Improvement of the Global Quasi-Linearisation (GQL) model reduction method

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1 Introduction

Chemical kinetics of complex oxidation processes in homogeneous systems is described by systems of ordinary differential equations (ODEs). These systems are tend to be very complex in terms of dimensionality (number of species involved), non-linearity and stiffness (due to elementary reaction rates) [1].

The computational demand (CPU time, memory) required to treat such systems can be very high. In order to integrate such systems numerically within reasonable time reduction methods are required. These need to provide us with models of lower dimension and less stiff in comparison to the detailed model [2].

The Global Quasi-Linearisation approach makes use of the time scale hierarchy [3] inherent in the system to reduce the model by using fast and slow invariant manifolds. The method allows to identify existing decomposition of time scales and to split the system into sub-systems of low-dimension, which govern the thermo-chemical processes at different characteristic time scales. By using these sub-systems the dimension of the original detailed model can be reduced [4, 5].

The method identifies automatically the reduced system dimension. It provides with the slow manifold equation. Though it is given in an implicit form it nevertheless can be used to investigate a chemical reaction mechanism [6] and to project the system onto this manifold [5, 7]. However, the method might not be accurate when the time scale separation is not asymptotically small, which is typically the case in applications. The decomposition between time scales of fast and slow subsystems might exist, but it is never infinitely large. Thus, in actual implementations one can face this problem which may lead to accuracy loss of the reduced model solutions.

At present, there is no appropriate solution to cope with this problem. Typically, increasing of the slow manifold dimension is implemented to improve the accuracy and performance of the reduced model in such cases.

In this work, the increase of the order of the approximation of the Global Quasi-Linearisation (GQL) approach is suggested to cope generically with this problem. The resulting manifold equation is improved by considering a first order correction. By using the implicit implementation strategy suggested to the

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first order correction to the manifold equation the reduced model is integrated in the same manner as the zeroth order model reduction.

For simplicity an isobaric closed reacting system of hydrogen / air is considered. The problem of auto-ignition is in the focus of the study. The results illustrate that the accuracy of the reduced model can be significantly improved by using the suggested first order correction to the GQL slow manifold.

2 Mathematical modeling for homogeneous reacting system

The considered system is described by a spatially homogeneous adiabatic system with constant volume condition. It is isolated with no mass and energy transfer with the surrounding. The corresponding mathematical model is represented by

$$\frac{\mathrm{d}\vec{\psi}}{\mathrm{d}t} = \vec{F}\left(\vec{\psi}\right), \tag{1}$$

where $\vec{\psi}$ is the vector of the thermo-kinetic states [8]. For an isochoric closed homogeneous reacting system, $\vec{\psi} = (u, V, \phi_i)^{\mathrm{T}}$ with u the specific internal energy, V the system volume, ϕ_i the specific mole number defined as the ratio of mass fraction w_i to molar mass M_i ($\phi_i = w_i/M_i$). The term $\vec{F}$ stands for the source term, and $\vec{F} = (0, 0, \dot{\omega}_i/\rho)^{\mathrm{T}}$ for the isochoric closed homogeneous reacting system with $\dot{\omega}_i$ as the molar rates of formation of a chemical species due to chemical reactions. Note that the choice of a different thermo-kinetic states (e.g. using mass fraction instead of specific mole number) does not affect the methodology of the GQL method, which will be proposed in the next section.

3 Global Quasi-Linearisation (GQL)

3.1 Methodology of the GQL

The detailed description for the Global Quasi-linearisation (GQL) can be found in e.g. [4].

Two main assumptions underlie the GQL methodology:

- there exists a decomposition of fast and slow processes of the system;
- the decomposition of fast and slow processes is valid everywhere inside our domain of interest.

