DRG AND GQL REDUCTION METHODS FOR A H2/AIR AUTO-IGNITION PROBLEM

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Abstract
Two advanced reduction methods for systems of chemical kinetics, Global Quasi-linearization (GQL) and Directed Relation Graph (DRG), are implemented to reduce a homogeneous combustion system. In this work, the auto-ignition process of H2-air system is considered for a wide range of system parameters and initial conditions. Results based on GQL and DRG reduced chemistry are compared.

Introduction

The interest in the numerical simulation of combustion processes has grown considerably during the last decade due to strict regulations with respect to pollutant formation and efficiency of combustion processes. An auto-ignition problem represents one of the most important and studied topics in combustion theory used for combustion mechanisms development and for their validation [1]. In order to accurately model the auto-ignition a large number of different elementary reactions between many species has to be accounted for. Vastly different characteristic time-scales induced by elementary reactions due to the non-linearity of elementary reaction rates result in stiffness of the governing system of ODEs [1] [2]. Both high dimensionality of such systems and their stiffness lead to an extreme complexity and high computational load with respect to the CPU time and memory storage required to treat combustion systems numerically. Therefore, there is a need to reduce these detailed mechanisms.

At present, significant progress in developing model reduction methodology has been made in a number of studies. It was originated by the Quasi-Steady state Approximation (QSSA) [3] and Partial Equilibrium Approximation (PEA) [4] approaches. The methods treat either species in quasi-steady state or elementary reactions in partial equilibrium [1] [5]. A very transparent example of such approaches is the Directed Relation Graph (DRG) [8], which is used in the current study. All these methods eliminate species and, hence, the resulted reduced models compute only some of the species, thus, reducing the dimension of governing equation systems [1].

Other model reduction approaches use low-dimensional invariant manifolds, namely, the Intrinsic Low-Dimensional Manifold (ILDM) method [6], the Global Quasi-linearization (GQL) approach [7], the Computational Singular Perturbation (CSP) method [9], which allow to keep information about all species involved and

nevertheless reduce the dimension of kinetic models.
In this work, two model reduction techniques, that belong to these different groups: the GQL method and the DRG method, are investigated and compared. The GQL method explores globally the multiple time-scale hierarchy of the system and the reduction algorithm follows the fast and slow invariant manifold concepts [7], while the DRG is based on the construction of skeletal mechanisms, aiming to remove those species having negligible influence on the important species [8].

Mathematical Model
In this work, auto-ignition processes are described by a pure homogeneous systems of ODEs. The mathematical model can be expressed in a general vector form:

$$\frac{d\boldsymbol{\psi}}{dt} = \mathbf{F}(\boldsymbol{\psi}), \quad \boldsymbol{\psi} \in R^n \tag{1}$$

Here ψ is the n-dimensional thermos-kinetic state vector $\boldsymbol{\psi} = \left(h, p, \frac{w_1}{M_1}, \frac{w_1}{M_1}, \cdots, \frac{w_{ns}}{M_{ns}}\right)^T$, where h is the enthalpy, p the pressure and w_i and M_i the mass fractions and molar masses of species. $\mathbf{F}(\boldsymbol{\psi})$ is the n-dimensional vector of thermo-kinetic source term[4].

Global Quasi-linearization (GQL)
The Global Quasi-linearization (GQL) approach stems from the singular perturbation theory and aims at the appropriate approximation of the low-dimensional manifold describing the system dynamic generically [7] [10]. The GQL is based on an assumption that the system dynamic can be decomposed and a small "hidden" parameter of the system determining the main disparity of time-scales exists [7] [11]. According to this assumption, the vector field $\mathbf{F}(\boldsymbol{\psi})$ can be globally linearly approximated [7]:

$$\mathbf{F}(\boldsymbol{\psi}) \cong \mathbf{T}_{\mathrm{GQL}} \cdot \boldsymbol{\psi} \tag{2}$$

The Eq. (2) shows that within the GQL approach we look for a linear mapping $\mathbf{T}_{\mathrm{GQL}}$ that transforms some thermo-kinetic state vectors $\boldsymbol{\psi}$ to the vector field of source term $\mathbf{F}(\boldsymbol{\psi})$. Once this mapping is found we can follow the main idea of ILDM method [6] and a time-scale separation can implemented to yield the eigenvalue decomposition of the GQL matrix $\mathbf{T}_{\mathrm{GQL}}$ identifying the fast and slow invariant subspaces:

$$\mathbf{T}_{\mathrm{GQL}} = \begin{pmatrix} Z_s & Z_f \end{pmatrix} \cdot \begin{pmatrix} N_s & 0 \\ 0 & N_f \end{pmatrix} \cdot \begin{pmatrix} \tilde{Z}_s \\ \tilde{Z}_f \end{pmatrix}, \tag{3}$$

