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

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Abstract

In numerical simulation of combustion models, solution of the chemical kinetics is often the most expensive part of the calculation, since accurate description of kinetic mechanism involves large number of species and reactions, leading to a large set of coupled ODE's, often too complex to be considered in their entirity along with a detailed flow simulation. Hence the need for representing the complex chemical reactions by simple reduced models, which can retain considerable accuracy while rendering computational feasibility. In this paper a framework is presented for development of adaptive reduced mechanisms that describe different regions of the flow-field with high accuracy. A new approach is presented for the feasibility analysis of the reduced mechanism, along with an outline for selection of an apporpriate mechanism by the flow simulation.

Keywords

Feasibility analysis, Mechanism reduction, α shapes

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 are 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 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 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.

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 mecha-

nism which can be excluded while still retaining a desired accuracy in the prediction of important combustion characteristics. 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 :

$$\min_{\lambda \in \Lambda^{N_R}} \sum_{r=1}^{N_R} \lambda_r$$

$$subject\ to\ \chi \leq \delta \quad (1)$$

$$\chi = \Big(\sum_{k \in \kappa} \int_{t_I}^{t_F} \Big(\frac{y_k^{reduced}(t) - y_k^{full}(t)}{y_k^{full}(t)} \Big)^2 dt + \int_{t_I}^{t_F} \Big(\frac{T_k^{reduced}(t) - T_k^{full}(t)}{T_k^{full}(t)} \Big)^2 dt \Big)^{1/2} \quad (2)$$

$$\frac{dy_k(t)}{dt} = \frac{R_k M_k}{\rho} \qquad k = 1, ..., N_s \quad (3)$$

$$\frac{dT}{dt} = -\sum_{k=1}^{N_s} \frac{R_k M_k h_k}{\rho \overline{C}_p} \quad (4)$$

$$R_k = \sum_{i=1}^{N_r} \lambda_i \Big(\nu_{ki}^r - \nu_{ki}^f \Big) q_i \quad (5)$$

$$q_i = k_{f_i} \prod_{k=1}^{N_s} X_k^{\nu_{k_i}^f} - k_{r_i} \prod_{k=1}^{N_s} X_k^{\nu_{k_i}^r} \quad (6)$$

$$k_{f_i} = K_{f_i} e^{-E_{f_i}/RT}, k_{r_i} = K_{r_i} e^{-E_{r_i}/RT} \quad (7)$$

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 $\sum_{r=1}^{N_R} \lambda_r$ 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. For the present study the observables are taken to be fuel, oxygen and system temperature. The decision of the trajectories to be observed depends on the simulation in hand, for example, for an environmental simulation the trajectories of the pollutants will be the key observables. 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. R_k is evaluated from the knowledge of intrinsic rates q_i of individual reactions and stoichiometry, as given by Equation (5). For combustion systems, the basic form of q_i is expressed by the power law expression of mass-action kinetics, as given by Equation (6). The temperature dependence of the specific reaction rate constant is given by Arrhenius law (Equation (7)).

Range of validity of reduced mechanism

As mentioned in the introduction, the reduced kinetic mechanism obtained by the procedure discussed in the previous section, or any other reduction method, 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 as shown in Figure (1). The detailed mechanism used in this simulation is the GRI-3.0 methane combustion mechanism consisting of 53 species and 325 reactions, which was reduced to 17 species 59 reaction mechanism. 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, as illustrated in Figure (2). To have a mathematical description of the feasible region approximated by these sampled points, a shape reconstruction methodology is used, where an α shape is constructed using the sample points. The output from α shape gives the points from the sampled set S that forms triangles defining the outer surface of the sampled data set.

