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Distributional Uncertainty Analysis Using Polynomial Chaos Expansions

Zoltan K. Nagy, *Member, IEEE*, and Richard D. Braatz, *Fellow, IEEE*

Abstract—A computationally efficient approach is presented that quantifies the influence of parameter uncertainties on the states and outputs of finite-time control trajectories for nonlinear systems, based on the approximate representation of the model via polynomial chaos expansion. The approach is suitable for studying the uncertainty propagation in open-loop or closed-loop systems. A quantitative and qualitative assessment of the method is performed in comparison to the Monte Carlo simulation technique that uses the nonlinear model for uncertainty propagation. The polynomial chaos expansion-based approach is characterized by a significantly lower computational burden compared to Monte Carlo approaches, while providing a good approximation of the shape of the uncertainty distribution of the process outputs. The techniques are applied to the crystallization of an inorganic chemical with uncertainties in the nucleation and growth parameters.

I. INTRODUCTION

MANY real world problems contain uncertainty. Comprehensive uncertainty analysis of mechanistic models is crucially important, especially when these models are used in the optimal control of processes, which generally occurs close to safety and performance constraints. The model-based computation of optimal control policies of finite-time processes is of increasing interest due to industrial interest in improving productivity [1]. However, uncertainties in the observed data, in the model parameters, and implemented inputs, if not taken into account, may lead to the loss of the benefits of using optimal control [2]–[4]. This motivates the development of techniques to quantify the influence of parameter uncertainties on the process states and outputs [5], [6].

Stochastic uncertainty analysis generally consists of three main steps: (1) the characterization of uncertainty in model parameters or inputs based on their probability density functions (PDF), (2) propagating of these PDFs through the model equations to obtain the PDFs of selected model outputs, and (3) management of the resulted uncertainty (e.g. robust control design, new experimental design, etc.) [7], [8]. Uncertainty propagation methods available can be classified into three categories: (i) analytical methods [9], (ii) Monte Carlo simulation methods, and (iii) response surface methods [7], [8], [10]–[12]. Analytical methods are

the most efficient but can be applied for a very limited class of systems represented by simple models. Monte Carlo simulations in principle can be used to achieve an accurate propagation of the input uncertainties into a vector of output uncertainties through a physical model; however, for complex models the method can be computationally prohibitive, especially in real-time applications, such as in robust control. Another disadvantage of the classical Monte Carlo method is that it does not provide a manageable representation of the predicted process. Therefore there is a need to investigate computationally-efficient alternative techniques for uncertainty propagation, which also provide a simplified mathematical representation of the process. Several efficient uncertainty analysis approaches have been proposed based on power series expansions [2], [4]–[6] and have been applied to open-loop and closed-loop robust control design. An alternative approach, which has been intensively applied in the past decade, is based on stochastic response surface (SRS) approaches, which describe the performance function/model output as a sum of elementary functions (bases) of stochastic input parameters, representing the model as convergent expansions similar to power series.

The paper provides a tutorial introduction to a computationally efficient method for propagating parameter uncertainty to the states and outputs of generic batch processes. The approach belongs to the class of SRS approaches and uses polynomial chaos expansion (PCE) as a functional approximation of the mathematical model [7], [8], [10]–[12]. The approach is suitable for studying the uncertainty propagation in open-loop or closed-loop systems. The paper extends earlier work that focused on the uncertainty analysis of states and outputs at the end of the batch [13], by indicating how the efficient PCE-based approach can be used to analyze the dynamic evolution of the effects of uncertainties during the entire batch. After providing an overview of applications of PCE in the control and related fields, the technique is compared with Monte Carlo simulation based on the mechanistic model, 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.

II. POLYNOMIAL CHAOS EXPANSIONS

A. Review of Applications of PCE

The polynomial chaos expansion (PCE) was originally introduced by [14] for turbulence modeling and has started to gain large popularity only relatively recently. Since Ref.

