Development of Reduced Mechanisms for Numerical Modelling of Turbulent Combustion

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Abstract

Recent advances in automation of systematically reduced mechanisms are reported here with the aim to accelerate the development process. A computer algorithm has been developed enabling fast generation and testing of reduced chemistry. This algorithm has been used to develop various reduced mechanisms of methane-air combustion for modelling of turbulent combustion. A 10-step reduced chemistry has been extensively tested showing good performances in predicting a wide range of flame phenomena, including general flame characteristics, flame extinction limits, flame propagation speeds, and auto-ignition delay times. Strategies for using such extensive reduced chemistry for modelling turbulent combustion are briefly discussed.

Introduction

Modeling of coupling between turbulence and combustion is essential to applications which involve strong turbulence-chemistry interactions. For instance, in a lifted turbulent nonpremixed jet flame, strong turbulence-chemistry interactions are observed in regions near the jet nozzle exit where chemical reactions are not fast enough to compete with turbulent mixing. Partially mixed unburned mixtures of fuel and oxidizer are being formed at the base of jet and the unburned mixture is ignited later by burned products formed down stream of the jet. Several modes of combustion are possible, such as propagation of premixed flames with different mixtures and diffusion controlled flames at far downstream. Since turbulence-chemistry interactions play a decisive role in the stabilization of flame, modeling of lifted turbulent jet flames will need a reduced mechanism with sufficient description of chemistry important to ignition process. In addition, flame chemistry in diffusion controlled mode can be of different nature depending on the contents of the combustible mixture. The degree of turbulence-chemistry interaction in practical devices can vary from one mode to another depending on the operation regimes. Often a large detailed chemistry with several hundred steps is necessary to describe many different modes of chemistry regimes mentioned above. Consequently, development of reduced chemistry to cover a wide range turbulence-chemistry interactions

1

presents a challenge task as only a small number of scalars is feasible under current turbulent combustion models.

An Automatic Numerical Algorithm for Systematically Reduction of Mechanisms

While the general idea of reducing complex kinetic schemes by the introduction of quasi-steady-state (QSS) assumptions has been known to chemists some time, it has become fruitful for combustion applications only recently. A systematic approach has been proposed by Peters and Kee (1987) for constructing reduced reaction mechanisms with the application of the QSS assumption. Based on this approach, several reduced mechanisms have been developed for methane and summarized by Smooke (1991). A study of oscillatory oxidation of hydrogen has been demonstrated using reduced chemistry by Tomlin et al, (1992). For practical combustion, reduced mechanisms for a variety of fuels can be found in the book edited by Peters and Rogg (1993) with applications to premixed flames and counterflow diffusion flames. To further develop this approach toward automation, Chen (1988) formulated a general procedure in constructing reduced mechanism in terms of matrix operations so that it can be performed by a computer. A comprehensive review on the development prior to 1991 is given by Frenklach (1991). Other systematic approaches for developing reduced mechanisms have been proposed including the computer singular perturbation (CSP) by Lam and Goussis (1988), the intrinsic low-dimensional manifolds (ILDM) by Mass and Pope (1992), and the repro-modeling approach by Turanyi (1994). While the reduced reaction method has been shown feasible for the systematic reduction of complex chemical kinetics to a few global reaction steps, developing reduced mechanisms can be a time-consuming task if it is done manually. This is especially true when there are a large number of species and elementary reaction steps in the detailed mechanism. Therefore, there is a need to develop a computer algorithm that automatically constructs reduced mechanisms.

An interactive Computer Assisted Reduction Mechanism Code (CARM) has been developed for automatic generation of reduced chemistry with the ability to produce source codes needed for computing the chemical sources (Chang, 1996). Shown in Figure 1 is the flow chart of using the CARM with Chemkin (Kee, et al., 1989) and several flame codes to develop and test reduced mechanisms. For comparison and post-processing purposes, the numerical solutions of simple-geometry flames with the detailed mechanism are first required. Possible candidates include the zero-dimensional Perfectly-Stirred-Reactor (PSR) and other one-dimensional laminar flames. Currently, the PSR is used for its simplicity (Glarborg, et. al., 1986); however, CARM code can be modified to accept solution from other flame results. As illustrated in Fig. 1, a detailed mechanism is used to solve PSR for certain flame conditions of interest. After successful results are obtained, the CARM reads in the solution file, which contains information on species concentrations, rate of production, and species sensitivity coefficients. With given criteria for discerning the relative importance of the elementary reaction steps, a skeletal mechanism can be developed by eliminating unimportant steps and redundant species. It is

possible to bypass the above step to use the detailed mechanism as the starting mechanism for developing the global reduced chemistry.

