Multi-Scale Modeling with Dynamic Discrepancy

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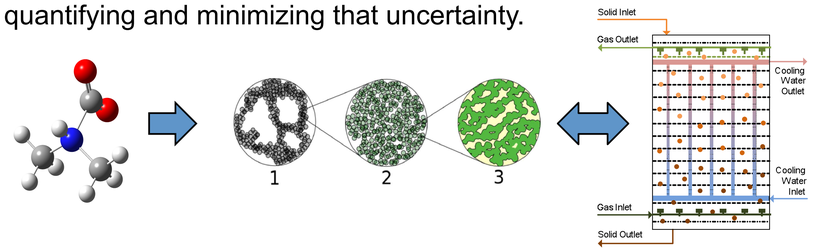
Multi-Scale Modeling in chemical process systems is an inherently statistical problem. Models for the chemistry at the scale of a catalyst or CO_2 sorbent are incorporated into process-level models. Almost invariably, some information must be left out at the larger scale, leading to uncertainty. Obtaining a good process-scale model means quantifying and minimizing that uncertainty.



Uncertainty Quantification and Model Reduction are combined in a novel Bayesian approach called *Dynamic Discrepancy*. Consider a dynamic system

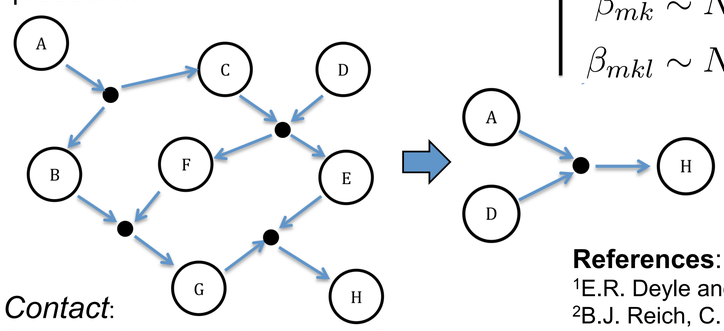
$$\dot{\mathbf{x}} = \mathbf{f}(\mathbf{x}; \mathbf{k}, \boldsymbol{\kappa})$$

where **x** is a vector of state variables and **k** and **κ** are kinetic and equilibrium parameters, respectively. A causal relationship between two states x_i and x_j is established when

$$\dot{x}_i = f_i(\cdots, x_j, \cdots; k_i, \kappa_i)$$
$$\Rightarrow x_j \rightarrow x_i$$

This suggests a method of reducing the order of a reaction network, based on its network topology. In the reaction network below, (circles and letters are states and dots are reactions or other interactions) intermediate states B, C, F, G and E – which propagate causality from the ultimate reactants A and D to the ultimate product H.

Takens has shown that the infinite timeseries of x_i will contain all of the information about the timeseries of x_j.[1]



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What the network map shows is that, if we are only interested in a subset of states in a chemical model, we can usually find some dynamic system containing only those states (with perhaps a few extras) that will provide the correct dynamic behavior.

The reduced dynamic system will be found through *Bayesian Nonparametric Regression*. We introduce a stochastic "discrepancy" function δ which is a Gaussian process:[2]

$$\delta \sim MVN(0, \Gamma)$$

where the covariance kernel Γ may be decomposed as:[2]

$$\Gamma(\vartheta, \vartheta') = \sigma_0^2 + \sum_{k=1}^{K} \sigma_k^2 \Gamma_1(\vartheta_k, \vartheta'_k) + \sum_{k=1}^{K-1} \sum_{l=k}^{K} \sigma_{kl}^2 \Gamma_2([\vartheta_k, \vartheta_l], [\vartheta'_k, \vartheta'_l]) + \cdots$$

with ϑ a functional input such as a reactant or temperature. The covariance is thus broken down in terms of interactions among inputs: first-order, second-order, etc. The kernels are defined as:[2]

$$\Gamma_1(\vartheta_k, \vartheta'_k) = \mathcal{B}_1(\vartheta_k)\mathcal{B}_1(\vartheta'_k) + \mathcal{B}_2(\vartheta_k)\mathcal{B}_2(\vartheta'_k) - \frac{1}{4!}\mathcal{B}_4(|\vartheta_k - \vartheta'_k|)$$

$$\Gamma_2([\vartheta_k, \vartheta_l], [\vartheta'_k, \vartheta'_l]) = \Gamma_1(\vartheta_k, \vartheta'_k)\Gamma_1(\vartheta_l, \vartheta'_l)$$

with $\mathcal{B}_i$ the i^{th} Bernoulli polynomial. A Karhunen-Loéve decomposition then leads to:[2]

