DISTRIBUTIONAL UNCERTAINTY ANALYSIS OF A BATCH CRYSTALLIZATION PROCESS USING POWER SERIES AND POLYNOMIAL CHAOS EXPANSIONS

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Abstract: Computationally efficient approaches are presented that quantify the influence of parameter uncertainties upon the states and outputs of finite-time control trajectories for nonlinear systems. In the first approach, the worst-case values of the states and outputs due to model parameter uncertainties are computed as a function of time along the control trajectories. The approach uses an efficient contour mapping technique to provide an estimate of the distribution of the states and outputs as a function of time. To increase the estimation accuracy of the shape of the distribution, an approach that uses second order power series expansion in combination with Monte Carlo simulations is proposed. Another approach presented here is based on the approximate representation of the model via polynomial chaos expansion. A quantitative and qualitative assessment of the approaches is performed in comparison to the Monte Carlo simulation technique that uses the nonlinear model. It is shown that the power series and polynomial chaos expansion based approaches require a significantly lower computational burden compared to Monte Carlo approaches, while give good approximation of the shape of the distribution. The techniques are applied to the crystallization of an inorganic chemical with uncertainties in the nucleation and growth parameters. *Copyright © 2003 IFAC*

Keywords: probabilistic analysis, distributional robustness analysis, worst-case analysis, crystallization, optimal control.

1. INTRODUCTION

Comprehensive uncertainty analysis of mechanistic models is crucial especially when these models are used in the optimal control of processes, which normally occurs close to safety and performance constraints. The model-based computation of optimal control policies for batch and semibatch processes is of increasing interest due to industrial interest in improving productivity (Barrera and Evans, 1989; Rippin 1983). However, uncertainty almost always exist in chemical systems in the observed data, in the model parameters, and implemented inputs, and its disregard may easily lead the loss of the benefits of using optimal control (Ma *et al.*, 1999). This motivates the development of techniques to quantify the influence of parameter uncertainties on the process states and outputs. Quantitative estimates obtained from robustness analysis can be used to decide whether more laboratory experiments are needed to provide better parameter estimates (Ma and Braatz, 1999; Miller and Rawlings, 1994). The traditional uncertainty analysis consists of the characterization of uncertainty in model parameters or inputs based on their probability density functions (*pdf*) and then propagating these *pdf*s through the model equations to obtain the *pdf*s of selected model outputs. The propagation of uncertainties via

traditional Monte Carlo methods, based on standard or Latin Hypercube sampling may require performing a large number of simulations, which can be prohibitive in most cases, especially if the propagation has to be performed in real-time. Therefore there is a need to study computationally efficient alternative techniques for uncertainty propagation.

The paper focuses on computationally efficient methods for propagating uncertainty in parameters to the states and outputs of generic batch processes. Two categories of approaches are corroborated. The first is based on first and second order power series approaches (Nagy and Braatz, 2003). The first order approach is based on the analytical computation of the worst-case values of the states and outputs due to the effects of model parameter uncertainties, and then using a contour mapping approach to compute the distribution. The second approach uses polynomial chaos expansion as a functional approximation of the mathematical model (Isukapalli, 1999; Isukapalli *et al.*, 1998; Pan *et al.*, 1997, 1998). Both approaches are suitable for studying the uncertainty propagation in open-loop or closed-loop systems. The techniques are compared with Monte Carlo simulations and applied to compute the distributions for the states and outputs for the batch crystallization of an inorganic chemical subject to uncertainties in the nucleation and growth kinetics.

2. DISTRIBUTIONAL UNCERTAINITY ANALYSIS

2.1 Uncertainty description

In the following we consider the class of finite time (batch) processes. Assume the model is described by the generic ODE vector equation:

$$\dot{x}(t) = f(x(t), u(t); \theta) \qquad (1)$$

with $x \in \mathbb{R}^{n_x}$ the state vector, $x \in \mathbb{R}^{n_x}$, $u \in \mathbb{R}^{n_u}$ vector of control inputs, and $\theta \in \mathbb{R}^{n_\theta}$ uncertain parameter vector, and f a vector function that is continuous wit respect to its elements. Characterizing the uncertainties enhances the value of the model by allowing the quantification of the accuracy of its predictions. This information can be used to assess whether the model is adequate for its intended purpose (e.g., for optimal control design) or whether more experimental data are needed to further refine the model.