To evaluate the performance of the skeletal mechanism, the CARM code facilitates direct comparisons between results obtained with the detailed mechanism and those with the skeletal mechanism. A skeletal mechanism is established if the differences between the two solutions are within the desired limits. The next step is to further reduce the number of species in the skeletal mechanism. The QSS species are selected based on their concentration levels as well as on the rate-of-production analysis. After the selection of the QSS species, a set of independent elementary reaction steps is chosen to eliminate the QSS species in order to permit systematic construction of the reduced mechanism. The detailed algebraic procedures have been formulated in terms of matrix operations (Chen, 1988) which are performed by a computer.

When reduced mechanisms are used to solve problems using the Chemkin package, there is only one subroutine needing modifications, namely the CKWYP routine. This subroutine returns the molar production rates of the species given the pressure, temperature, and mass fractions. Modifications are made for expressing the global reaction rates in terms of the elementary reaction rates which usually are functions of the QSS species. In the CKWYP subroutine the concentrations of the QSS species are solved numerically since they are often coupled nonlinearly. The latest refinement in CARM enables automatic generation of the source code for the modified CKWYP subroutine once the reduced mechanism is constructed. The source code is fully compatible with Chemkin but it can be used independently. Described below are the details of numerical implementation of the concepts outlined above.

Establishing the skeletal mechanism

The skeletal mechanism is developed by identifying and eliminating unimportant reaction steps in the detailed mechanism under a particular flame condition of interest. Let S_k^d, k=1,2,...,K+1 be the temperature and species composition obtained with the detailed mechanism and S_k^s be those obtained with the skeletal mechanism. The criterion to be satisfied by the skeletal mechanism is set as

$$\max_k \left| \frac{S_k^d - S_k^s}{S_k^d} \right| \le \varepsilon \tag{1}$$

where ε is a small number, e.g., 0.05. This number represents the relative differences in the solutions obtained with the detailed and the skeletal mechanisms. The strategy for identifying the unimportant reaction steps is to examine their normalized first-order sensitivity coefficients defined as

$$\left(\frac{\partial \ln X_k}{\partial \ln \alpha_i} \right) \bigg/ \max_i \left| \frac{\partial \ln X_k}{\partial \ln \alpha_i} \right| \text{ or } \left(\frac{\partial \ln T}{\partial \ln \alpha_i} \right) \bigg/ \max_i \left| \frac{\partial \ln T}{\partial \ln \alpha_i} \right|, \tag{2}$$

where α_i is the A-factor in the Arrhenius expression for the forward rate constant, $k_f = \alpha T^\beta \exp(\dfrac{-E_A}{RT})$, and X_k is the mole fraction of the k-th species. A skeletal mechanism consists of only those reaction steps having sensitivity coefficients greater than a certain level, e.g., usually a 5% cut-off level. Comparisons of flame solutions obtained with the skeletal mechanism to those obtained with the detailed mechanism indicate that the sensitivity coefficients are an accurate indicator of the relative importance of each elementary step.

The above criterion, when used alone, may have some shortcomings. The sensitivity coefficients are useful in identifying the 'limiting' steps in chemical processes. If the chemical process occurs in a sequence, the slowest step can be identified by the largest sensitivity coefficient. If the chemical process has several parallel steps, the largest sensitivity coefficient is often associated with the fastest step. In the latter scenario, using the sensitivity coefficient alone may not be sufficient in identifying the unimportant steps. The reason is that the fastest chemical step may have a fast reverse rate leading to a small net contribution to the production or destruction of the species. This situation is resolved by examining the normalized production or destruction rates; their definitions are given below

normalized production value

$$\overline{C}_{ki}^{p} = \frac{\max(\nu_{ki},0)q_i}{\sum_{i=1}^{I}\max(\nu_{ki},0)q_i} \tag{3}$$

