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Chunkan Yu, Viatcheslav Bykov

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GQL-RedChem: A MatLAB-based tool for the model reduction for chemical kinetics based on the Global Quasi-linearisation (GQL) approach

Chunkan Yu[1], ***Viatcheslav Bykov***[2]
Institute of Technical Thermodynamics (ITT)
Karlsruhe Institute of Technology (KIT)
Engelbert-Arnold-Str. 4, 76131, Karlsruhe, Baden Württemberg, Germany.

Abstract

The Global Quasi-linearisation (GQL) approach has been developed for the dimension reduction of the chemical kinetics, which aim at speeding up the numerical simulation of reacting flows. **GQL-RedChem** is a MatLAB-based package that integrates the homogeneous reacting systems, formulated mathematically as a system of Ordinary Differential Equations (ODEs). The package provides with an approximation of the fast/slow decomposition linear basis, describing a reacting source term by a system of Differential Algebraic Equations (DAEs). The GQL method can be applied for reacting systems with any complexity, and the GQL decomposition basis can be generated in a generic and automatic manner.

Keywords
chemical kinetics; ignition problem; model reduction; time scale analysis; invariant sub-spaces; slow invariant manifolds.

[1]chunkan.yu@kit.edu
[2]viatcheslav.bykov@kit.edu

Code metadata

Nr.	Code metadata description	Please fill in this column
C1	Current code version	*v 1.0*
C2	Permanent link to code/repository used for this code version	*https://github.com/ChunkanYu/GQL-RedChem*
C3	Permanent link to Reproducible Capsule	
C4	Legal Code License	MIT License
C5	Code versioning system used	none
C6	Software code languages, tools, and services used	MatLab, Cantera
C7	Compilation requirements, operating environments & dependencies	validated for MatLab 2020 - 2023a, Cantera 2.3.0 - 2.6.0
C8	If available Link to developer documentation/manual	*https://github.com/ChunkanYu/GQL-RedChem*
C9	Support email for questions	chunkan.yu@kit.edu; viatcheslav.bykov@kit.edu

1. Introduction

The simulation of reacting flow systems using detailed chemistry are typically high dimensional with respect to a number of species involved and highly stiff due to the non-linearity of the chemical reactions [1, 2]. Although the reacting systems of zero- & one-dimensional geometry by using the detailed chemistry is possible, the simulation of three-dimensional reacting flows with complex geometry by using the detailed chemistry leads to high computational cost, which even nowadays still represent a challenge. The model reduction for chemical kinetics provides a possibility to speed up the numerical calculation by reducing both the dimensionality and the stiffness of the governing equations.

The Global Quasi-Linearisation approach [3] makes use of the time scale hierarchy [4] inherent in the system to reduce the model by decomposing the original dynamic system into the fast and slow sub-systems. The method allows to find out existing decomposition of fast and slow time-scales, which can be used to identify the fast and slow manifolds, 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 [3, 5]. Compared to the standard Quasi-steady State Approximation (QSSA) method, the GQL approach follows a generic and automatic procedure, and no a priory information and empiric understanding of the chemical kinetic is required.

Furthermore, it has been shown that the GQL based reduced description on the slow manifold provides more accurate results compared to the QSSA reduced approach [6, 5] and has much lower dimension with high accuracy compared to the skeletal mechanism generation method [7]. From this perspective the GQL can be understood as an extension of the QSSA, where not species but their linear combinations are considered as an approximation of the fast motions and, consequently, this becomes more general and flexible in description of fast system dynamics and relaxations towards the slow manifold (see Fig. 1). The latter might be extremely important both for initial state projection [8] that captures the fast transient process and for improvement of a slow manifold equation.

2. Purpose and Functionality of the software

2.1 Purpose of the software

The **GQL-RedChem** package is a MatLab based code which generates the basis of the decomposition automatically. This software provides with the fast and slow invariant sub-spaces of the chemical systems. Moreover, the software also includes the numerical simulation based on the standard QSSA method, which can be used to compare with the results obtained from the GQL approach.

