A DESIGN PROCEDURE AND PREDICTIVE MODELS FOR SOLUTION CRYSTALLISATION PROCESSES

 Sean K. Bermingham, Andreas M. Neumann, Herman J.M. Kramer, Peter J.T. Verheijen, Gerda M. van Rosmalen and Johan Grievink
 Process Systems Engineering Group and Laboratory for Process Equipment Delft University of Technology, 2628 BL Delft, The Netherlands

Abstract

Useful components and deficiencies of conventional crystallisation design procedures and models are identified. A systematic procedure consisting of a hierarchical decomposition into design of the product, task, flowsheet and individual crystalliser is presented. At each level the relevant specifications, variables and necessary knowledge are identified. For the flowsheet and crystalliser level a predictive crystallisation model is presented to estimate kinetic parameters from experimental CSD data and to analyse the behaviour of design alternatives. Currently, the procedure is applicable to cooling, flash-cooling and evaporative crystallisation. The developed model is restricted to systems with little agglomeration. Validation of the proposed design procedure and models requires industrial case studies.

Keywords

hierarchical design, scale-up, compartmental models, draft tube baffle crystalliser.

Introduction

Crystallisation involves the formation of one or more solid phases from a fluid phase or an amorphous solid phase. It is applied extensively in the chemical industry, both as a purification process and a separation process. The main advantage of crystallisation over distillation is the production of substances with a very high purity, at a low level of energy consumption, and at relatively mild process conditions.

Although crystallisation is one of the older unit operations in the chemical industry, the design and operation of crystallisation processes still pose many problems.

Many crystallisation plants frequently produce crystals, which do not satisfy the defined quality specifications. For instance, an excess of fine particles will typically result in poor filterability characteristics. Consequently increasing the cost of the downstream solid/liquid processing. Another example is the inclusion of mother liquor. After solid/liquid separation and drying of the crystalline product, e.g. during transportation or storage, mother liquor may seep from broken crystals. Subsequent re-crystallisation may cement the crystals together; a process referred to as the caking of crystals.

Operational problems also constitute a large portion of the problems encountered in crystallisation processes. Firstly, scale growth or crystal deposition on heat exchanger surfaces often limits plant availability significantly. Secondly, plant availability may also be reduced by pipe blockages as a result of scale growth or high solids concentrations. Finally, many crystallisation processes suffer from open loop unstable behaviour.

Despite the importance of crystallisation, there is a relative lack of systematic design procedures and predictive models to help avoid or overcome the beforementioned problems. This relative lack in comparison with vapour/liquid processes is a major omission for the chemical engineering profession (Villadsen, 1997). It is however not a surprising omission, as the understanding of crystallisation processes, and of solids processes in general, is typically a degree more complex than that of vapour/liquid processes. This added complexity mainly results from:

- The fact that the product quality specifications cannot be solely defined in terms of chemical and phase composition. A crystalline product is also characterised by its size distribution, morphology, polymorphism and the amount of strain in the crystal lattice.
- The problems in dealing with the thermodynamic models of solid/liquid/-vapour systems and even of liquid/vapour systems when electrolytes are involved.
- The difficulties in predicting the hydrodynamics of a multi-phase flow as a function of crystalliser and impeller geometry, operating conditions, crystal properties and crystal concentrations.
- The fact that the rates with which crystals are born, grow, dissolve, are attrited, break, agglomerate, etc. are not only a function of liquid phase process conditions but also of distributed crystal properties such as size, surface structure and internal energy.

Knowledge of crystal properties, thermodynamic properties, hydrodynamic conditions and particle mechanics as well as understanding of their interactions is essential to predict the spatially distributed, size dependent and time dependent crystallisation kinetics. After all, it is the kinetics that ultimately determines the properties of the crystalline product. At present it is impossible to derive this knowledge and understanding from first principle models only. Consequently, heuristics, tabulated data, laboratory and pilot plant scale experiments still constitute a major part of the domain knowledge necessary for the design and optimisation of crystallisation processes.

However, improved design and operation of does systems not only crystallisation require developments in various fields of fundamental knowledge. Equally important is the availability of a systematic design procedure, which can simplify the design problem, can help organise the design tasks and will consequently improve the quality of designs and speed up the design process. An additional benefit of a systematic design procedure is reproducibility of the design process, which is essential to identify any remaining errors in the applied domain knowledge and/or design procedure.

It is our opinion that such a design procedure is not available yet, although some very relevant work has been done in both the fields of process design and crystallisation.

Based on the pioneering work of Douglas (1985) concerning the conceptual design of vapour/liquid processes, Rajagopal et al. (1992) developed a hierarchical design procedure for solids processes in general. More specific design procedures have been developed for fractional crystallisation (Dye and Ng, 1995), reactive crystallisation (Berry and Ng, 1997) and the interactions between the crystallisation step and the downstream processing (Rossiter and Douglas, 1986). These procedures mainly focus upon the synthesis and (economic) evaluation tasks of the design process. The lack of predictive models for analysis and optimisation of design alternatives limits widespread application of systematic design procedures for crystallisation processes.

Valuable work with respect to predictive models includes the development of kinetic models based upon first principles (Gahn et al., 1997), hydrodynamic studies of crystallisers (de Jong et al., 1998) and compartmental modelling to account for the interactions between kinetics and hydrodynamics (Kramer et al., 1998).

Scope of this paper

In the first part of this paper we will present a hierarchical procedure for the conceptual design of solution crystallisation processes. The proposed hierarchy consists of four design levels. The first two design levels have a product engineering character, whereas the last two design levels have a process engineering character. At each level of the design procedure we will (re-)consider the design specifications, design variables and the domain knowledge necessary to synthesise, analyse and optimise design alternatives. The design procedure is intended to cover various scenarios as regards the destination of the crystalline product (main product, byproduct or waste product) and the role of the crystallisation process (conversion, separation or purification).

The second part of this paper focuses on predictive models that are necessary for the design of crystallisation processes, but are not readily available. This is especially the case for the third and fourth level of the design procedure, where the performance of respectively flowsheet alternatives and individual crystalliser alternatives need to be analysed and optimised. For this purpose two model structures have been developed. The kinetic models required by these model structures cannot be determined purely from first principles and can only rarely be found in literature. The newly developed model structures are therefore intended for both the domain knowledge acquisition task, i.e. extracting intrinsic kinetic parameters from experimental data, as well as the analysis/optimisation

tasks of the design process. We will illustrate their use in the acquisition of domain knowledge (parameter estimation) and for analysis purposes (explorative simulations). The application of predictive models at levels one and two of the design procedure is not treated in this paper.

In its present form, the design procedure is applicable to cooling, flash-cooling and evaporative crystallisation processes. However, the developed model structures are currently still restricted to crystallisation systems, which exhibit negligible agglomeration.

A conventional crystallisation design procedure

In this section we will present a generalised picture of the conventional procedures for the design of a crystallisation process.

Synthesis

The starting point is the specification of the available feed composition, required product purity, yield, production capacity and a rough measure for the crystal size distribution (CSD), such as the median crystal size or the volume fraction crystals below or above a certain size. The next step involves collecting thermodynamic data and physical properties for the crystallisation system. Subsequently the synthesis phase is entered, which involves selection and sizing issues. The crystallisation method is mainly determined by the thermodynamics of the solid/liquid equilibrium. A heuristic scheme for this selection process is given in Fig. 1.



Figure 1: Crystallisation method selection (after Kramer et al. (1998)).