normalized destruction value

$$\overline{C}_{ki}^{d} = \frac{\min(\nu_{ki},0)q_i}{\sum_{i=1}^{I}\min(\nu_{ki},0)q_i}, \tag{4}$$

where ν_{ki} is the stoichiometric coefficient of k-th species in i-th reaction, q_i is the rate of progress variable for the i-th reaction step, and I is the total number of reaction steps. It is possible that the first-order sensitivity coefficient of a species with respect to a certain reaction step is small although the corresponding normalized production or destruction value of that species is large. In other words, this particular reaction step would be eliminated by using only the sensitivity analysis. A different approach is to use rate sensitivity analysis and then perform principal component analysis to the sensitivity matrix (Tomlin et al, 1992).

On the basis of the above observation, a procedure is proposed in establishing the skeletal mechanism:
(1) Select those reaction steps with $\partial \ln \dot{\omega}_k / \partial \ln k_i$ (it may be possible to use either $\partial \ln Y_k / \partial \ln k_i$ or $\partial \ln X_k / \partial \ln k_i$) larger than a certain level, e.g., usually a 5% cut-off level. This set of reaction steps forms the preliminary skeletal mechanism.

(2) Examine the remaining reaction steps that are not in the preliminary set to see if values of $\overline{C}_{ki}^p$ or $\overline{C}_{ki}^d$ for the species in the preliminary set are larger than a certain level, e.g., usually a 5% cut-off level. If so, those reactions will be added to the preliminary set.

(3) Check if there are additional species appearing in the reaction steps that are added to the preliminary set.

(4) Repeat (2) and (3) until no more additional species or steps.

It is noted that those eliminated steps may become important under different conditions. Therefore, applicability of the skeletal mechanism is limited to a specific range of conditions. It is possible to use the detailed mechanism to directly develop the reduced mechanism with a more complex form than using the skeletal mechanism.

Searching for the Quasi-Steady-State (QSS) Species

A species is approaching the QSS if its production and consumption rates satisfy the following criterion:

$$\frac{|\dot{\omega}_k^p - \dot{\omega}_k^c|}{\max(|\dot{\omega}_k^p|,|\dot{\omega}_k^c|)} << \delta \tag{5}$$
,

where the production and consumption rates of the k-th species are given by

$$\dot{\omega}_k^p = \sum_{i=1}^{I} \max(\nu_{ki},0)q_i \tag{6}$$

and

$$\dot{\omega}_k^c = -\sum_{i=1}^{I} \min(\nu_{ki},0)q_i, \tag{7}$$

and $\delta << 1$ and δ is usually set between $10^{-2} \sim 10^{-3}$. Equation (5) is not applicable for an inert species which has zero production and consumption rates. The inert gas is identified by its concentration and retained by CARM code automatically. The error introduced by the QSS approximation is proportional to the ratio of concentration of QSS species to the major species (Peters, 1991). The above criteria can be relaxed if the concentration of a certain species is relatively small compared to major species. A modified criterion which combines Eq. (5) and the level of concentration can be expressed as

$$X_k \frac{|\dot{\omega}_k^p - \dot{\omega}_k^c|}{\max(|\dot{\omega}_k^p|,|\dot{\omega}_k^c|)} << \delta, \tag{8}$$

where X_k is the mole fraction of k-th species.

Selecting the independent reaction steps

An independent set of reaction steps from the detailed mechanisms is used to eliminate the QSS species (Dixon-Lewis et al., 1975). This process is equivalent to combining elementary reactions in such a manner that the stoichiometry of the overall chemical system can be represented by a reduced mechanism. The strategy for selecting the independent set of reaction steps is to choose the least sensitive steps involving the

5

QSS species. CARM uses the following scheme to form an independent set of reaction steps.

(1) Begin by choosing the reaction step with the $\partial \ln \dot{\omega}_k / \partial \ln k_i$ value being the smallest for eliminating k-th species.
(2) Check if this reaction step is independent of the steps already selected. If not, it is replaced by one with next smallest value of $\partial \ln \dot{\omega}_k / \partial \ln k_i$. This step is repeated until all the available steps are exhausted.
(3) If none of the steps involving the k-th QSS species can form an independent set with the steps chosen previously, re-select a reaction step for the previous species (k-1 th species).
(4) Repeat (1) to (3) until all species have been assigned a reaction step.

