Model and Parameter Uncertainty in Distributed Systems

Kedar Kulkarni, Libin Zhang, and Andreas A. Linninger*

Laboratory for Product and Process Design, Department of Chemical and Bioengineering, University of Illinois at Chicago, Chicago, Illinois 60607

This paper formulates and solves the problem of model identification and parameter uncertainty in plutonium storage systems. A systematic procedure helps to choose among alternative mathematical models with different properties and degrees of freedom. Rigorous metrics for measuring the statistical alignment between five different physical models and given experimental data are discussed. On the basis of those metrics, the most adequate model is identified and the optimal parameter values for the heat source and the heat transfer coefficients in the packed plutonium bed are calculated. Model confidence and parameter uncertainty are accurately quantified by confidence regions. Rigorous bounds on the uncertain parameter range are computed. Mathematical framework for a flexible design approach and risk minimization is introduced. Flexibility analysis approach is used to detect the worst case temperature hotspots in plutonium storage containers.

1. Introduction

The importance of accurate risk assessment in uncertain systems is nowhere easier to demonstrate than with nuclear wastes. The use of nuclear energy has resulted in plutonium contaminated material that needs to be stored safely for up to fifty years before final disposition. A large part of this plutonium waste is stored as plutonium dioxide in cylindrical stainless steel (SS316) containers. The plutonium dioxide bearing containers run the risk of accidental release of their contents as a result of overpressurization. A temperature increase may lead to an elevated fill-gas pressure and to the deflagration of the vessel. Therefore, exact quantification of chemical and thermal processes in plutonium dioxide bearing cylindrical containers is a necessary step toward understanding, predicting, and assessing risk and risk tolerance of this safety-critical application.

Previous studies on thermal transport in plutonium dioxide powder have focused on predicting the temperature rise or on developing different mechanistic approaches for estimating the effective thermal conductivity of the packed-powder bed. Recently, new experimental data were obtained to better quantify heat generation, conduction, and radiation in the packed plutonium dioxide bed. Quantification of these properties involves solving a class of problems called inversion problems. These problems typically constitute a least squares optimization problem with partial differential equation (PDE) constraints.

Inversion problems have been formulated and solved in diverse fields ranging from aerodynamic design to heat transfer. In the past, robust numerical methods for least squares optimization with PDE and/or algebraic constraints were developed. Recently, problem inversion for reaction mechanisms in metallopane vapor decomposition processes microelectronic applications was studied. Prior research performed data assimilation for air pollution models. Inverse problems have been used to prevent excessive aerodynamic heating of space vehicles during re-entry into the earth’s atmosphere. Inversion also permits shape optimization in aerodynamic design or reconstruction of tissue optical property maps from photon migration experiments.

In previous work, uncertainty in property predictions was neglected. In this article, we shall demonstrate that the undue reduction of model’s spatial dimensionality can lead to substantial model uncertainties. We will further advocate the need to account for distributed system geometry to properly interpret existing experimental data. To justify this claim, we address the problem of model uncertainty by considering several candidate models to describe heat generation, conduction, and radiation in the storage vessel. We propose to identify the transport and reaction properties of the system by solving a large-scale distributed inversion problem. Rigorous metrics will be used to quantify the adequacy of these models.

In addition to model uncertainty, parameter uncertainty is a source for underperformance risk. We will show that the proposed problem inversion also contains information about the confidence of the parameter estimates. To preserve the statistical quality of the model—data agreement, we calculate the confidence bounds for the estimated parameters using the variance-covariance information. In conjunction, model and parameter uncertainty metrics assess the risk of failure of the distributed system. A flexible design accounting for the uncertainty sources minimizes the risk of failure.

An outline of this paper is as follows: Section 2 introduces a systematic approach for identifying the best physicochemical model given limited experimental data. Section 3 proposes a mathematical programming formulation for determining unknown heat transfer and chemical reaction properties in the plutonium storage system. Section 4 will solve a flexibility problem to identify the worst-case scenario of the process in steady operation. The paper closes with the conclusions and the future research direction.

