Systematic observability and detectability analysis of industrial batch crystallizers

Marcella Porru ∗Leyla Özkan ∗

∗Eindhoven University of Technology, De Zaale, 5612 AJ 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 (Houmanski 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

* This work has been done within the project Improved Process Operation via Rigorous Simulation Models (IMPROVISE) in the Institute for Sustainable Process Technology (ISPT).
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 (extendable 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:

\[
\frac{dT}{dt} = \frac{T}{V} \frac{dV}{dt} - \frac{F^w h^w}{\rho C_P V} + \frac{-\Delta H_0 \phi_s}{\rho C_P V} = f_T, \quad T(0) = T_0 \quad (1a)
\]

\[
\frac{dC}{dt} = -\frac{C}{\tau} \frac{d(log V)}{dt} - \frac{\phi_s}{V} = f_C, \quad C(0) = C_0 \quad (1b)
\]

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

\[
\frac{\partial n(L)}{\partial t} = -G \frac{\partial n(L)}{\partial L} - n(L) \frac{d(log V)}{dt} + B_0 + B - D = f_n,
\]

\[
n(0, L) = n_0, \quad n(t, 0) = n(t, \infty) = 0 \quad (1d)
\]

Where

\[
M_2 = \int_0^\infty n(L) L^2 dL, \quad \tilde{T} = T^R - T, \quad \phi_s = 3 \rho_s V k_0 GM_2.
\]

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 Houmslov 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.

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

\[
B_0 = k_{ci} C - C_{sat}(T) \frac{N_c G_{ci} \varepsilon}{N_p} \int_{L_{min}}^\infty n(L) L^3 dL \quad (2b)
\]

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

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

\[
\beta_a = a_1 G e \quad (2e)
\]

In \(2\) \(k_g, k_{ci}, L_{min}, a_i\) are kinetics parameters whose numerical values are estimated based on plant data. \(C_{sat}(T)\) is the solute concentration at saturation. \(N_c, N_p, \varepsilon\) 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 formulation (BDF) also known as upwind procedure.

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

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

\[
\frac{dn_i}{dt} = n_{i-1} \frac{G}{\Delta L} - n_i \frac{G}{\Delta L} + \frac{d(log V)}{dt} + B_0 \delta(L_i - L_0) + B_i - D_i = f_{n_i}, \quad n_0(i) = n_{i0}, \quad i = 0, ..., N_{max} \quad (4)
\]

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. \(B_0, B_i, D_i\) are the discretized versions of the kinetics equations (2b)-(2d). The Jacobian \(A_{BDF}(n_i, \Delta L, L_i, T, C)\)

497
of \( f \), with respect of the states \( n = [n_0, ..., n_{N_{\text{max}}}]' \) is shown in (5)

\[
A_{BDF} = \frac{\partial f_n}{\partial n} = \begin{bmatrix}
-\epsilon - d_1 & 0 & 0 & \cdots & 0 \\
\frac{G}{\Delta T} & -\epsilon - d_2 & 0 & \cdots & 0 \\
\vdots & \vdots & \vdots & \ddots & \vdots \\
0 & 0 & \cdots & \cdots & -\epsilon - d_{N_{\text{max}}} \\
\end{bmatrix}
\]

In (5) \( \epsilon = \left( \frac{G}{\Delta T} + \frac{d \log V}{dt} \right) ; \frac{d \phi_i}{dn} = \beta_n \frac{L_n^2}{2} \int_0^\infty n(\lambda) d\lambda \)

where the integral \( \int_0^\infty n(\lambda) d\lambda \) have to be integrated numerically from 0 to \( N_{\text{max}} \) based on (3).

3. ESTIMATION PROBLEM

In crystallization processes as well as in most of the particulate processes, the PSD influences the quality of the final product. However, the online monitoring of this performance indicator remains a challenge. Indeed, the measurement techniques are usually based on offline analysis of slurry samples (e.g., image analysis), while online instruments (e.g., focussed beam reflected measurement probes) are expensive and difficult to calibrate (Simon et al. (2015)). Thus, hardware analysers are not usually able to efficiently perform the monitoring task. Instead, state estimators or detectors can be used. These software sensors allow to infer the variables of interest based on the model of the process and the available measurements. The effectiveness of the process monitoring with this technology depends on basic properties of the model: the observability and the detectability. These concepts are explained in the following Subsection.