Define $\hat{\theta}$ as the nominal model parameter vector of dimension $(n \times 1)$, and $\delta\theta$ as the perturbation about $\hat{\theta}$. Then, the model parameter vector for the real system is:

$$\theta = \hat{\theta} + \delta\theta . \qquad (2)$$

We assume that the uncertainty in the parameter θ is characterized by the generalized ball

$$\mathcal{P} \triangleq \{ \theta \in \mathbb{R}^{n_\theta} \ \big| \| \theta - \hat{\theta} \| \leq 1 \} \qquad (3)$$

defined by using appropriate norm $|| \cdot ||$ in $\mathbb{R}^{n_\theta}$. One generic approach is to use a scaled Hölder p-norm ($1 \leq p \leq \infty$), given by $\| \theta \| = \| \mathbf{W}_\theta^{-1}\theta \|_p$, with the invertible weighting matrix $\mathbf{W}_\theta \in \mathbb{R}^{n_\theta \times n_\theta}$. This generalized description of the uncertainty set includes the case of a confidence hyper-ellipsoid $\varepsilon_\theta = \{\theta : (\theta - \hat{\theta})^T \mathbf{V}_\theta^{-1}(\theta - \hat{\theta}) \leq r^2(\alpha)\}$, (Beck and Arnold, 1985), for Gaussian random variable vector θ with expected value $\mathcal{E}(\theta) = \hat{\theta}$, the $(n_\theta \times n_\theta)$ positive definite variance-covariance matrix $\mathbf{V}_\theta$, and the scalar r which is the chi-square distribution function with n_θ degrees of freedom ($\chi^2_{n_\theta}(\alpha)$), for a chosen confidence level α:

$$\mathcal{P}_{\text{ellipsoid}}(\alpha) \triangleq \{ \theta \in \mathbb{R}^{n_\theta} \ \big| \| (1/r(\alpha))\mathbf{V}_\theta^{-1/2}(\theta - \hat{\theta}) \|_2 \leq 1 \} \quad (4)$$

The generalized ball described by (2) also includes the case of known lower and upper bounds θ_l, θ_u of the uncertain parameters, leading to a general uncertainty hyper-box:

$$\begin{aligned} \mathcal{P}_{\text{box}} &\triangleq \{\theta \in \mathbb{R}^{n_\theta} \,|\, \theta_l \leq \theta \leq \theta_u \} \\ &= \{\theta \in \mathbb{R}^{n_\theta} \ \big| \ ||\text{diag}(\frac{\theta_u - \theta_l}{2})(\theta - \frac{\theta_u + \theta_l}{2})||_\infty \leq 1 \} \end{aligned} \qquad (5)$$

2.2. Series expansion approaches for Worst-case and Distributional Robustness Analysis

Define $\hat{\psi}$ as the performance (describing an end point property) for the nominal model parameters $\hat{\theta}$, ψ as its value for the perturbed model parameter vector p, and the difference $\delta\psi = \psi - \hat{\psi}$. The worst-case robustness approach (Ma *et al.*, 1999; Nagy and Braatz, 2003) writes $\delta\psi$ as a power series in $\delta\theta$:

$$\delta\psi = L\delta\theta + \frac{1}{2}\delta\theta^T \mathbf{M}\delta\theta + \ldots , \qquad (6)$$

where the jacobian $L \in \mathbb{R}^{n_\theta}$, and hessian $\mathbf{M} \in \mathbb{R}^{n_\theta \times n_\theta}$ are:

$$L(t) = \left(\frac{\partial \psi(t)}{\partial \theta} \right)_{\hat{\theta}} , \qquad (7)$$

$$\mathbf{M}(t) = \left(\frac{\partial^2 \psi(t)}{\partial \theta^2} \right)_{\hat{\theta}} . \qquad (8)$$