2. Model Uncertainty and Parameter Estimation

In classical process design, model and parameter uncertainty is handled by overdesigning the system. As a result, systems with uncertain parameters are made wider, bigger, and stronger! When designing distributed systems with uncertainty, it is not obvious how to adopt the arbitrary overdesign practice “to be on the safe side”. To overcome this deficiency, we propose a rigorous design methodology for the flexible design of distributed systems. The proposed method depicted in Figure 1 will incorporate accurate mathematical relations for model uncertainty currently lacking in existing deterministic approaches.

The proposed flexible design methodology for distributed systems has two stages: (i) model identification and parameter
estimation and (ii) flexible design. In stage one, experimental observations about a distributed system are the basis for selecting suitable first principles models. It will be shown how to select the most appropriate model given the experimental data. A large-scale kinetic and transport inversion problem (TKIP) will render bounds and probability distribution functions for the unknown transport properties.

Stage two of the probabilistic design aims at choosing the system’s design variables to maximize performance, while satisfying desired risk tolerances against safety constraints. Mathematical programming approaches to identify the critical worst-case scenario will be proposed.

2.1. Experimental Setting and Data Acquisition. Many biological systems, chemically reacting microdevices and safety-critical applications cannot be studied in simplified one-dimensional lab settings. Due to the hazard associated with the plutonium containers, the distributed cylindrical storage device has to be studied as a whole and cannot be decomposed into parts. Therefore, a distributed system analysis is necessary. Figure 2 depicts a typical cylindrical vessel to store plutonium and its main dimensions. The container is filled with plutonium dioxide powder. Due to radioactive decay, heat is being generated constantly causing the temperature in the vessel to rise. In addition, helium gas is generated in the nuclear reactions causing a pressure increase in the vessel. The temperature and pressure rise are critical factors for the long-term safety of the storage device.

New thermal experiments will help identify the unknown extent of heat generation and to quantify the radiative and conductive heat transfer inside the vessel. To accomplish this task, only six radial temperature measurements taken 57 mm from the bottom of the vessel are available. Using more extensive instrumentation to acquire more accurate reaction kinetic and transport information is difficult due to the extreme hazard associated with plutonium. Given these limited measurements, can we quantify the unknown transport and reaction mechanisms? How big is the risk of failure in the next fifty years? We propose transport and kinetic inversion to do parameter identification and risk analysis.

2.2. Transport and Kinetic Inversion Problems. The mathematical program of (1) and (2) is a nonlinear constrained optimization problem with partial differential equation (PDE) constraints. The solution to this TKIP will render the parameter set \( \theta \) composed of the unknown heat source, \( q \), the conduction, \( k \), and the wall heat transfer coefficients, \( U_1 \) and \( U_2 \). The objective minimizes the least squares error between the experimentally measured temperatures \( T_{\text{exp}} \) and the computed temperature profile \( T_{\text{model}} \). The temperature profile is calculated by the spatially distributed heat generation, conduction, and radiation model in cylindrical coordinates. The constraints (2) are partial differential equations (PDEs).

\[
\min_{\theta} [T_{\text{exp}} - T_{\text{model}}(\theta)]^T V^{-1} [T_{\text{exp}} - T_{\text{model}}(\theta)]; \quad \theta = \{ \alpha(k), q, U_1, U_2 \} \tag{1}
\]

subject to

\[
\nabla^2 (\alpha T) + q = 0 \tag{2}
\]

Here, \( \alpha \) is the effective thermal conductivity of the system. The covariance matrix \( V \) accounts for measurement errors. As a first approximation, we assume that all temperature measurements have the same accuracy; thus \( V \) equals the identity matrix. The importance of the covariance matrix will be illustrated further in section 3.

2.2.1. Model Selection and Model Uncertainty. Stationary heat transfer in cylindrical coordinates can be written as in eq. (3).

\[
\frac{\partial}{\partial r} \left[ \alpha \frac{\partial T}{\partial r} \right] + r \frac{\partial}{\partial z} \left[ \alpha \frac{\partial T}{\partial z} \right] + qr = 0 \tag{3}
\]

Here, the scalar temperature field \( T \) is a function of the radial distance \( r \) and the height \( z \). \( q \) is the unknown heat source due to nuclear activity, and \( \alpha \) is the effective thermal conductivity of the system accounting for conductive and radiative contribution. However, five alternative models (A–E) as in eqs (4)–(8) seem reasonable to describe the thermal transport in the plutonium dioxide system. Which model will provide the best fit for the experimental data leading to the smallest possible model uncertainty?

