Joint PCE Surrogate Construction with Uncertainty Quantification for Parameterized Stochastic Processes

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Exascale Catalytic Chemistry

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Outline

1. Motivation and Background
2. Surrogate Construction
3. Application to Catalysis
4. Closure

Overview

- Stochastic models: viewed as functions

$$\mathcal{M}_s \; : \; \mathcal{D}_\Lambda \times \Omega \to \mathbb{R}^d$$
$$(\boldsymbol{\lambda}, \omega) \mapsto \mathcal{M}_s(\boldsymbol{\lambda}, \omega)$$

$\mathcal{D}_\Lambda$: domain of input parameters $\boldsymbol{\lambda}$
Ω: event space for intrinsic stochasticity ω

- Want to perform forward/inverse UQ studies of stochastic model $\mathcal{M}_s(\boldsymbol{\lambda}, \omega)$ when parameters are *uncertain*
  - Build approximation, e.g. $f(\boldsymbol{\lambda}, \omega)$, for model response (output QoI)
  - Polynomial chaos (PC) surrogate is the main workhorse
    - treat $\boldsymbol{\lambda}$, ω, $f(\boldsymbol{\lambda}, \omega)$ as random variables/vectors; represent as PCEs
    - global sensitivity analysis (GSA) is free bi-product

Challenges constructing PCEs & tools to tackle them

- Infinite dimensionality: temporal and/or spatial fields $f(\boldsymbol{\lambda}, \omega; \boldsymbol{t})$
  - ➤ Karhunen-Loève expansion
- High-dimensional input (parametric) space
  - ➤ sparse PC regression
- Multiple uncertainty sources in extracted QoIs
  - – assume input parameters $\boldsymbol{\lambda}$ are controllable but ω is not
  - – direct use of conventional PCEs not ideal:
    - ○ usually relies on *smoothness* of input/output map
    - ○ typically constructed for ensemble means/integrated QoIs (expensive, fails to represent intrinsic noise)
    - ○ ignores/fails to represent residual noise and influence of residual noise on model output

➤ represent random variables with stochastic/parametric PCEs[*,**]

[*] Lüthen *et. al.* A spectral surrogate model for stochastic simulators computed from trajectory samples. *Comput. Method. in Appl. Mech. Eng.*, 406:115875, 2023
[**] Zhu *et. al.* Stochastic polynomial chaos expansions to emulate stochastic simulators. *Int. J Uncert. Quantif.*, 2023

Challenges constructing PCEs & tools to tackle them

Idea of joint stochastic-parametric surrogate models is not new

- see Lüthen *et. al.**, Zhu *et. al.***
  - applied to scalar/vector random variables and random fields
  - give overview of general methodology and summary of GSA for stochastic surrogates
- Our focus
  - ensure surrogate remains in PCE framework
  - represent mapping from input to response PDF (generative)
  - compute Sobol indices for parametric/ noise contributions
  - apply methodology to catalysis

* Lüthen *et. al.* A spectral surrogate model for stochastic simulators computed from trajectory samples. *Comput. Method. in Appl. Mech. Eng.*, 406:115875, 2023

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From Parametric to Stochastic-Parametric PCEs

PCE surrogate *combines* representations of stochastic *and* parametric components to form *joint PCE* surrogate*

$$f(\boldsymbol{\lambda}, \omega) \approx f^{\text{PCE}}(\xi, \zeta) = \sum_{s=0}^{S-1} \underbrace{\left(\sum_{p=0}^{P-1} \boldsymbol{a}_{sp} \Psi_p(\xi) \right)}_{\boldsymbol{z}_s(\boldsymbol{\lambda})} \Psi_s(\zeta)$$

* **JNM** *et. al.* A joint PCE surrogate construction with uncertainty quantification for parameterized stochastic processes applied to heterogeneous catalysis. *In prep.*

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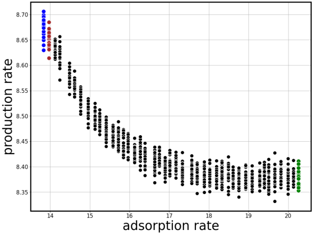
1. Build PCE representations of training samples $f(\boldsymbol{\lambda}^{(n)}, \omega^{(n,m)})$ ($m = 1, ..., M$, n fixed) using *inverse Rosenblatt transformation** *:*



