Analysis of an Archetypal Model of Homogeneous Isothermal Chain-Branched Explosions

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Abstract

Chain reactions play an essential role in chemical kinetics. Ignition of hydrocarbon (and hydrogen) fuels is controlled by this class of reactions. The complexities of ignition increase with the length/size of the fuel molecule, e.g., one of the reference mechanism for the oxidation of n-heptane [1], [2], [3] involves 560 species and 2600 reactions. The order of magnitude of the mechanism will become huge when oil sands derived fuel (like asphaltene, $C_{419}H_{498}N_6O_4S_8V$) will become a palatable alternative to the present fuels.

The Computational Singular Perturbation (CSP) Method has been used to analyze two-stage ignition of n-heptane by Kazakov, et al. [4], and Goussis, et al. [5]. They found that in spatially homogeneous systems, the occurrence of two branches of positive eigenvalues characterizes chain-branching and thermal ignition. Gupta, et al. [6] proposed a new criterion based on the CSP importance indices, the sum of the absolute values of the importance indices of diffusion and convection of temperature in the slow dynamics of temperature. They used this criterion to discriminate spontaneous ignition from deflagration in turbulent ignition problems for the automated detection of different ignition regimes at different times and location during the ignition events in HCCI applications. Lu, et al. [7] proposed an explosion index for explosive modes, indicating the contribution of species and temperature in the explosion process aimed at distinguishing radical and thermal runaway.

Given the theoretical and practical importance of identifying the key processes controlling the ignition, we decided to approach the analysis of Homogeneous Chain-Branched Explosions by means of tools developed on the basis of the Computational Singular Perturbation (CSP) Method, the *G-Scheme* [8], and a geometric characterization based on the concept of the local tangential stretching rate [9], which finds its theoretical justification in the theory of normal hyperbolicity viewed on a local level, i.e., pointwisely along system orbits/trajectories.

One outcome of these studies was the observation that the occurrence of two branches of positive eigenvalues is typical of the ignition (both one- and two-stages) of all the fuels we investigated (hydrogen, methane, propane, n-heptane). This indicated the existence of a relatively simple and universal pattern for ignition. Willing to address the details of this pattern with the simplest possible model, we decided to consider a two-dimensional (two unknowns) model for Homogeneous Isothermal Chain-Branched Explosions proposed by Williams [10], and to analyze it using CSP tools and the local tangential stretching rate concept.

The result of the analysis was rather surprising given that the controlling pattern of ignition in the complex hydrocarbon fuel ignition was also found in the model. This pattern involves a first stage during which the two eigenvalues are both real and positive; this stage ends when these two eigenvalues collide to form a complex conjugate pair with positive real parts; later, the complex conjugate pair cross the imaginary axis so that the real parts become negative; finally, the imaginary parts reduce to zero, and a pair of real negative eigenvalues emerge and remain until the system reaches its equilibrium state.

Having established the similarity between the real kinetic problems and the simple model, enabled us to identify different ignition regimes (subcritical and supercritical chain-branching ignition) in the toy model kinetics, and also different stages of ignition on the basis of the nature of the local eigenvalues. We will discuss the evolution along the trajectory of the fast and slow (local) subspaces, and of the occurrence of the pair of complex conjugate eigenvalues.

We also perform a nonlinear analysis of the toy model by examining the *stretching rates* along the system's evolution. Defining the tangential and normal unit vectors along the trajectory, we study the vector dynamics of a local perturbation. The stretching rates represent the rates at which the perturbation evolves in the tangential and normal directions. Finally, after having established the theoretical connection between

the local stretching rates concept and CSP, we will show how the tangential local stretching rate can give a direct indication of the key processes and the associated time scales during all phases of the ignition.

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