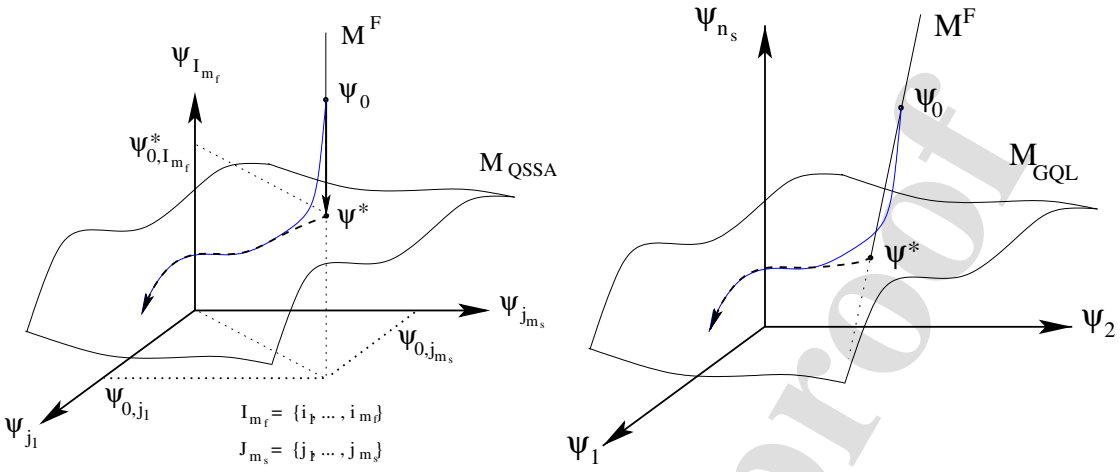

Figure 1: Schematic system decomposition structure in the state space. Left: slow/fast manifolds in the case of QSSA. Right: slow/fast manifolds in the case of the GQL, where the fast motion is not longer restricted to the original variables and can be represented via a linear combination - $M^F = \{\vec{\Psi} : \tilde{Z}_s(\vec{\Psi} - \vec{\Psi}_0) = 0\}$.

2.2 Functionality of the software

The **GQL-RedChem** integrates the systems of ordinary differential equation (ODE) describing the isochoric and isobaric homogeneous reacting systems. This package includes two main functions:

- main_0dSimulation.m: this main function handles the numerical simulation based on detailed chemistry, GQL and QSSA based reduced chemistry in form of (DAEs) system, where fast processes are relaxed and system solution is constrained on the slow manifold. For all three variants, the time evolution of the thermo-kinetic state vector $\vec{\Psi}$ reads:

$$\mathbf{M}_s \frac{\partial \vec{\Psi}}{\partial t} = \vec{F}(\vec{\Psi}) \tag{1}$$

  with the so-called mass matrix $\mathbf{M}_s$. For the detailed chemistry, the $\mathbf{M}_s$ is equal to the unit matrix. For the QSSA reduced chemistry, the diagonal elements are equal to 1 for non-QSS species and to 0 for QSS species. The QSS species must be user defined and specified in the input.

- main_GQL_RedChem.m: this main function aims at generating the GQL decomposition basis based on user defined dimension of fast invariant subspace N_f and the accepted relative errors ϵ_{tot} of GQL reduced chemistry model Eq. (1). For the GQL reduced chemistry, the mass matrix is calculated as

$$\mathbf{M}_s = \left(Z_s \; Z_f\right) \cdot \begin{pmatrix} \tilde{Z}_s \\ \mathbf{0} \end{pmatrix}, \tag{2}$$

  which is obtained during the generation of the GQL reduced chemistry in find_GQL_candidate.m.

Figure 2 summarizes the most important steps in the main_GQL_RedChem.m generating the possible GQL reduced chemistry. The block 1 involves all the required inputs to initiate the algorithm, and the block 2 focusing on the generation of a GQL reduced chemistry is fully automatic. Note that in the present version, the ignition delay time (IDT) is considered as the most important target quantity to check the accuracy of the GQL reduced chemistry and user specified tolerance. However, any other target function such as e.g., the maximum of the species concentrations, can also be considered, which can be implemented in the code.

The source terms of chemical reaction are evaluated through the Cantera [9]. The DAE system Eq.1 is integrated by using the ode15s, which can solve stiff differential equations and DAEs based on variable order method [10].