Another thermodynamically determined choice is the use of a certain solvent or additives to obtain a desired morphology. Subsequent selections involve the number of crystallisation steps, operation mode and number of stages. The first choice, the need for one or more recrystallisation steps is mainly determined by the amount of impurities in the feed and the desired final

product purity. The operation mode, batch or continuous, is typically dictated by the production capacity and the desired shape of the CSD. Multiple stage operation is of interest for evaporative crystallisation processes with a high energy consumption. The next stage of the design process involves selection of crystalliser type and sizing the crystalliser. Crystalliser type selection is often influenced by company preferences and experiences. The crystalliser volume, V, is a simple function of the desired production capacity, P, the maximum solids concentration that the equipment can handle, ϕ , the desired median crystal size, L_{50} , and an estimate for the crystal growth rate, G:

$$V = \frac{P}{\phi \cdot \rho_c} \frac{L_{50}}{3.67 \cdot G} \tag{1}$$

The area required for heat exchange is a function of the production capacity, the selected crystallisation method and the system's thermodynamics.

Analysis and optimisation

At this point a so-called base case design has been realised. Now the importance assigned to the CSD determines whether any effort needs to be put into the analysis and optimisation of the base case design with respect to the CSD. These design tasks require relations between nucleation and growth rates of crystals on the one hand and crystalliser geometry and operating conditions on the other hand. Over the years many kinetic models have been proposed for this purpose, e.g. Ottens et al. (1972), Ploß et al. (1989) and Ó Meadhra etal. (1996). All these models are power law models, ranging from purely empirical to partly fundamental. As a result, these models should have a limited predictive value for scale-up and design, i.e. when applied to another scale of operation, crystalliser type or operation mode than the configuration used to estimate the model parameters.



Figure 2: The 22 litre DT (left) and 1100 litre DTB evaporative crystalliser (right) at the Laboratory for Process Equipment.

We will illustrate this below with experimental results obtained on a 22 litre draft tube (DT) and 1100 litre draft tube baffle (DTB) crystalliser and the model framework of Ó Meadhra.

On both crystallisers experiments were performed with a residence time of 75 minutes, a specific heat input of $120 \text{ kW} \cdot \text{m}^{-3}$ and similar impeller tip speeds. Both crystallisers were operated continuously in an evaporative mode at a constant temperature of 50°C using ammonium sulphate/water as the model system. Differences in process conditions between the two experiments are summarised in Table 1. A more comprehensive coverage of these experiments is given by Bermingham et al. (1998).

Table 1: Differences in process conditions between experiments DT22 and DTB1100.

	DT22	DTB1100
Impeller frequency [s ⁻¹]	16.7	5.33
Tip speed impeller [m.s ⁻¹]	7.3	8.1
Circulation flow [m ³ .s ⁻¹]	0.015	0.200
Turnover time [s]	1.5	5.6
Mean spec. power input [W.kg ⁻¹]	4.5	1.4
Fines withdrawal flow [m ³ .s ⁻¹]	-	0.002

Neumann et al. (1998a) used experiment DT22 to estimate the attrition parameters of the Ó Meadhra model. The measured trend of the median crystal size and the trend simulated with the estimated parameters are shown in Fig. 3. The steady-state values are in good agreement, but the dynamics of the fitted trend are more pronounced than observed experimentally.



Figure 3: measured and fitted trend of the median crystal size for experiment DT22.

The predictive capabilities of the Ó Meadhra kinetic model are tested by performing a simulation using the conditions for experiment DTB1100 and the parameters estimated from experiment DT22. For this simulation (sim. 2) the model used for the simulation of DT22 (sim. 1) was extended with a fines classification model and an ideal fines dissolver. More simulation features are given in the appendix.



Figure 4: measured and predicted trend of the median crystal size for experiment DTB1100.

Comparison of the median crystal size trend from simulation 2 and from experiment DTB1100 reveal a difference in dynamic behaviour and a difference of approximately 160 μ m in expected steady state values (see Fig. 4). This difference may result from a number of factors, which are especially related to scale-up and crystalliser type:

- The spatial distribution of process conditions which is not accounted for, i.e. ideal mixing assumption.
- Incomplete dissolution of the fines removed from a DTB crystalliser. Conventional models assume complete dissolution.
- The kinetic model does not contain the right mechanisms to account for changes in nucleation rates due to differences in scale, geometry or operating conditions.

Evaluation of the conventional design procedures

Numerous heuristics exist for the development of a base case design. For mainly two reasons, heuristics for selection issues are also useful in a systematic design procedure. First of all, many heuristics cannot yet be replaced by fundamental knowledge. Secondly, the application of heuristics is usually relatively simple and rapid.

Reliable tools for the analysis and optimisation of the base case design with respect to the CSD are not readily available. Due to the absence of such tools full scale industrial processes cannot be designed on the basis of laboratory scale kinetic data only. In practice, additional experiments are therefore performed at one or two intermediate scales. Unless similar crystallisation processes exist, in which case new plants are often copied from existing ones. The development of reliable models for scale up purposes is thus essential in the development of a systematic design procedure for crystallisation processes.

Finally, when optimisation of the base case design fails to produce an adequate design, a design alternative with a different structure needs to be synthesised. To our knowledge none of the conventional methodologies cover this part of the design process.

A systematic design procedure

The main incentive for the development of a systematic design procedure is the need for crystallisation process designs of a consistently high quality (design effectivity). A second incentive is the available time-to-market. is which decreasing continuously. A systematic design procedure can help speed up the design process, e.g. by removing the need for pilot scale experiments for scale-up purposes (design efficiency). An additional benefit of a systematic design procedure was already stated in the introduction, namely the reproducibility of the design process (traceability of design decisions and rationales). This is essential to improve on existing designs and identify any remaining errors in the applied domain knowledge and/or design procedure. A final objective is to create mutual awareness of the tasks that people have in a design team.

To achieve the above-mentioned aims, we propose a design hierarchy (see Table 2) in order to simplify the design problem and help organise the design tasks. This decomposition consists of one level at which the initial design specifications are formulated and four design levels.

0	Initial design specifications
Ι	Design of the crystalline product
II	Physical/chemical design of the crystallisation task
III	Flowsheet design of the crystallisation process
IV	Design of a crystallisation stage

Table 3 shows the types of requirements considered at level 0. Depending on the destination of the crystalline product (main product, by product or waste product) and the role of the crystallisation process (conversion, separation or purification) some design specifications will be deemed necessary or desirable and others will be denoted irrelevant. Design process requirements are not treated in the remainder of this paper.

Design levels I through IV are aimed at finding design alternatives that meet the initial design specifications. As the designer progresses from one level to the next the emphasis shifts from product design (levels I and II) to process design (levels II and III) and ultimately to crystalliser design (level IV). In contrast with methodologies as proposed by Douglas (1985) the degree of detail does not automatically increase with each level. For product related issues it actually decreases, and for process and equipment related topics it increases.

Table 3: Level 0 - Initial design specifications.

Product performance requirements

In the crystalliser: no flotation, suspendability Downstream handling: filterability, washability, dryability, dissolution rate, pneumatic handling, freedom from dust, flowability, mechanical strength *Customer application*: no caking in storage, dissolution rate, mechanical strength, freedom from dust, bulk density or porosity, aesthetic appearance

Process requirements

production capacity, feed composition, yield, energy consumption, controllability, resiliency, availability, SHE considerations, battery limits and conditions

Design process requirements

design budget, time to market, in-house or licensed technology, available skilled design staff

At all four design levels the same tasks are performed, i.e. definition of design space and specifications, assessing domain knowledge, synthesis, analysis, evaluation and optimisation. (see Fig. 5). These tasks will be discussed briefly below and further on they will be exemplified at each level.

