

DEVELOPMENT AND VALIDATION OF MODELS FOR OPTIMISATION AND CONTROL OF BATCH AND CONTINUOUS CRYSTALLISATION PROCESSES

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A generic modelling framework is presented for solution crystallisation processes. The framework covers various supersaturation generation methods (cooling, flash-cooling and evaporation), operation modes (batch, semi-batch and continuous), crystalliser types (DTB, FC, stirred vessel), etc. Next, the framework is applied to several industrial case studies from the SINC-PRO project (IMS project focussing on crystalliser design and control), to illustrate both model configuration and validation (parameter estimation and experiment design).

1 INTRODUCTION

Although crystallisation is one of the older unit operations in the chemical industry, the design and operation of crystallisation processes still pose many problems, mainly related to product quality and process reliability. Decision support on the basis of rigorous process and product models is considered key to making optimal crystalliser design and operation less of an art and more of a science. However, model-based decision support is currently used to a much lesser extent in crystallisation than is the case for most vapour/liquid processes of interest to the chemical engineering profession. One of the main reasons for the relative lack in the use of model-based decision support, is not the effort required to develop a process model, but the effort required to validate that model and subsequently apply it for optimisation purposes.

Investigating the feasibility of using available commercial tools, with some additional development where required, for effective and efficient model-based optimal design and operation of crystallisation processes is the subject of this paper and one of the aims of the European part of the SINC-PRO project.

2 MODELLING FRAMEWORK

In order to enable the rapid development of consistent process and product models, a process and product modelling framework has been developed with a hierarchical structure as depicted in Figure 1 (Bermingham, 2003). This framework has been implemented in gPROMS (Process Systems Enterprise Ltd, 2003), a commercial modelling and optimisation tool. Applying the framework for the development of a process and product model for a new case involves the following steps:

1. Create a system properties model containing relationships and/or calls to a physical property package for thermophysical properties, such as VLE, SLE, densities, specific enthalpies, transport properties, such as viscosities and diffusion coefficients.
2. Select an existing kinetic model from the provided library or create a new one. This model provides expressions for all rate terms in the population balance equation.

3. Select an existing crystalliser model from the provided library or create a new one. This model contains:
 - The topology of the zone or compartment network used to account for spatial distributions in process conditions throughout a crystalliser including its external loops. The importance of zone network models is discussed by Kramer et al. (1999).
 - A number of selectors that allow the use of the same model for various operation modes (batch, semi-batch and continuous) and supersaturation generation methods (cooling, flash cooling and evaporation).

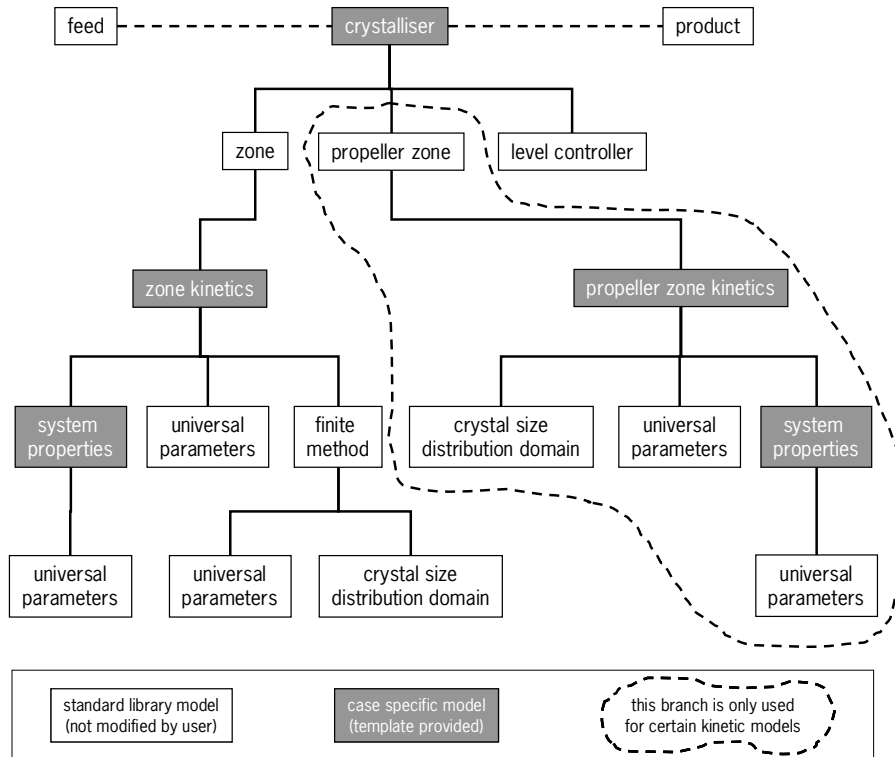


Figure 1: Hierarchical modelling framework for crystallisation processes.

