

# Continuous-discrete observer for crystal size distribution of batch crystallization process

Toufik Bakir and Sami Othman and François Puel and Hassan Hammouri

**Abstract**—A continuous-discrete observer was designed to estimate the CSD (Crystal Size Distribution) in batch crystallization processes. The observer is based on the discretization of PBE (population balance equations), it describes the evolution of the CSD with the finite difference method. Since the discretization leads a high order model, a reduced model was then constructed in order to simplify the observer synthesis and to reduce the computation time. The observer's output allows to calculate the moments of the CSD, which may be used for control purposes or process supervision.

**Keywords:** Crystallization processes, high gain observer, continuous-discrete observer, population balance equations, crystal size distribution, finite difference method.

## I. INTRODUCTION

Batch crystallization is the first and probably the main unit operation dealing with solids in chemical plants. There is a real challenge to produce crystals with specific properties allowing to perform downstream operations more easily thanks to mastered filterability, low fines concentration, low trend to cake on storage, good flowability, etc. Consequently, in the industrial practice, the control of the chemical purity is no more sufficient and the control of Crystals Size Distribution (CSD) can be critically important. However, measuring or estimating the CSD remains an open field of research. On line calorimetric or turbidimetric measurements have been used, together with off-line image analysis for the modelling of the time variation of CSD during batch solution crystallizations by [1] and [2]. However it is important to notice that no reliable online CSD measurements or estimates were available when such modelling studies were published.

Several sensors in situ technologies are actually available which allow to estimate, at least partially, the CSD during crystallization processes. However, none of these can really provide measurements of the whole CSD, moreover the physical significance of the measured sizes often remains questionable. The aim of the present work is to design an on line observer so as to reconstruct the whole crystal size distribution using population balance equations, and on-line partial measurements. For example, measuring the number of nuclei (birth of crystals) can be performed using the FBRM probe (Focussed Beam Reflected Measurement). Such sensor provides reasonable accuracy for the monitoring of the generation of nuclei, while bigger crystals cannot be satisfactorily measured. Therefore, one can expect the whole CSD to be reconstructed through the use of continuous discrete observers. As far as such observer is based on PBE

describing the time variations of the CSD, it is essential that the PBE captures the main dynamics features of the process in order to yield good estimates. Indeed, the crystallization rate (i.e. the rate of production of crystals) depends on two mechanisms : nucleation and crystal growth. With the assumption of invariant kinetics parameters, and knowing the different moments of the CSD, the control of such process had been explored by [3], [4] and [5]. In this work, it will be shown that a reduced continuous-discrete observer may give a very satisfactory results.

The paper is organized as follows, first the batch crystallization model is briefly described. The principle of discretization of the PBE is then exposed in section 3. Section 4 is devoted to the observer synthesis. In section 5, the estimation technique is validated through simulation.

## II. MODEL DEVELOPMENT

Modelling particulate processes such as crystallization is possible using the formalism of Population Balance Equations (PBE) which was developed by [6]. Applied to the batch crystallizer process, this approach yields the following partial differential equation :

$$\frac{\partial n(x,t)}{\partial t} + G(t) \frac{\partial n(x,t)}{\partial x} = 0, \quad (1)$$

$n(x,t)$  is a number population density function. It represents the number of crystals of size  $x$  per unit volume of volume of suspension and per unit of size. In this model, only nucleation and growth are considered, agglomeration and breakage are not taken into account. The growth kinetic  $G(t)$  is size independent.

