Analysis and Reduction of Chemical Models under Uncertainty

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***Abstract*— We discuss the motivation, utility, and challenges associated with analysis and reduction of chemical models under uncertainty. We outline a mathematical formulation for this area of study, focusing on a probabilistic uncertainty framework and dynamical analysis/reduction methods for ordinary differential equation systems. We highlight our recent progress in this area, and discuss opportunities and challenges going forward.**

I. Introduction

Detailed chemical kinetic models for hydrocarbon fuels are typically complex, involving a large number of reactions and species. These elementary-step kinetic models are constructed to include chemical reaction pathways and intermediate/radical species that are judged to be important/relevant. Typically, each reaction involves at least 3 parameters, being the Arrhenius rate expression coefficients. These parameters are either measured experimentally, estimated from *ab-initio* quantum computations, or derived using rate rules from other rate coefficients. In any of these scenarios, these rate parameters are known only to within a certain degree of uncertainty. Broadly speaking, chemical kinetic models are characterized by degrees of both model and parametric uncertainty. In the present discussion, we focus primarily on parametric uncertainty.

Given the above, there is a need for chemical model analysis and reduction strategies to take these uncertainties into account, beside the need to accomodate a broad range of state values and operating conditions. This argument is driven by both accuracy and efficiency considerations. From an accuracy perspective, it is of interest to ensure that conclusions regarding the acceptable performance of a given reduced model are valid over the range of uncertainty in the parameters of the starting detailed model. Just like a reduced model is evaluated against the detailed model over a range of states, this ought to also span a degree of uncertainty in detailed model parameters. In principle, this translates to a higher computational cost in the analysis/reduction strategy. However, it can also translate to considerable savings if/when the choice of reduced model accuracy thresholds is informed by the degree of uncertainty in the detailed model parameters. It does not make sense to insist on reduced model accuracy requirements that are significantly tighter than the uncertainty in the detailed model predictions. The model reduction error budget ought to take both sources of error into account.

These observations motivate the development of model analysis/reduction methods that take uncertainties into account. In the present context, we give a brief outline of first steps along the path towards this goal, with a particular focus on dynamical analysis methods, specifically computational singular perturbation (CSP), relying on eignanalysis of the Jacobian of the chemical source term under parametric uncertainty.

II. Formulation

We consider chemical reaction processes in a spatially homogeneous mixture, where the time evolution of the system state vector is described by a system of ordinary differential equations (ODEs). In the deterministic context, one can employ a number of available strategies for model analysis and reduction. We consider specifically CSP analysis and associated model reduction strategies. This method has been used to provide a range of feasible simplified chemical models with roughly monotonous variation of error relative to a given detailed model [6,10,11].

Further, we employ a probabilistic uncertainty quantification (UQ) framework, where we represent uncertain quantities as random variables. We rely on spectral polynomial chaos (PC) representations of random variables [3,5]. Forward propagation of uncertainty in this context can be done using sampling-based *non-intrusive* methods [5]. This involves evaluation of the deterministic model for a set of parameter values, and assembling the resulting predictions and analysis results to arrive at uncertain outputs of interest. While this is certainly viable, it can be severely taxed by the curse of dimensionality when there is a large number of uncertain parameters. This is particularly true when eigenanalysis is required for CSP analysis with each sample. In the present context, we focus on *intrusive* PC UQ methods [2,5], where the uncertain/random chemical ODE system is transformed via Galerkin projection into a deterministic ODE system for the spectral PC expansion (PCE) coefficients of the uncertain system state. In this context, a *single* solution of this (larger) ODE system (system size grows with both dimensionality and order) suffices to provide the full uncertain solution picture.

Extending model analysis/reduction to the uncertain ODE context has received some attention in recent literuare. In [4], small parametric perturbations were considered in the Proper Orthogonal Decomposition (POD) context. Sonday *et al.* [9] studied the eigenstructure of the Galerkin ODE system Jacobian of model uncertain/random ODE systems. Salloum *et al.* [8] provided a joint CSP-UQ study of a model uncertain ODE system with approximate characterization of the eigenstructure of the the Galerkin ODE system Jacobian.

III. Stochastic Eigenvalue Problem

In [1,9], we established a number of facts regarding the eigenstructure of the Galerkin ODE system Jacobian and its relationship to the statistics of eigenvalues/eigenvectors, hence dynamics, of the original random ODE system. Results indicate that, in the limit of infinite PC order, the eigenvalues of the Galerkin system tend to the distribution of eigenvalues of the original system. Further, for finite order, these two representations of the uncertain eigenvalues are close, and convergence with PC order, in the sense of measures, can be observed. Moreover, we outlined a constructive strategy for the eigenvectors of the Galerkin system Jacobian, that approximates that of the sampled eigenvectors of the original system. Accordingly, it is clear that there is a path towards learning the dynamical landscape of an uncertain chemical system by analysis of the eigenstructure of the corresponding Galerkin Jacobian.

IV. CSP Analysis of Uncertain ODEs

Given the above, we now have the beginnings of a strategy towards CSP analysis of uncertain chemical ODEs, and associated model reduction strategies. Further work is required to extend the various elements of the deterministic analysis/reduction method to the stochastic setting. Thus, while we have means of evaluating the random system eigenstructure, we need to establish robust methods for analyzing the random CSP vectors/co-vectors and associated modal expansion, to arrive at meaningful characterizations of uncertain low-dimensional manifold structure, fast and slow subspaces, classification of major/minor/radical spsecies, and importance and participation indices. We note for example, that uncertainty in fast processes can translate into uncertainty in slow manifold structure, while uncertainty in slow processes would primarily influence the evolution of the system along a deterministic manifold. Further, uncertain/random CSP Indices have to be compared to each other, and to specified thresholds, employing statistical measures of distance. Similarly, in the model reduction context, comparisons between model predictions, and estimates of error measures, have to rely on probabilistic analogues to the original deterministic procedures.

From another perspective, the Galerkin-projected ODE system, whose purpose is to allow the exploration of the dynamics of the original uncertain ODE system, is itself a candidate for CSP analysis and model reduction, in order to facilitate its time-integration. Given high-order and dimensionality, this system can become quite large and computationally expensive to solve/analyze. Its time integration is challenging, both from accuracy and stability viewpoints [7,12]. Accordingly, there are opportunities for using CSP, or other dynamical analysis methods, along with associated model reduction strategies, in this context.

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VI. References

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