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Comparison of different DRG-based methods for the skeletal reduction of JP-8 surrogate mechanisms

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ABSTRACT

Directed relation graph (DRG) techniques are used to generate small skeletal mechanisms capable of accurately simulating the combustion of a two-component surrogate for JP-8 jet fuel. Within the DRG framework, six different reduction techniques are considered, and the effectiveness of different definitions for the connection weights and error propagation is evaluated. The use of DRG reduction techniques for aided sensitivity analysis (DRGASA) and for on-the-fly reduction (flux-based DRG) is studied in detail. An optimal reduction approach based on the sequential use of DRG, DRGASA, and flux-based DRG is proposed. When global reduction is applied to a detailed mechanism of 234 species and 6997 reactions, the six reduction techniques result in very different skeletal mechanisms, but all of them are essentially equivalent in terms of accuracy and number of retained species (82–92 species). Finally, for two-dimensional coflow flame test problems, on-the-fly DRG techniques are investigated. Error-propagation-based methods are found to extinguish the flame artificially and cannot be used in an on-the-fly implementation. Conversely, normal DRG methods greatly improve the mechanism reduction, and accurate solutions are obtained using about 15% of the detailed mechanism.

1. Introduction

In the last five years, the sizes of chemical mechanisms used in combustion modeling have grown by orders of magnitude [1], increasing from 20–30 species for methane combustion to more than 1000 in some recent detailed mechanisms [2]. This growth is driven by interest in the combustion of ever more complex fuel mixtures and in the formation of pollutants. However, numerical solutions are still mostly limited to a few simplified problems such as adiabatic ignition, freely propagating flames, and counterflow flames, in which the fluid-mechanical portion of the problem can be simplified to 1D or even 0D models. The simulation of truly multidimensional flames is still a great challenge, and the solution of even moderately turbulent flames with simple kinetic models approaches the limit of the world's largest computational facilities [3].

A way to bridge the gap between computational fluid dynamics and complex chemistry is to replace the full set of chemical species and reactions by a simpler one. The new smaller model is optimized for a given problem and can thus generate results of the same accuracy at a much smaller computational cost. Some techniques try to eliminate unimportant species, obtaining a so-called *skeletal mechanism* (e.g., sensitivity analysis [4], directed relation graph reduction [5], and path flux analysis [6]). Others operate a *mechanism reduction* in which different timescales present in the chemical reactions are separated and used to eliminate unnecessary degrees of freedom (e.g., quasi-steady-state assumption [7], computational singular perturbation [8], and intrinsic low dimensional manifold [9]). Okino and co-workers survey different reduction techniques in [10], and a recent review by Lu and Law focuses more explicitly on combustion applications [1].

After the pioneering work of Bendtsen et al. [11] who first analyzed the oxidation pathways in combustion using graph structures (see also the earlier related work by Turányi [12]), the idea of using connectivity structures to produce reduction schemes was explored by many authors. In 2005, the original *directed relation graph (DRG)* method of Lu and Law [5] was proposed, in which a connectivity structure is built to quantify the direct coupling between species in the mechanism. DRG-based methods are known for not providing the optimal (i.e., smallest) reduced mechanism [13], but their conceptual simplicity and low computational cost (compared, for example, with sensitivity analysis approaches) have made this reduction approach widely popular.

Many modifications and improvements have followed the initial work of Lu and Law [5].

- Pepiot-Desjardins and Pitsch [14] proposed a different definition for the coupling norm that accounts separately for rates of formation and destruction.

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- In the same publication [14], Pepiot-Desjardins and Pitsch introduced the concept of error propagation in the graph search (DRGEP).
- More recently, Luo et al. [15] used a modified definition of the connection edges to increase the robustness of the method when dealing with mechanisms having many isomer molecules.

The overall implementation of DRG has also been investigated in the literature. Lu and Law [16] found that restarting the reduction procedure can be beneficial, and Zheng et al. [17] designed the DRGASA algorithm, which uses DRG to aid sensitivity analysis reduction. DRG-based methods are automatic (i.e., they require minimal user input) and very computationally cheap; for this reason they are well suited for on-the-fly implementation. Liang et al. [18] used DRGEP reduction to obtain time-dependent chemical mechanisms adaptively. Recently, their approach has been extended by our research group using a flux-based DRG approach that explicitly considers the contribution of transport to the directed graph and allows both time- and space-dependent on-the-fly reduction [19].

An approach that is somewhat similar to DRG methods is the so-called *path flux analysis* (PFA) method [6], in which the mass transfer between species is used to assemble a graph structure for the skeletal reduction. Alternatively, the *error minimization* method of Nagy and Turányi [13] uses the off-diagonal entries in the Jacobian matrix to cluster the species to eliminate. Both PFA and error minimization have been demonstrated to produce slightly smaller mechanisms but require more computationally intensive calculations. In any case, these methods are not considered in the present study because they do not belong to the DRG family of methods in a strictly mathematical sense, i.e., they are not based on the definition of a local coupling norm and the study of its propagation across a directed graph (as explained in Section 2).

