Global Quasi-Linearization (GQL) for model reduction of reaction diffusion systems

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1 Introduction

Numerical integration of reacting flows of 2D or 3D combustion systems with complex geometries using detailed chemistry are possible but typically limited to academic problems [1]. To achieve reliable results detailed mechanisms of chemical kinetics containing a large number of species need to be implemented. A high dimension together with very high stiffness (due to the high non-linearity of rates of chemical reaction) of the corresponding governing equation systems still essentially limits computations of processes taking place at engineering scales. The development of reduction methods for chemical kinetics represents an attractive and perspective way [2, 3] to cope with this problem.

The problem of model reduction has attracted attention and significant progress on a theoretical level has already been made (see a review of methods presented and discussed in e.g. [2, 3]). A number of very interesting and promising approaches have been proposed and implemented (see e.g. [2, 3] for more details).

Methods aiming to produce very low dimensional models and taking into account molecular diffusion and its influence onto system dynamics in the system state space are the flamelet [4], the Flame Prolongation of the ILDM (FPI) approach [5], the Flamelets Generated Manifold (FGM) [6] and the Reaction-Diffusion Manifolds (REDIM) [7]. These methods are very accurate, but depend strongly on a specific configuration and can perform very well within a narrow range of system parameters.

In this study, the Global Quasi-linearisation (GQL) method [8] is extended to treat reaction-diffusion systems. The method follows the main idea of ILDM [9], namely, it is assumed that the system can be decomposed into slow and fast subsystems by applying an eigenvalue decomposition.

The GQL method was already verified for isobaric homogeneous systems (auto-ignition problems) [10], and now, it is applied to premixed flames where physical processes such as convection and diffusion processes perturb the chemical kinetics. In order to simplify the presentation the same reacting system, namely, the hydrogen / air mixture is considered as in the previous study [10], which was devoted to model reduction of homogeneous combustion systems.

The suggested approach is based on a Differential and Algebraic Equations (DAEs) system where the slow manifold of the reduced model is treated in an implicit form. This means the system will be integrated in the whole state space but the system's thermo-chemical states are constrained to an implicitly defined manifold. The results of several premixed flames for varying equivalence ratio and using different mechanisms, but possessing the same GQL basis matrix are provided. It is shown that an optimized 4D reduced GQL manifold based on auto-ignition problem for a single mechanism and for only one set of initial and system parameters performs very well in a wide range of system parameters, initial conditions and different mechanisms.

2   Mathematical model

The partial differential equations system describing a combustion processes can be cast in vector notation in the following form [7]

$$\frac{\partial \Psi}{\partial t} = F(\Psi) - v \operatorname{grad}\Psi + \frac{1}{\rho}\operatorname{div}\left(D \operatorname{grad}\Psi\right), \tag{1}$$

with the state vector $\Psi = (h, p, w_1/M_1, ..., w_{n_s}/M_{n_s})$, the source term F, the density ρ, the velocity v and the diffusion matrix D (a detailed description of some terms is presented below). Additionally, h denotes the enthalpy of the system, p the pressure, while specific mole numbers defined by $\phi_i = w_i/M_i$ in [mol/g], where w_i and M_i are the mass fraction and the molar mass of specie i respectively, are used to describe the mixture composition of a combustion system. The three terms on the RHS of Eq. (1) describe the main physical processes of reaction, advection and diffusion and governing the reacting flow. This form is used to describe the suggested approach and to explain the observations made.

2.1   Identification of the GQL basis

The time scale analysis of the GQL model reduction is based on an adiabatic homogeneous system at a constant pressure with the source term given by

$$F = \left(0, 0, \frac{\dot{\omega}_1}{\rho}, \frac{\dot{\omega}_2}{\rho}, ..., \frac{\dot{\omega}_{n_s}}{\rho}\right)^{\mathrm{T}}. \tag{2}$$

Here $\dot{\omega}_i$ represents the molar rate of formation of a chemical species i due to chemical reaction in mol/(s·m^3). The source term of this system is used for the GQL analysis to set up the system decomposition into slow and fast sub-spaces [10].