3.1 Statement of the observability and detectability analysis problem

Let us consider the system (1a)-(1c), (4) consisting of \( N = 3 + N_{\text{max}} \) ODEs, which are nonlinear and coupled. Let us rewrite the nonlinear system in the standard form:

\[
\dot{x} = f(x,u(t)), y = h(x); x(0) = x_0
\]

**Observability:** The observation space \( \phi \) of a nonlinear system in the form (6) is the space of functions containing the output map \( h \) and all the repeated Lie derivatives:

\[
\phi(x, U(t)) = [h, L_1h, ..., L_\kappa h], \kappa = N, \dim(\phi) = N
\]

where \( U(t) = [u_1, u_2, ..., u_1^{v_1-1}, ..., u_1 u_2^{v_2-1}, ..., u_1 u_2 u_3^{v_3-1}], v_1 + ... + v_p = p \). The observability space \( \phi \) defines the (input derivative-independent) observability codistribution \( O(x, u(t)) \) (Porru (2015)):

\[
O(x, u(t)) \cong \Xi(x, U(t)) = \frac{\partial}{\partial x} \phi, \dim(O) = N \times N
\]

Based on (8) the system (6) is robustly exponentially (RE-) observable with the measurement \( y \) (Alvarez (2000)) if the observability conditions (9) hold:

\[
\text{rank}(O) = N, \text{cond}(O) < 1
\]

**Geometric Observer:** When the observability conditions (9) hold, the states of the systems can be reconstructed from the measurement and its time derivatives \( \Upsilon(t) = [y, y^{\kappa-1}] \) solving the algebraic system \( \Upsilon(t) = \phi(x, U(t)) \) for \( x: U(t) = \phi^{-1}(\Upsilon(t)) \), or alternatively, through the state geometric observer (Alvarez (2000)):

\[
\dot{x} = \hat{f}(\hat{x}, u(t)) + \Phi K(y - h(\hat{x})), \hat{x}(0) = \hat{x}_0
\]

where \( \Phi = O^{-1} \) and \( K \) is a proper tuning matrix.

**Detectability:** When the conditions (9) do not hold, one might do a partition between distinguishable \( x \) and undistinguishable \( x_s \) states so that the least strict robust exponential (RE-)detectability conditions (11) hold (Alvarez (2000)).

\[
\text{rank}(O_i) = \kappa_i, \text{cond}(O_i) < 1
\]

with \( O_i(x, u(t)) \approx \Xi(x, U(t)) = \partial \phi_i / \partial x_i \),

\[
\phi_i = [h, L_1h, ..., L_{\kappa_i} h], \dim(\phi_i) = \dim(x_i) = \kappa_i < N
\]

where \( \Phi_D = O_i^{-1} \) and \( K_D \) is a proper tuning matrix.

More than one measurement: \( y = h(x), y = [y_1, ..., y_m]' \). In this case the previous discussion can be generalized substituting:

(i) eq.(7) with eq.(14)

\[
\dot{x}_v = \dot{\hat{x}}(\hat{x}, u), \hat{x}_v(0) = \hat{x}_0
\]

(ii) eq.(12) with eq.(15)

\[
\phi = [h_1, L_1h_1, ..., L_{\kappa_1} h_1], ..., h_m, L_1h_m, ..., L_{\kappa_m} h_m]
\]

where \( \dim(\phi) = \kappa_1 + ... + \kappa_m = N \) and \( \kappa_i \) is the observability index related to the measurement \( y_i \),

(iii) and substituting \( (y - h(x)) \) instead of \( (y - h(x)) \) into the observer (10) and the detector (13a).

This concepts constitute the basis of the proposed methodology for systematic observability and detectability analysis described in Section 4.