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TABLE I
MOST COMMON DISTRIBUTIONS OF RANDOM PARAMETERS WITH
CORRESPONDING POLYNOMIAL BASES

Random variable	Polynomials
Beta	Jacobi
Gamma	Laguerre
Gaussian	Hermite
Uniform	Legendre

[15] implemented PCE in terms of Hermite polynomials for linear elastic problems, PCEs have become widely used in a large variety of fields including stochastic differential equations [16], computational fluid dynamics with applications to finite deformations [17], [18], or transport in porous media [19], as well as in environmental and acoustics applications [20]. Exploiting the fact the PCE provides the possibility to define the uncertain model as a deterministic model with an extended number of variables, several interesting applications have been proposed more recently. PCE has been applied for uncertainty analysis in electrical measurement [21], chemical processes [13], robot manipulators [23] and mechanical systems [24], and for parameter estimation [22], robust control design [25], stability analysis [26], [27], sensitivity analysis [28], and observer design [29], [30].

B. Uncertainty Analysis Using PCE

If the parameter uncertainties are described in terms of standard normal random variables, the polynomial chaos expansion (PCE) can describe the model output ψ as an expansion of multidimensional Hermite polynomial functions of the uncertain parameters θ . For other types of random variables, either different polynomial bases or an appropriate transformation can be used [15]. Table I summarizes the correspondence between the choice of polynomials and the type of distribution of the random variables.

Using the Hermite bases in the PCE, the output can be expressed in terms of the standard random normal variables $\{\theta_i\}$ using an expansion of order d :

$$\begin{aligned} \psi^{(d)} = & \underbrace{a_0^{(d)}\Gamma_0}_{\text{constant}} + \underbrace{\sum_{i_1=1}^{n_\theta} a_{i_1}^{(d)}\Gamma_1(\theta_{i_1})}_{\text{first-order terms}} + \underbrace{\sum_{i_1=1}^{n_\theta} \sum_{i_2=1}^{i_1} a_{i_1 i_2}^{(d)}\Gamma_2(\theta_{i_1}, \theta_{i_2})}_{\text{second-order terms}} \\ & + \underbrace{\sum_{i_1=1}^{n_\theta} \sum_{i_2=1}^{i_1} \sum_{i_3=1}^{i_2} a_{i_1 i_2 i_3}^{(d)}\Gamma_3(\theta_{i_1}, \theta_{i_2}, \theta_{i_3}) + \dots}_{\text{third-order terms}} \end{aligned} \quad (1)$$

where n_θ is the number of parameters, the $a^{(d)}$ with various subscripts are deterministic coefficients in $\mathbb{R}$ to be estimated, and the multidimensional Hermite polynomials of degree $m = i_1, i_2, \dots, i_{n_\theta}$, $\Gamma_m(\theta_{i_1}, \dots, \theta_{i_m})$ are

$$\Gamma_m(\theta_{i_1}, \dots, \theta_{i_m}) = (-1)^m e^{(1/2)\theta^T \theta} \frac{\partial^m e^{-(1/2)\theta^T \theta}}{\partial \theta_{i_1} \dots \partial \theta_{i_m}}. \quad (2)$$

TABLE II
HERMITE POLYNOMIALS FOR THE SECOND-ORDER FOUR-DIMENSIONAL PCE

i th polynomial chaos	Γ_i	Order of polynomial chaos
0	1	0
1	θ_1	1
2	θ_2	1
3	θ_3	1
4	θ_4	1
5	$\theta_1^2 - 1$	2
6	$\theta_1 \theta_2$	2
7	$\theta_1 \theta_3$	2
8	$\theta_1 \theta_4$	2
9	$\theta_2^2 - 1$	2
10	$\theta_2 \theta_3$	2
11	$\theta_2 \theta_4$	2
12	$\theta_3^2 - 1$	2
13	$\theta_3 \theta_4$	2
14	$\theta_4^2 - 1$	2

