Directed Relation Graph Generation Based Method for Efficient Large Kinetic Mechanism Reduction

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***Abstract*— Detailed kinetic mechanisms, for large chain hydrocarbons, contain hundred to thousands chemical species and reactions. However, it is difficult and more often impossible to use large mechanisms to conduct combustion CFD simulations for complex geometries. The objective of the current study is to develop algorithms that automatically and efficiently reduce big detailed kinetic mechanisms to the smallest possible size without losing important details of the flame structure and accuracy of predicted parameters such as flame speed or ignition-delay time. New algorithms have been developed based on the Directed-Relation Graph-Generation method, (DRG-G). Also other methods have been implemented namely Sensitivity Analysis (SA) and Unimportant Reaction Elimination (URE). Specifically, an n-heptane mechanism containing 561 species and 2539 reactions was reduced for temperatures in the range from 600 to 1800 K, pressures in the range from 1 to 50 atm, and equivalence ratios in the range from 0.5 to 1.5. Ignition- delay time was the validation parameter. The mechanism was successfully reduced to 93 species and 295 reactions. Very good agreement between numerical results with the skeletal and detailed mechanism was obtained.**

I. Introduction

Generally, combustion simulation is a rather complex field. It requires chemical understanding of the fuel and physical understating of the flow. For combustion in three dimensional geometry, numerical analysis become a critical issue. As far as the chemistry part is concerned, a detailed kinetic mechanism would be required. As the size of a hydrocarbon chain increases, the number of reactions will increase. As a consequence, even if high computing power is available, simulation costs for a complex system will be a concern. Hence mechanism-reduction algorithms are required that identify key species and reactions needed to describe the combustion process sufficiently accurately.

Generally, detailed kinetic mechanisms reduction is conducted in two steps. In the first step redundant species and reactions are removed, e.g. by sensitivity analysis (SA) and directed-relation graphs (DRG) [1]. In the second step, a time scale analysis, e.g. by computational singular perturbations (CSP) [2], and Int or intrinsic low-dimensional manifolds (ILDM) [3], is carried out. The focus in this study will be on removing redundant species and reaction. Thus, a Directed Relation Graph Generation based method (DRG-G) is developed and implemented or combined with other methods, namely sensitivity analysis (SA) and Unimportant Reaction Elimination (URE). Moreover, computational singular perturbations (CSP) are implemented, details of which, however, are not within the scope of this presentation.

II. Reduction Methodology

A. Directed Relation Graph Generation Based Method

DRG is a graph-based method introduced by Lu and Law [1]. Is suggests that for any two species A and B can be represented as vertex, and the line connecting A and B is the reactions involving them. The strength of the connection between species A and B (r_{AB}) can be calculated via an objective function. Pepiot and Pitsch [4] introduced DRG with Error-Propagation DRG-EP.

In the current study, DRG-G is used. DRG-G introduces two modification over DRG, viz., one for the objective function and another for the species marking. As a first modification, for transient systems DRG-G calculates (r_{AB}) at each time step according to the objective function formulation introduced by Lu and Law [1], but takes only the maximum (r_{AB}) over the entire time steps, as in (1). In a second modification, DRG-G separate species according to their relevance to a root species such as fuel. Then, at each generation, the species with maximum species mark (m) are adopted as new root species, as shown in Fig. 1. In Fig. 1, species A is root species, species B and C considered first generation, and species D is considered the second generation. The species mark evaluation is similar to DRG-EP [4] which considers the relative importance of each species on another rather than the direct importance as in DRG [1].

$$(r_{AB})_{max} = \frac{\sum_{i=1}^{I} |\nu_{A,i}\omega_i\delta_{B,i}|}{\sum_{i=1}^{I} |\nu_{A,i}\omega_i|} . \qquad (1)$$

$$\delta_{B,i} = \begin{cases} 1, & \text{if the ith reaction involves species A and B ,} \\ 0, & \text{Otherwise .} \end{cases}$$

where $\nu_{A,i}$ is the stoichiometric coefficient of species A in *i*th reaction, and ω_i the reaction rate of reaction *i*. Species A and B considered weakly connected whenever (r_{AB}) is smaller than user threshold value ϵ.