The solute concentration balance describing the mass transfer from the liquid to the solid phase is:

$$\frac{dV_l(t)C(t)}{dt} + \frac{dV_T(t)C_s(t)}{dt} = 0, \quad (2)$$

$C(t)$  represents the solute concentration,  $V_T(t)$  is the suspension volume, the variation of this volume due to the mass transfer is very small, this variation can be neglected.  $C_s(t)$  being the solid concentration, it is deduced from the crystal size distribution (CSD) :

$$C_s(t) = \frac{K_v \rho_s}{M_s} \int_0^\infty x^3 n(x,t) dx, \quad (3)$$

where  $K_v$  is a shape factor, for a sphere  $K_v = \frac{\pi}{6}$ ,  $M_s$  the molecular weight of solid,  $\rho_s$  the density of the solid, and

$V_i(t)$  is the solution volume (i.e. the continuous phase), which is calculated from the following expression :

$$V_i(t) = V_T(t) \left(1 - \frac{M_s}{\rho_s} C_s(t)\right) \quad (4)$$

The crystallizer temperature is described by the energy balance around the jacket wall :

$$\sum_{i=1}^3 C_{p_i} n_i \frac{\partial T_{cr}}{\partial t} = -\Delta H_c V_T \frac{dC_s}{dt} - U A_c (T_{cr} - T_j) \quad (5)$$

where  $C_{p_i}$  and  $n_i$  represent respectively the thermic molar capacities and the number of moles of the different components in the crystallizer.  $T_{cr}$  and  $T_j$  are respectively crystallizer and jacket temperatures.  $\Delta H_c$  is the crystallization enthalpy.  $U$  and  $A_c$  are respectively the transfer energy and the contact surface.

The solubility, which refers to the solute concentration under saturated conditions, is assumed to obey to Van't Hoff equation:

$$C_{sat}(T) = A \exp\left(\frac{-\Delta H_c}{RT}\right) \quad (6)$$

The absolute supersaturation ( $C - C_{sat}$ ) is the driving force of the crystallization process. The overall growth rate, including possible diffusive limitations, is assumed to be represented by the following simple power law. Exponential values of exponent  $g$  where generally found to lie between 1 and 2 :

$$G(t) = K_c \frac{M_s}{2\rho_s} \eta (C - C_{sat})^g \quad (7)$$

where  $K_c$  represents the kinetic growth rate coefficient,  $\eta$  represents the effectiveness factor which is the solution of the following equation :

$$\frac{K_c}{K_d} (C - C_{sat})^{g-1} \eta + \eta^{\frac{1}{g}} - 1 = 0 \quad (8)$$

$K_d$  represents the mass transfer coefficient by diffusivity and convection. This coefficient is assumed to be the same for all crystal sizes. Analytical solution of equation (8) is available if  $g$  is equal to 1 or 2, a numerical solution can be considered in the other case.  $B$  is the result of two competitive nucleation mechanisms. Primary nucleation takes place in the absence of any crystal in the solution :

$$B_1 = A_1 \exp\left(\frac{B_0}{\ln^2\left(\frac{C}{C_{sat}}\right)}\right), \quad (9)$$

and secondary nucleation, which may occur at lower supersaturation level, is favored by the presence of solid in suspension (i.e. added in the crystallizer through seeding or generated through primary nucleation) :

$$B_2 = A_2 M_T^j (C - C_{sat})^j \quad (10)$$

$A_1$ ,  $A_2$  and  $B_0$  are constants and  $M_T$  is the crystal mass in the solution. The boundary condition for equation (1) is usually set as follows, where only crystal nuclei of critical size  $x^*$  are assumed to grow :

$$n(x^*, t) = \frac{B(x^*)}{G(x^*)} \simeq \frac{B}{G} \quad (11)$$

$B$  is the sum of primary and secondary nucleation. more details concerning the boundary condition can be found in [7]. The modelling of such process is very difficult and the different parameters present uncertainties, an on line identification of these parameters can be done in order to cope with modelling errors. Industrial procedures involve the use of a seeding procedure, in order to avoid the development of primary nucleation. Two cases arise for managing the process, related to clear solution (case 1) and seeded solution (case 2). The first one (case 1) produces huge number of nuclei compared to the second one (case 2).

