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Towards improved automatic chemical kinetic model reduction regarding ignition delays and flame speeds

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ABSTRACT

In chemical kinetic model reduction under internal combustion engine conditions, most implementations only consider ignition related chemistry without consideration of flame speed prediction. In practice, flame propagation commonly exists in spark ignition engines, dual-fuel with pilot injection compression ignition engines, reactivity controlled compression ignition engines, and etc. Due to the inherent time-consuming nature, it is impractical to run a 1-D flame code with trial-and-error methods for model reduction, especially when starting with a large chemical kinetic model. In this paper, an improved reduction methodology is proposed for construction of a small set of species that give accurate predictions of both flame speeds and ignition delays. First, a strong correlation is found between the errors of maximum H radical and the errors in prediction of laminar flame speeds. Addition of H to the search targets in graph-based methods is conducted showing improvement in accuracy of flame speed prediction. Second, the normalized flame speed sensitivity with rate constants is analyzed for identifying a set of species that strongly influences the prediction of flame speeds. Finally, a trial-and-error based method is used for further reduction with a 0-D testbed for prediction of ignition only, while keeping the species important to flame chemistry. The newly proposed reduction methodology is used for development of accurate skeletal models predicting both ignition and flame speeds for several hydrocarbon fuels. These skeletal models include methane (27 species), propane (32 species), n-heptane (126 species), and primary reference fuel gasoline surrogates (207 species) with high fidelity to be used in engine simulations.

1. Introduction

Chemical kinetic modeling plays an essential role in reactive flow simulations, which are routinely applied in assisting designs of efficient combustion devices. However, large-scale simulations with detailed reaction mechanisms are prohibitively expensive in terms of memory and CPU time. Stiffness caused by the wide range of timescales is another issue limiting the use of large reaction mechanisms. Therefore, it is imperative to reduce the complexity of the chemical kinetic model by retaining sufficient species and reaction steps to adequately reproduce relevant physical phenomena under target conditions [1]. Various approaches have been proposed and combined to determine the unimportant species or reaction pathways in a detailed reaction model [1,2]. As the demand to develop a fairly small and accurate reduced model for a particular simulation grows, how to effectively recognize redundant species and reactions according to our needs is the goal to be achieved during the model reduction.

Compared to the approaches with hierarchical group combination by sensitivity analysis or graphical reaction pathway analysis [3–6], an automatic reduction technique is much more efficient and intelligent requiring little effort and expert knowledge form the user. With the detailed chemistry information under the targeted conditions of interest, a small set of species satisfying a preset level of error can be identified automatically without the need to tune the species list after running the software program. In the last decade, several improvements were made in the development of skeletal model by removing the redundant species and their associated reactions. The graph-based methods, such as the directed relation graph (DRG) [7], DRG with error propagation (DRGEP) [8], and path flux analysis (PFA) [9], are often employed as the first-step reduction due to their efficient features. After a satisfactory skeletal model has been developed, say using a DRG based method, further reduction is possible by using trial-and-error methods, such as Target Search Algorithm (TSA) [10], DRG-aided sensitivity analysis (DRGASA) [11], and etc. The main difference among various methods is how to rank the species list to decide which species

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is to be checked first. Niemeyer et al. [12] combined multiple methods into an automatic reduction scheme, the mechanism automatic reduction software (MARS). Lu et al. [13] extended the DRG method with expert knowledge (DRGX) by adding targets of species concentrations, allowing smaller reduction errors for the species of interest while larger errors for other species. Also, packages with other reduction schemes were developed such as simulation error minimization (SEM) [14]. In this study, we will mainly improve the reduction methodology combining graph-based methods and trial-and-error methods. Chen and Chen [15] demonstrate that for a fixed error, the combined scheme can achieve a large degree of reduction than simply using the trial-and-error method.

Under the condition of interest, the chemistry information of the detailed mechanism needs to be sampled for the graph-based method to calculate pairwise relation between species from a testbed. A closed homogeneous reactor [16], a perfectly stirred reactor [17], a premixed flame [18], or a Homogeneous Charge Compression Ignition (HCCI) simulation [19], can be used as the testbed in model reduction. In general, running a flame code is time-consuming with large detailed chemistry models to extract samples under a wide range of thermochemical states. The skeletal models generated by the graph-based approaches are often still large, thus further reduction conducted by the trial-and-error methods to assess whether the error induced by removing a species is unacceptable. The repeated running for all targeted conditions will make the reduction work prohibitively lengthy if one case takes too much time to run. The zero-dimension (0-D) closed homogeneous reactor is advantageous over others due to its efficient feature by evaluating global parameters such as ignition delay time.

