Computers and Chemical Engineering 81 (2015) 376–388

Contents lists available at ScienceDirect

Computers and Chemical Engineering

journal homepage: www.elsevier.com/locate/compchemeng

Bayesian estimation of parametric uncertainties, quantification and reduction using optimal design of experiments for CO_2 adsorption on amine sorbents

Jayashree Kalyanaraman, Yanfang Fan, Ying Labreche, Ryan P. Lively, Yoshiaki Kawajiri, Matthew J. Realff*

School of Chemical & Biomolecular Engineering, Georgia Institute of Technology, 311 Ferst Drive, Atlanta, GA 30332, United States

ARTICLE INFO

Article history:
Received 11 November 2014
Received in revised form 16 April 2015
Accepted 23 April 2015
Available online 16 May 2015

Keywords:
Bayesian inference
Adaptive metropolis
Parallel propagation
Optimal experimental design
CO_2 adsorption

ABSTRACT

Uncertainty quantification plays a significant role in establishing reliability of mathematical models, while applying to process optimization or technology feasibility studies. Uncertainties, in general, could occur either in mathematical model or in model parameters. In this work, process of CO_2 adsorption on amine sorbents, which are loaded in hollow fibers is studied to quantify the impact of uncertainties in the adsorption isotherm parameters on the model prediction. The process design variable that is most closely related to the process economics is the CO_2 sorption capacity, whose uncertainty is investigated. We apply Bayesian analysis and determine a utility function surface corresponding to the value of information gained by the respective experimental design point. It is demonstrated that performing an experiment at a condition with a higher utility has a higher reduction of design variable prediction uncertainty compared to choosing a design point at a lower utility.

1. Introduction

Models play a key role in the design of chemical processes. Mathematical models, although meant to represent and predict system behavior accurately, are compromised due to uncertainties of our knowledge of the system. Uncertainty in model, and hence its predictions, arises either due to uncertainties associated with the model representation itself, causing *model uncertainty* or due to errors in the parameter values used in the model, leading to *parametric uncertainty*. Model uncertainty occurs primarily because of inaccurate understanding of the underlying system, or due to over-simplifying assumptions used in the model while representing the system behavior. Parametric uncertainty, on the other hand, occurs due to errors associated with the measurement variables or because of the sparsity of the available measurements used to estimate the model parameters. While it is theoretically possible to reduce or even completely remove the uncertainties in the model parameters, it can often be very challenging due to the inherent physical limitations associated with the measurement devices or the sheer complexity of performing an experiment to collect data. Nevertheless, it is important to quantify the uncertainties and their impact on the model predictions, in order to use the model reliably in process design.

Uncertainty quantification (UQ) has grown in importance across many fields including chemical engineering (Duran and White, 1995; Coleman and Block, 2006; Hermanto et al., 2008; Najm et al., 2009; Angelikopoulos et al., 2012). There have been a number of methods developed to characterize, quantify, or propagate uncertainties including probabilistic methods, such as classical or frequentist inference and Bayesian inference (Omlin and Reichert, 1999), sampling methods such as Monte Carlo (Basil and Jamieson, 1998) and bootstrapping (Efron, 1979), and response surface methods (intrusive methods) such as polynomial chaos expansions (Xiu and Karniadakis, 2003) and Kriging approach (Yuan et al., 2008). Among these different methods, Bayesian inference offers several advantages over other methods as it does not require modification to the model and provides a comprehensive treatment of parametric and model uncertainties without any simplifying assumptions about their distributions (Omlin and Reichert, 1999; Alfaro et al., 2003). However, applying Bayesian inference and propagating the uncertainties using Monte Carlo sampling of the posterior parametric distribution or using polynomial chaos expansions leads to prohibitively expensive computation when applied to large models, as is commonly observed with any physio-chemical process,

* Corresponding author. Tel.: +1 404 862 0440.
E-mail address: matthew.realff@chbe.gatech.edu (M.J. Realff).

