G-Scheme-based Mechanism Simplification for Hydrogen and Hydrocarbon Ignition

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Abstract

In a multi-scale problem, when the instantaneous temporal gaps between slow and active, and between fast and active time scales become large, the system dynamics can be simplified: (i) the vanishing of the *fast* mode amplitudes related to the fast time scales constrains the active dynamics within a **s**low **i**nvariant **m**anifold (SIM) where the fast processes are in a near-equilibrium regime; (ii) the *slow modes* related to the slow time scales control the long term system dynamics; they are nearly-frozen on the time scale of the currently active time scale; (iii) subsequently, the processes neither in near-equilibrium nor in nearly-frozen states drive the system's dynamics at rates based on the currently active time scales.

The *G-Scheme* [1] is a time accurate computational tool that exploits, adaptively, opportunities for reduction from both or either the manifestation of fast/active and slow/active spectral gaps (which can be fairly small). The characterization of the local structure (local tangent space) of the subspaces associated with the slow, active, and fast scales can be of great significance in the analysis of the dynamics with the aim of achieving a low-dimensional description. The specific features of the *G-Scheme* allow a time-scale-aware sensitivity analysis of the problem, which may be used to simplify/reduce/understand the problem at hand.

In this work, we analyze and construct simplified (skeletal) mechanisms of a number of fuels on the basis of information extracted from the numerical solution of the ignition process by using the *G-Scheme* solver. To identify the reactions mostly contributing to the local dynamics, we generalize the original definition of the CSP *Participation Index* (PI) of a reaction towards a mode to a definition relating the participation of a reaction to the slow, active, and fast subspaces, respectively. This way we can identify the most important reactions associated to each of the 3 subspaces at each step of the numerical integration of the system of equations that describes the autoignition process.

A simplified (skeletal) mechanism for hydrogen, methane, propane, and n-heptane is obtained by taking the union of all the reactions that have a value of the PI above a selected threshold in at least one *G-Scheme* subspace. The accuracy of the results depends on the selected PI threshold: increasing the threshold decreases the number of reactions in the skeletal mechanism and also decreases the accuracy of the results. However, above a certain threshold the skeletal mechanism is not able to reproduce the explosion. Therefore,we will look for the threshold value corresponding to the smallest skeletal set of reactions able to reproduce the explosion stage within the given error tolerance. These simplified mechanisms are shown to be robust with respect to different initial conditions.

In the homogeneous chain-branched explosion, it is possible to identify a short period, the *explosive* stage, in which most of the chemical heat is released. Before this occurs, two other stages are noted: in the first, the *initiation* stage, the change of state is very slow, while in the second, the *chain-branching* stage, there is a rapid increase in the chain-carrier species concentration. The explosive stage itself is followed by the *termination* stage, where stable species are ultimately produced.

Applying the *G-Scheme* analysis to the four stages allows the identification of a simplified mechanism that reproduces the complete combustion process. By applying the same analysis focused only on the *explosion stage*, it is possible to identify a subset of species and reactions that play crucial roles in the explosion mechanism. Such analysis allows us to obtain a much smaller skeletal kinetics mechanism that reproduces the explosion process.

In focusing on the *explosive* stage, we find that it is characterized by a very short time scale, with the dynamics controlled by a pair of real distinct positive eigenvalues. During this period, the two eigenvalues are responsible for the increase in the chain-carrier species (and a decrease of the associated time scales) until they merge to form a pair of complex conjugate eigenvalues with positive real parts. Eventually their real parts become negative and the imaginary parts reduce to zero and thus, subsequently, become a pair of real distinct negative eigenvalues. This dynamics is found in all cases studied. When we evaluate the PI's only for the modes associated with the positive eigenvalues, we are able to determine a specific subset of

reactions that qualitatively capture the explosion stage. We will compare this subset with the one selected on the basis of the *G-Scheme*-based PI to identify similarities and discrepancies.

The simplified (skeletal) mechanisms are compared with other reduced mechanisms that have been proposed. For the low temperature, two-stage ignition of n-heptane, we are able to identify a skeletal mechanism for each explosion stage.

References

[1] Valorani and Paolucci, The G-Scheme: A framework for multi-scale adaptive model reduction. Journal of Computational Physics (2009) vol. 228 (13) pp. 4665-4701