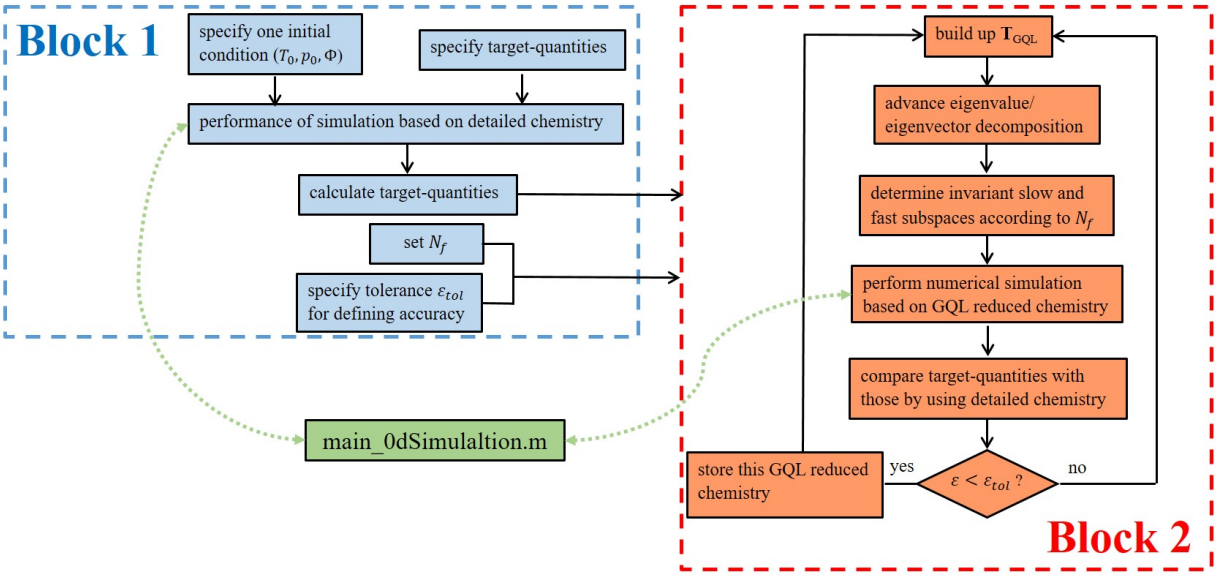

Figure 2: Flow chart of the main_GQL_RedChem.m code.

2.3 How to use

The GQL_RedChem software is highly automatic and generic. The main_0dSimulation.m requires only the initial conditions such as the initial temperature, initial pressure and fuel/air equivalence ratio. The chemical mechanism must be provided in Cantera format (.cti). The option *chemistry* defines whether and which detailed or reduced chemistry should be used.

- If *chemistry='detailed_chemistry'*, the detailed chemistry simulation is chosen, and nothing else is required.
- If *chemistry='QSSA_chemistry'*, the QSSA reduced chemistry simulation is chosen, and the QSS species are required to defined in variable *QSS_species*. Example could be *QSS_species*={*'OH','O'*} defining the OH and O species as QSS species. Note that the order of the QSS species is not important.
- If *chemistry='GQL_chemistry'*, the GQL reduced chemistry simulation is chosen, and the mass matrix defining the GQL reduced chemistry will be read from the file *GQL_Ms.mat*, which will be obtained from the main_GQL_RedChem.m.

The main_GQL_RedChem.m stores the GQL reduced chemistry in *GQL_Ms.mat*. Besides the initial conditions such as the initial temperature, initial pressure and fuel/air equivalent ratios, several inputs for the GQL generation algorithm are required: *n_GQL_attempt* defines the maximum number of attempt to find out the GQL reduced chemistry; *n_GQL_max* defines the maximum number of found/stored GQL reduced chemistry for a certain reduced dimension; *error_GQL* defines the accuracy of the found/stored GQL reduced chemistry in percentage; and *Nf* defines the dimension of fast invariant subspace $\tilde{Z}_f$.

In *GQL_Ms.mat* different possible GQL reduced models (as corresponding mass matrices) are stored, and they can be used further in main_0dSimulation.m for validation of wider application range.

3. Impact overview

3.1 Research impact

The **GQL-RedChem** has been developed at the Institute of Technical Thermodynamics (ITT) in Karlsruhe Institute of Technology (KIT) since 2015 during the first author's master dissertation, under the supervision of

the second author. After then, the code has been continuously developed and applied to test combustible gas mixtures with different level of complexity, from hydrogen-air system (9 species) [6, 5] to methane-air system (50 species) [7]. Based on this package, the fast and slow invariant sub-spaces can be identified automatically, which can be used for the following practical applications to address variety of scientific and engineering questions

