of time, due to sensor failure, an open-loop model prediction of the process output signal trend can be used. This will prevent a controller from undesired upsets.

• Physical quantities like the supersaturation, the crystal birth rate and the crystal growth rate, that cannot be measured directly, are estimated to support operators and engineers to gain knowledge on the process physics.

As described, the availability of a good model and covariance matrices for the different noise sources play a decisive role in the filter gain design procedure. This can be seen as a disadvantage, as it is often difficult or even impossible to obtain sufficiently accurate representations of the nominal model and the corresponding noise models. However, when a model is available, it may enhance the quality of reconstructed distributions, compared to the direct inversion approach.

This chapter has indicated how a nonlinear state estimator for a crystallization process that is designed from a linearized model can be applied for reconstruction of the CSD and related quantities from raw plant data. This approach forms a good basis for dynamic estimator applications on industrial crystallization plants.
Chapter 8

Controllability analysis and selection of control structures

In this chapter controllability aspects of a continuous crystallization process are considered. A main point is that the success of plant control mainly depends on inherent limitations in the plant (including sensors), rather than the controller that is used.

8.1 Introduction

Prior to the actual design of a controller it is important to know how well the plant can be controlled and what factors limit the achievable control performance. Furthermore a choice has to be made on what plant inputs and outputs are to be used for control, and how these inputs should be paired. These topics introduce the field of process controllability assessment. Controllability in the sense used here, denotes input-output controllability, which is defined by Skogestad [221] as:

The ability to achieve acceptable control performance, that is, to keep the outputs of a process within specified bounds or displacements from their setpoints, in spite of unknown variations such as disturbances and plant changes, using available inputs and available measurements.

In loose terms a plant is denoted as controllable if there exists a controller — connecting measurements and inputs— that yields acceptable performance for all expected plant variations. Notice that controllability addresses both the steady-state and dynamic plant behaviour. A key notion is that controllability is an inherent plant property, which is determined by the plant design, for example, the internal couplings, recycle loops, the sizing of the equipment, and the location of sensors. We will further explain this below.

In industry the field of controllability assessment (in an early stage of design) has gained an increased interest over the last twenty years [258]. The main incentive for this
is the increased complexity of plants (e.g., the high level of energy integration), which introduces additional uncertainties in the process environment. If no adequate control is designed to lower these uncertainties the merits of for instance energy integration can be lost. To achieve effective control it is therefore important to prevent potential control difficulties, already at the design stage of a plant. In relation to this the insight has grown that even small changes in a plant’s design can give large improvements in its controllability. Some examples illustrating these points are presented in [161, 259].

In the field of crystallization no studies are known which explicitly address the controllability of a crystallization process as a function of its design. Most work elaborates on the assessment of good steady-state performance whereas the question what design achieves the best performance at \textit{both} steady-state and dynamic process conditions, are not provided in literature. The aim of this chapter is to study controllability as a function of specific design choices related to process actuators and nominal process conditions to achieve both a good stationary product CSD, and to suppress effectively steady-state and dynamic process disturbances.

In general the design of a crystallization process involves decisions on several micro and macro scale process conditions, of which the most important are:

1. the effective crystallizer volume;
2. suspension mixing, as a limiting factor for both micro and macro scale transport characteristics;
3. the supersaturation of mother liquor, as the main driving force for crystallization;
4. the effective nucleation (birth) or seeding rate, which provides a boundary condition to the crystal population;
5. and the residence time that is needed to allow the crystals to grow to the desired product size;

The most basic completion of these functionalities is the so-called \textit{Mixed Suspension Mixed Product Removal} (MSMPR) crystallizer, which has been introduced by Randolph [190]. This is a theoretical concept which can only be approximated in reality. Several extensions to this concept and more practical guide lines for crystallizer selection and design are described in the literature. A number of papers [18, 166, 153, 174] presents important heuristics for the selection of a crystallizer type, the process scale, and for nominal process conditions like the pressure in the boiling zone, the slurry residence time and the heat input or the cooling rate. In [197], idealized models are used to study the effect of residence time of fine and large particles on the steady-state performance and on the stability of crystallizers. In [199] the so-called double drawoff principle (DDO) is considered. With this principle an additional flow containing only fines or even no crystals (clear liquor advance) is withdrawn from the crystallizer volume and not returned. In this way an independent process input is created that can be used to influence the magma density in the crystallizer.
This chapter will focus on design and control issues related to nucleation and residence time at steady-state and dynamic conditions. In particular, fines removal will be studied as an actuation mechanism for the number of small particles, with the objective to improve the stationary CSD, and to achieve stability and disturbance rejection. Secondly, product classification, will be studied for its ability to enhance the steady-state product quality and its ability to reject process disturbances. With respect to the other three aforementioned design issues, we assume that a well-mixed constant effective volume is present, in which a supersaturation is maintained by evaporation of solvent.

In the sequel to this chapter first some theoretical concepts and tools for model based controllability analysis will be outlined. Using these tools three different practical designs for a crystallizer will be studied, e.g. a basic MSMPR crystallizer, a crystallizer equipped with a fines removal system and a crystallizer equipped with a fines and a product removal system. For each design we will show how it affects the steady-state performance of the crystallizer and what the inherent limitations are to suppress disturbances at both steady-state and dynamic process conditions. In the last section of this chapter the role of the sensor on the achievable control performance will be addressed.

8.2 Controllability analysis

Regarding terminology, the term controllability is used here in a different sense than Kalman's state controllability concept. State controllability denotes the ability to tailor the system from a given initial state to any final state. This concept only address the performance of the plant at the final time, whereas for a chemical plant the performance over the entire time is important. Rosenbrock [213] introduced the more useful theoretical concept of functional controllability. Functional controllability investigates the conditions (process characteristics) under which an input trajectory exists that generates the desired output trajectory. Perkins et al., further discuss the relation between functional and state controllability [182, 215]. Morari [160] introduced the term dynamic resilience. This term as well as the term input-output controllability are used within the control engineering context to denote functional controllability. Here we follow the argument of Skogestad [221] who proposes to use input-output controllability instead of dynamic resilience for semantic reasons. Finally instead of input-output controllability some authors use the term dynamic operability [6]. This term places controllability in the wider context of process operability [162]. A plant is denoted as operable if it performs satisfactory under conditions different from the nominal design conditions. Operability analysis thus seeks for a level of flexibility in a plant design that is sufficient both to obtain the desired steady-state and dynamic performance in the presence of uncertainties in the process environment.

In the literature many tools have been developed that enable screening of a plant's
controllability on the basis of models. The traditional method for evaluating the controllability properties of a process is to select a control structure; to tune the controller "optimally"; and then to judge the performance of the system under closed-loop control by rigorous simulation. Unfortunately, this will be a costly exercise if there are several process configurations possible. In addition this method does not allow for a separate analysis of the effects of controller parameters and the design parameters. Therefore more effective methods have been developed in the past fifteen years to evaluate a plant's controllability.

To enable separate analysis of the effect of design parameters, Morari [160] makes use of the IMC concept, as introduced by Garcia and Morari [78]. Here some main results, which we describe in Section 10.2.1 are used. As shown in [160] if a perfect model is used in the IMC control structure (see also Fig. 10.2) than the closed-loop system is described by Eqn. 10.6. Hence if we choose for the controller the right inverse of the model, the perfect controller is obtained which gives perfect tracking of the setpoint, that is: $y = r$. This leads to the important notion that the factors that limit the achievable control performance are the factors that limit the invertibility of the plant. Hence the designer of a plant should aim at the invertibility of the plant, as it would give perfect tracking of setpoints. Furthermore this result explains in mathematical terms the key notion that controllability is an inherent plant property, which can be affected only by changing the design.

In practice the invertibility of a plant is limited by two inherent process characteristics: [160]:

**Physical constraints on manipulated variables.** These cause saturation of the manipulated variables especially at high frequencies or large excursions from the nominal behaviour.

**Non minimum phase elements.** A system is denoted as *minimum phase* when no other system with an equal gain exists that has smaller phase. Systems with delays and right half plane zeros are non-minimum phase systems. The inverse of these systems results in a predictive and unstable controller, respectively. As shown in [137] the zeros are invariant for feedback control. Thus unlike poles they can not be shifted by the controller —only by changing the design the location of zeros can be affected. An exhaustive discussion on the role of delays and right half plane zeros on controllability is given by Morari and co-workers [109, 110, 165].

Till here a perfect controller is assumed. In practice however the model that is used to design a controller is uncertain. Therefore the third factor that limits the achievable control performance is the uncertainty of the model that is used for controller design [222]. This factor is however not an inherent limitation of the plant. We further address the role of model uncertainty on the achievable control performance in Chapter 9 and 10.
Notice that the open-loop stability of a plant is not a limiting factor for the achievable control performance. Open-loop unstable plant behaviour is not an issue for design as long as sufficient actuators and sensors exist that can be used to cancel out instabilities by applying feedback compensation.

For SISO systems these factors can be translated easily to the following heuristic design rules: avoid systems with varying parameters, strong nonlinearities and inverse response characteristics, select short time constants and short dead-times between the actuator move and the actual effect on the output quantity, and finally select systems where the manipulated variable has a large effect on the controlled output. The latter rule implies a Bode plot for the open-loop transfer-functions with a large cross-over frequency and a large open-loop gain.

For MIMO systems such heuristic rules cannot be devised, mainly because the concept of gain is less transparent in this case. Methods and tools for analysis are presented by different authors. A comprehensive review of these is given by [161]. Here some main issues will be reviewed.

### 8.2.1 Analysis tools for MIMO systems

The tools for controllability assessment for MIMO processes are mainly frequency domain tools developed for linear models. The tools described here are incorporated in an easy to use Matlab toolbox, which is commercially available [147]. Because linear models are used, which provide a good description only in a neighbourhood of the nominal plant behaviour, a simulation of the lumped nonlinear process model including the controller should conducted as a final check of the controllability of a certain design.

**Singular values and condition number**

A basic tool used to study the controllability of a plant is the singular value decomposition [89]. Using the transfer function model for a multivariable system: \( G \), the singular value decomposition is written as:

\[
G = U \Sigma V^T \tag{8.1}
\]

where \( U \) and \( V \) are unitary matrices (\( U^T = U^{-1} \)) and \( \Sigma \) is a diagonal matrix (when \( G \) is square) containing the singular values \( \{ \sigma_1, \sigma_2, \ldots, \sigma_n > 0 \} \) on its diagonal. If \( G \) is non-singular all singular values are greater than zero. Generally \( \sigma \) is used to denote the largest singular value and \( \sigma \) as the smallest singular value.

For a certain input vector \( v \) it can be shown that:

\[
\sigma \leq \frac{\|Gv\|_2}{\|v\|_2} \leq \bar{\sigma} \tag{8.2}
\]
where $\| \cdot \|_2$ denotes the Euclidean vector norm. So the gain of a multivariable system is located between the largest and the smallest singular value. In particular the largest and smallest singular value are important because of the properties:

$$\max_{v \neq 0} \frac{\| Gv \|_2}{\| v \|_2} = \frac{\| Gv_{\max} \|_2}{\| v_{\max} \|_2} = \sigma(G)$$  \hspace{1cm} (8.3)

$$\min_{v \neq 0} \frac{\| Gv \|_2}{\| v \|_2} = \frac{\| Gv_{\min} \|_2}{\| v_{\min} \|_2} = \sigma(G)$$  \hspace{1cm} (8.4)

Consequently, if the input vector is in the direction of $v_{\max}(G)$, than the largest possible effect on the outputs will result, and in the direction $v_{\min}(G)$ the smallest effect is obtained. Conversely it can be shown that if a disturbance corrupts the output in the direction of $u_{\max}(G)$, the disturbance acts on the plant in the "best" direction, because the least strong action of the manipulated variables is needed to compensate the disturbance. The worse direction of a disturbance is $u_{\min}(G)$ where the largest action of the actuator is needed for compensation. The columns of $U$ and $V$ contain the right and left singular vectors $\{u_j\}$ and $\{v_j\}$ respectively. From Eqn. 8.1 it is found that:

$$Gv_j = \sigma_j(G)u_j$$  \hspace{1cm} (8.5)

Thus $v_{\max}$ and $v_{\min}$ are the columns of $V$ that correspond to the smallest and largest singular value.

For controllability analysis of a dynamic system the singular value decomposition of the process transfer function matrix is evaluated on a finite set of (logarithmically spaced) frequencies within the control relevant frequency range.

A plant with a large difference between the largest and smallest singular values has a large "directionality". The class of processes for which this occurs in the frequency range of interest are in general difficult to control.

As an indicator for directionality the condition number of the plant’s transfer function, denoted as $\gamma(G)$, is often used. The condition number equals the ratio of the largest and the smallest singular value: $\gamma(G) = \sigma(G)/\sigma(G)$. As a general rule a condition number lower than 10 will indicate an easy controllable system, whereas values larger than 25 are indicative for difficulties in assessing the desired closed-loop performance. Notice however that interpretation of the condition number should be done with care because it is a scalings depend measure. So these rules only apply provided that proper scaling is applied (see also Section 8.2.2). Often also the visual inspection of the plot of the principal gains can be used to study the plant’s controllability.

A non-scalings dependent quantity is the minimal condition number [146, 93]. The minimal condition number is lowest possible condition number, which is calculated by minimizing over all possible input and output scalings. For most cases only an approximation to the minimal condition number is available as it is difficult to compute [147].
Relative gain array (RGA)

A well known tool is the relative gain array (RGA) [26], which is used for problems related to control structure design, for example, the pairings of input-output variables in a so called multi-loop decentralized controller. This control structure is most attractive for its simplicity, however, it cannot deal with interaction explicitly. The RGA is widely applied in process industry [220], as an indicator for the interaction present in a certain plant. At the design stage the RGA is used to select plant designs that give a minimum of interaction.

The RGA can be calculated easily. Taking the element \( g_{ij} \) of the transfer function matrix \( G \) that corresponds to input \( j \) and output \( i \), when all loops are open, and \( h_{ij} \) when all outputs except the \( i \)th output are tightly controlled. Then the \((i,j)\) element of the RGA is denoted as:

\[
\gamma_{ij} = g_{ij}/h_{ij}
\]  
(8.6)

In terms of a transfer matrix, the relative gain array is defined by:

\[
\Gamma(s) = G(s) \times (G^{-1})^T
\]  
(8.7)

The symbol \( \times \) denotes the element by element multiplication.

Some important properties are (cf. [220, p. 295] and [146, 224]) that the RGA is invariant for input-output scaling and if \( G(s) \) is triangular or diagonal than \( \Gamma(s) = I \). The existence of large (>10) and negative values of the RGA is indicative for difficulties in controlling the plant. For \( \Gamma(s) \approx I \) no control difficulties are expected.

Effect of disturbances

Skogestad and Morari [223] argue that solely for the purpose of disturbance rejection the presence of directionality may not be a problem as long as the disturbances are in the “strong direction” of the plant. Consequently, the ability of a plant to reject specific disturbances may be further refined by taking into account the direction of the disturbances that act on the system. For this Skogestad and Morari use a disturbance model of the form:

\[
y = Gu + G_dz
\]  
(8.8)

in which \( G \) is the transfer function introduced above and \( G_d \) is the transfer function that relates the disturbance vector \( z \) to the output vector \( y \). Using this model the so called disturbance condition number, denoted as \( \gamma_d \), of the plant is introduced. This condition number is equal to the aforementioned condition number, however, it is corrected for the direction of the disturbances. From the same contribution it can be concluded that the disturbance condition number is a refinement of the condition number because: \( 1 \leq \gamma_d(G) \leq \gamma(G) \). So only if the plant condition number points at a large directionality it may be useful to calculate the disturbance condition number. In addition, the usefulness will also depend on the availability of accurate disturbance models.
Table 8.1: Dependence of indicators on scaling (After [147]).

<table>
<thead>
<tr>
<th>Indicator</th>
<th>depends on scaling ?</th>
</tr>
</thead>
<tbody>
<tr>
<td>RGA</td>
<td>no</td>
</tr>
<tr>
<td>RDG</td>
<td>no</td>
</tr>
<tr>
<td>Singular values</td>
<td>yes</td>
</tr>
<tr>
<td>Condition number</td>
<td>yes</td>
</tr>
<tr>
<td>Disturbance condition number</td>
<td>no</td>
</tr>
<tr>
<td>Minimal condition number</td>
<td>no</td>
</tr>
<tr>
<td>Stability</td>
<td>no</td>
</tr>
<tr>
<td>Right half plane zeros</td>
<td>no</td>
</tr>
</tbody>
</table>

Analogue to this the so called *relative disturbance gain* (RDG) is introduced as an extension to the RGA that accounts for the disturbance direction as well. The advantage of this measure is that it does not depend on scaling. We refer to [227] for details.

Because the disturbances acting on a crystallizer are generally difficult to model and because these are expected to have a limited accuracy, we will rely on the plant's condition number, in the first instance, as an indicator for controllability problems.

### 8.2.2 Scaling

Proper judgement of results from model based controllability analysis can only be achieved when sensible scalings to the model inputs and outputs are applied. In general it is convenient to scale the plant such that the expected variation in each output or input are comparable. For the results presented here the inputs were scaled so that the range -1 to +1 represents the available range for each manipulated variable. Scaling of the outputs is performed such that an equal change in each output variable is "equally bad" for the control of the process. This choice is subjective, but in many cases sufficient. Alternative scalings may be applied [147], however, also these will generally involve subjective judgements.

Table 8.1 (obtained from [147]) summarizes the dependence of the applied analysis method on input-output scaling.

### 8.3 Controllability analysis of continuous crystallizers

Using the crystallizer model described in Chapter 2, the described tools are applied to study the controllability of continuous crystallizers. We will do this for three physically
attainable design configurations, which differ in the actuation mechanism that is used to influenced the CSD at both steady-state and dynamic conditions.

First the process is described. Subsequently we will evaluate the steady-state controllability of the plant. At this stage also the merits of the different actuation mechanisms for the steady-state product quality are illuminated. Thereafter controllability analysis is used to determine for what input-output sets controllable processes are obtained. Appendix E summarizes the main software tools used.

8.3.1 Process description

The following three crystallizer designs are studied in this chapter.

**Design 1** represents a standard MSMPR crystallizer with no fines classification. Further its is assumed that no growth dispersion occurs in the system. The heat input and the product removal rate are used as process inputs.

**Design 2** represents a MSMPR crystallizer equipped with a fines removal and a fines destruction system, as introduced in Section 3. Further it is assumed that crystal growth retardation occurs due to the phenomena described in Section 2.2.3. As process actuators, respectively the fines removal rate $Q_f$, the total heat input defined by: $P_{tot} = P_{in} + P_{ex}$, and the unclassified product removal rate $Q_p$, are considered.

**Design 3** represents a crystallizer equipped with a fines removal and a classified product removal system. Product classification is achieved with a vibrating screen (Fig. 3.3). Growth and nucleation kinetics are the same as in design 2.

The analysis performed here is based on the pilot crystallizer model for which the basic assumptions are summarized in Appendix C. The specific model parameter values (see Chapter 2) used in each design are summarized in Table 8.2. The numerical values are estimated from the data as described in Chapter 5. The other constant operating conditions are described in Appendix D. The range over which the nominal values of the process input parameters are varied for steady-state analysis are provided in Table 8.3.

The steady-state CSD as a function of the process input parameters is estimated according to the method described in Section 2.3.6. For the dynamic controllability analysis, which is performed in the succeeding sections, the process model is linearized to a 100th order continuous time state-space model (see Chapter 2.3.7). Only constant scaling factors are applied to the different input and output parameters. These are summarized in Table 8.4. For nonlinear simulation and calculation of the steady-state properties a 200th order model is used.
### Table 8.2: Nominal parameters and process conditions for three designs.

<table>
<thead>
<tr>
<th>parameters</th>
<th>unit</th>
<th>equation</th>
<th>Design 1</th>
<th>Design 2</th>
<th>Design 3</th>
</tr>
</thead>
<tbody>
<tr>
<td>$p_1$</td>
<td>[-]</td>
<td>Eqn. 2.23</td>
<td>1.1</td>
<td>←</td>
<td>←</td>
</tr>
<tr>
<td>$p_2$</td>
<td>[-]</td>
<td>Eqn. 2.23</td>
<td>1.0</td>
<td>←</td>
<td>←</td>
</tr>
<tr>
<td>$p_3$</td>
<td>[-]</td>
<td>Eqn. 2.23</td>
<td>$3.0 \cdot 10^6$</td>
<td>←</td>
<td>←</td>
</tr>
<tr>
<td>$p_4$</td>
<td>[μm]</td>
<td>Eqn. 2.23</td>
<td>650</td>
<td>←</td>
<td>←</td>
</tr>
<tr>
<td>$p_5$</td>
<td>[-]</td>
<td>Eqn. 2.23</td>
<td>2.0</td>
<td>←</td>
<td>←</td>
</tr>
<tr>
<td>$p_6$</td>
<td>[-]</td>
<td>Eqn. 2.24</td>
<td>1.0</td>
<td>←</td>
<td>←</td>
</tr>
<tr>
<td>$p_7$</td>
<td>[μm]</td>
<td>Eqn. 2.24</td>
<td>1000</td>
<td>←</td>
<td>←</td>
</tr>
<tr>
<td>$p_8$</td>
<td>[-]</td>
<td>Eqn. 2.26</td>
<td>8</td>
<td>←</td>
<td>←</td>
</tr>
<tr>
<td>$p_9$</td>
<td>[μm]</td>
<td>Eqn. 2.26</td>
<td>1750</td>
<td>←</td>
<td>←</td>
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<tr>
<td>$p_{10}$</td>
<td>[-]</td>
<td>Eqn. 2.26</td>
<td>$1.0 \cdot 10^{-8}$</td>
<td>←</td>
<td>←</td>
</tr>
<tr>
<td>$p_{f1}$</td>
<td>[-]</td>
<td>Eqn. 2.11</td>
<td>-</td>
<td>$2.6 \cdot 10^{-6}$</td>
<td>←</td>
</tr>
<tr>
<td>$p_{f2}$</td>
<td>[-]</td>
<td>Eqn. 2.9</td>
<td>-</td>
<td>4.57</td>
<td>←</td>
</tr>
<tr>
<td>$p_{p1}$</td>
<td>[-]</td>
<td>Eqn. 2.14</td>
<td>-</td>
<td>-</td>
<td>6.0</td>
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<tr>
<td>$p_{p2}$</td>
<td>[μm]</td>
<td>Eqn. 2.14</td>
<td>-</td>
<td>-</td>
<td>800</td>
</tr>
<tr>
<td>$p_{p3}$</td>
<td>[%]</td>
<td>Eqn. 2.14</td>
<td>-</td>
<td>-</td>
<td>2.9215e-02</td>
</tr>
<tr>
<td>$Q_p$</td>
<td>[l/s]</td>
<td>Eqn. 2.29</td>
<td>0.215</td>
<td>←</td>
<td>←</td>
</tr>
<tr>
<td>$Q_f$</td>
<td>[l/s]</td>
<td>Eqn. 2.29</td>
<td>-</td>
<td>1.0</td>
<td>←</td>
</tr>
<tr>
<td>$P_{tot}$</td>
<td>[KW]</td>
<td>Eqn. 2.29</td>
<td>120</td>
<td>←</td>
<td>←</td>
</tr>
<tr>
<td>$Q_{pf}$</td>
<td>[l/s]</td>
<td>Eqn. 2.29</td>
<td>-</td>
<td>-</td>
<td>0.75</td>
</tr>
</tbody>
</table>

### Table 8.3: Range of nominal values of process input parameters.

<table>
<thead>
<tr>
<th>Inputs</th>
<th>$Q_f$ [l/s]</th>
<th>$Q_p$ [l/s]</th>
<th>$Q_{pf}$ [l/s]</th>
<th>$P_t$ [KW]</th>
</tr>
</thead>
<tbody>
<tr>
<td>range</td>
<td>0.30 - 3.0</td>
<td>0.10-0.35</td>
<td>0.20-1.4</td>
<td>25-250</td>
</tr>
</tbody>
</table>
Table 8.4: Constant scaling factors for model inputs and outputs used for
dynamic controllability analysis.

<table>
<thead>
<tr>
<th>Inputs</th>
<th>$Q_f$ [l/s]</th>
<th>$Q_P$ [l/s]</th>
<th>$Q_{pf}$ [l/s]</th>
<th>$P_t$ [KW]</th>
</tr>
</thead>
<tbody>
<tr>
<td>scaling</td>
<td>0.60</td>
<td>0.10</td>
<td>0.30</td>
<td>60</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Outputs</th>
<th>$y_{r1}$ [-]</th>
<th>$X_{50}$ $\mu$m</th>
<th>$M_t$ [kg/s]</th>
<th>$1 - \epsilon$ [-]</th>
<th>$\Delta C$</th>
</tr>
</thead>
<tbody>
<tr>
<td>scaling</td>
<td>0.60</td>
<td>80</td>
<td>20.10^{-3}</td>
<td>10</td>
<td>10</td>
</tr>
</tbody>
</table>

### 8.3.2 Process outputs

As process outputs the CSD in the crystallizer vessel and the product discharge line are considered. Note that without the use of product classification these quantities are assumed to be equal. The mass flow of produced solid crystal material, which is calculated from

$$Q_M = Q_P\rho_c\frac{M_t - \rho_t}{\rho_c - \rho_t}$$  \hspace{1cm} (8.9)

$$= Q_P\rho_c k_* m_3 = Q_P\rho_c(1 - \epsilon)$$  \hspace{1cm} (8.10)

is taken as a measure for the yield. Eqn. 8.9 is used to calculate $Q_M$ from the measured magma density $M_t$. In the simulation model $Q_M$ is calculated from the third moment of the distribution, $m_3$. Notice that $m_3$ directly determines the solids fraction in the crystallizer: $k_* m_3 = (1 - \epsilon)$.

Eqn. 8.10 also shows that if the product removal flow is constant, the mass flow $Q_M$ and the solids fraction $(1 - \epsilon)$ will only differ by a constant factor. In practice both parameters are important: the mass flow equals the yield and the solids fraction of the product determines the transportability of the slurry. Above 50% solids the product will be a viscous cake which cannot be transported by pumps.

To judge the stationary crystal size distribution in view of optimal solid-liquid separation we use both the median crystal size and the spread of the distribution. Also supersaturation is taken into account as it affects the purity of crystals, their mechanical properties, and their morphology. The supersaturation is given as a percentage of the saturation concentration $(\Delta C/C_s)$. Generally the supersaturation is not allowed to exceed a certain level.

For (dynamic) input-output controllability analysis the variable $y_{r1}$, which is introduced in Chapter 4, is considered. As shown in Section 4.5.3 this parameter has the largest sensitivity for CSD fluctuations. Besides it is close to the region where the fines removal system as a CSD actuator exerts its largest effect on the CSD. Thus in the face of the heuristics rules for controllability assessment presented on page 156,
this parameter is attractive. However, the question remains how the "location" of the sensor affects the achievable control performance. Therefore this topic is elaborated further in Section 8.6 of this chapter. It will be shown that the selection of a sensor location is non-trivial. First, however the role of actuators on the achievable control performance is outlined.

8.4 Results steady-state analysis

The results of steady-state analysis for the three designs is presented in Fig. 8.1 to Fig. 8.4. In Fig. 8.1 it is shown how different input parameter values affect the log-population density. In Fig. 8.2 relative volume distributions are used to present how the same variations affect the shape of the CSD in the middle size range. In Fig. 8.3, the effect of input parameter variations on the median crystal size ($X_{50}$), the spread denoted as ($X_{75} - X_{25}$), and the supersaturation level in percentage of the saturation concentration are given. Finally, in Fig. 8.4 the effect of feed flow and heat input variations on the stationary product flow, the median crystal size, and the solids fraction $(1 - \epsilon)$ of the product leaving the screen deck, are given. The arrows indicate the direction of the main effect in the stationary distributions. In the plots of Fig. 8.2 we have added the relative volume distribution that exists for design 2 as a solid line.

