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Skeletal mechanism reduction through species-targeted sensitivity analysis

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ABSTRACT

The use of simplifying techniques to obtain skeletal kinetic mechanisms with the required accuracy is often a necessary step when computationally demanding simulations are concerned. In this work, a novel approach for an automatic mechanism reduction, aimed at retaining accuracy on specific target species, is proposed. Starting from the consolidated coupling between flux analysis and sensitivity analysis, a methodology based on curve matching and functional data analysis was developed, through which the importance of a species in the target accuracy is assessed via a proper metric. The error associated with the removal of uncertain species from the detailed mechanism is quantified in terms of distance and similarity indices before and after such a removal, within a Species-Targeted Sensitivity Analysis (STSA) framework. A species ranking is then generated, and the original mechanism is progressively reduced. The whole algorithm also implements several improvements to enhance a faster convergence, and adds a novel criterion to remove unimportant reactions, based on sensitivity analysis to kinetic parameters.

The capability of this algorithm was tested through two case studies in this work. A kinetic mechanism for a Toluene Reference Fuel (TRF) was first obtained, with the overall reactivity as reduction target. The numerical procedure allowed to obtain a compact skeletal mechanism (115 species and 856 reactions), able to retain good accuracy in ignition delay time and laminar flame speed predictions of both fuel mixture and pure compounds. More important, two skeletal mechanisms for methane combustion, including chemistry of nitrogen oxides (NO_x), were developed, with different degrees of reduction. The agreement between the original and the skeletal mechanisms in terms of NO formation was successfully assessed with satisfactory results. Attention was also dedicated to the choice of the type of reactor where undertaking reduction, which turned out to play a major role in the overall process.

1. Introduction

In order to ensure the necessary accuracy, high-fidelity models of combustion phenomena require a complete representation of the elementary reactions taking place among the different species involved in the modeled system. For this purpose, detailed kinetic mechanisms are developed for several classes of hydrocarbon fuels, which are validated in simple systems (e.g. ideal reactors) through comparison with experimental data. As one can imagine, their ultimate application lies in the numerical simulation of more complex flames in real systems. Here, the sole use of simplified mechanisms prevents a comprehensive study of such phenomena like the formation of pollutants (soot [1], NO_x [2], etc.), low-temperature combustion [3] and so on. Moreover, the implementation of detailed chemistry can significantly improve the reliability of turbulent simulations, which are based on a non-negligible number of hypotheses, and whose turbulent combustion models would greatly benefit from such an introduction [4].

Nevertheless, the complexity of detailed mechanisms may turn out as the limiting factor when they are applied to multidimensional models of combustion devices. Although the recent advancements in computation technology have allowed to use large kinetic schemes in the modeling of combustion chemistry, the detailed mechanisms of practical fuels, e.g. large hydrocarbons, are still limitedly applicable to 0D and 1D simulations. Moreover, for mechanisms also involving soot, aromatics or oxygenated species, the size of the model may easily explode, such that even the simulation of 1D systems become impractical. In this regard, the 2-methyl alkanes kinetic scheme developed by Sarathy et al. [5], made up of 7175 species and 31,669 reactions, sets up an extreme example.

The impossibility to use most detailed kinetic models in complex simulations has paved the way, in the latest decades, to the research field of mechanism reduction. Several methodologies were developed

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Nomenclature

Roman symbols

P	Pressure
T	Temperature

Acronyms

DIC	Direct Interaction Coefficient
DRG	Directed Relation Graph
DRGASA	DRG-Aided Sensitivity Analysis
DRGEP	DRG with Error Propagation
DRGEPSA	DRG with Error Propagation and Sensitivity Analysis
MAD	Median Absolute Deviation
MPI	Message Passing Interface
NO_x	Nitrogen Oxides
ODE	Ordinary Differential Equation
OIC	Overall Interaction Coefficient
PAH	Polycyclic Aromatic Hydrocarbons
PRF	Primary Reference Fuel
PSR	Perfectly Stirred Reactor
STSA	Species-Targeted Sensitivity Analysis
TRF	Toluene Reference Fuel

Greek symbols

$\hat{\varepsilon}_D$	Normalized L^2-distance index
$\hat{\varepsilon}_S$	Normalized similarity index
ρ	Restart factor in sensitivity analysis
τ	Reactor residence time
ε_D	L^2-distance index
ε_S	Similarity index
Φ	Equivalence ratio