4. METHODOLOGY FOR SYSTEMATIC OBSERVABILITY AND DETECTABILITY ANALYSIS

A systematic methodology for the observability and detectability analysis is proposed based on the concepts previously reviewed. The methodology consists of the steps:

1. The analysis is performed considering each available measurement one at a time.
2. The \( -1 \) Lie derivatives of the output map \( h \) of the first measurement are computed.
3. If the related \( O(x, \Upsilon(t)) \approx \partial \phi / \partial x \) (8) satisfies the observability conditions (9) then it must be concluded that the system is RE-observable with the measurement.
4. If not, a \( \phi \) matrix with \( 1 < \kappa < N \) may exists in such a way that the (RE-)detectability conditions (11) are satisfied. Here, the state partition \( x = [x_k, x_s] \) of distinguishable and undistinguishable states is easily identified.
5. If \( \kappa = 0 \) the states of the system are neither observable nor detectable with the considered measurement.
6. Repeat the steps 1-5 for the entire set of measurements.

At this point the effect of the combination of the entire set of measurements must be considered. This can be done as:

7. Consider the entire set of output maps \( h \) and their \( \kappa_{i_1} + 1 + ... + \kappa_{i_m} = 1 \) Lie derivatives previously calculated. If there exists a combination of them (eventually discarding the highest order Lie derivatives) in such a way that a \( \phi \) matrix
in the form (14) can be found with $\sum_{i=1}^{m} \kappa_i = N$, and satisfying the observability conditions (9), then the states of the system are (RE-)observable with the selected measurements.

8. If instead a $\phi$ matrix in the form (15) is found, with $\sum_{i=1}^{m} \kappa_i \leq N$ and satisfying (11), then only a subset $x_i$ can be distinguished with the available measurements.

5. OBSERVABILITY AND DETECTABILITY ANALYSIS FOR THE BATCH CRYSTALLIZER

In this Section, the observability and detectability analysis is performed for the crystallization model (6), with the main goal to investigate the distinguishability of the subset of states $n_i, i = 1, ..., N_{\text{max}}$. The analysis follows the procedure proposed in Section 4, assuming that temperature, concentration and volume measurements are available.

5.1 Temperature measurements

In case of temperature measurements the output map assumes the linear form: $y_1 = C_1 x, C_1 = [1, 0, 0, 0, 0]^T$. The corresponding repeated Lie derivatives sequence in the $\phi_T$ matrix is:

$$
\phi_T = \begin{bmatrix}
L h(T, C, V) \\
L^2 h(T, C, V) \\
\vdots \\
L^{N-1} h(T, C, V)
\end{bmatrix}
= \begin{bmatrix}
\frac{\partial f_T}{\partial T} + \frac{\partial f_C}{\partial C} + \frac{\partial f_V}{\partial V} \\
\frac{\partial f_T}{\partial T} + \frac{\partial f_C}{\partial C} + \frac{\partial f_V}{\partial V} \\
\vdots \\
\frac{\partial f_T}{\partial T} + \frac{\partial f_C}{\partial C} + \frac{\partial f_V}{\partial V}
\end{bmatrix}
= \begin{bmatrix}
T \\
T \\
\vdots \\
T
\end{bmatrix}
$$

(16)

Thus, it is easy to see that the observability codistribution $O_T(x, u(t)) = \partial \phi_T(x, u(t))/\partial x$ has rank 3 and it is $n_i$ independent. However, $\phi_{i,T}(x, u(t)) = [h, L_i h, L_i^2 h, \ldots]$ with $\kappa_{i,T} = 3$ generates a $O_{i,T}$ satisfying the detectability conditions (11) for the distinguishable states $x_i,T = [T, C, V]^T$. This in turn means that (i) the subset of state $T, C, V$ is distinguishable with temperature measurements while (ii) the particle size distribution $n_i, i = 1, ..., N_{\text{max}}$ is not. Moreover, from (16) one can note that the solute concentration $C$ is undistinguishable with temperature measurements when $\Delta H_c = 0$ or $G \to 0$, verified for $C = C_{\text{sat}} \to 0$. In this case the set of distinguishable states reduces to $x_{i,T} = [T, V]^T$.