- Ignition problem in homogeneous reacting systems. The generated GQL reduced chemistry can be used for both isochoric and isobaric homogeneous reacting sytems (both can be simulated in main_0dSimulation via option *reactor_system*. Moreover, it has already been shown that the GQL reduced basis is invariant with respect to the chemical kinetics [5]. In other words, the GQL reduced chemistry generated by considering one chemical mechanism can also be applied for other chemical mechanisms;
- Premixed combustion systems including some critical phenomena such as diffusive-thermal instabilities, e.g., in [11] it was demonstrated that the mass matrix defined and optimized for a homogeneous system can also be used to capture the onset of pulsations and describe properties of oscillating solutions of a distributed one (i.e. system of PDFs);
- Turbulent simulations based on tabulated chemistry. The use of tabulated chemistry requires the definition of the progress variables, which enable to describe the fast relaxation process. According to the study in [8], the progress variables can be determined as $\theta = \tilde{Z}_s\vec{\Psi}$, where $\tilde{Z}_s$ can be obtained from the GQL approach;
- In general, in situations when initial states are not on the relevant slow manifold, parameterisation along the identified fast subspace might solve the problem with projecting initial data automatically.
- It also can be used to verify any reduced model based on the slow manifold, which can be represented by an implicit function in the form of DAEs, i.e. Eq. (1).

This GQL_RedChem package has also been used in several other research groups such as Prof. Felipe Minuzzi (Universidade Federal de Santa Maria) who combines the GQL approach with his own DRG code (generation of the skeletal mechanism), Prof. Liming Dai (Jiangsu University) who aims at using the GQL_RedChem package for the ammonia/DME system and identifying important species and Prof. Vladimir Goldshtein (Ben-Gurion University of the Negev) who compares the performance of the GQL and QSSA reduced chemistries using the GQL_RedChem package [5].

3.2 Educational Impact

The current package can be used for training and study purposes. The method can be implemented to treat academic benchmark models, to check assumptions made, prove and confirm certain asymptotic limits. It can be useful to verify and to compare performance of the standard (e.g., QSSA or PEA) methods to reduce and to approximate invariant slow manifolds introduced in these benchmark, toy models.

4. Future improvements

Though provided routines can already be used for investigations in the present form, future developments can significantly increase functionality and applicability range and might include

- detection of conserved or quasi-conserved sub-spaces and implicit decoupling of these sub-spaces;
- automatic detection of a decomposition dimension;
- implementation of a first order correction of the slow invariant manifold during the integration procedure;
- automatic identification of the extrapolation power of the basis for a wider range of system parameters (i.e. for pressures, temperatures and equivalence ratios).

5. List of all scholarly publications enabled by the software

(i) C. Yu, V. Bykov, U. Maas, Global quasi-linearization (gql) versus qssa for a hydrogen–air auto-ignition problem, *Physical Chemistry Chemical Physics* 20 (16) (2018) 10770–10779.

(ii) V. Bykov, C. Yu, V. Gol'dshtein, U. Maas, Model reduction and mechanism comparison of hydrogen/oxygen auto-ignition, *Proceedings of the Combustion Institute* 37 (1) (2019) 781 – 787.

(iii) C. Yu, V. Bykov, U. Maas, Coupling of simplified chemistry with mixing processes in pdf simulations of turbulent flames, *Proceedings of the Combustion Institute* 37 (2) (2019) 2183–2190.

(iv) C. Yu, F. Minuzzi, V. Bykov, U. Maas, Methane/air auto-ignition based on global quasi-linearization (GQL) and directed relation graph (DRG): Implementation and comparison, *Combustion Science and Technology* 192 (9) (2020) 1802–1824.

(v) V. Bykov, S. Shashidharan, E. Berszany, V. Gubernov, U. Maas, Model reduction of rich premixed hydrogen/air oscillatory flames by global quasi-inearization (GQL), *Combustion Science and Technology* 194 (12) (2022) 2377–2394.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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[8] C. Yu, V. Bykov, U. Maas, Coupling of simplified chemistry with mixing processes in pdf simulations of turbulent flames, Proceedings of the Combustion Institute 37 (2) (2019) 2183–2190.

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Highlights of the paper

- A MatLab-based code for the automatic generation of reduced chemistry based on Global Quasi-linearization (GQL) methodology
- Comparison of the GQL reduced chemistry with other standard reduced chemistry (e.g. QSSA)
- The reduction methodology is useful for any combustion system
- The generated GQL reduced chemistry can also be applied for more complex reacting flows such as laminar and turbulent flames

Declaration of interests

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☐The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: