Time Integration of Reacting Flows with CSP Tabulation

Youssef Marzouk*, Bert Debusschere†, Habib Najm†, Dimitris Goussis‡, Mauro Valorani§, Michael Frenklach¶
*Massachusetts Institute of Technology, Cambridge, MA, USA
†Sandia National Laboratories, Livermore, CA, USA
‡National Technical University of Athens, Athens, Greece
§Università di Roma “La Sapienza”, Rome, Italy
¶University of California at Berkeley and Lawrence Berkeley National Lab., Berkeley, CA, USA
bjdebus@sandia.gov

Abstract— **This paper presents recent progress on the use of Computational Singular Perturbation (CSP) techniques for time integration of stiff chemical systems. The CSP integration approach removes fast time scales from the reaction system, thereby enabling integration with explicit time stepping algorithms. For further efficiency improvements, a tabulation strategy was developed to allow reuse of the relevant CSP quantities. This paper outlines the method and demonstrates its use on the simulation of hydrogen – air ignition.**

I. INTRODUCTION

The dynamics of chemical systems exhibit a wide range of time scales, with associated stiffness of the governing equations. This stiffness, and the significant complexity of chemical kinetic models, both lead to substantial challenges with the computation of chemical systems. Chemical model simplification and reduction strategies typically target these challenges by reducing the number of reactions and/or species in the model, with associated reduction in model complexity. When done properly, this strategy also reduces the system stiffness. Alternatively, the Computational Singular Perturbation (CSP)-based time integration construction of [1] uses CSP analysis to project out the fast time scales from the detailed chemical source term, thereby rendering the equations non-stiff. The promise of this approach is that explicit time integrators can be used for large-time step integration of the resulting non-stiff source terms, with associated computational speedup as compared to implicit time integration of the non-filtered detailed source term. Further, this can very well eliminate the need for operator-split time integration of reaction-diffusion source terms. Moreover, by tailoring the projection operators to the local chemical state, optimized adaptive strategies can be implemented.

The key challenge with this time integration approach, however, is the large computational cost of solving for the requisite CSP information and the resulting projection matrices. This is where tabulation comes in. By adaptively storing and reusing the CSP information, the significant CSP overhead can be drastically reduced, leading to an efficient overall implementation. We have explored the utility of tabulation of CSP quantities and their reuse for time integration in earlier works on elementary model problems [2], [3]. Our tabulation strategy is based on the Piecewise Reusable Implementation of Solution Mapping (PRISM) [4] technique, whereby the chemical configuration space is suitably and adaptively subdivided into hypercubes within which low order polynomial response surfaces are used to represent the quantities of interest. In the current work, we extend this approach by using kd-trees [5] to efficiently store the CSP information along manifolds in the chemical configuration space, without requiring *a priori* partitioning of this space.

The paper first outlines the use of CSP analysis for integrating chemically reacting systems, illustrated on the simulation of H_2 – air ignition. Next, the tabulation approach is introduced and employed on this same reacting system. We illustrate the performance of the integrator, and highlight the role of the CSP homogeneous projection operation in the table construction and subsequent time integration of the system.

II. BASICS

Consider the chemical system described by

$$\frac{\mathrm{d}\boldsymbol{y}}{\mathrm{d}t} = \boldsymbol{g}(\boldsymbol{y}) \tag{1}$$

where $\boldsymbol{y} \in \mathbb{R}^N$, and $\boldsymbol{g}(\boldsymbol{y})$ is the chemical source term. The CSP basis vectors $\{\boldsymbol{a}_k\}_{k=1}^N$ and covectors $\{\boldsymbol{b}^k\}_{k=1}^N$, all in $\mathbb{R}^N$, enable the decoupling of the fast and slow processes, and the identification of low dimensional slow invariant manifolds (SIMs) [6]. Thus, we have

$$\begin{aligned}\frac{\mathrm{d}\boldsymbol{y}}{\mathrm{d}t} = \boldsymbol{g} &= \boldsymbol{g}_{\text{fast}} + \boldsymbol{g}_{\text{slow}} &(2)\\ &= \boldsymbol{a}_1 f^1 + \boldsymbol{a}_2 f^2 + \cdots + \boldsymbol{a}_N f^N &(3)\end{aligned}$$

where $f^i = \boldsymbol{b}^i \cdot \boldsymbol{g}$, for $i = 1, 2, \ldots, N$. After relaxation of fast transients, with M modes exhausted,

$$\boldsymbol{g}_{\text{fast}} = \sum_{r=1}^{M} \boldsymbol{a}_r f^r \approx 0 \tag{4}$$

$$\boldsymbol{g}_{\text{slow}} = \sum_{s=M+1}^{N} \boldsymbol{a}_s f^s = (I - \sum_{r=1}^{M} \boldsymbol{a}_r \boldsymbol{b}^r)\boldsymbol{g} = \boldsymbol{P}\boldsymbol{g} \tag{5}$$

