Global Quasi Linearization (GQL) for the automatic reduction of chemical kinetics

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Abstract
In the present work a new approach for automatic reduction of chemical kinetics models, the so-called Global Quasi-Linearization (GQL) method is discussed. The suggested approach is a result of an analytical study of the ILDM method from the point of view of the general framework of singular perturbation theory. This method has been developed in order to overcome difficulties of the theoretical concept of slow manifolds in simulations of reacting flows. The method is implemented within the standard ILDM method and applied to a simple, but meaningful combustion chemistry model, namely, the carbon-monoxide system. The results of the simulations show a good agreement of the detailed model with reduced one and demonstrate the potential of the suggested method.

1. Introduction

The fact that the use of detailed chemical kinetics in complex combustion phenomena is very important for accurate predictions has been commonly accepted in the last [1]. However, due to the large number of species and reactions, the computation of cost is typically high with detailed mechanisms and prohibits the use of detail chemistry in engineering applications especially with complicated geometries. Moreover, the mathematical model for the conservation laws of reacting flows is highly nonlinear with governing processes (i.e. hydrodynamics, molecular transport, chemical reactions) and the underlying processes occur at time scales differing by many orders of magnitude. This drastic disparity in time-scales and nonlinearities result in the so-called stiffness of the mathematical model and, therefore, courses serious difficulties in the numerical solution [2-4].

Therefore, simpler overall chemical kinetic models, which reduce stiffness and the system dimension, are desirable. Mechanism reduction has been extensively explored, and a range of methodologies has been developed in the past several decades to obtain simplified models of chemical kinetic (see e.g. [4] for more references and a detailed discussion).

In the last decade substantial progress has been made in model reduction methodologies both theoretically (see e.g. [5] for more references) and for practical/numerical applications (see e.g. [6, 7]). In general, model reduction can be achieved by a number of different mathematical methods. Chemical model reduction is often based on the observation that the full chemical kinetics accesses a small part of the state space during the combustion process. This is because time scales differing by orders of magnitude characterize the detailed system of equations, which governs the reacting flow. The wide range of time scales appearing in a typical combustion system causes the existence of low-dimensional manifolds in the state space, which possess very important properties like invariance and exponentially attractiveness for the system trajectory flow. The solution trajectories of such systems evolve from an arbitrary initial state with domination of the fast reactions, which equilibrate quickly. Thereafter, the evolution of the system is governed by the slow reactions along what is called as the slow manifold. Thus, when the fast part of the system dynamics is not so important, the composition can be assumed to be limited only to the slow manifold during the whole reaction/combustion process.

Accordingly, if the slow manifold in the state space is described by means of a smaller number of species concentrations or other parameters, then the governing system of differential equations are required to describe merely the behaviour of those parameters. The other species concentrations or the whole state space can be recast from values of these parameters by using functional relations defining the manifold and, therefore, do not require the solution of expensive differential equations system of higher dimension. The use of the slow manifold reduced chemistry provides an additional computational advantage, because the elimination of the fast reactions reduces the stiffness permitting larger time steps during numerical integration.

2. Motivation and Specific Objectives

As a motivation to the present work, let us list the following questions and problems, which arise in the application of the slow manifold concept. The most important problem concerns the local character of all known methods [4], which are based on a linearization of the non-linear vector field defined by Right Hand Side (RHS) of the system of governing equations. Hence, the global behavior of the system might not be represented appropriate by such an analysis. Note that a global analysis is very important form a point of view of

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correct representation of the reduced system of differential equations in terms of reduced parameters or variables.

A next disadvantage and drawback of current methods is a lack of proper tools for the identification of the fast system behavior. Obviously, the slow manifold does not reflect the fast subsystem behavior. Moreover, most reduction techniques completely neglect the fast transient period of the system behavior. It is often assumed that, after a short transient period of time the system relaxes onto the low-dimensional slow manifold, but where the fast par of the system trajectory ends up on the manifold and how its behavior is projected (as a system of differential equations) onto the manifold are open questions for many automatic reduction methods. In this work, we address these issues and, in particular, solve the problem with projection of initial conditions onto manifolds.

