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A dynamic adaptive chemistry scheme for reactive flow computations

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Abstract

An on-the-fly kinetic mechanism reduction scheme, referred to as dynamic adaptive chemistry (DAC), has been developed to incorporate detailed chemical kinetics into reactive flow computations with high efficiency and accuracy. The procedure entails reducing a detailed mechanism to locally and instantaneously accurate sub-mechanisms at each hydrodynamic time step of the calculation, and consequently no *a priori* information regarding simulation conditions is needed. The reduction utilizes an extended version of the directed relation graph (DRG) method in which the edges are weighted by a value that measures the dependence of the tail species (vertex) on the head species. An R-value is then defined at each vertex as the maximum of the products of these weights along all paths to that vertex from an initiating species. Active species are identified by their R-values exceeding a threshold value, ε_R, using a modified breadth-first search (BFS) that starts from a pre-defined set of initiating species. Chemical kinetics equations are then formulated with respect to the active species, with the inactive species considered only as third body collision partners. The DAC method is implemented into CHEMKIN and tested by simulating homogeneous charge compression ignition (HCCI) combustion using detailed and pre-reduced n-heptane mechanisms (578 species and 178 species, respectively) as the full mechanisms. The DAC scheme reproduces with high accuracy the pressure curves and species mass fractions obtained using the full mechanisms. The on-the-fly mechanism reduction scheme introduces minimal computational overhead and achieves more than 30-fold time reduction in calculations using the 578-species mechanism.

Keywords: Detailed chemistry; Mechanism reduction; Directed relation graph; Adaptive chemistry

1. Introduction

Incorporating detailed chemistry into computational fluid dynamics (CFD) calculations is widely recognized as necessary for developing predictive combustion models. The use of detailed chemical kinetics gives rise to a large system of nonlinear stiff ordinary differential equations (ODE) for each CFD computational cell. The numerical solution of the large number of such systems within the CFD framework results in extremely long CPU times. Consequently, large-scale three-dimensional reactive flow simulations become computationally prohibitive with detailed kinetic mechanisms developed for most hydrocarbon fuels.

Comprehensive chemical kinetic mechanisms for realistic fuels often incorporate reaction pathways valid over a wide range of operating condi-

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tions and may include hundreds of species and thousands of elementary reactions. However, for most practical combustion simulation problems, a relatively smaller number of species and reactions participate in the processes of pivotal importance such as heat release and emissions formation. This observation forms the basis of several approaches that endeavor to eliminate unimportant species and reactions from the detailed mechanisms. A recent review of these methods can be found in [1]. Most methods reduce the detailed mechanism to a single skeletal mechanism, either by defining a problem-specific merit function and carrying out global optimization, or by generating reduced sub-mechanisms at a group of sampled conditions and assembling the final reduced mechanism by taking the union of the sub-mechanisms.

Adaptive chemistry (AC) is an alternative strategy that has been explored by several researchers [2–5]. The principle of AC is to apply a locally accurate reduced mechanism, instead of the full mechanism or a fixed skeletal mechanism. This locally accurate mechanism is chosen from a library of reduced mechanisms based on the specific thermo-chemical conditions encountered in simulating the reactive flow. Compared to applications in which the single skeletal reduced model must capture the relevant chemistry over potentially vastly different conditions, it is easier for the sub-mechanisms in an AC library to achieve high accuracy with a smaller number of species because a given reduced mechanism is used only within a narrow range of thermo-chemical conditions. However, most of the AC approaches in the literature generate a library of reduced mechanisms before reactive flow calculations, assuming that the union of the feasibility ranges of sub-mechanisms in the library can cover all thermo-chemical conditions that will be encountered in the calculations. In reality, this requirement is challenging for most transient problems due to the great diversity of conditions encountered. The other challenge to employing a pre-generated library of reduced mechanisms is the efficient identification via high-dimensional searching of the most feasible mechanism to use based on the ‘real-time’ conditions. For example, in our previous work [5], we associated each pre-reduced mechanism with a characteristic state point, and used a nearest neighbor search (NNS) method to locate the ‘closest’ characteristic point to the local conditions encountered during combustion simulation, thus identifying the optimal mechanism to be used. Although modest CPU time savings were obtained, we found that the high-dimensional ‘query’ procedure could result in significant computational overhead.

In the present work, we propose an on-the-fly mechanism reduction scheme, termed dynamic adaptive chemistry (DAC), in which the adaptively reduced mechanisms are rigorously valid for their local and instantaneous conditions, and which incurs only minor computational overhead. We have based our approach on the DRG method developed by Lu and Law [6,7], which offers fast reduction speed with high accuracy. In the original DRG approach, each vertex represents a species present in the full mechanism and each directed edge represents the immediate dependence of one species on another. This dependence is quantified by the normalized contribution of B to A, defined by

$$r_{AB} \equiv \frac{\sum_{i=1,I} |\upsilon_{Ai}\omega_i\delta_{Bi}|}{\sum_{i=1,I} |\upsilon_{Ai}\omega_i|}, \qquad \delta_{Bi} = \begin{cases} 1 & \text{if reaction } i \text{ involves } B \\ 0 & \text{otherwise} \end{cases} \tag{1}$$

where i is the reaction index ($i = 1,\ldots,I$), υ_{Ai} is the stoichiometric coefficient of species A in the ith reaction, and ω_i is the progress variable of reaction i. r_{AB} is a measure of the error introduced to the production rate of A due to elimination of all the reactions that contain B. Then a directed relation graph is obtained in which there is a directed edge from A to B if and only if r_{AB} is greater than or equal to a user specified threshold value, ε. A search procedure is then carried out starting with selected major species to incorporate all the reachable species, i.e., those species that lie on a directed path emanating from one of the species in the chosen initial set. We note that a complete weighted directed relation graph is obtained if, for all species A and B, the directed edge from A to B is included in the graph, weighted by the value r_{AB}.

We have implemented the DRG-based scheme into the DAC framework and applied it to simulating homogeneous charge compression ignition (HCCI) combustion of *n*-heptane using the Lawrence Livermore National Laboratory (LLNL) mechanisms [8,9]. A breadth-first search (BFS) algorithm [10] is used in this implementation. However, we have found that the selection of fuel (*n*-heptane) as the only search-initiating species leads to fuel as the sole active species retained after high temperature ignition. This happens even when ε is ten times smaller than the value suggested in [6]. Since fuel fails to connect to any other species following ignition, additional search-initiating species are needed. We initially selected CO as an additional initiating species because CO oxidation reactions are essential to post-ignition kinetics. However, the inclusion of CO results in a large number of active species that include most of the heavy hydrocarbon species. This species proliferation is undesirable because hydrocarbon decomposition reactions generally do not dominate post-ignition kinetics. In fact, as shown below, the use of 25 species is sufficient