# DECISION MAKING WITH HYBRID MODELS UNDER PARAMETER AND EPISTEMIC UNCERTAINTY: REACTOR OPTIMIZATION CASE STUDY

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*Abstract*

In science and engineering, multiscale computer models are used to investigate, simulate, and predict behaviors of complex systems. These models, however, are subject to epistemic (i.e., model-form) uncertainty as they require approximations or simplifications to maintain computational tractability. Kennedy-O'Hagan (KOH) hybrid models, which augment first-principles formulations with data-driven Gaussian processes, account for missing phenomena due to these approximations, simplifications, or lack of mechanistic knowledge. While there exists a corpus of literature on KOH hybrid models for prediction and uncertainty quantification, decision making under uncertainty is largely not considered. In this work, we present a reactor optimization case study to demonstrate how KOH hybrid models can be used to inform decision making when the underlying phenomena are unknown. We show that despite parametric uncertainty and model inadequacy, the hybrid model captures the unknown kinetics with less data than the pure machine learning approach. Further, we demonstrate how the superior predictive of performance of the hybrid model translates to decision making when optimizing reactor design and operation. We posit that KOH hybrid models offer multiscale modeling in process systems engineering a new approach to incorporate quantified aleatoric (e.g., parametric, random) and epistemic uncertainties into decision making frameworks.

*Keywords*

Discrete & Hybrid Systems, Uncertainty & Stochastic Systems, Data Science & Machine Learning

## Introduction

Since the mathematization of process systems engineering (PSE) in the 1960's, first-principles, "white-box", or "glass-box" models have been the dominant framework for guiding process development and discovery (Sansana, et al., 2021). These complex models, however, require expert knowledge to formulate and approximations to ensure computational tractability thereby introducing epistemic (i.e., model-form) uncertainty. These shortcomings coupled with the growing availability of data in recent years have catalyzed the engineering research community to utilize machine learning techniques to gain insight into system behaviors (Beck, Carothers, Subramanian, & Pfaendter, 2016). These so-called "black-box" models have excellent predictive capabilities when rich, large data sets are available, however, they fail to utilize prior systems knowledge, have poor predictive performance for out-of-control samples, and are limited in interpretability (Shulkind, Horesh, & Avron, 2018). At the intersection of these models are "grey-box" or "hybrid" models which synergize white- and black-box constructs to yield models with better prediction accuracy, model stability, or both.

In their seminal work, statisticians Kennedy and O'Hagan presented a framework for Bayesian calibration of computer (hybrid) models (Kennedy & O'Hagan, 2002). Here, the relationship between the observation $y_i$, the true process $\zeta(\cdot)$, and the computer model output $\eta(\cdot)$ is given by

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$$y_i = \zeta(\mathbf{x}_i) + \epsilon_i = \eta(\mathbf{x}_i, \boldsymbol{\theta}) + \delta(\mathbf{x}_i; \boldsymbol{\omega}) + \epsilon_i, \quad (1)$$

where $\epsilon_i$ is the observation error of the $i^{\text{th}}$ observation, $\delta(\cdot)$ is a model discrepancy function, $\boldsymbol{\theta}$ is a vector of physically meaningful parameters, $\boldsymbol{\omega}$ is a vector of hyperparameters, and $\mathbf{x}_i$ is a vector of decision variables. It is assumed that the discrepancy follows a Gaussian process (GP) $\delta \sim \mathcal{GP}\big(m(\cdot), k(\cdot,\cdot)\big)$ where $\mathrm{m}(\mathbf{x}; \boldsymbol{\omega}) = \mathbb{E}[\delta(\mathbf{x}; \boldsymbol{\omega})]$ (expected value) and $k(\mathbf{x}, \mathbf{x}'; \boldsymbol{\omega}) = \mathrm{Cov}[\delta(\mathbf{x}; \boldsymbol{\omega}), \delta(\mathbf{x}'; \boldsymbol{\omega})]$ (covariance).