The elements of the time-varying sensitivity vector $L(t)$ and matrix $\mathbf{M}(t)$ can be computed using finite differences or by integrating the model's differential-algebraic equations augmented with an additional set of differential equations known as sensitivity equations (Caracotsios and Stewart, 1985):

$$\dot{L} = \mathbf{J}_x L + \mathbf{J}_\theta \qquad (9)$$

with the matrixes $\mathbf{J}_x = df/dx \in \mathbb{R}^{n_x \times n_x}$ and $\mathbf{J}_\theta = df/d\theta \in \mathbb{R}^{n_x \times n_\theta}$.

When a first-order series expansion is used, analytical expressions of the worst-case deviation in

the performance index ($\delta\psi_{w.c.}$) can be computed and the analysis can be performed with low computational cost (Matthews, 1997). In the case of an ellipsoidal uncertainty description the worst-case deviation is defined by

$$\delta\psi_{w.c.}(t) = \max_{\|\mathbf{W}_\theta \delta\theta\|_2 \leq 1} |L(t)\delta\theta| . \qquad (10)$$

The analytical solution of this optimization problem is given by:

$$\delta\psi_{w.c.}(t) = (r(\alpha)L(t)\mathbf{V}_\theta L^T(t))^{1/2} , \qquad (11)$$

$$\delta\theta_{w.c.}(t) = \frac{(r(\alpha))^{1/2}}{(L(t)\mathbf{V}_\theta L^T(t))^{1/2}} \mathbf{V}_\theta L^T(t) . \qquad (12)$$

A probability density function (PDF) for the model parameters is needed to compute the PDF of the performance index. More than 90% of the available algorithms to estimate parameters from experimental data (Beck and Arnold, 1977) produce a multivariate normal distribution:

$$f_{p.d.}(\theta) = \frac{1}{(2\pi)^{n_\theta/2} \det(\mathbf{V}_\theta)^{1/2}} \exp\left(-\frac{1}{2}[(\theta-\hat{\theta})^T \mathbf{V}_\theta^{-1}(\theta-\hat{\theta})]\right). \quad (13)$$

When a first-order series expansion is used to relate $\delta\psi$ and $\delta\theta$, then the estimated PDF of ψ is

$$f_{p.d.}(\psi) = \frac{1}{V_\psi^{1/2}\sqrt{2\pi}} \exp\left(-(\psi-\hat{\psi})^2 / (2V_\psi)\right) \quad (14)$$

where the variance of ψ is

$$V_\psi = L\mathbf{V}_\theta L^T . \qquad (15)$$

The distribution is a function of time since the nominal value for ψ and the vector of sensitivities L is a function of time.

2.3 Propagation of probability distribution

When the first order power series expansion is used the probability density function (*p.d.f.*) can be estimated very efficiently using a contour mapping approach, which instead of mapping the whole parameter space to the output space by performing exhaustive Monte Carlo simulations, maps only the contours of the uncertainty hyperellipsoid obtained for different α levels as shown on Figure 1. The mapping is performed via the worst-case analysis techniques by obtaining the worst-case $\delta\psi_{w.c}$ for different α-levels. Note that with this approach the mapping of the α levels is performed from the n_θ dimensional space characterized by a chi-square distribution of n_θ degrees of freedom ($\chi^2_{n_\theta}(\alpha)$) to an n_ψ dimensional space in which the same α-levels are characterized by a chi-square distribution but with n_ψ degrees of freedom ($\chi^2_{n_\psi}(\alpha)$), with usually $n_\psi=1$. Hence the probability mapping between the two spaces characterized by different degrees of freedom can be captured by multiplying the obtained worst-case deviations ($\delta\psi_{w.c.}$) with the ratio $(\chi^2_{n_\psi}(\alpha)/\chi^2_{n_\theta}(\alpha))^{1/2}$.
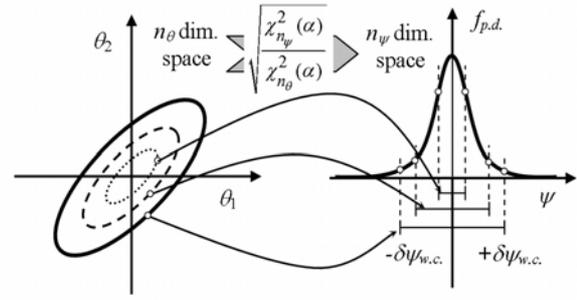
Fig.1. Distributional robustness approach based on a contour mapping technique.