Concentrations of the QSS Species

By applying the QSS approximation, one can obtain algebraic equations expressing the concentrations of the eliminated species in terms of major species. This set of equations is often coupled nonlinearly among those eliminated species. An iteration algorithm is employed in CARM to solve for the nonlinearly coupled equations for the QSS species. Functional and Newton iteration schemes are available and the iteration procedure is stopped when all the species satisfy the following convergence criteria

$$\max_k \left(\frac{\left| C_k^{n+1} - C_k^n \right|}{\max\left(C_k^{n+1}, C_k^n \right)} \right) \leq \varepsilon \tag{9}$$

where C_k is the concentration of the k-th species and superscript n and n+1 represent the solutions at previous and present iterations, respectively. The default value of ε is set to be 10^{-5}.
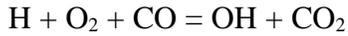
Applications to Methane-Air Combustion

Using the CARM and based on the detailed GRI1.2 and GRI2.1 mechanisms (Frenklach et al., 1994), several reduced mechanisms have been developed and tested. In particular, a 10-step mechanism has been developed to cover a wide range of flame conditions including rich combustion and auto-ignition. The global steps of this 10-step mechanism is summarized below:
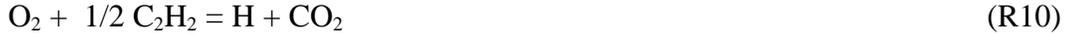
$$O_2 + 2CH_3 + C_2H_4 = H + 2CH_4 + CO_2 + 1/2 \, C_2H_2 \tag{R1}$$
$$O_2 + 2CH_3 = H_2 + CH_4 + CO_2 \tag{R2}$$
$$O_2 + 3CH_3 = H + 2CH_4 + CO_2 \tag{R3}$$
$$O_2 + CH_3 + CO = H + CO_2 + CH_2O \tag{R4}$$

$$O_2 + CO + C_2H_6 = CH_4 + CO_2 + CH_2O \qquad \text{(R5)}$$
$$O_2 + 2CO = 2CO_2 \qquad \text{(R6)}$$
$$H + O_2 + CO = OH + CO_2 \qquad \text{(R7)}$$
$$H_2 + O_2 + CO = H + OH + CO_2 \qquad \text{(R8)}$$
$$O_2 + H_2O + CO = 2OH + CO_2 \qquad \text{(R9)}$$
$$O_2 + 1/2\ C_2H_2 = H + CO_2 \qquad \text{(R10)}$$
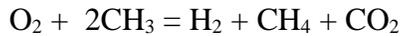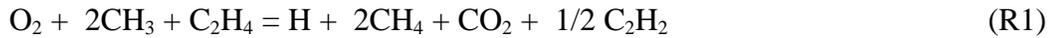
Two other reduced mechanisms will be included for comparisons. By using the QSS approximation for C_2H_2, a 9-step reduced mechanism without NOx chemistry has been developed. Using GRI2.1, two more steps can be added to the above mechanism to include prompt and thermal NO formation leading to a 12-step reduced chemistry. The total number of species in the reduced chemistry is the sum of the total number of steps and the total number of elements. If equal diffusivity of heat and species is assumed, the mixture fraction concept can be used for modeling turbulent combustion. The total number of independent variables in the reduced chemistry is the total number of steps plus one.

Assessment of Reduced Chemistry

The performance of reduced chemistry will be examined over a wide range of turbulence-chemistry interactions. Described below are comparisons of reduced chemistry and detailed chemistry with steady and transient flame models. These comparisons intend to assess the performances of reduced chemistry under widely different flame regimes encountered in turbulent flames.

Perfectly-Stirred-Reactor (PSR):

The Sandia PSR code is the steady state version of a Continuously Stirred Tank Reactor (CSTR) and it is considered the simplest but yet a useful tool for studying chemical kinetics. The reactor consists single inlet and outlet with constant mass flow though the reactor. The mixing of incoming reactants with the fluids inside the reactor is assumed to be infinitely fast such that the mixture inside the reactor is homogeneously uniform. Such simplification leads to a unique time scale associated with the through-flow of the reaction, namely, the residence time. The response of chemical kinetics with residence times is thus a useful test for reduced chemistry . At large residence times, the reactor properties approach the chemical equilibrium limits. Flame extinction occurs when the residence time is reduced to a point at which chemical reaction can not be sustained. Due to its simplicity, numerical calculations of PSR with detailed chemistry up to several thousand steps can be routinely performed. Figure 2 illustrates a sample comparison of temperature, CO, and NO for methane air combustion at equivalence ratios of 0.8, 1.0, and 1.4 using the detailed GRI 2.1 mechanism and a 9-step reduced chemistry. Excellent agreement between the detailed and reduced mechanisms is seen.