Based on these two assumptions, the following procedure allows us to find a global linear approximation of the vector field defined by right hand side of (1) that results in the dynamical decomposition. If matrix $\mathbf{T}_{\mathrm{GQL}}$ defines a valid linear approximation of the right hand side of (1)

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

we can say that $\mathbf{T}_{\mathrm{GQL}}$ has to reproduce the asymptotical decomposition, which is present in the original vector field F. Thus, this global linear transformation matrix $\mathbf{T}_{\mathrm{GQL}}$ aims at finding out the fast and slow invariant subspaces by using an algorithm similar to the ILDM method. Namely, the eigenvalue/-vector decomposition allows us obtaining two groups of eigenvalues consisting different slow and fast time scales:

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

3.2 Implementation: Zeroth order of the GQL

Once the decomposition Eq. (3) is defined, then, similar to the ILDM method, the slow manifold is the manifold where the reaction rates in direction of the fast invariant subspaces vanish:

$$\mathcal{M}^{0,s}_{GQL} = \{\vec{\psi} : \widetilde{Z}_f \cdot \vec{F}(\vec{\psi}) = 0\} \tag{4}$$

The original ODE system can be transformed into the following DAE when the GQL slow manifold is implicitly used

$$\mathbf{Q}_s \cdot \frac{\mathrm{d}\vec{\psi}}{\mathrm{d}t} = \vec{F}(\vec{\psi}), \quad \text{where: } \mathbf{Q}_s = \begin{pmatrix} Z_s & Z_f \end{pmatrix} \cdot \begin{pmatrix} \tilde{Z}_s \\ 0 \end{pmatrix}. \tag{5}$$

3.3 Implementation: First order of the GQL

The linear decomposition of the dynamical system obtained though (3) can be used to determine the fast and slow manifolds. The implicit form of the invariant equation can be used to improve the manifold Eq. (4), namely, when $\Phi_0(\vec{\psi}) = \tilde{Z}_f \cdot F(\vec{\psi}) = 0$ represents the zeroth order approximation of the invariant manifold defined for $F(\vec{\psi})$

The first order approximation would then be defined by

$$\Phi_1(\vec{\psi}) = D_\psi(F(\vec{\psi}))\Phi_0(\vec{\psi}) = 0. \tag{6}$$

Substituting $\Phi_0(\vec{\psi}) = \tilde{Z}_f \cdot F(\vec{\psi})$ into the above equation and considering that $\tilde{Z}_f$ is constant, the first order approximation to a leading order magnitude is obtained as

$$\mathcal{M}^{1,s}_{GQL} = \{\vec{\psi} : \tilde{Z}_f \cdot D_\psi(F(\vec{\psi})) \cdot F = 0\}. \tag{7}$$

Then the first-order approximation of slow manifold is simply given by

$$\mathcal{M}^{1,s}_{GQL} = \{\vec{\psi} : \widetilde{Z}_f \cdot \mathbf{J}(\vec{\psi}) \cdot \vec{F}(\vec{\psi}) = 0\}. \tag{8}$$

Here $\mathbf{J}(\vec{\psi}) = D_\psi(F(\vec{\psi}))$ is the Jacobian matrix of the system's source term. Therefore, using the first-order slow manifold based on GQL, the time evolution of species in original coordinate is slightly different from (5) following the DAE system:

$$\mathbf{Q}_s \cdot \frac{\mathrm{d}\vec{\psi}}{\mathrm{d}t} = \begin{pmatrix} \mathbf{I} \\ \mathbf{J}(\vec{\psi}) \end{pmatrix} \cdot \vec{F}(\vec{\psi}), \quad \text{where: } \mathbf{Q}_s = \begin{pmatrix} Z_s & Z_f \end{pmatrix} \cdot \begin{pmatrix} \tilde{Z}_s \\ 0 \end{pmatrix}, \tag{9}$$

where $\mathbf{I}$ is the identity matrix.

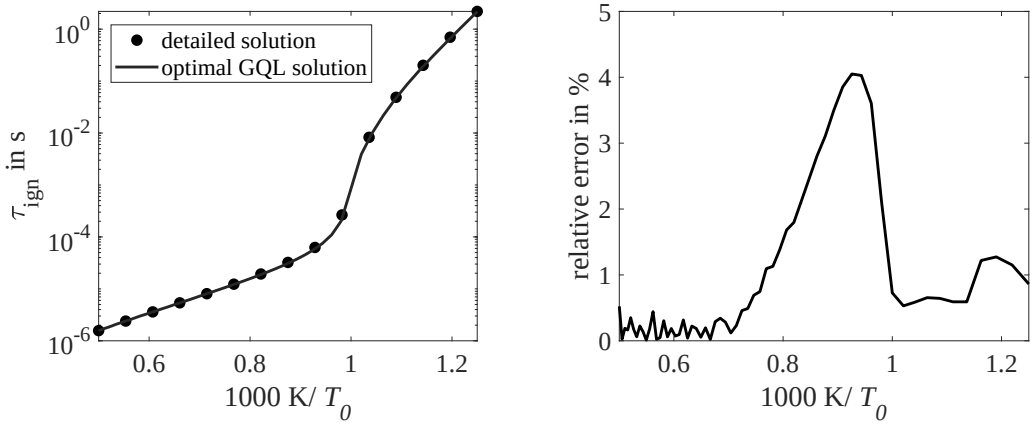
3.4 A general comment on the GQL methodology

