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Adaptive Explicit Integration of Stiff Chemical Systems

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***Abstract*— We discuss recent progress on the use of Computational Singular Perturbation (CSP) analysis for time integration of stiff chemical systems. We present results pertaining to the use of the CSP integrator in dynamical systems exhibiting limit cycle behavior. We also present an adaptive tabulation scheme that enables storage and reuse of relevant CSP quantities, allowing efficient use of the CSP time integration strategy.**

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 stratgies 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 ends up with reduced 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]. The 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 present context, we extend this approach in two ways. First, we outline results of time integration of a chemical system exhibiting limit cycle oscillations using the CSP integrator, without tabulation. Second, we apply the tabulation strategy coupled with the CSP integrator on a model problem with a complex manifold structure. 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) [5]. Thus, we have

$$\frac{\mathrm{d}\boldsymbol{y}}{\mathrm{d}t} = \boldsymbol{g} = \boldsymbol{g}_{\rm fast} + \boldsymbol{g}_{\rm slow} \tag{2}$$

$$= \boldsymbol{a}_1 f^1 + \boldsymbol{a}_2 f^2 + \cdots + \boldsymbol{a}_N f^N \tag{3}$$

where

$$f^i = \boldsymbol{b}^i \cdot \boldsymbol{g} \qquad i = 1, 2, \ldots, N \tag{4}$$

and where, after relaxation of fast transients, and with M modes exhausted,

$$\boldsymbol{g}_{\rm fast} = \sum_{r=1}^{M} \boldsymbol{a}_r f^r \approx 0 \tag{5}$$

$$\boldsymbol{g}_{\rm slow} = \sum_{s=M+1}^{N} \boldsymbol{a}_s f^s = (I - \sum_{r=1}^{M} \boldsymbol{a}_r \boldsymbol{b}^r)\boldsymbol{g} = \boldsymbol{P}\boldsymbol{g} \tag{6}$$

The CSP integrator [1] proceeds in each time step using a homogeneous correction followed by a time integration

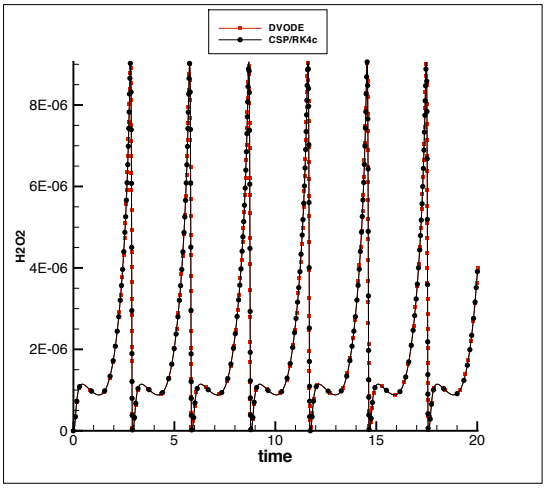

Fig. 1. H2O2 versus time, highlighting the agreement between the CSP integrator and DVODE over a long time horizon.

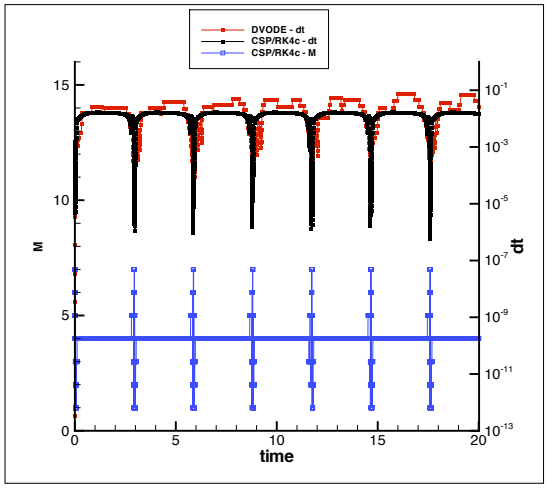

Fig. 2. Time step for DVODE and CSP, along with the time evolution of M.

of the slow dynamics, namely :

$$\tilde{\boldsymbol{y}}(t) = \boldsymbol{y}(t) - \sum_{m,n=1}^{M} \boldsymbol{a}_m \tau_n^m f^n \tag{7}$$

$$\boldsymbol{y}(t+\Delta t) = \tilde{\boldsymbol{y}}(t) + \int_t^{t+\Delta t} \boldsymbol{P}\boldsymbol{g}\,\mathrm{d}t' \tag{8}$$

where $\tau_n^m = 1/\lambda_n^m$, and, given the Jacobian $\boldsymbol{J}$ of $\boldsymbol{g}$,

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

Note that the time integration of the slow dynamics can be done using any suitable time integration procedure. Note also that the matrix $\{\tau_n^m\}_{m,n=1}^N$ is diagonal with entries the time scales $\{\tau_k\}_{k=1}^N$ when the CSP basis vectors are chosen as 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 Systems with Limit Cycle Dynamics

We applied the CSP time integrator to a CO-H_2 Continuously Stirred Tank Reactor (CSTR) computation using an 11-species, 33-reaction chemical kinetic model [6], with initial conditions $P = 10$ Torr, $T = 650$ K. We present in Figures 1-3 results comparing the CSP integrator and DVODE [7] as applied to this problem.

Fig. 1 indicates, using the time evolution of H_2O_2, the accuracy of the CSP integrator result compared to DVODE over a long time horizon comprised of several limit cycle oscillations.