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1. Build PCE representations of training samples $f(\boldsymbol{\lambda}^{(n)}, \omega^{(n,m)})$ ($m = 1, \ldots, M$, n fixed) using *inverse Rosenblatt transformation**:
   - ➤ inverse CDF functions $\mathcal{F}^{-1}(f)$ capture intrinsic noise ω influence (mapping through $\mathcal{F}$ defined using standard germ ζ)

$$\mathcal{R}^{-1}(\zeta) \; : \; f_i = \mathcal{F}^{-1}_{i|i=1,\ldots,1}(\zeta_i|\zeta_{i-1},\ldots,\zeta_1), \quad i = 1,\ldots,d$$

$$\Longrightarrow f(\boldsymbol{\lambda}^{(n)},\omega) \approx \mathcal{R}^{-1}(\zeta) = \sum_{s=0}^{S-1} \boldsymbol{z}_s(\boldsymbol{\lambda}^{(n)}) \Psi_s(\zeta)$$

* Rosenblatt. Remarks on a multivariate transformation. *Annals of Mathematical Statistics*, 23(3):470 – 472, 1952

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$$f(\boldsymbol{\lambda}, \omega) \approx f^{\mathrm{PCE}}(\xi, \zeta) = \sum_{s=0}^{S-1} \underbrace{\left(\sum_{p=0}^{P-1} \boldsymbol{a}_{sp} \Psi_p(\xi) \right)}_{\boldsymbol{z}_s(\boldsymbol{\lambda})} \Psi_s(\zeta)$$

2. Define mapping between model inputs $\boldsymbol{\lambda}$ and output $\boldsymbol{z}_s(\boldsymbol{\lambda})$:

$$\boldsymbol{\lambda} \approx \sum_{p=0}^{P'-1} \boldsymbol{c}_p \Psi_p(\xi), \quad \boldsymbol{z}_s(\boldsymbol{\lambda}) \approx \sum_{p=0}^{P-1} \boldsymbol{a}_{sp} \Psi_p(\xi(\boldsymbol{\lambda}))$$

- cast $\boldsymbol{\lambda}$ in terms of germ ξ
- constructed associated PCE for $\boldsymbol{z}_s(\boldsymbol{\lambda})$
- select PCE orders using model evidence
- construct PCE via sparse regression*

* Babacan *et. al.* Bayesian compressive sensing using Laplace priors. *IEEE Trans. Img. Proc.*, 19(1):53–63, 2010

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$$f(\boldsymbol{\lambda}, \omega) \approx f^{\text{PCE}}(\xi, \zeta) = \sum_{s=0}^{S-1} \underbrace{\left(\sum_{p=0}^{P-1} \boldsymbol{a}_{sp} \Psi_p(\xi) \right)}_{\boldsymbol{z}_s(\boldsymbol{\lambda})} \Psi_s(\zeta)$$

- represents *random variables* $f(\boldsymbol{\lambda}, \omega)$ as polynomial functions of *two* sources of randomness ξ (maps to $\boldsymbol{\lambda}$) and ζ
- PCE surrogate can be *resampled*
- analytic expressions for moments and sensitivity indices, GSA for *both* parametric elements and intrinsic noise
- extends to random fields via Karhunen-Loève expansions (KLE)

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Extending joint PCE to random field QoIs

- Find representation for fields, e.g. Karhunen-Loève expansions*

$$f(\boldsymbol{\lambda}, \omega; \boldsymbol{t}) = f_0(\boldsymbol{t}) + \sum_{\ell=1}^{L} \eta_\ell(\boldsymbol{\lambda}, \omega) \sqrt{\mu_\ell} \phi_\ell(\boldsymbol{t})$$

Represent with joint PCE $\eta_\ell(\boldsymbol{\lambda}, \omega) = \sum_{j=0}^{J-1} \tilde{\boldsymbol{a}}_j \Psi_j(\boldsymbol{\xi}, \zeta)$