One-Dimensional Flame Propagation:

Calculations of freely propagating one dimensional flame have been extensively performed by combustion researchers to study chemical kinetics. Due to its importance to many combustion applications, laminar flame speed is one of the parameters essential to combustion engineers. Figure 3 presents comparisons of results obtained with detailed and two reduced mechanisms for methane air combustion over equivalence ratios from 0.6 to 1.4. Near the stoichiometric the flame speeds are over-predicted by the reduced chemistry by about 10 to 15% but very good agreement is obtained for rich and lean mixtures.

Opposed Flow Flames:

Opposed flow flames are widely used for studying the effect of stretch rate on flames. In this section, the Tsuji type opposed flow flames are modeled using the Sandia flame code (Miller et al, 1984). The predicted peak flame temperatures versus the strain rate using the detailed GRI2.1 mechanism and a 12-step reduced chemistry including NOx reactions are presented in Figure 4 and the results from the 10-step and 9-step mechanisms are similar (no presented). The predicted extinction limit by the reduced chemistry is seen in very good agreement with the detailed chemistry. The peak flame temperatures are slightly over-predicted by the reduced chemistry at low strain rates. Figure 5 presents the predicted flame structures in the mixture fraction space for a=100/s. As seen in the comparisons, the 9-step reduced chemistry in which the C_2H_2 species is removed predicts much higher CO and C_2H_4 levels in fuel rich parts of the flame. The comparisons of radicals and intermediate species, H, OH, CH_3 and CH_2O are presented in Figure 6 showing very good agreement among the reduced mechanisms and the detailed mechanisms.

Transient PSR (or CSTR):

Transient flames are important to modeling of turbulent flames which are highly transient in nature. One of the possible ways to study transient flame is to extend the steady state flame models to transient situation. However, the extension may need significant modification of the solution techniques as one-dimensional steady state flames are often solved by Newton iterations. The equations describing transient PSR can be expressed by

$$\frac{dY_k}{dt} = -\frac{1}{\tau(t)}(Y_k - Y_k^*) + \frac{\dot{\omega}_k W_k}{\rho}, \tag{10}$$

$$C_p \frac{dT}{dt} = -\frac{1}{\tau(t)}\sum_{k=1}^{K} Y_k(h_k - h_k^*) + \sum_{k=1}^{K}\frac{h_k \dot{\omega}_k W_k}{\rho} - \frac{\dot{Q}}{\rho V}, \tag{11}$$

where superscript * denotes properties of reactants entering the reactor, $\dot{Q}$ is heat loss and V is the volume of the reactor. The residence time has the following function form to mimic turbulence as

$$\log_{10} \tau(t) = \log_{10} \tau_{mean} + \log_{10}(\frac{\tau_{mean}}{\tau_{ext.}})\sin(2\pi ft), \qquad (12)$$

where τ_{mean} is the mean residence time and $\tau_{ext.}$ is the extinction limit based on steady state solution, and *f* is the frequency. A typical comparison of temperature and CO is shown in Figure 7 showing good agreement and the corresponding results for radicals are presented in Figure 8 with quite satisfactory performances from the reduced chemistry. It is interesting to note that both detailed mechanism predicts lower levels of NO when unsteady effects are introduced.

Auto-Ignition Delay Times:

Using the Plug-Flow-Reactor (PFR), the auto-ignition delay times have been computed for a stoichiometric mixture of methane air with initial temperatures ranging from 1000K to 1800K. Figure 9 summaries the predicted delay times using detailed GRI1.2 mechanism and the 10-step reduced chemistry. The errors in the predicted delay times by the 10-step reduced chemistry are within 15% as seen in the figure.