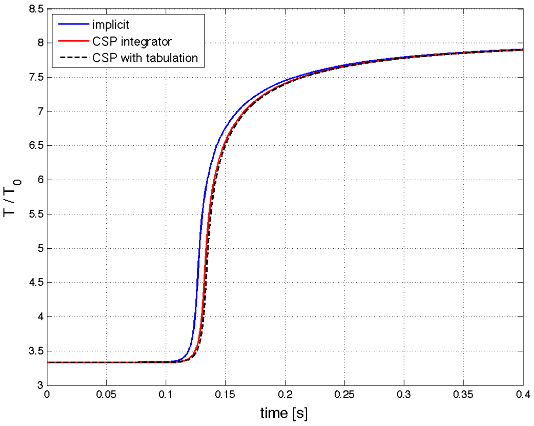

Fig. 1. Evolution of temperature (normalized by $T_0 = 300$ K) in an igniting lean H_2–air system, using an implicit solver (blue solid line), the CSP solver (red solid line), and the CSP solver with tabulation (black dashed line). All three approaches are in good agreement, except for minor differences in the ignition time delay.

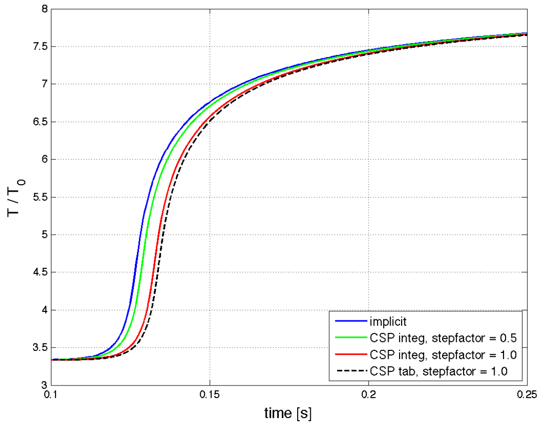

Fig. 2. Close-up of the ignition zone of Fig. 1. The CSP approximation converges to the implicit solution with decreasing time step factor α : $\Delta t = \alpha \cdot \tau_{M+1}$.

The CSP integrator [1] proceeds in each time step by first integrating the slow dynamics of the system, followed by a homogeneous correction (HC) to correct for the fast time scales:

$$\tilde{\boldsymbol{y}}(t+\Delta t) = \boldsymbol{y}(t) + \int_t^{t+\Delta t} \boldsymbol{P}\boldsymbol{g}\,\mathrm{d}t' \quad (6)$$

$$\boldsymbol{y}(t+\Delta t) = \tilde{\boldsymbol{y}}(t+\Delta t) - \sum_{m,n=1}^{M} \boldsymbol{a}_m \tau_n^m|_t \hat{f}^n \quad (7)$$

$$\hat{f}^n = \boldsymbol{b}^n \cdot \boldsymbol{g}[\tilde{\boldsymbol{y}}(t+\Delta t)] \quad (8)$$

where τ_n^m is the inverse of λ_n^m, given by

$$\lambda_n^m = \left(\frac{\mathrm{d}\boldsymbol{b}^m}{\mathrm{d}t} + \boldsymbol{b}^m \boldsymbol{J} \right) \boldsymbol{a}_n \quad (9)$$

and $\boldsymbol{J}$ is the Jacobian of $\boldsymbol{g}$. Note that the time integration of the slow dynamics can be done using any suitable time integration procedure. Also, the matrix τ_n^m is diagonal with entries the time scales $\{\tau_k\}_{k=1}^N$ when the CSP basis vectors are chosen to be the eigenvectors of $\boldsymbol{J}$ and the curvature of the SIM is neglected, *i.e.* $\mathrm{d}\boldsymbol{b}^m/\mathrm{d}t = 0$.

