Chemical Reductions Do Not Necessarily Lead to Computational Reductions

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Abstract— The comparison of solutions of the shock-ignition of a hydrogen bubble in air using detailed and reduced chemical mechanisms is presented. The reduced chemical kinetics mechanism with 9 species and 12 irreversible reactions is obtained using the *G-Scheme*. The test shows that the simulation becomes computationally more expensive using the reduced mechanism.

I. Introduction

Detailed and reliable numerical simulations of compressible reactive flows are particularly challenging because of their strong multiscale characteristics. Chemical, diffusive and viscous phenomena occur on molecular scales. Thus, spatial scales span five orders of magnitude for the simulation of a device having the size of the order of centimeters.

In this work we use the *p*arallel *W*avelet *A*daptive *M*ultiscale *R*epresentation (pWAMR) method [1], [2] for controlling the grid adaptivity based on magnitudes of wavelet amplitudes. The amplitude of the wavelet transform provides a direct measure of the local error at each associated collocation point. Applying a threshold and omitting those points with small amplitudes, a dynamically adaptive sparse grid is generated.

The numbers of species and reactions included in a chemical mechanism are other factors that lead to an increase in computational cost. The use of a reduced mechanism can help in reducing the computational cost at the sacrifice of introducing a small inaccuracy in the solution. We use a reduction method based on the *G-Scheme* [3]. The methodology proposed [4] identifies the most important reactions in the process using an objective index.

We perform the numerical simulation of a shock impacting a hydrogen bubble.

II. Test case description

The pWAMR method is used to solve the same problem proposed by Billet *et al.* [6]. The domain is two-dimensional with height of 0.75 cm and length of 5 cm. The ambient gas is air, represented by a $78N_2 : 22O_2$ mixture in mole fraction. A hydrogen bubble of radius $r_0 = 0.28$ cm is centered at $x = 0.8$ cm. A Mach 2 shock is located at $x = 0.4656$ cm. The unshocked gas has a uniform pressure of 1 atm and temperature of 1000 K. The post-shock state has a pressure of 4.41 atm, temperature of 1559 K, and velocity

of 798 m/s. The left boundary is a supersonic inflow with the post-shock conditions prescribed, and top and bottom boundaries are symmetry planes.

The problem is modeled using the reactive Navier-Stokes equations for a multicomponent gas mixture. It includes the evolution equations for species' partial density. The production rate for the species for the kinetic mechanism considered is evaluated using Chemkin. The constitutive relations include detailed multi-component diffusion, Sorét and Dufour effects and state dependent transport properties. The pWAMR method is implemented as a method of lines algorithm. Here we use a standard 4th/5th-order Runge-Kutta-Fehlberg scheme to integrate the equations.

III. *G-Scheme* reduction method

A separate analysis using the *G-Scheme* is performed to obtain the reduced chemical kinetics mechanism. The ODEs representing just the chemical reactions constitute a multi-scale system. The *G-Scheme* is a time accurate computational tool that exploits, adaptively, opportunities for reduction of a multi-scale system in order to integrate only a non-stiff subset of equations. The space is decomposed in 4 subspaces according to the time scale of each mode: the active subspace $\mathbb{A}$ contains all the active scales, the subspaces $\mathbb{H}$ has all scales slower than the active ones, while faster scales are in the subspaces $\mathbb{T}$, and the invariant subspace $\mathbb{E}$ contains the invariant modes. Only the active subspace is numerically integrated using a time step of the order of the time scale of the fastest mode in this subspace. The contributions of the slow and fast subspaces are evaluated using a asymptotic approximations.

The standard CSP Participation Index [5] provides a non-dimensional measure of the contribution of the k-th reaction to the i-th mode at the state $\mathbf{x}$. In the *G-Scheme* we modify it to define the Participation Index of the k-th reaction relative to the dynamics of the subspace[1] s:

$$\widehat{\mathscr{P}}_k^s(\mathbf{x}) = \frac{\sum_{i=N_{s,b}}^{N_{s,e}} C_k^i(\mathbf{x}) r^k(\mathbf{x})}{\sum_{i=N_{s,b}}^{N_{s,e}} \sum_{k'=1}^{\mathbb{R}} |C_{k'}^i(\mathbf{x}) r^{k'}(\mathbf{x})|}, \quad (1)$$

[1]The index $s = h/a/t$ identifies one of the four subspaces as follows: h for $\mathbb{H}$, a for $\mathbb{A}$, and t for $\mathbb{T}$.