Hybrid models have been predominantly used in PSE for process monitoring (Bui, et al., 2022; Wang, et al., 2022; Puliyanda, Srinivasan, Sivaramakrishnan, & Prasad, 2022), control & optimization (Paulson & Lu, 2022; Paulson, Shao, & Mesbah, 2021), and uncertainty quantification (Ostace, et al., 2022). Largely missing in the literature, however, is deploying Kennedy-O'Hagan (KOH) hybrid models for decision making under epistemic uncertainty. In this work, we apply the KOH hybrid framework to a reaction engineering case study to quantify the epistemic uncertainty of a mechanistic kinetic model to inform reactor design and operation. We show that KOH hybrid kinetic models have superior predictive power to their mechanistic counterparts alone. Lastly, we demonstrate how KOH hybrid models can be used in a decision making context to optimize reactor design and operation despite model inadequacy.

## Methods

In this case study, we assume the true kinetics underlying the noisy observations are generated with time-series, isothermal, batch kinetics of the form

$$\mathrm{A} \underset{\kappa_3}{\overset{\kappa_1}{\rightleftharpoons}} \mathrm{B} \overset{\kappa_2}{\rightarrow} \mathrm{C}. \quad (2)$$

Model inputs $\mathbf{x}_i = [\mathrm{t}, \mathrm{T}, \mathrm{c_{A0}}]^\intercal$ represent time (h), temperature (K), and initial concentration of species A (M), respectively. The rate equations of this reaction are

$$\frac{dc_A}{dt} = -\kappa_1 c_A + \kappa_3 c_B, \quad (3)$$

$$\frac{dc_B}{dt} = \kappa_1 c_A - \kappa_2 c_B^3 - \kappa_3 c_B, \quad (4)$$

$$\frac{dc_C}{dt} = \kappa_2 c_B^3, \quad (5)$$

where $\kappa_1, \kappa_2$, and $\kappa_3$ are rate constants. Eqs. (3)-(5) are solved numerically using initial conditions

$$c_A(t=0) = c_{A0}, c_B(t=0) = 0, c_C(t=0) = 0, \quad (6)$$

yielding the full kinetic model $\zeta(\cdot)$. We restrict the outputs of this model to consider only species B giving rise to the set of $n$ truth observations $\{y_i\}_{i=1}^n$

$$y_i = \zeta(\cdot) + \epsilon_\mathrm{i}, \quad (7)$$

where $\epsilon \sim \mathcal{N}(\mathbf{0}, \sigma_\epsilon^2 \mathbf{I})$. Alternatively, we consider a simple reaction mechanism

$$\mathrm{A} \overset{\mathrm{k_1}}{\rightarrow} \mathrm{B} \overset{\mathrm{k_2}}{\rightarrow} \mathrm{C}\,, \quad (8)$$

which yields rate equations

$$\frac{dc_A}{dt} = -k_1 c_A, \quad (9)$$

$$\frac{dc_B}{dt} = k_1 c_A - k_2 c_B, \quad (10)$$

$$\frac{dc_C}{dt} = k_2 c_B. \quad (11)$$

Here, $k_1$ and $k_2$ are solved with the Arrhenius equation

$$k = \alpha \exp\left(\frac{-E}{RT}\right), \quad (12)$$

where $\alpha$ is the pre-exponential factor ($\mathrm{s^{-1}}$) and $E$ is the activation energy (kJ/mol) of reactions one and two. Employing the initial conditions in Eq. (6), Eqs. (9)-(11) have a known analytical solution

$$c_A = c_{A0} \exp(-k_1 t), \quad (13)$$

$$c_B = \frac{k_1}{k_2 - k_1} c_{A0} [\exp(-k_1 t) - \exp(-k_2 t)], \quad (14)$$

$$c_C = c_{A0} - c_B - c_A, \quad (15)$$

where $c_B$ is taken as the simple kinetics model $\eta(\cdot,\cdot)$ with parameters $\boldsymbol{\theta} = [\alpha_1, \alpha_2, E_1, E_2]^\intercal$. Nonlinear least squares regression is used to fit $\{y_i\}_{i=1}^n$ to $\eta(\cdot,\cdot)$ yielding parameter and noise variance estimates $\widehat{\boldsymbol{\theta}}$ and $\widehat{\sigma_\epsilon}^2$ given by

$$\widehat{\sigma_\epsilon}^2 = \frac{\sum_{i=1}^n \big(y_i - \eta(\mathbf{x}_i, \widehat{\boldsymbol{\theta}})\big)^2}{n-4}. \quad (16)$$

