Adaptive chemical model reduction

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Abstract. We briefly review various chemical model reduction strategies with application in reacting flow computations. We focus on systematic methods that enable automated model reduction. We highlight the specific advantages of computational singular perturbation (CSP) analysis. We outline a novel implementation of CSP, with adaptive tabulation of the basis vectors, that enables fast identification of the reduced chemical model at any point in the chemical phase space, and efficient integration of the chemical system. We describe this implementation in the context of a particular model problem that exhibits stiffness typical of chemical kinetic systems.

1. Introduction

Chemical model reduction strategies have been of significant interest in combustion research over the past few decades, and there is a wealth of associated literature [1,2]. These techniques continue to be of high relevance today, despite the phenomenal rise in available computational power, because of the similarly formidable increase in complexity of known detailed chemical models for fuels.

One early reduction strategy involves order reduction by 'lumping' of variables. The general character of the approach involves the transformation of the reactant vector to a lower dimensional vector of pseudospecies [2]. However, acceptable lumping schemes become harder to identify with increasing chemical complexity. Other approaches have been used to eliminate unimportant species, e.g. based on sensitivity and/or principal component analysis [3]. Alternate techniques eliminate unimportant reactions based on elementary reaction fluxes [4–6]. These methods have been found to be effective in handling various chemical systems. However, one challenge with reaction flux approaches is that a "small" reaction flux is not in general an indication that a reaction step can be discarded. Thus care has to be taken to avoid eliminating important steps.

One of the most commonly used reduction methods, based on time-scale analysis, employs the quasi-steady state approximation (QSSA) [2, 7, 8]. QSSA identifies radical species whose chemical source term is dominated by nearly-equal fast and opposed reactions, such that the net rate of change in their concentration in time is "small". A significant challenge with QSSA
is precisely the steady-state species selection, which can be shown to lead to wrong answers under some conditions [9]. In recent work, sensitivity analysis has been used to aid in the identification of the steady state species [10]. Other QSSA challenges have also been discussed in the literature [2].

Another approach, the partial equilibrium approximation [11, 12], identifies elementary reactions whose forward and reverse rates are nearly equal. By setting them strictly equal, one arrives at corresponding algebraic constraints among the participating species. In this context, the identification of the species whose concentration is to be found from the algebraic constraint is not uniquely established [9]. Moreover, the formal accuracy of the procedure requires a specific set of of constraints on the algebraic relationships among the elementary rates [13].

Yet another strategy, that has been developed over the past couple of decades, is computational singular perturbation (CSP) analysis [9,14–16]. CSP is a general formal procedure both for analysis and reduction of chemical systems. It enables automatic identification of fast and slow reaction processes, the elimination of fast-exhausted and dormant modes, and the identification of the slow modes that drive the slow evolution of the chemical system. The method relies on the identification of a suitable set of basis vectors that enable the decoupling of fast and slow processes. A leading-order approximation of these vectors is provided by the eigenvectors of the Jacobian of the chemical source term. CSP provides a refinement procedure that iteratively adjusts the basis vectors to further improve the decoupling of the fast and slow subspaces. Moreover, CSP has been used as a basis for an efficient time integration strategy [17] that filters out the fast time scales enabling large time-step explicit integration of the slow modes, with a requisite projection to correct for the contribution from the fast modes.

The intrinsic low dimensional manifold (ILDM) method [18] uses the eigenvectors of the Jacobian to enable the above decoupling of fast and slow processes, as is done in the first-order CSP implementation. However, the typical utilization of ILDM, as coupled with tabulation, has relied on the assumption that the low-dimensional manifold has the same dimension everywhere in the reacting flow system, an assumption that has been shown to be grossly inaccurate by the analysis of premixed methane-air flame-vortex interactions with CSP [19].

The low dimensional slow manifold ideas behind CSP and ILDM are also at the basis of the rate-controlled constrained-equilibrium (RCCE) technique [20, 21]. RCCE asserts that a stiff chemical system relaxes toward equilibrium through a sequence of rate-controlled constrained-equilibrium states. These states can be determined by entropy maximization, subject to instantaneous values of slowly changing constraints imposed by a relatively small number of rate-controlling reactions. However, there is no unique or automated choice of the RCCE constraints in general. Yet the specific choice does seem to affect the computed time histories of species [1].