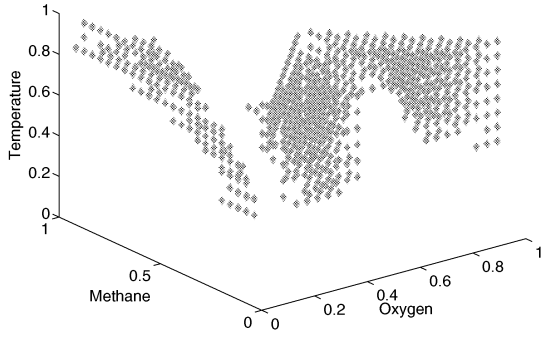

Figure 1: Feasible region of a reduced model of GRI-3.0 mechanism involving 17 species and 59 reactions

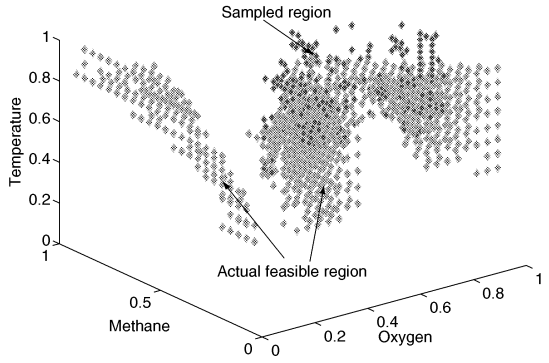

Figure 2: Sampled feasible region of a reduced model of GRI-3.0 mechanism

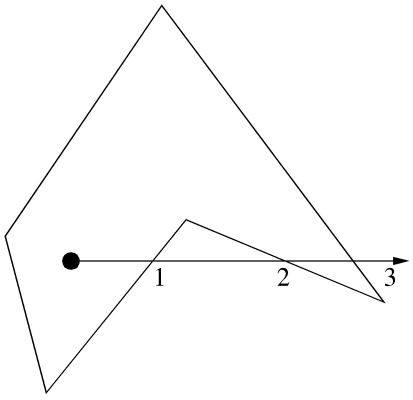

Figure 3: Point-in-polygon test: odd number of intersections means point is inside, even number of intersections means point is outside

Using this procedure, the predicted feasible region is constructed (Figure (4)) using the point-in-polygon test for each point for the reduced model of the GRI-3.0 mechanism used for Figures (1) and (2).

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 Mcke(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 notion of α shape has been extensively applied in automatic mesh generation and geometric modeling, molecular structure and protein folding analysis, distribution of point sets, e.g., galaxies on the universe, etc.

Having defined the surface or shape of the feasible region, the next step involves determination of whether a particular point belongs to the feasible region. This can be done by using one of the point-in-polygon tests (Haines (1994)) used to determine whether a point is inside a polygon or outside it. One way to determine whether a point is inside a region is the *Jordan Curve Theorem*, which states that a point is inside a polygon if, for any semi-infinite ray from this point, there is an odd number of intersections of the ray with the polygon's edges (Figure (3)). Conversely, a point is outside a polygon if the ray intersects the polygon's edges an even number of times, or does not intersect at all.

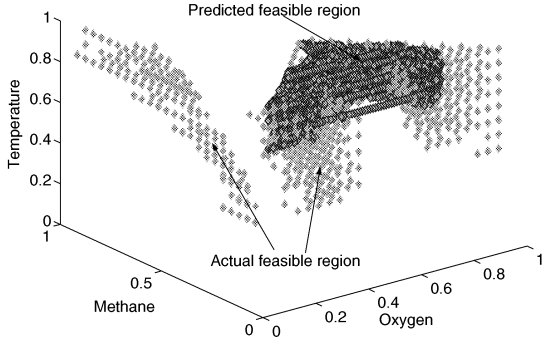

Figure 4: Predicted feasible region of a reduced model

Adaptive reduction technique

To successfully integrate the adaptive reduction with a detailed flow field one needs a set of reduced models which covers the entire range of conditions accessed by the flow simulation. However, this requires *apriori* 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 reduced 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), which is known to provide a stringent test for combustion chemistry. At any time t, the PMSR consists of an even number of particles

(N), the i^{th} particle having composition $\phi^{(i)}(t)$. With Δt being the specified time step, $\phi^{(i)}(t)$ can change discontinuously at the discrete times $k\Delta t$, due to inflow, outflow and pairing of particles. Between these discrete times, the composition evolves by a mixing fractional time step and a reaction fractional time step.