We proceed with a discussion of these results for the three different designs that are introduced in the previous section.

8.4.1 MSMPR crystallizer (design 1)

The standard MSMPR crystallizer has only two inputs, i.e the total heat input and the mean residence time of slurry determined by the product removal rate (cf. Eqn. 2.1). The stationary solution of the logarithmic population density is always proportional to the crystal size. In the upper left subplots of Fig. 8.1 the stationary log-population density functions, calculated from the nonlinear model, for different values of the heat input and the residence time, are given. Clearly, the two actuators only provide a decrease or increase of the distribution, which is proportional over the whole size domain. Hence only the solids fraction, in the crystallizer (and the product) is influenced and no effect on the shape of the CSD resorts. This is also depicted by the relative volume distributions given in Fig. 8.1. In the latter plots also the relative volume distribution of Design 2 is given. Clearly, the latter design outperforms the standard MSMPR crystallizer, because much larger particles can be produced.

Further it was found that choosing $p_1$ (see Table 8.2) equal to 1, exactly, no effect is found of the heat and residence variations on the stationary CSD. For values different from 1 a small effect is observed, which is insignificant compared to the effect on the mass production rate.

This result indicates that there are adequate manipulated variables to correct for disturbances or load changes that affect the mass flow and/or the fraction of solids in
Fig. 8.1: Effect of process inputs on stationary ln(n) – x plots. The strips in each subplot represents the effect of the fines flow (upper), the product flow (middle) and the heat input (lower). The arrows indicate the direction of change of the actuator (small arrows) and the CSD (large arrows).
Fig. 8.2: Effect of process inputs on steady-state relative volume CSD. The strips in each subplot represents the effect of fines flow (upper) product flow(middle) and heat input (lower). In each plot the corresponding nominal relative volume distribution for Design 2 (solid) is added for comparison. The arrows indicate the direction of change of the actuator (small arrows) and the CSD (large arrows).
Fig. 8.3: Effect of process inputs on stationary properties inside the crystallizer vessel. The strips in each subplot represent the effect on the relative supersaturation ($\Delta C/C_{sat}$) (upper), the median crystal size ($X_{50}$) (middle) and the spread ($X_{75} - X_{25}$) (lower) on design 1 (solid), design 2 (dashed), design 3 (dash-dotted).
Fig. 8.4: Effect of process inputs on stationary properties for design 3. The strips in each subplot represent the effect on the actual product flow (upper) the median crystal size (middle) and the solids fraction (lower). In the middle and the lower plots strips the effect on the product properties in the product is added as a dotted line.

the crystallizer (=fraction of solids in product), however there are no adequate inputs to compensate for changes in the steady-state CSD. Thus the MSMPR crystallizer is steady-state uncontrollable for disturbances that affect the CSD.

8.4.2 Fines removal and unclassified product removal (design 2)

To improve the steady-state CSD and to make the CSD controllable as well, the fines removal and fines destruction systems, as described in Section 3, are added to the process. Notice that this system introduces a recycle loop.

For design of this actuator the cross sectional area of the annulus (Fig. 3.9) and the nominal fines removal flow are considered as the main design parameters to optimize steady-state and dynamic controllability. Eqn. 2.11 and Eqn. 2.9 describe the relation between the cutsize of the classifier and the surface area $A$. The size of the annular zone is 600 litres, which is large in comparison to the crystallizer volume.

In the upper right subplot of Fig. 8.1, the normalized stationary population density functions, for different values of the fines flow, the mean residence time, and the heat input are given. The upper right subplot in Fig. 8.2 depicts the corresponding relative volume distributions. In the upper left subplot in Fig. 8.3 the effect on the
supersaturation, the median crystal size and the spread is described. Except for the heat input, now the actuators have a significant influence on the shape of the crystal size distribution. The strongest effect is a shift of the CSD from the region of small particles to the region of large particles, induced by fines removal flow variations. The effect of the product flow is still limited and mainly caused by the addition of length dependent growth to the model. An important observation from the upper strip of the upper right plot in Fig. 8.1 is that fines removal counteracts itself in the region of small crystals ($x < 30 \mu m$). This mechanism is explained by additional secondary nucleation that is caused by the increase of supersaturation (see also the upper left subplot in Fig. 8.3) and the increase of large mother crystals. In Section 8.6 it is shown that this counteracting effect has large implications for the selection of the sensor that is used for control.

In comparison to the MSMPR case it can be concluded that when using fines removal also a "rotation" of the CSD can be accomplished. Due to this one additional degree of freedom is added to the process, that relates to the slope of the stationary CSD. Fig. 8.3 describes the range over which the median or the spread can be changed. An additional advantage of fines removal is that it solely affects the shape of the CSD without disturbing the mass production rate or the magma density — provided that heat input variations in the external loop are compensated with an internal heat exchanger, as is done in the 970 litre crystallizer.

Notice that with the additional degree of freedom, in principle only one additional process output parameter can be steered to an independent value. For steady-state optimization the choice of output parameter to represent this additional degree of freedom should be located in the range of 200 to 600 $\mu m$ as the largest effect on the CSD is observed in this range. For control other requirements exist. These will be discussed in Section 8.6.

Another point to note is that fines removal will always increase the level of supersaturation in the crystallizer (cf. Fig. 8.3). This may be compensated by changing the residence time, however this will disturb the magma density in the crystallizer for which no independent actuator is present. Adding an independent actuator for the magma density as present in a double drawoff DDO crystallizer [199] may solve this problem.

**Equipment sizing**

A lower cross sectional area will lower the size of annular zone and thereby lower the equipment cost. It is found that for decreasing values of the cross sectional area of the annular zone the solids concentration increases if the removal flow rate is not altered. For values of $A = 1.5, 0.75$ and $0.375 m^2$ the corresponding values for the solids concentration in the fines loop were: $52 \times 10^{-3}, 130 \times 10^{-3}$, and $300 \times 10^{-3} \text{ kg/m}^3$, respectively. Therefore, the minimum value for $A$ should be based on the maximum capacity of the fines dissolving system. A factor that limits the maximum capacity is
the maximum temperature increase of fines.

Increasing the removal flow will also increase the solids fraction in the fines loop will. For values of $Q_f = 0.5$, 1.0, and 2.0 l/s, the corresponding values for the solids concentration were $61 \cdot 10^{-3}$, $130 \cdot 10^{-3}$, and $189 \cdot 10^{-3}$ kg/m$^3$. Hence, also the maximum nominal fines flow should be selected on the basis of the maximum capacity of the fines dissolver as well. Below the influence of these design parameters on the dynamic behaviour is further explored.

### 8.4.3 Fines removal and classified product removal (design 3)

The third design, which includes both a fines removal system and a product classifier is expected to enhance the steady-state CSD further. Notice that this configuration adds a second recycle loop to the flow sheet. In comparison to design 2 the recycle flow $Q_{pf}$ instead of the product flow $Q_p$ is taken as an input parameter.

The crystals leaving the deck of the vibrating screen (Fig. 3.3) are now considered as product crystals. Notice that the size distribution of product crystals is different from the CSD in the crystallizer. The crystals and liquor passing the screen apertures are returned to the crystallizer. The product is a viscous cake which does not disperse. Experimental results show that the solids fraction in this cake is approximately 55%, and seems fairly constant over time.

With the model used here, a constant solids fraction can only be obtained when the parameter $p_{p3}$, which is used in Eqn. 2.14, is adapted (see also Eqn. 2.20). We applied an iterative procedure for this. At steady-state conditions parameter $p_{p3}$ was found to be $2.92 \cdot 10^{-2}$ (cf. Table 8.2). Only for feed flow variations this parameter had to be adapted significantly. For $Q_{pf} = 0.2$ l/s the value for $p_{p3}$ is found to be 0.12. For an increasing feed flow this value thus also increases. Notice that we assume that the void fraction is constant however only limited experimental results are available, to validate this assumption.

In the lower left and lower right subplots in Fig. 8.1 the effects on the population density in the crystallizer vessel and the product flow are shown, respectively. In the lower subplots in Fig. 8.2 the corresponding effects on the relative volume distribution are shown. Additional product and process properties as a function of different input parameter values are shown in Fig. 8.3 and Fig. 8.4.

The effects have the same directions as found for design 2: fines removal causes a decrease in the small particle size range and an increase in the large particle size range; enlarging the classifier feed flow $Q_{pf}$ mainly causes a decrease in the large particle size range; and heat input variations cause a proportional change in all crystal sizes, without affecting the shape of the CSD. The results also show that product classification has a small effect on the CSD in the size range till 700 μm. A small increase of the CSD is observed in the region of small particles when the flow $Q_{pf}$ is increased. Notice further that the steady-state product CSD is not monotonically decreasing any more as in the previous designs.
Another observation is that the median crystal size in the crystallizer vessel is lower than the median found for design 2, whereas the spread of the distribution is drastically lower. For an increased feed flow the median further decreases. Due to the lower number of large mother crystals and the low value for the supersaturation we found that the nucleation rate also decreases, which is highly desirable. This is also confirmed by experimental results as shown in Fig. 5.20 and 5.22. Increasing the feed flow to the screen lowers the median, which is expected as crystals get a smaller chance to grow to larger sizes. The spread of the distribution is significantly lower than for the other two designs. The spread is found to be independent from the feed flow and the heat input. The supersaturation in the crystallizer is fairly low, this value only increases slightly when we increase \( Q_{pf} \). Note that the increase is much smaller than the increase obtained with unclassified product removal. This is mainly due to the fact that the total surface area of crystals that consume supersaturation is located in the size range from 50 to 750 \( \mu m \).

The solids fraction in the crystallizer for the nominal case is found to be 11\% which is acceptable. An important effect is seen in Fig. 8.4 where it is shown that the solids fraction in the crystallizer increases when the feed flow to the screen is decreased and when the heat input is increased. The solids fraction in the crystallizer is an important parameter as for values (>20 \%) slurry transportability problems may occur.

In summary, product classification mainly decreases the spread of the product CSD, whereas the level of supersaturation in the crystallizer decreases, and the density of fines decreases. Both effects are highly desirable.

### 8.4.4 Equipment sizing

It is found that also the aperture size of the screen has a large influence on the solids fraction in the crystallizer. In many cases, where a coarse product is desired, the aperture size of the screen should be chosen as large as possible. However, it is found that increasing \( p_{p1} \) (cf. Eqn. 2.14) with 100 \( \mu m \), roughly increases the solids fraction in the crystallizer with 10\%. Therefore, the upper limit for \( p_{p1} \) should be determined from the maximum allowed solids fraction in the crystallizer.

### 8.5 Selection of control structures

In this section the controllability is further analyzed for the three designs at dynamic process conditions. On the basis of the results the feasible and infeasible control structures will be indicated.

#### 8.5.1 MSMPR crystallizer (design 1)

For the MSMPR crystallizer the heat input and the product removal rate, which determines the mean residence time, are taken as inputs. Using these inputs two
different sets of output parameters are considered.

A CSD parameter and the mass flow as output

First the CSD parameter \( y_{r1} \) and the mass flow \( M_t \) are considered as output parameters. The largest element of the RGA was larger the \( 1.0 \times 10^5 \), which is highly unacceptable, and also the condition number is larger than \( 10 \times 10^5 \) over the whole frequency domain. This confirms the result from the steady-state analysis that the system only has one degree of freedom, which is the mass production rate or alternatively solids fraction, which can be manipulated easily using the heat input, or the product flow, or both. As shown by De Wolf [49] the dynamics of this system can be described in terms of moments and is governed by a single bilinear first order differential equation [144].

Supersaturation and mass flow as outputs

Replacing the CSD related output \( y_{r1} \) by the supersaturation again a two input two output system is obtained. The singular values are shown in Fig. 8.5. The condition number of this system is lower than 1.8, the RGA is close to an identity matrix, and the largest element is 1.001. Hence, this system has negligible interaction and appears to be easily controllable. Using a multi-loop control structure will suffice. Notice that the dynamics of the loop with \( \Delta C \) and \( Q_p \) is determined by parameter \( p_0 \) (cf. Section 5.2.2). A problem is however to measure the supersaturation on-line. The nonlinear state estimator described in Chapter 7 may be used to estimate the supersaturation from on-line light scattering data on the basis of a first principles model.

8.5.2 Fines removal and unclassified product removal (design 2)

For design 2, control structures which use \( Q_f \), \( Q_p \) and \( P_t \) as inputs and different sets of outputs are considered.
A badly controllable three-input-three-output configuration

At first the three input and three output system with the inputs \( \{Q_f, Q_p, P_t\} \) and the outputs \( \{y_{r1}, X_{50}, M_t\} \) is considered. In Fig. 8.7 the singular values are given. The steady-state condition number is 231, the largest element of the RGA is 33, and the minimal condition number is 136. Over the whole frequency range the condition number is larger than 200, except in a narrow frequency band around \( 2 \cdot 10^{-3} \) Hz., where the condition number is 20.

These results indicate that the system has large directionality. Further it is found that the system is non-minimum phase as right half plane zeros exist. Step responses as indicated in Fig. 8.8 show that the median will only show a delayed response in comparison to \( y_{r1} \), for both step changes in the product flow and fines flow (Notice that for presentation purposes the response of the median is inverted). The stationary value of the median will however always be coupled to \( y_{r1} \) with a constant factor. Consequently, the median cannot be controlled independently and should be omitted as output parameter.

An easy controllable three-input-three-output configuration

For this the median crystal size is replace by the supersaturation. Thus a process with \( Q_f, Q_p \) and \( P_t \) as inputs and \( y_{r1} \) and \( \Delta C \) and \( M_t \) as outputs is considered. The singular values for this system are given in Fig. 8.6. The minimal condition number is between 1.09 and 1.82. The steady-state condition number is 1.28 and the RGA is again close to unity with a 1.002 as the largest diagonal element. The condition number in the frequency range of interest is smaller than 10. Therefore this system has limited directionality. And a multi-loop control configuration with the inputs paired directly with the corresponding output is expected to give good closed-loop performance. Control of this process is further explored in Section 10.3.4.
8.5 Selection of control structures

![Graph showing gain versus frequency](image)

Fig. 8.7: Singular values of process with \(\{Q_f, Q_{pf}, P_{tot}\}\) as inputs and \(\{y_{r1}, X_{50}, M_t\}\) as outputs.

**An easy controllable two-input-two-output configuration**

If the product removal flow and the median or supersaturation are removed as parameters, a two input two output system remains. The singular values of this system closely resemble the two dominant singular values in Fig. 8.7. Besides the system is minimum phase. The condition number of this system is lower than 15 over the whole frequency domain, the RGA is close to an identity matrix, and the largest element found at any frequency is 1.033. Also for this system a multi-loop standard PI control structure is recommended, with \(y_{r1}\) coupled to \(Q_f\) and \((1 - c)\) coupled to the heat input. If disturbances affecting the solids fraction are small the control structure may be further reduced to a simple SISO controller, which is used to stabilized the process, that is, suppress the resonant mode that is clearly visible in the singular value plot of Fig. 8.7. This simple structure has been used for stabilizing control in Section 6.3 and will be further evaluated in Chapter 9 and 10.

**Effect of equipment sizing**

An important question related to equipment sizing, is how the dynamics are affected when the cross sectional area of the annular zone is lowered or when the nominal fines flow is increased. In Fig. 8.9, the magnitude plots of a SISO linear model is shown, with fines removal \(Q_f\) as input and \(y_{r1}\) as output, for different values for the cross sectional area \(A\) of the fines classifier, and for different removal flows. From these plots it can be concluded that lowering the cross sectional area of the settling zone slightly increases the bandwidth of the open-loop process, which is desired if a fast reaction on disturbances is required. As indicated in the section on steady-state analysis increasing the fines flow increases the solids fraction. Because the achievable control performance is only slightly affect by this parameter, its minimum should be based on the maximum capacity of the fines dissolving system rather than on the basis of process dynamics. As shown in the lower plot of Fig. 8.9, increasing the removal flow enlarges the bandwidth, besides in the previous section it is shown that the steady-state CSD improves, whereas
the supersaturation increases, and the solids concentration as well. Therefore the best
nominal fines flow should be a compromise between these factors.

**Stability**

The experimental results shown in Section 5.3 reveal that the stability is affected
when increasing the fines flow. With the results presented here it is found that stability
is not an issue as long as the model is operated in a closed-loop. Consequently, the
designer can focus solely on the steady-state and dynamic controllability of the plant
as long as sufficient actuators and sensors are present to design the stabilizing feedback
loop.

### 8.5.3 Fines removal and classified product removal (design 3)

The third case is a crystallizer equipped with a vibrating screen to enable product
classification. As shown above product classification using a vibrating screen strongly
improves the quality of the steady-state product CSD. For control we expect the feed
flow to the screen to be an additional actuator for the CSD dynamics.
8.5 Selection of control structures

Fig. 8.9: Bode plots for different removal flows and surface areas of the fines annulus.

A badly controllable three-input-three-output configuration

To study the controllability the model with $u = \{Q_f, Q_{pf}, P_{tot}\}$ as inputs and $y = y_{r1}, X_{50}, M_t$ as outputs is used. Also for this system a condition number larger than 100 is found which points at a large directionality. The largest element in the steady-state RGA is 33, whereas the largest RGA element at any frequency is found to be 5260. This system thus appears to be hard to control. Step responses calculated with the nonlinear model, as given in Fig. 8.10, show that again the median shows a delayed response in comparison to $y_{r1}$ and cannot be used as an independent input (cf. Fig. 8.8). This figure also confirms what is observed experimentally (cf. Fig. 5.21) that the dynamic response of the magma density in the crystallizer vessel is also oscillating with a time period equal to that of the CSD.

An easy controllable three-input-three-output configuration

If again the output variable $X_{50}$ is replaced by $\Delta C$ an easy controllable system is obtained with acceptable values for the steady-state RGA and condition number (4.8). The condition number in the relevant frequency range is smaller than 20, and the largest element in the RGA at any frequency is 2.2. The system has two unstable eigenvalues and no right half plane zeros. The singular values for this system are depicted in Fig. 8.11.
Fig. 8.10: Simulated step response of design 3 with \{Q_f, Q_{p_f}, P_{tot}\} as inputs and \{y_{r1}, X_{50}, M_t\} as outputs.

Stability

From the experimental results described in Section 5.4.1 it is found that the operating the screen at values where the desired product is expected, e.g. a large cutsize and a low feed flow, enhance cycling of the CSD. With the linearized simulation model the instabilities observed during RUN45 and RUN54 could be predicted reasonably. Also for this case the same argument applies that stability is not an issue as long as a stabilizing loop can be devised that provides sufficient stability to the process. In Section 9.4 we show that a simple stabilizing feedback loop which uses \(Q_f\) as input and \(y_{r1}\) as output effectively stabilizes the process with classified product removal.

8.6 The role of sensors on the achievable control performance

So far it has been found that merely one CSD parameter is controllable. In addition it is shown that using \(y_{r1}\) as an output parameter leads to a controllable structure. What has not yet been addressed is how control performance is affected when the detection efficiency function, which characterizes a sensor, is changed. For instance when another sensor is installed.
Fig. 8.11: Singular values for design 3 with \( \{Q_f, Q_{pf}, P_{tot}\} \) as inputs and \( \{y_{r1}, \Delta C, M_t\} \) as outputs.

To study this, two basic design parameters related to the sensor will be studied:

1. The location of the detection efficiency curve.

2. The shape of the detection efficiency curve.

The discussion is limited to fines removal based controllers. First, it will be shown that a lower bound exists, below which detection of fines for the purpose of control will introduce a right half plane zero in the controlled system which strongly limits the achievable control performance.

### 8.6.1 Lower bound for detection range

In most crystallization processes fines removal will counteract itself by the mechanism of secondary nucleation. For example, increasing the fines flow will lower the surface area of particles, which increases the supersaturation and growth rate, and thereby the total number of large mother crystals. Both the increase of large mother crystals and the increase of supersaturation will cause additional secondary nuclei by the mechanism of surface and mechanical breeding [40]. This phenomenon is further illuminated by the experimental results presented in Section 9.4, and experimental data presented by [127].

Due to this mechanism there will always be a region in the stationary CSD which will not be affected by fines removal, that is \( \frac{dn}{dQ_f} = 0 \). This is illustrated in Fig. 8.12, where 6 stationary crystal size distributions (calculated from the stationary model equations), are displayed for 6 different fines removal rates in the range of 0.5 to 3.0 litres/s (notice that we focus on fine particles only). This plot shows that in the neighbourhood of \( \approx 35 \mu m \) the sensitivity of the stationary distribution to changes in the fines removal flow is extremely low \( \frac{dn}{dQ_f} \approx 0 \) as the stationary distribution "rotates" around this location. In addition, if the sensor is sensitive to CSD fluctuations below the size \( x \approx 35 \mu m \) only, and a step change is made in the fines removal rate than inverse response dynamics occur: right after the step a rapid
(desirable) decrease of the number of fines occurs, also in the range below 35 μm, however, after a few hours also secondary nucleation starts to rise, and finally drives the response in the opposite direction. From control theory, inverse response dynamics (for SISO systems) corresponds to a right halve plane zero, which is undesired as it is a non-minimum phase element (see also Section 8.2). Due to the large phase lag, which is larger than 180 degrees in the control relevant frequency range, the controller gain has to be lowered to achieve stable closed-loop dynamics. Physically this will mean that the additional nucleation which responds to changing fines removal rate slowly, is now the quantity that is controlled. This result shows that measurement should occur above 35 μm.

Surprisingly, Randolph [192] claims in his patent that a fines removal based controller, which extracts CSD information in the size range of 16 to 32 μm effectively stabilizes a crystallizer. Fig. 8.13 depicts the controller response on a step change in the counter setpoint of 10%, for the case that the sensor is focused in this size range. Clearly, in the first instance, the increased fines removal flow lowers the measured number of fines rapidly, however on a much larger time scale, (t > 2 hours) the signal starts drifting from the setpoint slowly, indicating that the loop is unstable.

As a rule of thumb, \( x_d > 50 \) μm is recommended for the detection location. Notice, however that this value will depend on the process design and the substance that is crystallized. An exact value for the lower bound of the detection range can be calculated from the steady-state model by calculating \( dn(x = x_d)/dQ_f = 0 \). This is however an elaborate calculation. Alternatively, the following simple calculation can be applied. Consider a stationary crystallizer with a volume \( V \) for which the countereacting mechanism of secondary nucleation increases the nuclei density from \( n_{01} \) to \( n_{02} \) after a step of \( \Delta Q_f \) on the nominal fines flow. Using a simplified MSMPR model [197] for the stationary solution in the size region of small crystals, and assuming that the stationary growth rate \( g \) of particles is not affected by fines removal, the following relation holds.
Fig. 8.13: Unstable behaviour when sensor is focused at particles in the size range 16 to 32 µm, after step change in counter setpoint (dashed).

for the location at which the distribution does not change \((dn/dQ_f \approx 0)\):

\[
n_{01} \exp\left(-\frac{Q_f}{gV} x_d\right) = n_{02} \exp\left(-\frac{Q_f + \Delta Q_f}{gV} x_d\right).
\]  

(8.11)

Thus, if the condition

\[
x_d > \frac{gV}{\Delta Q_f} \ln\left(\frac{n_{01}}{n_{02}}\right),
\]  

(8.12)

is not fulfilled, the effect of secondary nucleation will take precedence over the effect of fines removal, and unstable behaviour may easily occur.

If a step change \(\Delta Q_{pf}\) is applied an estimate can be obtained for \(\frac{n_{01}}{n_{02}}\). For example the data in Fig. 9.10 shows that for a fines flow increase of roughly 1 l/s an inverse response of the fines density is observed which is related to an increase of the nuclei density with a factor of 2. Using a growth rate of \(5 \times 10^{-8}\) m/s, Eqn. 8.12 predicts \(x_d = 35\) which compares to the value estimated from Fig. 8.12.

### 8.6.2 Location and shape of detection efficiency curve

The question now remains what is the best location and shape of the detection efficiency curve in the range \(x_d > 35 \mu\text{m}\). In the upper plot of Fig. 8.14 different detection efficiency curves are depicted.

**Curve 1** represents the nominal design with a log-normal distribution for the detection curve and the parameter values given in Table 9.1.
Curves 2, 3 and 4 represent mono-size detection at different locations (indicated with arrows) and

Curve 5 corresponds to the case where all particles in the range of 0 to 1500 μm have an equal efficiency to be detected, for example, the sensor output equals the zeroth moment of the whole distribution.

Although mono-size detection can only be realized for practical purposes with a sensor that detects particles in a narrow size range, it is applied here merely to indicate the effect of the detection location. The lower plot in Fig. 8.14 represents the curves subdivided by the stationary crystal size distribution. Scaling is important for proper judgement because the distribution we measure is an exponential distribution in which the number of small particles greatly outnumbers the number of large particles. Consequently, the sensitivity of the sensor output for the number of larger particles will be negligible.

In Fig. 8.15, the Bode plot of the loop-gain of the system is given, and in Fig. 8.16 step responses are given on a positive step change in the counter setpoint of 10%. These results were obtained using a PI-controller. The sampling period for the controller T was 60 seconds. The integral time was fixed at 3600 seconds and the proportional gain k_p was chosen in a way that the gain or phase margins for the different systems were comparable. For systems with an infinite gain margin the proportional gain was chosen in a way that the sensitivity to a measurement noise, is equal. For the different cases a proportional gain value representing an increase of 2.0 litres/s of the removal rate for a 100% change in the number of counts gave good responses.
8.6 The role of sensors on the achievable control performance

Fig. 8.15: Bode plots of loop-gain for five different detection efficiency curves.

Fig. 8.16: Step responses of closed-loop process for five different detection efficiency curves.
As shown by the Bode plot in Fig. 8.15, the dynamic behaviour corresponding to the curves 3 and 4 suffers from a large delay, which is evidently caused by the time that CSD fluctuations, caused by actuator manipulations, need to grow into the region where they can be detected. Hence, detection of particles with a size that is outside the region where they can be removed, easily causes instability of the closed-loop system. Therefore, we recommend to select the location or maximum value of the detection efficiency curve inside the region where the fines removal system is active.

Fig. 8.15 and Fig. 8.16 further reveal that the other three detection curves (1.2 and 5) give rise to comparable closed-loop dynamic behaviour. Besides, all three cases have an infinite gain margin, and could therefore be designed with the same sensitivity to noise. The best behaviour is obtained when the peak of the detection efficiency curve is located between 60 and 90 $\mu$m. Below 60 $\mu$m instability may occur and above 90 $\mu$m a phase lag is introduced, which may cause instabilities as well. This detection location achieved easily with particle counting, however, not with a sensor which outputs a quantity that is related to the total fines surface area of small particles (the second moment of the fines distribution) as proposed by [141] or fines density (the third moment of the fines distribution) as proposed by [211]. These outputs will always correspond to monotonically increasing detection efficiency functions for which the maximum value is located on the right boundary of the detection region.

To sum up, detection of fines in the region $50 < x_d < 150$ $\mu$m will in our case, irrespective of the shape of the detection efficiency curve lead to a stable closed-loop behaviour, provided that the controller is well tuned. For other crystallizers the formulated heuristic rules will provide a good starting point.

### 8.7 Conclusions

In this chapter we have shown that control issues should be integrated in the design stage. When appropriate sensors and actuators are present to achieve stability the design should be done on the basis of steady-state process performance, rather than stability.

The steady-state and dynamic controllability for three designs are compared. With a standard MSMPR crystallizer an inappropriate product is obtained with a low mean and a large spread. Further it is found that no actuators are present for this process to reject disturbances or to speed-up the startup dynamics.