Model A: \( \frac{\partial}{\partial r} [\alpha \frac{\partial T}{\partial r}] + qr = 0; \quad \alpha = k_1 \) (constant) \tag{4}

Model B: \( \frac{\partial}{\partial r} [\alpha \frac{\partial T}{\partial r}] + qr = 0; \quad \alpha = k_1 + k_2 T^3 \) \tag{5}

Model C: \( \frac{\partial}{\partial r} [\alpha \frac{\partial T}{\partial r}] + r \frac{\partial}{\partial z} \left[ \alpha \frac{\partial T}{\partial z} \right] + qr = 0; \quad \alpha = k_1 \) (constant), \( U_1 = U_2 = U \) \tag{6}

Model D: \( \frac{\partial}{\partial r} [\alpha \frac{\partial T}{\partial r}] + r \frac{\partial}{\partial z} \left[ \alpha \frac{\partial T}{\partial z} \right] + qr = 0; \quad \alpha = k_1 + k_2 T^3, U_1 = U_2 = U \) \tag{7}

Model E: \( \frac{\partial}{\partial r} [\alpha \frac{\partial T}{\partial r}] + r \frac{\partial}{\partial z} \left[ \alpha \frac{\partial T}{\partial z} \right] + qr = 0; \quad \alpha = k_1 + k_2 T^3, U_1 \neq U_2 \) \tag{8}

The boundary conditions, neglecting the steel shell, enforced for models A–E imply zero heat flux at the center line and finite heat transfer across the mantle of the cylindrical vessel as summarized in Table 1. \( T_s \) is the cylinder’s surface temperature, \( T^o \) is the ambient temperature, \( U_1 \) is the heat transfer...
coefficient of the upper/lower lids, and $U_2$ is the heat transfer coefficient of the cylinder mantle.

The five models are compared on the basis of two criteria: (i) total least squares (TLSQ) error and (ii) F-test. 16 The total least squares error is represented by the objective function of eq (1). Low TLSQ errors suggest small model-data mismatch. However, models with more degrees of freedom (more model parameters) have smaller TLSQ errors without necessarily reflecting better the underlying physico-chemical process. Therefore, a second criterion is needed. The second criterion, the F-ratio, accounts also for number of parameters necessary to achieve the TLSQ.17

There are $n$ temperature measurements available; the number of estimated parameters is $p$. A low value of the TLSQ error coupled with a high value of the F-ratio will help identify the best model for the given experimental data set.

2.2.2. Discretization of Partial Differential Equation Constraints. The PDE constraints (2) are discretized using the finite volume method. 18,19 Figure 3 depicts the control volume in cylindrical coordinates. Accordingly, the heat balance over each control volume can be written in terms of five temperatures.

$$a_P T_P = a_N T_N + a_E T_E + a_W T_W + a_S T_S + b \quad (10)$$

where $a_P = a_N + a_E + a_W + a_S$

The derivation of the discretization equations can be found elsewhere. 18,19 Hence, eq (2) can be written as a set of algebraic equations $h$ in terms of the state variables, $T$, defined at the center of each finite volume, and the model parameters, $\theta$.

$$h(T, \theta) = 0 \quad (11)$$

More details of the finite volume approach are discussed in the Appendix.

2.3. Results: Parameter Inversion for Plutonium Storage.

2.3.1. Model Selection. For each model in eqs (4)-(8), problem (1) with discretized transport equations in eq (11) was solved using an SQP algorithm on a Pentium IV PC (3.06 GHz). When reasonable initial values were chosen for the parameters and the initial guess of the temperature field was set consistently, the inversion problems converged typically within 10 s. However, odd assignments of parameters and temperatures can cause convergence failure. The solutions of the optimal parameter sets are listed in Table 2.

By solving the distributed inversion problem, the unknown parameters, TLSQ error, and F-ratio can be calculated as shown.