By Eq. (1), the model discrepancy function $\delta(\cdot)$ represents the difference between $\zeta(\cdot)$ and $\eta(\cdot,\cdot)$. We assert that $\delta(\cdot)$ follows a GP with mean zero and kernel function $k(\cdot,\cdot)$, $\delta(\cdot) \sim \mathcal{GP}(0, k(\cdot,\cdot))$. This choice in mean function is satisfied by normalizing the model inputs prior to training.

Moreover, we specify the covariance $k(\cdot,\cdot)$ with the $\nu = 3/2$ Matern kernel with hyperparameters $\boldsymbol{\omega} \in \mathbb{R}^4$.

Figure 1 demonstrates the main steps of the proposed hybrid modeling framework. In the first step, the set of noisy observations $\{y_i\}_{i=1}^n$ is generated from $\zeta(\cdot)$ with known model inputs $\{\mathbf{x}_i\}_{i=1}^n$ in lieu of conducting experiments. We then propose and formulate a (simplified) kinetic model $\eta(\mathbf{x}_i, \boldsymbol{\theta})$ and obtain parameter and error estimates with nonlinear least squares regression. The noisy discrepancy is computed by taking the difference between the observations and the simple kinetic model estimates, $\delta(\mathbf{x}_i) + \epsilon_i = y_i - \eta(\mathbf{x}_i, \widehat{\boldsymbol{\theta}})$. For GP regression, a training data set $\mathcal{D} = \{\mathbf{X}, \boldsymbol{\Delta} + \boldsymbol{E}\}$ is assembled where $\mathbf{X} = [\mathbf{x}_1, \dots, \mathbf{x}_n]^\intercal \in \mathbb{R}^{n\times 3}$ is a matrix of the model inputs and $\boldsymbol{\Delta} + \mathbf{E} = [\delta_1 + \epsilon_1, \dots, \delta_n + \epsilon_n]^\intercal \in \mathbb{R}^n$ is a vector of noisy discrepancy observations. Priors are posed for the hyperparameters $\boldsymbol{\omega}$ and the measurement variance $\sigma_\epsilon^2$ which we refer to collectively as $\boldsymbol{\psi} = [\boldsymbol{\omega}^\intercal, \sigma_\epsilon^2]^\intercal$. The maximum a posteriori (MAP) estimates of the hyperparameters $\widehat{\boldsymbol{\psi}}_{\text{MAP}}$ are obtained with Bayes' rule to inform the discrepancy posterior $\delta|\mathcal{D}$ with mean $\mu_\delta$ and variance $\sigma_\delta^2$. This concludes hybrid kinetic model calibration, enabling predictions $y^*$ to be made over model inputs outside the training set $\mathbf{x}^*$.

We quantify parametric uncertainty in predictions $y^*$ using nonlinear error propagation. We denote the set of functions given in Eqs. (13)-(15) as $\{\mathbf{f}(\widehat{\boldsymbol{\theta}})\}$ such that

$$\mathbf{c}^* = \mathbf{f}(\mathbf{x}^*, \widehat{\boldsymbol{\theta}}), \tag{17}$$

where $\mathbf{c} = [c_A, c_B, c_C]^\intercal$ is a vector of the concentrations. A first-order Taylor series expansion yields

$$\mathbf{f} = \mathbf{f_0} + \mathbf{J}\widehat{\boldsymbol{\theta}}, \tag{18}$$

where $\mathbf{J}$ is the Jacobian which is approximated with a centered finite difference. The variance-covariance of the estimated parameters $\boldsymbol{\Sigma}_{\widehat{\boldsymbol{\theta}}}$ is used to compute the variance-covariance of predictions