### III. DISCRETIZATION OF THE PBE "POPULATION BALANCE EQUATION"

Many discretization methods were developed for solving PBE by [7] and [8]. To our concern, two methods are applied : finite difference method which is widely developed in numerical analysis books, and collocation method supported by [9] where the discretization of the PDE (Partial Differential Equation) using this method is sufficiently developed. The first method corresponds exactly to the observer structure. Indeed, the state matrix has tri-diagonal form. Moreover it agrees with the physical behavior of the system. Collocation method gives a polynomial approximation with correlation between different states without any link with physical phenomenon.

After the discretization of the PBE, the final system of equations is :

$$\begin{cases} \dot{n}_x(t) = \alpha(t) A n_x(t) \\ y(t) = C_1 n_x(t) \end{cases} \quad (12)$$

with:  $\alpha(t) = \frac{G(t)}{\Delta x}$ ,

$$n_x = \begin{pmatrix} n_{x_1} \\ n_{x_2} \\ n_{x_3} \\ \vdots \\ n_{x_{N-1}} \\ n_{x_N} \end{pmatrix}, \quad A = \begin{pmatrix} 1 & -1 & 0 & \dots & 0 \\ \frac{1}{2} & 0 & -\frac{1}{2} & \ddots & \vdots \\ 0 & \ddots & \ddots & \ddots & 0 \\ \vdots & \ddots & \frac{1}{2} & 0 & -\frac{1}{2} \\ 0 & \dots & 0 & 0 & 0 \end{pmatrix},$$

$$C_1 = (1 \ 0 \ \dots \ \dots \ 0),$$

where  $n_x \in \mathbb{R}^N$ ,  $A \in \mathbb{R}^N \times \mathbb{R}^N$  and  $C_1 \in \mathbb{R}^N$ .  $n_x$  represents the value of the function  $n(x, t)$  for a group of crystal sizes ( $x_1, x_2, x_3, \dots, x_N$ ) at time  $t$ .

The moment's equations of the CSD are given by :

$$\mu_i = \int_{x^*}^{x_{\infty}} x^i n(x, t) dx \quad i = 0, 1, 2, 3 \quad (13)$$

The moments computation allows the evaluation of certain values that characterizes the CSD like the number mean size  $L_{pop}$  and the variation coefficient  $VC_{pop}$  which are calculated as follow :

$$L_{pop} = \frac{\mu_1}{\mu_0} \quad (14)$$

$$VC_{pop} = \sqrt{\frac{\mu_0 \mu_2}{\mu_1^2} - 1} \quad (15)$$

#### IV. HIGH GAIN CONTINUOUS- DISCRETE OBSERVER SYNTHESIS

In the case of single output systems, the high gain observer is dedicated to the class of uniformly observable systems of the following form :

$$\begin{cases} \dot{z} = f(z) + \sum_{i=1}^N u_i g_i(z) \\ y = h(z) \end{cases} \quad (16)$$

where  $z(t) \in \mathbb{R}^N, y \in \mathbb{R}, u \in \mathbb{R}^p$

System (16) is said to be uniformly observable if for two initial states  $z \neq \bar{z}$  and all admissible inputs defined on any  $[0, T]$ , there exists  $t \in [0, T]$  such that  $y(z, u, t) \neq y(\bar{z}, u, t)$ , where  $y(z, u, t)$  is the output associated to the initial state  $z$  and the input  $u$ .

In our case, system(16) takes the particular form of system (12). This system is clearly observable due to its triangular form. The canonical form may be used to construct an exponential observer for system (12) under the following assumption :

$0 < \gamma \leq \alpha(t) \leq \xi \quad \forall t \geq 0$ , for some constants  $\gamma$  and  $\xi$ .