Although the GQL has been proven to be more accurate compared to the QSSA method, as it is shown in e.g. [8], we would like to mention here that finding out an optimal GQL basis requires some computational effort, because in the current implementation one uses a statistical approach and tests and validates a number of candidates for linearisation matrix for different system conditions and reduced dimensions. However, if one uses the first order approximation as suggested, the GQL basis does not need to be optimal to reach higher accuracy (see subsections below).

4 Implementation: Hydrogen-Oxygen Isochoric Homogeneous Reaction System

In this section, the hydrogen-oxygen isochoric homogeneous reacting system is considered as a simple but representative example. This system has been intensively investigated in our previous work [8], showing the advantage of GQL over QSSA. There are several further studies [5, 7, 9] where one looks for an optimal GQL reduced chemistry and implement it for different mechanisms and systems.

In the following, we show the results using an optimal GQL reduced chemistry as described above in subsection 4.1, but also the results of the improved manifold by using the first order approximation (see subsection 4.2).



(a) Predicted ignition delay time over temperature

(b) Relative errors in %

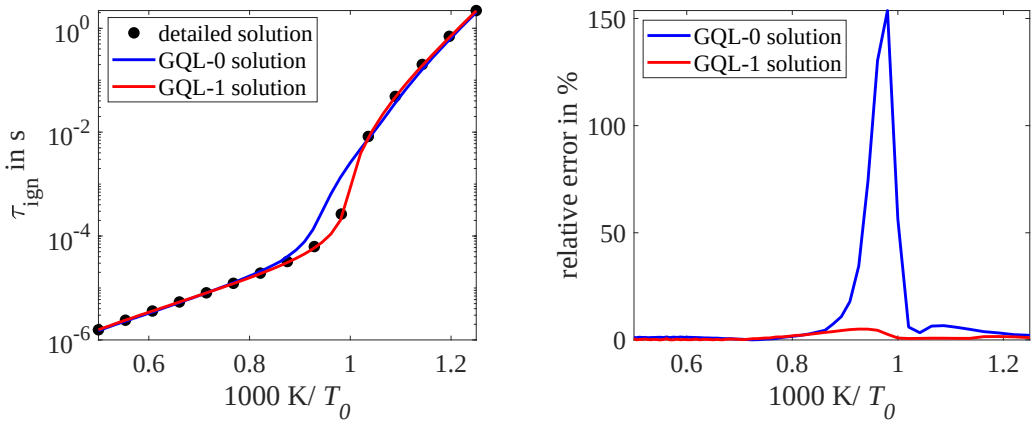
Figure 1: Dependence of the ignition delay times on the initial temperature T_0 at p_0 = 1 bar and Φ = 1.0.

4.1 Results using an optimal GQL with zeroth order approximation

The results using an optimal GQL, which was intensively studied in our previous works, and an optimal 4D GQL reduced model (see e.g. [5]) which describes the auto-ignition process with very high accuracy are shown in Fig. 1. In this figure, an optimal 4D GQL model is selected and used for the prediction of the ignition delay times for different initial temperature ranging from 800 K to 2000 K. It shows that the relative errors of the reduced solution based on the optimal 4D GQL model are less than 5% for all temperatures. It should be mentioned here that during the generation procedure for the 4D GQL reduced chemistry, the 4D-GQL with zeroth order approximation has the maximal errors for temperatures between 900 K and 1100 K. This is due to the competition between the chain branching reaction $O_2 + H = OH + O$ and the chain termination reaction $O_2 + H + M = HO_2 + M$ that the GQL reduced model should be able to take into account. Numerical experiments showed that it costs some computational effort to find out an optimal 4D GQL reduced model which is valid for the whole range of temperatures.

