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Systematic observability and detectablity analysis of industrial batch crystallizers ⋆

Marcella Porru * **Leyla Özkan** *

* *Eindhoven University of Technology, De Zaale, 5612AJ Eindhoven, The Netherlands (e-mail: m.porru;l.ozkan@tue.nl).*

Abstract: Motivated by the lack of hardware analysers for particle size distribution (PSD) and solute concentration measurements in industrial crystallizers, this work investigates the feasibility of designing alternative monitoring tools based on state observers. The observability and detectability properties of the discretized population balance equation accounting for crystal growth, attrition and agglomeration coupled with energy and solute mass balances are studied. A systematic methodology for sensor selection based on nonlinear observability and detectability principles is proposed and applied. Results are corroborated by a machine learning technique (the self-organizing map), leading to the fact that the solute concentration is distinguishable with temperature measurements, while the PSD is not. The results represent the starting point for future detector design where temperature measurements are used to infer composition, while the estimation of the PSD is done in "open loop" fashion.

Keywords: Nonlinear observability, nonlinear detectability, batch crystallization monitoring, sensor selection, geometric estimation, self-organizing map.

1. INTRODUCTION

Batch crystallization is an important separation process to obtain high value-added chemicals in crystalline form from liquid solutions in pharmaceutical, food and fine chemical industries. As most of the particulate processes, the quality of the solid product is determined by its particle size distribution (PSD), which is the result of the combination of events at the microscopic and macroscopic scale during the batch run. The microscopic events are governed by complex kinetic interaction between the solute molecules and the crystal lattice, and diffusion mechanisms, whereas the macroscopic events are related to the crystallization operation (solute concentration, temperature profile, mixing). Thus, the achievement of the desired yield and quality targets of the final crystalline product relies on an efficient monitoring tool for both separation supervision and control. However, online measurements of the solute concentration and PSD are not often available due to technological and economical limitations (Simon et al. (2015)). These unmeasurable process variables can be estimated by real time simulation models in parallel to the process. However, models even based on first-principles typically exhibit structural and parametric mismatch with respect to the real process. Thus, the quality of the estimation of the process variables tend to degrade. A remedy for this deficiency is the use of state observers that combine information from two sources, namely a process model and available online measurements.

The natural framework for particulate process modelling is the population balance equation (PBE) (Randolph and Larson (1971)) in the form of a partial differential equation (PDE) describing the evolution of the number of crystals along the size and time domain. However, the PBE cannot be easily employed for estimator implementation purposes, because its analytical solution may not be obtained. Two main approaches have been proposed to overcome this problem: (i) the use of a reduced model in terms of a finite number of its moments (Randolph and Larson (1971)), (ii) and the discretization of the PBE (Hounslow et al. (1988)) resulting in a set of ordinary differential equations.

The problem of designing state observers for monitoring and/or control of the time evolution of the crystal phase has been extensively addressed with the use of the moment model accompanied by: (i) moment measurements (Mesbah et al. (2011)), (ii) solute concentration measurements (Shi et al. (2005)), (iii) moment, composition and temperature measurements (Nagy and Braatz (2003)). This model guarantees simplicity and tractability for online use due to its low dimensionality. However, the reconstruction of the PSD from a finite number of its moments is still an open problem (Cogoni and Frawley (2014)) and nonlinear crystallization phenomena such as agglomeration cannot be incorporated. On the other side, only a few papers deal with the discretized PBE to derive soft sensors for monitoring and control strategies. Mesbah et al. (2012) use it for a moving horizon observer driven by the online measurements of the PSD; Bakir et al. (2006) propose a high gain observer assuming that the measurement of the number of nuclei is available. Finally, Abbas and Romagnoli (2007) propose the use of the model without any innovation term. However, the monitoring strategies mentioned above can be unlikely implemented in industrial scale, because, as highlighted at the beginning of the introduction, online PSD measurements are rarely available.

In this scenario the identification of secondary measurements to derive monitoring schemes based on states observers or detectors for industrial applications still remains an open issue. This motivates a more accurate and systematic analysis of the observability and detectability properties of the crystallization model. In particular, the

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answers to the following questions are researched: (i) can the entire set of performance and process variable be reconstructed through measurements of secondary variables? (ii) can a subset of states be detected through measurements of secondary variables? (iii) what should be the sensor configuration to extract proper information regarding the dynamical evolution of the system?
This paper proposes a systematic methodology amenable to perform the sensor selection (extendible to any process) based on robust exponential (RE-)observability and detectability arguments (Álvarez and Fernández (2009)). The methodology is applied to an industrial batch crystallization system which accounts for growth, attrition and agglomeration phenomena through the discretized PBE. The results are corroborated using a data-derived technique, with the data generated by gCRYSTAL simulations of the model under various operating conditions. For this purpose the self-organizing map (SOM) (Alhoniemi et al. (1999)) accompanied by measures of topological relevance (MTR) (Corona et al. (2012)) has been selected allowing the visualization and quantification of the relationship between primary (i.e. to be estimated) and secondary (i.e. measured) variables. In process engineering the SOM technique has been applied in online and offline fashion for fault detection, modelling, sensitivity analysis (Alhoniemi et al. (1999)), process analysis and, sensor selection coupled with MTR (Corona et al. (2012)) for continuous processes. In this work, the SOM is applied to batch processes for observability and detectability analysis purposes.
Our systematic analysis leads to the following results: the concentration (or temperature) is distinguishable with temperature (or concentration) measurements, while the subset of number of particles per each class of length is not distinguishable with composition and/or temperature measurements. Consequently, the most effective monitoring scheme for the process may be a state detector with innovation based on temperature and/or concentration (if available) measurements on the dynamics of the above mentioned states, while the estimation of the discrete PSD should be performed in open loop fashion. The assessment of the performance of this monitoring scheme is out of the scope of this work.
The paper is organized as follows: the model of the industrial crystallizer is presented, and nonlinear observability and detectability concepts are introduced. Then, the proposed systematic analysis of the observability and detectability properties is explained and applied to the crystallization process. Finally, the results of the approach are corroborated by using the SOM.