Remark: Since in this approach the sampling is always performed over one parameter (the confidence level α) a significantly smaller number of Monte Carlo simulations are required than using classical sampling of the usually larger dimensional parameter space. Additionally, while the classical sampling procedure requires large number of samples to capture accurately the distribution for values with low probability, in the contour mapping approach samples span over all confidence levels and generate enough data to give insight into the tails of the *pdf* in a similar way as in the Latin Hypercube sampling method.

For increased accuracy of the estimated shape of the *pdf*, the second order series expansion can be used in a classical or Latin Hypercub type Monte Carlo simulation. This Monte Carlo approach is computationally much more efficient than applying the Monte Carlo method to the original nonlinear model. The computational burden of the second order approximate Monte Carlo approach is higher than in the case of the contour mapping approach but provides better estimate of the shape of the output distribution.

2.4 Uncertainty Analysis Using Polynomial Chaos Expansions

An alternative to power series is to use polynomial chaos expansions (PCEs). The PCE (Wiener, 1938) describes the model output ψ as an expansion of multidimensional Hermite polynomial functions of the uncertain parameters θ in the form:

$$\psi = \underbrace{a_0\Gamma_0}_{\text{constant}} + \underbrace{\sum_{i_1=1}^{n_\theta} a_{i_1}\Gamma_1(\theta_{i_1})}_{\text{first order terms}} + \underbrace{\sum_{i_1=1}^{n_\theta}\sum_{i_2=1}^{i_1} a_{i_1 i_2}\Gamma_2(\theta_{i_1},\theta_{i_2})}_{\text{second order terms}}$$

$$+ \underbrace{\sum_{i_1=1}^{n_\theta}\sum_{i_2=1}^{i_1}\sum_{i_3=1}^{i_2} a_{i_1 i_2 i_3}\Gamma_3(\theta_{i_1},\theta_{i_2},\theta_{i_3})}_{\text{third order terms}} + \ldots \qquad (16)$$

where the $\Gamma_a(\theta_{i_1},\theta_{i_2},\ldots,\theta_{i_a})$ are polynomials, n_θ is the number of parameters, and the $a_0, a_1, a_2, \ldots, a_{11}, a_{12}, \ldots$ are constants in $\mathbb{R}$. Polynomial chaos terms of different order are orthogonal to each other as are polynomial chaos terms of the same order but with a different argument list. The orthogonal polynomials are derived from the

probability distribution of the parameters. In PCE any form of polynomial could be used but the properties of orthogonal polynomials make the uncertainty analysis more efficient. The number of coefficients in the PCE depends on the number of uncertain parameters and the order of expansion. In principle there are two main methods for computing the coefficients of the PCE, (i) the probabilistic collocation method (PCM) (Pan *et al.*, 1997, 1998), and (ii) the regression method with improved sampling (RMIS) (Isukapalli *et al.*, 1998). In both methods the coefficients are calculated from the model at a set of sample points using regression based technique.