III. APPLICATION TO H_2–AIR IGNITION SYSTEM

The CSP integration method outlined in the previous section was applied to the simulation of ignition of a lean homogeneous H_2–air mixture at a temperature of $T = 1000$ K. The system is modeled using a 9 species reaction mechanism, resulting in a total state space dimension of $N = 10$ (9 species + temperature) [7]. Fig. 1 compares the predicted temperature evolution obtained with a full, implicit integration (using CVODE), and with the CSP integrator (using the explicit RKC integration scheme).

The CSP integrated solution is in good agreement with the full solution, except for a small difference in the ignition time delay. The CSP approximation can be improved by selecting a smaller time step factor α, where α is the integration time step as a fraction of the first non-exhausted time scale τ_{M+1}: $\Delta t = \alpha \cdot \tau_{M+1}$. As shown in Fig. 2, cutting the time step factor to 0.5 greatly improves the accuracy of the predicted ignition time.

As the reaction progresses, the number of exhausted modes M, and the associated CSP radicals change according to the reaction dynamics. Fig. 3 indicates that the system initially has two exhausted modes, followed by a time window during ignition where all modes are active, after which M gradually increases up to three at late time, as more and more modes become inactive.

IV. TABULATION

While the CSP integrator provides an effective way to remove stiffness from the reaction system, the approach is still computationally expensive given the high cost of determining the CSP vectors and covectors $\boldsymbol{a}$ and $\boldsymbol{b}$, as well as the associated time scales. To improve the efficiency of the CSP integrator, a tabulation approach has been developed to enable reuse of the essential CSP quantities: the M fast CSP vectors and covectors, as well as the $M+1$ fastest time scales, which are sufficient to assemble the slow-manifold projector $\boldsymbol{P}$ needed for the HC and CSP integration, and to select the time step along the slow manifold.

While other methods have relied on tabulation of integrated source term data over the chemical state space before [4], [8], the combination of CSP with tabulation is particularly powerful as CSP effectively reduces the needed dimension of the tabulation. As the CSP vectors, covectors and time scales can be modeled as functions of the active modes only, it is sufficient to tabulate these quantities in an $N - M$ dimensional table, rather than having to cover the full N-dimensional state space. For example, for the H_2–air system studied here, tabulation in a 7-dimensional table is sufficient for the section(s) of the 10-dimensional state space where 3 modes are exhausted (see Fig. 3).

Starting from a number of *design points* in state space, successive HCs are applied to identify SIMs in the system. Each SIM is characterized by a unique value of M and the

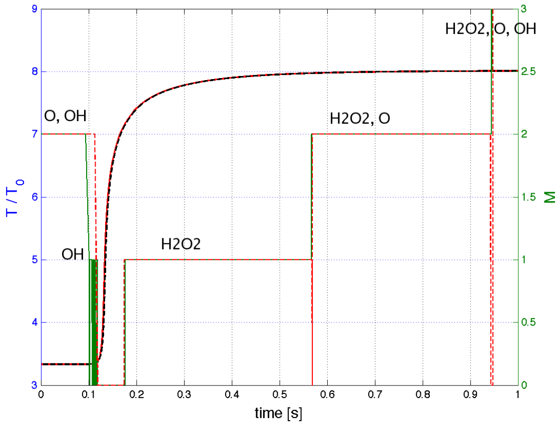

Fig. 3. Evolution of the temperature and the number of exhausted modes, M, as obtained by the CSP integrator (green) and the CSP integrator with tabulation (red). The species labels indicate the CSP radicals corresponding with the exhausted modes.