The decomposition and a basis for slow and fast subsystems (see e.g. [11]) is provided by the canonical eigensystem decomposition for the $\mathbf{T}_{\mathrm{GQL}}$ linear map ($\mathbf{T}_{\mathrm{GQL}} \cdot \Psi \sim F(\Psi)$) as

$$\mathbf{T}_{\mathrm{GQL}} = \mathbf{V}\Lambda\mathbf{V}^{-1} = \begin{pmatrix} Z_s & Z_f \end{pmatrix} \cdot \begin{pmatrix} N_s & \mathbf{0} \\ \mathbf{0} & N_f \end{pmatrix} \cdot \begin{pmatrix} \tilde{Z}_s \\ \tilde{Z}_f \end{pmatrix}, \tag{3}$$

where on the diagonals of matrices N_s and N_f one finds eigenvalues sorted by magnitude having relatively small and large absolute values.

The spectrum of the $\mathbf{T}_{\mathrm{GQL}}$ eigenvalues is investigated and the reduced dimension and relevant invariant sub-spaces are identified. The relevant invariant eigenspaces can now be employed to separate the slow subsystem. The basis for the invariant eigenspaces can be implemented to integrate the system on the slow manifold given by

$$\tilde{Z}_f \cdot F(\Psi) = 0. \tag{4}$$

For further details see e.g. [10–12].

2.2   Integration on the slow manifold

In order to integrate the slow sub-system on the manifold Eq. (4) an implementation scheme was introduced in [10, 11, 13]. Below we shortly outline the implementation scheme for completeness of the exposition. In order to apply the concept to treat a general combustion reacting flow, the evolution of thermo-kinetic state can be described to be constrained on the manifold Eq. (4) [12]. The decomposition is given by the linear transformation Eq. (3) and the matrix of the basis of sub-spaces that define the slow manifold can now be used in Eq. (1) in a very generic manner. Namely, the dynamics of the system Eq. (1) constrained on the slow manifold is governed by

$$\begin{bmatrix} \tilde{Z}_s \\ \mathbf{0} \end{bmatrix} \cdot \frac{\partial \Psi}{\partial t} = \begin{bmatrix} \tilde{Z}_s \\ \tilde{Z}_f \end{bmatrix} \cdot F(\Psi) + \begin{bmatrix} \tilde{Z}_s \\ \mathbf{0} \end{bmatrix} \cdot v \operatorname{grad} \Psi + \begin{bmatrix} \tilde{Z}_s \\ \mathbf{0} \end{bmatrix} \cdot \frac{1}{\rho} \operatorname{div}(D \operatorname{grad} \Psi). \tag{5}$$

Now, multiplying both sides with the non-degenerate matrix $\mathbf{V} = \begin{pmatrix} Z_s & Z_f \end{pmatrix}$ a vector form of the reduced and fully equivalent system as

$$\mathbf{Q}_s \frac{\partial \Psi}{\partial t} = F(\Psi) - \mathbf{Q}_s \, v \operatorname{grad} \Psi + \mathbf{Q}_s \frac{1}{\rho} \operatorname{div}(D \operatorname{grad} \Psi), \tag{6}$$

where the so-called mass matrix $\mathbf{Q}_s$

$$\mathbf{Q}_s = \begin{pmatrix} Z_s & Z_f \end{pmatrix} \cdot \begin{pmatrix} \tilde{Z}_s \\ \mathbf{0} \end{pmatrix}, \tag{7}$$

can be directly used in the DAEs system numerical integrator packages, where the time- and spatial derivative terms are multiplied by this mass matrix Q_s.

There are advantages and disadvantaged of the suggested implementation scheme. First of all this method can be used to verify and to validate reduced modes given / provided by a manifold equation in the algebraic implicit form as e.g. in Eq. (4). This opens a perspective for comparison and verification of different reduced manifolds without the need to really reduce the system dimension. This, however, represents also the drawback of the proposed approach because one has only an implementation scheme with the original dimension kept.