Table II lists the form of Hermite polynomials for a second-order PCE. The polynomial chaos terms are random variables, since they are functions of the random variables, and terms of different order are orthogonal to each other (with respect to an inner product defined in Gaussian measures as the expected value of the product of the two random variables, i.e., $\mathcal{E}[\Gamma_i \Gamma_j] = 0$ for $\Gamma_i \neq \Gamma_j$). In addition, polynomial chaos terms of the same order but with a different argument list are also orthogonal ($\mathcal{E}[\Gamma_m(\{\theta\}_i) \Gamma_m(\{\theta\}_j)] = 0$, $i \neq j$). In PCE any form of polynomial could be used but the properties of orthogonal polynomials make the uncertainty analysis more efficient. For example, calculating the expected value of both sides of (1) results in the expected value of ψ being simply $\mathcal{E}[\psi^{(d)}] = a_0^{(d)} \Gamma_0$. The calculation of other statistical measures is also significantly simplified using the properties of orthogonality. The orthogonal polynomials are derived from the probability distribution of the parameters ($f_{d.f.}(\theta)$) using the orthogonality condition:

$$\int_{\theta} f_{d.f.}(\theta) \Gamma_i(\theta) \Gamma_j(\theta) d\theta = \delta_{ij}, \quad (3)$$

where δ_{ij} is the Kronecker delta function given by $\delta_{ij} = 1$ if $i = j$ and $\delta_{ij} = 0$ if $i \neq j$. Since $\Gamma_0(\theta) = 1$, the first-order Hermite polynomial can be calculated from $\int_{\theta_1} f_{d.f.}(\theta_1) \Gamma_1(\theta_1) d\theta_1 = 0$ and the procedure can be repeated to obtain all terms in the PCE. The number of coefficients (N_a) in the PCE depends on the number of uncertain parameters and the order of expansion and can be calculated as $N_a = (n_\theta + d)! / n_\theta! d!$. For example, there are 6 coefficients for two parameters in a second-order PCE and 15 coefficients for a fourth-order PCE, whereas for four

uncertain parameters there are 15 coefficients for a second-order PCE and 70 for a fourth-order PCE. For most engineering applications it is not necessary to use higher order than three or four. The polynomial chaos expansion is convergent in the mean-square sense [15], so the coefficients in the PCE can be calculated using least-squares minimization considering sample input/output pairs from the model, to achieve the best fit between the PCE and nonlinear model (or experimental data). Because all parameters are random variables, for a more accurate determination of the PCE coefficients, the probability distribution of $\{\theta_i\}$ has to be considered in the selection of the sampling points. There are two main methods for computing the coefficients of the PCE: (i) the probabilistic collocation method (PCM) [10], [11], and (ii) the regression method with improved sampling (RMIS) [8]. Both methods are weighted-residual schemes, which differ in the way the sampling points are chosen. The methods use the principle of collocation, which imposes that ψ is exact at a set of chosen collocation points, making the residual between the output of PCE and complex nonlinear model at those points equal to zero. In PCM, the number of collocation points is

set equal to the number of unknown coefficients, which are found by solving a set of linear equations generated from the outputs from the original simulation model. In RMIS, additional collocation points are selected to improve the accuracy of the computed coefficients. The two methods also differ in how the collocation points are chosen. In PCM, the collocation points are selected from the roots of the orthogonal polynomial of a degree one higher than the order of the PCE [12]. Which roots are selected affects the accuracy of the approximation. In this paper the probability collocation method was used to calculate the coefficients. An iterative approach to select the order of the PCE was implemented (see Fig. 1), with automatic derivation of the Hermite polynomials using an algorithm based on ORTHPOL [31].

Generally, the approaches based on PCE are similar to the power series expansion techniques [5]-[7], which also utilize a simpler representation of the simulation model that can be used to compute the PDF of the outputs (either directly via the Monte Carlo method or via a contour mapping approach). The PCE can be used to analytically compute statistical measures, such as the mean, variance, or higher order moments of the outputs.

III. UNCERTAINTY ANALYSIS OF A BATCH CRYSTALLIZATION PROCESS USING PCE

Crystallization is an industrially important separation unit. 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 optimal control studies of crystallisation processes compute the temperature profile that optimizes some property of the CSD. Uncertainties in the model parameters can lead to large variations in the final product quality and potential loss of benefits of optimal control. The moment model of the crystallisation process is represented by a set of ordinary differential equations (ODEs) [32]:

$$\dot{x}(t) = f(x(t), u(t); \theta) \quad (4)$$

$$y(t) = g(x(t), u(t); \theta) \quad (5)$$

with $x^T = [\mu_0, \dots, \mu_4, C, \mu_{seed,1}, \dots, \mu_{seed,3}]$ and

$$f(x, u, \theta) = \begin{bmatrix} B \\ G\mu_0 + Br_0 \\ 2G\mu_1 + Br_0^2 \\ 3G\mu_2 + Br_0^3 \\ 4G\mu_3 + Br_0^4 \\ -\rho_c k_v (3G\mu_2 + Br_0^3) \\ G\mu_{seed,0} \\ 2G\mu_{seed,1} \\ 3G\mu_{seed,2} \end{bmatrix} \quad (6)$$