Models developed using this framework are applicable for simulation, model validation and optimisation activities without requiring any further modifications.

3 CASE STUDIES AND MODEL DEVELOPMENT

In order to validate the usefulness of this modelling framework, a number of case studies have been brought forward by the SINC-PRO partners (see Table 1). The case studies have been selected such that the range of possible operation modes, supersaturation generation methods and crystalliser types are well covered. For each case study, the three steps outlined in the previous section were performed to create a crystalliser process model from the general modelling framework. The correlations and data used for the system properties model (step 1) were provided by the partner contributing the case study. The kinetic model (step 2), the zone network model (step 3) and the selector values for operation mode and supersaturation generation method (step 3) for each case study can be found in Table 1.

4 MODEL VALIDATION

This section deals with the validation of the crystalliser process models developed for two of the four SINC-PRO case studies. Model validation typically covers both parameter estimation and the design of experiments with a maximum information content. In this contribution we will only focus on

parameter estimation, however, research into the use of the same models presented here for experiment design purposes is also part of the still ongoing SINC-PRO project.

Table 1: Case studies for validation of the crystallisation process and product modelling framework.

Case	ammonium sulphate	lactitol monohydrate	potassium diphosphate	dextrose
Partner	Delft University of Technology	PURAC Biochem	Kemira Oy	Roquette Italy
Purpose of model development	prediction of scale-up effects	optimal recipe and process automation	scale-up and optimal recipe	aid design of new crystalliser
<i>Process characteristics</i>				
Operation mode	continuous	semi-batch	batch	batch/continuous
Supersaturation generation	evaporation	evaporation/cooling	evaporation	cooling
Crystalliser type	0.022 m ³ draft tube (DT) crystalliser 1.1 m ³ draft tube baffle (DTB) crystalliser	14 m ³ forced circulation (FC) crystalliser with propeller in main body	0.022 m ³ stirred vessel	0.010 m ³ stirred vessel (batch) 200 m ³ column crystalliser (continuous)
<i>Modelling characteristics</i>				
Crystallisation phenomena	Secondary nucleation and growth	Secondary nucleation and growth	Primary nucleation and growth	Secondary nucleation and growth
Kinetic model	Gahn and Mersmann (1999)	based on Eek et al. (1995)	power law model	power law model
Zone network model	1 zone for DT and 5 zones for DTB	2 zones	1 zone	1 zone for stirred vessel and 30 zones for column crystalliser
<i>Experimental data</i>				
PSD measurement device	Sympatec Helos (laser diffraction)	Sympatec Opus (acoustic attenuation)	LS Analyser (laser diffraction)	UR turbidimeter (turbidimetry)
Other measurements		brix, a measure for the supersaturation	yield	supersaturation

4.1 Continuous evaporative crystallisation of ammonium sulphate

Although the process is continuous and has a steady-state operating point for given process inputs, dynamic process data measured during start-up of the 0.022 m³ DT crystalliser is used for parameter estimation purposes. The reason for this is not that the developed crystalliser process models cannot be used for steady-state simulation and parameter estimation (they can), but that the information content of dynamic process data is typically much higher than that of steady-state process data.

4.2 Batch cooling/evaporative crystallisation of lactitol monohydrate

Measurements from three batch experiments were used to validate the model developed for PURAC's Lactitol monohydrate process. The parameter estimation was performed using the data from all three experiments simultaneously. Figure 4 and Figure 5 show the fits for the quantiles and the brix for one of these experiments. Both fits are considered adequate, although at certain points during the batch there is considerable mismatch between measurements and predictions. The lack of fit of the quantiles at the beginning of the batch are mainly attributed to the fact that acoustic attenuation does not provide reliable CSD measurements at low solids concentrations. As regards the brix, the model predicts the trend quite well but not the oscillations. These oscillations are a result of a fluctuating temperature in the real plant, which is currently not captured by the model.

5 CONCLUSIONS AND OUTLOOK

The effort required for the development and validation of crystallisation process and product models has been significantly reduced through the development of a generic crystallisation modelling framework and the use of commercially available tools that allow the same model to be used for simulation, parameter estimation, experiment design and optimisation. The next step in the SINC-PRO project involves applying the same models, either directly or after model reduction/linearisation, for soft sensing and model predictive control purposes. This activity is already underway for the Lactitol

monohydrate case at PURAC Biochem. Albeit outside the scope of the SINC-PRO project, some of the partners are already investigating the use of the same models for operator training purposes.

