European Symposium on Computer Aided Process Engineering – 15
L. Puigjaner and A. Espuña (Editors)

An Adaptive Reduction Scheme to Develop Flexible Reduced Chemistry Models for Reactive Flow Simulations

Ipsita Banerjee and Marianthi G. Ierapetritou*,
Department of Chemical and Biochemical Engineering
Rutgers University, Piscataway, NJ 08854

Abstract

Detailed simulation of reactive flow systems using complex kinetic mechanisms consisting of hundreds of species is a computationally demanding task. In practice, to alleviate the computational complexity, the reactive flow models use a skeletal kinetic model instead of the detailed mechanism. However, the reduced chemistry models can accurately predict the detailed model only over limited range of conditions. Since the reactive flow simulation encounters a broad range of conditions, using a single reduced model throughout the simulation incorporates inaccuracy in the predictive capacity of the flow model. In this paper, an adaptive scheme is presented, which aims at developing different reduced models to address the changing conditions of the flow simulation, thereby maintaining high accuracy throughout the simulation with relatively simple chemistry models. A new approach is developed for the analysis of the range of validity of the reduced model, which is highly non-convex, for which the conventional techniques do not perform well. Finally, a procedure to implement the adaptive chemistry in different flow simulations is presented.

Keywords: Kinetic Model reduction, Feasibility Analysis, Mixed Integer Optimization.

1. Introduction

Over the past couple of decades kinetic mechanisms have been developed to model combustion processes. Although kinetic simulations comprising hundred of species and thousand of reactions is not a constraint in homogenous systems, detailed simulation of complex reacting flow systems having strong coupling of heat and mass transfer is still computationally prohibitive. Such simulation of reacting flow involves the integration of a system of partial differential equations comprising of equations of mass, momentum and energy, equation of state along with kinetic source terms. The stiffness of the embedded kinetics often makes the integration of chemical source term the most expensive step. In practice, to alleviate the computational complexity, the reacting flow models use a skeletal kinetic model instead of the detailed mechanism. However, the accuracy compromised by this skeletal mechanism or the range of its validity is not quantified. Although the simplified kinetic model may represent the chemical activity tolerably well in a limited region of the flow field, it is rarely accurate over the entire

---

* Author to whom correspondence should be addressed: marianth@soemail.rutgers.edu

temperature/composition space of interest. By applying the reduced model in regions where it is not valid will significantly lower the predictive value of the detailed reactive flow model. The adaptive chemistry approach proposed here addresses this issue by replacing the detailed reaction mechanism which is valid over the entire flow field, by a set of reduced mechanisms, each valid over a limited region of the flow field. A very important step in this approach is the accurate representation of the range of conditions in the species composition/ temperature space over which a particular reduced mechanism is valid. As the flow simulation proceeds, an appropriate reduced mechanism is selected and integrated depending on the local conditions of temperature and species concentrations.

2. Mathematical formulation of adaptive reduction

The reduction technique adopted in this work follows the mathematical programming approach proposed by Androulakis (2000). This approach is based on determining the reactions of the detailed mechanism, which can be excluded while still retaining a desired accuracy in the prediction of the profiles of certain important species. The reduction process begins with the choice of a reactor model and a discrepancy function, which is a measure of the error incurred in excluding reactions from the mechanism. The optimization problem for an isobaric batch reactor can be formulated as:

$$\min_{\lambda \in \Lambda^{N_R}} \sum_{r=1}^{N_R} \lambda_r \quad \text{subject to: } \chi \le \delta \tag{1}$$

$$\chi = \sqrt{\sum_{k \in K} \int_{t_I}^{t_F} \left(\frac{y_k^{reduced}(t) - y_k^{full}(t)}{y_k^{full}(t)} \right)^2 dt + \int_{t_I}^{t_F} \left(\frac{T^{reduced}(t) - T^{full}(t)}{T^{full}(t)} \right)^2 dt} \tag{2}$$

$$\frac{dy_k(t)}{dt} = \frac{R_k M_k}{\rho} \tag{3}$$

$$\frac{dT(t)}{dt} = -\sum_k \frac{R_k M_k h_k}{\rho \bar{C}_p}, \; R_k = \sum_{i=1}^{N_r} \lambda_i \left(v^r_{k_i} - v^f_{k_i} \right) q_i \tag{4}$$