Fig. 2 shows the time evolution of the time step used by DVODE and by the CSP integrator, along with the time evolution of the number of exhausted modes M found by CSP. The CSP integrator time step is chosen as a fraction of the driving time scale, $\tau_{M+1} = 1/\lambda_{M+1}$, where λ_{M+1} is the $(M+1)$-th eigenvalue of $\boldsymbol{J}$. The results indicate similar oscillatory behavior of the time step between the two schemes, and the associated evolution of M. The

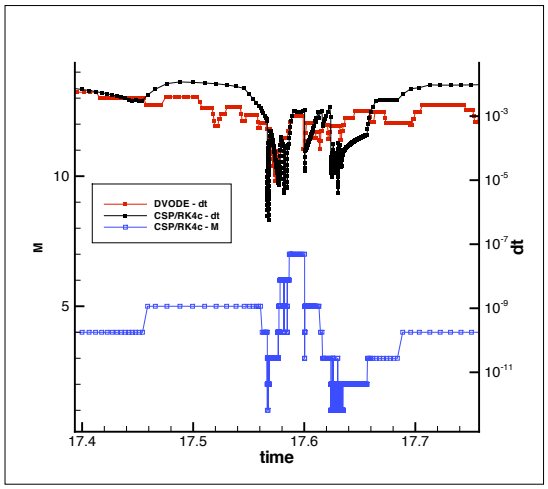

Fig. 3. A detailed inspection of the time step history in a narrow time region where significant oscillation in the integrator time step is observed.

number of exhausted modes is constant at $M = 4$ over long periods of time, with cyclic oscillation to higher and lower values over brief time intervals. A detailed examination of these time traces in one of these time intervals is shown in Fig. 3. This plot shows that both DVODE and CSP exhibit rapid changes in the integration time step in this region. In the case of CSP, those changes are in accordance with the changes in τ_{M+1} and the observed changes in M.

IV. Adaptive Tabulation

The adaptive tabulation procedure seeks to save and reuse the information required for the CSP integrator. This includes,

- The CSP basis vectors $\boldsymbol{a}_1, \boldsymbol{a}_2, \ldots, \boldsymbol{a}_M$
- The CSP covectors $\boldsymbol{b}^1, \boldsymbol{b}^2, \ldots, \boldsymbol{b}^M$
- The $\{\tau_n^m\}_{k=1}^M$ and the driving time scale τ_{M+1}.

These quantities are tabulated using a local low-order polynomial response surface within each hypercube. The tabulation is over $(N-M)$-dimensional space in each hypercube, namely over the $(N-M)$ active (non-CSP-radical) species. A key gain in tabulation efficiency is provided by the reduced dimensionality. The table construction process involves decisions on the proper size of each hypercube, and the value of M within it. Various considerations are relevant in this regard. In effect, we seek to maximize the hypercube size and the assigned value of $M = M^* \in [0, N]$ subject to

- Identical M^* CSP radicals and $N - M^*$ active species $\forall \boldsymbol{y}$ in the hypercube
- Homogeneous corrections computed with M^* do not take the state vector outside the hypercube
- The fast amplitudes f^k vanish to within given tolerances after a specified number of homogeneous corrections
- Response surface goodness-of-fit statistics are satisfied

Moreover, in the time integration process, using the tabulated information, we employ multiple homogeneous corrections to proceed directly to the SIM without the costly time integration of the fast time scales, before proceeding along the SIM.

We applied the tabulation procedure to the 3-dimensional Oregonator model problem analyzed in [8]. The right hand side of this model problem reads:

$$\boldsymbol{g} = \begin{bmatrix} -\frac{5\,y_1}{\varepsilon} - \frac{y_1 y_2}{\varepsilon} + y_2 y_3 + \frac{5\,y_2^2}{\varepsilon} + \frac{y_3}{\varepsilon} - y_1 \\ 10\frac{y_1}{\varepsilon} - \frac{y_1 y_2}{\varepsilon} - y_2 y_3 - 10\frac{y_2^2}{\varepsilon} + \frac{y_3}{\varepsilon} + y_1 \\ \frac{y_1 y_2}{\varepsilon} - y_2 y_3 - \frac{y_3}{\varepsilon} + y_1 \end{bmatrix} \quad (10)$$

where ϵ 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.

We explored the accuracy and computational savings using the tabulated CSP integration procedure with 4^{th}-order Runge-Kutta time integration of the slow dynamics, as compared with CVODE [7]. We found generally that, after 2 homogeneous corrections, the fast amplitudes are exhausted and CSP decouples fast and slow processes efficiently. We found excellent accuracy of the CSP integrator results after the fast transients are exhausted, with maximum error less than 0.2%. As regards computational efficiency, we found that the CPU cost of the tabulated CSP integration procedure is of the same order as that of CVODE. Clearly therefore, on this simple problem, there is no direct computational time integration advantage in using the tabulated CSP integration approach as compared to the implicit time integration strategy used in CVODE. It remains to be seen how this comparison looks for more complex high-dimensional stiff chemical kinetic problems. Moreover, it is noteworthy that the promise of this technique, using explicit integration of the slow dynamics, is the elimination of the need for operator-splitting of the reaction and diffusion terms [2]. Therefore, comparisons between operator-split flame computations using implicit time integration of the chemical source terms, and non-split explicit coupled time integration of both diffusion and reaction terms will be of interest.

V. Conclusion

We have explored the application of the CSP integrator to oscillatory chemical systems, specifically the CO-H_2 CSTR system, and found excellent accuracy over a long time horizon covering a number of limit cycle oscillations. We also explored the utility of tabulation of CSP information for purposes of adaptive chemical reduction based on filtering-out of the fast time scales. We applied the tabulation procedure to a model 3D dynamical system, and demonstrated algorithms for table construction and utilization. Results indicate excellent accuracy of the tabulated integrator results, and computational performance that is comparable to CVODE. Followup work is in progress, extending this application to a H_2-O_2 system as an intermediate step before tackling hydrocarbon fuels.

VI. Acknowledgement

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