In the case of continuous measurements, a candidate exponential observer for this system is given by [10] and [11] :

$$\dot{\hat{z}}(t) = \alpha(t)A\hat{z}(t) - \alpha(t)S_{\theta}^{-1}C_1^T(C_1\hat{z}(t) - Y(t)), \quad (17)$$

where  $S$  is symmetric positive definite matrix given by the following equation :

$$\dot{S}_{\theta}(t) = -\theta S_{\theta}(t) - A^T S_{\theta}(t) - S_{\theta}(t)A + C_1^T C_1 \quad (18)$$

If  $\alpha(t)$  is negative for all the time, it implies the change of the correction term sign :

$$\begin{cases} \dot{\hat{z}}(t) = \alpha(t)A\hat{z}(t) + \\ \alpha(t)S_{\theta}^{-1}C_1^T(C_1\hat{z}(t) - Y(t)) \end{cases} \quad (19)$$

Considering now that the measurements of the system (12) are given at periodic intervals, i.e.  $y(t_k) = C_1 z(t_k)$ , with  $t_k = kT_s$  where  $T_s$  is the sampling time period. A continuous discrete observer algorithm was proposed by [12] and [13]. The principle of this observer is the following :

1) a prediction step for  $t \in [t_k, t_{k+1}[$

$$\begin{cases} \dot{\hat{z}}(t) = \alpha(t)A\hat{z} \\ \dot{S}_{\theta}(t) = -\theta S_{\theta}(t) - A^T S_{\theta}(t) - S_{\theta}(t)A \end{cases} \quad (20)$$

In this step, there is no correction term. So the estimator is a copy of the process model.

2) a correction step at  $t = t_{k+1}$

$$\begin{cases} S(t_{k+1}) = S(t_{k+1}^-) + T_s C_1^T C_1 \\ \hat{z}(t_{k+1}) = \hat{z}(t_{k+1}^-) - k\alpha(t)S_{k+1}^{-1}C_1^T \\ (C_1\hat{z}(t_{k+1}^-)(t) - Y(t_{k+1})) \end{cases} \quad (21)$$

In the second step, correction term is explicitly given.  $S(t)$  is a time varying, symmetric positive definite matrix

and  $k$  and  $\theta$  are a fixed tuning parameters. The expression  $(t_{k+1}^-)$  represents the limit value of a variable at time  $t_{k+1}$  :

$$S(t_{k+1}^-) = \lim_{t \rightarrow t_{k+1}} S(t) \quad (22)$$

In [13], it was shown that this observer converges exponentially for  $T_s$  small enough and  $\theta \in [\theta_0, \theta_1]$ , where  $\theta_0$  and  $\theta_1$  are two positive constants.

Seeded solution deals with the use of the model (12). Here the discretization is based upon about 50 samples. This number was found to allow satisfactory accuracy of the model representation. Thus we used the same number of samples in the observer synthesis. While in case 1 (unseeded solution) we took  $N$  equal to 100 to improve the model precision. This number could be reduced for the observer to a quarter ( $N_{observer} = 25$ ). This simplification implies an important reduction of the observer variables, and consequently allows an important gain of computation time. Variable calculated by the model and the corresponding estimated ones given by the observer will be compared on the next section. For case 1 (unseeded solution), the matrix  $A$  in equation (12) is replaced by :

$$A_r = \begin{pmatrix} \frac{1}{rat} & -\frac{1}{rat} & 0 & \dots & 0 \\ \frac{1}{2rat} & 0 & -\frac{1}{2rat} & \ddots & \vdots \\ 0 & \ddots & \ddots & \ddots & 0 \\ \vdots & \ddots & \frac{1}{2rat} & 0 & -\frac{1}{2rat} \\ 0 & \dots & 0 & 0 & 0 \end{pmatrix},$$

with :

$$rat = \frac{N}{N_{observer}}$$

In other words:

$$A_r = \frac{A}{rat}$$

In order to apply the previous results concerning the observer, we consider the following change of variables :

$$\alpha_1(t) = \frac{\alpha(t)}{rat} = \frac{G(t)}{\Delta x rat} \quad (23)$$

The new system is given by :

$$\begin{cases} \dot{n}_{x_{observer}}(t) = \alpha_1(t)A n_{x_{observer}}(t) \\ y(t) = C_r n_{x_{observer}}(t) \end{cases} \quad (24)$$

with:

$$C_r = (1 \ 0 \ \dots \ \dots \ \dots \ 0), C_r \in \mathbb{R}^{N_{observer}}.$$

The structure remains unchanged. As we mentioned above, the aim of this reduction is to reduce the number of the different equations of the matrix  $S$ . The new number of  $S$  equations is  $(1 + N_{observer}) \frac{(N_{observer}-1)}{2}$ .