5.2 Composition measurements

In this case the output map takes the linear form: $y_2 = C_2 x, C_2 = [0, 1, 0, 0, 0]^T$. The corresponding Lie derivatives sequence in the $\phi_C$ matrix is:

$$
\phi_C = \begin{bmatrix}
L h(T, C, V) \\
L^2 h(T, C, V) \\
\vdots \\
L^{N-1} h(T, C, V)
\end{bmatrix}
= \begin{bmatrix}
\frac{\partial f_C}{\partial T} + \frac{\partial f_C}{\partial C} + \frac{\partial f_C}{\partial V} \\
\frac{\partial f_C}{\partial T} + \frac{\partial f_C}{\partial C} + \frac{\partial f_C}{\partial V} \\
\vdots \\
\frac{\partial f_C}{\partial T} + \frac{\partial f_C}{\partial C} + \frac{\partial f_C}{\partial V}
\end{bmatrix}
= \begin{bmatrix}
C \\
C \\
\vdots \\
C
\end{bmatrix}
$$

(17)

Thus, it is easy to see that $O_C(x, u(t)) = \partial \phi_C(x, u(t))/\partial x$ has rank 3 and it is $n_i$ independent. Similarly to the previous case, one can identify the subset of state $x_{i,C} = [T, C, V]^T$ which is distinguishable, while the particle size distribution $n_i, i = 1, ..., N_{\text{max}}$ cannot be distinguished by measuring the solute concentration. It must be pointed out that the temperature $T$ is not distinguishable with concentration measurements when $G \to 0$.

5.3 Volume measurements

In this case the output map takes the linear form: $y_3 = C_3 x, C_3 = [0, 0, 1, 0, 0]^T$. The $\phi_V$ matrix is:

$$
\phi_V = \begin{bmatrix}
h = V \\
L h \\
L^2 h \\
\vdots \\
L^{N-1} h
\end{bmatrix}
= \begin{bmatrix}
0 \\
\frac{\partial f_V}{\partial x_i} f_i = 0
\end{bmatrix}
$$

(18)

Accordingly $O_V(x, u(t)) = \partial \phi_V(x, u(t))/\partial x$ has rank 1, leading to the conclusion that for the purposes at hand the volume measurement can only be used to monitor the evolution of the volume itself.

5.4 Temperature, concentration, and volume measurements

Finally, the effect of the entire set of measurements must be considered. From the results obtained in the previous Subsections one can easily conclude that neither with single nor multiple measurements the subset of $n_i$ states is distinguishable.

5.5 Observability of the subset of states $n_i$

For the sake of completeness, it should be also mentioned here that the dynamics of the subset of $n_i, i = 1, ..., N_{\text{max}}$ states can be theoretically observed with the only measurement of the number of particles of length corresponding to the class $N_{\text{max}}$, due to the lower bidiagonal structure of the $A_{\text{BDF}}$ (5) if and only if the terms $\epsilon - d_i$ appreciably differ from zero, which might be easy to verify. It must be also pointed out that the choice of the discretization scheme modifies the structure of the $n_i$ state matrix (5), which in turn affects the observability properties of the submodel.

5.6 Remarks

This Section has shown the application of the systematic procedure for observability and detectability analysis proposed in Section 4. Through this methodology, the set of distinguishable states $x_i$ can be obtained looking at the output map and its sequence of Lie derivatives, identifying the functionality of the $\phi$ matrix with the states, and considering one measurement at a time. Moreover, the proposed analysis allows the identification of regions of operating conditions that generate deficiency in observability or detectability, as it happens when $G \to 0$ (which is in general verified at the end of the batch run). In the following Section, the result of the analysis are corroborated with the use of a data-derived technique, able to give a quantification of the information transmitted from the measurement to the process states and of easy interpretation by practitioners.

6. DETECTABILITY RESULTS ASSESSMENT THROUGH DATA-DERIVED APPROACHES

In Section 5 suggestive detectability results are given based on nonlinear observability and detectability concepts. However, one might not be able to draw conclusions in those cases when the Lie derivative sequence is difficult to compute (as mentioned in Subsection 5.5), which can happen especially for highly coupled, high dimensional,
structurally uncertain, or unsteady nonlinear ODE systems. Moreover, the analysis based on observability and detectability concepts may lack of intuitive interpretation, which is important at the moment of the commissioning of the software sensor to support the academic research to its final applications and uses. In this Section, a data-derived technique able to assist and corroborate the observability and detectability analysis is applied.

Data is obtained through numerical simulation of the model (6) with the software gCRYSTAL. The values of the process variables and the state trajectories during several batch runs with different initial conditions are collected. The data set is employed into the framework of a classical machine learning method: the self-organizing map (SOM). Measure of topological relevance (MTR) on the SOM are employed to quantify the relationship between primary and secondary process outputs.