The next question concerns the needed reduced dimension, which is also a weak point of current approaches. Usually, it is defined according to the user specified tolerances [8, 9], but again has local character and might be different in different parts of the state space.

Additionally, it is of great interest in many engineering applications to construct an appropriate tool, which would have only moderate accuracy, but allows to calculate the manifold very fast and robust with a good parameterization for a correct projection.

In this work we address these issues and discuss the following aspects of the model reduction:

- the problem of global identification of the fast and slow decomposition of the original system,
- a significant simplification of the procedure of the slow manifold identification and description,
- an approximation of the fast subsystem, which allows the application of the fast subsystem variables for parameterization of the slow manifold permitting more robust projection procedures.

In particular, we propose an algorithm solving at least to a large extent these problems, which is highly efficient, simple to apply and can be employed for fast reduction when the dynamical identification of the slow manifold is needed.

3. Methodology

3.1 Mathematical model

Mathematically, the decomposition means that we are looking for new coordinates representing the original system in the form of a Singular Perturbed System (SPS) [10, 11]. A key mathematical concept for the novel reduction is that of Singular Perturbed Vector Fields (SPVF) notion [12]. In a number of works [12-14], the theory of SPS [10] is reviewed and extended with special emphasis on a coordinate free approach. The main idea of our approach is an evaluation of such change of the system coordinates that would provide us with a necessary decomposition. For this decomposition all the machinery of powerful methods of SPS can be applied to analysis, formulation of the reduction method and evaluation of the reduced system dynamics.

Outline the main stages of the approximation of the new coordinate system and describe the implementation scheme. In this work we restrict ourselves by considering homogeneous systems described by autonomous ODE (1). An extension to more general systems of PDEs is the subject of our future research. Let us consider very general form of the system of governing equations

$$\frac{d\psi}{dt} = F(\psi), \quad \psi \in \Omega \subset R^n . \qquad (1)$$

Here the state vector ψ is the n-dimensional vector $\psi = \left(h, p, \frac{w_1}{M_1}, ..., \frac{w_{n_s}}{M_{n_s}} \right)$, where h denotes the enthalpy, p the pressure, $w_1, ..., w_{n_s}$ are the species mass fractions and $M_1, ..., M_{n_s}$ the molar masses ($n = n_s + 2$). $F(\psi)$ is the n-dimensional vector of the thermo-chemical source term and t denotes the time.

The suggested approach is based on two main assumptions, first of all on the assumption that there exist a decomposition because of different time scales and, secondly, we suppose that it is valid (within a certain accuracy) everywhere inside our fixed domain Ω, which we call as the domain of interest. The following construction allows us define a linear approximation to the vector field defined by RHS of (1) (so called Global Quasi-Linearization) that leads to the desired decomposition.

3.2 Suggested method

In the present work, a variation of the GQL procedure [14] is discussed, which was motivated by numerical simulations. A modification needed, because in many practical situations the system has a dimensional form and, therefore, cannot be used for a global multi-scale analysis without a proper non-dimensionalization procedure. Otherwise the system hierarchy is perturbed by naturally different scales of the system variables. We overcome here this problem suggesting a proper normalization procedure for systems in dimensional form.

Suppose $F(\psi)$ is a vector field, which satisfies our assumptions about a "hidden" small parameter of the system, which determines the main disparity of time scales. The main steps of the reduction are the

following.

- First, we select n linearly independent points (vectors) $\psi_1,...,\psi_n \in \Omega$ in such a way that the set of vectors defined by the vector field $F(\psi_1),...,F(\psi_n)$ is also linearly independent; this is possible because we assume additionally that all linear integrals define the conserved subspace of the system were removed from its final form (1).