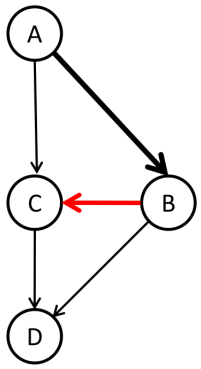

Fig. 1. Schematic graph of DRG-G, each vertex indicates a species, and each line indicates the dependency of each species on another.

B. Sensitivity Analysis and Unimportant Reaction Elimination

Sensitivity analysis methods are implemented by taking the relative error in a target parameter caused by removing one species at a time from the mechanism. In the present study, the target parameter is ignition-delay time (2). Accordingly we define

$$err_k = \frac{|\tau - \tau_k|}{\tau} . \tag{2}$$

where err_k is a relative error, τ the ignition-delay time when all species are involved, and τ_k the ignition-delay time when species k is removed.

Unimportant reaction elimination (URE) is employed in conjunction with the importance-index function defined by Lu and Law [5], which is based on CSP (3). Specifically,

$$I_{k,i} = \frac{|\nu_{k,i}\omega_i|}{\sum_{i=1}^{I} |\nu_{k,i}\omega_i|} , \tag{3}$$

where I is the importance index, $\nu_{k,i}$ the stoichiometric coefficient of th species k in reaction i, ω_i the reaction rate of reaction i, and I indicates the total number of reactions.

C. Validation

An n-heptane mechanism [6] containing 561 species and 2539 reactions was reduced successfully to a skeletal mechanism containing 93 species and 295 reactions for temperatures in the range from 600 to 1800 K, pressures in the range from 1 to 50 atm, and equivalence ratios in the range from 0.5 to 1.5. The reduction sequence was two stages DRG-G, followed by SA and URE, finally one more DRG-G stage, as shown in 2. Very good agreement between the skeletal mechanism and the detailed mechanism was obtained, as shown as in 3.

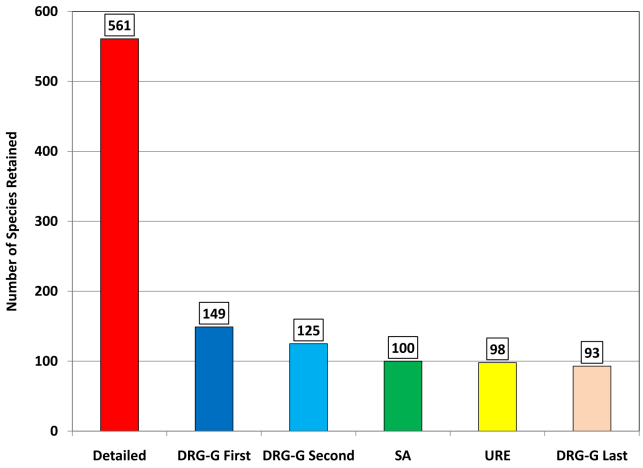

Fig. 2. Number of species retained at each reduction stage.

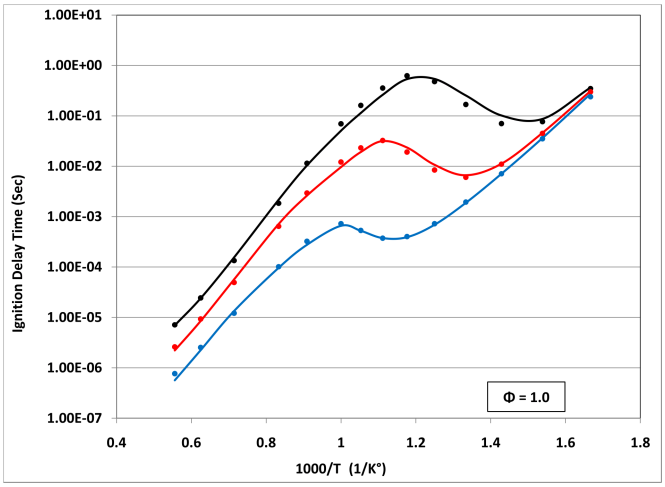

Fig. 3. Ignition-delay time obtained with 561 species detailed mechanism (lines) and 93 species skeletal mechanism (dots), at equivalence ratio 1.0, temperature 600-1800, and pressure 1 (black), 5 (red), and 50 (blue) atm.

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