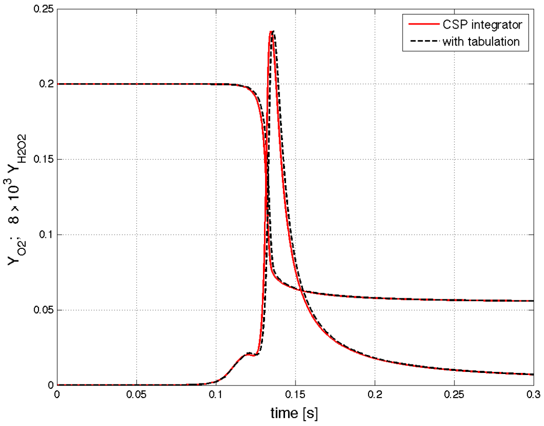

Fig. 4. Mass fractions of O_2 and H_2O_2 using the CSP integrator with (black dashed line) and without tabulation (red solid line). The tabulation approximation is in excellent agreement with the non-tabulated integration.

associated CSP radicals. The extent in $\mathbb{R}^N$ within which each manifold is valid is assumed to be the smallest box, aligned with the Cartesian coordinates $(T, Y_1, \ldots, Y_k)$, that encompasses all design points that are attracted by the manifold.

For each identified SIM, the tabulation of the associated CSP information relies on a nonparametric regression approach, which offers a high degree of flexibility as it does not depend on any pre-determined spatial partitioning. Information is retrieved using efficient nearest-neighbor searches through kd-tree data structures that cover the $N - M$ dimensional slow species space [5], and new data can readily be added to the table as it becomes available. In the current implementation, the CSP information is approximated with the corresponding values at the nearest neighbor point in the table, which amounts to a 0^{th}-order interpolation. Higher order interpolations, relying on interpolation between nearest neighbors or on polynomial response surfaces [2], [4], are the subject of ongoing work.

As indicated in Figs. 1 and 2, the temperature profile in the ignition simulation with the tabulation approximation matches the predictions with the regular CSP integrator very well. While showing overall good agreement, Fig. 3 does reveal small differences in the number of exhausted modes with and without tabulation in a small time span right before ignition, at $t \approx 0.1$ s. However, those differences seem to have no significant effect, as illustrated by the overall good agreement between the CSP predictions with and without tabulation of the temperature profile, as well as the O_2 and H_2O_2 species mass fractions shown in Fig. 4.

In [2], the CSP integrator combined with a tabulation approach was shown to be competitive with direct CVODE integration in terms of CPU cost. The performance of the present tabulation scheme, as a function of the table size, system and manifold dimensionality, degree of stiffness, and desired accuracy is currently under study.

V. CONCLUSIONS

This paper demonstrated the use of CSP to automatically reduce the stiffness in reaction mechanisms, enabling their integration with efficient explicit time stepping algorithms. To improve the computational efficiency of this approach, a tabulation method has been developed that enables efficient reuse of CSP information. The approach was shown to give good results for the simulation of ignition in a H_2–air mixture with a 9 species model. Ongoing work focuses on further improvement of the efficiency and accuracy of the tabulation approach.

ACKNOWLEDGMENT

This work was supported by the US Department of Energy (DOE), Office of Basic Energy Sciences (BES), SciDAC Computational Chemistry program. Sandia National Laboratories is a multiprogram laboratory operated by Sandia Corporation, a Lockheed Martin Company, for the United States Department of Energy under contract DE-AC04-94-AL85000. MYF acknowledges the financial support of DOE, BES under contract no. DE-AC03-76SF00098. MV acknowledges the support of the Italian Ministry of University and Research (MIUR).

REFERENCES

[1] Valorani, M., and Goussis, D.A., *J. Comput. Phys.*, 169:44–79 (2001).
[2] Lee, J.C., Najm, H.N., Lefantzi, S., Ray, J., Frenklach, M., Valorani, M., and Goussis, D., *Combustion Theory and Modeling*, 11(1):73–102 (2007).
[3] Lee, J.C., Najm, H.N., Lefantzi, S., Ray, J., and Goussis, D.A., in *Computational Fluid and Solid Mechanics 2005* (K. Bathe, Ed., Elsevier Science, pp. 717–720, (2005).
[4] Tonse, S.R., Moriarty, N.W., Brown, N.J., and Frenklach, M., *Israel Journal of Chemistry*, 39:97–106 (1999).
[5] Arya, S., Mount, D. M., Netanyahu, N. S., Silverman, R., and Wu, A. Y., *J. ACM*, 45(6):891–923 (1998).
[6] Lam, S.H., and Goussis, D.A., *Proc. Comb. Inst.*, 22:931–941 (1988).
[7] Yetter, R.A., Dryer, F.L., and Rabitz, H., *Combust. Sci. Technol.*, 79:97 (1991).
[8] Pope, S.B., *Combust. Theory and Modelling*, 1:41–63 (1997).