- These vectors form the columns partitioning of the matrices

$$\bar{\bar{F}} = [F(\psi_1),...,F(\psi_n)],\ \bar{\bar{\Psi}} = [\psi_1,...,\psi_n].$$

The matrix $T = \bar{\bar{F}}\,\bar{\bar{\Psi}}^{-1}$ is the GQL of the system (1). It has a simple geometrical interpretation. It is the matrix for the linear mapping that transforms the vectors $\psi_1,...,\psi_n$ to $F(\psi_1),...,F(\psi_n)$;

- Finally, we exploit the invariant eigenspaces of the the matrix T and define the decomposition. If the matrix T is decomposed into invariant subspaces

$$T = \begin{pmatrix} Z_s & Z_f \end{pmatrix} \begin{pmatrix} N_s & \\ & N_f \end{pmatrix} \begin{pmatrix} \tilde{Z}_s \\ \tilde{Z}_f \end{pmatrix} \qquad (3)$$

then, similar to the assumption of the ILDMs method (see e.g. [15, 16] for more details), the fast relaxed processes define an approximation of the low dimensional manifold within the state space as

$$\tilde{Z}_f F(\psi) = 0, \qquad (4)$$

i.e. the manifold where the reaction rates in direction of the fast processes vanish defines an approximation to the slow manifold. The invariant eigenspaces define a reduced system dimension since Z_s and $\tilde{Z}_s$ are the right and left invariant subspaces belonging to the m_s eigenvalues having the smallest real parts (N_s) and Z_f and $\tilde{Z}_f$ are the right and left invariant subspace, correspondingly, related to the fast relaxing processes with m_f eigenvalues (N_f) having the largest negative real parts respectively ($m_s + m_f = n$). The equation (4) is then defined globally in Ω and it implicitly represents an m_s-dimensional slow manifold in the state space.

It has to be mentioned that the present algorithm is similar to the ILDMs concept with the difference of using GQL - T instead of the system Jacobian matrix. Consequently, all previous developments of the ILDM can be applied to implement the method with only minor changes in the numerical code structure. Furthermore, because T is defined globally, the time consuming decomposition of the matrix has (in contrast to ILDM) only to be performed once in the generation of the manifold. The reduced system dimension is defined as a gap between the N_s and N_f eigenvalues, in such a way that the absolute value of the ratio of the maximal eigenvalue by absolute value in small group N_s to minimal in large part N_f stands for a small system parameter. Thus, after we get the linear decomposition, high order approximations according to standard SPS approach allow improving the decomposition itself and increase performance of the method. The last is the subject of forthcoming works.

3.3 Choice of reference vectors/points

Of course the efficiency and the accuracy of this procedure depends essentially on choice of $\psi_1,...,\psi_n$. Roughly speaking, practical recommendations for the choice are following: $F(\psi_1),...,F(\psi_n)$ should not be too close because it can result in degeneration of the matrix $\bar{\bar{\Psi}}$. Additionally, the values of vector field $F(\psi_i)$ should represent "different" behavior for different $i = 1,..,n$. This choice is a crucial point of the algorithm and must be adapted to every particular model. At present, the following algorithm for choice of reference set of points/vectors is proposed. Note, however, that other possibilities of the choice of reference points can exist, but theoretically, their results differ only in higher orders of approximations with respect the system small parameter (see e.g. [13, 14] for details and the definition (6)).