$$q_i = k_{f_i} \prod_{k=1}^{N_s} X_k^{v^f_{k_i}} - k_{r_i} \prod_{k=1}^{N_s} X_k^{v^r_{k_i}}, \; k_{f_i} = K_{f_i} \exp\left(-\frac{E_{f_i}}{RT}\right), k_{r_i} = K_{r_i} \exp\left(-\frac{E_{r_i}}{RT}\right) \tag{5}$$

where, λ_r is a binary variable used to denote the presence ($\lambda_r = 1$) or absence ($\lambda_r = 0$) of a particular reaction. Hence, the objective function represents the total number of reactions in the reduced set, which has to be minimized, subject to a specified accuracy δ determined by the integral error measure χ, given by Equation (2). χ defines the approximation error of the trajectories of the key observable quantities for the interval of interest. In the above formulation, y_k denotes the mass fraction whereas X_k represents the molar concentration of specie (k) used in the calculation of the reaction rates. Equations (3) and (4) represent the material and energy balances for the reactor model, where R_k is the net rate of production of specie (k); M_k is the molecular weight of specie k; ρ denotes the mixture density, which is a function of composition and temperature. For combustion systems, the basic form of q_i is expressed by the power law expression of mass-action kinetics, and the temperature dependence of the specific reaction rate constant described by Arrhenius law (Equations (5)). Formulation (1)-(5) of mechanism reduction corresponds to an integer nonlinear programming problem, which has been solved using Genetic Algorithm (GA). GA (Goldberg, 1989, Michalewicz and Schoenauer, 1996) represents a class of search and optimisation procedures that are patterned after the biological process of natural selection and they lend themselves to solution of a wide range of optimisation problems. When GA is applied to optimization problem, each optimization variable is typically encoded as a string of bits, and these strings are appended together to form a chromosome. Each individual in a population has a particular chromosome value that can be decoded to evaluate the parameter values and objective function, also called the fitness function. Populations are evolved through several generations until the objective function cannot be improved any further. In the mathematical model (1)-(5) the optimization variables are the N_r binary variables associated with the reactions. Hence parameterisation for GA is straightforward since each binary variable λ_r becomes a bit in the GA chromosome. For a particular combination of λ_r the reduced differential equation sets are integrated to evaluate the discrepancy function.

2.1 Range of validity of reduced mechanism

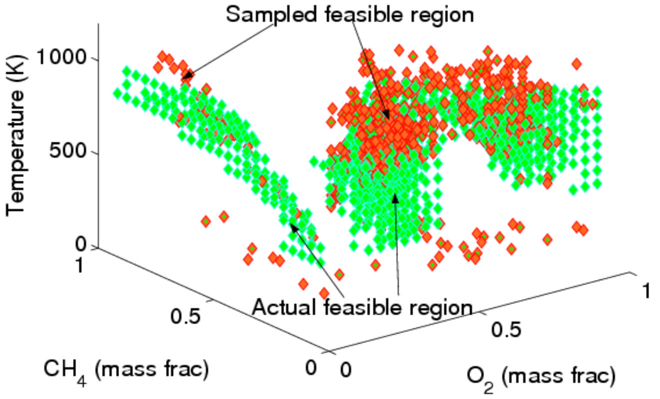

Figure 1. Feasible region of a reduced model of GRI-3.0 mechanism involving 17 species and 59 reactions together with the sampled feasible region

As mentioned in the introduction, the reduced kinetic mechanism obtained by the procedure discussed in the previous section is valid over a limited range of temperature and species concentration and will incorporate significant inaccuracy if used outside this range. Hence accurate estimation of the feasible region is of paramount importance. An examination of the actual valid range of the reduced set, obtained by performing grid search, showed that the feasible region is highly non-convex, non-smooth, sometimes even disjoint (Figure 1). However, since the evaluation of the error function is an expensive operation, performing grid search for every reduced set is not feasible. Hence a sampling scheme is used to obtain an approximate representation of the feasible region. To have a description of the feasible region approximated by the sampled points, a shape reconstruction methodology is used, where an α shape is constructed using the sample data points.

The α shape of a finite point set S is a polytope that is uniquely determined by S and a parameter α. It expresses the intuitive notion of the shape of S, and controls the level of detail reflected by the polytope. The original paper on α shapes (Edelsbrunner et al. 1983) defines the concept in the plane. An extension to three dimension together with an implementation is reported in (Edelsbrunner and Mücke, 1994)

The value of α controls the level of details of the reconstructed shape. For $\alpha \to 0$, the α shape of S degenerates to the point set S, and $\alpha \to \infty$, reproduces the convex hull of the point set. The output from α shape gives the points from S, which forms triangles defining the outer surface of the data set S. Figure (1) illustrates the feasible region of a reduced mechanism as obtained by grid search together with the representative data set obtained from a search technique. This data set is given as an input to the α shape program, the output from which is the information of the data points forming vertices of the triangles defining the surface of the data set.