here N_s and N_f denote the diagonal matrices of the two groups of eigenvalues on the diagonal, and Z_s and Z_f the relevant base of slow and fast eigenspaces. Then, similar to the assumption of the ILDM method [6], the fast relaxed processes define an approximation of the low-dimensional slow manifold within the thermo-kinetic system state space as $\tilde{Z}_f \cdot \mathbf{F}(\boldsymbol{\psi}) = 0$. Then, the implementation scheme of

the GQL approach can be applied to solve the fast and slow subsystems. For the fast subsystem, one can solve [11]:

$$\begin{cases} \dfrac{d\boldsymbol{\psi}}{dt} = Z_f \cdot \tilde{Z}_f \cdot \mathbf{F}(\boldsymbol{\psi}) \\ \tilde{Z}_s \cdot \psi = \tilde{Z}_s \cdot \psi_0 \end{cases}, \tag{4}$$

and the slow subsystem governs the system dynamics on the low-dimensional slow manifolds [11]:

$$\begin{cases} \dfrac{d\boldsymbol{\psi}}{dt} = Z_s \cdot \tilde{Z}_s \cdot \mathbf{F}(\boldsymbol{\psi}) \\ \tilde{Z}_f \cdot \mathbf{F}(\boldsymbol{\psi}) = 0 \end{cases}. \tag{5}$$

Directed Relation Graph (DRG)

The empirical Directed Relation Graph (DRG) approach, developed by Lu and Law in 2005 [8], is a reduction method for the construction of skeletal mechanisms. The aim of the method is to efficiently resolve the species coupling, such that those who have a little or none influence on the important species can be removed.

The DRG is applied to identify the unimportant species and remove the elementary reactions that are not associated with them, using an empirical criterion. Accordingly, the contribution of species B in the production/consumption rate of species A can be quantified through the normalized index r_{AB} [12], given by

$$r_{\mathrm{AB}} = \frac{\sum_{i=1}^{n_r} |\nu_{A,i} \cdot \dot{\omega}_i \cdot \delta_{B,i}|}{\sum_{i=1}^{n_r} |\nu_{A,i} \cdot \dot{\omega}_i|} \tag{6}$$

where $\nu_{A,i}$ is the stoichiometric coefficient of A in reaction I, $\dot{\omega}_i$ is the reaction rate and $\delta_{B,i}$ is

$$\delta_{B,i} = \begin{cases} 1, & \text{if the i}^{\text{th}} \text{ elementary reaction involves species B} \\ 0, & \text{otherwise} \end{cases} \tag{7}$$

The terms in the denominator of Eq.(6) measure the contribution of reactions to the production/consumption of species A, and the terms in the numerator are those from the denominator involving species B only [12].

Defining a threshold limit value ε ($0 < \varepsilon < 1$), and if index r_{AB} is larger compared to this threshold, then removing species B, can induce error in the production of species A, so that species B must be maintained in the skeletal mechanism. Usually, species A are chosen as those who have some desirable chemical attributes that the reduced mechanism should reproduce [13]. The resulting skeletal mechanism obtained has errors according to the user-specified threshold ε for the conditions under which it is developed [8]. Therefore, mechanisms with different levels of accuracy can be obtained by assigning different values for ε.

From a practical point of view, the index r_{AB} defines an error estimate in predicting

species A if B is neglected. Therefore, consumption and production reactions must be considered altogether [13].

Results and Discussion

As a test example, the hydrogen-air system represents a simple combustion system but still sufficient for comparison of these two approaches: GQL and DRG. The detailed chemical kinetic model [4] consists of 9 species (including the inert gas N_2 but NO_x formation is not taken into account).

For DRG calculation applied in this work the following procedures are conducted: first, the target species, which will play the role as A species in the index r_{AB}, are defined, consisting of H_2, O_2 and H_2O. The detailed mechanism is used to simulate an ideal gas reactor and to estimate ignition delay times. For each time step, the DRG is applied using concentrations and temperature from the reactor calculations, and the species with the index greater than the threshold are retained and stored. The final set of species consists in the union of the specie's set from each time step of the reactor calculation. The reactions containing only those species then are selected to be in the skeletal mechanism. Note that the DRG calculation is problem dependent. Therefore, if one only focuses on high temperature range or normal pressures, DRG calculation shows that H_2O_2 can be removed. In this case, one obtains skeletal mechanism with 13 elementary reactions and 8 species (without H_2O_2). However, since H_2O_2 plays important roles in high pressures [4], if one should obtain a satisfactory accuracy over a wider range including high pressures, H_2O_2 should be considered as well and, in this case, all species should be retained in the reduced skeletal model.