6.1 Brief description of the self-organizing map

The self-organizing map (SOM) is a type of artificial neural network (ANN) which is able to deal with unsupervised problems and is thus applicable to process monitoring tasks. SOMs consist of an usually high dimensional input layer (the data set) and an output layer which is arranged into a two-dimensional grid. The grid has $N$ nodes. A prototype vector $m_R$ with dimension equal to the number of measured variables is associated to each node. During the training the informative content of the data is projected into the $m_R = [m_R_1,...,m_R_n]$ vectors. In this way, the grid is arranged so that the low dimensional representation of the process data preserves its high dimensionality topology (see Corona et al. (2012) and Alhoniemi et al. (1999) for more details about the training procedure). In process engineering the SOM has been applied in online and offline fashion for fault detection, modelling, sensitivity analysis, process analysis and, coupled with MTR, sensor selection for continuous processes. In this work, the field of application is extended to batch processes for observability and detectability analysis purposes. In particular the SOM is used for data visualization and variables ranking. The visualization technique used is the component plane. A component plane shows the coordinates of the prototype vectors along a specific direction in the data space; that is, each component plane is associated with one original variable. The SOM tool offers the possibility to order component planes in such a way that planes (and thus process variables) that show high similarities are placed near each other. Moreover, similarities are quantified calculating MTR on SOM for each possible-variable coupling. This allows to rank the variables according to their relevance with the variable to be estimated. SOM and MTR are computed using the SOM Toolbox for Matlab available from http://cis.hut.fi/projects/somtoolbox/.

6.2 Implementation on the batch crystallization model

A data set consisting of $J = 20$ process variable (listed in Table 1) is taken for $K = 24$ time intervals with duration of 300 s from the simulation of $I = 19$ batch runs based on the model (6), simulated in gCRYSTAL. The batch runs differ in initial seeding, solute concentration, impeller conditions and temperature profiles. The data set is organized in a three-way array $D(I \times J \times K)$ which has been unfolded with respect to the $K$ dimensions, to analyse the variability among the batches with respect both to variables and their time variation (Nomikos and MacGregor (1994)). According, 24 SOMs (one per sample time) are trained with the two-way array $D(I \times J)$. Ordered component planes are computed and analysed. Fig. 1 shows the ordered component plane at time interval $k = 15$; the ordered planes for the other sample times lead to similar results and are not presented here. The ordered component plane

| Relevant Time-Varying Process Variables | States | Initial Conditions |
|----------------------------------------|--------|--------------------|
| Temperature (Temp), Solute concentration (Conc) | $n_1_{1um}$ (Cla1), $n_{100um}$ (Cla2), $n_{300um}$ (Cla3) | Impeller frequency (Ifre), Solute concentration (Icon) |
| $n_{700um}$ (Cla4), $n_{1500um}$ (Cla5), Volume (Vol) | Seeds SD mean (Iloc), Seeds SD spread (Idev) Seed mass (Imass) |
| | 50% quantile (D50), Supersaturation (Ssat), Vapor Flow (Vapf) | |
| Mass of crystals (Mercy), Solid fraction (Sfac) | Second Moment of the PS distribution (M2) |

Table 1. List of the process variables and tags

![Fig. 1. Ordered component map at time interval $k = 15$](image-url)
identifying which measurement is more suited to infer the variable of interest. Histograms report that: (i) the temperature can be inferred with concentration measurements and vice versa. (ii) Even if in the correlation map (Fig.1) the temperature, composition and volume planes where clustered in the same region, the MTR shows that the measurement of the volume does not inject information for temperature and concentration detection. (iii) During the batch run the mutual relevance is varying. In particular, at the beginning of the batch run the correlation between composition and temperature (and vice versa) is low, then passes through a maximum and decreases again. This may be related with the variation of the supersaturation, as discussed in Subsections 5.1 and 5.2. This may result in time varying performance of a state detector, when implemented. (iv) Finally, the PSD is uncorrelated with all the considered variable, confirming that the subset of \( n_{i}, i = 1, ..., N_{\text{max}} \) states is undistinguishable with any considered measurement.