Most past development of reduced chemical kinetic models under engine conditions doesn't take low-temperature ignition and flame propagation into account simultaneously [12,16,20,21]. Auto-ignition delay time under high pressure, especially in the negative temperature coefficient (NTC) regions [4,6,16], are the most important for HCCI engines. In contrast, flame propagation is the most salient feature in combustion devices of spark ignition engines. In new combustion concepts in engines, such as dual-fuel with pilot injection compression ignition engines, and reactivity controlled compression ignition engines, flame propagation can be also important under certain operating conditions. More importantly, most flamelet-based tabulation methods [22] require fast computations of laminar flames over the engine conditions. In this study, the existing issues in current automatic reduction approaches considering flame speeds are discussed first. Then, an improved automatic reduction methodology is proposed to accurately retain both flame speed and ignition related chemistry. To demonstrate the proposed improvement, several fuel kinetic models including methane, propane, n-heptane and primary reference fuel (PRF) gasoline surrogates are reduced and validated.

2. Automatic chemical kinetic model reduction methodology

2.1. Directed relation graph with error propagation (DRGEP) method

Due to their efficient feature, graph-based methods are now routinely used for development of skeletal models from detailed mechanisms. In this study, the DRGEP approach (Pepiot-Desjardins and Pitsch [8]) is used as a representative method. Several studies [12,15] have shown that the DRGEP method can achieve more accurate searching than the earliest graph-based method, namely, the DRG method developed by Lu and Law [7] but at the price of losing control over the worst-case errors. The detailed procedures in the DRGEP reduction are summarized as follows.

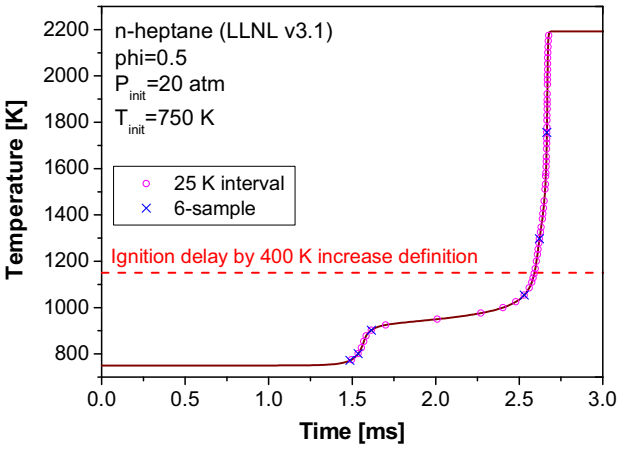

Fig. 1. An example case of 0-D n-heptane simulation using the WMR model with the scatters of x-mark for 6-sample extraction, and circles for 25 K temperature interval sampling. The detailed model is the LLNL v3.1 [24] with 654 species.

(1) Chemical states of a certain combustion process need to be computed and sampled. For HCCI applications, the auto-ignition delays are most relevant features to be reproduced accurately by the reduced model. An in-house computer code for a well-mixed reactor (WMR) [23] is used for computing auto-ignition delays and writing input files for automatic reduction code. Herein, the issue of adequate sampling chemical states during auto-ignition simulations is discussed. Figure 1 shows a typical n-heptane auto-ignition case under the conditions of equivalence ratio (φ) of 0.5, initial temperature and pressure of 750 K and 20 atm, respectively. The Lawrence Livermore National Laboratory (LLNL) v3.1 n-heptane mechanism [24] with 654-species is used in the simulation. The auto-ignition delay time is defined as 400 K above the initial temperature. Six chemical states (denoted by x symbols) at temperatures of 20 K, 50 K, 150 K, 300 K, 550 K, and 1000 K increment above the initial temperature were chosen to cover low and high temperature combustion processes. The circles represent the uniform 25 K temperature interval sampling. In order to examine the difference between the two types of sampling methods, thirty target cases including the NTC regions are selected in the range of $P_0 = \{20\ \text{atm}, 40\ \text{atm}\}$, $\varphi = \{0.5, 1.0, 2.0\}$ and $T_0 = \{750\ \text{K}, 825\ \text{K}, 875\ \text{K}, 950\ \text{K}, 1100\ \text{K}\}$ for the DRGEP reduction. Figure 2 plots the worst case relative error (by comparing the detailed and skeletal chemical model running among the thirty cases) against the species number of each skeletal model during the reduction. Based on the past experience and analysis on the reduction process [15,25], a 10% relative error tolerance which is a post-reduction criterion to select a desired skeletal model is considered reasonable for subsequent reduction. The plot indicates that small differences exist in the DRGEP reduction between the two sampling methods when the 10% error tolerance is considered; thus the 6-sample scheme is found adequate and will be used in each target case in this study.

(2) The pairwise relation between species needs to be determined using the sampled chemistry information. In this study, the Jacobian defined pairwise relation shown in Eq. (1) [15] is used with n_{sp} denoting total number of species in the detailed model. The Jacobian matrix elements are formulated by defining a semi-normalized sensitivity coefficient, which is solved numerically using Eqs. (2)–(4). Y_i and Y_j are the mass fraction of the ith and jth species. W_i and $\dot{\omega}_i$ are the molecular weight and molar production rate of the