In this paper, we focus our attention on a particular set of methods for skeletal reduction that are based on the DRG representation of the coupling between chemical species. We provide a unified mathematical formulation for different DRG methodologies and present a careful comparison among the different methods. More specifically, we consider three different definitions of the graph structure, in conjunction with both standard DRG and error propagation (DRGEP) reduction. We also consider aided sensitivity analysis (DRGASA) as an effective way to shrink further the mechanism size for any method of the DRG family.

This paper is divided into five sections. In Section 2, different DRG and DRGEP methods are defined, and Section 3 discusses on-the-fly implementations. In Section 4, different reduced mechanisms for the combustion of JP-8 are generated via a sequential reduction approach and the results are examined, as follows.

- In Section 4.1, the original DRG reduction method is considered.
- In Section 4.2, further reduction is obtained using DRG to aid sensitivity analysis (the so-called DRGASA method).
- In Section 4.3, the skeletal mechanisms are verified on the solution of two-dimensional (2D) axisymmetric laminar coflow JP-8 flames.
- In Section 4.4, further numerical speedups obtained by on-the-fly reduction techniques are analyzed.

Finally, Section 5 presents conclusions.

2. DRG methods

All DRG-based reduction methods can be summarized in five main steps.

(1) Define a norm (or *connection weight*) to quantify direct coupling between pairs of unknowns.
(2) Use the norm to define a directed relation graph structure that connects the unknowns. In the graph, every chemical species maps to a vertex, and an edge is present between two species if and only if direct coupling exists (i.e., if the species have a reaction in common).
(3) Define a set of *target species* that are necessary for the accurate solution of the problem. A typical choice for combustion problems is the H radical; alternatively, a combination of fuel molecules, oxygen, and combustion products can be used. Pollutants and secondary products should be added to the list of target species if their accurate prediction is of interest.
(4) Starting from the target species, search the directed graph and attribute to each of the vertices an *importance coefficient*. This coefficient quantifies how strongly a given species is connected to the target species.
(5) Eliminate from the mechanism any species whose importance coefficient is below a user-defined threshold, because such species are only loosely connected to the main combustion pathway.

DRG-based methods differ in their definitions of connection weights and importance coefficients.

2.1. Definition of connection weights

The original DRG method of Lu and Law [5] defines the connection weight from species i to species j as a ratio between the chemical activity of the couple i,j compared to the total chemical activity of species j, namely

$$R_{i\to j}^{(\mathrm{Lu})} = \frac{\sum_{\alpha\in\mathcal{C}(i,j)} |\nu_{i\alpha} r_\alpha|}{\sum_{\alpha\in\mathcal{R}(i)} |\nu_{i\alpha} r_\alpha|}, \tag{1}$$

where $\mathcal{R}(i)$ is the set of reactions that pertain to species i, $\mathcal{C}(i,j) = \mathcal{R}(i)\cap\mathcal{R}(j)$ is the set of reactions in which both species i and j participate, $\nu_{i\alpha}$ is the stoichiometric coefficient of species i in reaction α, and r_α is the net reaction rate (forward minus reverse).

Pepiot-Desjardins and Pitsch [14] noticed that Eq. (1) does not distinguish between reactions that create or destroy species i, which they argued could result in a low extent of reduction. They suggest a possible alternative definition:

$$R_{i\to j}^{(\mathrm{Pep.})} = \frac{\left|\sum_{\alpha\in\mathcal{C}(i,j)} \nu_{i\alpha} r_\alpha\right|}{\max\left(\sum_{\alpha\in\mathcal{R}(i)} (\nu_{i\alpha} r_\alpha)^+, \sum_{\alpha\in\mathcal{R}(i)} (\nu_{i\alpha} r_\alpha)^-\right)}, \tag{2}$$

where the operator $(\cdot)^+$ selects only the positive terms in the summation and the operator $(\cdot)^-$ selects only the negative terms and makes them positive. Eq. (2) quantifies the contribution of the species couple i,j to the total rate of formation or destruction of species i. Note that all forward and backward rates must be considered as a single reaction when using the connection weights (2), or else partial equilibrium reactions could result in artificially low connection weights.

Finally, in recent work, Luo et al. [15] noticed that the original formulation (1) can be improved if the detailed mechanism contains many isomers. For this reason, a variation of the method is introduced, in which the maximum norm is used instead:

$$R_{i\to j}^{(\mathrm{Luo})} = \frac{\max_{\alpha\in\mathcal{C}(i,j)} |\nu_{i\alpha} r_\alpha|}{\max_{\alpha\in\mathcal{R}(i)} |\nu_{i\alpha} r_\alpha|}. \tag{3}$$

Eqs. (1)–(3) enable the definition of a directed graph structure in which every species maps to a vertex of the graph and a directed edge (connection) is present only if its R coefficient is not null. A