An improved stationary CSD with a larger mean and a smaller spread can be achieved with an MSMPR crystallizer equipped with a fines removal system, however at the expense of open-loop cycling. Using fines removal as actuator stability can be achieved easily. Main design parameters are the nominal flow and the cross sectional area of the classifier. A lower cross sectional area of the annular settling zone does not hamper the dynamic performance, but only increases the amount of solids that have to be dissolved. A larger removal flow also increases the solids fraction, however, also
8.7 Conclusions

increases the bandwidth of the system and thereby the speed at which disturbances can be compensated.

Classified product removal with a vibrating screen further improves the product quality. In particular the spread of the distribution is lowered, while the supersaturation in the crystallizer also drops. This extension to the simple design, however, enhances cycling of the CSD especially at low feed flows to the screen. We found that the same stabilizing loop which uses information on the fines density can be used to achieve stability.

Main factors that should be considered for the design of a product classification system are the nominal flow to the screen and the aperture size. Both a low feed flow and a large aperture size will increase the product quality, but also increase the solids fraction in the crystallizer.

For the presented continuous ammonium sulphate crystallizer, closed-loop stability was not an issue when the sensor counts particles in the size range of 60 to 90 μm only. In this range controller tuning is performed to reduce the sensitivity to measurement noise, and to limit the allowable control input energy, to achieve a certain performance. Closed-loop performance may be bad and stability problems may occur when the detection region is chosen outside the region where the fines removal system is active. In particular, a right half plane zero — that strongly hampers control performance — is introduced if the sensor is focused specifically onto particles in the size range $x < 35 \, \mu\text{m}$. Physically the right half plane zero is caused by a secondary nucleation effect that counteracts the effect of fines removal.
Chapter 9

Design of simple feedback controllers

In this chapter simple stabilizing feedback controllers are analyzed and verified experimentally. Some heuristic design rules are given that may be useful for industrial applications where rigorous models are absent.

9.1 Introduction

In the literature, many simple heuristic schemes are proposed for control of crystallizers. The work of Randolph et al., [14, 198, 195] represents the first milestone in this field. With experimental results from small laboratory scale reactors they showed that simple proportional feedback controllers, which manipulate the fines removal rate based on information of the fine crystals in the CSD, can be used to control the CSD. The main hypothesis underlying this control approach, is that a stabilized number of fine crystals in the crystal population will in the longer term lead to a stable overall crystal population. This control approach, for which strong analogies can be found in the literature on population biology [168], was introduced into the field of crystallization by Beckman and Randolph [14]. A recent patent has been issued on this scheme to Randolph [192]. Also Rohani et al., [212, 211] have presented experimental results obtained with a similar control scheme. Reviews of published work on the topic of CSD control are given by [201, 49].

The experimental evaluation of CSD controllers on larger scale crystallizers, however, has not yet been addressed in the literature. This is mainly due to limitations in applying robust on-line CSD sensing over a sufficiently long time span, and the large probability of disturbances and other operational problems that corrupt the desired experimental process trends. Besides, no discussion is found in the literature addressing questions about what is the best sensor, from an economic and operational point of view (e.g., accuracy and robustness for disturbances), and what model accuracy is needed to design this controller on the basis of a dynamic process model.

The purpose of this chapter is to study design and implementation aspects of simple
single-input single-output stabilizing controllers for continuous crystallizers, based on an actuator and a sensor that both operate in the region of fine crystals. In Section 8.6 we have studied how the sensor may limit the achievable control performance. It was found that a lower bound exists below which control difficulties (right half plane zeros) occur which strongly limit the achievable control performance.

This chapter first starts with a discussion on robustness issues, that are highly important for industrial applications. The robustness of a counter based controller will be discussed. Lengthy pilot plant runs are performed to show the feasibility of the control system. First, the ability to stabilize the CSD after startup is considered. Second, the ability to track setpoint changes for both the proportional and the proportional integral controlled process is considered by changing the setpoint of the controller. The experimental results are compared with the signal trends generated with the simulation model. Furthermore, the disturbance rejection capability of the PI-controlled process for a deliberate disturbance is tested. Finally some perspectives and some heuristic controller design rules are given for industrial applications.

9.1.1 Control structure

The control scheme is outlined in Fig. 9.1. In this figure $d(t)$ represents a possible disturbance, and $m(t)$ the measurement noise. Only SISO PI-controllers are considered, as they are most common in standard process control systems.

The controller gain $k_p$ and the integration time $T_i$, which are used here, are defined by:

$$Q_f = k_p \{ \epsilon_k + \frac{T}{T_i} \sum_{n=0}^{k} \epsilon_n \} , \quad \text{with, } \epsilon_k = N_f - N_{f_{setp}} \quad (9.1)$$

The error $\epsilon_k$ is the difference between the current counter output $N_f$ and the setpoint value $N_{f_{setp}}$. $T$ is the sample time and $T_i$ is the integral time or reset time. Raw sensor readings are available every 10 seconds. A first order pre-sampling filter is applied. The actual controller sampling time $T$ was taken 60 seconds. This value roughly equals 10 times the fastest time constant of the relevant process dynamics.
9.1.2 Sensors

Particle counter

The fines counting technique is a useful sensing tool in a control system, because it is inexpensive and robust, for example, the sensitivity to fluctuations in the laser intensity is low, and to the optical transmittivity of the liquid phase is low. In addition, the fine particles and insoluble dust particles in the micron and sub-micron range can be excluded from the signal easily, by applying a threshold to the raw signal as described in Section 3.3.4. Moreover, as was shown in Section 8.6, the desired closed-loop characteristics of a counter based controller can be established more easily, compared to other sensing techniques like obscuration or density based measurements. For the sampling location, the crystallizer vessel is chosen. Measurement in the fines loop is an alternative, however, here transport delays of approximately 10 minutes can occur, which will deteriorate the controller performance. Besides, it should be noted that control of the removed (number) flow of fines will in principle not lead to effective control of CSD, as it stabilizes the removed number of fines instead of the density of fines in the crystallizer.

The detection efficiency function is described by the function Eqn. 2.37. Estimating $p_{s1}$ and $p_{s2}$ is described in Section 4.4 and the numerical values are given in Table 4.1. Below, the term location denotes the maximum value of other detection efficiency curves as well. The constant $p_{s3}$ in Eqn. 2.37 is used to match the simulated response to the measured response. A nominal value of $p_{s3} = 3.6 \times 10^{-13}$ is found. Fig. 9.12 illustrates how the process model including the sensor model describes the experimental data well.

Malvern particle sizer

A Malvern particle sizer, located in the main slurry discharge line (SL1 in Fig. 3.1), is used in the first instance to monitor the CSD in the crystallizer. Herewith the performance of the simple particle counter could be judged independently. One additional experiment is performed where the first principal component of the raw Malvern output (see Section 4.5.2) is used as a process output parameter.

9.1.3 Controller design using the first principles model

First a controller will be designed using the first principles model. A robustness analysis will be performed which will show what uncertainties in the process and the model limit the achievable control performance. In the first instance the counter based controller is considered.

The constant process conditions used in the simulation study are summarized in Table 9.2. For frequency domain analysis the 100th order (the last element represents
the linearized supersaturation) model is used and for simulation a 200th order nonlinear model is used.

With the controller given by Eqn. 9.1, and the state space model Eqn. 2.66, the so called return ratio of the controlled system is calculated to visualize the Bode or Nyquist plot of the system, and to judge the corresponding stability and robustness properties. The return ratio $K(s)$ is determined by the product of the process and controller transfer-functions $K(s) = G_p(s)G_c(s)$ with $G_p(s) = C(sI - A)^{-1}B$ and $G_c(s) = k_p \left(1 + \frac{T_i}{s}\right)$. On the basis of these frequency domain models the sensitivity and complementary sensitivity have been calculated. For a description see Section 10.2.1.

### 9.2 Tuning the controller

The eigenvalues of the open-loop system matrix $A$, reveal that the open-loop system is stable. Visual inspection was used to tune the controller. For a closed-loop system with an infinite gain margin, the controller sensitivity to an uncorrelated measurement noise added to the signal at each sampling time instant of 60 seconds, was taken as a tuning criterion. The selected proportional gain causes a change in the removal flow of 2.0 litres/second for a 100% change in the number of counts. The selected integral action $T_i$ was chosen 3600 seconds. In Fig. 9.2, the Bode plot of the open and closed-loop system are given. The gain at zero frequency was scaled to unity for the open-loop plant in this plot.

As can be concluded, both the P- and PI-controller reduce the resonant peaks

### Table 9.1: Nominal parameter values and estimated upper and lower uncertainty bounds

<table>
<thead>
<tr>
<th>parameter</th>
<th>equation</th>
<th>nominal value</th>
<th>lower bound</th>
<th>upper bound</th>
</tr>
</thead>
<tbody>
<tr>
<td>$p_1$</td>
<td>Eqn. 2.23</td>
<td>1.00</td>
<td>0.80</td>
<td>1.5</td>
</tr>
<tr>
<td>$p_2$</td>
<td>Eqn. 5.4</td>
<td>0.34-10^8</td>
<td>0.05-10^8</td>
<td>0.6-10^8</td>
</tr>
<tr>
<td>$p_4$</td>
<td>Eqn. 2.23</td>
<td>650-10^{-6}µm</td>
<td>550-10^{-6}µm</td>
<td>750-10^{-6}µm</td>
</tr>
<tr>
<td>$p_5$</td>
<td>Eqn. 2.23</td>
<td>2.15</td>
<td>1.5</td>
<td>3.0</td>
</tr>
<tr>
<td>$p_8$</td>
<td>Eqn. 2.26</td>
<td>8.00</td>
<td>6.0</td>
<td>11.0</td>
</tr>
<tr>
<td>$p_{f1}$</td>
<td>Eqn. 2.11</td>
<td>4.6</td>
<td>4.1</td>
<td>4.9</td>
</tr>
<tr>
<td>$p_{f2}$</td>
<td>Eqn. 2.9</td>
<td>0.24</td>
<td>0.22</td>
<td>0.26</td>
</tr>
<tr>
<td>$p_{s1}$</td>
<td>Eqn. 2.37</td>
<td>120-10^{-6}µm</td>
<td>85-10^{-6}µm</td>
<td>155-10^{-6}µm</td>
</tr>
<tr>
<td>$p_{s2}$</td>
<td>Eqn. 2.37</td>
<td>1.8</td>
<td>1.4</td>
<td>2.1</td>
</tr>
<tr>
<td>$p_{s3}$</td>
<td>Eqn. 2.37</td>
<td>3.6-10^{-13}</td>
<td>2.5-10^{-13}</td>
<td>4.7-10^{-13}</td>
</tr>
</tbody>
</table>
Table 9.2: Constant process conditions.

<table>
<thead>
<tr>
<th>Property</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>temperature crystalline</td>
<td>50 °C</td>
</tr>
<tr>
<td>saturation temperature of feed</td>
<td>50 °C</td>
</tr>
<tr>
<td>product flow</td>
<td>0.215 l/s</td>
</tr>
<tr>
<td>temperature feed</td>
<td>55 °C</td>
</tr>
<tr>
<td>heat input</td>
<td>120 kw</td>
</tr>
<tr>
<td>effective volume</td>
<td>0.970 m³</td>
</tr>
<tr>
<td>temperature fines</td>
<td>60 °C</td>
</tr>
<tr>
<td>fines flow particle counter</td>
<td>15 ml/s</td>
</tr>
</tbody>
</table>

Fig. 9.2: Bode plot for the normalize open-loop plant (solid), and both a closed-loop P- (dashed) and PI-controlled (dotted) plant.
present in the open-loop transfer-function. Clearly, the integral action eliminates the steady-state offset. Note, however, that the analysis we apply here is based on a lumped and linearized population balance model. Uncertainties, introduced in the linearization or lumping step, are not accounted for.

9.3 Robustness analysis

Robustness of the designed controller for uncertain model parameters is studied by perturbing the empirical model parameters around their nominal values. For this study, the designed PI-controller using information from the particle counter was used. The controller settings selected for the nominal plant as described above were used. Further details on robustness analysis are provided in Section 10.2.3.

Uncertain empirical parameters are used in the relation for nucleation (Eqn. 2.23), the length dependent growth function (Eqn. 2.26), the fines removal efficiency function (Eqn. 2.9), and the fines detection efficiency (Eqn. 2.37). Table 9.1 summarizes the nominal values of these parameters and their uncertainty bounds. The parameter uncertainty bounds were taken equally to the largest and smallest estimated values, which were obtained for seven independent process startup responses at different process conditions (see Chapter 5). These bounds are chosen conservatively, as it was not possible to estimate them accurately. The other model parameters are kept constant as they do not contribute to the process input-output behaviour, significantly ($p_2 = 1.0$, $p_6 = 1.0$, $p_7=1.0\cdot10^{-8}$ and $p_9=1000$ μm). Further, it is assumed that parameters related to kinetics (i.e., \(\{p_1, p_3, p_4, p_5, p_8\}\), are not correlated with the parameters related to the crystal removal and detection, (i.e. \(\{p_{f_1}, p_{f_2}, p_{s_1}, p_{s_2}, p_{s_3}\}\), see also Table 9.1).

Robustness is studied for these two sets of 5 uncertain parameters, by perturbing each parameter to its lower bound, its nominal value, and its upper bound. Linearizing of the corresponding nonlinear models and calculating the total set of return ratios in the frequency domain for all possible 243 (≡5^3) combinations of these parameters, gives an indication of the controller robustness for both modelling errors and changes in the process parameters.

The results obtained for both sets of parameters are depicted in Figs. 9.3, 9.4 and 9.5. Fig. 9.6 depicts the simulated performance of the worst case closed-loop plant in Fig. 9.4. These results show that the parameter uncertainties can give rise to bad closed-loop performance or even instability. However, comparing Fig. 9.3 with Fig. 9.4 shows that the robustness is mainly affected by the uncertainties in the parameters related to particle detection and removal. Hence, the closed-loop properties are most sensitive to errors in the parameter values for the spread and the height of the detection efficiency.

The results further show that varying the kinetic parameters \(\{p_1, p_3, p_4, p_5, p_8\}\), mainly affects the gain of the transfer function at low frequencies, while the stability of the system is not affected. In particular it is found that parameter $p_1$ (see Eqn. 2.23)
Fig. 9.3: Nyquist contour of return ratios for the set of uncertain parameters related crystallization kinetics, together with $M=1.3$ circle.

Fig. 9.4: Nyquist contour of return ratios for the set of uncertain parameters related to particle detection and removal, together with $M=1.3$ circle.
Fig. 9.5: Gain of sensitivity (|S|), and complementary sensitivity (|T'|), for the set of uncertain parameters related to crystallization kinetics (left), and the set of uncertain parameters related to particle detection and particle removal (right).

Fig. 9.6: Comparison of simulated step responses on the counter setpoint $N_{I_{set}}$ (dotted) for the closed-loop nominal plant (solid) and the worst case (unstable) model (dashed) in the set of uncertain parameters related to particle detection and particle removal.
Table 9.3: Overview of experimental results.

<table>
<thead>
<tr>
<th>Experiment</th>
<th>Description</th>
<th>Controller</th>
<th>Sensor</th>
</tr>
</thead>
<tbody>
<tr>
<td>RUN42</td>
<td>startup behaviour</td>
<td>PI</td>
<td>counter</td>
</tr>
<tr>
<td>RUN57</td>
<td>startup behaviour vibrating screen</td>
<td>PI</td>
<td>Malvern</td>
</tr>
<tr>
<td>RUN41</td>
<td>Setpoint tracking</td>
<td>P</td>
<td>counter</td>
</tr>
<tr>
<td>RUN43</td>
<td>Setpoint tracking</td>
<td>PI</td>
<td>counter</td>
</tr>
<tr>
<td>RUN50</td>
<td>Open-loop disturbance response</td>
<td>open</td>
<td>counter</td>
</tr>
<tr>
<td>RUN53</td>
<td>disturbance response</td>
<td>PI</td>
<td>counter</td>
</tr>
</tbody>
</table>

is responsible for the largest fluctuations in the return ratio. Because the gain at low frequencies is large, due to the integral action, the uncertainties in these parameters do not affect the closed-loop performance. Also, the sensitivity and complementary sensitivity plots in Fig. 9.5 confirm that the ability to attenuate disturbances or to track the setpoint is hardly affected by the uncertainties in the kinetic parameters. However, the uncertainties related to removal and detection can cause deviations of up to 50% in the ability to suppress disturbances in a specific frequency range.

These results further suggest, that for identification of the empirical model parameters most (experimental) efforts should be spent to obtain a small error in the estimates for the parameters related to removal and detection of particles. Accurate estimates of the kinetic parameters, which are also difficult to obtain, is less stringent. In addition, the results show that it is desirable to select a sensor for which the detection characteristics do not alter during operation, for example, due to fouling or changing temperatures.

Because the uncertainty bounds on the parameters are conservative, and because only three specific parameter combinations caused instability we decided to implement the controller developed for the nominal plant on the pilot crystallizer.

9.4 Experimental results and discussion

Most experiments are performed using the particle counter, except for one experiment where the first principle component of the output if the Malvern particle sizer has been used. The complete set of experiments, using a P- and PI-controller, is tabulated in Table 9.3. Table 9.2 summarizes the nominal process conditions.
9.4 Experimental results and discussion

9.4.1 Stabilizing control using Malvern particle sizer

The controller using the parameter $y_{r1}$ as its output is evaluated first. In Section 6.3 the results of a closed-loop startup response and two successive setpoint changes are described. The same chapter introduces a feedback controller that has been used to identify the process dynamics in a closed-loop manner. As may be concluded from these first results effective stability could be achieved.

One additional experiment has been conducted with the same control setup, however now the process is operated with the vibrating screen. The same procedure as in the open-loop counterpart of this experiment (RUN54, see Fig. 5.21 and 5.21) is followed: first the process is started without product classification; after a sufficient level of stability has been reached, the classifier is switched on and the response to the CSD and the magma density $M_1$ has been measured. After switching on the product classification the controller has been kept in cascade and the closed-loop response has been measured for approximately 30 hours in time. The integral time $T_i$ and the proportional feedback gain $k_p$ (cf. Eqn. 9.1) were: 2400 s and 4.0e-3, respectively.

Fig. 9.7 depicts the controlled variable and some related process parameters. In Fig. 9.8 the corresponding manipulated variable is given. As is shown an effective reduction of the open-loop cycles is achieved. Furthermore also cycling of the magma density is eliminated, however, still some undesired transients are visible. These are due to the product flow changes (cf. C and D in Fig. 9.7), and may be cancelled by adding one loop to the controller, as is done in Chapter 10.

9.4.2 Startup behaviour using particle counter

In Fig. 9.9, the startup behaviour of the process with the counter based controller (RUN42) is compared to an open-loop startup. As can be seen, the counter based controller reduces the oscillations in the median crystal size, induced from the initial distribution. The initial CSD of the non seeded crystallizer results from heterogeneous primary nucleation occurring in the supersaturated mother liquor. Due to the limited dynamic range of the actuator (particles can only be removed, not created), the controlled response has significant overshoot. Therefore, better results may be expected when besides a fines removal system also a fines generation system is implemented. This can be achieved with a pump that causes strong particle abrasion. Addition of fines will, however, only be attractive at the dynamic startup phase, or right after a disturbance has occurred, as it will always increase the total number of fines, which is not desired in view of optimal solid-liquid separation in the process downstream.

9.4.3 Setpoint tracking

Two control experiments (RUN41, RUN43) have been performed to find out to what extend the proposed controller is able to move the CSD from one stationary state to another.
Fig. 9.7: Open-loop (RUN54) and closed-loop (RUN57) responses of the controlled variable: \( y_{r1} \) (upper), the median crystal size and the magma density. The screen has been switched on at A; at B the setpoint of the fines controller is lowered from 0 to -0.2; at C a step changes is made to the product withdrawal flow \( (Q_{p,f}) \) from 0.75 to 1.0 l/s; at D this flow is returned to 0.75 l/s.

Fig. 9.8: Manipulated variable measured during RUN57 (see also Fig. 9.7).
Setpoint tracking with P-controller (RUN41)

In Fig. 9.10, the results of a first experiment lasting 35 hours, where a proportional controller was used, are depicted. In Fig. 9.11 the corresponding fines removal flow is given. As shown, a significant response to these step changes is obtained. In Fig. 9.10, the response of the product density signal is also given. As expected, there is a negligible interaction between the fines flow and the product slurry density.

As shown by the other plots, the setpoint changes greatly affect the counted number of fines. The controller moves the process rapidly, within one hour, to the new setpoint in a first stage of the response. This behaviour resembles the first order behaviour of a well-stirred tank, as the fines concentration follows an exponential response with a time constant that equals the mean residence time constant of fines: $\tau = V/Q_f \approx 500\text{s}$. In a second stage of the response, however, a dominant inverse response is developed that drives the system back to its original value. This drift is explained by the additional secondary nucleation. In Section 8.6 it was shown that this counteracting mechanism may introduce a right half plane zero in the loop if the sensor location is chosen improperly.

This counteracting mechanism also indicates that fines removal can move the CSD within a limited setpoint range only. For large changes in the stationary fines flow of 1.5 l/s, an increase in the median crystal size of approximately 100 $\mu$m is observed. Imposing a setpoint change of 125 counts, near the end of the run, shows that the range of attainable setpoint values is limited: This low value cannot be reached and the process becomes unstable again. From these results it can be concluded that fines removal can move the median crystal size to values in the range of 600 to 800 $\mu$m. If a coarser product is desired, other actuators like a product classifier should be considered.
Fig. 9.10: Measured responses (solid) of the counter signal (upper), the median crystal size and the magma density (lower), for a P-controlled process (RUN41), on step changes in the counter set-point $N_{f_{step}}$ (dotted).

Fig. 9.11: Measured response of fines removal flow, on step changes in the counter set-point (RUN41).
Fig. 9.12: Measured responses (dashed) of the median crystal size (upper), the counter signal and the fines flow, for a PI-controlled process (RUN43), on step changes in the counter set-point $N_{f, \text{step}}$ (dotted), and the simulated response (solid).

Setpoint tracking with PI-controller (RUN43)

A second experiment is performed to study the reproducibility of the results. Integral action was added to the controller to compensate for the slow drift of the signal caused by additional secondary nucleation. According to Eqn. 9.1, an integration time constant of 60 minutes and a proportional gain giving rise to a change of 2.0 litres/second in the removal flow rate for a 100% change in the detected number of counts, or the counter setpoint. In Fig. 9.12, the results of this experiment are given. Significant responses to step changes are obtained with a negligible static deviation. Notice that at 13 and 18 hours, undesired disturbances occurred. These were due to blockage of the fines sampling system. In these plots, the setpoint values are added as a dotted line. Note that the counteracting mechanism of secondary nucleation causes the integrator to be active over a period of approximately six hours.
Comparison with simulated behaviour

The experimental results are compared with data from the simulation model to check the model validity. In Fig. 9.12 the predictions of the simulation model, for the nominal design, described in the previous section, are added as a solid line to the plots. The same controller as implemented on the process was implemented on the simulation model. As can be concluded, the results are well predicted by the model. Comparable model responses were found for the other data sets depicted in this section.

9.4.4 Disturbance behaviour

Another important feature to be evaluated is the ability of the controller to reject process disturbances. As a representative disturbance, a product removal blockage is simulated by blocking the product discharge for one hour, while the other process quantities were kept at their setpoints. This causes the process to run in batch mode for one hour. In a first experiment the open-loop behaviour of the process is tested for this disturbance. In a second experiment the closed-loop behaviour is evaluated. For both experiments the disturbance is repeated twice to evaluate the reproducibility.

Open-loop disturbance behaviour (RUN50)

In Fig. 9.13, the magma density of the product is given as a function of time. As expected the density increases linearly and decreases exponentially after releasing the product discharge line. During this open-loop run, the stabilizing controller is active between B and C to stabilize the response before the next disturbances, which was necessary to limit the experimental duration. The same figure depicts the responses of the median crystal size, measured with the Malvern, and the counted number of fines from the particle counter. Around 10 hours an undesired disturbance occurred in the signal trend and at 15 hours right before the start of the second disturbances fouling of the detector occurred which caused an offset in the signal trend.

Despite these undesired process disturbances, it is observed that the counter signal first increases after closing the product discharge pipe (see also Fig. 9.14) and then decreases rapidly to reach a minimum of nearly zero counts. After that, a dampened oscillation occurs which brings the signal back to its original value. The increase of the counter signal is caused by the sudden decrease of product removal, which will decrease the removal of fines via the unclassified removal of product crystals. The sharp decrease is caused by a decrease in the supersaturation and the growth rate, which results from an increase of the total surface area. Due to the lower growth rate the chance that a fines particle survives the fines trap drops drastically (cf. Eqn. 5.6) and thereby the counted number of fines drops.
Fig. 9.13: Measured open-loop responses of the counter signal, the median crystal size (middle), and the product magma density (lower) due to a product flow disturbance at A and C. The stabilizing controller has been active between B and C and after D (RUN50).

**Closed-loop disturbance behaviour (RUN53)**

In Figs. 9.14 and 9.15, the PI-controlled response of the process for the described product flow disturbance is given. In Fig. 9.14 the process outputs are given and Fig. 9.15 describes the response of the fines flow. As can be concluded from these experiments, the control system attenuates the oscillations induced by the disturbance. However, a dynamic effect in the CSD remains. This can be explained by the limited size range of the distribution at which the fines detection and actuation mechanism is active. Disturbances which affect the distribution outside the range of the controller can only be corrected when they appear, with a certain delay, in the control region via a growth, or a nucleation related mechanism.
Fig. 9.14: Measured closed-loop responses (solid) of the PI-controlled counter signal (RUN53), the median crystal size (middle) and the product magma density (lower), for product flow disturbances at A and B (dotted).

Fig. 9.15: Measured closed-loop response (solid) of the fines removal flow, for product flow disturbances at A and B (dotted) (RUN53).
9.5 Perspectives for stabilizing control of industrial crystallizers

Based on the results presented in this chapter and Section 8.6 heuristics are developed that may be useful for industrial control applications where stabilizing control is the objective. We assume that rigorous models are not available. Clearly, if an accurate model is available controllability analysis described in Chapter 8 and the design procedure reported in this chapter should be applied.

The design of a control system will involve three major steps, selection and installation of a sensor, selection and installation of an actuator, and tuning of the controller. These steps are shortly addressed below.

9.5.1 Selection of a sensor

A simple sensor outputting a scalar measure will generally be sufficient. To locate the sensor in the crystal size domain is, however, most critical. Thus it is important to identify, a priori, the detection efficiency function. In Section 8.6 it is shown that a lower bound exists below which the achievable control performance is hampered due to a right half plane zero. While measuring too large particles will introduce a growth time delay that causes an additional phase shift in the loop that also prohibits tight control responses. It is found that the shape of the detection efficiency curve is less stringent.

When the presented requirements on the location of the detection efficiency curve are fulfilled, the selection of a sensor for CSD control should primarily be based on economic factors, for example, price, lifetime and maintenance cost; operational robustness, for example, sensitivity to changing optical properties of the liquid phase, temperature and pressure changes; and statistical properties, that is, a good signal to noise ratio. Currently, the availability of such sensors is limited.

For a fines removal system, which has a cut-size of 100 μm, a sensor described by the log-normal function Eqn. 2.37, with a location of 120 μm and a spread of 1.8, gives good results.

9.5.2 Actuator

The selection of an effective actuation mechanism for the CSD is also highly important. It has been shown that fines removal is an effective actuation mechanism. Note however, that the results in Chapter 8 show that fines removal should be regarded as an actuation mechanism used to achieve stable operation; to achieve a coarse product it is better to install a product classifier.