#### V. SIMULATION RESULTS

The simulation results concerns the first case (unseeded solution). Where states number reduction and also distribution estimation are showed. this simulation was performed using a concentration measurement with added noise of about ( $\pm 2\%$ ), a reasonable choice because of the range of concentration values  $(0 - 1500) \frac{mol}{m^3}$ . The solubility curve ( $C_{sat}$  and

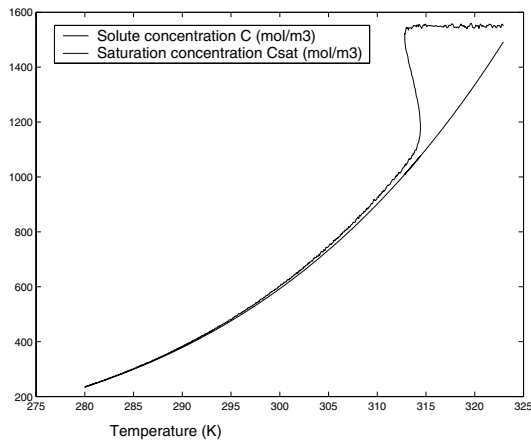


Fig. 1. solubility and solute concentration versus temperature

the solute concentration profile during the crystallization are presented in the Fig. 1.

When the supersaturation involved by the temperature decrease becomes enough important, nuclei which are produced by the primary nucleation grow. Temperature increases in this time interval because the crystal growth is exothermic. The secondary nucleation also starts because of crystal presence. A simulation problem due to noise appears when the supersaturation becomes small. In fact, some noisy solute measurements are lower than the concentration at saturation ( $C_{sat}$ ). This error introduces a fundamental change of the model (from positive to negative growth). Knowing the process behavior, this error is set equal to zero.

Since the production of nuclei occurs essentially at the beginning of the crystallization, the Fig. 2 shows this production during an adequate interval. The generation of nuclei in the remaining time interval is less important. Sampling period  $T_s$  is equal to 3 seconds. In continuous observer, it is

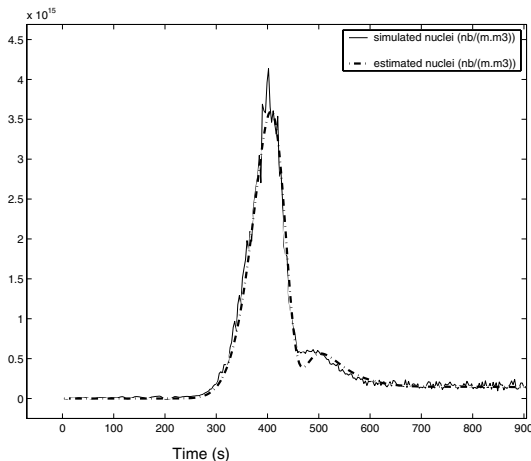


Fig. 2. nuclei

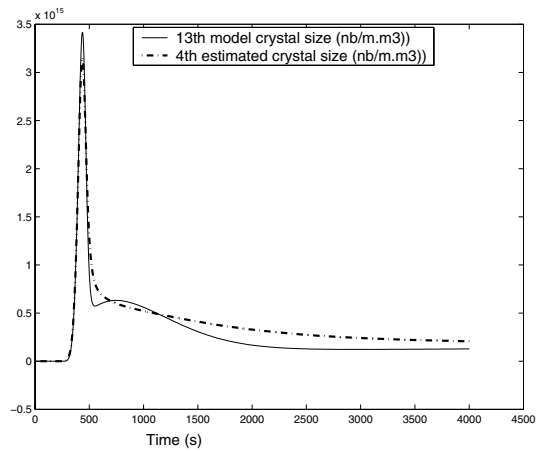


Fig. 3. 13th model crystal size and equivalent 4th observer crystal size

equal to 2 seconds.  $T_s$  is increased but remains small because of fast system dynamics.