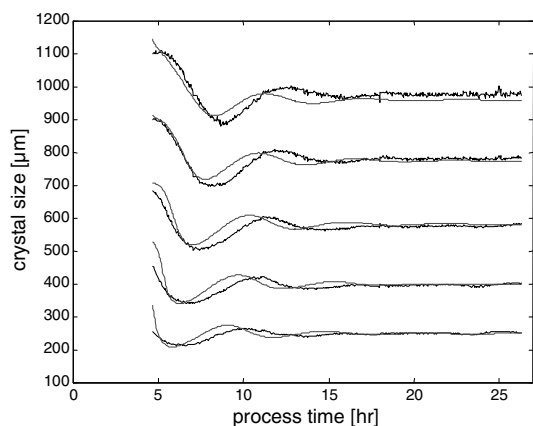


Figure 2: Ammonium sulphate case - Model predictions versus measurements for the 10, 25, 50, 75 and 90% quantiles of the crystal size distribution following start-up of the crystalliser.

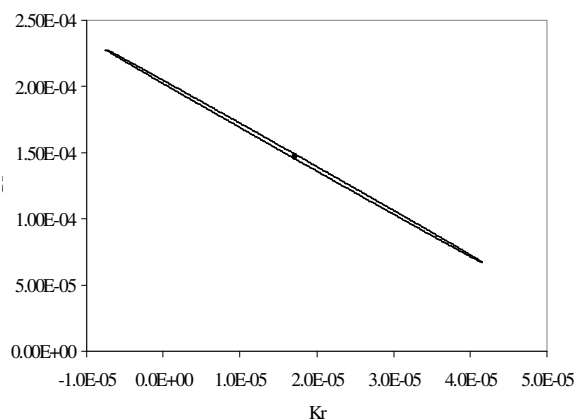


Figure 3: Ammonium sulphate case - Cross-correlation between the two kinetic parameters of the Gahn and Mersmann model (1999) that were estimated to obtain the fit shown in Figure 2.

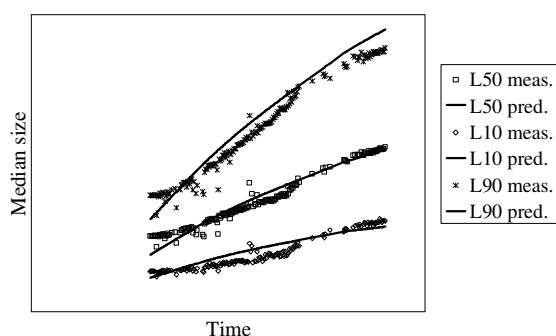


Figure 4: Lactitol case - Model predictions versus measurements for the 10, 50 and 90% quantiles of the crystal size distribution during one of the three batch experiments.

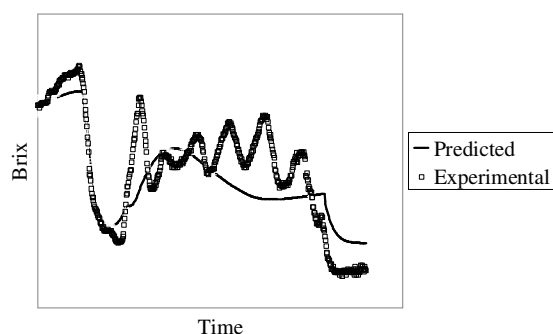


Figure 5: Lactitol case - Model predictions versus measurements for the brix (measure for sugar concentration in solution) during one of the three batch experiments.

6 REFERENCES

- Bermingham, S.K., 2003, A design procedure and predictive models for solution crystallisation processes: development and application, PhD Thesis, Delft University of Technology, Delft University Press, ISBN 90-407-2395-8.
- Eek, R.A, Dijkstra, S., and Van Rosmalen, G.M., 1995, Dynamic modelling of suspension crystallizers using experimental data, *AIChE J.*, 41:571.
- Gahn, C. and Mersmann, A., 1999, Brittle fracture in crystallisation processes. Part A. Attrition and abrasion of brittle solids, *Chem. Engng Sci.*, 54:1273.
- Kramer, H.J.M., Bermingham, S.K., and Van Rosmalen, G.M., 1999, Design of industrial crystallisers for a required product quality, *J. Crystal Growth*, 198/199:729.
- Process Systems Enterprise Ltd, 2003, gPROMS Advanced User Guide, London, UK.