3. APLICATION TO A BATCH CRYSTALLIZATION PROCESS

Crystallization from solution is an industrially important unit operation due to its ability to provide high purity separation. The control of the crystal size distribution (CSD) can be critically important for efficient downstream operations (such as filtration or drying) and product quality (e.g., bioavailability, tablet stability, dissolution rate). Most studies on the optimal control of batch crystallizers focus on computing the temperature profile that optimizes some property of the CSD. The problem of computing the optimal temperature profile can be formulated as a nonlinear optimization problem, which is then solved using general-purpose optimization algorithms. A convenient way to describe the temperature trajectory is to discretize the batch time and consider the temperatures at every discrete time k as the optimization variables. In this case the optimal control problem can be written in the following form:

$$\underset{T(k)}{\text{optimize }} J \qquad (17)$$

subject to:

$$
\begin{bmatrix}
\dot{\mu}_0 \\
\dot{\mu}_1 \\
\dot{\mu}_2 \\
\dot{\mu}_3 \\
\dot{\mu}_4 \\
\dot{C} \\
\dot{\mu}_{seed,1} \\
\dot{\mu}_{seed,2} \\
\dot{\mu}_{seed,3}
\end{bmatrix}
=
\begin{bmatrix}
B \\
G\mu_0 + Br_0 \\
2G\mu_1 + Br_0^2 \\
3G\mu_2 + Br_0^3 \\
4G\mu_3 + Br_0^4 \\
-\rho_c k_v (3G\mu_2 + Br_0^3) \\
G\mu_{seed,0} \\
2G\mu_{seed,1} \\
3G\mu_{seed,2}
\end{bmatrix}
\qquad (18)
$$

$$T_{\min}(k) \le T(k) \le T_{\max}(k),$$

$$R_{\min}(k) \le \frac{dT(k)}{dt} \le R_{\max}(k), \qquad (19)$$

$$C_{final} \le C_{final,\max},$$

where the objective function J is a function of the states, and usually it is a representative property of the final CSD. The equality constraints (18) represent the model equations, with initial conditions given in (Chung *et al.*, 1999), where μ_i is the ith moment ($i = 0, \ldots, 4$) of the total crystal phase (resulted from

growth from seed and nucleation) and $\mu_{seed,j}$ is the jth moment ($j = 0, \ldots, 3$) corresponding to the crystals grown from seed, C is the solute concentration, T is the temperature, r_0 is the crystal size at nucleation, k_v is the volumetric shape factor, and ρ_c is the density of the crystal. The rate of crystal growth (G) and the nucleation rate (B), respectively, are given by (Nyvlt, *et al.*, 1985):

$$G = k_g S^g, \qquad (20)$$

$$B = k_b S^b \mu_3, \qquad (21)$$

where $S = (C-C_{sat})/C_{sat}$ is the relative supersaturation, and $C_{sat} = C_{sat}(T)$ is the saturation concentration. The model parameter vector consists of the kinetic parameters of growth and nucleation:

$$\theta^T = [g, k_g, b, k_b], \qquad (22)$$

with nominal values (Rawlings *et al.*, 1993):

$$\hat{\theta}^T = [1.31, 8.79, 1.84, 17.38], \qquad (23)$$

with the uncertainty description of the form (4) characterized by the covariance matrix (Miller and Rawlings, 1994):

$$
V_\theta^{-1} =
\begin{bmatrix}
102873 & -21960 & -7509 & 1445 \\
-21960 & 4714 & 1809 & -354 \\
-7509 & 1809 & 24225 & -5198 \\
1445 & -354 & -5198 & 1116
\end{bmatrix}.
\qquad (24)
$$

In the inequality constraints (19) T_{min}, T_{max}, R_{min}, and R_{max} are the minimum and maximum temperatures and temperature ramp rates, respectively, during the batch. The first two inequality constraints ensure that the temperature profile stays within the operating range of the crystallizer. The last inequality constraint ensures that the solute concentration at the end of the batch C_{final} is smaller than a certain maximum value $C_{final,max}$ set by the minimum yield required by economic considerations.

