On a number of degrees of freedom of a homogeneous combustion system

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Abstract—**In order to model ignition processes accurately a very detailed description of the chemical kinetics is needed. Therefore, typical combustion mechanisms have become impractically large and detailed during last few decades. The problem of an optimal dimension (number of parameters to consider) to model the ignition process has become very important but is not answered yet in general case. In the current work we discuss coordinate free approaches of model reduction to access this problem. A combination of local (ILDM based) and global (GQL based) characteristic time scale analysis for mechanism reduction is outlined. Results and methodology are benchmarked and illustrated by considering rich and lean methane/air combustion systems.**

I. Introduction

The question about actual number of degrees of freedom (that corresponds to the number of equations to be solved) exhibited by a reacting system is a very important and yet an open question of model reduction (see e.g. [1-2]). This question, however, cannot be answered in the general case. It strictly depends on the phenomena under consideration and on the appropriate level of accuracy required in the application.

In engineering applications this problem leads to the usage of very large, detailed mechanisms. Usually, these have not only a very high dimension but are extremely stiff and non-linear (see e.g. [3]). This is because recently the mechanism development process mostly relies on compilation of different detailed mechanisms and uses mostly empirical generic roles [4,5].

However, it seems that (depending on the problem at hand) it is not necessarily to describe the chemical kinetics with such very high level of sophistication. This would mean that reduced reaction mechanisms have to be devised for any particular application maintaining the optimal balance between the accuracy and complexity (in terms dimensionality, non-linearity, functional simplicity etc.).

Several methods have been devised to support optimal mechanisms development (constructing reduced models) often by using different post-processing procedures [6]. For example by implementing so-called sensitivity analyses and analyzing the reaction paths species can be identified, which are only by-products or reaction, i.e. which do not govern the overall dynamics of the system. Therefore, they can be excluded from the mechanism. In this way the so-called skeletal mechanisms are developed (see e.g. [6] for details and additional references).

Nowadays, efficient computational tools are available, which perform this post processing automatically. Depending on the level of simplification one can use them to obtain either skeletal mechanisms or global mechanisms (oversimplified mechanisms where several reaction steps are used only).

In our opinion reduced models processed in this way have a natural limit: There is no much hope to extend and improve the performance of the existing reduced models if one requires the relatively high accuracy for a wide range of system parameters, and whenever one is at the same time restricted to the original coordinate systems (i.e. if one tries to formulate the reduced description by manipulating the species and elementary reactions only). At some point, and most investigators agree on that, no further reduction is possible in this way without significant damage to the overall mechanism performance. For instance, in combustion of low hydrocarbons like methane the limit is about 25-30 dimensions / species [3], i.e. ~30 parameters are needed to be modeled to obtain reliable numerical results with such mechanisms, for n-heptane the limit is about 35-40 etc.

Hence, in order to overcome these limits, one has to develop more comprehensive tools and mathematical methods to handle this problem without being restricted to a particular system representation. In the current work we address this problem. The main emphasis of the study is made on the estimation of number of real degrees of freedom (i.e. reduced model dimension) to be accounted for a reliable description of chemical kinetics in the ignition problem.

II. ILDM and GQL

A. Homogenous system - chemical kinetic source term

The equations of the system governing the chemical reaction in homogeneously stirred reactor can be described by the evolution of thermo-chemical state vector $\psi = (\psi_1, \ldots \psi_n)^T$ in time:

$$\frac{d\psi}{dt} = F(\psi) = S \cdot R(\psi), \quad \psi \in R^n, \quad n = n_s + 2 . \qquad (1)$$

where ψ_j represents such quantities as the pressure of the mixture - p , the enthalpy - h , i.e. two thermodynamic quantities ($n = n_s + 2$), and mixture composition (e.g. by using specific mole numbers $\psi_i = w_i / M_i$). F represents the chemical kinetics mechanism of n_s chemical species

participating in n_r elementary chemical reactions. $S_{n_s \times n_r}$ is the stoichiometric matrix and $R(\psi) = (R_1(\psi), \ldots R_{n_r}(\psi))^T$ is the vector of elementary reaction rates, which are highly non-linear functions of certain type depending on components of the system state vector [3]. The chemical reaction source term (1) represents a composition of linear with matrix *S* and non-linear (elementary reaction rate functions *R*) mappings.

B. IDLM – local chatrachteristic time scale analysis

One of major breakthroughs in model reduction was the development of local characteristic time scale analysis and development of the manifold's based model reduction concept [7,8]. The problem of model reduction was formulated by using the concept of decomposition of motions and was based on coordinate invariant properties of the vector field defined by the system of ODEs (1) describing the mechanism of chemical kinetics. The eigenvalues of the system's Jacobian matrix are used to estimate characteristic time scales while the corresponding eignespaces are used to decompose the vector field and define coordinate free representation of the Intrinsic Low-Dimensional Manifolds (ILDMs) [8]. The geometrically invariant structure of the system Jacobian

$$J(\psi) = \left(dF_i / d\psi_j\right)_{i,j=1}^{n}$$

with respect to coordinate representation and the invariance of the ILDM equation (with respect to the first order correction to the manifold invariance condition) made the ILDM approach robust and efficient tool for model reduction. However, the strict dependence on the state of the characteristic time scales (eigenvalues) and invariant subspaces (eigenspaces) of the Jacobi matrix makes the problem of definition of reduced model dimension very difficult especially in the context of the ignition problem. Typically, one has a situation that the so-called gap condition [8,9] (time scale gap between the eigenvalues) is not uniformly valid in the considered part of the system state space.

C. GQL and coordinate free singular perturbations

The framework of Singularly Perturbed Vector Fields (see [9] for more references and details) represents a coordinate free generalization of the method of singular perturbations [10]. It assumes that for any given problem there is suitable (non-linear in general) frame of coordinates in which the decomposition into so-called fast and slow motions can be represented explicitly. In this respect fast relaxation processes will be realized along the coordinates that can be used to parameterize the so-called fast manifolds globally (manifolds describing the fast system motions).

Accordingly, the Global Quasi-Linearization (GQL) technique was emerged from the SPVF framework [9] as a natural extension of the idea to use original variables as e.g. QSSA, but now linear combinations are permitted (in the line with the method briefly described in [11]). This additional assumption about linearity (which, however, should not be restricted to any original coordinate sub-spaces) of fast manifolds simplifies considerably the application and analysis of the fast and slow manifolds and the decomposition itself.

Thus, in current work we show how these two complimentary methods can be combined to resolve the question of optimal reduced model dimension. The local analysis (ILDM) is used to estimate possible dimension (by the gap condition [9]) and global analysis (GQL) is used to verify it for a wide range of system parameters and equivalence ratio. The methane/air rich and lean systems in the auto-ignition problem are used to benchmark the method. We also show how to identify most suitable linear combinations of the original variables, which can be used to identify the slow manifold and represent the decomposition explicitly. Furthermore, it provides with the detailed implementation scheme, allows us rigorous treatment of reduced models and their systematic improvement.

III. SUMMARY

In this work, the problem of optimal reduced model dimension is discussed. The coordinate free methodology to deal with natural assumption of the existence of multiple-scales and the decomposition of motions are in the focus of the study.

Two complimentary coordinate invariant approaches aiming at definition of the decomposition and of the invariant manifold of fast/slow motions are discussed. The suggested combination of the ILDM and GQL approximates not only the slow system dynamics / manifolds, but the fast system dynamics as well. It can be used to identify the reduced model dimension globally and improve considerably the model reduction of mechanisms without significant damage to their overall performance.

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