In the present work, we focus on the use of CSP as a tool for efficient integration of chemical models. In particular, we use it to automatically identify the underlying low-dimensional manifold at any location in chemical composition space (CCS), and to use this information to efficiently integrate the chemical system [17]. However, given the cost of the CSP analysis, such a straightforward implementation is clearly not feasible. Instead, we use adaptive tabulation of the CSP basis vectors so that they can be reused whenever the system lands in a previously visited (and tabulated) hypercube in the CCS. In this manner we arrive at an effective adaptive chemistry construction. The implementation of adaptive chemical model reduction using reaction flux analyses has already been demonstrated [5], and other adaptive strategies have also been outlined [22]. However, this is the first such implementation in association with the formal manifold identification and efficient time integration enabled by CSP.

There are two commonly used solution tabulation strategies in the combustion literature. One method, "in situ adaptive tabulation" (ISAT) [23] is based on the use of Taylor series for evaluation of the solution in the vicinity of previously visited points in the CCS. On the other hand, "piecewise reusable implementation of solution mapping" (PRISM) [24, 25] uses
polynomial regression to construct a response surface for the solution within each hypercube in the CCS. In the present context, we use PRISM to tabulate the CSP basis vectors. One of the challenges with PRISM is the rise in cost of response surface construction with increased dimensionality of the chemical system. This is adequately addressed in the present context, as CSP effectively uses a reduced order model valid within each hypercube, thereby allowing lower dimensionality tabulation. Thus, the present approach couples CSP and PRISM to arrive at efficient adaptive integration of chemical models.

In the following, we illustrate this construction in the context of a 4D ordinary differential equation (ODE) model problem, and examine the integration of the system using both the detailed and CSP-PRISM-reduced constructions. The fundamentals of CSP are well documented in the above cited literature, and are therefore not discussed here.

2. Model Problem
We define a 4D model ODE system, which we integrate using the CSP–projection scheme of Valorani and Goussis [17]. The system is given by:

\[
\frac{dY_i}{dt} = \frac{1}{\epsilon^{N-i}} \left( -Y_i + \frac{Y_{i+1}}{1 + Y_{i+1}} \right) - \sum_{j=i+1}^{j=N} \frac{Y_{j+1}}{(1 + Y_{j+1})^2}, \quad i = 1, \ldots, N; \quad \epsilon \ll 1
\]  

where \( N = 4 \). By design, this system exhibits the time scales: \( \{\tau_i\}_{i=1}^{N} = \epsilon^{N-i} \). When the \( i^{th} \) mode becomes exhausted, the CSP manifold is defined (to leading order) by \( Y_i - Y_{i+1}/(1 + Y_{i+1}) = 0 \). Let us consider the case where \( \{Y_i\}_{i=1}^{4} = Y^{*} \) is in a range where the number of exhausted modes is \( M = 1 \). Assuming a partitioning of the chemical composition space, we tabulate the CSP basis vectors by taking samples in the hypercube containing \( Y^{*} \).

This hypercube is expanded only in the 3D space of \( \{Y_2, Y_3, Y_4\} \), while \( Y_1 \) is evaluated from \( Y_1 = Y_2/(1 + Y_2) \). Obviously, if \( M = 2 \), we can use a 2D hypercube (expanding only \( \{Y_3, Y_4\} \)) and an analogous expression to evaluate \( Y_2 \). In any case, given the chosen dimension of the hypercube, a response surface is constructed for each component of the tensor quantities evaluated from the time scales and CSP basis vectors corresponding to the fast/exhausted modes, and used in the time integration procedure [17]. The construction of the response surface in PRISM relies on an optimal choice of design points. Table 1 presents a 3-parameter 18-run \( d \)-optimal design [26]. We construct a single hypercube tabulation for the \( M = 1 \) case, where \( X_i = \log(Y_i/Y_i^c)/\log f, \quad i = 2, 3, 4 \) (a log–scaled design); \( Y_i^c \) is the center point of the hypercube; the design factor is \( f = 10 \), which corresponds to a very large hypercube; and the dimension of \( X_1 \) is not expanded.

