Parameter Identification, Reliability & Model Discrimination of Population Balance Models for Crystallization

EPSRC Centre for Innovative Manufacturing in Continuous Manufacturing and Crystallisation

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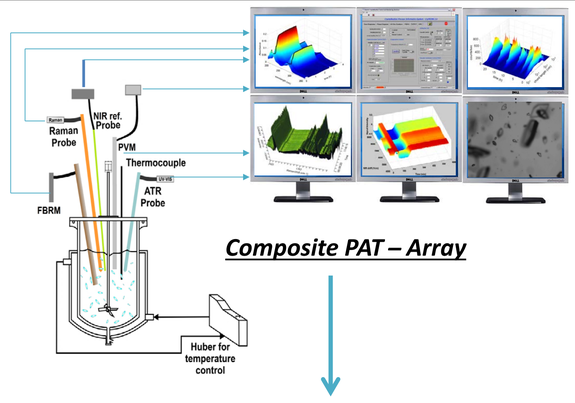
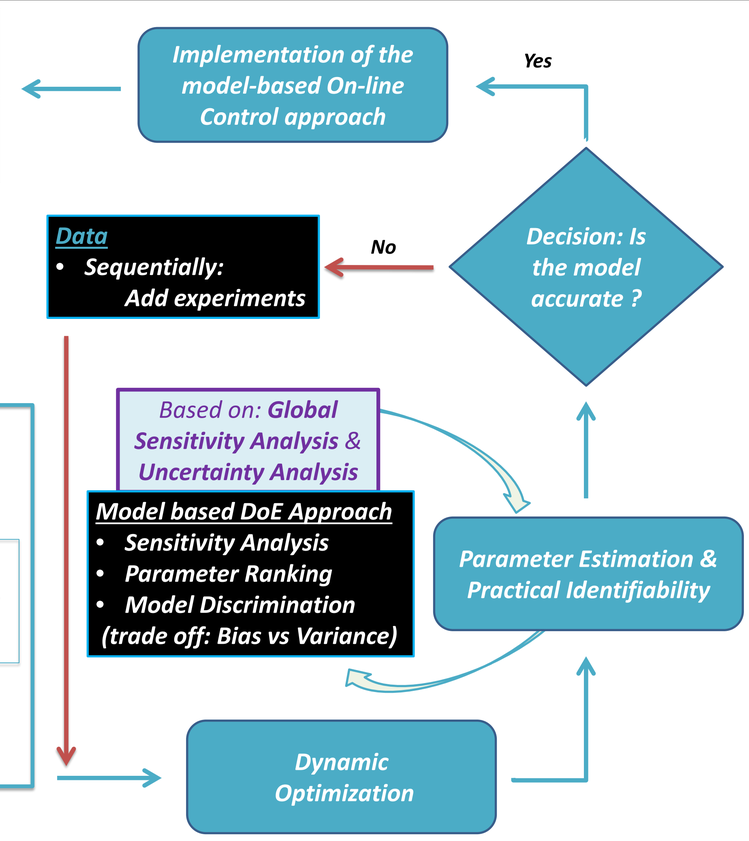
Overview

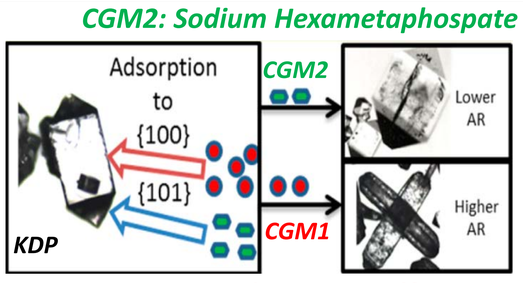
- *Population Balance Modelling (PBM) framework has been accepted as the most fundamental approach for dynamic modelling of numerous particulate processes.*
- ***The functional form of the model (PBM) is well – established but kinetic parameters need to be estimated from experimental data [1].***
- ***The PBM model may contain more parameters that can be accurately identified from the available experimental data (e.g. limited experimental data, correlation between parameters or between their effects with respect to the outputs, structure of the mathematical model) [2].***
- ***This challenge can be solved by identifying the subset of most influential parameters and estimating their values accurately from the measurements, while the parameters with the least estimability potential are fixed at nominal values [2].***
- *This approach has been applied to a multi-dimensional, multi-impurity adsorption (MIAM) PBM model that accounts for the combined effects of crystal growth modifiers (CGM) on the crystal size and shape distribution (CSSD) of needle-like crystals **[1]**.*