http://dx.doi.org/10.1016/j.compchemeng.2015.04.028
0098-1354/

Nomenclature

A, B	constants defining the temperature dependency of n
b	Toth isotherm affinity constant [Pa^{-1}]
b_0	parameter defining the temperature dependency of b [Pa^{-1}]
C_{br}	limiting CO_2 gas phase concentration at breakthrough [mol/m^3]
C_g	CO_2 gas phase concentration [mol/m^3]
C_p	specific heat capacity [J/kg K]
e_d	multiplicative factor defining the expansion rate of proposal covariance matrix
h_g	flue gas convective heat transfer coefficient [W/m^2 K]
h_t	convective heat transfer coefficient between thermocouple and module [W/m^2 K]
L	length [m]
n	Toth isotherm heterogeneity parameter
$\mathcal{N}$	normal distribution
P	pressure [Pa]
q_{avg}	average CO_2 loading in sorbent [mmol/g fiber]
q_{bulk}	CO_2 loading in sorbent within the bulk PEI sites [mmol/g fiber]
q_{br}	CO_2 loading in sorbent at breakthrough time t_{br} [mmol/g fiber]
q_{eq}	CO_2 loading at equilibrium [mmol/g fiber]
q_m	maximum possible CO_2 loading at a given temperature [mmol/g fiber]
q_{m0}	maximum possible CO_2 loading at the reference temperature T_0 [mmol/g fiber]
$q_{surface}$	CO_2 loading in sorbent within the surface PEI sites [mmol/g fiber]
r_o	outer fiber radius [m]
r_i	inner fiber radius [m]
r_{fs}	radius of free space surrounding fiber [m]
s_d	multiplicative factor defining the shrinking rate of proposal covariance matrix
T	temperature [K]
u_g	bulk gas velocity [m/s]
$U(d)$, $U'(d)$	utility function describing the information gain of experiment at d
U	overall heat transfer coefficient of the module [W/m^2 K]
$\mathcal{U}$	uniform distribution
$1/K_{ov,surface}$	overall mass transfer resistance for CO_2 adsorption on surface PEI sites [s]
$1/K_{ov,bulk}$	overall mass transfer resistance for CO_2 adsorption on bulk PEI sites [s]

Greek symbols

α	parameter defining the rate of increase of mass transfer resistance [g fiber/mmol]
ϵ	porosity [–]
μ	sensitivity parameter mean/distribution mean [–]
σ	standard deviation of sensitivity/distribution [–]
Θ	set of adsorption isotherm parameters and hyperparameter [–]
ΔH_0	isosteric heat of adsorption at zero loading [kJ/g mol]
ΔH_{avg}	average heat of adsorption [kJ/g mol]

Subscripts

f	fiber
g	gas
t	thermocouple

involving coupled partial differential equations. In effect, most of the studies on quantifying uncertainties in the field of chemical engineering have been restricted to characterizing uncertainties (parametric inference problems), most of which involves simple batch system models such as kinetic models (Najm et al., 2009; Hsu et al., 2009; Albrecht, 2013), adsorption isotherms (Anagu et al., 2012; Mebane et al., 2013), fed batch fermentation reactors (Coleman and Block, 2006) and microbiology models (Pouillot et al., 2003), and only few studies have been performed on larger models such as packed bed adsorbers (Duran and White, 1995; CSTRs (Chen et al., 2004) and crystallization processes (Hermanto et al., 2008).

While it is desirable to reduce the uncertainties associated with the model and parameters, it may often be difficult and expensive to perform experiments and collect data that are required to reduce the uncertainties. Under such scenarios, it is advantageous to determine the conditions at which performing experiments would provide maximum information gain. Such a strategy is termed as optimal experimental design (OED). By optimally designing the experiments, one can gain maximum amount of information about the system using least number of experiments.

Model-based design of experimental methods using the Fisher Information Matrix (FIM) (Atkinson et al., 2007) have been well studied for models with linear parametric dependence, employing the well known optimal design conditions termed as *alphabetical optimality criteria*, which are evaluated as functionals of FIM (Franceschini and Macchietto, 2008). In case of models with nonlinear parametric dependence, a number of simplifying assumptions are made to employ FIM based optimal experimental design, including linearization of the model response and Gaussian approximation of the parametric distributions (Chu and Hahn, 2007). Bayesian experimental design, on the other hand, does not require, in general (Chaloner and Verdinelli, 1995) any simplifying assumptions on the parametric distribution or the model linearization. Measure of the information gain using Bayesian analysis, is represented by an objective function called the *utility function*, which includes an optimality criteria that is maximised to determine the optimal design condition (Lindley, 1972). A detailed review on the Bayesian experimental design methods and the variations of the utility function can be found in Chaloner and Verdinelli (1995).

Bayesian experimental design was first introduced by Lindley (1972), where Shannon information gain is used from posterior to prior distribution for the experimental design; followed by several others using variations of optimality criteria (Brooks, 1977; Shewry and Wynn, 1987). The computational expense of evaluating the utility function has been a major challenge in deploying Bayesian design to determine the optimal experiment as most of the real world models are complex and cannot be analytically evaluated (Ryan, 2003; Terejanu et al., 2012). In effect, most of the reported work on Bayesian experimental design have been using linear models and in the few studies having non-linear models, approximations of the utility function or Gaussian approximations of the posterior distributions are used (Russi et al., 2008; Mosbach et al., 2012). In that context, Muller and Parmigiani (1995) suggested using a Monte Carlo estimator and simulation based optimal design by fitting the Monte Carlo samples of utility surface. However, they concluded that the evaluation becomes computationally prohibitive for large dimensions of design parameters. There have been very limited studies on Bayesian experimental design in the chemical engineering literature. In recent work, Solonen et al. (2012) applied the simulation based optimal design for a CSTR model using variance of predictions as the utility function. They sidestepped the computational complexity of posterior distribution evaluation after every added experiment, by weighing the parameters with the corresponding likelihood of the new measurements. Even with such methodology, the likelihood evaluation could turn out be