- First, by performing quasi-stochastic uniform distribution, an "initial set" $S_N = \{\psi_1,...,\psi_N\}$ consisting of points ($N >> n$) uniformly distributed in the domain Ω is formed.
- Then we calculate mean value of the vector field over the sequence S_N as $\bar{F} = \frac{1}{N}\sum_{i=1}^{N} F(\psi_i)$, and take a subset of S_N as following

$$S_K = \{\psi_i \in S_N : \|F(\psi_i)\| > \|\bar{F}\|\} \qquad (5)$$

where $i = 1,..,K$, $K = k \cdot n,\ k >> 1$. The set S_K is called the "control set". It consists of the points, which are sufficiently far away from the slow manifold and, therefore, can be safely used to estimate the fast subspace (see e.g. [13, 14] for more details). Note that any subset of length of n of the control set S_K can be

used as the reference set to get T, but there is a degeneration problem of the chosen subset of the control set if some of points/vectors are close one to another or some points are still close to the slow manifold. This can lead to a degeneration of the matrix $\overset{=}{\Psi}$ and, consequently, to a wrong decomposition. Accordingly, not every subset of S_K of length of n can be used as the reference one. To overcome this problem we need to take a subset of vectors such that it spans the simplex of volume that can be compared to the volume of the domain Ω. To resolve this problem we suggest further exploiting existing decomposition. Accordingly, we form arbitrarily sequences of length of n from the control set and choose the best one, which gives the best decomposition (small parameter). Namely,

- we build up the sequence of GQL approximations $T_i, i = 1,..,k$ based on subsets of the control set

$$\{\psi_{(i-1)n+1},...,\psi_{in}\}, i = 1,...,k;$$

- The final reference sequence $\{\psi_{(i^*-1)n+1},...,\psi_{i^*n}\}$ and the final GQL approximation are found simultaneously as $T = T_{i^*}$ by a maximal gap for given dimension of the reduced model m_s:

$$i^* : \max_i \left(\frac{|\lambda_{m_s+1}(T_i)|}{|\lambda_{m_s}(T_i)|} \right), \qquad (6)$$

where $\lambda_j(T_i), j = 1,...,n$ are eigenvalues of T_i ordered in increasing order by absolute values. Then, the reciprocal value $\varepsilon = \left(\frac{|\lambda_{m_s+1}(T_{i^*})|}{|\lambda_{m_s}(T_{i^*})|} \right)^{-1}$ defines a small system parameter. As it was mentioned above, once we have the decomposition and the small parameter then, following the SPVF, its improvement up to arbitrary order of magnitude is possible [14]. The suggested algorithm has been successfully implemented in the numerical code for the standard ILDM.

4. Results and Discussion

As a test example the syngas/air combustion system has been chosen. This model is not too complicated, but nevertheless, sufficient for illustrations and introduction of the suggested method. The chemical kinetic model consists of 13 species [17]. The overall dimension of the considered example is then equals to 15. A detailed system solution and the manifold mesh are generated by using the in-house HOMREA code [18, 19]. The program was originally developed to simulate homogeneous reactors, later it was extended to tackle the ILDM table generation and then used for the IC engine cycle simulation. In numerical simulations we define the domain of interest on a basis of the system solution trajectories. Namely, we take a sequence of the system trajectories $\psi_j(t) = \psi(\psi_j^0; t)$ starting at different initial states ψ_j^0 and calculate the maximal values for the species molar numbers over this sequence

$$\psi^i_{max} = \max_{j,\, t>0} \psi^i(\psi_j^0; t). \qquad (7)$$

The maximal values then determines the boundary of the domain $\psi_{max} = (\psi^1_{max},...,\psi^n_{max})$, except the conserved quantities which values remain equal to the initial ones. Thus, the domain of interest in our case is

$$\Omega = \{\psi \in R^n : 0 < \psi^i < \psi^i_{max}, i = 1,..,n\}.$$

The implementation scheme introduced above one can evaluate the GQL manifold by equation $\tilde{Z}_f F(\psi) = 0$, fast subsystem (up to a leading order of approximation)

$$\begin{cases} \frac{d\psi}{dt} = Z_f \tilde{Z}_f F(\psi) \\ \tilde{Z}_s \psi = \tilde{Z}_s \psi_0 \end{cases} \qquad (8)$$

and the slow subsystem governs the system dynamics on the manifold

$$\begin{cases} \frac{d\psi}{dt} = Z_s \tilde{Z}_s F(\psi) \\ \tilde{Z}_f F(\psi) = 0 \end{cases} \qquad (9)$$