The first step at each level is to make an inventory of applicable design specifications and to identify the design space. The specifications consist of relevant initial design specifications from level 0 and eventual design specifications propagated from a previous level. The design space is defined by the available design variables and operational variables.

The second step is to gather domain knowledge which relates the design variables and operational variables to the behaviour of a design alternative. This knowledge may consist of heuristics, experimental data and behavioural models. If parts of the domain knowledge are considered inadequate, additional experimental and modelling efforts may be required.

The third step, the synthesis task involves the creation of design alternatives. A design alternative is characterised by its structure and its scale. In this design procedure we will classify design and operational variables which determine the structure as discrete, and those that define the scale as continuous design and operational variables.

The fourth step is the analysis of the design alternatives. Subsequently, the set of behavioural results is evaluated (step five). The evaluation first of all concerns the compliance of the design alternatives' behaviour with the design specifications. If this evaluation is positive, the design alternative is propagated to a next design level, accompanied by new design specifications. These propagated specifications are in fact design variables and operational variables set at a previous level. If the design alternatives fail the evaluation step, their performance is optimised by modifying their structure and scale, i.e. by returning to the synthesis step. Normally, a screening of the economic potential will be part of any evaluation step. However, such screening is not covered in this paper. Secondly, the evaluation phase may be used to judge the quality of the applied domain knowledge. If a part of the knowledge is considered inadequate, additional experimental and modelling efforts may again be called for.



Figure 5: Typical sequence of design tasks and the outcome of these tasks. The superscripts refer to the column numbers of Tables 4 through 7.

A comprehensive execution of all the abovementioned tasks at all four design levels, while considering all the design specifications mentioned in this paper, will obviously lead to very lengthy design trajectories. Assigning importance to the various initial design specifications is thus crucial, as the specifications taken into consideration largely determine the amount of effort put into each design level.

Design of the crystalline product

At this design level (see Table 4) the aim is to determine which product composition is required to meet the product performance criteria, i.e. the product related initial design specifications. For instance, filterability and freedom of dust are strongly related to the content of fine particles, aesthetic appearance is usually related to particle size uniformity, the bulk density is determined by the polymorphism and morphology of the crystals and the caking tendency in storage is related to the liquor inclusion content.

Most of the relevant domain knowledge for this level belongs to the field of particle technology, and has

a strong empirical character. This is mainly due to the fact that product performance criteria are often equipment specific and hence cannot be defined generically. The development of fundamental knowledge in this field is further complicated by the need to account for distributed properties. Because a collection of particles can rarely be described by one size, one morphology, one purity, etc. Depending on the importance of the product performance criteria, much experimental work may be needed for this design level.

The design alternatives, i.e. sets of design variables, are propagated to design level II, where they are treated as design specifications.

Physical/chemical design of the crystallisation task

The design specifications of level II, the physical/chemical design of the crystallisation task, are composed of process requirements from level 0 and propagated product composition characteristics from level I. Consult Table 5 for a list of specifications.

The polymorphism and morphology of the crystalline product are influenced by the choice of solvent and additives. The domain knowledge required for this selection issue was traditionally obtained from experimental work but is increasingly being replaced by molecular modelling. These tools can perform first principles calculations for the adsorption energy of a component on a specific crystal face.

The same knowledge is also essential to determine the sensitivity of the crystal purity, morphology and polymorphism for impurities in the feed, and hence determine the need for feed purification.

Similarly to the conventional design procedure, the crystallisation method is mainly selected on the basis of the thermodynamics of the solid/liquid equilibrium (see Fig. 1). For many systems, e.g. those involving electrolytes, these equilibria still need to be determined experimentally. Other factors influencing this selection are the scaling tendencies of components present in the solution and the production capacity.

Design specifications Objectives and Constraints		Design Variables	Domain knowledge
<i>Product:</i>FilterabilityNo caking in storage	 SHE considerations Pneumatic handling Freedom from dust 	Discrete: • Polymorphism	• Filterability tests and models: permeability and compressibility
 No flotation Suspendability Washability Dryability Dissolution rate 	 Flowability Mechanical strength Abrasion resistance Bulk density or porosity Aesthetic appearance 	 <i>Continuous:</i> Morphology Crystal size distribution Purity Maximum inclusion content 	 Shear tests Indentation tests Caking tests Flowability tests Safety aspects

 Table 4: Design Level I - Design of the crystalline product.

Design specifications	Design and Operational Variables	Domain knowledge
Objectives and Constraints		
Process:	Discrete:	• Thermodynamic activity of
Production capacity	Crystallisation method	species/components in solid,
Feed composition	Feed purification	liquid and vapour phase
• Yield	Recrystallisation step	• Adsorption (energy) of
Energy consumption	• Solvent(s)	components/species on the
Availability	• Additive(s)	various crystal faces
SHE considerations	• Material of construction	Scaling or encrustation tendency of components/species
Product:	Continuous:	• Metastable zone with respect to
Polymorphism	Pressure range	homogeneous and heterogeneous
Morphology	Temperature range	primary nucleation
Crystal size distribution	• Concentration solvent(s)	Safety aspects
Purity	• Concentration additive(s)	
Maximum inclusion content		

Table 6: Design Level III - Flowsheet design of the crystallisation process.

Design specifications	Design and Operational Variables	Domain knowledge		
Objectives and Constraints	Design and Operational Variables	Domain Knowledge		
 Process: Production capacity Feed composition Crystallisation method Yield Pressure range Temperature range Supersaturation range Energy consumption Availability, controllability and Resiliency SHE considerations <i>Product:</i> Crystal size distribution 	 Discrete: Operation mode Number of stages Feed configuration Recycle structure Location purge stream(s) Continuous: Residence time in each stage or batch time Recycle flow rates Purge flow rate Pressure and/or temperature in each stage Heating/cooling duty or trajectory Heat exchange rates 	 Thermodynamic activity of species/components in solid, liquid and vapour phase Crystallisation kinetics, i.e. rate expressions for the nucleation, growth, attrition, agglomeration and breakage of crystals Fouling kinetics Shape factors of the crystalline components Fire and explosion index 		

	Design specifications Objectives and Constraints	Design and Operational Variables		Domain knowledge
Pro	Production capacity Feed composition Crystallisation method Yield Operation mode Pressure and temperature range	 Discrete: Crystalliser type Fines classification and dissolution/clear liquor advance Product classification Heat exchanger type Circulation device 	•	Equipment characteristics Hydrodynamics Thermodynamic activity of species/components in solid, liquid and vapour phase Crystallisation kinetics,
• • • • •	Supersaturation range Residence time (distribution) Heat exchange rates No boiling in heat exchanger No entrainment of droplets by vapour Suspension criterion Availability, controllability and resiliency SHE considerations	 Continuous: Equipment dimensions Feed location Product removal location Solids concentration Circulation flow rate Operating conditions of classification devices Flow rate through heat 	•	i.e. rate expressions for the nucleation, growth, attrition, agglomeration and breakage of crystals Fouling kinetics Shape factors of the crystalline components
•	Crystal size distribution	exchanger		

Table 7: Design Level IV - Design of a crystallisation stage.

For instance, the availability of a direct cooling crystallisation process can be reduced significantly by scaling on the cooling surface. Depending on the production capacity this loss in availability may or may not be a problem. Another example, a crystallisation method requiring a vacuum system is very inconvenient for low capacity processes. Operating conditions such as pressure and temperature are chosen such to obtain the highest possible yield, while obeying SHE constraints. Purity considerations determine the necessity for recrystallisation steps and the maximum crystal growth rate. With increasing growth rates the tendency of components to co-crystallise and entrapment of mother liquor increase. This introduces an upper limit for the maximum supersaturation. The allowable supersaturation also depends on the metastable zone. When the concentration exceeds this zone primary nucleation occurs, which is usually unwanted as it decreases the average crystal size.

