Adaptive Chemistry Computations of Reacting Flow

J.M. Ortega¹, H.N. Najm¹,², J. Ray¹, M. Valorani³, D.A. Goussis¹, and M. Frenklach⁵

¹ Sandia National Laboratories, Livermore, CA, USA
² Università di Roma “La Sapienza”, Rome, Italy
³ National Technical University of Athens, Athens, Greece
⁵ Univ. of California at Berkeley & Lawrence Berkeley Nat. Lab., Berkeley, CA, USA

E-mail: hnnajm@sandia.gov

Abstract. We present a new tabulation strategy for the numerical integration of chemical reacting flow processes on the basis of a non-stiff system of equations. Both the tabulation and the identification of the non-stiff system are adaptive and are based on the Computational Singular Perturbation (CSP) method. The tabulation strategy is implemented in order to store and reuse the CSP quantities required for the construction of the non-stiff model. In this paper we describe a particular feature of this algorithm, the “homogeneous correction”, that allows for an accurate and efficient identification of the manifold on which the solution moves according to the slow time scales. The improved efficiency in constructing the slow model and simulating the system dynamics along the manifold during run-time calculations is demonstrated.

1. Background
The dynamics of chemical kinetic systems exhibit a wide range of time scales. Computational Singular Perturbation (CSP) [1, 2] analysis facilitates the study of chemical systems by decomposing the system dynamics into fast and slow dynamics, where the fast dynamics constrain the system evolution within a low-dimensional manifold, usually referred to as the Slow Invariant Manifold (SIM), and the slow dynamics drive the evolution of the system along the manifold. The key advantage of CSP-based reduction strategies is the ability to carry out the fast/slow decomposition automatically allowing for the replacement of a stiff system of large dimension with one which is non-stiff and involves a reduced number of evolution/differential equations. The numerical integration of the reduced, non-stiff model can be carried out by resorting to an explicit time-scale split integration algorithm [3]. However, a straightforward implementation of CSP is not affordable when dealing with systems of large dimension because of the high computational cost associated with the CSP analysis. One way to reduce this cost is to implement a tabulation strategy to store and reuse the manifold information generated by CSP.

Our tabulation borrows from the “Piecewise Reusable Implementation of Solution Mapping” (PRISM) [4] technique the utilization of hypercubes in the Chemical Configuration Space (CCS) and the polynomial regression of response surfaces, and adapts them to address the specific challenges of CSP, while exploiting the reduction of dimensionality offered by CSP. In contrast to conventional PRISM implementations, where the solution is mapped against the full state...
vector, we seek to store and reuse information of a lower, if existing, \((N - M)\)-dimensional surface within the \(N\)-dimensional hypercube. Here, \(N\) is the number of unknowns in the state vector \(y\) and \(M\) is the number of fast time scales that, at some point in the CCS, are found “exhausted”, and therefore not contributing to the (slow) dynamics of the system. Specifically, we tabulate the CSP basis vectors/covectors \(\{a_r, b^r\}_{r=1}^M\) spanning the \(M\)-dimensional fast subdomain. This choice is based on the fact that the eigenvalue analysis of the Jacobian of the source term and the refinements needed for the associated calculations of \(\{a_r, b^r\}\) are computationally expensive. Moreover, the CSP vectors/covectors are at the core of the computation of important quantities such as the projection matrix \(Q_s = I - \sum_{r=1}^M a_r b^r\) or the radical correction in the CSP integrator [3].

The implementation of the tabulation consists of building local –low-order– polynomial response surfaces of the elements of the first \(M\) columns of \(a\) and rows of \(b\) as a function of the \(N - M\) major species. The effectiveness and feasibility of the approach relies ultimately on the ability to identify the value \(M\) and the \(N - M\) variables in order to characterize the SIM accurately when the table is being constructed.

In this paper we show how the concept of the CSP homogeneous (radical) correction [5, 6] can be used to project any state vector picked at random in an \(N\)-dimensional hypercube onto a neighborhood of an \((N - M)\)-dimensional SIM. The action of the homogeneous correction is to move the state vector along the \(M\) fast directions and bring it closer to the SIM. A measure of the approach to the SIM is the magnitude of the amplitudes of the \(M\) fastest modes, which decay exponentially as the state vector approaches the SIM. We will adopt the definition of the homogeneous correction used in [3], that is:

\[
\delta y = - \sum_{m,n=1}^{M} a_m \tau^m_n f^n
\]

where \(\delta y\) is the displacement of the state vector in the fast subspace produced by non-vanished fast mode amplitudes \(\{f^n = b^r \cdot g^r\}_{r=1}^M\), with \(g\) denoting the right hand side (RHS) of the kinetic model. The matrix \(\tau^m_n\) is the inverse of \(\lambda^m_n\) where

\[
\lambda^m_n = \left( \frac{db^m}{dt} + b^m J \right) a_n
\]

and \(J\) is the Jacobian matrix.

