MODEL REDUCTION IN REACTING FLOW

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ABSTRACT

In this abstract, I present a brief overview of model analysis and reduction methods as applied to chemically reacting flows. I highlight specifically the computational singular perturbation method for analysis of chemical systems, and associated model reduction strategies. I outline our use of this method for analysis and model reduction in a range of chemically reacting flow systems. I also touch on challenges and opportunities associated with the use of CSP for analysis and model reduction in both macroscale and stochastic chemical systems, as well in uncertain chemical systems in general.

INTRODUCTION

Chemically reacting flows exhibit a range of computational challenges associated with their large range of length and time scales, as well as underlying chemical model complexity. In particular, chemical model complexity leads to range of complications. The large number of chemical species involved in detailed elementary reaction-step mechanisms, for even the simplest hydrocarbon fuels, leads to an associated large number of partial differential equations that need to be handled in a reacting flow computation. Further, the large number of reactions results in complex chemical source terms with a significant associated computational cost. Moreover, strong nonlinearity and fast time scales, in chemical rate expressions, lead to the formation of sharp reaction fronts and high spatial gradients of species concentration profiles. The resulting range of length scales necessitates fine mesh discretizations with associated computational loads. Similarly, the resulting range of time scales leads to extreme levels of temporal stiffness, and the need for specialized stable and accurate time integration strategies.

Given this set of difficulties, it is clear that chemical model reduction, to say nothing of efficient turbulence modeling requirements, is a necessary step towards enabling the utilization of chemically reacting flow computations in studies of practical combustion systems. In the following I will present brief highlights of existing work in chemical model reduction, with a more focused discussion of our work, followed by an outline of interesting challenges and opportunities in the field.

BACKGROUND

There has been extensive development of chemical model reduction methods targeting detailed hydrocarbon kinetics over a number of decades [1, 2]. Available techniques include lumping methods [2], sensitivity and principal component analyis [3], and elementary reaction flux based methods [4–6]. Given the strong stiffness and nonlinearity of typical chemical mechanisms for oxidation of hydrocarbon fuels, however, it is important that comparisons of sensitivity and reaction fluxes be done with care. In particular, brief dominance of reaction processes with fast time scales can obscure the importance of slower reactions that can ultimately be of much higher relevance. Therefore, ideally, chemical model analysis ought to be done with proper attention to the dynamical landscape of the associated equation system. Time-scale analysis has been accordingly used from an early time, starting with the quasi-steady state approximation (QSSA) [2, 7–10]. The partial equilibrium approximation (PEA) [10–13] has also been used to identify effective algebraic constraints relevant in model reduction. A detailed analysis of the conditions under which QSSA and PEA are valid has been recently presented [14].

The computational singular perturbation (CSP) method [10, 15–17] has also been used successfully for analysis and reduction of a range of chemical systems [18–25]. The underlying stiffness and separation of time scales in chemical systems results in dynamics that are characterized by distinct fast and slow subspaces. These systems exhibit slow invariant manifolds defined by the equilibration of fast processes. Given an arbitrary initial state, the system evolves quickly towards a nearby slow manifold according to the fast time scales, and then evolves along the manifold according to the slow processes. CSP relies on eigenanalysis of the Jacobian of the equation system to allow analysis of the dynamics separately in the fast or slow subspaces, identifying important processes [26–30]. This analysis has provided means for development of a chemical model reduction strategy that retains important slow *and* fast reactions, and associated species. Further, the identification of the fast and slow directions enabled the development of time integration strategies that eliminate the fast time scales and enable time integration along the slow manifold with large explicit time steps [31, 32]. Identification of slow invariant manifolds, and their use for efficient time integration of chemical systems, is also at the core of the intrinsic low dimensional manifold (ILDM) method [33]. Similarly, the rate-controlled constrained-equilibrium (RCCE) technique [34, 35], based on the Second Law of thermodynamics, describes the evolution of a chemical system along rate-controlled constrained equilibrium paths. The invariant constrained equilibrium edge preimage curve method (ICE-PIC) [36] is based on RCCE, and makes use of trajectory-generated low-dimensional manifolds.