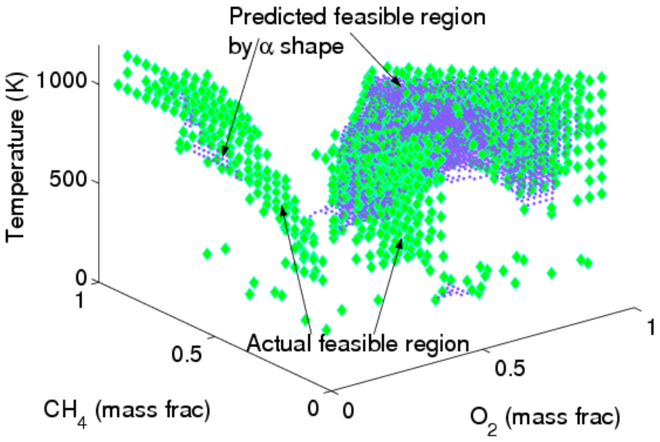

Figure 2. Predicted feasible region of a reduced model

Having defined the surface or shape of the feasible region, the next step involves determination of whether a particular point is inside the feasible region or outside it. This can be done by using one of the point-in-polygon tests (Haines 1997) used to determine whether a point is inside a polygon or outside it. Using this procedure, the predicted feasible region is constructed (Figure 2) using the point-in-polygon test for each point.

2.2 Adaptive reduction technique

To successfully integrate the adaptive reduction with a detailed flow field one needs a library of reduced models which covers the entire range of conditions accessed by the flow simulation. However, this requires a-priori knowledge of the accessed region of the flow simulation, which is not possible. An alternative method is to have an estimate of the accessed region by solving the detailed flow simulation with a skeletal kinetic mechanism or a simplified flow simulation with detailed kinetic mechanism. In the present work, an estimate of accessed region is obtained from homogenous flow simulation of a pairwise mixed stirred reactor (PMSR) (Pope 1997), which is known to provide a stringent test for combustion chemistry.

To generate reduced mechanisms to cover the entire accessed space, one needs to identify nominal points where reduction needs to be performed. This can be accomplished by clustering the data of temperature and species concentration to identify patterns in the data set and representative points for each cluster. Having obtained such representative points, mechanism reduction is performed at each of the cluster centers, to obtain reduced reaction sets, which can describe the accessed region. The next step is the determination of the range of validity of the reduced model within which it retains the desired accuracy. This requires an estimate of the feasible region, which can be obtained by sampling the temperature-composition space.

In order to obtain a mathematical definition of this feasible space, an α shape is constructed, which identifies the boundaries of the feasible space. The accessed data points of PMSR simulation are then checked to identify points which lie outside the generated feasible region using the point-in-polygon strategy. This uncovered set is reclustered to obtain further nominal points for reduction. This procedure is followed iteratively, until enough reduced mechanisms are generated to cover the entire accessed region. The reclustering procedure is necessary since the feasible points for a particular reduced mechanism can cover an entirely different region than defined by the initial cluster for which the nominal point was the cluster center.

For the present simulation for methane combustion using GRI-3.0 (Smith et al. web page), a total of 18 reduced sets were generated covering 76 % of the region accessed by the PMSR model. All the reduced models were obtained by running Genetic Algorithm with a population size of 10, evolved through 500 generations, with an allowable error of 0.01. Once sufficient number of reduced reaction models have been developed to cover the entire estimated accessed region, the next step is the incorporation of the reduced sets in the reactive flow models.