Subscripts

i	Species index
j	Reaction index
k	Reactor index

to this purpose, and complete reviews of the state of the art were carried out by Lu and Law [6], and more recently by Turanyi and Tomlin [7]. As long as skeletal reduction approaches are concerned, they can be focused on the elimination of either reactions or species, which are considered as unimportant in the whole operating space of temperature, pressure, and equivalence ratio. Detecting the reactions which do not contribute to the formation of the investigated species is usually more direct, and several methods were developed to the purpose: among the others, Wang and Frenklach [8] proposed the detailed reduction method, while procedures based on sensitivity analysis were developed by Rabitz et al. [9] and Tomlin et al. [10]. Brown et al. [11] and Vajda et al. [12] applied the Principal Components Analysis (PCA) for reaction elimination. On the other hand, species-based elimination is more challenging because of the mutual coupling among species themselves, but it is much more effective from a computational point of view because it directly reduces the number of equations to be solved in the codes where the kinetic mechanism is applied. Therefore, significant research in this direction has been carried out in the latest decades. The most noticeable works in this field are based either on the analysis of the Jacobian matrix [13,14] or, more recently, on flux analysis. In this regard, the method developed by Lu and Law [15] and known as Directed Relation Graph (DRG) made a significant breakthrough in mechanism reduction: by fixing a group of target species, this method estimates the importance of the remaining species in contributing to their formation. In the wake of DRG, other flux-based methods were developed, including DRG-Aided Sensitivity Analysis (DRGASA) [16], DRG with Error Propagation (DRGEP) [17], Path Flux Analysis (PFA) [18] and DRG with Error Propagation and Sensitivity Analysis (DRGEPSA) [19]. Each of them successfully improved the original formulation of the methodology, thus providing a reference framework for automatic mechanism reduction. On-the-fly techniques based on such approaches were also developed [20], and successfully coupled [21] to pre-existing tabulation-based methodologies [22].

Sensitivity-based reduction approaches like DRGASA and DRGEPSA were able to overcome the weaknesses of flux analysis. If on the one hand flux-based approaches are particularly fast, on the other they are of limited use when a very high level of reduction is required. Therefore, at the price of a higher computing demand, sensitivity-based reductions are able to further push the degree of reduction with an acceptable preservation of accuracy. Nevertheless, the main limitation of such techniques lies in being targeted at fuel/air mixture reactivity (estimated through ignition delay time) as the only property to be preserved. Doing so, all the properties not directly related to it, like flame speed or species formation, are not under control and can then be compromised. A representative example in this direction is set up by the formation of pollutant species like Polycyclic Aromatic Hydrocarbons (PAH) and Nitrogen Oxides (NO_x), whose time scales are orders of magnitude higher than fuel ignition time.

The purpose of this work stems from the need to overcome such a limitation, and proposing a more comprehensive approach able to automatically keep the desired accuracy of skeletal mechanisms on the dynamics of defined target species, in addition to mixture reactivity. Starting from the described background, this paper presents a newer, generalized framework for the generation of skeletal mechanisms, which improves the established techniques through an extended sensitivity analysis, in order to achieve a final optimal reduction. The different steps of the proposed methodology are first described in Section 2, along with the respective strengths and weaknesses. A deeper insight into the theoretical aspects of Species-Targeted Sensitivity Analysis is then provided in Section 3. The whole approach is validated in Section 4, where two case studies are presented. Finally, conclusions are drawn in the last Section.

2. A divide-and-conquer, multi-step approach to skeletal reduction

As pointed out by Xin et al. [23], when a detailed mechanism is to be reduced, the original species may be divided into three subsets: critical, marginal, and nonessential. Once the range of operating conditions (i.e. temperature, pressure, and equivalence ratio) is identified, nonessential species are detected and eliminated quite straightforwardly, and a proper selection of marginal species is carried out, also according to the desired level of accuracy. As expectable, there is an opposite trend between the effectiveness of a given reduction technique and its computational demand. For this reason, the most performing strategies often consist of faster (and rougher) approaches on the initial mechanism, and computationally heavier (but more accurate) techniques on already reduced models.

This procedure was followed in this work, too. As a first step, the detailed mechanism is reduced through DRGEP, thus cutting out the totally unnecessary species and retaining almost the total accuracy of the original model. Then, a dynamic sensitivity analysis on species is carried out on the uncertain species. As a last step, a sensitivity analysis on the kinetic parameters of the reactions allows the removal of the unimportant reactions. Each of these steps is described in this section, recalling the consolidated theoretical concepts and better focusing on the introduced novelties.

2.1. DRG with error propagation

A complete description of the original approach can be found in the work by Pepiot-Desjardins and Pitsch [17]. Therefore, only the main concepts are recalled, and framed into the overall procedure.

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