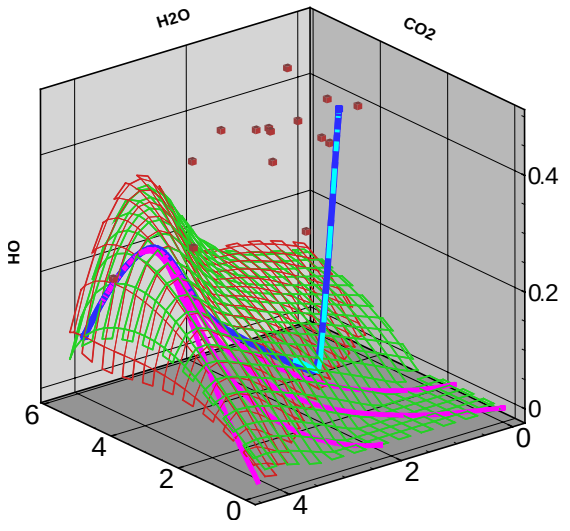

Fig. 1. The state space projection onto the so-called major species molar numbers H_2O-CO_2-OH.

In the case of the standard ILDM and similar approaches uses Jacobian matrix, this representation is defined locally, because the dependence of the transformation matrices $\tilde{Z}_s, \tilde{Z}_f$ on the point in the state space.

In figures 1-3 a typical structure of 2D slow manifold together with results of application of the method is presented in projections to different species subspaces. The solid blue line is the solution trajectory of the detailed system (1); the initial state for this solution was taken to be close to a boundary of existence of ILDM manifold, which is represented here by red mesh. The solid cyan line shows the fast subsystem solution (8). The green mesh shows the slow manifold based on GQL method, where by red cubes the final set of reference point is presented, so exactly those points were used to obtain the decomposition.

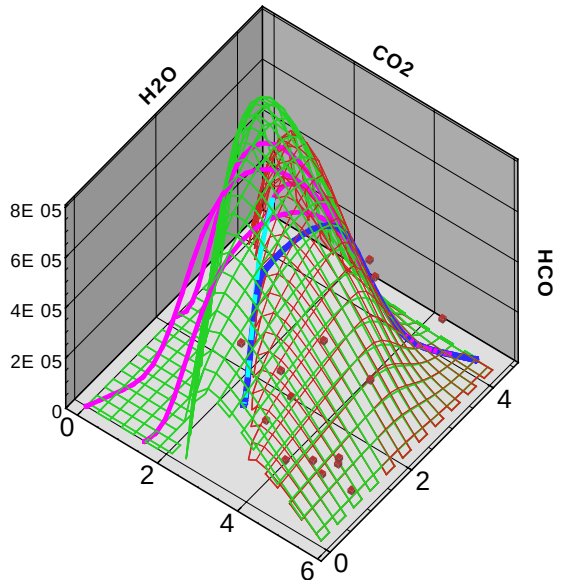

Fig. 2. The state space projection onto the so-called minor species molar numbers H_2O-CO_2-HCO.

At first glance, we can see that the GQL manifold is close to the ILDM manifold where it exists, but starting near the boundary of existence it deviates from the ILDM. This fact is in agreement with theoretical predictions [22]. The explanation is that the ILDM manifold, as it was shown in a number of works [21, 22], defines the first order approximation being applied to the standard SPS system, while our current methods provides us with the zero order approximation. If the small system parameter tends to zero we should not see a difference between two approaches, but because the assumption is only asymptotically valid, we obtain these differences. The small parameter for the 2D slow manifold varies in this case from 0.005 to 0.02 depending on realization of the algorithm due to stochastic nature of the reference point's choice. The slow manifold, however, does not reflect this. In different realizations, there were insignificant deviations from one realization to another near the boundary only.