The crystal size distribution (CSD) parameters of interest are: nucleation to seed mass ratio ($J_{n.s.r.}$), coefficient of variations ($J_{c.v.}$), and weight mean size of the crystals ($J_{w.m.s.}$), given by the following expressions:

$$J_{n.s.r.} = (\mu_3 - \mu_{seed,3}) / \mu_{seed,3} \qquad (23)$$

$$J_{c.v.} = (\mu_2\mu_0 / (\mu_1)^2 - 1)^{1/2} \qquad (24)$$

$$J_{w.m.s.} = \mu_4 / \mu_3 \qquad (25)$$

The optimal temperature trajectory that minimizes the nucleation mass to seed mass ratio at the end of the batch was computed setting $J = J_{n.s.r.}$, and solving the optimal control problem (17)-(19) for the nominal parameter $\hat{\theta}$.

The aforementioned uncertainty analysis approaches are used to assess the effect of parameter uncertainty on the nominal control performance. The distributional uncertainty analysis approaches based on power series approximation and polynomial chaos expansion, respectively, were evaluated in comparison to Monte Carlo (MC) simulations. A
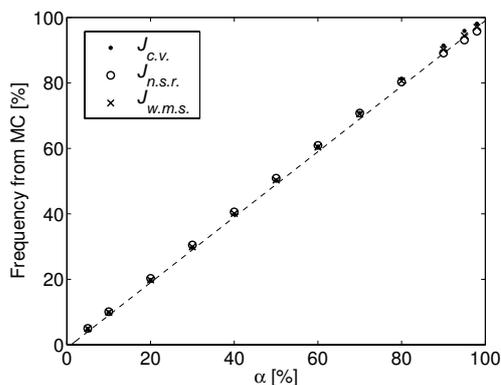
Fig. 2. Comparison between the first-order power series analysis and Monte Carlo simulations. The dashed line corresponds to the ideal fit.

Method	Computational time
Monte Carlo with dynamic model (80,000 data)	8 hours
First order approach (50 α levels, every 10 min)	1 second
Monte Carlo with 2nd order series expansion model (80,000 data)	4 minutes
Polynomial chaos expansion	2 seconds

number of 80,000 random parameter sets with mean $\hat{\theta}_i$ and covariance $\mathbf{V}_\theta$ were generated from the multivariate distribution using the Cholesky decomposition of the covariance matrix. With the random parameter vectors, Monte Carlo simulation was performed using the dynamic model of the process. Then, the frequency that the simulation output from the Monte Carlo simulation falls in the confidence interval obtained for a certain α level with the power series approach was computed for all states and outputs. The results obtained with the first order analysis approach for the output variables are presented in Figure 2. The accuracy of the first-order approach is very good with a slightly decreasing tendency for large α values. This can be explained by the cumulative effect of the truncation error due to the first-order approach (which increases when α increases) and the tail effect of the Monte Carlo simulation. Although there is a slight decrease in the accuracy of the first order approach for large α in the case of certain process outputs the first-order technique gives good result for practical purposes.

The first-order power series approach approximates the output distributions with a normal distribution. To obtain a more accurate representation of the output PDF, the uncertainties can be propagated through the nonlinear dynamic model via Monte Carlo simulations. To avoid the prohibitively high computational time required in this case, an alternative approach that performs Monte Carlo simulation using a higher order power series expansion or polynomial chaos expansion in place of the dynamic simulation model can be used. Using a second-order power series expansion in the Monte Carlo simulations gives an accurate approximation of the nonlinear distribution with a low computational cost (Table 1). PCE can be used instead of the power series expansion, resulting in similarly good computational efficiency. The advantages of the aforementioned uncertainty analysis approaches compared to classical sensitivity analysis consist in that they provide the variation of the whole distribution for all states and outputs along the entire batch.. Figure 3 shows the results obtained with the two computationally efficient approaches in comparison with the Monte Carlo simulation. In the simulation results presented here a second order PCE was used. Since there are four uncertain parameters the second order PCE requires the determination of 15 coefficients. The probabilistic collocation method was used as described in (Tatang *et al.* 1997, Webster *et al.*, 1996). The obtained PCE provide accurate estimation of the effects of parameter uncertainties at a computational burden similar to the first order power series approach. The power series and PCE based robustness analysis approaches can be used for the assessment and efficient synthesis of robust control approaches. Since the robustness analysis is a convex problem, it is significantly easier to solve it than the direct robust controller synthesis, which usually leads to nonconvex problem formulations. The robust controller synthesis is formulated as a classical minmax optimization or as a multiobjective optimization problem where one of the objective accounts for the nominal term and the other (usually a measure of the variance of the distribution) accounts for robustness. Both uncertainty analysis approaches can be used as an efficient tool of calculating the robustness term.