- Construct joint PCE for random coefficients $\eta_\ell(\boldsymbol{\lambda}, \omega)$, rewrite

$$f(\boldsymbol{\lambda}, \omega; \boldsymbol{t}) = \sum_{j=0}^{J-1} \underbrace{\left(f_0(\boldsymbol{t})\delta_{0j} + \sum_{\ell=1}^{L} \tilde{\boldsymbol{a}}_{\ell j} \sqrt{\mu_\ell} \phi_\ell(\boldsymbol{t}) \right)}_{\boldsymbol{c}_j(t)} \Psi_j(\boldsymbol{\xi}, \zeta)$$

* Sargsyan *et. al.* Spectral representation and reduced order modeling of the dynamics of stochastic reaction networks via adaptive data partitioning. *SIAM J Sci. Comput.*, 31(6):4395–4421, 2010

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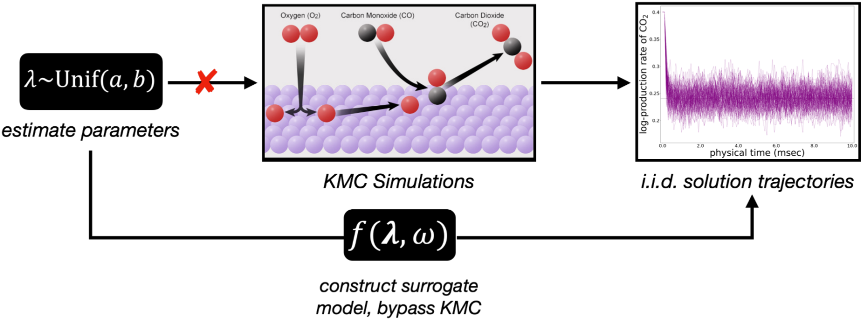
Modeling oxidation of CO on RuO_2 with joint PCEs

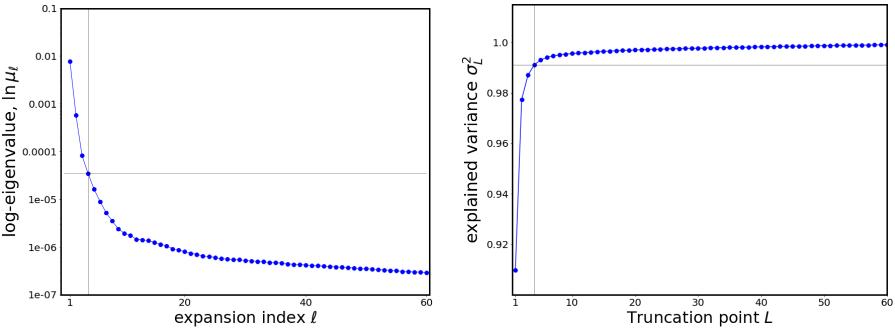


- Multiple molecular species CO, O_2, and CO_2
- *Stochastic* system due to randomness of chemical rections
  - uncertain forward rates collected in vector $\boldsymbol{\lambda} = [\ln k_1, \ldots, \ln k_{15}]$
  - intrinsic stochasticity ω due to randomness of chemical reactions
  - QoI: consumption and production rates over time

Image: https://derekcarrsavvy-chemist.blogspot.com/2017/05/transition-metals-heterogeneous.html

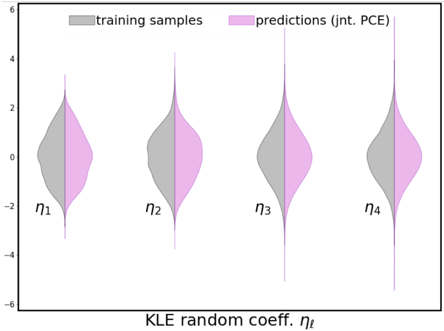
KLE eigenvalue decay indicates room for significant dimension reduction