Let us consider this part of the domain in more detail, according the SPS theory, in the vicinity of the ILDM boundary there is so called turning manifold that separates two distinct parts of the slow manifold its attractive and repulsive parts [12], moreover, close to the turning manifold first order approximation is far away from the real system dynamics.

Now, to illustrate this, let us compare the constructed slow manifolds with the stationary flame structure. In figures 1-3 by magenta lines stationary solutions of the free flat laminar flames [20] are presented. It is seen, in particular for minor species (Fig. 1 and 2), that stationary system solutions approach the slow system manifold along the repulsive part of the system invariant manifold (see close to the boundary, where the ILDM vanishes). In this region the 2D dimensional manifold does not approximate properly the system dynamics and we should increase the dimension.

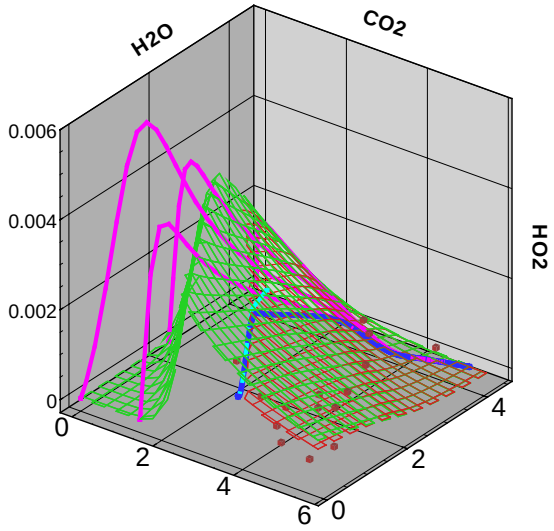

Fig. 3. The state space projection onto the so-called minor species molar numbers H_2O-CO_2-HO_2.

Although, there are some problems with our new method right near the origin (see [23] for instance), it nevertheless allows better approximation of the reduced system dynamics without any increase of the system dimension in the so-called boundary domain [23].

Finally, even if we do not take into account the fact of wider range of the manifold existence, the new method (of course for relatively small system parameter) being much simpler then the other methods, agrees well with both the ILDM and detailed system solutions. Hence, there is a potential of this approach and we are going to improve it in our further works in order to increase its accuracy. This is possible by considering higher order approximations according to the standard SPS theory, which now can be applied with all its powerful, sophisticated and well developed methods.

Furthermore, the application of the suggested

method to more complex reacting flows and geometries as well as to turbulent flows will also be subject of our future research.

5. Conclusions

A new method of automatic mechanism reduction is developed in the current work. It is a realization of the general scheme of an automatic reduction procedure based on the ILDM approach, with the difference that an invariant manifold of low dimension and system decomposition is approximated by the developed GQL procedure instead of the system Jacobian.

In principle, the suggested approach follows well-known ideas from the standard ILDM method, but there are some important novelties making it very efficient in realization of the general scheme of automatic reductions. First of all, the global decomposition is found using invariant subspaces of the GQL in the whole domain of interest of the state space. This allows us to use developed ILDM codes to implement the method. A next, the reduced system dimension is defined by the spectrum analysis of the GQL and the structure of the decomposition remains constant throughout the whole domain of interest. This permits identification of a fast subsystem behavior such that correct projection becomes available. Thus, numeric for evaluation of the manifold's equation is significantly simplified because of computationally expensive procedure of decomposition into invariant subspaces is not longer needed. Finally, an estimation of the system small parameter (6) is obtained by the suggested approach.

Reduced and detailed calculations were performed in order to verify the method and compare it to the standard ILDM method. The simulations show that relatively simple flame structures yield a good agreement. Furthermore, the fast part of the detailed system solution trajectory is approximated surprisingly well not only by the local ILDM method, but by the constant GQL decomposition showing the potential of the suggested method.

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