$$\boldsymbol{\Sigma}_{\mathbf{f}} = \mathbb{E}[(\mathbf{f} - \mathbb{E}[\mathbf{f}]) \otimes (\mathbf{f} - \mathbb{E}[\mathbf{f}])] \tag{19}$$

$$= \mathbf{J}\mathbb{E}\left[\left(\widehat{\boldsymbol{\theta}} - \mathbb{E}[\widehat{\boldsymbol{\theta}}]\right) \otimes \left(\widehat{\boldsymbol{\theta}} - \mathbb{E}[\widehat{\boldsymbol{\theta}}]\right)\right]\mathbf{J}^\intercal \tag{20}$$

$$= \mathbf{J}\boldsymbol{\Sigma}_{\widehat{\boldsymbol{\theta}}}\mathbf{J}^\intercal. \tag{21}$$

We are interested in the objective function $\phi$ to maximize the value of the products

$$\phi = \mathbf{w}^\intercal \cdot \mathbf{c}(\mathbf{x}^*), \tag{22}$$

where $\mathbf{w} = [-0.5, 1, -0.5]^\intercal$ is a vector of weights. We seek

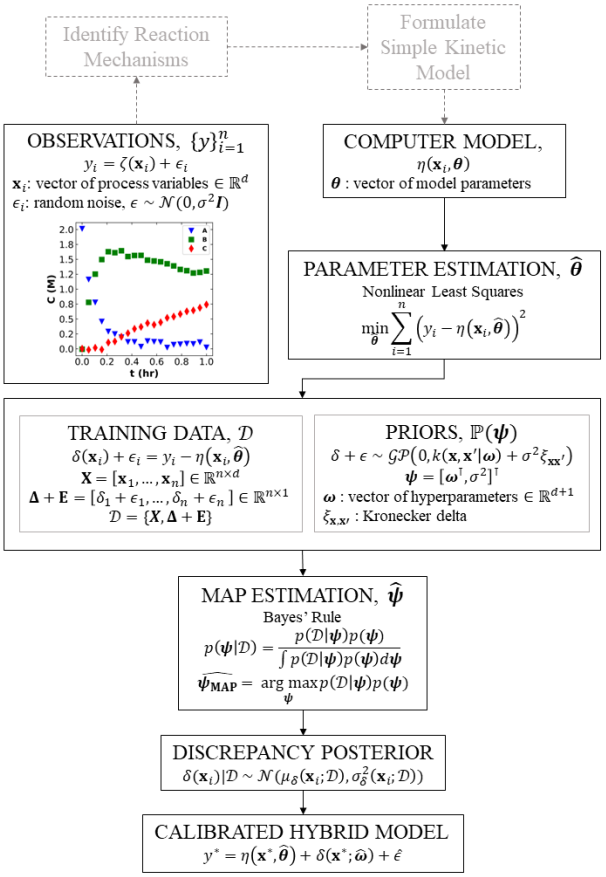



*Figure 1. Main steps of the hybrid modeling framework.*

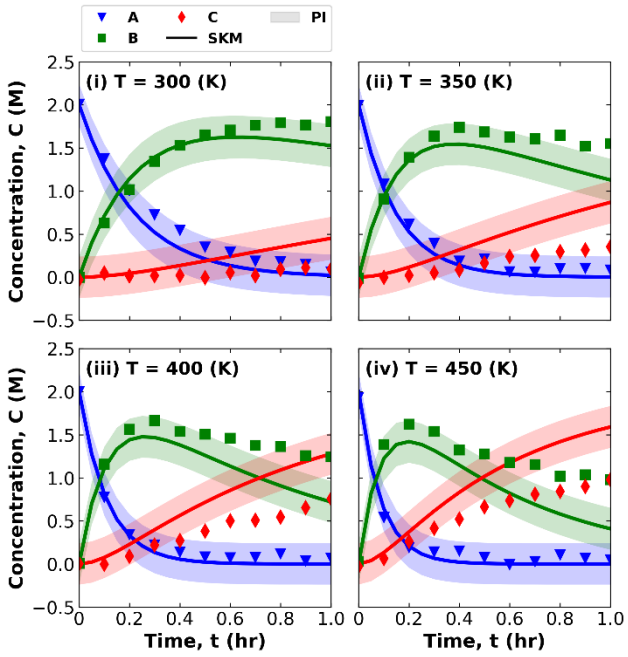
to optimize the batch reaction time and temperature to maximize the value of the objective function