MIAM PBM Model

CGM2: Sodium Hexametaphospate



CGM1: Aluminum Sulfate

PBM for Dynamic evolution of the crystal shape distribution

Characteristic lengths $x=\{x_1,x_2\}$; ***Distribution function*** n; ***Nucleation rate*** B_p ; ***Growth rates*** G_i

$$\frac{\partial n(t,x)}{\partial t}+\frac{\partial[G_1\,n(t,x)]}{\partial x_1}+\frac{\partial[G_2 n(t,x)]}{\partial x_2}=B_p\,\delta(x_1-x_{1,0})\,\delta(x_2-x_{2,0})$$

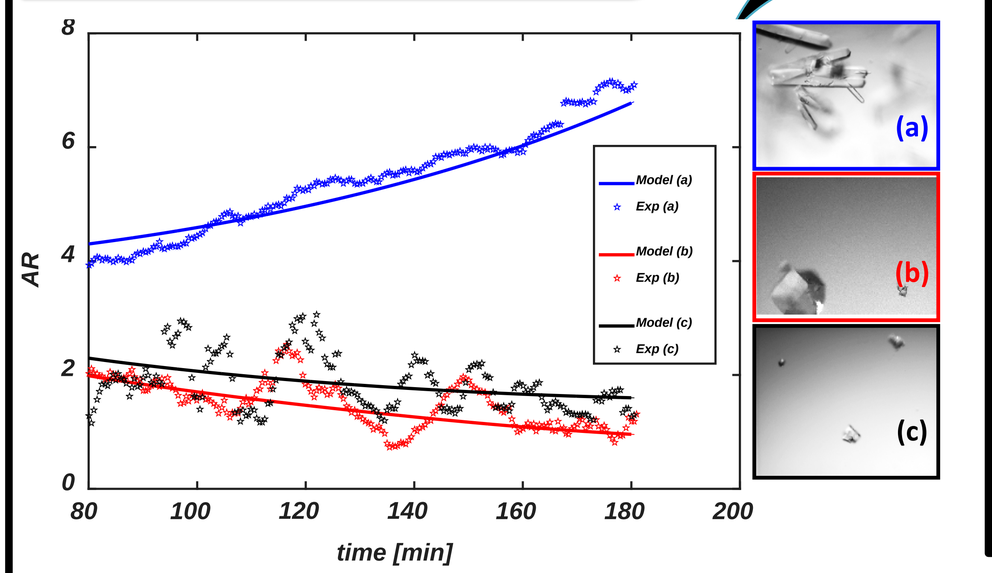
Parameters:

$\theta_{G1}=[k_{CGM1,ads,0}, k_{CGM1,des,0}, \beta_1, G_{min,1}, k_{CGM1,m,0}, k_{CGM1,e}, \Delta G_{min,ads}, \Delta G_{min,ads}]$;

$\theta_{G2}=[k_{CGM2,ads,0}, k_{CGM2,des,0}, \beta_2, G_{min,2}, k_{CGM2,m,0}, k_{CGM2,e}, \Delta G_{min,ads}, \Delta G_{min,ads}]$;

$\theta_{B_p}=[k_{p,0}, E_p, k_e]$;

Results



LSA	GSA
15	14
10	8
14	16
16	15
8	3
9	6
11	2
12	7
13	1
4	13
5	11
3	10
6	9
7	5
1	4
2	12

***Figure 2**: Comparison between the experimental and simulated mean aspect ratio evolution for the data used in identification of kinetics.*

Local Sensitivity Analysis (LSA)

$$\frac{\partial y(t)}{\partial \theta_j}=[S(y_1;\theta_j),S(y_2;\theta_j),\dots,S(y_n;\theta_j)]^T$$

$$S_{ij}=\begin{pmatrix}\frac{\partial y_{11}}{\partial \theta_1} & \cdots & \frac{\partial y_{1d}}{\partial \theta_p}\\ \vdots & \ddots & \vdots\\ \frac{\partial y_{1n}}{\partial \theta_1} & \cdots & \frac{\partial y_{dn}}{\partial \theta_p}\end{pmatrix}$$

y_n: Outputs
θ_j: Parameters
S_{ij}: Sensitivity Matrix

Sensitivity Analysis

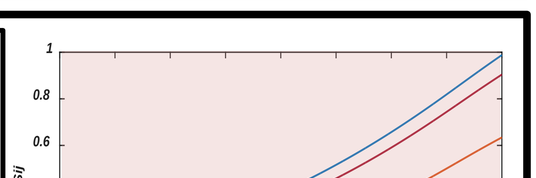
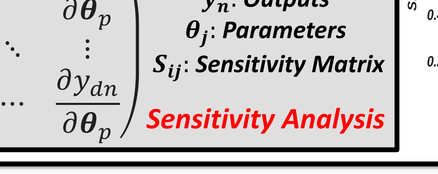