From a control point of view the fines removal setup of the UNIAK pilot crystallizer is most profitable as changes in the fines removal rate solely effect the CSD, while the magma density and the mass production rate remain unaffected. The range over which the fines removal rate should be variable for this configuration is recommended to be more than ± 0.5 l/s for a 1 m³ crystallizer.
In many industrial plants (e.g. Oslo and Forced Circulation type crystallizers, [17]) all heat is supplied externally in the fines loop. These processes differ from the UNIAK setup in that the actuator range is more limited and that alternating the circulation flow will affect the heat and mass balances as well. For these systems the following alternative actuation mechanisms are proposed:

**Mechanism 1** If the external loop contains only fine crystals, then use the flow rate as the manipulated variable and keep the heat input in the external loop constant to avoid interaction with the mass balances in the main volume. Now the temperature of dissolved fines will be related to the fines flow. Consequently, with low flows the temperature rises, which may cause flashing, and with large flows the fines may only partially dissolve. Thus the range over which the flow can be varied is limited.

**Mechanism 2** A configuration similar to the UNIAK set-up can be achieved by using two external loops. One is used to supply heat to a nonclassified slurry, and one is used to supply heat to a classified fines flow. With this setup a constant fines temperature can be established in the external fines loop, and a constant heat input can be established with external loop containing unclassified slurry. In this way dissolution of fines is always guaranteed and interaction with the heat balance is avoided.

**Mechanism 3** When all heat is supplied externally the problem of oscillatory behaviour may be more due to a lack of fines than an abundance of fines. Hence, fines addition instead of fines removal is expected to be an effective actuation mechanism in this case. Fines addition can be achieved with a granulation device or with a small reactor which is placed in the feed-line where small particles are bred.

**Mechanism 4** Dissolve fines by injection of solvent in an external loop. Now the removal flow rate may be used as an actuator, while the amount of solvent is controlled by measuring the magma density in the loop. This may be useful when the temperature-solubility curve is too flat. Because the volumetric fraction of solids (<0.1%) in the fines loop is low the economic loss may be limited.

Notice that all configurations introduce the lower bound as described in Section 8.6 the value of this bound may however differ. Using the model described in Chapter 2 the effectiveness of these configurations may be assessed prior to implementation.

### 9.5.3 Tuning the controller

Usually simple PI controllers will be sufficient (see also Chapter 10). For controller tuning it is recommended to select a large integral time $T_i$ (i.e., larger than 60 minutes) and a low proportional gain (Eqn. 9.1). For the UNIAK set-up good results have been
obtained with an integration time constant of 30 to 60 minutes and a proportional gain, of 2.0 litres per second fines removal rate (for an effective crystallizer volume of 1 m$^3$ slurry), per 100% change in the detected number of counts. If after applying a step change oscillations occur as the one described in Fig. 9.6 then lower the proportional gain. If the response is desirable in the first instance, but a slow drift occurs at a larger time scale, (see Fig. 8.13 ) then the sensor should be focused more on larger particles.

9.6 Conclusions

A simple PI-controller has been designed for a continuous crystallizer, that acts on the number of fines by means of a fines removal system, on the basis of a measured number of fines inferred from a simple on-line particle counter and a Malvern particle sizer. From the results the following conclusions are drawn:

1. From a stability and operational point of view, particle counting is a valuable technique to serve as a sensor for a stabilizing controller that acts on the fines removal rate in a continuous crystallization process.

2. Disturbances affecting the whole crystal size distribution can be suppressed to a certain extend with the proposed control scheme. In particular the influence of product flow disturbances which affects both small and large particles, could be suppressed.

3. With the described controller the median crystal can be varied in the range of 600 to 800 $\mu$m.

4. The development of an accurate model is in general expensive, however valuable insight into the dynamics of the underlying process may be obtained. From the robustness evaluation it is found that modelling effort should aim primarily on the fines removal and particle detection sub-systems, as errors in these models can strongly determine the stability robustness of the controller. Accurate estimates of kinetic parameters is less important as the modelling errors will not affect robustness and also not the performance of the designed control system.

With results from a semi-industrial evaporative crystallization process, we have shown that stabilizing control is in principle possible. Good results are obtained, with a standard PI controller and a simple sensor, provided that certain requirements related to the characteristics of the sensor are fulfilled.

For the general class of industrial suspension crystallizers some heuristics have been developed. These come down to the selection of a suitable sensor and an effective actuation mechanism.
Chapter 10

Model based control design

In this chapter it is studied to what extend advanced model based feedback controllers can improve the closed-loop performance of the process, compared to the simple feedback controller described in the previous section.

10.1 Introduction

In the previous chapter it is shown that already with a simple PI controller stable operation of the process can be achieved. In Chapter 8 it is stated that also a multi-loop configuration using simple PI-controllers may give satisfactory results for the multiple input multiple output (MIMO) behaviour of the plant. However, these simple solutions may yield less satisfactory performance in case:

- tight input/output constraints exist;
- significant interactions occur;

In some sense the first limitation is always present in reality, whereas the second only plays a role for MIMO systems with a large directionality (cf. Chapter 8). For roughly 20% of the industrial control problems both these factors require the use of advanced model based controllers. The benefits and the status of advanced process control in the process industry are discussed in [5, 151]. For crystallization process both factors play a role, though significant interaction only exists for some specific input/output configurations (cf. Chapter 8).

The purpose of this chapter is to study the merits of model based feedback control. This will be done by comparing model based controllers with the simple feedback controllers introduced in the previous chapter. As a tool for model based feedback control model predictive control (MPC) is chosen. The main motivation is that MPC handles constraints flexibly and optimally. For instance changing process constraints due to changing actuator or sensor characteristics (for example due to fouling or failure of a sensor) can be accounted for on-line, from the operator console. In addition MPC is
widely accepted in nowadays process industry. This popularity is due to its constrained handling capability, however, also due to the low maintenance requirements and the fact that tuning can be done by people at a medium level of technical education. Examples of successful applications are reported among others in the oil refinery and the pulp and paper industry [31, 206, 205, 209, 247]. No applications are known where MPC is used for feedback control of CSD properties in industrial crystallization processes.

With model predictive control an internal model representation of the plant dynamics is used to estimate future process inputs, that minimize a performance criterion over a finite time in the future, subject to constraints. The latter can be, constraints on the inputs, the outputs, the rate of change of input variations, and internal or state constraints [81]. The criterion function penalizes both the costs of deviations of the process outputs from the setpoint values, and the process input moves, to obtain a feasible and economic control action.

MPC is a generic name for a large collection of model based control techniques that were developed over the last 20 years. Cutler and Ramaker [38] developed one of the most popular schemes among these, which is known as dynamic matrix control (DMC). Garcia and Morshed [80] added constraints explicitly to the optimization procedure in DMC. Numerous variants and extensions have been developed since then. Garcia provides a review of these [81].

Most MPC approaches, reported for industrial processes, employ step or impulse response models as the internal model. Recently, however, different authors [19, 142, 171, 208] have proposed MPC algorithms on the basis of state space models. The use of state-space models is advantageous as these models compactly describe the process dynamics, and by that lower the computational burden. More important, however, is that within the powerful framework of Linear Quadratic Gaussian (LQG) control [4, 137], which is fully based on state-space models, many well-defined concepts are developed, which translate easily to the MPC framework. A basic feature, for instance, of LQG controllers is that they include a state estimator, which adds a tunable expression for the disturbance behaviour to the controller. In this way an optimal trade-off can be found between noise sensitivity and setpoint tracking.

The early MPC schemes, which employ step or impulse response models, should be regarded as open-loop observer forms of MPC as they do not include an independent tunable expression for the disturbance behaviour of the controller. This motivated different authors [19, 142, 171, 208] to apply so called closed-loop observer forms of MPC that incorporate an estimator for the actual state of the process —equivalent to the one used in LQG controllers. It was found that with a well designed (stable) observer also open-loop unstable systems can be handled.

Recently, the closed-loop observer form of MPC based on state-space models, is formalized by Ricker [208], and implemented in a commercially available Matlab toolbox [163]. The state-space approach is extended to nonlinear process by Gattu and
Zafiriou [85]. They propose a computation scheme that should be denoted as semi-nonlinear as a repeated linearization is made of the nonlinear equations that govern the plant dynamics. With respect to this, their scheme extends a nonlinear QDMC (denoted NLQDMC) approach, proposed by Garcia [82]. The latter also employs successive linearization of the plant model along the trajectory that is followed. Complete nonlinear model predictive (NMPC) controllers are in most industrial cases, however, not attractive due to their large computational burden. With the growing improvement and decreasing price of computing facilities, however, nonlinear predictive control is expected to be of increasing importance in the future. Examples of process control with NMPC are given among others by [183, 181, 46].

Currently two major issues are studied in the field of MPC, which are robustness and feasibility of the minimization problem in case input/output constraints are active. These issues point at the major drawbacks of current MPC schemes, that is, that no unified theory is available for robustness and feasibility when constraints are active. However, recently important progress is made [257, 202, 47].

We proceed this chapter with a short outline of some model based control concepts. In particular LQG control will be discussed, as MPC inherits most of its features. Model predictive controllers will be designed for different process input and output configurations. Results from both simulation and pilot plant experiments will be used to evaluate the performance and the stability of the designed controllers. A comparison with conventional PI control is made to judge whether the improvements obtained with advanced model based control are significant.

## 10.2 Controller design

### 10.2.1 Some basic concepts

The purpose of the control system to be designed is to guarantee stability, to give good regulatory behaviour (i.e., disturbance suppression), and to track setpoint changes (i.e., servo behaviour), in the face of uncertainties, present in both the model used for controller design, and the process itself (e.g., failure of actuators or sensors). A classical control structure is given by Fig. 10.1. In this figure $r$ represents the reference input,
which should be followed by the plant. $e$ is the controller input, $d$ is a disturbance that corrupts the process output trend, $u$ and $y$ are the process input and output, respectively, and $m$ is the measurement noise. Some important relationships in the frequency domain between inputs and outputs are given by [146, 164]:

$$
\frac{e}{d - r} = \frac{y}{d} = (I + PC)^{-1} \triangleq S(s) \tag{10.1}
$$

$$
\frac{y}{r - m} = PC(I + PC)^{-1} \triangleq T(s) \tag{10.2}
$$

The transfer functions $S(s)$ and $T(s)$ are defined as the sensitivity and the complementary sensitivity function, respectively. Another important measure is the return ratio, which is the transfer function of the elements present in the open feedback-loop. The return-ratio of the controlled plant in Fig. 10.1 is given by $PC$. It is common to use the open-loop return-ratio of a plant to assess the closed-loop performance and stability properties.

Generally, one requires the feedback loop to attenuate the disturbances $d$, and to track setpoint changes. Therefore, $S(s)$ should be close to zero and $T(s)$ should be close to one. However, because $S(s) + T(s) = I$ and

$$
\lim_{s \to \infty} S(s) = I \tag{10.3}
$$

one strives to make $S(s)$ close to zero, and thus $T(s)$ close to one, in the low frequency range only. Consequently, the frequency range over which disturbances can be suppressed is limited. For a SISO system the upper limit of this range is often taken at $\|S(s)\| = 1/\sqrt{2}$. The corresponding frequency is also denoted as the bandwidth. Notice that for good measurement noise suppression, that is $y/m \approx 0$, $T(s)$ should be made small. Consequently, in designing a feedback controller a good compromise is search for between effective measurement noise suppression and good setpoint tracking. However, if measurement noise is small or has high frequency components outside the bandwidth only, it will not pose a problem.

An important variant on the classical structure is the so called Internal model control IMC structure, which is depicted in Fig. 10.2. The IMC concept has been introduced by Garcia and Morari [78]. It is important here as IMC is inherent in all MPC structures.

For the IMC controller, three important facts are raised [78]:

1. The IMC controller and the classical control structure are equivalent when:

$$
Q = C(I + \tilde{P}C)^{-1} \tag{10.4}
$$

$$
C = Q(I - \tilde{P}Q)^{-1} \tag{10.5}
$$

with $C$ the controller in Fig. 10.1.
2. If $P = \hat{P}$ than the relation between any input and output in Fig. 10.2 is affine\(^1\) in the controller $Q$. In particular: $y/r = PQ$ and $e/d = I - PQ$. This is in contrast with the classical control structure where the fundamental input-output properties (Eqn. 10.1 and Eqn. 10.2) are a nonlinear function of the controller. From a mathematical point of view it is more efficient to optimize an affine or linear function than a nonlinear function.

3. If $P$ is stable than the IMC controller is internally stable if and only if the classical control system with $C$ is internally stable. If $P = \hat{P}$ and stable, than the IMC controller is stable. For this case the relation between the inputs and outputs is given by:

$$y = PQ(r - d) + d \quad (10.6)$$

if also non-minimum phase elements are absent than controller is obtained when $Q$ equals the inverse of the plant.

Studying Eqn. 10.1 and Eqn. 10.2 reveals that the response to errors $d$ and setpoint changes $r$ cannot be influenced separately as both are determined by the same transfer relationship. So if setpoint changes are different in nature than disturbances (for example when the reference input is a ramp and the disturbance input is a step) it is not possible to design the controller such that both good setpoint tracking and disturbance rejection are achieved. To avoid this limitation an additional degree of freedom is added to the controller [4, 81], as shown in Fig. 10.3. This structure then denotes a two degrees of freedom control structure. Note that for $C_2 = C_3$ and $C_1 = I$ this controller is equivalent to the one degree of freedom controller. As derived by Zafiriou and Morari [256] the two degrees of freedom controller—in the absence of modelling errors—allows for independent tuning of the noise and the setpoint

\(^1\)The relation between $x$ and $y$ is affine when $y = A = Bx$. 
Fig. 10.3: Two degrees of freedom control structure.

responses. Tuning of multivariable two degrees of freedom IMC controllers is treated extensively by Morari and Zafiriou [164].

Standard LQG control schemes [19, 4] employ a two degrees of freedom structure as well. In the sequel we will focus further on some properties of LQG controllers, as MPC based on state-space models is closely related to the LQG controller. The relation between LQG and MPC is explored in detail by Bitmead et al., [19]. For control we employ discrete time models, which relate the process inputs and outputs only at the sampling instants. Between two sampling instants the process inputs are assumed to be constant.

Linear Gaussian Quadratic (LQG) control

The problem addressed by LQG control theory is the following: assume a linear discrete time and time invariant model

\[
x(k + 1) = \Phi x(k) + \Gamma u(k) + v(k)
\]

(10.7)

\[
y(k) = C x(k) + D u(k) + w(k)
\]

(10.8)

with the process noise \(v(k)\) and sensor noise \(w(k)\) that corrupts the states and the measurements, respectively. These are assumed to be independent with a zero mean and uncorrelated.

When full state information is available, a so-called LQ controller can be designed. The problem then is to find a sequence of future controller input values \(u(k), \ldots, u(t + N - 1)\) that minimizes the following finite horizon quadratic criterion:

\[
H(N-1) = \sum_{j=0}^{N-1} x(k+j) \phi_j E \{ \bar{y}^T(k+j) | P_0 \} \phi_j \bar{y}(k+j)
\]

\[
+ u^T(k+j) R_c (N - j - 1) u(k+j) \{
\}

with \(P_c, Q_c\) and \(R_c\) weighting matrices. \(E\) denotes the expected value of the stochastic parameters.
The last term in Eqn. 10.9 penalizes the control input energy to obtain a feasible control action. The term \( x^T(k+N)P_0x^T(k+N) \) in the performance criterion expresses the objective to have a final state \( x(t+N) \), which is as close to the origin as possible. For optimization of batch systems it is often required to have a fixed non-zero terminal state. Then the terminal condition \( x(k+N) = x_f \) should be used in the criterion.

The optimal solution of this feedback control law, is described by the so called Ricatti Difference Equation [4, 137]:

\[
P(j + 1) = \Phi^T P(j) \Phi - \Phi^T P(j) \Gamma (\Gamma^T P(j) + R_c(j))^{-1} \Gamma^T P(j) \Phi + Q_c(j)
\]  

(10.10)

with

\[
u(k + N - j) = -(\Gamma^T P(j - 1) \Gamma + R_c(j - 1))^{-1} \Gamma^T P(j - 1) \Phi x(t + N - j)
\]

\[
= K(j - 1)x(t + N - j), \quad j = 1, \ldots, N
\]

(10.11)

(10.12)

This equation is a recursive equation that generates controller values at each new sampling interval, only via standard linear matrix manipulations. Consequently, it may be used to calculate the time-varying gain values \( K(j - 1), \quad j = 1, \ldots, N \), based on the system matrix \( \Phi \), the input matrix \( \Gamma \), and the weighting matrices \( Q_c, R_c \) and \( P_0 \). The latter also being the initial condition for the Ricatti equation Eqn. 10.10.

A control law that minimizes Eqn. 10.9 can be derived in three different ways:

1. **Finite horizon.** Implementing the finite optimal sequence \( u(k), \ldots, u(t + N - 1) \) that minimizes Eqn. 10.9, directly to the plant, would yield an open-loop strategy, which does not anticipate on unknown future disturbances or load changes.

2. **Infinite horizon.** Calculating:

\[
J(x(k)) = \lim_{N \to \infty} \frac{1}{N} J(N, x(k))
\]

(10.13)

gives the infinite horizon, time invariant LQ controller. For this case the Ricatti Difference Equation converges to an Algebraic Ricatti Equation [19]. For this case the optimal input \( u(k) \) is a constant linear function of the process state \( x(k) \).

3. **Receding horizon.** A third alternative is to implement the finite horizon controller as a receding horizon controller. Now only the first calculated input value \( u(k) = K(N-1)x(k) \), from the optimal sequence \( u(k), \ldots, u(t+N-1) \) is implemented to the plant. At the next control instant \( t + 1 \) the procedure is repeated. Thus the receding horizon also minimizes Eqn. 10.9, however, in comparison to the finite horizon controller it is a feedback control strategy — the minimization procedure is repeated at each time instant based on new measurements of the state \( x(k) \).
Notice that stability considerations only apply to the latter two approaches. The receding horizon form of the LQ controller is important here as the repeated minimization of Eqn. 10.9 over a finite horizon enables to incorporate constraints in the control law computation, as is done by the MPC controller.

**State estimator design**

Lacking directly measurable noise free state variables, the implementation of an LQ control law requires the availability of a state estimator that provides estimates of the current state on the basis of past process input and output values. Therefore, the state variable \( x \) in the above equations should be replaced by its estimate \( \hat{x} \).

The design of an optimal state estimator is dual to the design of an optimal state-variable feedback controller and described for a continuous time model in Chapter 7. The extension to discrete time is straightforward, and is given among others in [19].

The state estimator is a dynamic model of the form:

\[
\hat{x}(k+1) = (\Phi - K_f C \Phi)\hat{x}(k) + (I - K_f C)\Gamma u(k) + K_f y(k+1)
\]

which has both the current process output \( y(k+1) \) and the process input \( u(k) \) as its inputs. The feedback gain \( K_f \) is designed such that the expected state estimation error \( E((x - \hat{x})(x - \hat{x})) \) is minimized. Such an estimator is also known as a Kalman filter.

A Linear Quadratic Gaussian (LQG) controller is obtained by interconnection of the state variable feedback control law and the linear state estimator. An important characteristic of the design procedure is the so-called separation principle [137]. This principle says that the optimal state feedback is achieved from the successive and separated design of an optimal state estimator and a linear quadratic control problem.

**Disturbance modelling**

The model given by Eqn. 10.8 assumes that the noise contributions are zero mean and uncorrelated white noise. For the state noise \( v \) this may be a valid assumption, however, the output noise assumption is too strict for many industrial control applications, with disturbances consisting of ramps, steps, sinusoid's and coloured noise. For these cases it is possible to augment the filter model with a disturbance model to obtain a Kalman filter that eliminates the influence of these disturbances on the estimated state in a desired manner.

The output noise may be generated with a noise generating model, which is driven by uncorrelated zero mean noise sequences \( p(k) \) and \( q(k) \):

\[
x^d(k+1) = \Phi^d x(k) + \Gamma^d p(k)
\]

\[
v(k) = C x(k) + q(k)
\]

Introducing the composite state vector \( x(k) = [x \ x^d]^T \), the augmented
model is written as:

\[
x(k+1) = \begin{bmatrix} \Phi & 0 \\ 0 & \Phi^d \end{bmatrix} x(k) + \begin{bmatrix} \Gamma \\ 0 \end{bmatrix} u(k) + \begin{bmatrix} I & 0 \\ 0 & I^d \end{bmatrix} \begin{bmatrix} w(k) \\ p(k) \end{bmatrix}
\]

(10.16)

\[
y(k)(k) = \begin{bmatrix} C \\ C^d \end{bmatrix} x(k) + q(k)
\]

(10.17)

which is again a standard state-space model.

Anderson [4] describes different augmented model configurations. A common application of the plant model augmentation, is to add integral action to the estimator. When it is assumed that the noise is a Gaussian random signal with a non-zero mean the noise model Eqn. 10.15 is specified with \( \Phi^d = I, \Gamma^d = I, q(k) = 0 \), thus:

\[
x^d(k+1) = x^d(k) + w(k)
\]

(10.18)

For this case the original plant is augmented with an integrator, which will effectively compensate the static disturbance, and therefore does not bias the estimated state. Instead of a proportional feedback, a proportional integrating estimator is thus created, that estimates both the plant states and the deterministic part of the disturbances.

Maciejowski [146] points at a complication with this particular augmentation: the solution to the Ricatti equation only exists provided that the plant is stabilizable and detectable, which is clearly violated in this specific case. A solution to this problem is to place the poles not exactly at the unit circle but just inside. A more common solution to obtain a stabilizable and detectable model, which is standard in LQ and predictive control is to difference the model Eqn. 10.17. This results in:

\[
\Delta x(k+1) = \begin{bmatrix} \Phi & 0 \\ 0 & \Phi^d \end{bmatrix} \Delta x(k) + \begin{bmatrix} \Gamma \\ 0 \end{bmatrix} \Delta u(k) + \begin{bmatrix} I & 0 \\ CA & I^d \end{bmatrix} \begin{bmatrix} w(k) \\ p(k) \end{bmatrix}
\]

(10.19)

\[
y(k)(k) = \begin{bmatrix} C \\ C^d \end{bmatrix} \Delta x(k) + q(k)
\]

(10.20)

This form, which employs \( \delta u \) instead of \( u \), is denoted as the incremental form, has the added advantage that the step size \( \Delta u \) can be penalized in the criterion easily. This is important because the maximum rate of change of an actuator within one control interval is limited.

Dual to the estimator design, for which the model was augmented to obtain a desired response to disturbances, we may wish to augment the model used for state feedback to enhance the controlled response to setpoint changes. Usually, only integral action is added similar as in the estimator case to remove static offsets of the process output from the setpoint.
10.2.2 Model predictive control

Model predictive controllers employ the receding horizon form of LQ control. However, the standard criterion Eqn. 10.9 is extended with a set of constraints to yield the following nonlinear minimization problem:

\[
J(t) = \min_{\Delta u(k), \ldots, \Delta u(k+N-1)} \sum_{j=k+1}^{k+P} \left[ \hat{y}(j) - r(j) \right]^T Q \left[ \hat{y}(j) - r(j) \right] + \Delta u(j-1)^T R \Delta u(j-1)
\]

subject to the following linear inequality constraints:

\[
u(k) \geq u_{\text{min}}(k)\]

\[
u(k) \leq u_{\text{max}}(k)\]

\[y(k) \geq y_{\text{min}}(k)\]

\[y(k) \leq y_{\text{max}}(k)\]

\[|\Delta u(k)| \leq \Delta u_{\text{max}}(k)\]  

(10.21)

(10.22)

(10.23)

(10.24)

(10.25)

(10.26)

Thus on sample time \(k\) we seek a sequence of input moves \(\Delta u(i)\) over \(i = k, \ldots, k + N - 1\) instants in the future that minimize the criterion Eqn. 10.21 over \(P\) samples in time. Generally \(N\) is denoted as the control horizon and \(P\) is the prediction horizon. The matrices \(Q\) and \(R\) are diagonal weights that have the same meaning as in the standard LQ criterion (cf. Eqn. 10.9). Only the first move is implemented and at the next control instant the minimization is repeated. The parameters involved are further explained in Fig. 10.4.

Note that the constraints are time-varying and thus adaptable from the operator console, if required. Another point to note is, that other non-quadratic criterion functions may be used as well.

To find the solution to this criterion, the process output trend \(\hat{y}\) should be computed. This value is given by

\[
\hat{Y} = H \mu(k) + Y_x \hat{x}(k|k - 1) + Y_d \hat{d}(k|k)
\]

(10.27)

where \(\hat{x}(k|k - 1)\) denotes the estimate value of \(x\) at sampling period \(k\) based on information at sampling period \(k - 1\). Three different terms are distinguished in this equation.

1. The forced response This is the effect of present and future manipulated variable moves. The future manipulated variables are given by:

\[
\mu(k) = [m^T(k|k)m^T(k + 1|k), \ldots, m^T(k|k + N - 1)]
\]

(10.28)
and the matrix $H$ is the pulse response matrix given by:

$$H = \begin{bmatrix}
H_1 & 0 & \ldots & \ldots & 0 \\
H_2 & H_1 & 0 & \ldots & 0 \\
H_1 & \ldots & \ldots & \ldots & \ldots \\
H_{N-1} & H_{N-2} & \ldots & H_1 & 0 \\
H_N & H_{N-1} & \ldots & \ldots & H_1
\end{bmatrix}$$

(10.29)

where the matrices $H_i$ are given by:

$$H_k = C\Phi^{k-1}\Gamma$$

(10.30)

2. The free response. This the effect of past values of the manipulated variables on the process output. As argued in [142, 171, 208] applying an open-loop model to generate the contribution of the past inputs results in a standard IMC control structure, which requires a stable open-loop plant model to achieve a stable controller (cf. Fig. 10.2). Introduction of a closed-loop observer introduces an additional degree of freedom to shape the response to disturbances independently. In addition, when the feedback gain is properly designed it can be used to stabilize
an unstable or boundary stable internal model. In this way MPC including a state estimator can be applied to unstable processes as well. From the estimated state \( \hat{x}(k|k-1) \) the free response is calculated from:

\[
Y_x = [(C\Phi)^T (C\Phi^2)^T \cdots (C\Phi^N)^T]^T
\] (10.31)

In the semi-nonlinear MPC controllers proposed by [82, 85] this contribution is calculated with the nonlinear model.

3. **Future disturbances**. These are generally unknown and assumed to be constant:

\[
\hat{d}(k+i|k) = \hat{d}(k|k) \quad \text{for} \quad i = 1, \ldots, N
\] (10.32)

This is also necessary to remove steady-state offset. The matrix \( Y_d \) is given by [208]:

\[
Y_d = [(C K)^T (C(\Phi + I)K)^T \cdots (C(\sum_{k=1}^N \Phi^{k-1}K)^T)^T
\] (10.33)

With the linear model, that is used to compute the forced response, the minimization of Eqn. 10.21 results in a so called quadratic programming (QP) problem for which algorithmic solutions are available [207]. When constraints are not taken into account, the minimization of the quadratic criterion can be done in one step.

Details on MPC algorithm including state space models are given by [208], on standard DMC employing step response models by [188], on nonlinear QDMC by [82], and on nonlinear QDMC including a state estimator by [85].

### 10.2.3 Computation scheme

The MPC controller including a state-estimator is used for control of the crystallizer. The following computation scheme has been applied:

**Step 1:** Linearize the simulation model (see Section 2.3.7).

**Step 2:** Reduce the continuous time model. For high dimensional state-space systems \( n > 25 \) the control computational burden and the numerical condition of the problem will prevent on-line use of the model. Therefore the model is reduced to according the method described in Section 2.3.8

**Step 3:** Discretize the reduced continuous time model. This yields a discretized state-space model \( \{\Psi, \Gamma, C\} \) [9]. An important design choice that has to be made at this point is the control interval \( \Delta T \). As a rule of thumb this time time is taken 10 times smaller than the smallest open-loop time constant in the system [8].

**Step 4:** Compute the Kalman Filter gain.

**Step 5:** Calculate the free response using Eqn. 10.31.
Step 6: Predict the output. The predicted output is computed as the sum of the effect of past and future manipulated variables and the future predicted disturbances.