7. CONCLUSIONS
In this paper a systematic methodology for observability and detectability analysis of nonlinear systems has been proposed. The methodology has been used to study the detectability properties of an industrial batch crystallization process accounting for growth, attrition and agglomeration phenomena, modelled with the discretized PBE. The outcome of this study shows that the solute concentration is distinguishable through temperature measurements, while the PSD is undistinguishable either with temperature and concentration measurements. To the best of our knowledge there are no papers reporting this result. The results are corroborated using a data-derived technique: the SOM. In previous studies, the SOM has been applied to continuous processes: here its use has been extended to batch processes through an unfolding procedure of the training dataset. The obtained results represent the starting point for the design of a monitoring tool based on state detectors, when measurements of the temperature (and concentration if available) can be used to correct the prediction of the temperature, concentration and volume given by an estimation model, while the PSD is inferred through the model in open loop fashion.

REFERENCES
Abbas, A. and Romagnoli, J.A. (2007). Multiscale modeling, simulation and validation of batch cooling crystallization. Separation and purification technology, 53(2), 153–163.
Alhonimi, E., Hollmén, J., Simula, O., and Vesanto, J. (1999). Process monitoring and modeling using the self-organizing map. Integr. Comput. Aided Eng, 6(1), 3–14.
Alvarez, J. (2000). Nonlinear state estimation with robust convergence. Journal of Process Control, 10(1), 59–71.
Álvarez, J. and Fernández, C. (2009). Geometric estimation of nonlinear process systems. Journal of Process Control, 19(2), 247–260.
Bakir, T., Othman, S., Fovette, G., and Hammouri, H. (2006). Nonlinear observer of crystal-size distribution during batch crystallization. AIChE Journal, 52(6), 2188–2197.
Cogoni, G. and Frawley, P. (2014). Particle size distribution reconstruction using a finite number of its moments through artificial neural networks: A practical application. Crystal Growth & Design, 15(1), 239–246.
Corona, F., Mulas, M., Baratti, R., and Romagnoli, J.A. (2012). Data-derived analysis and inference for an industrial deethanizer. Industrial & Engineering Chemistry Research, 51(42), 13732–13742.
Evans, T., Sarofim, A., and Margolis, G. (1974). Models of secondary nucleation attributable to crystal-crystallizer and crystal-crystal collisions. AIChE Journal, 20(5), 959–966.
Houlsow, M., Ryall, R., and Marshall, V. (1988). A discretized population balance for nucleation, growth, and aggregation. AIChE Journal, 34(11), 1821–1832.
Mesbah, A., Huesman, A.E., Kramer, H.J., and Van den Hof, P.M. (2011). A comparison of nonlinear observers for output feedback model-based control of seeded batch crystallization processes. Journal of Process Control, 21(4), 652–666.
Mesbah, A., Nagy, Z.K., Huesman, A.E., Kramer, H.J., and Van den Hof, P.M. (2012). Nonlinear model-based control of a semi-industrial batch crystallizer using a population balance modeling framework. IEEE Trans. on Control Systems Technology, 20(5), 1188–1201.
Nagy, Z.K. and Braatz, R.D. (2003). Robust nonlinear model predictive control of batch processes. AIChE Journal, 49(7), 1776–1816.
Nomikos, P. and MacGregor, J.F. (1994). Monitoring batch processes using multiway principal component analysis. AIChE Journal, 40(8), 1361–1375.
Porru, M. (2015). Quality regulation and energy saving through control and monitoring techniques for industrial multicomponent distillation columns. Ph.D Thesis, Universita’ degli Studi di Cagliari.
Porru, M., Alvarez, J., and Baratti, R. (2013). A distillate composition estimator for an industrial multicomponent i4c4-n4c4 splitter with experimental temperature measurements. IFAC Proceedings Volumes, 10(part1), 391–396.
Randolph, A.D. and Larson, M.A. (1971). Theory of particulate processes, analysis and techniques of continuous crystallization. Academic Press.
Shi, D., Bhaskar, P., El-Farra, N.H., and Christofides, P.D. (2005). Predictive control of crystal size distribution in protein crystallization. Nanotechnology, 16(7), S562–S574.
Simon, L.L. et al. (2015). Assessment of recent process analytical technology (pat) trends: a multiauthor review. Organic Process Research & Development, 19(1), 3–62.