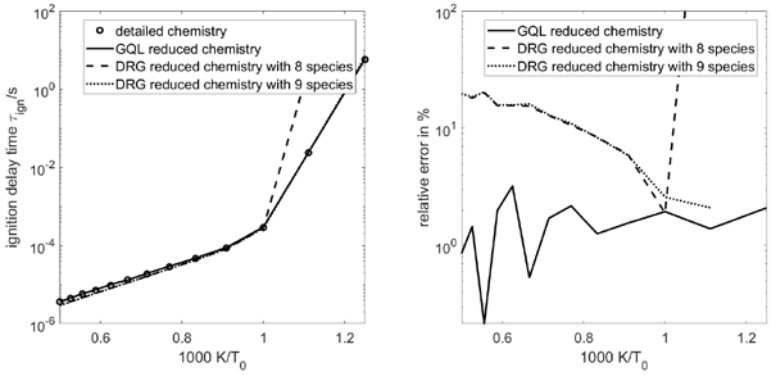

Figure 1. Dependence of the ignition delay time on the initial temperature at p_0=1bar and $\Phi = 1.0$.

The GQL approach for hydrogen-air system has been discussed in [10], where a 4-dimension (4D) reduced model was achieved by using the low-dimensional manifold concept. At present the system (5) represents the reduced model, where the manifold in an implicit form is taken into account and it comprises a reduced model. The system is integrated as a system of Differential and Algebraic

Equations (DEAs). Then, the solution of the system (5) is compared to the solution of the system (1).

The figures illustrate the performance of both approaches and show the dependence of the ignition delay time on the initial temperatures (Fig. 1) and initial pressures (Fig. 2). At first, one can see for the studied range that the GQL reduced model (solid lines) can predict the ignition delay time very well with relative errors less than 2%. The DRG method performs as following. If H_2O_2 was not considered (dashed lines), then relative errors are relatively large at low temperatures (T_0 <1000 K) and at high pressures (p_0 >10 bar). However, if H_2O_2 is considered (point lines), then relative errors remain rather small (less than 15%) for the whole studied range. It should be emphasized that the hydrogen mechanism is already a small mechanism (19 reversible elementary reactions) and all species are strongly coupled. As it is shown by the results outlined, the GQL can predict the auto-ignition processes for a wide range of initial conditions with less number of reduced dimension and more accurately. The DRG, on the other hand, has the advantage of being represented by the skeletal mechanisms, which can be straightforwardly implemented.

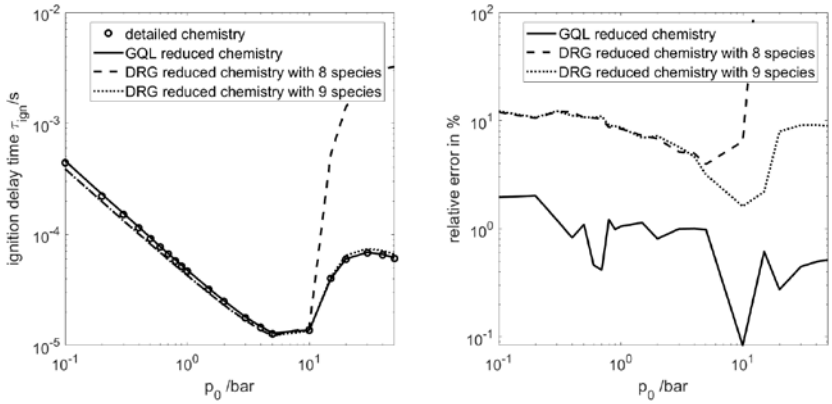

Figure 2. Dependence of the ignition delay time on the initial pressure at T_0=1200 K and $\Phi = 1.0$.

Conclusion

The Global Quasi-linearization (GQL) and Direct Relation Graph (DRG) methods were discussed and their performance was compared for the auto-ignition problem of hydrogen-air system. The results of the comparison were validated in a very wide range of initial conditions ($800 \leq T_0 \leq 2000$ K, $0.1 \leq p_0 \leq 50$ bar). It was shown that for the whole considered range, the relative errors by using the 4D GQL reduced chemistry were often less than 2%. For the DRG method, since hydrogen mechanism is already small, all species are strongly coupled and the only specie that could be withdrawn from the detailed mechanism to obtain the least number of species and elementary reactions is the H_2O_2. But in this case, the results will show less accuracy for high pressures and low temperatures. However, for large

mechanisms, DRG will produce a higher reduction in the dimension of the resulting skeletal mechanisms.

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