Step 7: Calculate M future manipulated variables by solving QP. If constraints are absent, this solution can be written in the form of a state space equation. An advantage in this case is that all classical robustness analysis techniques apply. When input/output constraints are set to tight the optimization problem may be infeasible. If this problem is structural the design of the process should be changed. For output constraints a way to overcome this problem is to apply constraint "softening".

Step 8: Apply inputs to the process over one control interval and collect the output response. If the controller is tested with the simulation model the outputs of the plant at the next control instance are calculated by integrating the model \( \dot{x} = f(x,u) \) over one control interval. The inputs are kept constant over this interval.

Notice that the first three steps can be skipped when a system identification procedure as described in Chapter 6 is applied. Another point to note is that the first four steps are executed only once if the process model is not time-varying. This is a valid assumptions for most continuous process. If, however, a trajectory is followed, as in many batch reactors it may be necessary to integrate the first four steps in the control loop as well. The repeated adaptation of the model then accounts for possible changing process dynamics along the trajectory that is followed. Such a scheme is described by [85].

The described procedure has been applied to the continuous crystallizer as described below. The steps 5, 6 and 7 are computed using standard tools present in the Matlab MPC toolbox [163]. An overview of the main M-files used is given in Appendix E.

**Tuning of MPC controller**

For the presented procedure the main tuning parameters are:

**Diagonal weighting matrices \( Q \) and \( R \) (cf. Eqn. 10.21)**. These have the same meaning as in the standard LQ criterion refeq1qcon.

**State estimator feedback gain \( K \)**. When exact knowledge is present on the noise terms an optimal estimator feedback gain is obtained. However, often one treats the noise covariance matrices as design parameters that are adapted until a sufficient speed of response on errors is obtained. Ricker [208] provides some ad-hoc rules of thumb, which can be used to determine the feedback gain. Generally increasing the state estimator gain will increase the speed of response to disturbance, however, it will also increase the sensitivity to measurement noise.
The control horizon $N$. Increase the control horizon to infinite should yield the LQG solution. If inverse response characteristics are present the horizon should sufficiently exceed the negative part of the response.

The prediction horizon $P$. Increasing the control horizon to infinite should yield the LQG solution. If inverse response characteristics are present the horizon should sufficiently exceed the negative part of the response.

The number of control moves $k$. The number of moves strongly determines the time needed to calculate a solution. So called blocking can be applied as suggested by Ricker [207]. With blocking the number of independent future manipulated variable moves is limited by keeping them constant over a block of sampling intervals. For the crystallizer control applications it is found that blocking decreases the computational load and that varying only two or three blocks of manipulated variables over the control horizon is generally sufficient to gain the desired response characteristics. In [163] it is shown that a large control horizon without blocking can cause a large variance in the manipulated variable — an effect that is known as "ringing".

In Garcia and Morari [79] some additional guidelines are presented for tuning of IMC controllers.

Robustness analysis

The controller designed on the basis of a single nominal model may exhibit excellent performance. However, due to inevitable plant model mismatch the same controller can have unacceptable performance or even be unstable when implemented to the plant. In general we have to detune the controller, that is, lower its nominal performance, by changing the weights as described in the previous section, to obtain sufficient robustness for modelling errors.

To analyze the robustness the model uncertainties have to be represented in a suitable manner. Uncertainty representations are generally subdivided into structured and unstructured uncertainties. An example of an unstructured uncertainty is a certain phase or gain margin. Structured uncertainties are for instance the uncertainties in the parameters as represented in Table 9.1.

Here the set of possible frequency responses of the return ratio of the plant, are used as an uncertainty representation. This set of frequency responses is being generated by varying the uncertain model parameters over the ranges which determine their extend of uncertainty. If this set of frequency response is displayed (for instance as Nyquist curves) each element of the nominal frequency response is surrounded by a region in which all possible responses are located. Such regions are often denoted as uncertainty templates [146].

This way of representing the uncertainty is straightforward, however, as argued by [146], a possible problem is that the bounds of the parameters do generally not coincide
with the bounds of the uncertainty regions in the frequency domain. Therefore, both the boundary values and nominal values of the parameters are taken into account here.

To check whether a designed feedback controller is robust in the face of these uncertainties, the classical Nyquist stability criterion has been applied. For a SISO controlled plant, which is open-loop stable the Nyquist curve of the return ratio $G(s)$ should not encircle the point (-1,0) in the complex plane. In the MIMO case the generalized Nyquist criterion is applied. If the open-loop plant is stable, the generalized nyquist criterion states that the closed-loop system is also stable, if the so called characteristic loci do not encircle the point (-1,0) in the complex plane. The characteristic loci are the graphs of $\lambda_i(s)$, where $\lambda_i(s)$ is the $i$th eigenvalue of $G(s)$, (cf. [146, p. 61]). We assume that constraints do not become active.

A more profound method exists for robustness analysis which is known as $\mu$-analysis [54]. To perform this analysis both structured or unstructured uncertainties are pulled out of the system and represented in a separate block. If an accurate uncertainty description is available, this analysis leads to a non-conservative guess of the robustness of the plant. Pretti and Garcia have applied this to a heavy oil refractionator [188, p. 146]

Hakvoort [94] and De Vries [48] recently developed methods to estimate an uncertainty model from plant input-output data directly.

Tools present in the Matlab MPC-toolbox have been used here to calculate the frequency response of the return ratio of the applied MPC controller, In addition to frequency domain analysis, also time domain responses to a disturbance have been simulated for the set of possible plants in closed-loop using the designed controller.

### 10.3 MPC applied to crystallization processes

The MPC scheme including a state estimator as presented in the previous section is applied for control of the crystallizer. The designed controller is tested for its ability to stabilize the process, to reject disturbances and to follow setpoint changes. A comparison with a standard multi-loop PI controller will be made.

On the basis of controllability analysis discussed in Chapter 8 three different control configurations are selected. These are depicted in Fig. 10.5. Configuration a and b will be discussed first. These configurations have been evaluated with the pilot crystallizer as well. The latter has been evaluated by simulation only. First the results of two open-loop experiments are described, which are used subsequently to study the disturbance rejection capability of the designed controllers. Both the linearized, reduced and discretized nonlinear model as well as the identified input-output model have been used as internal models. The robustness of the controller is evaluated by considering the parameters: \{p_1, p_3, p_{f1}, p_{f2}, p_{f3}\} as uncertain parameters (see also Table 9.1). From the analysis performed in Section 9.3 it is found that these parameters have the largest influence on the controller stability and performance. The nominal
10.3 MPC APPLIED TO CRYSTALLIZATION PROCESSES

Fig. 10.5: Three input/output configuration for which an MPC controller has been devised.

process conditions applied during both experiments and simulations are summarized in Appendix D. Appendix E summarizes the main software tools used.

10.3.1 Open-loop plant behaviour

To study the disturbance rejection capabilities, the effect of two plant disturbances on the open-loop plant dynamics are considered.

1. blockage of the product removal flow. In Fig. 10.6 the process response to a blockage of the product discharge flow for one hour is depicted. After 18 hours this disturbance is repeated. As expected the magma density increases linearly after closing the valve because the process runs in batch mode while the heat input is not affected. After releasing the valve an exponential response is observed back into the direction of its value before the disturbance. The other two strips of this plot reveal that also the CSD is strongly disturbed. The signal $y_{r1}$ reduces in the first stage after the disturbance because the removal of particles is stopped, and the supersaturation decreases. After releasing the product discharge line this quantity increases again because an additional number of fines are created due to the increased magma density. The same mechanism explains the dynamics of the median crystal size. The free process response is dampened in between time B and C in this plot with a simple proportional controller, that was described in Chapter 9 to create a sufficient stationary condition at the start of the second disturbance. This was necessary to limit the duration of the experiment.

2. water injection The second disturbance was a pulse wise injection (injection
Fig. 10.6: Experimental open-loop response on two successive disturbances of the product flow (A and B). Stabilizing controller is used between B and C and after D.

time was < 5min) of 40 litres water into the system, while it operated at a stationary condition. The temperature of the water is equal to the slurry, further no significant disturbance of the basic process quantities was observed. In industrial practice the injection of water frequently occurs to remove salt incrustated on the crystallizer internals, or to free blocked slurry handling systems such as centrifuges. The nominal solids fraction in the slurry of the crystallizer is approximately 11%. Using the physical properties listed in Appendix D, injection of 40 litres in a volume of 1 m$^3$ causes a decrease of the solids fraction to 8.5%. The main effect of this disturbance is that the smaller particles directly dissolve and the magma density drops directly —provided that the system is sufficiently well mixed. In Fig. 10.7 the response on the median crystal size, the signal $y_1$, and the magma density are given. Clearly, water injection affects both the mass and the shape of the CSD. The responses indicate that an oscillation is induced which damps after approximately 12 hours. However, after $t=7$ hours problems occurred with the process computer. This caused a non-quantified additional disturbance to the process —the temperature, the level, and some other basic process quantities were upset for at least 1 hour— which obscures the signal
Fig. 10.7: Experimental open-loop disturbance response on 40 litres pulse wise injected water A; effect of undesired disturbance from process computer after B.

trend after \( t = 7 \) hours.

10.3.2 Design of SISO fines removal based controller

At first case the simple SISO MPC controller which uses \( y_{r1} \) as its input and the fines removal flow as its output (a) in Fig. 10.5, is designed. In Fig. 10.8 the Nyquist contour plot of the return ratio of the tuned 5th order MPC controller is given. In the same plot a PI controller is plotted which has the same phase margin as the MPC controller. The internal model is a fifth order model whose input and output have been scaled with the values given in Table 8.4. The tuning parameters have been chosen: \( Q = \text{diag}(100), R = \text{diag}(50) \) (cf. Eqn. 10.21), and the prediction horizon and the control horizon are both 2 hours, however, the 24 values for the manipulated variable are blocked to 4,8 and 12 blocks, respectively.

As can be seen the largest differences occur in the low frequency range. In Fig. 10.9 the closed-loop response of both controllers on a disturbance in the product removal flow (see previous subsection) is given with the open-loop response. The results show that the disturbance is effectively suppressed. Furthermore, both the PI and MPC
controller have a comparable closed-loop responses, however, the response of the MPC controller is slightly more tight. For the response to setpoint changes a similar observation is made. The sensitivity to measurement noise is the same for both designs.

In Fig. 10.11 the Nyquist curves of all 243 return ratios are given that are estimated by perturbing 5 uncertain parameters in the plant model. The results show that the stability margins are not violated and also that the performance is not affected significantly. Consequently we may conclude that the controller is robust for uncertainties in the described parameters.

**Manipulated variable Constraint handling**

MPC is chosen for its capability to handle constraints explicitly. However, this does not mean that simple control PI controller cannot be applied when input or output constraints are present. The constraint handling capabilities of MPC have been compared to the PI controller. To prevent so called *wind-up* [220] of the PI-controller, accumulation of the integrator is stopped when the constraint becomes active. The results are depicted in Fig. 10.13. As can be seen the PI controller still has a comparable performance. Consequently, also for its ability to handle constraints the MPC controller has not stronger performance.
Fig. 10.9: Responses in time domain on disturbance in product removal flow for open-loop system (dotted) and closed-loop with a 5th order MPC controller with state estimator (solid) and a PI controller (dashed).

Fig. 10.10: Region within which all possible closed-loop responses, obtained with a 5th order MPC controller for 243 different plant models, are located.
**Fig. 10.11:** Nyquist curves corresponding to return ratio using a 5th order MPC controller for different perturbed plants.

**Fig. 10.12:** Sensitivity and complementary sensitivity of 5th order MPC controller for different perturbed plants.
Fig. 10.13: Simulated closed-loop responses of 5th order MPC controller and PI controller with a manipulated variable constraint.

Role of state estimator

In Fig. 10.14 the standard DMC solution (\( K = 0 \)) is compared to the optimal tuned MPC solution (\( K \neq 0 \)). Clearly with the standard DMC solution the open-loop poles are present in the feedback loop and limit the systems speed of response to the response time related to the slowest dynamics. The two degrees of freedom structure provided by the MPC approach allow for independent tuning of the state response and the sensitivity for noise. Hence we may conclude that the standard DMC performs much worse than a PI controller and the MPC controller including a state estimator.

Experimental evaluation of SISO MPC controller

During an experiment that lasted 50 hours, the closed-loop performance of this controller is first tested for the open-loop product flow disturbance. In Fig. 10.15 the closed-loop responses on \( y_1 \), the median crystal size and the magma density are given. Note that the controller does not affect the magma density response. In Fig. 10.16 the controller output signal is given. During this experiment a lower actuator constraint is maintained at 0.3 l/s. Clearly the constraints are active at a large part of the run. The closed-loop output responses show that the disturbance is effectively suppressed.

At the end of RUN53 a small experiment has been conducted to test its response to a step change in the setpoint from -0.5 to -0.9. Fig. 10.17 the effect of this step on the median crystal size and the output are depicted. As can be seen the setpoint change is tracked rapidly and smoothly. The controller is able to reach the setpoint within one hour without overshoot. The median exhibits a delayed response. It starts to rise
Fig. 10.14: Simulated closed-loop response with standard DMC controller with different gains.

Fig. 10.15: Experimental closed-loop response with MPC on $y_1$, median and magma density (RUN53), for two disturbances at A and B and a constant controller setpoint (dash-dotted).
after the step, it did just reach a steady at the end of the experiment. As expected the magma density is not affected.

10.3.3 Design MIMO controller

Applying SISO control to the signal \( y_{r-1} \) effectively controls CSD properties, however, the magma density is not affected. Therefore, a MIMO controller is designed that affects both the magma density and the CSD. The process to be controlled is depicted in Fig. 10.5 As was found in Chapter 8 this system is easily controllable. The interaction between inputs and outputs as expressed by the RGA is low. The first input solely influences the first output, while the second input mainly influences the second output and in the high frequency range also the first output.

**Tuning and robustness of MIMO controller**

For design of the two input two output controlled system two different internal models are applied:

1. A reduced order linear version of the first principles model, which is derived according to the method described in Section 2.3.8,

2. An input-output model that is estimated directly from process input and output data measured under closed-loop conditions.

The latter model is expected to give a more accurate representation of the dynamics to be controlled. The use of identified input-output models has an appeal for certain industrial applications where sufficient accurate first principle models are absent. This route is especially attractive when applied in conjunction with MPC as it is relatively easy to tune. A disadvantage, which was already addressed in a previous section is that a non-conservative uncertainty description cannot be obtained easily. In Fig. 10.21 the simulated response of the 2 input 2 output controller is given. The internal model is
Fig. 10.17: Experimental step response (at A) on controller setpoint (dash-dotted) for $y_1$, the median crystal size, and the magma density.

Fig. 10.18: Experimental response (at A) of controller output after step change in setpoint.
Fig. 10.19: Bounds of regions in which the two characteristic loci for the two input two output system are located.

scaled with the constant values given in Table 8.4. Using this model the following tuning parameters have been chosen: $Q = \text{diag}(100100)$, $R = \text{diag}(5050)$ (cf. Eqn. 10.21), the prediction horizon and the control horizon are both 2 hours, however, the 24 values for the manipulated variable are blocked to 4.8 and 12 blocks, respectively.

The characteristic loci of the possible return ratios are given in Fig. 10.19. As can be seen from this plot the generalized Nyquist stability criterion is not violated.

Comparison with multi-loop PI-control

In Fig. 10.20 the MIMO MPC response is compared with the response of the multi-loop PI controlled process. Surprisingly a similar response is obtained, however, the constraint handling is superior.

Experimental evaluation of 2in-2out controller, using first principles model

Two sets of MIMO experiments are performed with the process configuration depicted in Fig. 10.5. Fig. 10.22 depicts the response of the crystallizer, controlled on the basis of a first principles model as internal model, for the disturbances discussed in Section 10.3.1 and a step change on the setpoint of the product magma density. Because the product flow was kept constant the latter step directly influences the mass production rate. In between 5 and 6 hours after startup some undesired disturbances occurred in the heat supply system which caused a reduction of the effective heat in-
Fig. 10.20: Comparison of multi-loop PI controlled response (dotted) and MPC response (solid); inputs: $Q_f$ and $P(k)$ (left); outputs: $y_{r1}$ and $m_3$ (right).

Fig. 10.21: Comparison of multi-loop PI controlled response (dotted) and MPC response (solid); inputs: $Q_f$ and $P(k)$ (left); outputs: $y_{r1}$ and $m_3$ (right).
Fig. 10.22: Experimental response of process outputs for 2in 2out MPC controlled process, on the basis of first principles model, for startup, blockage of the product flow (A), injection of 40 litres water (B), and a step change in the setpoint (dashed) for the product magma density (C)

put with -40 KW during 30 minutes. However, this had a minor affect on the desired response.

In Fig. 10.23 the corresponding process input signals are given. As can be concluded the response is satisfactory. Control of the CSD properties is achieved with comparable performance as in the case of the SISO controller (see Fig. 10.15). However, the disturbance on the magma density is strongly suppressed in this case. Near the end of the run (C) the setpoint of the magma density is increased from 1305 kg/m$^3$ to 1320 kg/m$^3$, which is equivalent to an increase of the mass production rate with 40 kg/hr. from 147 kg/hr. to 187 kg/hr. Note that this disturbance drives the actuator to its upper constraint. The response on the response on the CSD is effectively suppressed by the controller.
Fig. 10.23: Experimental response process inputs for 2in 2out MPC controlled process, on the basis of first principles model, for startup, blockage of the product flow (A), injection of 40 litres water (B), and a step change in the setpoint (dashed) for the product magma density (C), with actuator constraints (dash-dotted).

Experimental evaluation of 2in-2out controller, using identified model

Fig. 10.24 depicts the output response of the crystallizer, controlled on the basis of an identified model used as internal model, for the two disturbances. Also for this case the product flow was maintained at a constant value. In Fig. 10.25 the corresponding inputs are given. Comparing these figures with Fig. 10.22 and Fig. 10.23 reveals that the response are even more tight. However, note that the different weighting that is applied may be responsible for this, rather then the improved accuracy of the model.

An important fact is that no simulation study was done with the controller designed. As can be seen in the first stage of the response between A and B after switching on the controller, oscillatory behaviour is induced in the closed-loop because the initial diagonal elements of Q and R (see Eqn. 10.21) were not appropriate. Therefore, we detuned the controller in the first five hours of the response by increasing the input penalty with a factor of 2 (roughly) and reducing the output weighting also with a factor of 2 (roughly), while observing the process behaviour. A desired closed-loop response could be obtained in this way. Hence this procedure shows that, in case no strong requirements with respect to safety exist, on-line tuning of the MPC controllers can be very effective and easy to perform.
10.3 MPC applied to crystallization processes

![Graph](image.png)

**Fig. 10.24:** Experimental response of process outputs for 2in 2out MPC controlled process, on the basis of identified model for startup (A), injection of 40 litres water (B), and a blockage of the product flow (C), for a constant setpoint (dashed).

### 10.3.4 Control of three input three output process

The last configuration which was verified experimentally is the three input three output plant (see Fig. 10.5). Compared to the previous case now the product flow is added as an input and the supersaturation is added as an output. In Chapter 8 we have shown that also this process is relatively easy controllable however a significant interaction is present.

In tuning this controller a relatively large weight is applied to the supersaturation. In Fig. 10.26 the responses of the plant inputs and outputs are depicted for a disturbance in the product removal rate for the open-loop plant, the MPC controller and the PI controller. Comparison with the previous plots reveals that the responses of the PI and MPC controller are different. In effect the interaction between the heat input and the supersaturation is used to lower the decrease of the supersaturation supersaturation. Hence for this specific case we see that the use of a multivariable controller is meritorious compared to a multi-loop controller.

In the simulation we have assumed that the supersaturation is directly measurable.
Fig. 10.25: Experimental response of process inputs for 2in 2out MPC controlled process, on the basis of identified model for startup (A), injection of 40 litres water (B), and a blockage of the product flow (C), for a constant setpoint (dashed), with actuator constraints (dash-dotted).

Fig. 10.26: Comparison of open-loop and closed-loop response of MPC controller using three inputs (left) and three outputs (right).
In practice this will be difficult to achieve, therefore it should be inferred from the CSD sensor and magma density measurements. This has been described in Chapter 7.

10.4 Perspectives for batch crystallizer control

With batch crystallization a certain level of supersaturation is maintained, during the so called batch time, to grow an initial population of (seed) crystals to their final product specifications. In some cases when an additional feed is maintained the process is called to operate semi-batch. Although the operation seems rather uncomplicated the industrial practice shows that the final distribution of crystal sizes does often not agree with the product specifications. For example the spread of the product CSD may be too large or the mean size is unacceptable low. In addition the batch time needed too achieve the product quality may be too large.

In the past twenty years a lot of research has been done in the field of optimization and control of batch crystallizers. Control strategies for batch crystallizers mainly focus on influencing the supersaturation level during the batch program, in order to optimize the final product specifications. For batch cooling crystallization basically three different modes of operation exist: natural cooling, linear cooling and controlled cooling.

With natural cooling the transient temperature is ruled by Newtons law of cooling: The cooling rate is proportional to the excess temperature. In case of linear cooling, the temperature decreases linearly, and with controlled cooling the transient temperature is steered along a set point temperature profile. These policies are open-loop control policies, because there is no feedback from the actual variable (or state) to be optimized to the inputs of the process.

The main, often conflicting, objectives of the open-loop policies are: to achieve optimal final CSD characteristics by keeping the supersaturation level within the metastable zone width (i.e., the area under the line in the supersaturation-temperature diagram that bounds the region of excessive nucleation), and to keep supersaturation, and thus the growth rate, as high as possible to obtain a maximum production rate.

Many authors have addressed the problem of calculation of optimal cooling profiles [2, 33, 103, 125, 124, 123, 157, 159, 167, 173, 212, 250]. Most authors use simplified models and aim at reduction of the secondary nucleation rate, by keeping the supersaturation at a predefined level. A review of early attempts in this field is given by Jones [123]. The resulting trajectories implied on the process resulted in most cases in an improved final product CSD. A great disadvantage, however, is that the criteria to be optimized are often not directly stated in terms of economic cost or product specifications.

An improvement was attained with optimal control theory which exploits the Pontryagin Maximum principle to maximise an economic performance criterion, explicitly. As discussed by Jones [123], Ajinkya and Ray [2] and Morari [159] the resulting pro-
files lead to a further improvement of the product. In [157] optimal control theory is applied and the influence of uncertainties in the estimated model parameters on the estimated cooling trajectories is analyzed. Typically the optimal temperature trajectory for these approaches show a slow reduction in the early stage of the batch program. A low crystal surface area results in a low deposition rate and therefore in a high sensitivity of the supersaturation and nucleation for temperature changes. Towards the end of the program the temperature will reduce more rapidly, as an increasing crystal surface area now causes a low sensitivity of supersaturation and nucleation for temperature changes. A similar strategy can be developed for an evaporative crystallizer. Some authors discuss the effect of fines destruction on the final CSD properties as well [124, 203].

A disadvantage of open-loop approaches is that there is no feedback compensation is added to compensate for errors related to improper seeding, and alternating purity or solvent composition which may cause unacceptable batch to batch variations. These deficiencies can be alleviated by the MPC including a nonlinear state estimator which was introduced Section 10.2. As feedback compensation is not applied in the literature a short example will be given. More results on this topic may are expected from the UNIAK-3 research program [22].

10.5 Conclusions

The aim of this chapter has been to study the merits of advanced model based control for crystallization processes. As a tool model predictive control including a state estimator has been used. For analysis of the performance and robustness of the closed-loop system a linearized model has been used.

The results given in this chapter give an incentive for using simple PI controllers instead of advanced model based controllers for the control of a crystallization process. For SISO systems a PI controller acting on the same input and output has a similar performance as the MPC controller even when constraints are active. The PI controller is tuned to have the same phase margin as the MPC controller. An MPC controller without a state estimator, representing a standard DMC controller, was found to have a much worse performance than the standard PI controller.

When applied to a two input two output plant with small interaction the MPC controller was found to have a slightly better performance than a multi-loop PI controller. This is mainly because the MPC controller takes into account the process interaction implicitly. However, the decentralized PI controller is much simpler and is therefore presented as a good alternative. For experimental evaluation of this MIMO plant, both a simple identified process model and a linearized and reduced first principles model have been used as internal models. As expected the identified closed-loop model performs slightly better.

Both the robustness of the SISO and MIMO plant have been investigated in both
the time and the frequency domain. It is found from this that the designed MPC controller is robust for uncertainties in the parameters related to fines removal and nucleation and growth kinetics.
Chapter 11

Conclusions and perspectives

11.1 Conclusions

The main result of this thesis is that decisive evidence is given, on the basis of experimental results, for the fact that effective control of CSD dynamics can be achieved, using an actuator and a simple sensor that both operate in the fines region, provided that the detection location of the sensor is chosen properly.

With respect to the latter condition it is found that neither the type of controller, nor the controller parameters, nor the complexity of the sensor, but merely the crystal size range at which the sensor is focused is the major parameter that determines closed-loop stability and performance.

Experiments have been conducted with a 970 litres DTB crystallizer using ammonium sulphate as a substance system. This crystallizer is equipped with a fines removal system and a vibrating screen to achieve product classification. Accurate measurement of CSD dynamics is achieved with a sensor based on forward light scattering in combination with a continuous slurry diluter. A simple particle counter is found to provide sufficient information on the control relevant dynamics.

From lengthy pilot plants runs the following important insight is gained on the dynamics of crystallization processes.

- Both fines removal and product classification promote CSD cycling, while increasing the fines flows and lowering the classifier feed flow intensifies cycling.

- Right after the onset of primary nucleation at unseeded startup significant agglomeration may occur when the heat input exceeds a certain critical value. For the pilot crystallizer agglomeration occurred for heat inputs larger than 80 KW.

- The time period of CSD oscillations is proportional to the mean residence time of slurry in the crystallizer.
11.1 Conclusions

- Heat input variations only affect CSD dynamics at higher frequencies.

- With product classification additional interaction is introduced between the CSD and the magma density and thereby the mass production rate. The time period of cycles in these quantities equals the CSD cycle time.

- Due to settling of particles significant segregation may occur in the crystallizer vessel.

- When applying product classification with a vibrating screen the fines density drops significantly, which indicates a reduced secondary nucleation rate.

- The reproducibility of startup experiments is good.

Rigorous process models have played an important role in further analysis of the experimental results and the design of controllers. We have shown that a macroscopic population balance model, including models for crystal birth and growth, reasonably describes the process dynamics. For proper validation of the model, explicit modelling of the sensor and the particle shape is found to be important. In verifying the model, using experimental data from the pilot crystallizer, the following conclusions are drawn:

- Describing secondary nucleation as a function of the surface area of particles with a size larger than a certain critical size, which is found to be 500 \( \mu m \) for ammonium sulphate, effectively describes CSD cycling.

- Errors in the parameters related to crystal growth and crystal birth mainly affect the low frequent dynamics of the CSD.

- When product is removed unclassified and the stirrer speed is kept constant a simple first order model reasonably describes the dynamics of the solids phase.

- Supersaturation has a low influence on the secondary nucleation rate.

- The initial distribution that exists after the primary nucleation phase at unseeded startup, can be described with a Rosin-Rammler distribution having three degrees of freedom.

In conjunction with process modelling we have shown that the desired model accuracy depends on the intended use of the model. When the model is used solely for stabilizing feedback controller design, accurate estimates for the kinetics are not necessary. On the other hand for process design or observer design often more detailed models with a larger accuracy are needed.

System identification is applied for verification of the process model, and as an alternative to process modelling as well. It is found that identification experiments to achieve input-output data, have to be conducted in a closed-loop fashion to prevent the process dynamics to be dominated by slowly cycling behaviour. The well known
problem of identification in a closed-loop could be circumvented by applying identification in two steps. The results show that low order linear models give a fairly accurate description of the process dynamics, when the process is operated close to its nominal behaviour.