$$\delta(\vartheta; \beta) = \beta_0 + \sum_{k=1}^{K} \sum_{m=1}^{M} \beta_{mk} \varphi_{m1}(\vartheta_k) + \sum_{k=1}^{K-1} \sum_{l=k+1}^{K} \sum_{m=1}^{M} \beta_{mkl} \varphi_{m2}(\vartheta_k, \vartheta_l) + \cdots$$

here the basis functions φ are deterministic and known, while coefficients β are independently distributed:

$$\beta_{mk} \sim N(0, \lambda_{m1}\sigma_k^2) \quad \text{with } \lambda_{ij} \text{ the eigenvalue corresponding to the basis function } \varphi_{ij}.[2]$$

$$\beta_{mkl} \sim N(0, \lambda_{m2}\sigma_{kl}^2)$$

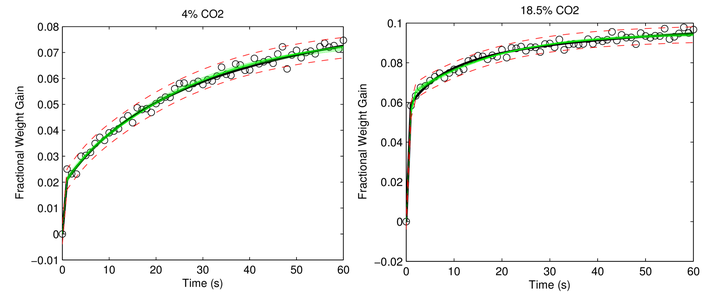
$$\dot{x}_i = f_i(x_i, x_j; k_i, \kappa_i)$$

$$k_i = k_i^0 \exp\left[\delta_{k_i}(x_i, x_j, T; \beta_{kijT})\right]$$

$$\kappa_i = \kappa_i^0 \exp\left[\delta_{\kappa_i}(x_i, x_j, T; \beta_{\kappa ijT})\right]$$

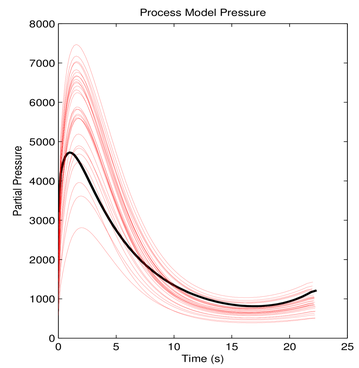
The reduced dynamic system is then constructed from the reduced set of states and discrepancy functions as shown at left.[3]

Bayesian calibration (Markov Chain Monte Carlo) is then employed to find the distribution of **β** that fits bench-scale data. The uncertainty contained in the distribution can then be propagated to the next scale.

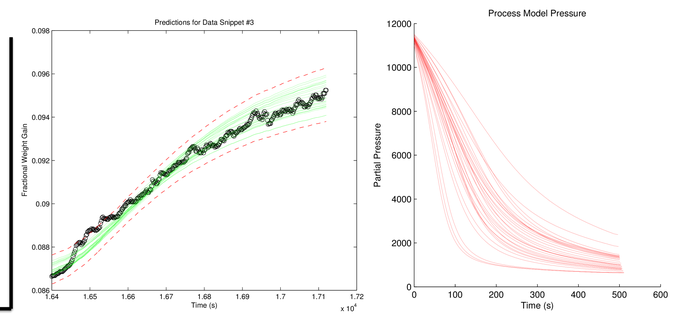


Simulated TGA data generated for a hypothetical CO_2 sorbent, with draws (green) from the discrepancy model distribution.[3]

The discrepancy model was then implemented in an Aspen-based model of a bubbling fluidized bed adsorber.[4]

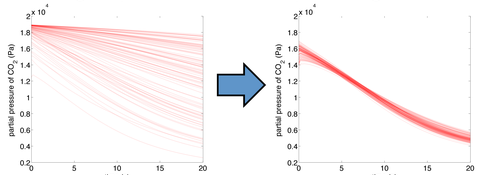


Uncertainty propagation to the process scale for a hypothetical CO_2 sorbent. Model runs are in red; the "real" process behavior is the black curve. Units are Pa; time is convertible to bed depth.[3]



Above: application of the method to a real PEI-silica sorbent (measurement by James Hoffman, NETL). Left: TGA data calibration. Right: process model upscaling.[3]

Dynamic Discrepancy is a foundation for machine learning in process design and control. Quantification of uncertainty leads to methods that can minimize uncertainty.



References:
[1]E.R. Deyle and G. Sugihara, PLoS One 6 (2011) e18295.
[2]B.J. Reich, C. B. Storlie and H.D. Bondell, Technometrics 51 (2009) 110.
[3]K.S. Bhat, D.S. Mebane, C.B. Storlie, P. Mahapatra, submitted.
[4]A. Lee and D.C. Miller, Ind. Eng. Chem. Res. 52 (2013) 469.