Although the implementation is straightforward there are several issues of the general reacting flows that complicate the integration and need to be treated to make computations less sensitive to the boundary and initial conditions of the system solution profiles in the case when artificial (igniting solutions) initial profiles are implemented to integrate the system Eq. (6). In order to cope with this problem in the implicit settings one can modify the mass matrix Q_s. This is implemented and accounted for by using modified mass matrices for time derivative, advection and diffusion terms in Eq. (1)

$$Q_s = \begin{pmatrix} Z_s & Z_f \end{pmatrix} \cdot \begin{pmatrix} \mathbf{I}_s & \mathbf{0} \\ \mathbf{0} & \varepsilon \, \mathbf{I}_f \end{pmatrix} \cdot \begin{pmatrix} \tilde{Z}_s \\ \tilde{Z}_f \end{pmatrix}. \tag{8}$$

This modification do not perturb the system solution profile for $t \gg \varepsilon$. The artificial small parameter ε of order of fast time scales and the outcome typically is not affected the the choice. In the current

study $\varepsilon = 10^{-8}$ was chosen, which means that after a short integration time of order of $t > 10^{-6}$ this parameter can be efficiently set to zero. In current computations, however, we have used fixed value of ε throughout the time interval. All numerical computations is implemented using in-house codes HOMREA for homogenoues systems and INSFLA for 1D flames [14] based on the linear extrapolation integrator for stiff differential/algebraic systems LIMEX [15].

3 Results and discussion

3.1 Premixed flame with the Warnatz mechanism

In order to illustrate additionally the suggested approach the detailed and reduced models are compared to the QSSA. The results of the 4D GQL and the QSSA implemented for OH and O species [10], which are the best choice for the considered system, are compared. Because the reaction space dimension of the considered system is $n_r = n_s - n_e = 9 - 3 = 6$, where n_e is the number of elements considered (H, O, N), specifying only two constraints of QSSA species correspond to a 4D manifold in the system reaction space.
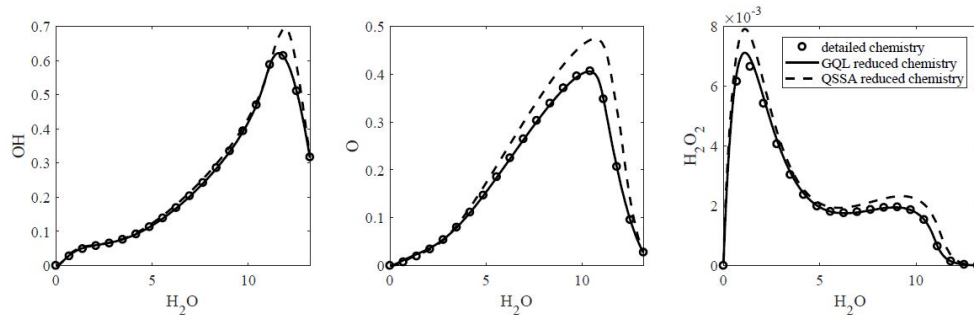

Figure 1: Species mole numbers some radicals as a function of water specific mole number for the steady state solution profiles for $T_u = 298$ K, $p = 1$ bar and $\Phi = 1.0$ for the Warnatz mechanism. Comparison of detailed model (circles), GQL reduced model (line) and QSSA (dashed line) implemented for OH and O species shown by dashed line.

Figure 1 shows the comparison of the species mole numbers of major radicals OH, O, as well as minor H_2O_2 computed along the steady state solution profile (for $T_u = 298$ K, $p = 1$ bar and $\Phi = 1.0$) as function of water specific mole number. The figure justifies that not only major species are captured very well along the steady state profile, but also minor species like H_2O_2. Again the profiles predicted by 4D GQL reduced chemistry performs better then those profiles predicted by 4D QSSA reduced chemistry.