The 4th and the 15th observer's variables (crystal sizes) and those corresponding of the model crystal are compared by simulation (to deduce the corresponding variable in the model, we apply the simple expression :  $Variable_{model} = 1 + (V_{observer} - 1)N_{reduction}$ ), in our case  $N_{observer}$  is equal to 4. As an example, the corresponding model variable for the 15th observer variable is :  $Variable_{model} = 1 + (15 - 1)4 = 1 + 56 = 57th$ . The choice of this variables as an examples was arbitrary, the aim is to show the convergence for the small and the big crystal sizes. Their evolutions with time are presented in the Fig. 3 and the Fig. 4. It is shown that these two crystal sizes are estimated with an acceptable precision. The overall precision of the observer is showed by the Fig. 5 which represents number mean size. This value depends on the different moments which are computed from all of the CSD.

The convergence time of the observer concerning the number mean size is very small. The model CSD and its

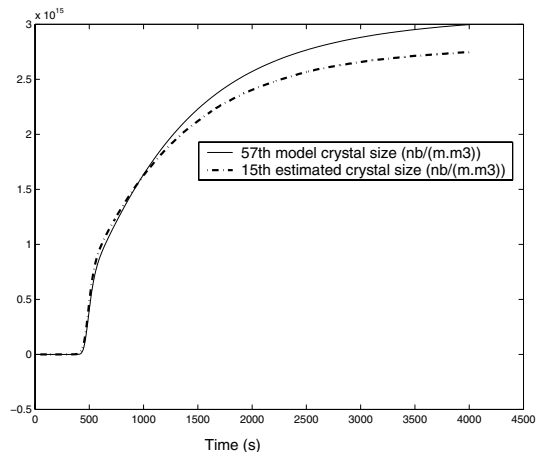


Fig. 4. 57th model crystal size and equivalent 15th observer crystal size

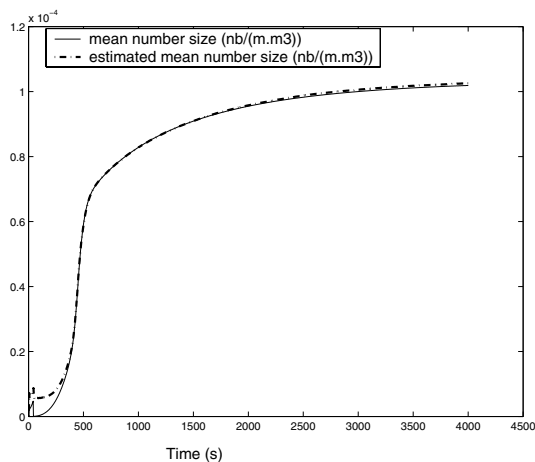


Fig. 5. number mean size

estimation are respectively plotted on 3D graphs (Fig. 6 and Fig. 7). An error estimation can be seen, it is justified by the huge number of estimated variable.

## VI. CONCLUSION

In spite of the great number of variables (crystal sizes) to be estimated, and the single output measurement (nuclei number), continuous-discrete observer allowed the estimation of the CSD with acceptable accuracy. The reduction of the model's order involved a lower number of variables to integrate, and then a smaller computation time. The greater sampling period we used in the case 1 (unseeded solution) and case 2 (seeded solution) are respectively 3 and 8 seconds, this limitation is due to high dynamics behavior. Such work allows the supervision and quality control of the final products, it can be used also to perform control law using the different moments calculated, the adequate observer for this task must be an implicit observer. As prospect, it is conceivable to use this software sensor in an industrial scale for batch operation. The next step will be dedicated to extent it to continuous crystallization process, for which some transient states are recurrent [14].