***Figure 1**: Relative Sensitivity trajectories of 16 Parameters for the MIAM model.*

Modified Gram Schmidt Orthogonalization algorithm



Parameter Ranking

- *Approach to determine **how sensitive a system is with respect to the change of parameters.***
- ***Parameters that have high impact** on the system variables **can be identified.***

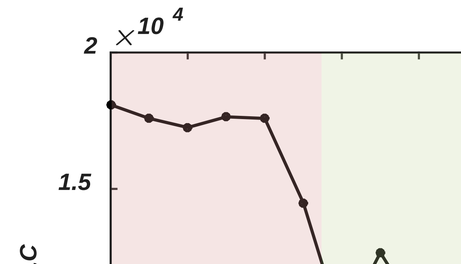


***Figure 3**: Number of selected parameters vs cut – off value criterion.*

- *Each number corresponds to one of the parameters (e.g. number 1 corresponds to the adsorption rate constant for the CGM1).*
- *Optimization: **Minimizing the Maximum Likelihood Criterion (MLC)**: Trade – off between Bias & Variance.*
- ***Calculate sequentially the MLC** by estimating certain sets of parameters while the others are fixed at initial guesses.*
- ***The 4 last parameters could be neglected** or fixed at certain nominal values without compromising the accuracy of the model.*

Global Sensitivity Analysis (GSA)

$$\frac{Var_x[E(y|\theta)]}{Var(y)}$$

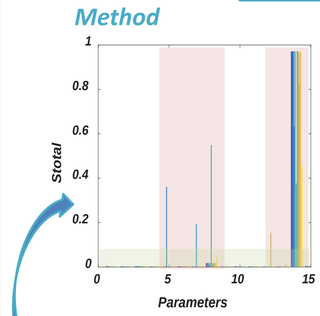
Variance – Based Method

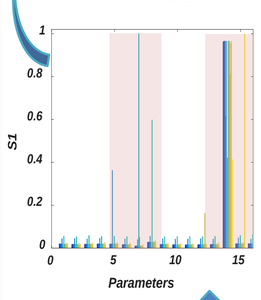
$$y(\theta_1,\dots,\theta_k)=f_0+\sum_{i=1}^{k}f_i(\theta_i)+\sum_{1\le i\le j\le k}f_{ij}(\theta_i,\theta_j)+\dots+f_{1,2,\dots,k}(\theta_1,\theta_2,\dots,\theta_k)$$

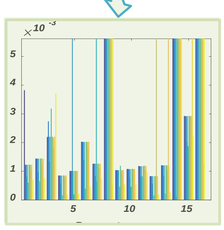
Decomposition of Variance

Global Sensitivity Analysis (Sobol Method)

Parameter Selection & Probability Distribution Allocation







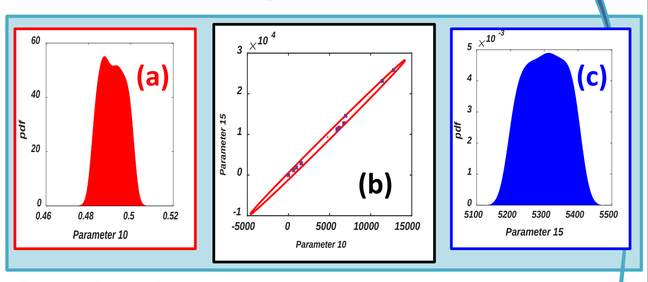


***Figure 4**: (a) Probability density function for parameter 10, (b) Confidence domain between parameter 10 & 15, (c)Probability density function of parameter 15.*

Parameter Sensitivity & Ranking

Model $y=f(\theta_1,\theta_2,\dots,\theta_n)$

GSA analysis is implemented to the MIAM PBM model.

Conclusions & Future Work

- ***Both local and global estimability & identifiability concepts has been applied to the MIAM PBM model** that accounts for the combined effects of different crystal growth modifiers on the crystal size and shape distribution (CSSD) of needle-like crystals.*
- ***Although noisy data were used, the most influential and the least correlated parameters can be identified, providing sufficiently accurate inputs for the dynamic evolution of the CSSD.***
- *Implementation of this workflow as an **on-line parameter identification approach**.*
- *Further development of the model discrimination (Cut-off value) strategy for GSA analysis.*
- ***Integrate the algorithm in a control design strategy for parameter tuning.***

References

[1] A., Borsos, A., Majumder, Z.K., Nagy, *Cryst. Growth. Des.* 2016, 16, 555-568.

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