During the tabulation stage, given a starting point inside a hypercube, repeated application of the homogeneous correction is used to attract the system state to the manifold along the fast directions. We compute \(M\) inside the hypercube by monitoring when \(\delta y\), resulting from each application of the homogeneous correction, is sufficiently small such that the state is “close” to the manifold. The selection of the proper size of the hypercube is carried out adaptively during the table construction, although this issue is not discussed in this paper. By projecting the state vector onto the SIM during the integration stage, substantial computational savings can be achieved, since the exhausted fast scales are eliminated and the number of time steps needed to accurately integrate the slow dynamics of the original system of differential equations is significantly smaller.

2. CSP “homogeneous correction”

Typically, a state vector \(y\) landing in a hypercube where a SIM of unknown dimension \((N - M)\) exists, is found significantly off the SIM, causing the \(M\) fastest time scales to be active and forcing the state vector to move towards the SIM. Thus, one way to identify the SIM dimension within a hypercube is to compute trajectories starting from different points in the hypercube and
to monitor how many fast amplitudes are vanished when the trajectories leave the hypercube. However, computing the trajectories soon becomes prohibitively expensive.

As an alternate strategy, we choose to project the state vector onto a SIM of presumed dimension \((N - M)\) by resorting to the homogeneous correction, Eq.(1), which allows skipping the computation of the fast dynamics. We take the largest value of \(M\) for which the projected state vector lands inside the same hypercube as the SIM dimension within a hypercube.

Repeated application of the homogeneous correction, for an assumed value of \(M\), can bring the state vector arbitrarily close to the nearest \((N - M)\)-D SIM. The homogeneous correction mostly affects the variables identified as CSP radicals, whereas the remaining (major) species are relatively unaffected. The ratios \(\{dy_i/y_i\}_{i=1}^N\) monitor the relative changes of each solution component after each correction, which can be used as a stopping criterion for the homogeneous correction iterates.

Clearly, the projected state vector on the SIM is not the same as the point that the integration trajectory would reach, starting from the same initial conditions. This only occurs if the ratio \(\tau_{\text{fast}}/\tau_{\text{slow}} = 0\), otherwise the two points differ by an amount which is a function of the time, \(\Delta t\), elapsed to reach the SIM from the starting point. In the next section, we show with an example that this difference can be made small without affecting significantly the accuracy of the integration of the slow dynamics, while the computation of the large number of very small integration time steps required to describe the fast approach to the SIM is avoided.

### 3. Example: a 3 species kinetics problem

To illustrate the application of the homogeneous correction, we consider the 3-species kinetics problem analyzed in [2]. The right hand side of this model problem reads:

\[
g = \begin{bmatrix}
-\frac{5y_1}{\epsilon} - \frac{y_1y_2}{\epsilon} + y_2y_3 + \frac{5y_3}{\epsilon} + y_1 - y_1 \\
10\frac{y_2}{\epsilon} - \frac{y_1y_2}{\epsilon} - y_2y_3 - 10\frac{y_2}{\epsilon} + \frac{y_3}{\epsilon} + y_1 \\
\frac{y_1}{\epsilon} - y_2y_3 - \frac{y_3}{\epsilon} + y_1
\end{bmatrix}
\]  

(3)

where \(\epsilon\) is a small parameter controlling the stiffness of the system. The solution trajectories of this system are asymptotically attracted towards a 1-D SIM, a line in a 3-D phase space.

Consider the hypercube defined by the vertex \([y_1, y_2, y_3] = [0.875, 0.875, 0.875]\) and edge lengths \([\delta y_1, \delta y_2, \delta y_3] = [0.125, 0.125, 0.125]\), plotted in Fig. 1. Small black dots represent values of the state vector selected randomly within the limits of the hypercube. For all these “experimental design” points we computed two successive homogeneous corrections for both \(M = 1\) and \(M = 2\). The homogeneous correction in Eq.(1) is calculated using the eigenvectors of the Jacobian of the RHS as CSP vectors. Table 1 shows the state vector after one and two corrections for the same initial point, the relative change of each component of the state vector, and the magnitude of the fast modal amplitudes. The relative changes between initial and final points of the second correction are significantly smaller than those due to the first correction, indicating convergence towards the SIM as also confirmed by the vanishing values of the fast modal amplitudes.