Fig. 1. Flowchart with the steps of the polynomial chaos expansion (PCE)-based distributional robustness analysis with the algorithm to obtain the PCE approximation of the nonlinear model.

where μ_i is the i th moment ($i=0, \dots, 4$) of the CSD resulting from growth from seed and nucleation, $\mu_{seed,j}$ is the j th moment ($j=0, \dots, 3$) 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:

$$G = k_g S^g, \quad (7)$$

$$B = k_b S^b \mu_3, \quad (8)$$

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 $\theta^T = [g, k_g, b, k_b]$, with nominal values given by $\hat{\theta}^T = [1.31, 8.79, 1.84, 17.38]$, with the uncertainty description characterized by the covariance matrix [32]

$$V_{\theta}^{-1} = \begin{bmatrix} 102873 & -21960 & -7509 & 1445 \\ -21960 & 4714 & 1809 & -354 \\ -7509 & 1809 & 24225 & -5198 \\ 1445 & -354 & -5198 & 1116 \end{bmatrix}. \quad (9)$$

An optimal control problem is solved with the objective of computing the optimal temperature profile. This problem can be formulated as a nonlinear optimization problem, which is then solved using general-purpose optimization algorithms. Various objective functions can be used that are properties of the crystal size distribution and are functions of the moments. The most commonly used CSD properties are the nucleation-to-seed-mass ratio ($J_{n.s.r.}$), coefficient of variation ($J_{c.v.}$), and weight-mean size ($J_{w.m.s.}$), which are given by

$$J_{n.s.r.} = (\mu_3 - \mu_{seed,3}) / \mu_{seed,3} \quad (10)$$

$$J_{c.v.} = (\mu_2 \mu_0 / (\mu_1)^2 - 1)^{1/2} \quad (11)$$

$$J_{w.m.s.} = \mu_4 / \mu_3 \quad (12)$$

The optimal temperature trajectory that minimizes the nucleation-to-seed-mass ratio at the end of the batch was computed solving the optimal control problem for the nominal parameter $\hat{\theta}$. The optimal temperature trajectory is used in the uncertainty analysis to assess the effect of parameter uncertainty on the nominal control performance. The distributional uncertainty analysis approach based on polynomial chaos expansion was evaluated using Monte Carlo (MC) simulations in comparison to MC simulations using the first-principles nonlinear model.

The entire PDFs of the states and outputs of interest were computed based on the frequency histograms for all model states and outputs at the end of the batch. The histograms were obtained by splitting the range of the values in equal-sized bins (called classes), and then counting the number of

TABLE III
COMPUTATIONAL COST OF THE VARIOUS APPROACHES TO COMPUTE THE DISTRIBUTION FOR ALL PROCESS OUTPUTS DURING THE ENTIRE BATCH (ON A P4-1.4 GHZ COMPUTER)

Method	Computational time
Monte Carlo simulation with full dynamic model (80,000 points)	8 hours
Monte Carlo applied to second- Second-order polynomial chaos expansion (80,000 points)	20 seconds