$$\max_{t,T} \phi, \tag{23}$$

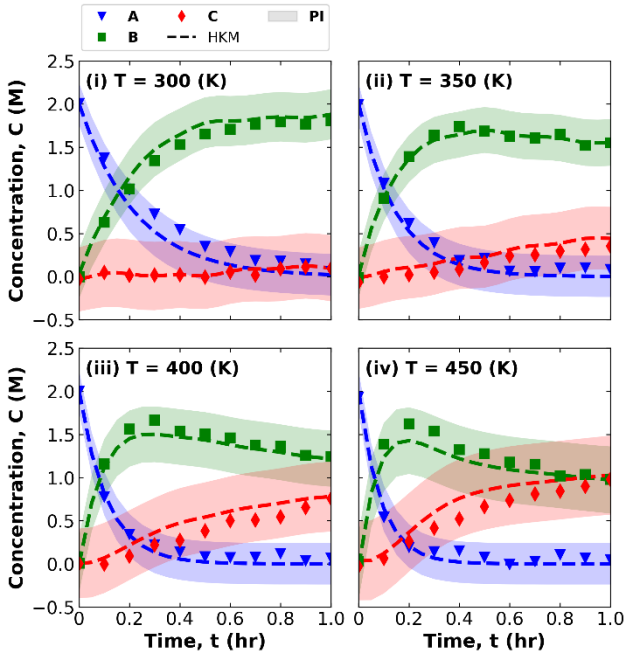
$$\text{s.t.} \quad t \in [0, 1], T \in [293, 493]. \tag{24}$$

## Results

Estimated parameters $\widehat{\boldsymbol{\theta}}$ were obtained via nonlinear least squares regression to benchmark the predictive performance of the hybrid model. Figure 2 shows simple (a) and hybrid (b) model kinetic predictions for temperatures 300 (i), 350 (ii), 400 (iii), and 450 (iv) (K). Shaded regions represent time-series propagated uncertainty. Figure 2 (a) demonstrates that the simple kinetic model prediction accuracy decreases at longer times and hotter temperatures. Moreover, the mean squared error (MSE) and mean absolute error (MAE) for the simple kinetic model increase with temperature ranging from 0.45-2.35 and 0.41-0.71, respectively. We compute the percent outside the prediction




*(a)*



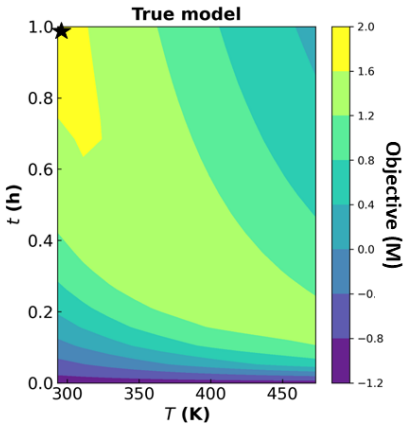

*(b)*

*Figure 2. Isothermal, time-series kinetics of (a) simple (SKM) (-) and (b) hybrid (HKM) (--) kinetic models considering $c_{A0}$ = 2.0 (M) across T= (i) 300, (ii) 350, (iii) 400, & (iv) 450 (K). Concentrations of A, B, and C are shown with red diamonds, green squares, and blue triangles, respectively. Shaded regions represent prediction intervals (PI, $\pm\hat{\boldsymbol{\sigma}}$).*