An important application of the dynamic process model is to serve as a state estimator. With a state estimator, dynamic process data stored implicitly in a dynamic model is combined with on-line process information to reconstruct quantities that cannot be measured on-line or only with great difficulty. We have shown that the error feedback law, may be designed on the basis of a linear model and implemented to the nonlinear model to derive a nonlinear observer. Verification with experimental data revealed that the designed estimator performs well and has a large robustness for sensor failure and errors in the initial conditions.

Controllability analysis has shown that crystallizers in general have limited degrees of freedom. For the pilot crystallizer equipped with a fines removal and a product removal system, where the heat input to the crystallizer, the fines removal rate, and the unclassified withdrawal flow are considered as inputs, a single CSD parameter, the total mass flow rate, and the level of supersaturation can be controlled independently. The amount of interaction between these three quantities is found to be limited. Consequently, a control structure with which interaction is not taken into account explicitly may be sufficient.

With respect to controller design it is found that several design related questions, such as where to place the sensor, are important, and mainly determine the closed-loop performance that can be achieved. When simple PI-controllers are chosen we found that unstable behaviour occurs when the sensor is focused onto particles in the size range of 0.40 μm. In addition, focusing above 100 μm gives bad results as well even if the controller was tuned optimally. Stability and good disturbance rejection and rapid setpoint tracking is achieved with a SISO PI controller using the output of the simple particle counter, as its input, and the fines removal as its output. It is found that the particle counter is sensitive to CSD fluctuations in a range around 60 μm.

Analysis of the process and controllers gives a strong incentive for designing stabilizing feedback controllers directly on a heuristic basis. However, the use of a more advanced Model Predictive Controller may be legitimate, when very tight control within process input and output constraints is desired and when significant interaction between multiple process inputs outputs exists. Experimental results have shown that low-order models derived from both the first principles model and the closed-loop system identification procedure, give good closed-loop performance when they are used as internal model in the MPC controller.
11.2 Perspectives

The work presented in this thesis has provided a good basis for implementation of (feedback) controller for the CSD and the supersaturation in industrial crystallizers. A large challenge that remains for process industry and sensor suppliers is to develop and install a cheap and simple sensor, with a properly known and unchanging particle detection characteristics, with a large robustness for operational condition such as soluble and insoluble foreign material and with low maintenance requirements.

For more fundamental research there are numerous other issues which pose new challenges. These are located more in the field of equipment design and control of other properties such as morphology and crystal purity, which were only touched upon in this work. In addition, if accurate models for particle shape and purity are desired also models for mixing both on a micro and macro scale will be necessary.

For verification of more detailed models, more accurate sensors are needed. For this image analysis is expected to have good perspectives. In particular for on-line detection of particle shape, this technology is attractive. In line with this, the development of robust image analysis schemes is also seen as a major challenge.

When models are made more detailed, generally their complexity and dynamic order will increase as well. For the purpose of analysis and controller design, the allowed model complexity is limited, hence the topics of model reduction and model linearization will remain important. In particular the development of reduction methods that preserve the main nonlinearities is important. With respect to model linearization, modern tools available in flowsheeting packages may give ambiguous or wrong solutions when applied to highly dimensional (lumped parameter) models. Because analytic solutions are too elaborate the development of robust linearization schemes is important.

Finally, the model based control schemes developed so far, are not validated experimentally for batch wise operation. In addition, it will be important to elaborate further on the development of a general control algorithm which can be customized easily for a wide variety of process configurations and substance systems. In particular for control of multi-purpose batch plants, such a controller may be profitable.
Appendix A

Population, mass and heat balances

A.1 General balance equations

All physical slurry flows introduced below, are defined per cubic meter of slurry. Except some slight modifications, the equation in this section are after De Wolf, (1990). Note that the solid phase is assumed to contain only a single pure component. Consequently, the density \( \rho_c \) instead of a concentration vector is used to denote the solids phase in the mass balances and the heat balance.

Population balance

The population balance describes the CSD dynamics in the crystallizer, [197]:

\[
\frac{\partial V(t)n(x,t)}{\partial t} + V(t)\frac{\partial G_c(x,t)n(x,t)}{\partial x} + Q_f(t)\gamma_f(t)h_f(x,t)n(x,t) \\
+ Q_{pf}(t)h_p(x,t)n(x,t) - Q_s(t)n_s(x,t) = 0,
\]

(A.1)

with the boundary condition:

\[
n(x = x_0, t) = B(t)/G_c(x_0, t)
\]

(A.2)

and the parametrized initial condition: \( n_s(x, t = t_0, \theta_S) \).

Crystal mass balance

Two additional equations are introduced that describe the solids mass production rates, determined by crystal birth (denoted as \( \Psi_1(t) \)) and crystal growth (denoted as \( \Psi_2(t) \)), respectively:

\[
\Psi_1(t) = \rho_c(t)k_vV(t)B(t)x_0^3
\]

(A.3)

\[
\Psi_2(t) = 3\rho_c(t)k_vV(t) \int_0^\infty G_x(x)x^2n(x,t)dx
\]

(A.4)
these equations are used to include the effects of crystal birth and crystal growth in the mass balances. The crystal mass in the crystallizer is described by the following equation:

\[
\frac{d}{dt} \left[ V(t) \left[ 1 - \epsilon(t) \right] \rho_c(t) \right] = \\
+ Q_i(t) \left[ 1 - \epsilon_i(t) \right] \rho_c(t) - Q_f(t) \left[ 1 - \epsilon_f(t) \right] \rho_c(t) \\
- Q_p(t) \left[ 1 - \epsilon_p(t) \right] \rho_c(t) + \Psi_1(t) + \Psi_2(t) \tag{A.5}
\]

The void fractions \( \epsilon \) are calculated according to Eqn. 2.7, and \( \rho_c \) is the density of the crystals. The crystal mass balance can be derived directly from the population balance, Eqn. A.1; consequently, it does not add new information to the model (De Wolf, 1990).

**Salt mass balance**

The dynamic equation describing the mass of salt (dissolved crystal material and crystals) present in the crystallizer is given by [49]:

\[
\frac{d}{dt} \left[ V(t) \left[ \epsilon(t)C(t) + [1 - \epsilon(t)]\rho_c(t) \right] \right] = \\
+ Q_i(t) \left[ \epsilon_i(t)C_i(t) + \left[ 1 - \epsilon_i(t) \right] \rho_c(t) \right] \\
- Q_f(t) \left[ \epsilon_f(t)C(t) + \left[ 1 - \epsilon_f(t) \right] \rho_c(t) \right] \\
- Q_p(t) \left[ \epsilon_p(t)C(t) + \left[ 1 - \epsilon_p(t) \right] \rho_c(t) \right] \\
+ Q_r(t)C_r(t). \tag{A.6}
\]

**Total mass balance**

The balance for the total mass (clear liquid mass and crystal mass) is given by [49]:

\[
\frac{d}{dt} \left[ V(t) \left[ \epsilon(t)\rho(t) + [1 - \epsilon(t)]\rho_c(t) \right] \right] = 
\]
A.1 General balance equations

\begin{align}
+ Q_i(t) \left[ \epsilon_i(t) \rho_i(t) + \left[ 1 - \epsilon_i(t) \right] \rho_c(t) \right] \\
- Q_f(t) \left[ \epsilon_f(t) \rho(t) + \left[ 1 - \epsilon_f(t) \right] \rho_c(t) \right] \\
- Q_p(t) \left[ \epsilon_p(t) \rho(t) + \left[ 1 - \epsilon_p(t) \right] \rho_c(t) \right] \\
+ Q_r(t) \rho_r(t) - Q_v(t) \rho_v(t), \tag{A.7}
\end{align}

where \( Q_v(t) \) is the vapour flow, and \( \rho \) and \( \rho_i \) are the density of mother liquor in the crystallizer volume and the feed flow, respectively.

**Heat balance**

The heat balance of the crystallizer is given by the equation [49]:

\[
\frac{d}{dt} \left[ V(t) T(t) \left[ \epsilon(t) \rho(t) c_{pl}(t) + [1 - \epsilon(t)] \rho_c(t) c_{pc}(t) \right] \right] =
\]

\[
+ Q_i(t) \left[ \epsilon_i(t) \rho_i(t) c_{pl}(t) + \left[ 1 - \epsilon_i(t) \right] \rho_c(t) c_{pc}(t) \right] T_i(t) \\
- Q_f(t) \left[ \epsilon_f(t) \rho(t) c_{pl}(t) + \left[ 1 - \epsilon_f(t) \right] \rho_c(t) c_{pc}(t) \right] T(t) \\
- Q_p(t) \left[ \epsilon_p(t) \rho(t) c_{pl}(t) + \left[ 1 - \epsilon_p(t) \right] \rho_v(t) c_{pc}(t) \right] T(t) \\
+ Q_r(t) \rho_r(t) c_{pr}(t) T_r(t) - Q_v(t) \rho_v H_v \\
+ P_{imp}(t) + P_{in}(t) + P_{cryst}(t) - P_{loss}(t), \tag{A.8}
\]

where \( T \) and \( T_i \) are the temperature in the crystallizer volume and the feed flow, respectively, the term \( P_{in} \) is the heat supplied at the internal heat exchanger, \( H_v \) is the specific enthalpy of vapour and \( c_{pc} \) and \( c_{pl} \) are the specific heat of crystals and mother liquor, respectively. \( P_{cryst} \) is the heat of crystallization (calculated from Eqn. 2.28), \( P_{loss} \) is the heat loss to the surrounding, \( P_{imp} \) is the heat supplied by pumps and stirrers, and \( P_{in} \) is the heat supplied with the internal heat exchanger.
Equations that relate the density, the specific heat and the heat of crystallization, to the temperature and the concentration, depend on a substance system that is crystallized. For ammonium sulphate these values are listed in Appendix D.

**Fines dissolver**

Assuming negligible holdup or delay in the fines removal section, the following two mass balances and heat balance are used to describe the ideal fines dissolver [49]:

\[
Q_r(t)\rho_r(t) = Q_f(t) \left[ \epsilon_f(t)\rho(t) + [1 - \epsilon_f(t)]\rho_c(t) \right]. \tag{A.9}
\]

\[
Q_r(t)C_r(t) = Q_f(t) \left[ C(t)\epsilon_f(t) + \rho_c(t)[1 - \epsilon_f(t)] \right]. \tag{A.10}
\]

The heat balance for the fines dissolver is given by:

\[
P_{ex}(t) = Q_f(t)\xi(t) \left[ T_r(t) - T(t) \right]. \tag{A.11}
\]

\(T_r\) is the temperature of the returned fines, and \(\xi\) a factor that is an estimated value of the specific heat of the fines flow. In the following it is assumed that this factor is constant.

**A.2 Derivation of simplified equations**

The above presented balance equations describe the general class of well-mixed, non-isothermal, non constant volume crystallizers. Additional assumptions can be introduced to obtain a simplified set of equations. This will be illustrated below for the case of the evaporative pilot crystallizer which is described in Chapter 3.

The main simplifying assumptions for this case are that the volume and the slurry temperature are constant and that the nuclei and are born at a negligible size: \(\Psi_1 = 0\). In addition it is assumed that the density of mother liquor does not depend on the concentration. As process input parameters, the fines flow, the product flow, and the heat input, are taken.

**A.2.1 Simplified balance equations**

A concentration balance is obtained by substitution of the crystal mass balance, Eqn. A.5, into the saltmass balance, Eqn. A.6. The concentration balance is given by:

\[
\epsilon(t)V\frac{dC(t)}{dt} = Q_i(t)C_i + Q_f(t)(\rho_c - C(t))[1 - \epsilon_f(t)] - Q_p(t)C(t) - \frac{\rho_c - C(t)}{\rho_c}\Psi_2(t). \tag{A.12}
\]

In this equation the term \(Q_r(t)C_r(t)\) is replace by Eqn. A.10.
Substitution of the crystal mass balance, Eqn. A.5, in the total mass balance, Eqn. A.7, results in the following equation for the feed flow \( Q_i(t) \):

\[
Q_i(t) = \frac{1}{\rho_i} \left[ Q_p(t)\rho + Q_v(t)\rho_v + Q_f(t)(\rho - \rho_c)[1 - \epsilon_f(t)] - \frac{\rho - \rho_c}{\rho_c} \Psi_2(t) \right] \tag{A.13}
\]

In this equation the term \( Q_r(t)\rho_r \) is replaced by Eqn. A.9.

A relation for the vapour flow \( Q_v(t) \) is obtained by substitution of the crystal mass balance, Eqn. A.5, into the heat balance, Eqn. A.8:

\[
Q_v(t) = \frac{1}{\rho_v H_v} \left[ + Q_i(t)\rho_i c_{pi} T_i - Q_p(t)\rho c_{pi} T - Q_f(t)\rho c_{pi} T + Q_r(t)\rho_r c_{pr} T_r \\
- Q_v(t)\rho_v H_v + T(\rho c_{pi} - \rho c_{pc}) \frac{\Psi_2(t)}{\rho_c} + P_{tmp}(t) + P_{in}(t) + P_{cryst}(t) - P_{loss}(t) \right] \tag{A.14}
\]

Substitution of Eqn. A.11, and Eqn. A.9 into Eqn. A.14 gives:

\[
Q_v(t) = \frac{1}{\rho_v H_v} \left[ P_{tot}(t) - Q_f(t)\xi(T_r - T) + Q_f(t) \left( \epsilon_f(t)(\rho - \rho_c) + \rho_c \right) c_{pr} T_r \\
+ Q_i(t)\rho_i c_{pi} T_i - Q_p(t)\rho c_{pi} T - Q_f(t)\rho c_{pi} T + T \frac{\Psi_2(t)}{\rho_c}(\rho c_{pi} - \rho c_{pc}) \right] \tag{A.15}
\]

Substitution of Eqn. A.15 into Eqn. A.13 eliminates the vapour flow from the set of equations and results in an equation for the feedflow:

\[
Q_i(t) = \frac{H_v}{\rho_i (H_v - c_{pi} T_i)} \left[ Q_p(t)\rho + Q_f(t)(\rho - \rho_c)[1 - \epsilon_f(t)] - \frac{\rho - \rho_c}{\rho_c} \Psi_2(t) \\
+ \frac{1}{H_v} \left[ P_{tot}(t) - Q_f(t)\xi(T_r - T) + Q_f(t) \left( \epsilon_f(t)(\rho - \rho_c) + \rho_c \right) c_{pr} T_r \\
- Q_p(t)\rho c_{pi} T - Q_f(t)\rho c_{pi} T + T \frac{\Psi_2(t)}{\rho_c}(\rho c_{pi} - \rho c_{pc}) \right] \right] \tag{A.16}
\]

Substitution of this equation into the concentration balance A.12 gives:

\[
V \epsilon(t) \frac{dC(t)}{dt} = \frac{C_i H_v}{\rho_i (H_v - c_{pi} T_i)} \left[ Q_p(t)\rho + Q_f(t)(\rho - \rho_c)[1 - \epsilon_f(t)] - \frac{\rho - \rho_c}{\rho_c} \Psi_2(t) \\
+ \frac{1}{H_v} \left[ P_{tot}(t) - Q_f(t)\xi(T_r - T) + Q_f(t) \left( \epsilon_f(t)(\rho - \rho_c) + \rho_c \right) c_{pr} T_r \right] \right]
\]
\[-Q_p(t)\rho_c \rho_i T - Q_f(t)\rho_c \rho_i T + T \frac{\Psi_2(t)}{\rho_c}(\rho_c \rho_i - \rho_c e_{pc}) \]

\[+ Q_f(t)(\rho_c - C(t))(1 - \epsilon_f(t)) - Q_p(t) C(t) - \frac{\rho_c - C(t)}{\rho_c} \Psi_2(t) \quad (A.17)\]

The assumptions on a constant volume and isothermal operation have reduced the set of balance equations to one single equation for the salt concentration in the mother liquor.

### A.2.2 The Class I model

To simplify Eqn. A.17 the following parameters are introduced:\(^1\):

\[k_0 = H_v - c_{pi} T_i\]
\[k_1 = \rho_c \rho_i - \rho_c e_{pc}\]
\[k_2 = H_v\]
\[k_3 = \rho - \rho_c\]
\[k_4 = \rho_c - C(t)\]
\[k_5 = \frac{1}{3k_v V}\]
\[k_6 = \frac{\rho_i k_0 k_5}{k_4 \rho_i k_0 - (k_1 T - k_3 k_2) C_i}\]
\[k_7 = \frac{C_i k_2}{\rho_i k_0}\]
\[k_8 = \frac{1}{k_2}\]
\[k_9 = \rho_c \rho_i T\]
\[k_{10} = \xi(T_r - T)\]
\[k_{11} = c_{pr} T_r\]

Substitution in Eqn. A.17 results in:

\[Ve(t) \frac{dC(t)}{dt} = k_7 \left[ Q_p(t) \rho + k_3 Q_f(t)[1 - \epsilon_f(t)] - \frac{\Psi_2(t)}{\rho_c} \right]\]

\(^1\)parameter \(\rho_4\) is only used in the Class II model discussed below.
\[ + k_8 \left[ \frac{P_{\text{tot}}(t) - k_{10}Q_f(t) + k_{11}Q_f(t)(\epsilon_f(t)k_3 + \rho_c)}{\rho_c} \right] \]

\[- k_9Q_p(t) - k_9Q_f(t) + k_1T \frac{\Psi_2(t)}{\rho_c} \left] \right. \]

\[ + [\rho_c - C(t)]Q_f(t)[1 - \epsilon_f(t)] - Q_p(t)C - [\rho_c - C(t)]\frac{\Psi_2(t)}{\rho_c}. \quad (A.18) \]

Further simplification gives:

\[ V(c(t) \frac{dC(t)}{dt} = k_7Q_p(t)\rho + k_7k_3Q_f(t)[1 - \epsilon_f(t)] - k_7k_3\frac{\Psi_2(t)}{\rho_c} \]

\[ + k_7k_8P_{\text{tot}}(t) - k_7k_8k_10Q_f(t) + k_7k_8k_11Q_f(t)(\epsilon_f(t)k_3 + \rho_c) \]

\[ - k_7k_8k_9Q_p(t) - k_7k_8k_9Q_f(t) + k_1k_7k_8T \frac{\Psi_2(t)}{\rho_c} \]

\[ + [\rho_c - C(t)]Q_f(t)[1 - \epsilon_f(t)] - Q_p(t)C - [\rho_c - C(t)]\frac{\Psi_2(t)}{\rho_c}. \quad (A.19) \]

The absolute super saturation concentration is defined by:

\[ \Delta C(t) \triangleq C(t) - C_s \quad (A.20) \]

Substitution into Eqn. A.19 results in the following supersaturation balance:

\[ V(c(t) \frac{d\Delta C(t)}{dt} = \]

\[ + \left[ - Q_p(t) - Q_f(t)[1 - \epsilon_f(t)] + \frac{\Psi_2(t)}{\rho_c} \right] \Delta C(t) \]

\[ + \left[ k_7\rho - k_7k_8k_9 - C_s \right] Q_p(t) \]

\[ + \left[ k_7k_3 - k_3k_7k_8k_11 + \rho_c - C_s \right] Q_f(t)[1 - \epsilon_f(t)] \]

\[ + \left[ - k_{10} + k_{11}(k_3 + \rho_c) - k_9 \right] k_7k_8Q_f(t) \]

\[ + \left[ - k_7k_3 + k_1k_7k_8T - \rho_c + C_s \right] \frac{\Psi_2(t)}{\rho_c} \]

\[ + k_7k_8P_{\text{tot}}(t) \quad (A.21) \]

Further simplification is achieved by introducing the following variables:

\[ k_{c1} = \frac{1}{V} \left[ k_7\rho - k_7k_8k_9 - C_s \right] \]
\[ k_{c2} = \frac{1}{V} \left[ k_7k_3 - k_3k_7k_8k_{11} + \rho_c - C_s \right] \]
\[ k_{c3} = \frac{1}{V} \left[ k_7k_8k_{10} + k_{11}(k_3 + \rho_c) - k_9 \right] \]
\[ k_{c4} = \frac{1}{V} \left[ k_7k_8 \right] \]
\[ k_{c5} = \frac{1}{V} \left[ -k_7k_3 + k_1k_7k_8T - \rho_c + C_s \right] \]

Now Eqn. A.21 is simplified to:

\[
\frac{d\Delta C(t)}{dt} = \frac{1}{V\epsilon(t)} \left[ -Q_p(t) - Q_f(t)[1 - \epsilon_f(t)] + \frac{\Psi_2(t)}{\rho_c} \right] \Delta C(t) \\
+ k_{c1}Q_p(t) + k_{c2}Q_f(t)[1 - \epsilon_f(t)] + k_{c3}Q_f(t) \\
+ k_{c4}P_{tot}(t) + k_{c5} \frac{\Psi_2(t)}{\rho_c} \right] \]

(A.22)

The growth rate is described by the powerlaw Eqn. 2.24. Substitution of this equation in the production term A.4 gives:

\[
\Psi_2(t) = 3V\rho_c k_v p_0 \Delta C^p(t) \int_0^\infty G_x(x,t)n(x,t)x^2dx \]

(A.23)

The dynamics of the crystal free phase in the crystallizer is now fully described by Eqn. A.22 and Eqn. A.23.

### A.2.3 The Class II model

The Class I model is further simplified by assuming that the saturation is constant in time: \(dC(t)/dt = 0\). Changes in the crystal mass production rate will give an immediate change of the crystal growth rate. The resulting model is denoted as the Class II model [197]. This assumption is valid for fast growing substances [154]. The concentration balance A.17 can now be written as a constraint for the growth rate. assuming \(dC/dt = 0\) in Eqn. A.17 gives:

\[
[k_3k_7 - k_1k_7k_8T + k_4] \frac{\Psi_2(t)}{\rho_c} = \\
+ k_7Q_p(t)\rho + k_7k_3Q_f(t)[1 - \epsilon_f(t)] \\
+ k_7k_8P_{tot}(t) - k_7k_8k_{10}Q_f(t) + k_7k_8k_{11}Q_f(t)(\epsilon_f(t)k_3 + \rho_c) \\
- k_7k_8k_9Q_p(t) - k_7k_8k_9Q_f(t) \\
+ k_4Q_f(t)[1 - \epsilon_f(t)] - Q_p(t)C \]

(A.24)
substitution of the general equation for the production rate Eqn. A.4 where \( G_e(x, t) \) is given by Eqn. 2.25 results in:

\[
G_k(t) = \frac{k_5}{[k_3 k_7 - k_1 k_7 k_8 T + k_4] m_{2x}} \left[ k_7 k_8 P_{tot}(t) + k_7 k_8 Q_f(t) \left[ k_{11}(k_3 + \rho_c) - k_9 - k_{10} \right] + \left[ - k_3 k_7 k_8 k_{11} + k_4 + k_3 k_7 \right] Q_f(t) \left[ 1 - \epsilon_f(t) \right] + \left[ k_7 \rho - k_7 k_8 k_9 - C \right] Q_p(t) \right]
\]

(A.25)

with

\[
m_{2x} = \int_0^\infty G_x(x, t) n(x, t) x^2 dx
\]

(A.26)

Further simplification is achieved by introducing the following constant factors:

\[
k_{g_0} = \frac{k_5}{k_3 k_7 - k_1 k_7 k_8 T + k_4}
\]

\[
k_{g_1} = \frac{1}{k_{g_0}} \left[ - k_3 k_7 k_8 k_{11} + k_4 + k_3 k_7 \right]
\]

\[
k_{g_2} = \frac{1}{k_{g_0}} \left[ k_7 k_8 k_{11}(k_3 + \rho_c) - k_9 - k_{10} \right]
\]

\[
k_{g_3} = \frac{1}{k_{g_0}} \left[ k_7 \rho - k_7 k_8 k_9 - C \right]
\]

\[
k_{g_4} = \frac{1}{k_{g_0}} \left[ k_7 k_8 \right]
\]

This results in the following simplified algebraic equation for the kinetic growth rate of crystals, which obeys the Class II growth rate constraint:

\[
G_k(t) = \frac{1}{m_{2x}(t)} \left[ (k_{g_1} m_3 f + k_{g_2}) Q_f(t) + k_{g_3} Q_p(t) + k_{g_4} P_{tot}(t) \right]
\]

(A.27)

Note that \( k_{g_3} \) solely accounts for the additional heat input when the feed temperature \( T_i \) is larger than the slurry temperature. Thus, for \( T = T_i \), \( k_{g_3} \) will be zero.
Appendix B

Model linearisation

A linearized version of the crystallizer model is obtained analytically by assuming small perturbations around the stationary solution. The nonlinear elements in the model are approximated with a first order Taylor expansion. First the linear population balance is derived, next the linearized submodels are given.

B.1 Linearisation of the population balance

The stationary solution of Eqn. G.1 is given by: All time and size variables are omitted for ease of notation. Perturbations are denoted with the $\Delta-$symbol. Introduction of a first order approximation to the population balance gives:

$$V \left[ \frac{\partial \Delta n}{\partial t} + \frac{\partial (\bar{G}_e + \Delta G_e)(\bar{n} + \Delta n)}{\partial x} \right]$$

$$+ \left[ (\bar{Q}_f + \Delta Q_f)(\bar{h}_f + \Delta h_f) + (\bar{Q}_p + \Delta Q_p)(\bar{h}_p + \Delta h_p) \right](\bar{n} + \Delta n) = 0. \quad (B.1)$$

where the overbar denotes a stationary value. Subtraction of the stationary solution given by Eqn. 2.59 and neglecting all second order $\Delta$’s gives:

$$\frac{\partial \Delta n}{\partial t} + \frac{\partial \Delta G_e \bar{n}}{\partial x} + \frac{\partial \bar{G}_e \Delta n}{\partial x}$$

$$+ \left[ \Delta Q_f \bar{h}_f + \bar{Q}_f \Delta h_f + \Delta Q_p \bar{h}_p + \bar{Q}_p \Delta h_p \right] \frac{\bar{n}}{V} + \frac{\bar{Q}_f \bar{h}_f + \bar{Q}_p \bar{h}_p}{V} \Delta n = 0. \quad (B.2)$$

Rewriting this equation and substitution of:

$$\frac{\partial \Delta G_e \bar{n}}{\partial x} = \Delta G_e \frac{d\bar{n}}{dx} + \bar{n} \frac{d\Delta G_e}{dx} \quad (B.3)$$

$$\frac{\partial \bar{G}_e \Delta n}{\partial x} = \bar{G}_e \frac{\partial \Delta n}{\partial x} + \Delta n \frac{d\bar{G}_e}{dx} \quad (B.4)$$

\[last printed: \text{April 24, 1995}\]
and substitution of the linearized model for both the effective growth function and its first derivative:

$$\Delta G_e = \Delta G_k G_x,$$  \hspace{1cm} (B.5)  

$$\frac{\partial \Delta G_e}{\partial x} = \Delta G_k \frac{dG_x(x)}{dx},$$  \hspace{1cm} (B.6)  

then gives the following linearised population balance:

$$\frac{\partial \Delta n}{\partial t} + \bar{G}_e \frac{d\Delta n}{dx} + \left[ G_x \frac{d\bar{n}}{dx} + \bar{n} \frac{dG_x}{dx} \right] \Delta G_k$$

$$+ \left[ \Delta Q_{jf} \bar{h}_f + \bar{Q}_f \Delta h_f + \Delta Q_{pf} \bar{h}_p + Q_{pf} \Delta h_p \right] \frac{\bar{n}}{V}$$

$$+ \left[ \frac{Q_{jf} \bar{h}_f + \bar{Q}_f \bar{h}_p}{V} + G_k \frac{\partial \bar{G}_x}{\partial x} \right] \Delta n = 0$$  \hspace{1cm} (B.7)  

This equation is treated further to end up with a linear time invariant state-space model:

$$\frac{dx}{dt} = Ax + Bu$$

$$y = Cx + Du.$$  \hspace{1cm} (B.8)  

The process input vector $\Delta u$ is given by:

$$\Delta u = [\Delta Q_f \hspace{0.5cm} \Delta Q_{pf} \hspace{0.5cm} \Delta P_t]^T$$  \hspace{1cm} (B.9)  

To arrive at this expression a lumping step is involved. First, however, linear expressions should be derived for the nonlinear internal variables, e.g. the growth rate $\Delta G_k$ and the boundary condition. Below this problem will be solved for two different cases:

- A Class II type of system in which the growth rate is described by an algebraic constraint (cf. Eqn. A.27).