where $C_k^i = \mathbf{b}^i(\mathbf{x}) \cdot \mathbf{S}_k$, $\mathbf{b}^i$ is the contravariant basis vector, and $\mathbf{S}_k$ the stoichiometric vector. Note that $0 < P_k^i(\mathbf{x}) < 1$. The CSP Participation Index provides a non-dimensional measure of the contribution of the the k-th reaction to the i-th mode at the state $\mathbf{x}$ normalized by the contribution of all reactions at the state $\mathbf{x}$. In contrast, equation (1) provides a measure of the contribution of the k-th reaction to the subspace s normalized by the contribution of all reactions in the same subspace. In other words, it provides a criterium to identify if the k-th reaction is *active*, *slow*, *fast*, or as *negligible* if it doesn't have a relevant value in any subspace. The concept of an *important reaction* is not an inherent property of the reaction but a function of the local state.

We can rank the reactions according to the maximum value of $\widehat{\mathscr{P}}_k^s$ in the time interval considered as follows:

$$\overline{\mathscr{P}}_k^s = \max_{t_b < t < t_e} \left(|\widehat{\mathscr{P}}_k^s(\mathbf{x})| \right). \qquad (2)$$

Equation (2) provides a value that summarizes all the information given by (1). By identifying the maximum value of the participation index calculated for any reaction along the process, we obtain an objective measure of the importance of the reaction for the analyzed problem.

Based on (2), we can create a very simple iterative procedure to obtain a simplified mechanism that satisfies an objective criteria defined by the analyst. In this study we choose the error in the equilibrium temperature and ignition time as criteria, defined as:

$$err_T = \frac{T_{f,s} - T_{f,c}}{T_{f,c}} \quad \text{and} \quad err_t = \frac{t_{e,s} - t_{e,c}}{t_{e,c}}, \qquad (3)$$

where $T_{f,s}$ and $T_{f,c}$ are the equilibrium temperatures for the simplified and complete mechanism respectively, while $t_{e,s}$ and $t_{e,c}$ are the ignition times for the simplified and complete mechanism respectively. Then the algorithm finds iteratively the threshold value of ParticipationIndex giving the smallest mechanism that satisfies a user-defined value for these error criteria. In this study we have used $err_T = err_t = 0.02$.

The simplified mechanism found contains 16 irreversible reactions. In comparing it with the full mechanism, it yields a maximum error of 1.76% for the equilibrium temperature and 1.27% for the ignition time in the range of initial temperature, pressure and mixture ratio of $T_i = 700 - 1500\ K$, $p_i = 0.5 - 100\ atm$, and $\phi = 0.5 - 2$.

IV. RESULTS

We perform the numerical simulation of a shock impacting a hydrogen bubble. Since the bubble is less dense than the surrounding air, large bubble distortions result and reactions start on the deformed bubble surface.

The mechanisms used for the test both have 9 species; the detailed one includes 19 reversible elementary reactions
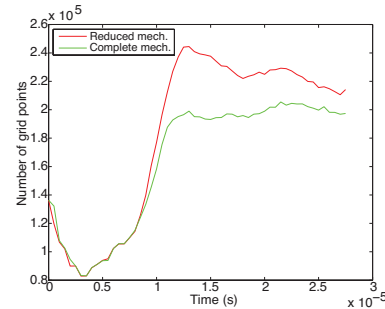

Fig. 1.   Comparison of the umber of grid point required for the simulation using the complete and the reduced mechanism.

while the reduced one 16 irreversible reactions. The solutions obtained in both cases are very accurate and do not show any sensible difference.

Most interestingly, Fig. 1 shows that the number of grid points used in the simulation with the reduced mechanism is *larger* than in the one with the detailed mechanism. The average number of grid points with the detailed mechanism is 164,369, while it increases to 182,635 when using the reduced mechanism.

The test shows that the simulation becomes computationally more expensive using the reduced mechanism. For a small mechanism, like hydrogen/air with 9 species, it is not possible to reduce the number of species without introducing a relatively large error. The present computational study shows that just a reduction in the number of reactions does not necessarily produce a significant saving in the computational cost.

We observe that a larger number of points at the finest scale is necessary to solve the problem with the same accuracy. We note that the finest resolution required by the solutions is below 1 μm for both detailed and reduced mechanisms. An explanation of this increase based on analysis of the solution, with particular attention to the species involved in the deleted reactions, will be discussed.

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