The particles are arranged in pairs: particles 1 and 2, 3 and 4, ..., $N-1$ and N are partners. The mixing fractional step consists of pairs (p and q) evolving by the following equation :

$$\frac{d\phi^{(p)}}{dt} = -(\phi^{(p)} - \phi^{(q)})/\tau_{mix} \qquad (8)$$

where τ_{mix} is the specified mixing timescale. With τ_{res} being the specified residence time, outflow and inflow consists of selecting $1/2N\Delta t/\tau_{res}$ pairs at random and replacing their compositions with inflow compositions, which are drawn from a specific distribution. With τ_{pair} being the specified pairing timescale, $1/2N\Delta t/\tau_{pair}$ pairs of particles (other than the inflowing particles) are randomly selected for pairing. Then these particles and the inflowing particles are randomly shuffled so that they have the probability of changing partners. This simulation is performed untill a stochastic steady state is reached, and all the species compositions and temperature conditions encountered in the simulation are recorded, to have an estimate of the accessed region.

To generate reduced mechanisms to cover the entire accessed space, the nominal points where reduction needs to be performed have to be identified. This is accomplished by clustering the data of temperature and species concentration to identify patterns and obtain 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 to perform feasibility analysis of each reduced set, and describe an α shape around the sampled data points representing the feasible region. This procedure has to be performed iteratively to cover the entire required data space. Once sufficient number of reduced reaction models have been developed to cover the entire estimated accessed region, it can be easily incorporated in the reactive flow models. Following the fractional time stepping approach, an additional step is required in order to evaluate which of the reduced models is feasible in the conditions obtained for the flow time step. Since each reduced model is characterised by a feasible region described by the α shape, this evaluation involves checking the feasible region of all the reduced sets by using the point-in-polygon technique at the current condition, to find one which is feasible. This procedure is followed for the PMSR simulation with 4 adaptive reduced models with an average set size of 50 reactions, which were found to be feasible for 70 % of the accessed conditions. Figure (5) compares the profiles obtained by using adaptive reduced model with a single reduced model consisting of 59 reactions, and the adaptive model was found to have greater accuracy in predicting the profiles.

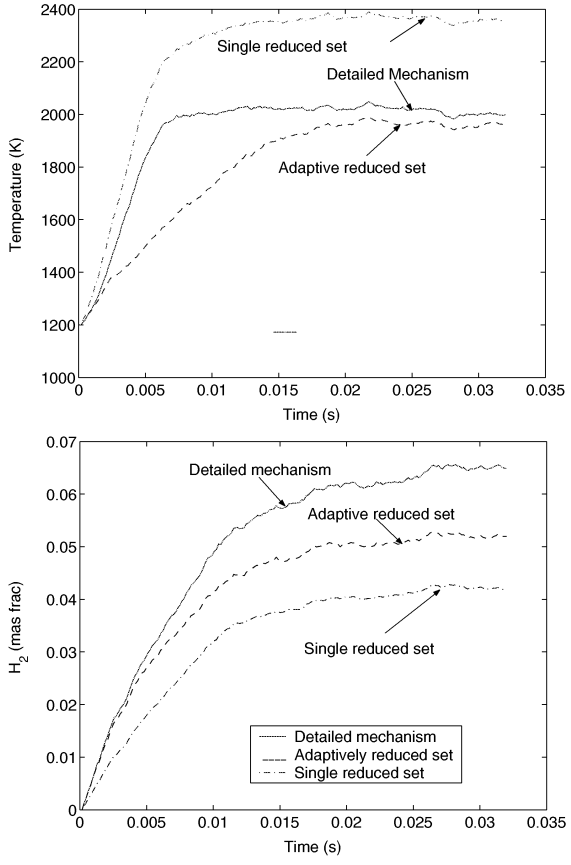

Figure 5: Performance of adaptive reduction in PMSR simulation

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