| run # | \( X_2 \) | \( X_3 \) | \( X_4 \) | run # | \( X_2 \) | \( X_3 \) | \( X_4 \) | run # | \( X_2 \) | \( X_3 \) | \( X_4 \) |
|-------|---------|---------|---------|-------|---------|---------|---------|-------|---------|---------|---------|
| 1     | 0       | -1      | -1      | 7     | -1      | 1       | -1      | 13    | 1       | 1       | 0       |
| 2     | 1       | -1      | 1       | 8     | -1      | 0       | 1       | 14    | 0       | 1       | 1       |
| 3     | 1       | -1      | 0       | 9     | 1       | 1       | -1      | 15    | 1       | 0       | 1       |
| 4     | -1      | -1      | 1       | 10    | 1       | -1      | -1      | 16    | 0       | -1      | 1       |
| 5     | 1       | 1       | 1       | 11    | 1       | 0       | -1      | 17    | -1      | 1       | 1       |
| 6     | -1      | -1      | -1      | 12    | -1      | -1      | 0       | 18    | 0       | 0       | 0       |

Table 1. A three-variable fractional factorial \( d \)-optimal design with 18 runs

We now consider the tabulation of the CSP tensors using the CSP–PRISM technique deployed in a reduced–dimension hypercube. We construct second-order polynomial curve fits using the standard least-squares method, with the sampling points shown in Table 1, for components
of the CSP tensors; and store the polynomial coefficients. Here, the time scales $\tau_i$ need not be tabulated since they are constant. In the time integration of the model problem, when the variables fall within the range of validity of a hypercube tabulation, the CSP tensors are reconstructed using these polynomials, which can be evaluated at a CPU cost substantially lower than the direct CSP analysis, and used in the time integration scheme. The tabulation can also be pre-constructed to provide additional savings in CPU time, as was done in the present work.

The single hypercube tabulation thus created is valid in the time range $0.001 \leq t \leq 2.34$ for $\epsilon = 10^{-2}$ and $\Delta t/\tau_{M+1} = 0.05$. We plot in Fig. 1 the solution from $t = 0$ to $t = 5$ obtained with the original CSP projection scheme [17]. The results highlight the four operative time-scales $\{\tau_i\}_{i=1}^4$ in the problem, with $Y_i$ decaying slower than $Y_{i-1}$ for $i = 2, 3, 4$. The solution in $0.001 \leq t \leq 2.34$, computed with the tabulated CSP tensors, and the relative difference between the results obtained with the two schemes, are also reported in the figure. It can be seen that the solution obtained exhibits good accuracy, and that the tabulation-based solution at the end point $t = 2.34$ agrees with that obtained with the full CSP-projection scheme to within $10^{-5}$, which is a very promising for a computation performed with a single large hypercube.

**Figure 1.** Comparison of the solutions of the model problem defined in Eqn. 1 obtained with the original time integration scheme, with the CSP analysis carried out at every time step for $0 < t < 5$, and the one obtained with the CSP-PRISM strategy i.e. with CSP tensors tabulated using the $d$--optimal design points found in the reduced--dimension hypercube (without any CSP analysis) during time integration in the duration $0.001 \leq t \leq 2.34$. The parameters defining the model problem and the time integration procedure are: $\epsilon = 10^{-2}$ and $\Delta t/\tau_{M+1} = 0.05$. We plot in Fig. 1 the solution from $t = 0$ to $t = 5$ obtained with the original CSP projection scheme [17]. The results highlight the four operative time-scales $\{\tau_i\}_{i=1}^4$ in the problem, with $Y_i$ decaying slower than $Y_{i-1}$ for $i = 2, 3, 4$. The solution in $0.001 \leq t \leq 2.34$, computed with the tabulated CSP tensors, and the relative difference between the results obtained with the two schemes, are also reported in the figure. It can be seen that the solution obtained exhibits good accuracy, and that the tabulation-based solution at the end point $t = 2.34$ agrees with that obtained with the full CSP-projection scheme to within $10^{-5}$, which is a very promising for a computation performed with a single large hypercube.
3. Conclusions
We have presented a brief overview of chemical model reduction strategies, and outlined the specific advantages of CSP for analysis and reduction of chemical models. We have demonstrated that the combination of the CSP-projection method and PRISM offers a new way to construct an adaptive reduced-order model for a stiff dynamical system. CSP allows a reduction in dimensionality of the hypercubes in the PRISM tabulation procedure. By constructing tabulations for the requisite CSP tensors, we arrive at an efficient explicit time integration construction based on the CSP solver in [17]. The test performed on a stiff model problem demonstrated the feasibility of this strategy. The results indicate that a high level of accuracy is readily achievable. Future work will concentrate on the construction of such an adaptive model for chemical kinetic systems that are of direct relevance to combustion.

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