Flowsheet design of the crystallisation process

The realisations of the design and operational variables from design level II are propagated to design level III, the flowsheet design of the crystallisation process. Together with relevant initial design specifications from level 0, they constitute the objectives and constraints for level III. An overview of the design specifications, design and operational variables and domain knowledge for this level is given in Table 6. Note that at this level all product related specifications except the CSD have disappeared. If relevant, they are now present as operating windows for pressure, temperature and supersaturation.

Criteria to select batch or semi-batch operation include a low production capacity, a short time-tomarket, a short product lifetime, high value products and a narrow CSD. A narrow CSD is also a reason to opt for multiple stage over single stage operation. The feed configuration of a multiple stage process is governed by the feed temperature and the required CSD. The heat exchange rates are determined by the production capacity.

To analyse the consequences of the abovementioned choices on the CSD a predictive process model consisting of thermodynamics, kinetics, mass, energy and population balances is required. Such a model should also be applicable to kinetic parameter estimation. As mentioned before kinetic models cannot vet be derived from first principles only. Experimentation and parameter estimation are hence an intrinsic step in crystallisation design. In the second part of this paper we will present a general crystallisation process model for analysis and optimisation with respect to the CSD as well as domain knowledge acquisition concerning the kinetics.

Another analysis, especially important for multiple stage evaporative crystallisation processes, involves the energy consumption and heat exchange surface area for the required heat exchange rates. Of an evaporative process consisting of N stages, the steam consumption and the total heat exchange surface area are proportional to respectively 1/N and N. Large deviations of the relationship for the surface area occur when a system exhibits significant boiling point elevations (BPE). When components with a high fouling tendency are present, time variant heat transfer coefficients need to be taken into account when sizing heat exchangers. For this purpose, another predictive model has been developed (Bermingham et al., 1999).

The resulting flowsheet design alternatives are propagated to level IV. If the suggested flowsheets contain multiple crystallisation stages, the relevant information for each stage is propagated to different instances of design level IV.

Design of a crystalliser stage

Practically all the design specifications from the previous level are present at this design level. The set is extended with equipment related specifications and with design and operational variables propagated from the previous level (see Table 7).

The first selection at this level involves the crystalliser type, e.g. fluidised bed crystalliser, DTB crystalliser. forced crystalliser, DT circulation crystalliser or simply a mixed tank. The order in which these crystallisers are mentioned here, usually coincides with decreasing crystal size for a certain crystallisation system. To increase the average crystal size and/or stabilise the crystalliser with respect to the CSD, certain crystallisers can be equipped with classification devices. The type of circulation device, impeller or pump, is determined by the crystalliser type and the mechanical properties of the crystals. For brittle materials a device with a high pumping number must be selected to prevent excessive attrition.

At this level, hydrodynamics is added to the fields of domain knowledge. Applications of this knowledge include the sizing and operation of classification devices, determination of minimum circulation rates for adequate particle suspension and optimisation of the product removal location. The use of computational fluid dynamics (CFD) packages to obtain hydrodynamic information is on the increase for crystallisation processes. However, it is not yet possible to combine CFD techniques with population balance modelling. To analyse the influence of kinetic-hydrodynamic interactions on the product CSD, the general crystallisation model mentioned at the previous design level can be used in a compartmental manner to account for spatially distributed process conditions.

The final design alternative can be propagated to the detailed engineering phase. This design level does not lie within the scope of this paper.

A predictive crystallisation model

A predictive crystallisation model with respect to the CSD has been developed for domain knowledge acquisition (*parameter estimation*) as well as analysis and optimisation purposes (*predictive modelling*) at design levels III and IV. The model consists of material balances, an energy balance, a population balance and a relation for the temperature dependent equilibrium solute concentration. The main features of this model are:

- Kinetic model from Gahn et al. (1997). Nucleation as a function of crystalliser/impeller geometry, impeller frequency and the CSD. Crystal growth rates determined by liquid phase composition and internal energy of crystals
- A fines dissolution model based on the kinetic parameters from the Gahn model.
- A conventional ideal fines dissolver for comparison reasons.
- A compartmental nature to describe the spatial distribution of the CSD, energy dissipation, supersaturation, etc.

Fig. 6 shows the compartment structures used for the simulations throughout this paper: (a) ideally mixed DT, (b) ideally mixed DTB with ideal fines dissolution, (c) ideally mixed DTB with real fines dissolution, (d) and (e) DTB with real fines dissolution and compartmentalised main body.



Figure 6: Compartment structures employed for simulations with the compartmental model.

The subdivision of a crystalliser main body into multiple compartments, as done in Fig. 6(d) and 6(e), is performed on the basis of hydrodynamic analysis and characteristic times of the crystallisation phenomena (Kramer et al. (1999)).

The compartmental model is applied in the following sections for parameter estimation and for analysis purposes related to design levels III and IV.

Modelling of single crystallisers.

Estimation of kinetic parameters

Before the crystallisation model can be applied to a specific crystallisation system, the parameters of the Gahn kinetic model need to be estimated. For ammonium sulphate this was done by Neumann et al. (1998b) using experiment DT22 (see Table 1).



Figure 7: measured and fitted trend of the median crystal size for experiment DT22.

The quality of fit (Fig. 7) is comparable to the one obtained with the Ó Meadhra kinetic model (Fig. 3), but the number of parameters involved is now only two as opposed to twelve.

Evaluation of the model's predictive value

Using the same kinetic parameters, but a different compartment structure to account for differences in geometry and scale, experiment DTB1100 was simulated (sim. 6). The expected steady-state median crystal sizes only differ approximately 100 μ m and the periods of the oscillations are in much better agreement than when using the Ó Meadhra model (Fig. 4).



Figure 8: measured and predicted trend of the median crystal size for experiment DTB1100.

No difference is seen between simulations 4, 5 and 6. By using three different compartment structures, these simulations show that incomplete fines dissolution

and/or a spatial distribution of process conditions are not significant in the case of experiment DTB1100.

Influence of crystalliser scale on the crystal size distribution.

Because the volume specific surface area of both the crystalliser and the impeller decrease with increasing scale, the nucleation rate per unit volume should decrease with size, thus leading to larger crystals.

Table 8: Scale dependent circulation quantities

Simulation nr.	7	8	9
Crystalliser volume [m ³]	0.022	1.1	360
Impeller frequency [s ⁻¹]	19.52	5.38	0.756
Spec. power input [W·kg ⁻¹]	7.27	1.52	0.21
Axial velocity [m·s ⁻¹]	1	1	1
Internal circulation $[m^3 \cdot s^{-1}]$	0.018	0.20	9.04
Turnover time [s]	1.2	5.6	40

The capability of the model framework to predict this trend is shown in Fig. 9 for different scale crystallisers operating at identical conditions, i.e. same residence time, specific heat input and axial velocity. As for all simulations in this paper, more details can be found in the appendix.



Figure 9: Median crystal size trends for DT crystallisers of 0.022, 1.1 and 360 m³.

Influence of fines removal design and operation on the crystal size distribution

The model framework will now be applied to investigate sizing issues related to the fines removal system a fictive 360 m^3 DTB crystalliser. First three fines removal characteristics are given:

- Fines residence time in the main body. This is the ratio of the main body volume and fines withdrawal rate.
- Vertical velocity in the annular zone. This is where the classification occurs
- Residence time in the dilution loop. This is the ratio of the dissolution loop volume and fines withdrawal rate.