Transient Well-Mixed-Reactor

In addition to the test of the auto-ignition delay times, turbulent flames are often stabilized by ignition of unburned mixtures by the burned products. The transient Well-Mixed-Reactor (WMR) provides a useful tool to study this phenomena. A conceptual picture of the WMR is presented in Figure 10 showing a well-mixed reactor with a injection stream which consists of burned products of a lean mixture. In the simulation, the reactor initially contains a premixed mixture of methane-air at equivalence ratio of 0.6. The injecting stream has the same composition except it is fully burned at the equilibrium state. The predicted time evolution of chemical kinetics is presented in Figure 11 with ignition occurring around 5 ms. The agreement between the reduced and detailed mechanisms is quite satisfactory.

Partially-Stirred Reactor (PaSR):

The PaSR relaxes the assumption of infinitely fast mixing and it better represents turbulent combustion in practical applications. A stochastic modeling approach for PaSR has been recently developed by Correa (1993) for premixed combustion and by Chen (1997) for nonpremixed combustion. In the PaSR, the mean thermo-chemical properties are assumed to be spatially homogeneous but not well-mixed at the molecular level. One additional parameter that describing the molecular mixing process is prescribed so that the

effect of finite-rate mixing on chemical kinetics can be systematically studied. The transport equation for the joint PDF for the PaSR is

$$
\frac{\partial \tilde{P}_{\bar{\phi}}(\overline{\psi},t)}{\partial t} = -\sum_{\alpha=1}^{k} \frac{\partial}{\partial \psi_{\alpha}} \left\{ S_{\alpha}(\overline{\psi})\tilde{P}_{\bar{\phi}}(\overline{\psi},t) \right\} + \frac{1}{\tau_{res}} \left\{ \tilde{P}_{\bar{\phi},inlet}(\overline{\psi}) - \tilde{P}_{\bar{\phi}}(\overline{\psi},t) \right\}
$$
$$
- \sum_{\alpha=1,\beta=1}^{k} \frac{\partial^2}{\partial \psi_{\alpha}\partial \psi_{\beta}} \left\{ \left\langle \varepsilon_{\alpha\beta} | \overline{\phi} = \overline{\psi} \right\rangle \tilde{P}_{\bar{\phi}}(\overline{\psi},t) \right\},
$$

(13)

where the properties and mass fluxes of the streams entering/leaving the reactor have been assumed constant. Furthermore, the sum of mass fluxes entering the reactor, $\dot{m}$, is set equal to the mass flux of the exit stream, and thus the mass of fluids inside the reactor remains constant. Under these conditions, the mean residence time can be identified as $\tau_{res} = m_{reactor} / \dot{m}$. In Eq. (13), the first two terms on the right hand side represent the effects of chemical reaction and the through-flow on the joint scalar PDF respectively. Both of these terms do not need modeling. The last term stands for the effect of micro-scale mixing on the PDF which requires modeling. The simplicity of this model permits us to carry out simulations with detailed chemistry without significant computing burden. From the general view of modeling turbulent reactive flows, PaSR can be viewed as a single grid cell embedded in a large computational scheme. Consequently, PaSR offers an ideal test bed for reduced chemistry in an environment closely resembles to the turbulent flows.

Figure 12 presents comparisons of computed mean temperature, CO and NO concentrations with the detailed and the reduced chemistry for methane air combustion showing good agreement. A comparison of mean statistics of the H radical and CO conditioned on mixture fraction is shown in Fig. 13. As seen in the figure, both the CO and H predictions are improved when C_2H_2 is included in the calculations.

Implementation of Reduced Mechanisms into Turbulent Combustion Models

The systematically reduced mechanisms can be readily used for modeling of turbulent flows with approaches, such as flamelet model or Conditional Moment Closure method. For Probability Density Function (PDF) approach, direct numerical integration of stochastic events may not feasible for two-dimensional flows. Methods of tabulation can be used to reduce the computational time by 1000 folds making the computation feasible. For reduced chemistry with a small number of independent scalars, a structured look-up table can be constructed and used during the numerical simulations of turbulent combustion with the PDF approach (e.g., Chen et al, 1989). A recent development of an in situ adaptive tabulation by Pope (1997) enables incorporation of up to 16 scalars in simulations of turbulent flames.