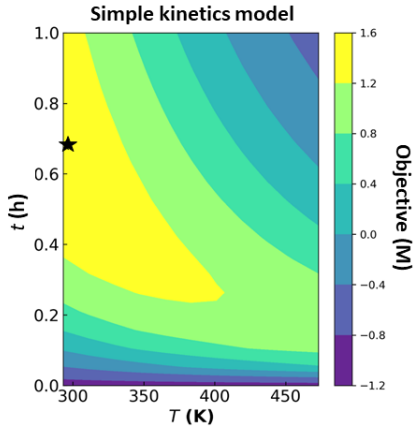
interval (POPI) as the percent of all observations outside the prediction $\pm\hat{\boldsymbol{\sigma}}$. For the simple kinetic model, this value ranges from 9.09%-42.42%. Taken as a whole, these results demonstrate poor predictive performance of the simple kinetic model at certain model inputs and failure of frequentist statistical methods to capture uncertainty.

In Figure 2 (b), we observe that the hybrid model prediction accuracy is robust to temperature fluctuations. In support of this observation, the MSE and MAE of the predictions do not demonstrate a relationship to model inputs, ranging from 0.10-0.31 and 0.17-0.27, respectively. The POPI is 0% at all four temperatures studied. Thus, prediction intervals for the hybrid model are sufficiently large to capture the variability in the training data, unlike the simple kinetic model. On average, the MSE, MAE, and POPI of the hybrid model are much less than those of its mechanistic counterpart alone, indicating superior predictive performance of the hybrid model across model inputs.

With the calibrated hybrid model, the objective function was computed for $t \in [0, 1]$ and $T \in [293, 493]$. Figure 3 shows contour plots of the objective function for the (a) true, (b) simple, and (c) hybrid kinetic models. As shown in Figure 3 (a), the true model predicts the maximum objective at low temperatures and long times. Comparing the true model with the simple kinetic model (Figure 3 (b)), we observe that the simple kinetic model optimum is also favored at low temperatures but underpredicts the time and value of the objective function. Lastly, the hybrid model presented in 3 (c) demonstrates results close to those of the true model. Taken as a whole, these results demonstrate how superior predictive capability of the hybrid model yields better decisions for optimizing the reactor controls under uncertainty.




*(a)*

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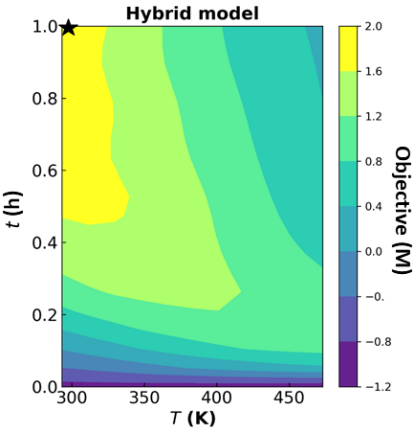


*(c)*

*Figure 3. Time-temperature contour plots of the objective function for the (a) true, (b) simple, and (c) hybrid kinetic models. Black stars represent the location of the maximum.*

## Conclusions

In this work, we utilized a batch reactor optimization case study to conceptualize hybrid model decision making under epistemic uncertainty. We found that the hybrid kinetic model demonstrates superior predictive performance when compared to its simple kinetic counterpart. Moreover, the GP component of the hybrid model successfully learns and corrects for severe model inadequacy. We show how superior predictive capability of the hybrid kinetic model yields a framework for reactor optimization under uncertainty. In future work, we plan to quantify the closed loop performance on the framework when new experiments are adaptively proposed and added to training data. In doing so, we will explore the tradeoff between exploration and exploitation for choice in acquisition function (e.g., expected improvement, lower confidence bound). In our previous work, we showed that a pure black-box surrogate (GP regression) approach requires careful tuning and struggles with limited data using a cannonball case study (Eugene, Gao, & Dowling, 2019). Moreover, we found that KOH hybrid models overcome limitations this approach. Toward extending this work, we will benchmark a GP-only model with the reaction case study. Lastly, we will demonstrate scalability of the hybrid kinetic model by considering discrepancy in all chemical species in the reaction network. Building on our previous work, we aim to embed our optimization problem in a scalable stochastic program to facilitate more sophisticated reactor design (Wang, Eugene, & Dowling, 2022).

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