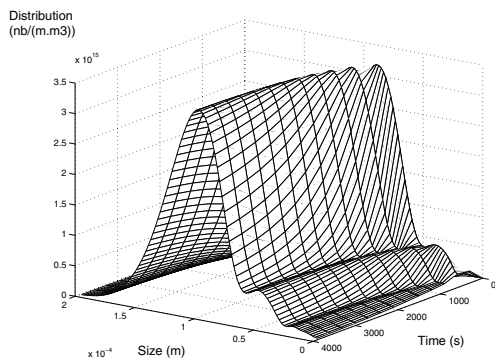


Fig. 6. model CSD

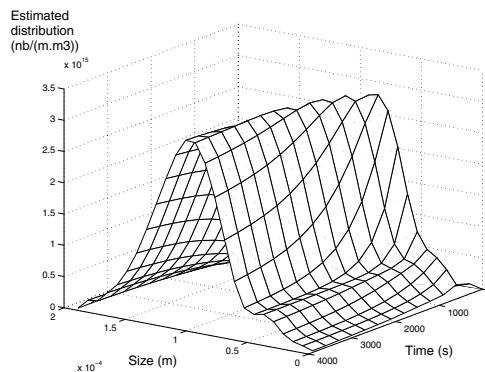


Fig. 7. Observer CSD

## REFERENCES

- [1] H. Kleizen, A. de Putter, M. van der beek, and S. Huynink, "Particle concentration, size and turbidity," *Filtration and Separation*, vol. 32, pp. 897–901, 1995.
- [2] O. Monnier, G. Fevotte, C. Hoff, and J. Klein, "Model identification of batch cooling crystallisation through calorimetry and image analysis," *Chemical engineering Science*, vol. 52, no. 7, pp. 1125–1139, 1997.
- [3] A. Jones, "Optimal operation of a batch cooling crystallizer," *Chemical engineering Science*, vol. 29, pp. 1075–1087, 1974.
- [4] S. Rohani, "Self-tuning control of crystal size distribution in a cooling batch crystallizer," *Chemical engineering Science*, vol. 45, no. 12, pp. 3457–3466, 1990.
- [5] K. Zoltan and D. Richard, "Open loop and closed-loop robust optimal control of batch processes using distributional and worst case analysis," *Journal of process control*, pp. 411–422, 2004.
- [6] A. Randolph and M. Larson, *Theory of particulate processes : analysis and techniques of continuous crystallization, second edition*. Academic edition, 1988.
- [7] S. Kumar and D. Ramkrishna, "On the solution of population balance equations by discretization-iii.nucleation, growth and aggregation of particles," *Chemical engineering Science*, vol. 52, pp. 4659–4679, 1997.
- [8] Y. Liu and I. Cameron, "A new wavelet-based adaptive method for solving population balance equations," *powder technology*, pp. 181–188, 2003.
- [9] J. Villadsen and W. Stewart, "Solution of boundary-value problems byorthogonal colocation," *Chemical engineering Science*, vol. 22, pp. 1483–1501, 1966.
- [10] M. Farza, H. Hammouri, S. Othman, and K. Busawon, "Nonlinear observer for parameter estimation in bioprocesses," *Chemical engineering Science*, vol. 52, no. 23, pp. 4251–4267, 1997.
- [11] J. Gauthier, H. Hammouri, and S. Othman, "A simple observer for non linear systems application to bioreactors," *IEEE Trans. Automat. Control*, vol. 37, pp. 875–880, 1992.
- [12] C.-D. Astorga, N. Othman, S. Othman, H. Hammouri, and T.-A. McKenna, "Nonlinear continuous-discrete observer : application to emulsien polymerization reactors," *Control engeneering practice*, vol. 10, pp. 3–13, 2002.
- [13] F. Deza, E. Busvelle, J. Gauthier, and D. Racotopara, "High gain estimation for non linear systems," *Systems and Control letters*, vol. 18, pp. 295–299, 1992.
- [14] F. Puel, P. Marshal, and J. Klein, "Habit transient analysis in industrial crystallization using two dimensional crystal sizing technique," *Trans IChemE*, vol. 75, pp. 173–205, 1997.