Simulation nr.	10	11	12	13
Fines withdrawal	0.655	2.62	2.62	2.62
rate $[m^3 \cdot s^{-1}]$				
Velocity in annular	0.263	0.263	2.63	2.63
zone $[cm \cdot s^{-1}]$				
Temperature	18.9	4.72	4.72	4.72
increase fines [K]				
Fines res. time in	100	100	100	10
dilution loop [s]				
Diss. rate $[kg \cdot s^{-1}]$	0.130	0.188	2.32	1.27
Diss. rate $[\# \cdot s^{-1}]$	2.2E9	5.0E9	11E9	3.7E9
Mass-based diss.	0.97	0.84	0.38	0.06
efficiency [kg·kg ⁻¹]				
Number-based diss.	0.97	0.89	0.79	0.30
efficiency [#·# ⁻¹]				

Table 9: Different configurations of the fines removal system of a $360 \text{ m}^3 \text{ DTB}$ crystalliser.

The first configuration (sim. 10) is a scale-up of experiment DTB1100, i.e. same fines removal characteristics. As the temperature increase of the fines flow is rather high (18.9 K) in this set-up, simulation 11 was performed for a lower (factor 4) residence time in the main body, while keeping other fines removal characteristics constant. This new configuration has a very large annular zone: a cross sectional area of approximately 10^3 m^2 . Therefore simulation 12 was performed with a higher velocity (factor 10) in the annular zone. The difference in classification behaviour is depicted in Fig. 10. For simulation 14, the residence time in the dilution loop was reduced tenfold to yield an industrial value.



Figure 10: calculated fines classification curves for velocities of 0.263 and 2.63 cm s⁻¹.

The effect of the different fines removal configurations on the expected median crystal size can be seen in Fig. 11 and 12. Simulation 9 was included in Fig. 11 for comparison with a 360 m³ DT crystalliser.



Figure 11: Influence of fines dissolution and withdrawal rate on the median crystal size.



Figure 12: Influence of the dissolution loop volume on the median crystal size.

The simulated effect of different fines removal configurations can be understood by investigating the mass-based and number-based dissolution rates given in Table 9. Because crystal dissolution is a size dependent phenomenon, these rates are not interchangeable. Clearly, The number based and not the mass based dissolution rate is of importance for the median size.

Influence of spatially distributed process conditions on the crystal size distribution

For the same fictive 360 m³ DTB crystalliser another simulation was performed to study whether the spatial distribution of process variables does play a role for this crystalliser. Recall that no effect was observed for experiment DTB1100. However, а 360 m^3 crystalliser has significantly larger turnover times (see Table 8) and the characteristic times for the crystallisation phenomena are scale independent. Comparison of simulations 13 and 14 (main body modelled respectively with one and ten compartments) shows a large effect (see Fig. 13 and Table 10).



Figure 13: Influence of spatially distributed process conditions on the median crystal size

Table 10: Spatial distribution of the crystal mass production and supersaturation.

	crys. mass production		Macroscopic
	absolute [kg·s ⁻¹]	specific [kg⋅m ⁻³ ⋅s ⁻¹]	supersaturation [kg·m ⁻³]
sim. 13			
MB-C1	15.64	0.0434	1.38
FD-C2	-1.252	-0.0478	-4.03
sim. 14			
MB-C1	0.015	0.0008	0.23
MB-C2	0.029	0.0007	0.24
MB-C3	0.030	0.0007	0.23
MB-C4	0.030	0.0007	0.23
MB-C5	0.030	0.0007	0.23
MB-C6	2.013	0.1118	2.66
MB-C7	3.905	0.0964	2.41
MB-C8	3.379	0.0834	2.18
MB-C9	2.935	0.0725	2.00
MB-C10	3.235	0.0799	1.78
FD-C11	-1.342	-0.0512	-3.60

Again, the effect can be best understood by a study of the number based and not the mass based dissolution rates. (see Table 11).

Table 11: Influence of spatial distribution on fines removal.

Simulation nr.	13	14
Dissolution rate $[kg \cdot s^{-1}]$	1.27	1.35
Dissolution. rate $[#\cdot s^{-1}]$	3.7E9	0.9E9
Mass-based diss. efficiency [kg·kg ⁻¹]	0.063	0.046
Number-based diss. efficiency [#·# ⁻¹]	0.30	0.14

Modelling multiple stage crystallisation systems

The influence of the number of stages on the crystal size distribution is investigated by comparing a single and triple stage configuration, both with a total volume of 360 m^3 . The triple stage configuration consists of identical 120 m^3 DT crystallisers as opposed to one

360 m³ DT crystalliser. Operating conditions were identical for both configurations, i.e. same residence time, specific heat input and axial velocity.



Figure 14: Simulated CSD's resulting from a single and triple stage process.

Fig. 14 shows a minor advantage of multiple stage operation for the CSD in case of ammonium sulphate.

Conclusions

Assessment of conventional design procedures reveals the value of heuristics for the rapid development of a base case design. Shortcoming are the lack of reliable analysis and optimisation tools and of a structuring of heuristics to provide design alternatives.

To systematically cope with the large number of initial design specifications, design and operational variables and the wide variety of domain knowledge involved, a hierarchical design procedure has been developed. Structuring of the relevant specifications, variables and knowledge at each design level simplifies the design problem and provides valuable insights for designers. It also highlights the major shortcomings in design knowledge, i.e. product performance/composition relations (level I) and predictive models for the concise analysis of flowsheets (level III) and comprehensive analysis of single crystallisers (level IV) with respect to the CSD and supersaturation. The hierarchical decomposition does not imply a oncethrough process with respect to the design levels.

To support the analysis step at levels III and IV, a crystallisation model has been developed. This model allows estimation of key kinetic parameters and prediction of the effect of crystalliser scale and type and of operating conditions on the local CSD and supersaturation without adjustment of kinetic parameters. However, the model's predictive capabilities need to be validated more decisively.

The ultimate aim of this design procedure, i.e. better designs in less time, cannot be guaranteed until the procedure as a whole is validated by means of industrial case studies. Main achievements so far are Finally, although the design procedure has been presented in the light of grassroots design, it can also be largely applied to retrofit and optimisation of process operation. One should think of synthesis at levels I and II and analysis tools at all levels.

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Appendix: Crystalliser specifications and simulation features

Crystalliser name	DT-1	DT-2	DTB-2	DT-3	DT-4	DTB-4
Volume main body [m ³]	0.022	1.1	1.1	120	360	360
Crystalliser diameter [m]	0.23	0.7	0.7	3.32	4.75	4.75
Draft tube diameter [m]	0.15	0.5	0.5	2.37	3.39	3.39
Impeller [‡] diameter [m]	0.14	0.485	0.485	2.32	3.34	3.34
Edge of impeller [m]	0.002	0.006	0.006	0.029	0.043	0.043
Breadth of impeller [m]	0.047	0.18	0.18	0.86	1.24	1.24
Power number [-] / Pumping number [-]	0.4 / 0.33	0.4 / 0.32	0.4 / 0.32	0.4 / 0.32	0.4 / 0.32	0.4 / 0.32
Volume dilution loop [m ³]	-	-	0.2	-	-	varied
Cross sectional area annular zone [m ²]	-	-	0.761	-	-	varied

Table A.I: Crystalliser specifications

^{*}All impellers are equipped with three blades; Angle of the blades is 0.436 rad.