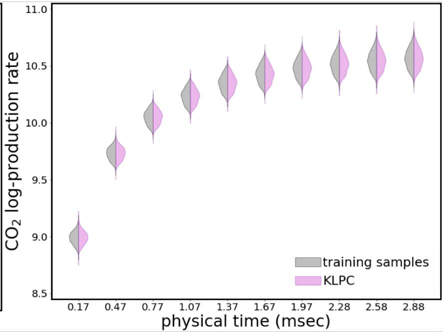
Explained variance in KLE computed from eigenvalues: $\frac{\sum_{\ell=1}^{L} \mu_\ell}{\sum_{\ell=1}^{\mathcal{L}} \mu_\ell}, \quad L \ll \mathcal{L}$



Joint PCE predictions agree in both spectral space and physical (QoI) space

PDF comparisons of combined stochastic-parametric samples





Spectral samples $\eta_\ell(\boldsymbol{\lambda}, \omega)$ *Physical samples* $f(\boldsymbol{\lambda}, \omega; \boldsymbol{t})$

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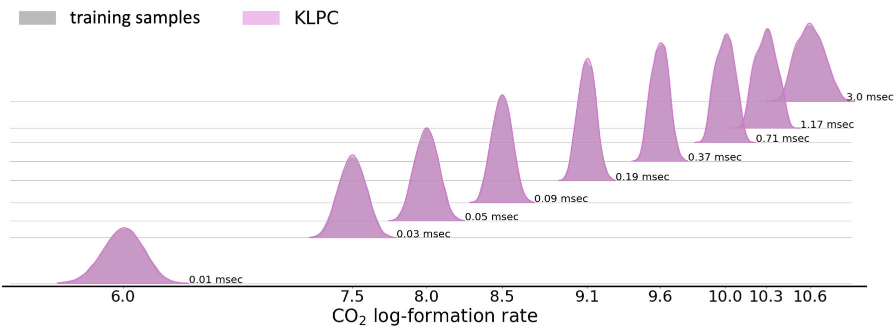
Predictions agree with training samples across time



KLPC surrogate model

- provides *reliable* representations physical model
- is *generative*, i.e. can be resampled
- provides samples from correct distribution *at fraction of cost* (sample-intensive UQ tasks are feasible)

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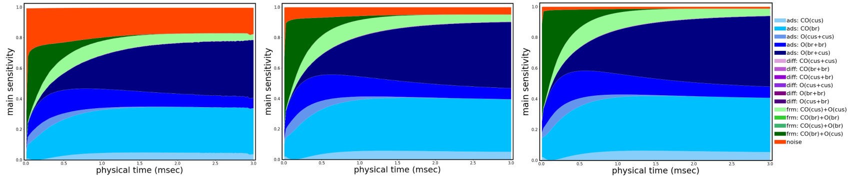
Sobol indices for output QoI reveal reduction in noise as lattice size increases.

$$S_i = \frac{1}{\mathbb{V}[f(\boldsymbol{t})]} \sum_{j \in \mathcal{J}_{S_i}} c_j(t)^2 ||\Psi_j||^2 = \frac{1}{\mathbb{V}[f(\boldsymbol{t})]} \sum_{j \in \mathcal{J}_{S_i}} \left(\sum_{\ell \le L} \tilde{\boldsymbol{a}}_{\ell j} \sqrt{\mu_\ell}\, \phi_\ell(\boldsymbol{t}) \right)^2 ||\Psi_j||^2$$



25 × 25 lattice   50 × 50 lattice   100 × 100 lattice

Sobol indices over time for adsorption rates (blues), diffusion rates (purples), formation rates (greens), and noise (red).

Zhu *et. al.* Global sensitivity analysis for stochastic simulators based on generalized lambda surrogate models. *Reliab. Eng. Syst Safe.*, 214:107815, 2021

Conclusion

Summary:

- PC-based method of constructing surrogate for stochastic model with parametric uncertainty
- Useful for representing both vector and field QoIs
- GSA attributes output uncertainty to *both* parameters and noise
- Successfully applied to a catalysis problem

On-going Work:

- Improvements to method
  - adaptive basis selection for parametric PCE construction
- Extend to chemical systems with correlated parameters λ
- Utilize information ‘thrown away’ in KLE dimension reduction step

...Thank you!