4.2 Results for GQL with zeroth and first order approximation

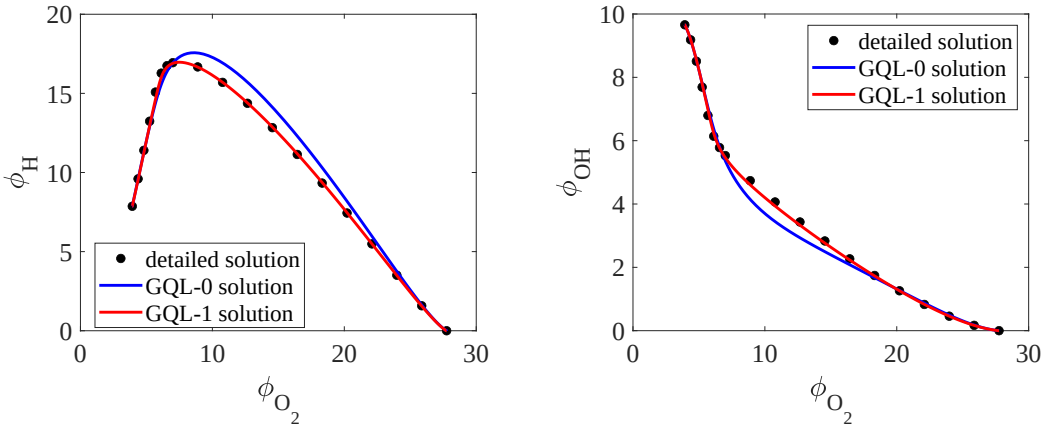
In this subsection, we apply an GQL based reduced chemistry model, which is not optimized, and focus on the comparison of the accuracy between GQL with 0^{th} order approximation and GQL with 1^{st} order approximation. In Fig. 2 we show the predicted ignition delay time over a wide range of temperatures



(a) predicted ignition delay time over temperature

(b) relative errors in %

Figure 2: Dependence of the ignition delay times on the initial temperature T_0 at p_0 = 1 bar and Φ = 1.0.



(a) $\phi_{O_2} - \phi_H$ state space

(b) $\phi_{O_2} - \phi_{OH}$ state space

Figure 3: Different projections of the state space in terms of the specific mole number with unit mol/kg. Initial conditions: $T_0 = 1050$ K, $p_0 = 1$bar, $\Phi = 1.0$.

using the 4D-GQL reduced chemistry based on zeroth order approximation Eq. 5 (blue line) and 1st order approximation Eq. 9 (red line). Note that, the same $\mathbf{Q}_s$ was used for both first and zeroth order cases. The result shows clearly that if the 4D GQL reduced model is not optimal, large errors up to 150% can be observed within the aforementioned initial temperature range between 900 K and 1100 K. However the corresponding 4D-GQL with 1st order approximation significantly improves the accuracy (red lines in Fig. 2), and the maximal error is now reduced to about 10%. The improvement of the accuracy using the first order approximation can also be observed in Fig. 3, where different projections of the system state space are presented. The initial temperature $T_0 = 1050$ K is chosen as a representative example, because the accuracy of the prediction for the ignition delay time by using the 4D GQL with first order approximation is largely improved (c.f. Fig. 2). The comparison of species time histories shows that the state spaces predicted by the 4D GQL with first order approximation better agree with those predicted by the detailed solution.

5 Conclusions

The implementation of a first order approximation of the GQL reduced chemistry was introduced. The problem of efficient implementation of the GQL based slow manifolds was treated by using the implicit scheme of the reduced model numerical integration. This approach can be used generically to verify any decomposition (into fast and slow) based manifolds. Moreover, we have suggested to use it to improve significantly the accuracy and performance of the GQL. The first order correction of the manifold equation was employed. The improved performance was illustrated by a benchmark model of the hydrogen-air ignition problem of combustion. Again of an order of magnitude in the relative error with respect to the ignition delay time was reported. The results show that not only the ignition delay time but also the course of the species state space (in terms of the species specific mole numbers in this work) are improved by using the first order manifold equation. The simplicity of the suggested correction of the manifold equation and no need in demanding improvement and optimization of the original GQL basis opens new perspectives in the implementation of the GQL model reduction approach.

Acknowledgments

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