3.2 Application to laminar premixed flame calculations using different reaction mechanisms

After testing and validation of the mass matrix $\mathbf{Q}_s$ found for only one set of system parameters for the Warnatz mechanism the method is applied to other mechanisms considered in [10]. A set of laminar premixed flat flames with an unburnt temperature $T_u = 298$ K and $p = 1, 20, 50$ bar is computed for a wide range of equivalence ratios $0.5 \leq \Phi \leq 4$ using both the detailed model Eq. (1) and the reduced model implementation scheme Eq. (6). The universal applicability of the GQL matrix for four other mechanisms is investigated in this way. The same trend as in [10] can be reported. Figure 2 demonstrates that there are almost no visible differences between the laminar flame velocities predicted by the different detailed mechanisms [10] and reduced models. Remember that the case of ambient pressure of 50 bar with the ideal gas equation might be in question, but it is used here for comparison
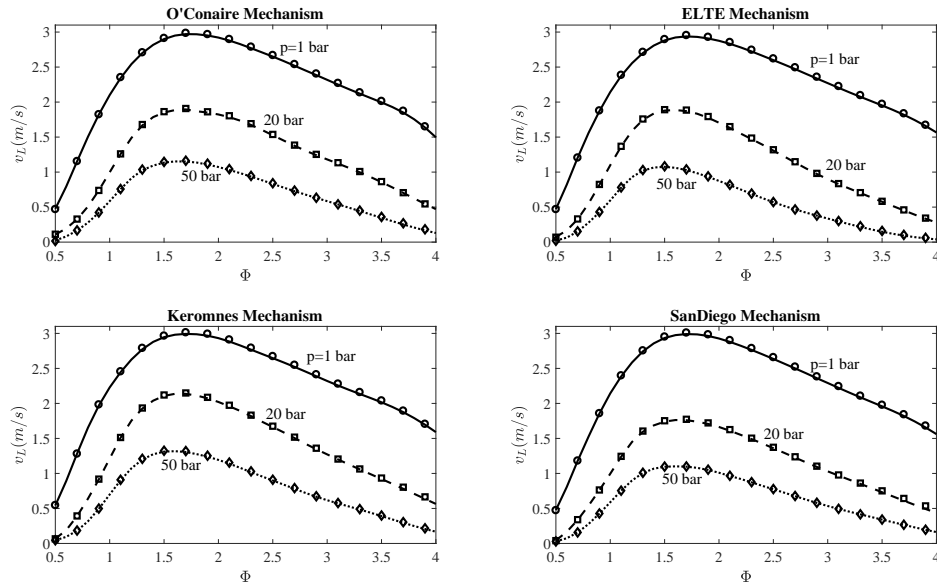
Figure 2: Laminar flame velocity as function of equivalence ratio and different pressures for four mechanisms from [10]. Comparison of detailed model (circles) and GQL reduced model (lines).

between detailed and reduced models and such large variations of the system parameters can still be used for validation purposes. One can see that in the whole considered range of system parameters, the relative error of the flame speed by using the 4D GQL reduced model is very small and remains smaller than the differences between the flames speeds predicted by different kinetic mechanisms. All relative errors remain of the order of 1% for all mechanisms considered.

4   Conclusions

In this study an extension of the GQL reduced model to calculate premixed flat flames was suggested and employed. It was shown how the 4D GQL reduced chemistry for a homogeneous system performs very well in describing premixed hydrogen/air flames. The slow manifold was constructed for an auto-ignition problem and it is based on only one mechanism for a single set of initial conditions and system parameters, namely, for Warnatz mechanism with $\Phi = 1$, $T_0 = 1800$ K and $p_0 = 1$ bar. This GQL reduced chemistry was applied to laminar premixed flames using several well established and validated hydrogen combustion mechanisms and a very wide range of system parameters and initial conditions.

The very good agreement of detailed and reduced model computations demonstrates clearly that

- the GQL method, which is based on an analysis of homogeneous systems, can be extended to allow the description of laminar premixed flames, which are governed by a strong coupling of chemistry with the molecular transport;

- a GQL mass matrix obtained for one specific set of parameters $(T, p,$ mixture composition) can be applied over a large range of different reaction conditions;

- the GQL matrix represents an invariant property of the combustion system such that a GQL basis obtained from an analysis of one mechanism can be used for model reduction of other mechanisms and performs very accurately in a wide range of system parameters.

Acknowledgments

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