Table A.II:	General	simulation	features

Crystalliser temperature [K]	323.15			
Feed temperature [K]	323.15			
Feed composition	saturated and crystal free			
Heat input per unit volume [W·m ⁻³]	$120 \cdot 10^3$			
Product residence time [s]	4500			
Solids concentration [vol. %]	10.2			
Initial crystal size distribution	Normal distribution (L ₀ = 300 μ m, σ = 70 μ m, 40 kg·m ³)			
Discretisation crystal size domain	Logarithmic with a lower bound of 10 µm			
Estimated parameters for Gahn kinetic model	$k_r = 10^{-5} \text{ m}^4 \cdot \text{mole}^{-1} \cdot \text{s}^{-1}; \qquad \Gamma_k = 1.9 \cdot 10^{-4} \text{ kg} \cdot \text{m}^3 \cdot \text{mole}^{-1} \cdot \text{s}^{-2}$			

Simulation nr.	1	2	3	4	5	6	7
Crystalliser name (see Table A.I)	DT-1	DTB-2	DT-1	DTB-2	DTB-2	DTB-2	DT-1
Impeller frequency [s ⁻¹]	16.67	5.33	16.67	5.33	5.33	5.33	19.52
Mean spec. power input [W·kg ⁻¹]	4.53	1.48	4.53	1.48	1.48	1.48	7.27
Axial velocity [m·s ⁻¹]	0.85	0.99	0.85	0.99	0.99	0.99	1
Internal circulation rate $[m^3 \cdot s^{-1}]$	0.015	0.19	0.015	0.19	0.19	0.19	0.018
Turnover time [s]	1.5	5.6	1.5	5.6	5.6	5.6	1.2
Product removal rate $[dm^3 \cdot s^{-1}]$	0.0049	0.244	0.0049	0.244	0.244	0.244	0.0049
Fines res. time in main body [s]	-	550	-	550	550	550	-
Fines withdrawal rate $[m^3 \cdot s^{-1}]$	-	0.002	-	0.002	0.002	0.002	-
Cross sect. area annular zone [m ²]	-	0.761	-	0.761	0.761	0.761	-
Velocity in annular zone [cm·s ⁻¹]	-	0.263	-	0.263	0.263	0.263	-
Temperature increase fines flow [K]	-	18.9	-	18.9	18.9	18.9	-
Fines res. time in dilution loop [s]	-	-	-	-	100	100	-
Volume dissolution loop [m ³]	-	-	-	-	0.2	0.2	-
Compartmental model (see Fig. 6)	а	b	а	b	c	d	a
L ₂₅ in final point of simulation	372	690	379	516	515	513	308
L ₅₀ in final point of simulation	543	982	515	726	724	721	438
L ₇₅ in final point of simulation	744	1344	644	949	949	945	543
CPU [hrs]	03	0.5	0.7	09	1.3	63	07
	0.2	0.0	0.7	0.2		0.5	0.7
Simulation nr.	8	9	10	11	12	13	14
Simulation nr. Crystalliser name (see Table A.I)	8 DT-2	9 DT-4	10 DTB-4	11 DTB-4	12 DTB-4	13 DTB-4	14 DTB-4
Simulation nr. Crystalliser name (see Table A.I) Impeller frequency [s ⁻¹]	8 DT-2 5.38	9 DT-4 0.756	10 DTB-4 0.756	11 DTB-4 0.756	12 DTB-4 0.756	13 DTB-4 0.756	14 DTB-4 0.756
Simulation nr. Crystalliser name (see Table A.I) Impeller frequency [s ⁻¹] Mean spec. power input [W·kg ⁻¹]	8 DT-2 5.38 1.52	9 DT-4 0.756 0.21	10 DTB-4 0.756 0.21	11 DTB-4 0.756 0.21	12 DTB-4 0.756 0.21	13 DTB-4 0.756 0.21	14 DTB-4 0.756 0.21
Simulation nr. Crystalliser name (see Table A.I) Impeller frequency [s ⁻¹] Mean spec. power input [W·kg ⁻¹] Axial velocity [m·s ⁻¹]	8 DT-2 5.38 1.52 1	9 DT-4 0.756 0.21 1	10 DTB-4 0.756 0.21 1	11 DTB-4 0.756 0.21 1	12 DTB-4 0.756 0.21 1	13 DTB-4 0.756 0.21 1	14 DTB-4 0.756 0.21 1
Simulation nr.Crystalliser name (see Table A.I)Impeller frequency $[s^{-1}]$ Mean spec. power input $[W \cdot kg^{-1}]$ Axial velocity $[m \cdot s^{-1}]$ Internal circulation rate $[m^3 \cdot s^{-1}]$	8 DT-2 5.38 1.52 1 0.20 1 0.20 1 <th1< th=""> <th1< th=""> <th1< th=""> <</th1<></th1<></th1<>	9 DT-4 0.756 0.21 1 9.04	10 DTB-4 0.756 0.21 1 9.04	11 DTB-4 0.756 0.21 1 9.04	12 DTB-4 0.756 0.21 1 9.04	13 DTB-4 0.756 0.21 1 9.04	14 DTB-4 0.756 0.21 1 9.04
Simulation nr. Crystalliser name (see Table A.I) Impeller frequency [s ⁻¹] Mean spec. power input [W·kg ⁻¹] Axial velocity [m·s ⁻¹] Internal circulation rate [m ³ ·s ⁻¹] Turnover time [s]	8 DT-2 5.38 1.52 1 0.20 5.6 1 <th1< th=""> <th1< th=""> <th1< th=""> <!--</td--><td>9 DT-4 0.756 0.21 1 9.04 40</td><td>10 DTB-4 0.756 0.21 1 9.04 40</td><td>11 DTB-4 0.756 0.21 1 9.04 40</td><td>12 DTB-4 0.756 0.21 1 9.04 40</td><td>13 DTB-4 0.756 0.21 1 9.04 40</td><td>14 DTB-4 0.756 0.21 1 9.04 40</td></th1<></th1<></th1<>	9 DT-4 0.756 0.21 1 9.04 40	10 DTB-4 0.756 0.21 1 9.04 40	11 DTB-4 0.756 0.21 1 9.04 40	12 DTB-4 0.756 0.21 1 9.04 40	13 DTB-4 0.756 0.21 1 9.04 40	14 DTB-4 0.756 0.21 1 9.04 40
Simulation nr.Crystalliser name (see Table A.I)Impeller frequency $[s^{-1}]$ Mean spec. power input $[W \cdot kg^{-1}]$ Axial velocity $[m \cdot s^{-1}]$ Internal circulation rate $[m^3 \cdot s^{-1}]$ Turnover time $[s]$ Product removal rate $[m^3 \cdot s^{-1}]$	8 DT-2 5.38 1.52 1 0.20 5.6 0.244	9 DT-4 0.756 0.21 1 9.04 40 80	10 DTB-4 0.756 0.21 1 9.04 40 80	11 DTB-4 0.756 0.21 1 9.04 40 80	12 DTB-4 0.756 0.21 1 9.04 40 80	13 DTB-4 0.756 0.21 1 9.04 40 80	14 DTB-4 0.756 0.21 1 9.04 40 80
Simulation nr.Crystalliser name (see Table A.I)Impeller frequency $[s^{-1}]$ Mean spec. power input $[W \cdot kg^{-1}]$ Axial velocity $[m \cdot s^{-1}]$ Internal circulation rate $[m^3 \cdot s^{-1}]$ Turnover time $[s]$ Product removal rate $[m^3 \cdot s^{-1}]$ Fines res. time in main body $[s]$	8 DT-2 5.38 1.52 1 0.20 5.6 0.244 -	9 DT-4 0.756 0.21 1 9.04 40 80 -	10 DTB-4 0.756 0.21 1 9.04 40 80 550	11 DTB-4 0.756 0.21 1 9.04 40 80 137.5	12 DTB-4 0.756 0.21 1 9.04 40 80 137.5	13 DTB-4 0.756 0.21 1 9.04 40 80 137.5	14 DTB-4 0.756 0.21 1 9.04 40 80 137.5