As mentioned before, the PMSR simulation is a time marching scheme, where the incremental time step is split into the mixing time step and the reaction time step. For such a fractional time stepping approach, the flow time stepping will remain unaltered. This time step will result in a set of species concentrations and temperature, which are passed on to the reaction time step, at which the system will react under the conditions obtained from flow time step. This reaction step requires the integration of a set of coupled stiff ODE's, which needs to be integrated over the reaction time step. While using the detailed model for kinetic source term, all the N_s ODE's consisting of N_r reaction terms need to be integrated. The objective of the reduced model is to reduce the number of ODE's and their coupling by eliminating species and reactions participating in the source term. While using a single reduced reaction model over the entire flow simulation, the same set of reduced ODE's needs to be integrated at the reaction step. In the adaptive chemistry scheme, however, there is a library of mechanisms from which the most appropriate one needs to be selected and integrated. Hence at every time it is likely that a different set of reduced ODE's are integrated. As discussed before, the accuracy of the reduced model depends solely on the initial condition of the species concentration and temperature at which it is used. Also each reduced set is characterized by an α shape defining the range of conditions over which it is accurate. Hence to select an appropriate reduced mechanism during the PMSR simulation, one needs to check the feasible region of all the mechanisms by performing the point-in-polygon test, to determine one, which is feasible at the local condition. This procedure was followed for the PMSR simulation of CH_4 combustion, when the 18 reduced models were found to cover 76% of the conditions encountered by the flow simulation. Similar simulation was also performed using a single reduced set for the entire flow simulation. Figure (3) illustrates the performance of adaptive reduction in the prediction of the species and temperature profiles as compared to using a single reduced model. The adaptive scheme predicted the temperature profile with an error of 0.75 %, whereas the single reduced model had an error of 11%. The prediction of CH_4 profile was comparable for both the cases. The adaptive scheme result in an error value of 0.1387, and the single reduced model introduced an error of 0.1125. The prediction of both the H and H_2 profiles was more accurate for adaptive scheme, having an error of 9% and 7% respectively, while the corresponding values for single reduced model was 32% and 21%. The average size of the reduced model used for the adaptive scheme is approximately 54, while that of the single model was 59. This shows that by adapting the reduced model to the changing condition of the flow simulation, greater accuracy can be obtained with the same dimension of the reduced model.

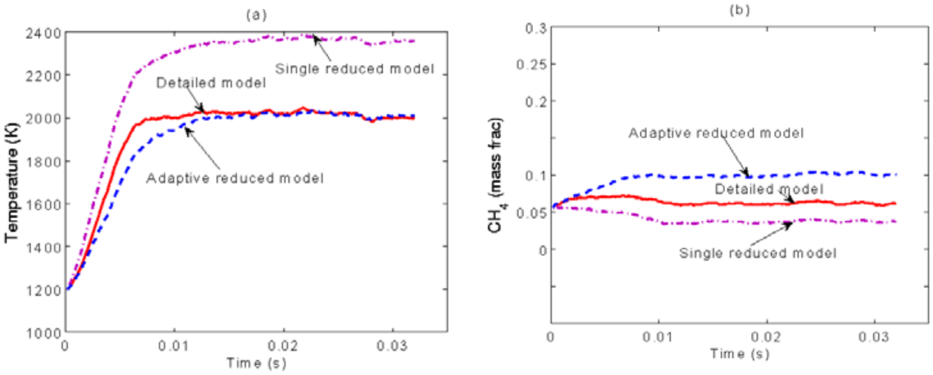

Figure 3. Performance of adaptive reduction in (a) temperature and (b) methane profile

References

Androulakis, I.P., 2000, AIChE J. 46, 361.

Bard,Y., 1974, Nonlinear parameter estimation, Academic Press, New York.

Vajda,S.,Valko,P.,Turanyi,T., 1985, Int. J. Chem. Kinet., 17, 55.

Edelsbrunner, H., Kirkpatrick, D.G., Seidel, R., 1983, IEEE Transactions on Information Theory, IT-29 (4), 551.

Edelsbrunner, H., Mücke, E.P. 1994, ACM Transactions on Graphics, 13(1), 43.

Goldberg, D. E. 1989, Genetic algorithms in search, optimization and machine Learning, Addison-Wesley, New York.

Haines, E., 1994, ed. Paul Heckbert, Academic Press, 24.

Michalewicz, Z. and Schoenauer, M, 1996, Evol. Comput., 4, 1.

Smith,G.P.and Golden,D.M. and Frenklach,M. and Moriarty,N.W. and Eiteneer,B. and Goldenberg,M. and Bowman,C.T. and Hanson,R.K. and Song,S. and Gardiner,W.C. and Lissianski,V.V. and Qin,Z., http://www.me.berkeley.edu/gri_mech/

Pope, S. B., 1997, Combustion Theory and Modeling, 1,41.

Acknowledgements

The authors gratefully acknowledge financial support from the Office of Naval Research under the contract N00014-03-1-0207 and National Science Foundation (CTS 0224745).