points that fall into each bin. The probability distribution functions were calculated from the frequency histograms of the simulations, as the count in the class divided by the number of observations times the class width. For this normalization, the area (or integral) under the histogram is equal to one, and the resulting normalized histogram resembles the real PDF. Fig. 2 shows the histograms obtained via the Monte Carlo method applied to the full nonlinear model, using 80,000 parameter sets. The resulting distributions are not Gaussian, showing the effect of the nonlinearity on the uncertainty propagation. Some of the states are closer to the form of the normal distribution, e.g. μ_3 , whereas others, e.g. μ_0 , show large deviations from a Gaussian distribution. Although the Monte Carlo method provides a good estimate for the shape of the nonlinear distributions, it requires a large number of sampling points and very high computational cost (see Table III), which makes it inappropriate for on-line applications such as real-time robust feedback control and inconvenient for off-line applications such as robust batch recipe design. Monte Carlo approaches that provide more efficient sampling (e.g. quasi-MC [33]) can reduce the computational time, but not to the same degree as PCE, and without producing as useful of a model representation. For fair comparison between the approaches, the same sampling approach was used. The order of the PCE was determined to be two using the iterative approach in Fig. 3 (including third-order terms did not increase the accuracy of the approximation significantly). Since there are four uncertain parameters, the second-order PCE required the determination of 15 coefficients (according to $N_c^{(2)} = 1 + n_{\theta} + n_{\theta}(n_{\theta} + 1)/2$). The second-order four-dimensional PCE is

$$\psi^{(2)}(\theta) = \sum_{i=0}^{14} a_i \Gamma_i, \quad (13)$$

where the Hermite polynomials are listed in Table II. The coefficients a_i were computed using the probabilistic collocation method with the collocation points obtained from the roots of the third-order Hermite polynomials. The second-order PCE very accurately captured the shape of the PDFs at the end of the batch (compare Figs. 2 and 3), at a significantly lower computational cost than the Monte Carlo simulations based on the nonlinear model (see Table III).

Fig. 2. Probability distribution functions at the end of the batch obtained via the Monte Carlo method applied to the full nonlinear model.

Fig. 3. Probability distribution functions at the end of the batch obtained via the Monte Carlo method applied to the second-order polynomial chaos expansion.

Fig. 4. Distribution bands for all process states and outputs obtained from the Monte Carlo simulation (80,000 parameter sets) using the PCE.

The PCE-based uncertainty analysis approach can be used to compute the trajectories for all states due to parametric uncertainties. Fig. 4 shows the distribution bands for all states and the three outputs of interest. These distribution bands indicate how the state trajectories develop during the entire batch due the parametric uncertainties in the model. Figure 4 indicates that for this particular process, for most of the sates, the effect of uncertainties increases towards the end of the batch. The effect of the uncertainties in the kinetic parameters varies significantly among the states and outputs.

Fig. 5. Dynamic evolution of the distribution of the coefficient of variation obtained using the PCE-based uncertainty analysis approach.

For example, while uncertainties lead to significant variation in the zeroth moment, much weaker effect can be observed for the third moment and concentration. This is due to the fact that uncertainties in the nucleation kinetics can significantly influence the number of particles forming during the batch, which is directly related to the zeroth moment. Although the variation in the number of particles can be large, these particles are very small and hence the variation in number has little effect on the total volume of the solid, which is mainly determined by the seed particles.

Variations in the seed moments are smaller compared to the zeroth moment indicating that uncertainties in the nucleation kinetics are more significant than in the growth parameters.

Similarly to the computation of the distributions at the end of the batch, the probability distribution functions can be also computed for all states and output variables for the entire duration of the batch. For illustration, the dynamic evolution of the PDF for the coefficient of variation during the entire batch computed using the PCE-based uncertainty analysis approach is presented in Fig. 5. Similarly to the corresponding plot in Fig. 4, the results indicate that the effect of parameter uncertainties on the coefficient of variation is significantly different during different phases of the batch. The PCE-based uncertainty analysis indicates significantly smaller variation in the coefficient of variation at about 140 min compared to the end of the batch. If the main objective is to achieve consistent size distribution the batch could be stopped earlier (at around 140 min). This would lead to slightly lower yield and smaller crystals for some of the batches, while producing a more consistent coefficient of variation for repeated batches.

IV. CONCLUSIONS

The paper provides an overview of the application of polynomial chaos expansions (PCE), with a particular example for the uncertainty analysis of a finite-time process. The approach is based on the approximate representation of the full process model using the PCE, and provides a qualitative and quantitative estimation of the effect of parameter uncertainties on the states and output variables along a batch. The computational cost of the robustness analysis method based on PCE is significantly lower than the classical Monte Carlo method applied to the full nonlinear model. The PCE can provide a very good approximation of the shape and tails of the output and states distributions, providing a generally applicable approach for uncertainty propagation in robust batch control and design.

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