Simulation nr.Crystalliser name (see Table A.I)Impeller frequency $[s^{-1}]$ Mean spec. power input $[W \cdot kg^{-1}]$ Axial velocity $[m \cdot s^{-1}]$ Internal circulation rate $[m^3 \cdot s^{-1}]$ Turnover time $[s]$ Product removal rate $[m^3 \cdot s^{-1}]$ Fines res. time in main body $[s]$ Fines withdrawal rate $[m^3 \cdot s^{-1}]$	8 DT-2 5.38 1.52 1 0.20 5.6 0.244 - -	9 DT-4 0.756 0.21 1 9.04 40 80 - -	10 DTB-4 0.756 0.21 1 9.04 40 80 550 0.655	11 DTB-4 0.756 0.21 1 9.04 40 80 137.5 2.62	12 DTB-4 0.756 0.21 1 9.04 40 80 137.5 2.62	13 DTB-4 0.756 0.21 1 9.04 40 80 137.5 2.62	14 DTB-4 0.756 0.21 1 9.04 40 80 137.5 2.62
Simulation nr.Crystalliser name (see Table A.I)Impeller frequency $[s^{-1}]$ Mean spec. power input $[W \cdot kg^{-1}]$ Axial velocity $[m \cdot s^{-1}]$ Internal circulation rate $[m^3 \cdot s^{-1}]$ Turnover time $[s]$ Product removal rate $[m^3 \cdot s^{-1}]$ Fines res. time in main body $[s]$ Fines withdrawal rate $[m^3 \cdot s^{-1}]$ Cross sect. area annular zone $[m^2]$	8 DT-2 5.38 1.52 1 0.20 5.6 0.244 -	9 DT-4 0.756 0.21 1 9.04 40 80 - - - -	10 DTB-4 0.756 0.21 1 9.04 40 80 550 0.655 249	11 DTB-4 0.756 0.21 1 9.04 40 80 137.5 2.62 996	12 DTB-4 0.756 0.21 1 9.04 40 80 137.5 2.62 99.6	13 DTB-4 0.756 0.21 1 9.04 40 80 137.5 2.62 99.6	14 DTB-4 0.756 0.21 1 9.04 40 80 137.5 2.62 99.6
Simulation nr.Crystalliser name (see Table A.I)Impeller frequency $[s^{-1}]$ Mean spec. power input $[W \cdot kg^{-1}]$ Axial velocity $[m \cdot s^{-1}]$ Internal circulation rate $[m^3 \cdot s^{-1}]$ Turnover time $[s]$ Product removal rate $[m^3 \cdot s^{-1}]$ Fines res. time in main body $[s]$ Fines withdrawal rate $[m^3 \cdot s^{-1}]$ Cross sect. area annular zone $[m^2]$ Velocity in annular zone $[cm \cdot s^{-1}]$	8 DT-2 5.38 1.52 1 0.20 5.6 0.244 - - - - - -	9 DT-4 0.756 0.21 1 9.04 40 80 - - - - - - - - -	10 DTB-4 0.756 0.21 1 9.04 40 80 550 0.655 249 0.263	11 DTB-4 0.756 0.21 1 9.04 40 80 137.5 2.62 996 0.263	12 DTB-4 0.756 0.21 1 9.04 40 80 137.5 2.62 99.6 2.63	13 DTB-4 0.756 0.21 1 9.04 40 80 137.5 2.62 99.6 2.63	14 DTB-4 0.756 0.21 1 9.04 40 80 137.5 2.62 99.6 2.63
Simulation nr.Crystalliser name (see Table A.I)Impeller frequency $[s^{-1}]$ Mean spec. power input $[W \cdot kg^{-1}]$ Axial velocity $[m \cdot s^{-1}]$ Internal circulation rate $[m^3 \cdot s^{-1}]$ Turnover time $[s]$ Product removal rate $[m^3 \cdot s^{-1}]$ Fines res. time in main body $[s]$ Fines withdrawal rate $[m^3 \cdot s^{-1}]$ Cross sect. area annular zone $[m^2]$ Velocity in annular zone $[cm \cdot s^{-1}]$ Temperature increase fines flow $[K]$	8 DT-2 5.38 1.52 1 0.20 5.6 0.244 - - - - - - - - - - -	9 DT-4 0.756 0.21 1 9.04 40 80 - - - - - - - - - - - -	10 DTB-4 0.756 0.21 1 9.04 40 80 550 0.655 249 0.263 18.9	11 DTB-4 0.756 0.21 1 9.04 40 80 137.5 2.62 996 0.263 4.72	12 DTB-4 0.756 0.21 1 9.04 40 80 137.5 2.62 99.6 2.63 4.72	13 DTB-4 0.756 0.21 1 9.04 40 80 137.5 2.62 99.6 2.63 4.72	14 DTB-4 0.756 0.21 1 9.04 40 80 137.5 2.62 99.6 2.63 4.72
Simulation nr.Crystalliser name (see Table A.I)Impeller frequency $[s^{-1}]$ Mean spec. power input $[W \cdot kg^{-1}]$ Axial velocity $[m \cdot s^{-1}]$ Internal circulation rate $[m^3 \cdot s^{-1}]$ Turnover time $[s]$ Product removal rate $[m^3 \cdot s^{-1}]$ Fines res. time in main body $[s]$ Fines withdrawal rate $[m^3 \cdot s^{-1}]$ Cross sect. area annular zone $[m^2]$ Velocity in annular zone $[cm \cdot s^{-1}]$ Temperature increase fines flow $[K]$ Fines res. time in dilution loop $[s]$	8 DT-2 5.38 1.52 1 0.20 5.6 0.244 - - - - - - - - - - - - - - - -	9 DT-4 0.756 0.21 1 9.04 40 80 - - - - - - - - - - - - - - - - -	10 DTB-4 0.756 0.21 1 9.04 40 80 550 0.655 249 0.263 18.9 100	11 DTB-4 0.756 0.21 1 9.04 40 80 137.5 2.62 996 0.263 4.72 100	12 DTB-4 0.756 0.21 1 9.04 40 80 137.5 2.62 99.6 2.63 4.72 100	13 DTB-4 0.756 0.21 1 9.04 40 80 137.5 2.62 99.6 2.63 4.72 10	14 DTB-4 0.756 0.21 1 9.04 40 80 137.5 2.62 99.6 2.63 4.72 10
Simulation nr.Crystalliser name (see Table A.I)Impeller frequency $[s^{-1}]$ Mean spec. power input $[W \cdot kg^{-1}]$ Axial velocity $[m \cdot s^{-1}]$ Internal circulation rate $[m^3 \cdot s^{-1}]$ Turnover time $[s]$ Product removal rate $[m^3 \cdot s^{-1}]$ Fines res. time in main body $[s]$ Fines withdrawal rate $[m^3 \cdot s^{-1}]$ Cross sect. area annular zone $[m^2]$ Velocity in annular zone $[cm \cdot s^{-1}]$ Temperature increase fines flow $[K]$ Fines res. time in dilution loop $[s]$ Volume dissolution loop $[m^3]$	8 DT-2 5.38 1.52 1 0.20 5.6 0.244 -	9 DT-4 0.756 0.21 1 9.04 40 80 - - - - - - - - - - - - - - - - - - -	10 DTB-4 0.756 0.21 1 9.04 40 80 550 0.655 249 0.263 18.9 100 65.5	11 DTB-4 0.756 0.21 1 9.04 40 80 137.5 2.62 996 0.263 4.72 100 262	12 DTB-4 0.756 0.21 1 9.04 40 80 137.5 2.62 99.6 2.63 4.72 100 262	13 DTB-4 0.756 0.21 1 9.04 40 80 137.5 2.62 99.6 2.63 4.72 10 26.2	14 DTB-4 0.756 0.21 1 9.04 40 80 137.5 2.62 99.6 2.63 4.72 10 26.2