We can observe that, when the homogeneous correction is calculated with \(M = 1\), the projected points are in the vicinity of a 2D surface (red points in Fig. 1). This surface is a 2-D SIM where the first modal amplitude \(f_1 \approx 0\). Similarly, when \(M = 2\) the corrections take the experimental design points to the vicinity of a 1-D SIM, the intersection of two surfaces at which \((f_1 \approx 0 \cap f_2 \approx 0)\) (green points in Fig. 1). The trajectory (black square symbols) found by prescribing constant time intervals starting from the initial point in Table 1, shows the behavior of the state vector under the influence of fast scales, described in the previous section.
and in [2]. The trajectory exhibits fast initial decay towards the (red) 2D manifold, followed by slow evolution along this surface towards the (green) 1D manifold, and proceeding finally with continued slow evolution along this line. The blue circles in Fig. 1 represent the final values of the state vector after the two homogeneous corrections computed with $M = 1$ and $M = 2$. One of these points ($M = 1$) lies on the (red) 2D manifold, close to the point where the fast trajectory lands on this manifold. The other one ($M = 2$) lies on the (green) 1D manifold, close to the point where the trajectory along the 2D manifold meets the 1D manifold.

Figure 2 shows the time evolution of the three species using the CSP integrator [3] and 4th-order Runge-Kutta (black line). We compare these results with those obtained after 1 (red) and 2 (green) homogeneous corrections computed with $M = 2$ (Table 1) followed by the same CSP integration. We note the high accuracy of the integration after a short initial period. The number of integration steps with CSP starting from the initial point until $\log_{10}(\text{time}) = -0.75$ is 170. After one homogeneous correction, most of the short integration steps of the rapid transit period are skipped and the number of steps drops to 65. Finally, with 2 consecutive homogeneous corrections, the modal amplitudes become negligible indicating close proximity to the 1D manifold. Under these conditions, CSP is especially effective in filtering out the fast scales, and only 1 time step is needed to integrate the system of ODEs with comparable accuracy.

### Table 1.

| $M = 1$ | $y_1$ | $y_2$ | $y_3$ | $dy_1/y_1$ | $dy_2/y_2$ | $dy_3/y_3$ | $f^1$ | $f^2$ |
|---------|-------|-------|-------|-------------|-------------|-------------|-------|-------|
| Initial | 0.97003 | 0.92696 | 0.99514 | -128.35270 |             |             |       |       |
| 1 hc    | 0.94726 | 0.97593 | 0.99401 | 0.02346     | -0.05283    | 0.00113     | 2.41797 |       |
| 2 hc    | 0.94768 | 0.97504 | 0.99403 | -0.00044    | 0.00091     | -0.00002    | 0.00454 |       |

| $M = 2$ | $y_1$ | $y_2$ | $y_3$ | $dy_1/y_1$ | $dy_2/y_2$ | $dy_3/y_3$ | $f^1$ | $f^2$ |
|---------|-------|-------|-------|-------------|-------------|-------------|-------|-------|
| Initial | 0.97003 | 0.92696 | 0.99514 | -128.35269 | -11.47343   |             |       |       |
| 1 hc    | 0.97782 | 0.99093 | 0.96854 | 0.00091     | 0.00160     | 0.00010     | 4.59536 | -0.06144 |
| 2 hc    | 0.97876 | 0.98934 | 0.96844 | -0.00096    | 0.00160     | 0.00010     | 0.00275 | -0.00033 |

4. Conclusions

The CSP homogeneous correction provides an efficient way to identify an accurate projection on a SIM of any state vector close to, but off, the SIM. This property can be used to identify and characterize the SIM dimension without resorting to expensive trajectory calculations. An effective dimensionality reduction is obtained as the CSP information can be computed as a function of just the $N - M$ major species. Significant CPU savings can be achieved by skipping the detailed calculation of the fast approach to the SIM at the cost of a minimal loss of accuracy. Further development of this methodology will allow the utilization of CSP to identify and use tabulated manifolds for time integration in general chemical kinetic systems, thereby enabling reacting flow computations with adaptively reduced chemistry.

Acknowledgments

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Figure 1. Black points are random design points in a hypercube defined by the vertex $[y_1, y_2, y_3] = [0.875, 0.875, 0.875]$ and edge lengths $[\delta y_1, \delta y_2, \delta y_3] = [0.125, 0.125, 0.125]$; red points are calculated with $M = 1$; green points are calculated with $M = 2$; both calculations involving two homogeneous corrections.

Figure 2. Time integration with CSP (black line) for an initial point $y = [0.97003, 0.92696, 0.99514]$. Red points correspond to the results of the integration after 1 homogeneous correction with $M = 2$ at $t = 0$, followed by CSP integration of the full system. Green points are the results after 2 homogeneous corrections.

5. References

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