- A Class I type of system in which the growth rate is determined from the supersaturation that is described explicitly with an ordinary differential equation (cf. Eqn. A.22).

### B.2 Linearisation of removal functions

The product removal efficiency function Eqn. 2.14 is a stationary functions which need not be to linearized. If we assume that the offset $p_{p3}$ (see Eqn. 2.14) to be constant, the product flow $Q_p$, is a nonlinear function of the classifier feedflow $Q_{pf}$, and should therefore be linearized.
Linearisation of product flow function

Linearization of Eqn. 2.19 gives:

\[
\Delta Q_p = \left[ (1 - k_v m_3) p_{p3} + k_v \int_0^\infty h_p \tilde{n} x^3 dx \right] \Delta Q_{pf} \\
- \bar{Q}_{pf} p_{p3} k_v \Delta m_3 + \bar{Q}_{pf} k_v \int_0^\infty h_p \Delta n x^3 dx
\] (B.10)

or

\[
\Delta Q_p = L_{Qp1} \Delta Q_{pf} + L_{Qp2} \Delta m_3 + L_{Qp3} \int_0^\infty h_p \Delta n x^3 dx
\] (B.11)

with

\[
L_{Qp1} = (1 - k_v m_3) p_{p3} + k_v \int_0^\infty h_p \tilde{n} x^3 dx \\
L_{Qp2} = -\bar{Q}_{pf} p_{p3} k_v \\
L_{Qp3} = \bar{Q}_{pf} k_v
\] (B.12)

Linearisation of fines classification function

The product classification function \( h_p \) is a static function, which does not need to be linearized. Linearization of the fines classification function (Eqn. 2.9), gives:

\[
\Delta h_f = \left. \frac{d h_f}{d Q_f} \right|_w \Delta Q_f
\] (B.13)

with

\[
\left. \frac{d h_f}{d Q_f} \right|_w = l_h(x) = \frac{p_f2 p_{f1} \bar{h}_f^2 x^{p_{f2}}}{2 x^{(p_{f2}+2)}}
\] (B.14)

B.3 Linearisation of moment equations

The standard moment equations are already linear, thus:

\[
m_i = \int_0^\infty n x^i dx, \Rightarrow \Delta m_i = \int_0^\infty \Delta n x^i dx
\] (B.15)

and,

\[
\Delta m_{2x} = \int_0^\infty \bar{G}_x(x) \Delta n x^2 dx
\] (B.16)

The linearised equation for the third moment of the fines distribution is given by:

\[
\Delta m_{3f} = \int_0^\infty h_f \Delta n x^3 dx + \int_0^\infty l_h(x) x^3 \tilde{n} dx \Delta Q_f
\] (B.17)
The latter term can be written as the product of the constant factor $l_{m3f}$ and the flow $\Delta Q_f$:

$$l_{m3f} = \int_0^\infty l_{hf}(x)x^3\tilde{n}dx$$  \hspace{1cm} (B.18)

### B.4 Linearisation of growth rate model

A linear relation for the first derivative of the length dependent growth rate model given by Eqn. 2.26 is given by:

$$\frac{dG_z}{dx} = -\frac{p_8x_a^{p_8}x_{p_8}\Delta Q_f}{x_2^{p_8}} \frac{x_{p_8}^{p_8} + x_e^{p_8}}{(x_{p_8}^{p_8} + x_a^{p_8})^2}$$  \hspace{1cm} (B.19)

Linear expressions for the kinetic growth rate model are derived for both the Class I and the Class II model.

#### Linear growth rate for Class II model

Linearizing the Class II growth rate constraint Eqn. A.27 gives:

$$\Delta G_k = \frac{\partial G_k}{\partial Q_p} \bigg|_w \Delta Q_p + \frac{\partial G_k}{\partial Q_f} \bigg|_w \Delta Q_f + \frac{\partial G_k}{\partial P_t} \bigg|_w \Delta P_t + \frac{\partial G_k}{\partial m_{2x}} \bigg|_w \Delta m_{2x} + \frac{\partial G_k}{\partial m_{3f}} \bigg|_w \Delta m_{3f}$$  \hspace{1cm} (B.20)

with:

$$\frac{\partial G_k}{\partial Q_f} \bigg|_w = lg_1 = \frac{kg_1m_{3f} + kg_2}{m_{2a}}$$  \hspace{1cm} (B.21)

$$\frac{\partial G_k}{\partial Q_p} \bigg|_w = lg_2 = \frac{kg_3}{m_{2a}}$$  \hspace{1cm} (B.22)

$$\frac{\partial G_k}{\partial P_t} \bigg|_w = lg_3 = \frac{kg_4}{m_{2a}}$$  \hspace{1cm} (B.23)

$$\frac{\partial G_k}{\partial m_{2x}} \bigg|_w = lg_4 = \frac{\bar{G}_k}{m_{2a}}$$  \hspace{1cm} (B.24)

$$\frac{\partial G_k}{\partial m_{3f}} \bigg|_w = lg_5 = \frac{kg_1\bar{Q}_f}{m_{2a}}$$  \hspace{1cm} (B.25)

$$lg_6 = lg_5l_{m3f}$$  \hspace{1cm} (B.26)

this equation can be written as:

$$\Delta G_k = lg_2\Delta Q_p + (lg_1 + lg_6)\Delta Q_f + lg_3\Delta P_t + lg_4\int_0^\infty G_z x^2\Delta ndx + lg_5\int_0^\infty \bar{h}_f x^3\Delta ndx$$  \hspace{1cm} (B.27)
Substitution of Eqn. B.11 gives:

\[
\Delta G_k = l g_2 \left[ L_{Qp1} \Delta Q_{pf} + L_{Qp2} \Delta m_3 + L_{Qp3} \int_0^\infty h_p \Delta n x^3 dx \right] \\
+ \left( l g_1 + l g_6 \right) \Delta Q_f + l g_3 \Delta P_t + l g_4 \int_0^\infty G_x x^2 \Delta n dx + l g_5 \int_0^\infty h_f x^3 \Delta n dx \tag{B.28}
\]

**Linear growth rate for Class I model**

Linearization of the Class I growth rate model (Eqn. 2.24), gives:

\[
\Delta G_k = \frac{dG_k}{dC} \bigg|_w \Delta C^* = p_7 p_6 \Delta \bar{C}^{(p_7 - 1)} \Delta C^* = L_{cg} \Delta C^*, \tag{B.29}
\]

with:

\[
L_{cg} = p_7 p_6 \Delta \bar{C}^{(p_7 - 1)} \tag{B.30}
\]

To prevent the use of a double \(\Delta\)-symbol, the perturbation of \(\Delta C\) around its average value is denoted as \(\Delta C^*\).

Prior to the linearization of the supersaturation the variable \(\Psi^*_2\) is introduced which is calculated from Eqn. A.4:

\[
\Psi^*_2 = 3 p_6 k_v V \int_0^\infty G_x n dx = \frac{\Psi_2}{\rho_c \Delta C^{p_7}} \tag{B.31}
\]

The supersaturation balance (Eqn. A.22) can be written as a general nonlinear function:

\[
\frac{\Delta C^*}{dt} = f(\Delta Q_p, \Delta Q_f, \Delta P_t, \Delta C^*, \Delta m_{3f}, \Delta m_{2x}, \Delta m_3) \tag{B.32}
\]

Linearization gives:

\[
\frac{\Delta C^*}{dt} = \frac{\partial f}{\partial Q_p} \bigg|_w \Delta Q_p + \frac{\partial f}{\partial Q_f} \bigg|_w \Delta Q_f + \frac{\partial f}{\partial P_t} \bigg|_w \Delta P_t + \frac{\partial f}{\partial C} \bigg|_w \Delta C \\
+ \frac{\partial f}{\partial m_{3f}} \bigg|_w \Delta m_{3f} + \frac{\partial f}{\partial m_{2x}} \bigg|_w \Delta m_{2x} + \frac{\partial f}{\partial m_3} \bigg|_w \Delta m_3 \tag{B.33}
\]

with,

\[
\frac{\partial f}{\partial Q_f} \bigg|_w = l c_1 = \frac{1}{\epsilon} \left[ (k_{c2} - \frac{\Delta \bar{C}}{V}) k_v \bar{m}_{3f} + k_{c3} \right] \tag{B.34}
\]

\[
\frac{\partial f}{\partial Q_p} \bigg|_w = l c_2 = \frac{1}{\epsilon} (k_{c1} - \frac{\Delta \bar{C}}{V}) \tag{B.35}
\]

\[
\frac{\partial f}{\partial P_t} \bigg|_w = l c_3 = \frac{1}{\epsilon} k_{c4} \tag{B.36}
\]
\[ \frac{\partial f}{\partial \Delta C^*} \bigg|_w = l_c_4 = \frac{\Psi_2}{V \varepsilon} \left[ (p_7 + 1) \Delta \tilde{C}^p_7 + V k_c 5 p_7 \Delta \tilde{C}^{(p_7 - 1)} \right] \]
\[ - \frac{1}{\varepsilon} (\tilde{Q}_p + \tilde{Q}_f k_v m_{3f}) \]
(B.37)

\[ \frac{\partial f}{\partial m_{3f}} \bigg|_w = l_c_5 = \frac{k_v Q_f}{\varepsilon} \left[ k_c 2 - \Delta \tilde{C} \right] \frac{\tilde{V}}{V} \]
(B.38)

\[ \frac{\partial f}{\partial m_{2f}} \bigg|_w = l_c_6 = \frac{1}{\varepsilon} \left[ k_c 5 + \frac{\Delta \tilde{C}}{V} \right] \left( 3 V k_v \tilde{G}_k \right) \]
(B.39)

\[ \frac{\partial f}{\partial m_3} \bigg|_w = l_c_7 = \frac{f}{\varepsilon} k_v = 0 \]
(B.40)

and substitution of the relation for \( \Delta Q_p \) given by Eqn. B.11, this equation can be written as:

\[ \frac{\Delta C^*}{dt} = l_c_2 \left[ L_{Qp1} \Delta Q_{pf} + L_{Qp2} \int_0^\infty \Delta n x^3 dx + L_{Qp3} \int_0^\infty h_p \Delta n x^3 dx \right] \]
\[ + l_c 5 \int_0^\infty h_f \Delta n x^3 + l_c 5 l_{m3f} \Delta Q_f + l_c_1 \Delta Q_f \]
\[ + l_c_3 \Delta P_t + l_c_4 \Delta C + l_c_6 \int_0^\infty G_x(x) \Delta n x^2 dx \]
(B.41)

Note that fluctuations of the void fraction around the stationary solution do not affect the supersaturation (\( l_c_7 = 0 \)).

## B.5 Linearisation of the boundary condition

From Eqn. 2.30 the following linearised boundary condition is obtained:

\[ \Delta n(x_0, t) = \frac{\partial n(x_0, t)}{\partial B(t)} \bigg|_w \Delta B(t) + \frac{\partial n(x_0, t)}{\partial G_k(t)} \bigg|_w \Delta G_k(t). \]
(B.42)

With

\[ \frac{\partial n(x_0, t)}{\partial B(t)} \bigg|_w = \frac{1}{G_k}, \text{ and } \frac{\partial n(x_0, t)}{\partial G_k(t)} \bigg|_w = \frac{-\tilde{B}}{G_k^2} = -\frac{\tilde{n}(0)}{G_k}, \]
(B.43)

This can be written as:

\[ \Delta n(x_0, t) = \frac{1}{G_k} \left[ \Delta B - \tilde{n}(0) \Delta G_k \right] \]
(B.44)

Below, linear boundary conditions are derived for both the ClassII and ClassI models.
Linear boundary condition for Class II model

Linearization of the secondary nucleation rate, given by Eqn. 2.23, with \( \Delta C \) replace by \( G_k \) gives:

\[
\Delta B = lb_1 \Delta G_k + lb_2 \int_{p_4}^{\infty} x^{p_5} \Delta ndx. \tag{B.45}
\]

with,

\[
lb_1 = p_2 p_3 \left[ \int_{p_4}^{\infty} \bar{n} x^{p_5} dx \right]^{p_1} \frac{G_k^{p_2 - 1}}{G_k} \tag{B.46}
\]

\[
lb_2 = p_1 p_3 \left[ \int_{p_4}^{\infty} \bar{n} x^{p_5} dx \right]^{p_1 - 1} \frac{G_k^{p_2}}{G_k}. \tag{B.47}
\]

Substitution of Eqn. B.45 into the linear boundary condition Eqn. B.44, gives:

\[
\Delta n(0, t) = \frac{1}{G_k} \left[ (lb_1 - \bar{n}(0)) \Delta G_k + lb_2 \int_{p_4}^{\infty} x^{p_5} \Delta ndx, \right] \tag{B.48}
\]

Substitution of Eqn. B.28, gives:

\[
\Delta n(0, t) = \frac{[lb_1 - \bar{n}(0)]}{G_k} \left[ lg_2 \left[ L Q P_3 \Delta Q_{p_f} + L Q P_2 \Delta n_3 + L Q P_3 \int_0^{\infty} h_p \Delta n x^3 dx \right]
\right.
\]

\[
+ (lg_1 + lg_6) \Delta Q_f + lg_3 \Delta P_1 + lg_4 \int_0^{\infty} G x^2 \Delta ndx + lg_5 \int_0^{\infty} h_f x^3 \Delta ndx
\left. \right] + \frac{lb_2}{G_k} \int_{p_4}^{\infty} x^{p_5} \Delta ndx. \tag{B.49}
\]

This equation is simplified to:

\[
\Delta n(0, t) = dn_2 \int_{p_4}^{\infty} x^{p_5} \Delta ndx, + dn_3 \int_0^{\infty} G x^2 \Delta ndx + dn_4 \int_0^{\infty} h_f x^3 \Delta ndx
\]

\[
+ dn_5 \Delta Q_f + dn_6 \Delta Q_{p_f} + dn_7 \int_0^{\infty} \Delta n x^3 dx
\]

\[
+ dn_8 \int_0^{\infty} h_p \Delta n x^3 dx + dn_9 \Delta P_1, \tag{B.50}
\]

with,

\[
dn_1 = \frac{[lb_1 - \bar{n}(0)]}{G_k}
\]

\[
dn_2 = \frac{lb_2}{G_k}
\]
\[ \begin{align*}
    dn_3 &= dn_1 lg_4 \\
    dn_4 &= dn_1 lg_5 \\
    dn_5 &= dn_1 (lg_1 + lg_6) \\
    dn_6 &= dn_1 lg_2 L_{Qp1} \\
    dn_7 &= dn_1 lg_2 L_{Qp2} \\
    dn_8 &= dn_1 lg_2 L_{Qp3} \\
    dn_9 &= dn_1 lg_3.
\end{align*} \]

**Linear boundary condition for Class I model**

Linearizing the nucleation model Eqn. 2.23 for a Class I system gives:

\[ \Delta B = lb_1 L_{cg} \Delta C^* + lb_2 \int_{p_4}^{\infty} x^{p_5} \Delta ndx \quad (B.51) \]

with,

\[ lb_1 = p_2 p_3 \left[ \int_{p_4}^{\infty} \tilde{n}_x^{p_5} dx \right]^{p_1} \tilde{G}_k^{p_2 - 1} \quad (B.52) \]

\[ lb_2 = p_1 p_3 \left[ \int_{p_4}^{\infty} \tilde{n}_x^{p_5} dx \right]^{p_1 - 1} \tilde{G}_k^{p_2}. \quad (B.53) \]

Substitution in the linearized boundary condition Eqn. B.44, gives:

\[ \Delta n(0, t) = \frac{1}{G_k} \left[ lb_2 \int_{p_4}^{\infty} x^{p_5} \Delta ndx + \left[ (lb_1 - \tilde{n}(0)) L_{cg} \Delta C^* \right] \right] \quad (B.54) \]

This is an equation is simplified to:

\[ \Delta n(0, t) = dn_1 \int_{p_4}^{\infty} x^{p_5} \Delta ndx + dn_2 \Delta C^* \quad (B.55) \]

with,

\[ \begin{align*}
    dn_1 &= \frac{1}{G_k} lb_2 \\
    dn_2 &= \frac{1}{G_k} \left[ (lb_1 - \tilde{n}(0)) L_{cg} \right]
\end{align*} \quad (B.56, B.57) \]

**B.6 Linearisation of output equations**

Several process output quantities are described in Chapter 4.
The mean crystal size

The mean crystal size $X_{43}$ given by Eqn. 4.14 can be linearized easily:

$$\Delta X_{43} = \frac{1}{m_3} \left[ \Delta m_4 - 2 \frac{m_4}{m_3} \Delta m_3 \right] \quad (B.58)$$

The coefficient of variation

For the same reason the coefficient of variation $cv_{43}$ instead of the quartile ratio is used as a measure for the spread. This gives:

$$\Delta cv_{43} = \frac{1}{2 \bar{c}_{43}} \left[ \frac{\bar{m}_5}{\bar{m}_4^2} \Delta m_3 + \frac{\bar{m}_3}{\bar{m}_4^2} \Delta m_5 - 2 \frac{\bar{m}_3 \bar{m}_5}{\bar{m}_4^3} \Delta m_4 \right] \quad (B.59)$$

The scattered light energy

To eliminate the influence of a fluctuating dilution factor (See Section 4.2), a normalization step is implemented in which each element of the scattered light energy vector generated by the Fraunhofer model is divided by the sum of all elements $e_i, i = 1, \ldots, n$:

$$e_i^* = \frac{e_i}{\sum_{j=1}^{n} e_j} = f_i(e_1, e_2, \ldots, e_n) \quad (B.60)$$

Linearization of this equation gives:

$$\Delta e_i^* = \left. \frac{\partial f_i}{\partial e_1} \right|_{w} \Delta e_1 + \left. \frac{\partial f_i}{\partial e_2} \right|_{w} \Delta e_2 + \cdots + \left. \frac{\partial f_i}{\partial e_n} \right|_{w} \Delta e_n \quad (B.61)$$

with

$$\left. \frac{\partial f_i}{\partial e_j} \right|_{w} = \frac{-1}{(\sum_{j=1}^{n} e_j)^2} \quad i \neq j \quad (B.62)$$

$$\left. \frac{\partial f_i}{\partial e_j} \right|_{w} = \frac{\sum_{j=1}^{n} (e_j - e_i)}{(\sum_{j=1}^{n} e_j)^2} \quad i = j \quad (B.63)$$

B.7 Collecting the terms

The terms presented in the linearized equations are collected to obtain a linear state-space structured model. However, due to the distributed parameters this model still has an infinite dimension. Therefore, a lumping step as described in Section 2.3.4 is applied to arrive at a finite dimensional linear state-space model.
The linear Class II model

For the Class II model the linearised equation for the population balance model can be written as:

\[ \frac{\partial \Delta n}{\partial t} + f_1(x) \frac{\partial \Delta n}{\partial x} + f_2(x) \Delta n + f_3(x) \Delta u = 0 \]  
(B.64)

and the boundary condition:

\[ \Delta n(x_0, t) = f_4(x) \Delta n + f_5 \Delta u \]  
(B.65)

The functions \( f_3 \) and \( f_5 \) are vector functions, which contain a number of elements equal to the number of input variables. The functions \( \{f_1, f_2, \ldots, f_5\} \) are given by:

\[ f_1(x) = G_e(x) \]  
(B.66)

\[ f_2(x) = \left[ \frac{dn}{dx} G_x + \bar{n} \frac{dG_x}{dx} \right] \left[ \frac{lg_1}{\Delta n} \int_0^\infty G_x x^2 \Delta ndx \right. \right. \]

\[ + \frac{lg_5}{\Delta n} \int_0^\infty \Delta n \bar{h}_f x^3 dx + \frac{L_Q p_2}{\Delta n} \int_0^\infty \Delta n x^3 dx + \frac{L_Q p_3}{\Delta n} \int_0^\infty h_p \Delta n x^3 dx \] \]

\[ + \frac{\bar{Q}_f \bar{h}_f + \bar{Q}_p \bar{h}_p}{V} + G_k \frac{\partial \bar{G}_x}{\partial x} \]  
(B.67)

\[ f_3(x) = \left[ \frac{dn}{dx} G_x + \frac{dG_x}{dx} \bar{n} \right] \left[ \frac{lg_1 + lg_6}{\Delta n} + \bar{h}_f + \bar{Q}_f h_f(x) \right] \frac{\bar{n}}{\bar{V}} \]

\[ f_4(x) = \frac{dn_2}{\Delta n} \int_{p_4}^{\infty} x^5 \Delta ndx + \frac{dn_3}{\Delta n} \int_0^\infty G_x x^2 \Delta ndx + \frac{dn_4}{\Delta n} \int_0^\infty \bar{h}_f x^3 \Delta ndx \]

\[ + \frac{dn_7}{\Delta n} \int_0^\infty \Delta n x^3 dx + \frac{dn_8}{\Delta n} \int_0^\infty h_p \Delta n x^3 dx \]  
(B.69)

\[ f_5 = \begin{bmatrix} dn_5 \\ dn_6 \\ dn_9 \end{bmatrix} \]  
(B.70)
With the second order central difference scheme the following difference matrix is obtained:

\[
H = \frac{1}{2\Delta x} \begin{bmatrix} 0 & 1 & \cdots & 0 \\ -1 & 0 & 1 & \vdots \\ -1 & 0 & 1 & \vdots \\ \vdots & \ddots & \ddots & \vdots \\ -1 & 0 & 1 & \vdots \\ 0 & \cdots & 1 & -4 & 3 \end{bmatrix}
\]  
(B.71)

Note that the first grid point \( x_0 \) at which the left boundary condition is defined is not yet included in the model.

A discretized model is obtained by discretizing the functions \( f_i \) on a set of discrete sizes: \( x_i, \ i = 1, \cdots, n \) and introduction of the diffraction matrix. The discretized functions are assumed to be row vectors. Eqn. B.64 can be written as:

\[
\frac{d\Delta n}{dt} = \left[ -\text{diag}(f_1(x))H - \text{diag}(f_2(x))I \right] \Delta n - f_3^T \Delta u 
\]  
(B.72)

The square matrices \( \text{diag}(f_1) \) and \( \text{diag}(f_2) \) contain the values of \( f_1(x) \) and \( f_2(x) \) for \( x_i, \ i = 1, \cdots, n \) on their diagonal. The linear boundary condition Eqn. B.65 is incorporated in the first row of the system matrix:

\[
A_i(1, i) = A(1, i) + \frac{1}{2\Delta x} f_4(x_i), \ i = 1, \cdots, n 
\]  
(B.73)

and the first row of the input matrix:

\[
B_i(1, i) = B(1, i) + \frac{1}{2\Delta x} f_5(i), \ i = 1, \cdots, 3 
\]  
(B.74)

**The linear Class I model**

For the Class I the linearised equation for the population balance model is written as:

\[
\begin{bmatrix} \frac{d\Delta n}{dt} \\ \frac{d\Delta C^*}{dt} \end{bmatrix} = \begin{bmatrix} \frac{\partial \Delta n}{\partial t} + f_1(x) \frac{\partial \Delta n}{\partial x} + f_2(x) \Delta n + f_3(x) \Delta C^* + f_4^T(x) \Delta u \\ f_5(x) \Delta n + f_6 \Delta C^* + f_7^T \Delta u \end{bmatrix} 
\]  
(B.75)

and the boundary condition:

\[
\Delta n(x_0, t) = f_8(x) \Delta n + f_9 \Delta C^* 
\]  
(B.76)
with \{f_1, f_2, \ldots, f_{10}\} given by:

\[
\begin{align*}
  f_1(x) &= \bar{G}_e(x) \\
  f_2(x) &= \frac{Q_f \bar{h}_f + Q_{pf} \bar{h}_p}{V} + G_k \frac{\partial \bar{G}_e}{\partial x} \\
  f_3(x) &= \left[ \frac{dn}{dx} G_x + \bar{n} \frac{dG_x}{dx} \right] L_{cg} \\
  f_4(x) &= \left[ \bar{h}_f + l_{hf} \right] \bar{n} / V \\
  f_5(x) &= \frac{lc_2 L_{Qp3}}{\Delta n} \int_0^\infty \bar{h}_p \Delta nx^3 dx + \frac{lc_3}{\Delta n} \int_0^\infty \bar{h}_f \Delta nx^3 dx + \frac{lc_6}{\Delta n} \int_0^\infty G_x(x) \Delta nx^2 dx \\
  f_6 &= lc_4 \\
  f_7 &= \begin{bmatrix}
    lc_1 + lc_5 l_{m3f} \\
    lc_2 L_{Qp1} \\
    lc_3
  \end{bmatrix} \\
  f_8(x) &= \frac{dn_1}{\Delta n} l_{b2} \int_{p4}^\infty \Delta nx^{p5} dx \\
  f_9 &= dn_2
\end{align*}
\]

Again using the difference matrices Eqn. B.71 and Eqn. B.71 gives:

\[
\begin{bmatrix}
  \frac{d\Delta n}{dt} \\
  \frac{d\Delta C^*}{dt}
\end{bmatrix} = \begin{bmatrix}
  H \text{diag}(f_1) - I \ast \text{diag}(f_2) & f_3(x) \\
  f_5(x) & f_6
\end{bmatrix} \begin{bmatrix}
  \Delta n \\
  \Delta C^*
\end{bmatrix} + \begin{bmatrix}
  f_4^T(x) \\
  f_7^T
\end{bmatrix} \begin{bmatrix}
  \Delta Q_f \\
  \Delta Q_{pf} \\
  \Delta P_f
\end{bmatrix}
\]

(B.86)
Note that the order of this model is \( n + 1 \) instead of \( n \) and that the system matrix has a sparse structure with one upper and a lower diagonal and a full first row, last row and last column.

The linear boundary condition Eqn. B.76 is incorporated in the first \( n \) elements of the first row of the system matrix:

\[
A(1, i) = A(1, i) + \frac{1}{2\Delta x} f_8(x_i), \; i = 1, \cdots, n
\]  

(B.87)

and the last element of the first row (\( n + 1 \)) of the system matrix:

\[
A(1, n + 1) = A(1, n + 1) + \frac{1}{2\Delta x} f_9(i),
\]

(B.88)
Appendix C

Model assumptions

Major assumptions general model:

1. The crystallizer volume is ideally mixed.
2. No solute or solid crystal material is present in the vapour line.
3. The particle shape is constant and rectangular with an aspect ratio $\alpha$.
4. The solids phase contains only a single pure component.
5. Primary nucleation does not occur.
6. Secondary nucleation takes place at a size $x_0$.
7. Crystals do not aggregate.
8. Breakage of particles does not occur.
9. Growth rate dispersion does not occur.
10. Attrition, length dependent growth and spatial segregation occur in the crystallizer, and are accounted for with a length dependent growth function.
11. The effective growth rate is the product of the length dependent growth rate and the kinetic growth rate determined by the supersaturation level.
12. The impeller speed is constant.
13. The kinetic growth rate does not depend on the crystal size.
14. Entrance classification at the slurry withdrawal locations does not occur.
15. No crystallization phenomena occur in the fines and product classifiers.
16. Hold-up in the classifiers is neglected.
17. All fines dissolve.