Simulation nr.Crystalliser name (see Table A.I)Impeller frequency $[s^{-1}]$ Mean spec. power input $[W \cdot kg^{-1}]$ Axial velocity $[m \cdot s^{-1}]$ Internal circulation rate $[m^3 \cdot s^{-1}]$ Turnover time $[s]$ Product removal rate $[m^3 \cdot s^{-1}]$ Fines res. time in main body $[s]$ Fines withdrawal rate $[m^3 \cdot s^{-1}]$ Cross sect. area annular zone $[m^2]$ Velocity in annular zone $[cm \cdot s^{-1}]$ Temperature increase fines flow $[K]$ Fines res. time in dilution loop $[s]$ Volume dissolution loop $[m^3]$ Compartmental model (see Fig. 6)	8 DT-2 5.38 1.52 1 0.20 5.6 0.244 - - - - - - - - - - - - - - - - - a	9 DT-4 0.756 0.21 1 9.04 40 80 - - - - - - - - - - - - - a	10 DTB-4 0.756 0.21 1 9.04 40 80 550 0.655 249 0.263 18.9 100 65.5 c	11 DTB-4 0.756 0.21 1 9.04 40 80 137.5 2.62 996 0.263 4.72 100 262 c	12 DTB-4 0.756 0.21 1 9.04 40 80 137.5 2.62 99.6 2.63 4.72 100 262 c	13 DTB-4 0.756 0.21 1 9.04 40 80 137.5 2.62 99.6 2.63 4.72 10 26.2 c	14 DTB-4 0.756 0.21 1 9.04 40 80 137.5 2.62 99.6 2.63 4.72 10 26.2 e
Simulation nr.Crystalliser name (see Table A.I)Impeller frequency $[s^{-1}]$ Mean spec. power input $[W \cdot kg^{-1}]$ Axial velocity $[m \cdot s^{-1}]$ Internal circulation rate $[m^3 \cdot s^{-1}]$ Turnover time $[s]$ Product removal rate $[m^3 \cdot s^{-1}]$ Fines res. time in main body $[s]$ Fines withdrawal rate $[m^3 \cdot s^{-1}]$ Cross sect. area annular zone $[m^2]$ Velocity in annular zone $[cm \cdot s^{-1}]$ Temperature increase fines flow $[K]$ Fines res. time in dilution loop $[s]$ Volume dissolution loop $[m^3]$ Compartmental model (see Fig. 6)L ₂₅ in final point of simulation	8 DT-2 5.38 1.52 1 0.20 5.6 0.244 -	9 DT-4 0.756 0.21 1 9.04 40 80 -	10 DTB-4 0.756 0.21 1 9.04 40 80 550 0.655 249 0.263 18.9 100 65.5 c 707	11 DTB-4 0.756 0.21 1 9.04 40 80 137.5 2.62 996 0.263 4.72 100 262 c 821	12 DTB-4 0.756 0.21 1 9.04 40 80 137.5 2.62 99.6 2.63 4.72 100 262 c 1433	13 DTB-4 0.756 0.21 1 9.04 40 80 137.5 2.62 99.6 2.63 4.72 10 26.2 c 822	14 DTB-4 0.756 0.21 1 9.04 40 80 137.5 2.62 99.6 2.63 4.72 10 26.2 e 664
Simulation nr.Crystalliser name (see Table A.I)Impeller frequency $[s^{-1}]$ Mean spec. power input $[W \cdot kg^{-1}]$ Axial velocity $[m \cdot s^{-1}]$ Internal circulation rate $[m^3 \cdot s^{-1}]$ Turnover time $[s]$ Product removal rate $[m^3 \cdot s^{-1}]$ Fines res. time in main body $[s]$ Fines withdrawal rate $[m^3 \cdot s^{-1}]$ Cross sect. area annular zone $[m^2]$ Velocity in annular zone $[cm \cdot s^{-1}]$ Temperature increase fines flow $[K]$ Fines res. time in dilution loop $[s]$ Volume dissolution loop $[m^3]$ Compartmental model (see Fig. 6) L_{25} in final point of simulation L_{50} in final point of simulation	8 DT-2 5.38 1.52 1 0.20 5.6 0.244 - - - - - - - - - - - - a 379 565	9 DT-4 0.756 0.21 1 9.04 40 80 - - - - - - - - - - - - - a 5553 809	10 DTB-4 0.756 0.21 1 9.04 40 80 550 0.655 249 0.263 18.9 100 65.5 c 707 1016	11 DTB-4 0.756 0.21 1 9.04 40 80 137.5 2.62 996 0.263 4.72 100 262 c 821 1170	12 DTB-4 0.756 0.21 1 9.04 40 80 137.5 2.62 99.6 2.63 4.72 100 262 c 1433 2044	13 DTB-4 0.756 0.21 1 9.04 40 80 137.5 2.62 99.6 2.63 4.72 10 26.2 c 822 1185	14 DTB-4 0.756 0.21 1 9.04 40 80 137.5 2.62 99.6 2.63 4.72 10 26.2 e 664 951
Simulation nr.Crystalliser name (see Table A.I)Impeller frequency $[s^{-1}]$ Mean spec. power input $[W \cdot kg^{-1}]$ Axial velocity $[m \cdot s^{-1}]$ Internal circulation rate $[m^3 \cdot s^{-1}]$ Turnover time $[s]$ Product removal rate $[m^3 \cdot s^{-1}]$ Fines res. time in main body $[s]$ Fines withdrawal rate $[m^3 \cdot s^{-1}]$ Cross sect. area annular zone $[m^2]$ Velocity in annular zone $[cm \cdot s^{-1}]$ Temperature increase fines flow $[K]$ Fines res. time in dilution loop $[s]$ Volume dissolution loop $[m^3]$ Compartmental model (see Fig. 6) L_{25} in final point of simulation L_{75} in final point of simulation	8 DT-2 5.38 1.52 1 0.20 5.6 0.244 -	9 DT-4 0.756 0.21 1 9.04 40 80 - - - - - - - - - - - - - - - - - - a 5553 809 1121	10 DTB-4 0.756 0.21 1 9.04 40 80 550 0.655 249 0.263 18.9 100 65.5 c 707 1016 1390	11 DTB-4 0.756 0.21 1 9.04 40 80 137.5 2.62 996 0.263 4.72 100 262 c 821 1170 1591	12 DTB-4 0.756 0.21 1 9.04 40 80 137.5 2.62 99.6 2.63 4.72 100 262 c 1433 2044 2760	13 DTB-4 0.756 0.21 1 9.04 40 80 137.5 2.62 99.6 2.63 4.72 10 26.2 c 822 1185 1623	14 DTB-4 0.756 0.21 1 9.04 40 80 137.5 2.62 99.6 2.63 4.72 10 26.2 e 664 951 1293

Table A.III: Features per simulation