4. CONCLUSIONS

An overview of several computationally efficient distributional robustness analysis approaches are presented. The approaches are based on the approximate representation of the process model using first or second order power series or polynomial chaos expansions, and provide a qualitative and quantitative estimation of the effect of parameter uncertainties on the states and output variable along the batch time. The computational burden of the robustness analysis approaches is significantly reduced compared to the classical Monte Carlo approach based on the nonlinear model. The algorithms are assessed via a simulated batch cooling crystallization process.

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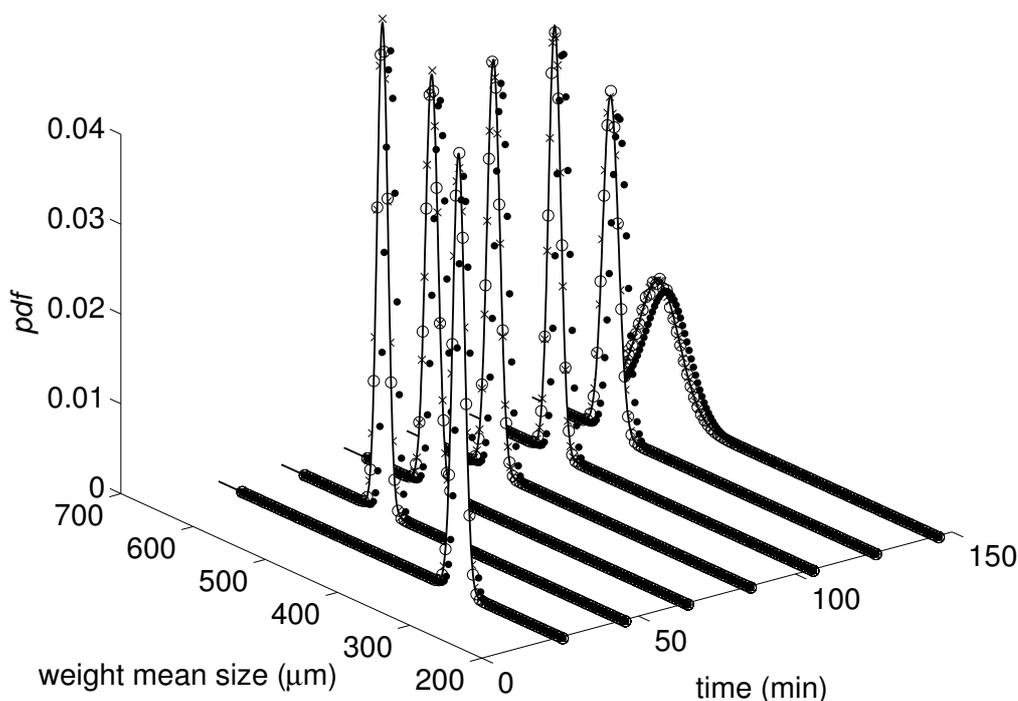
Fig. 3. Variation of the distribution of the weight mean size along the batch, computed using different uncertainty analysis approaches. Solid line is obtained from Monte Carlo simulations using 80,000 parameter sets and the nonlinear model; dots represent the results with first-order power series approach and contour mapping; circles correspond to the result from Monte Carlo simulations based on second order power series expansion; x-marks correspond to the results using the polynomial chaos expansion based approach.

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