CHALLENGES AND OPPORTUNITIES

We have used dynamical analysis methods, specifically CSP, for the analysis of laminar flame computational databases and chemical model reduction for a number of hydrocarbon fuels including methane, propane, n-heptane, and jet fuel. This included both 1D flames as well as edge and jet flames. We identified and studied the dynamical structure of these flames under different operating conditions, providing information on the relative importance of specific reaction pathways and transport processes in the dynamical response of species concentrations and temperature in specific flame layers. We have used these results to arrive at an understanding of flame structure and dynamics, identifying dominant processes that contribute to either the time-dynamics or the underlying partial-equilibria. We have also used them to identify simplified chemical mechanisms under a range of operating conditions, and in different flame locations. Looking forward, there are a number of challenges in the utilization of CSP for model reduction in chemical systems, along with opportunities for further investigation.

To begin with, it is worth noting that the existing CSP-based model simplification strategy [19,20] is global over the database of states, and therefore not necessarily optimal at any given state. Typical means of generation of such data bases have included computations of ignition using the detailed mechanism over a range of initial conditions. Thus, the database covers a range of behaviors from slow preheat-phase reactions, through fast ignition, and decay to equilibrium, over a range of initial states. In principle, tabulation of simplified mechanisms for ranges of state values would provide a more optimal construction, however one then has to deal with the change in the size of the state vector in time/space [5]. Similarly, CSP-based model reduction *on the fly*, based on the projection of the governing equations onto the slow manifold, is local and has some optimality properties, but is computationally prohibitive unless suitable solution tabulation techniques are employed [25]. Another point to note is that CSP-based model reduction does not *explicitly* enforce a specific *error* measure on the state space trajectories. Rather, it attains control on the state error employing an error threshold on the importance index set, *i.e.* essentially on the projected chemical source terms. However, there is no guarantee that any specific resulting simplified N-species mechanism has the smallest state-error, relative to the detailed model, among all N-species mechanisms. Further, given nonlinearity, there are no explicit guarantees on the accuracy of computations using the simplified model *outside* the range of database of states, with this or other reduction schemes. Finally, note that since, by design, the simplified mechanism has both fast *and* slow species, this mechanism does retain some stiffness, albeit typically to a smaller degree than the original detailed mechanism. Clearly, there is significant room for improvement of the present CSP-based model reduction methodology.

From another perspective, there is a significant need for extension of dynamical analysis methods, such as CSP, to the realm of stochastic chemical systems [37]. Stochastic effects are non-negligible in chemical processes at the microscale, *e.g.* in a biological cell or at a catalytic surface nano-scale feature, where molecular counts are small. Accordingly, depending on the range of molecule counts, these systems are modeled using jump Markov processes or stochastic differential equations [38–42]. In this broad context, there is a need for well-founded practical definitions of stochastic "slow" manifolds and fast/slow subspaces. With proper setup of the requisite stochastic dynamical analysis framework, one can then address analysis of stiff stochastic chemical system dynamics and associated model reduction strategies. While there has been work in this area [43–51], much more remains to be done.

Finally, the area of model reduction under uncertainty is of particular interest. Allowing for model error and/or noise in empirical measurements, it is clear that models generally involve some degree of uncertainty in their parameters. Accordingly, there is a need to account for this uncertainty in the model reduction context. In particular, user-specified error thresholds on reduced models ought to be informed by the degree of uncertainty in the starting detailed model predictions. Thus, for example, it does not seem justifiable to insist on reduced model errors that are orders of magnitude smaller than the uncertainty in the detailed model predictions. Rather, it is natural to consider all relevant sources of error and uncertainty in the model-reduction error budget. On the other hand, there are many challenges in this area related to the reformulation of dynamical analysis and model reduction techniques to account for uncertainty. Here again, the underlying dynamical objects, manifolds and fast/slow subspaces, need to be properly defined and represented before adequate model reduction strategies can be arrived at. Further, the very notion of *error* in a reduced model has to be revised, when both reduced and detailed predictions are uncertain. There has been some work with relevance in this context [52–54], but there is need for significantl more development.

CLOSURE

I have tried in the above to give a brief overview of methods for analysis and model reduction in reacting flow. I covered with some detail our experience with the use of CSP in this context. I also presented brief highlights of challenges and opportunities going forward in the context of both continuum and atomistic chemical systems, and the need for addressing uncertainties in both analysis and reduction of chemical systems. These challenges are of relevance in a number of applications, providing opportunities for advancing the state of the art and impacting scientific and technical progress.

ACKNOWLEDGMENT

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