2. MODEL OF THE BATCH CRYSTALLIZATION

Consider the seeded flash-cooling crystallization of the chemical a in the solvent s. The model of the process consists of material (for the liquid and solid phases) and energy balances. The particulate feature of the solid product is modelled with the PBE (Randolph and Larson (1971)). Under the assumptions of perfect mixing, size independent crystal growth rate, absence of crystals and solute in the vapour flow, dilute solution, the crystallizer model (1) follows:

$$\frac{dT}{dt} = \frac{\tilde{T}}{V}\frac{dV}{dt} - \frac{F^w h^w}{\rho C p V} + \frac{-\Delta H_c \phi_c}{\rho C p V} = f_T, T(0) = T_0 \quad (1a)$$

$$\frac{dC}{dt} = -C\frac{d(logV)}{dt} - \frac{\phi_c}{V} = f_C, C(0) = C_0 \quad (1b)$$

$$\frac{dV}{dt} = -\frac{F^w}{\rho} = f_V, V(0) = V_0 \quad (1c)$$

$$\frac{\partial n(L)}{\partial t} = -G\frac{\partial n(L)}{\partial L} - n(L)\frac{d(logV)}{dt} + B_0 + B - D = f_n,$$
$$n(0, L) = n_0, n(t, 0) = n(t, \infty) = 0 \quad (1d)$$

Where

$M_2 = \int_0^\infty n(L)L^2 dL$, $\tilde{T} = T^R - T$, $\phi_c = 3\rho_c V k_v G M_2$.
The model (1) accounts for the following the crystallization kinetics (2). The size independent power law kinetics for crystal growth G (2a) is widely used in crystallization modelling (Abbas and Romagnoli (2007); Mesbah et al. (2011)) because of its simplicity. The secondary nucleation B_0 (2b) is modeled through the Evans kinetics (Evans et al. (1974)) when only crystal-impeller collisions are considered. The birth B (2c) and death D (2d) functions due to agglomeration phenomena are modelled according to Hounslow et al. (1988). Note that the modelling of the agglomeration phenomena is a source of nonlinearities for the system. The agglomeration Kernel (2e) is calculated according to an empirical expression.

$$G = k_g \frac{C - C_{sat}(T)}{\rho_c} \quad (2a)$$

$$B_0 = k_{ci}\frac{C - C_{sat}(T)}{C}\frac{N_Q}{N_P}\rho_c k_v \varepsilon \int_{L^{min}}^\infty n(L)L^3 dL \quad (2b)$$

$$B(L) = \frac{L^2}{2}\beta_a \int_0^L \frac{n((L^3 - \lambda^3)^{1/3})n(\lambda)}{(L^3 - \lambda^3)^{2/3}} d\lambda \quad (2c)$$

$$D(L) = n(L)\beta_a \frac{L^2}{2}\int_0^\infty n(\lambda)d\lambda \quad (2d)$$

$$\beta_a = a_1 G \varepsilon \quad (2e)$$

In (2) $k_g, k_{ci}, L^{min}, a_1$ are kinetics parameters whose numerical values are estimated based on plant data. $C_{sat}(T)$ is the solute concentration at saturation. N_Q, N_P and ε are impeller parameters (power and flow numbers, energy dissipation rate respectively).

2.1 Discretization of the population balance equation along the internal coordinate L

The model (1) is a system of integro-partial differential equations which is solved by the use of numerical methods (Abbas and Romagnoli (2007)). The most adopted discretization scheme is the backward differentiation formulae (BDF) also known as upwind procedure.

$$\frac{\partial n}{\partial L} \approx \frac{n_i - n_{i-1}}{\Delta L}, \Delta L \to 0 \quad (3)$$

The application of (3) to (1d) leads to the subset of ODEs

$$\frac{dn_i}{dt} = n_{i-1}\frac{G}{\Delta L} - n_i\left(\frac{G}{\Delta L} + \frac{dlogV}{dt}\right) + B_{0i}\delta(L_1 - L_0) + B_i - D_i$$
$$= f_{n_i}, n_i(0) = n_{i0}, i = 0, ..., N_{max} \quad (4)$$

where the sub-index 0 refers to the minimum size of crystal taken into account (i.e. nuclei dimension), while the sub-index max refers to the maximum size considered. B_{0i}, B_i and D_i are the discretized versions of the kinetics equations (2b)-(2d). The Jacobian $A_{BDF}(n_i, \Delta L, L_i, T, C)$