Additional assumptions UNIAK pilot crystallizer model:

18. The temperature in crystallizer is constant.
19. The volume of the crystallizer is constant.
20. The temperature of the dissolved fines is constant.
21. The heat of crystallization is zero.
22. The heat loss to the surrounding is zero.
23. The efficiency of product classification does not depend on time.
Appendix D

Physical properties and operation conditions

Physical properties of the substance system

The following relations are used in the crystallizer model to calculate the properties of ammonium sulphate as a function of the temperature.

The solubility

\[ w = 0.41179 + 9.121 \cdot 10^{-4} T \]  
\[ -6 \degree C \leq T \leq 90 \degree C \]  
\[ \text{[kg/kg soln]} \]  
\[ \text{(D.1)} \]

The Dynamic viscosity of the solution:

\[ \eta = 10^{A_1 + A_2 w + A_3 w^2 + A_4 w^3 + A_5 T + A_6 T^2 + A_7 T^3} \]  
\[ \text{[mpas]} \]  
\[ \text{(D.2)} \]

with,

\[ A_1 = 0.22 \]
\[ A_2 = 5.70 \cdot 10^{-3} \]
\[ A_3 = 1.22 \cdot 10^{-4} \]
\[ A_4 = -1.90 \cdot 10^{-7} \]
\[ A_5 = -1.25 \cdot 10^{-2} \]
\[ A_6 = 7.81 \cdot 10^{-5} \]
\[ A_7 = -2.66 \cdot 10^{-7} \]

and

\[ 0 \degree C \leq T \leq 80 \degree C \]
\[ 0.02 \leq w \leq 0.42 \]
The density of crystals:

\[ \rho_c = 1777.5 - 0.19697T \quad [\text{kg/m}^3] \quad (D.3) \]

\[ 0^\circ C \leq T \leq 80^\circ C \]

The density of the solution:

\[ \rho_l = 1007.9 - 0.384T + 568.907w \quad [\text{kg/m}^3] \quad (D.4) \]

\[ 35^\circ C \leq T \leq 65^\circ C \]

This equation only holds near saturation [39]

The specific heat of a solution:

\[ c_{pl} = 4.259 - 3.0321w - 1.7668 \cdot 10^{-3}T + 4.2874 \cdot 10^{-6}T^2 \quad [\text{KJ/KgK}] \quad (D.5) \]

The specific heat of crystals:

\[ c_{pc} = 1.3760 + 2.13 \cdot 10^{-2}T \quad (D.6) \]

\[ 25^\circ C \leq T \leq 323^\circ C \]

Constant operation conditions

The 970 litre crystallizer is operated at the following constant conditions:

**Table D.1**: constant operation conditions 970 litres crystallizer.

<table>
<thead>
<tr>
<th>Condition</th>
<th>Nominal value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Crystallizer temperature</td>
<td>50 °C</td>
</tr>
<tr>
<td>Feed temperature</td>
<td>55 °C</td>
</tr>
<tr>
<td>Saturation temperature of feed</td>
<td>50 °C</td>
</tr>
<tr>
<td>Nominal heat input</td>
<td>120 KW</td>
</tr>
<tr>
<td>Temperature of dissolved fines</td>
<td>60 °C</td>
</tr>
<tr>
<td>Nominal product flow rate</td>
<td>0.215 l/s</td>
</tr>
<tr>
<td>Nominal fines removal rate</td>
<td>1.00 l/s</td>
</tr>
<tr>
<td>Volume of crystallizer</td>
<td>970 m$^3$</td>
</tr>
</tbody>
</table>
Appendix E

Software tools

Matlab tools

• Model predictive control toolbox (MPC) [163]
  purpose: Constrained multivariable predictive control of linear and nonlinear processes using a linear internal model in state-space and step format.
  applied modules: scmpc\(^1\), smpccon, smpccl, mod2ss
  used in: Chapter 10

• Multivariable Frequency Domain Toolbox (MFD) [74]
  purpose: Analysis of multivariable processes in the frequency domain for control purposes
  used in: Chapter 10
  applied modules: feig, dmv2fr, mv2fr, fmulf, fsvd

• Weighted Order Reduction toolbox (WOR) [255]
  purpose: Frequency weighted reduction of high order models in both closed and open-loop configuration.
  used in: Section 2.3.8, Chapter 6 and 10
  applied modules: balredf

• Process controllability toolbox (PCTB) [147]
  purpose: Analysis of several controllability aspects of linear processes using state-space models
  used in: Chapter 9
  applied modules: run_pctb, dynsvd, mincond, relgain

\(^1\)This module has been adapted for (realtime) control of the UNIAK pilot plant, and for simulation of the MPC controlled crystallizer model. The results are described in Section 10.3.
• System Identification Toolbox [145]
  
purpose: Identification of MIMO processes.
  used in: Chapter 6
  applied modules: arx, th2ss

• Control toolbox [91]
  
purpose: Common control system design, analysis and modelling tools, including tools for Linear optimal control, (e.g. state estimator design)
  used in: Chapter 7, 9, and 10
  applied modules: c2d, dlqe, lqe, bode, dbode, lsim, dlsim, balred

• Miscellaneous
  
  Matlab M-file ortfir. Used for identification with orthonormal basis functions as applied in Chapter 6 (see also [237]).

Other (Fortran) tools

• Crystallizer process simulator (CPS)
  
purpose: Simulation, of linear and nonlinear evaporative crystallizers with an option to output linearized state-space models (see also Appendix F)
  used in: all chapters

• ESSS, From Dumsi package [236]
  
purpose: Identification of state-space models in (pseudo)-canonical observability form, output error and innovation error identification criterion.
  used in: Chapter 6

• TMSS03, from Dumsi package [236]
  
purpose: Conversion of a state-space model into an observability canonical form representation. This program has been used in conjunction with the ESSS program.
  used in: Chapter 6
Appendix F

Simulation program

The simulation model is subdivided into five main modules (Fig. F.1):

Initial
Contains a menu for the selection of parameter values and constant process settings; to calculate the stationary solution of model as a function of process inputs and different parameters related to equipment sizing and crystallization kinetics; and to precalculated different quantities to speedup the dynamics simulation.

Main
In the main module all calls to sub-modules are presented and the exchange of data with the environment (e.g Matlab, MatrixX) is controlled.

Integ
This module integrates an nth-order system of initial-value ODE's (see e.g. Eqn. 2.55).

Derv
This module is called by Integ and calculates the right hand side of the ODE's as a function of constant parameter values and current process input values.

Print
Calculates the model output equations as a function of the simulated crystal size distribution further different numerical values are displayed on the screen.

The interconnection between these models and the flow of the main parameters is explained in Fig. F.1. The basic program structure, (i.e. the modules MAIN, DERV,
PRINT INTEG and INITIAL) is obtained from the DSS2 software package [184]. However, different extensions are made: A user interface is added with which all parameters can be changed interactively or read from a standard initial file (as in the standard DSS/2 implementation) and a direct interface with Matlab is implemented.

Matlab has served as a host for the simulator. This has the advantage that the model can be invoked iteratively, which is important for the robustness studies reported. In addition all graphic and signal analysis tools are directly available. Three different execution levels are programmed in the simulator which can be invoked from Matlab with a control parameter (1,2 or 3):

1. Go to INITIAL and set all constants at user chosen values

2. Initialize constant parameters needed for simulation, estimate the stationary process condition and store the results in an array, or linearize the model and write the linearized model to a file.

3. Simulate the linear or nonlinear equations for only one print interval $t_p$ and store the intermediate results in the results array $y_{res}$. The print interval is usually chosen equal to the sampling interval, to compare simulation results directly with historical process data. The latter is especially important for parameter estimation.
The user interactive menu programmed in the module INITIAL contains 11 main menu items that include several sub-items:

1. **Process input parameters.**
   Indicate whether process input parameters from the host (i.e. Matlab or MatrixX) or local (constant) values are used; select whether the model should operate in closed-loop as an observer. In the latter case the error correction signal is received from the host.

2. **Initial condition.**
   Select a stationary state or an analytical size distribution as initial condition for simulation. Calculation of the stationary state is described in Section 2.3.6.

3. **Integration parameters.**
   Change default values for: the simulation time step, the final time; the integration scheme (see also, [184]), and the intervals at which simulation results are outputted.

4. **Differentiation parameters.**
   Change the defaults values for: the number of lumping elements, the lower and upper size bounds of the simulation grid, and the discretisation scheme (i.e. a second or fourth order method of lines scheme).

5. **Classification parameters.**
   Here the empirical parameters \( \theta_C \) for the efficiency functions for fines and product classification are chosen.

6. **Growth models.**
   In this menu different types of length dependent growth curves and the empirical parameters theirin are chosen. Furthermore growth according to the Class I or Class II model can be selected.

7. **Kinetic parameters.**
   All default values for the parameters \( \theta_K \) related to crystallization kinetics (see Chapter 2) can be changed in this menu.

8. **Process conditions.**
   The parameters related to the stationary process conditions, i.e. the flows and the size of the vessel and the size of the annular zone are given here.

9. **Sensor model.**
   Select the output model (i.e. sensor model matrix) and enter a
name of the file including the model; select the location of the sensor, e.g. the product discharge or the well-mixed crystallizer vessel.

10. **Linear or nonlinear model.**

   Use the linear or the nonlinear model for simulation; select whether the linear model has to be written to a file. The latter option enables the use of linearized models in state-space format in Matlab, as is done extensively in this work.

11. **Miscellaneous.**

   Change defaults values for the scaling of physical parameters and the nucleation model (see Eqn. 5.4).

The full order linear models outputted to a file could be read into Matlab easily with a special purpose interface program. The high order models were used to analyze the stability of the system, and for state estimator design. Reduced order versions of the linear model were used for controller design.
Appendix G

Laplace domain solutions

To analyze some system properties a simple transport equation is used which describes the CSD dynamics in a finite size domain \((0 < x < x_e)\). This equation is derived from the general population balance model described by Randolph [197]. It is assumed that the growth rate of the particles is constant and that the probability that a particle with a size \(x\) is removed from the crystallizer volume is given by a time independent removal efficiency function \(h(x)\).

\[
\frac{\partial n(x,t)}{\partial t} + g \frac{\partial n(x,t)}{\partial x} + \frac{u_1(t)}{V} h(x)n(x,t) = 0, \quad 0 < x < x_e \quad (G.1)
\]

The size \(x_e\) is the final size of the region that can be influenced with the fines removal rate. The boundary condition is formulated as an independent disturbance input \(u_2(t)\):

\[
n(x = x_0, t) = u_2(t) \quad (G.2)
\]

The flow \(u_1(t)\) is considered as the control input. The initial condition is given by:

\[
n_s(x, t = t_0) = n_0(x). \quad \text{We consider that the behaviour of the process can be measured with a sensor that measures the population of fines with a certain time independent efficiency function } k(x), \quad \text{(cf. Eqn. 2.41). For an ideal particle counter which measures monosized particles with a size } x_a, \quad \text{the detection efficiency will be a dirac function.}
\]

A solution for this model can be derived after linearization of Eqn. G.1, followed by a Laplace transform which will lead to a linear first order distributed boundary value problem which can be solved analytically. When the solution to this boundary value problem is written as transfer functions between inputs and outputs in the frequency application of the classical Bode and Nyquist frequency domain analysis techniques can be applied to analyze different properties of this system.

The stationary solution of Eqn. G.1 is given by:

\[
\bar{n}(x) = \bar{u}_2 \exp \left( \frac{-\bar{u}_1}{g} \int_{\xi=0}^{x} h(\xi) d\xi \right). \quad (G.3)
\]
Eqn. G.1 is linearized by considering small disturbances around the stationary solution:

\[ n(x, t) = \bar{n}(x) + \Delta n(x, t) \quad \text{(G.4)} \]
\[ u(t) = \bar{u} + \Delta u(t) \quad \text{(G.5)} \]

after substraction of the stationary values this gives:

\[ \frac{\partial \Delta n(x, t)}{\partial t} + g \frac{\partial \Delta n(x, t)}{\partial x} + h(x) \bar{n}(x) \Delta u_1(t) + \bar{u}_1 h(x) \Delta n(x, t) = 0, \quad \text{(G.6)} \]

and the boundary condition:

\[ \Delta n(0, t) = \Delta u_2(t) \quad \text{(G.7)} \]

The Laplace transformation rules for the variables \( u(t) \) and \( n(x, t) \) are given by:

\[ \mathcal{L}\{u(t)\} = U(s) = \int_0^\infty u(t)e^{-st}dt \quad \text{(G.8)} \]
\[ \mathcal{L}\{n(x, t)\} = N(x, s) = \int_0^\infty n(x, t)e^{-st}dt \quad \text{(G.9)} \]
\[ \mathcal{L}\left\{ \frac{\partial n(x, t)}{\partial t} \right\} = sN(x, s) - n(x, 0) \quad \text{(G.10)} \]

The Laplace transform of Eqn. G.6 is written as:

\[ [s + \bar{u}_1 h(x)] \Delta N(x, s) + g \frac{\partial \Delta N(x, s)}{\partial x} = -h(x) \bar{n}(x) \Delta U_1(s) + \Delta n(x, 0), \quad \text{(G.11)} \]

with the boundary condition,

\[ \Delta N(0, s) = \Delta U_2(s) \quad \text{(G.12)} \]

This is a boundary value problem with non-homogeneous boundary conditions. Use is made of a solution to a similar problem for a continuous furnace, which is described in detail by Franke [76]. After substitution of the stationary solution \( \bar{n}(x) \) (Eqn. G.3), the solution is given by:

\[ \Delta N(x, s) = G_1(x, s) \Delta U_1(s) + G_2(x, s) \Delta U_2(s) \quad \text{(G.13)} \]

\[ G_{x1}(x, s) = -\frac{\bar{u}_2}{g} \exp \left( -\frac{\bar{u}_1}{g} \int_{\xi=0}^{x} h(\xi)d\xi \right) \int_{\zeta=0}^{x} h(\zeta) \exp \left( \frac{s}{g}(\zeta - x) \right) d\zeta \quad \text{(G.14)} \]

\[ G_{x2}(x, s) = 1 - \frac{1}{g} \int_{\xi=0}^{x} \left[ \exp \left( \frac{\bar{u}_1}{g} \int_{\xi=x}^{\zeta} h(\xi)d\xi \right) \exp \left( \frac{s}{g}(\zeta - x) \right) [s + \bar{u}_1 h(\zeta)] \right] d\zeta \quad \text{(G.15)} \]
Fig. G.1: Block scheme of simple population balance model in closed-loop.

This relation shows the important property that the CSD dynamics observed with a certain will depend on the crystal size region at which the sensor is focused. If it is assumed that the output is calculated with Eqn. 2.41, the transfer functions between process inputs and outputs are obtained from:

\[
\Delta Y(s) = \int_{x=0}^{x^*} k(x) \Delta N(x, s) dx
\]

\[
= \int_{x=0}^{x^*} k(x) G_{x1}(x, s) dx \Delta U_1(s) + \int_{x=0}^{x^*} k(x) G_{x2}(x, s) dx \Delta U_2(s)
\]

\[
= G_1(s) \Delta U_1(s) + G_2(s) \Delta U_2(s) \quad (G.16)
\]

With the equations presented here, the transfer functions \(G_{x1}, G_{x2}, G_1\) and \(G_2\) enable the analysis of the process and controller performance in the Laplace domain as a function of the removal and detection efficiency functions \(k(x)\) and \(h(x)\).

For control purposes, the described system can be studied in a closed-loop setting where a SISO controller \(G_c(s)\) is introduced to act on the input \(u_1\) on the basis of measurements \(y\), derived from a sensor characterized by a detection efficiency function \(k(x)\). A block-scheme is provided in Fig. G.1.

The closed-loop properties can be studied in the frequency domain using the following transfer functions:

\[
\Phi_1 = \frac{Y(s)}{Y_R(s)} = \frac{G_c(s)G_1(s)}{1 + G_c(s)G_1(s)} \quad (G.17)
\]

\[
\Phi_2 = \frac{Y(s)}{U_2(s)} = \frac{G_2(s)}{1 + G_c(s)G_1(s)} \quad (G.18)
\]

\[
\Phi_2 = \frac{Y(s)}{U_2(s)} = \frac{G_2(s)}{1 + G_c(s)G_1(s)} \quad (G.19)
\]

These relations are obtained from the above presented equations.
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Glossary of Symbols

Symbols not included in this list are defined locally and need no further explanation.

Symbols related to crystallization

- $A$: cross sectional area, [m$^2$]
- $B$: nucleation rate, [g/m-m$^3$]
- $C$: solute concentration, [kg/m$^3$]
- $C_s$: saturation concentration, [kg/m$^3$]
- $c_p$: specific heat, [kJ/(kg °C)]
- $G_k$: kinetic crystal growth rate, [m/s]
- $G_x$: length dependent crystal growth rate, [m/s]
- $G_e$: effective crystal growth rate, [m/s]
- $h$: particle removal efficiency function, [-]
- $k$: particle detection efficiency function, [-]
- $k_{cryst}$: heat of crystallization, [kJ/kg]
- $H_v$: specific enthalpy of vapor, [kJ/(kg)]
- $k_y$: constant related to Class II crystal growth, [-]
- $k_z$: constant related to Class I crystal growth, [-]
- $k_v$: volume based crystal shape factor, [-]
- $L$: vector containing light energy values from array detector, [-]
- $M_t$: magma density, [kg/m$^3$]
- $m_j$: j-th moment of crystal size distribution, [g/m$^3$]
- $n$: population density, [g/m$^3$]
- $n_0$: population density at smallest size, [g/m$^3$]
- $N$: cumulative number function, [z/m$^3$]
- $obsc$: obscuration, [-]
- $p$: empirical parameter, [-]
- $P$: power, [KW]
- $P_{cryst}$: heat consumption for crystallization, [KW]
- $Q$: volumetric slurry flow, [m$^3$/s]
- $Q_{ext}$: light extinction coefficient, [-]
- $Q_M$: solids mass flow, [kg/s]
Glossary of Symbols

$qr$ quartile ratio, [-]
$r$ retention time, [s]
$T$ temperature, [$°C$]
$t$ time, [s]
$y_{11}$ first principal component of dynamic light energy vector, [-]
$V$ volume of crystallizer vessel, [m$^3$]
$x$ crystal size, [m]
$x_c$ cutsize, [m]
$X_k$ $k\in\{25, 50, 75\}$ quartile of distribution, [m]
$X_{43}$ mass based mean crystal size, [m]

Greek

$\alpha$ aspect ratio of crystals, [-]
$\Delta$ difference operator, [-]
$\gamma$ flow split factor, [-]
$\gamma_d$ dilution ratio, [-]
$\rho$ density, [kg/m$^3$]
$\tau$ residence time, [s]
$\mu$ absolute viscosity, [kg/m · s]
$\Psi_1$ mass production rate due to crystal birth, [kg/s]
$\Psi_2$ mass production rate due to crystal growth, [kg/s]
$\varepsilon$ void fraction, [-]
$\xi$ constant factor related to specific heat, [kJ/(kg · °C)]

Sub- and superscripts

$a$ area based
$c$ solid phase (crystals)
$ex$ external
$f$ fines
$F$ filter
$HC$ heat exchanger
$i$ input flow
$imp$ impeller
$in$ internal
$k$ kinetic
$l$ liquid phase
$l$ length
$m$ measured
$p$ product
$pf$ product classifier feed
Glossary of Symbols

ff fines classifier feed
r return or recycle
s sensor
v volume based
vap vapour
x crystal size related
w width (of crystal)

System theoretic

A system matrix
B input matrix
C output matrix
D transfer matrix
e error signal
f process model
f_b model for boundary condition
F system matrix
g model measurement system
{g_k, b_k} expansion coefficients
{f_k} orthonormal basis functions
(G, H) model transfer functions
H observation matrix
G input matrix
k_p proportional gain PI controller, [-]
K observer feedback gain matrix
J criterion
P covariance of the estimated state
Q weight matrix states in LQG criterion
R weight matrix inputs in LQG criterion
R correlation matrix
T_i integral or reset time
T sampling interval
u process input signal
v process noise
w measurement noise
V process noise covariance matrix
W measurement noise covariance matrix
y process output
z principal component vector
Greek

Δ  difference operator
ε  residual
λ  eigenvalue
σ  standard deviation
Σ  covariance matrix
θ  set of unknown parameters
ζ  integration constant

Sub- and superscripts

c  controller
f  filter
R  reduced

Abreviations

CSD  Crystal Size Distribution
DMC  Dynamic Matrix Control
FDI  Fault Detection and Isolation
FIR  Finite Impulse Response
MIMO  Multi Input Single Output
MPC  Model Predictive Control
PID  Proportional Integral Differential
PC  Principal Component
PLC  Programmable Logic Controller
PMC  Process Monitoring and Control
PCA  Principal Component Analysis
PRBS  Pseudo Random Binary Sequence
RBS  Random Binary Sequence
SISO  Single Input Single Output
SL  Sample Location number j
UNIAK  Universal Instrumentation and Automation of Crystallizers
Samenvatting

Regeling en dynamische modellering van industriële suspensie kristallisatoren

R.A. Eek

Industriële kristallisatie vanuit een suspensie is nog steeds een moeilijk beheersbaar proces. De kristalgrootteverdeling (KGV) van de product kristallen, geproduceerd met een continue kristallisator, vertoont vaak slecht gedempt of oscillerend gedrag, terwijl het kristalproduct van zowel batchgewijs als continu gedreven kristallisatoren vaak niet aan de gewenste specificaties voldoet. Zowel het oscillerende gedrag als de constante afwijking verlagen de werkingsgraad van vast-vloeistof scheiders in de "downstream" van het proces, en verschillende eigenschappen die bij verdere verwerking van kristallen van belang zijn, bijv. het oplossen van kristallen. Tevens leiden KGV fluctuaties tot fluctuaties in de groeisnelheid van kristallen. Dit kan de zuiverheid van het product verlagen, bijvoorbeeld doordat bij te hoge groeisnelheden te veel vloeistof insluitingen worden gevormd.

Het onderwerp van dit proefschrift is het ontwerp van regelsystemen voor kristalgrootteverdelingen om daarmee een verbetering van de de productkwaliteit en de economische werkingsgraad van kristallisatieprocessen te bewerkstelligen. Een belangrijke eis hierbij is dat het te ontwerpen regelsysteem moet voldoen aan de eisen van de procesindustrie.

Als eerste wordt het dynamische procesgedrag gemodelleerd op grond van de "first principles" van kristallisatie. Dit levert een geparametrisernd, niet-lineair populatiebalance model waarmee het dynamische gedrag van de kristalgrootteverdeling kan worden beschreven als functie van kristalgeboorte, kristalgroei, attritie en geklassificeerde verwijdering van zowel kleine als grote product kristallen. Verschillende numerieke schema’s worden afgeleid die noodzakelijk zijn voor de simulatie van dit model, voor de analyse van de dynamische proceseigenschappen en voor het afleiden van lage orde modellen. Lage orde modellen zullen gebruikt worden voor het regelaarontwerp.

De empirische parameters in het rigoureuze dynamische model hebben een grote onzekerheid. Een belangrijke stap daarom is om een goede schatting van de model parameters te verkrijgen en het model te verifiseren op grond van experimentele proces
data. Hiertoe zijn relevante ingangs-uitgangs data gemeten aan een semi-industriële kristallisator die voorzien is van actuatoren voor de proces dynamica en van sensors voor het on-line meten van de KGV dynamica. Voor the meten van de KGV is gebruik gemaakt van voorwaartse licht verstrooiing. Bovendien is er gebruik gemaakt van een eenvoudige deeltjesstomer voor het meten van de "fines" dichtheid. Extra aandacht is besteed aan de monstername, en aan de interpretatie van de ruwe gemeten data.

De verificatie van het model met experimentele data is in drie stappen uitgevoerd. In de eerste stap zijn een aantal specifieke model aannamen geverifieerd door gebruik te maken van eenvoudige experimenten. In een tweede stap worden parameters voor de kristallisatie kinetiek geschat op grond van proces opstart data, zoals die in databases van industriële processen voorhanden zijn. Deze opstart data zijn gemeten aan de pilot kristallisator bij zowel geclasseerde als ook niet geclasseerde product verwijdering en bij verschillende proces kondities. In de derde stap wordt gebruik gemaakt van systeemidentificatie. Hierbij wordt het proces geëxciteerd door testsignaal die toegevoegd zijn aan de procesingangen. Het blijkt dat deze experimenten principieel in gesloten lus uitgevoerd dienen te worden om te voorkomen dat het oscillerende gedrag van de KGV dynamica de excitatie met de ingangssignalen overheerst. Met de verkregen meetdata wordt het nietlineaire proces model geverifieerd en wordt tevens een empirische ingangs-uitgangs model geschonken. Hierbij is met name aandacht besteed aan het probleem dat directe identificatie van modellen, op grond van gesloten lus data, tot model fouten kan leiden, omdat de ruis op de ingangs- en uitgangssignalen gecorreleerd is. Zowel het geïdentificeerde model als het geoptimaliseerde rigoureuse model zijn voor het regelaarontwerp gebruikt.

Met het toepassen van voorwaartse licht verstrooiing als meettechniek voor kristalgrootteverdelingen zijn de grootheden die voor regeling relevant zijn niet direct beschikbaar. Daarom is er een toestandsschatter ontwikkeld waarmee de relevante grootheden op grond van het dynamische model, ruimachtige proces ingangssignalen, en de gemeten ruwe sensor data kan worden geschät. Het gedrag van deze dynamische toestandschatter is vergeleken met een eenvoudige schatter die gebruik maakt van een geschatte inverse van het sensor model.

Voorafgaand aan het regelaarontwerp is de regelbaarheid van het proces bestudeerd. Het belangrijkste doel hierbij is om vast te stellen welke vrijheidsgraden er in het proces aanwezig zijn en welke combinaties van ingangs- en uitgangssignalen tot een effectieve beheersing van deze vrijheidsgraden leidt. Er is aangetoond dat voor de pilot kristallisator, die bedreven wordt met ammonium sulfaat, een goede regelbaarheid slechts verkregen kan worden idien de sensor gefocuseerd wordt op deeltjes met een grootte boven 30 micrometer. Het meten van deeltjes onder deze grens veroorzaakt een rechter half vlak nulpunt in de proces overdracht die de regelbaarheid van het proces sterk beperkt. Goed gesloten lus gedrag is echter gerealiseerd met een deeltjes teller die het aantal kristallen in het grootte gebied van 40 tot 100 micro meter telt.

Enkele richtlijnen zijn gegeven voor het ontwerp van stabiliserende regelaars voor
industriële kristallisatoren. Deze zijn beperkt tot eenvoudige enkelvoudige PI regelaars waarbij gebruik wordt gemaakt van fines vernietiging als actuator en een eenvoudige sensor die een scalaire waarde gerelateerd aan het aantal kleine deeltjes als uitgangssignaal heeft. Kort wordt uiteengezet hoe dit principe toegepast kan worden op industriële kristallisatoren. Met experimenten uitgevoerd met de pilot kristallisator is de validiteit van de voorgestelde regelaar aangetoond.

Het laatste onderwerp dat wordt behandeld is het ontwerp van een model gebaseerde regelaar. Hierbij is van een multivariabele voorspellende regelaar gebruik gemaakt. Deze regelaar houdt expliciet rekening met beperkingen in de procesingangen en uitgangen en bevat tevens een toestandsschatter. Met simulatie en experimentele resultaten is aangetoond dat hiermee een goede beheersing van het proces dynamische gedrag kan worden verkregen. Echter uit een vergelijking met het gedrag van een multi-loop PI regelaar blijkt dat ook met deze regelaar een vergelijkbaar gesloten lus gedrag kan worden verkregen, ook wanneer er beperkingen in het werkgebied van de actuator aanwezig zijn.
Curriculum Vitae

Rob Eek is geboren op 5 augustus 1965 in Eemnes en getrouwd met Anja van der Graaf.