Summary

A computer algorithm for automatic generation of reduced chemistry by systematically reduction is described in detail. With this program, several reduced mechanisms have been developed for methane air combustion. A 10-step reduced chemistry has been generated to cover a wide range of combustion regime. Extensive comparisons between reduced and the detailed GRI mechanisms have been conducted showing a very satisfactory agreement among the results. Recent advances in numerical algorithms for tabulation have made it possible to use an extensive reduced chemistry up to 16 species.

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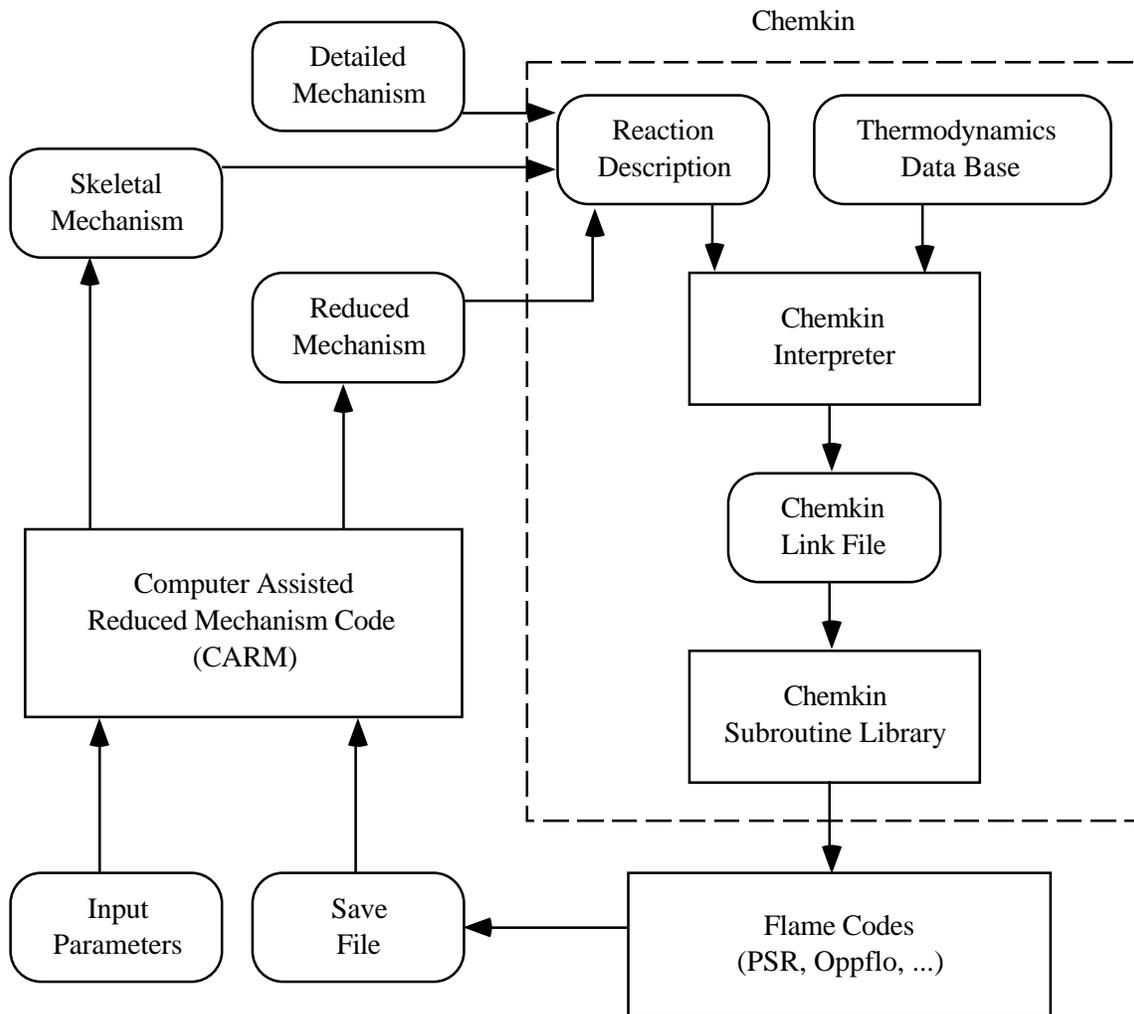
Fig. 1 Flow chart of using CARM with Chemkin and flame codes for automatic development of reduced mechanisms.

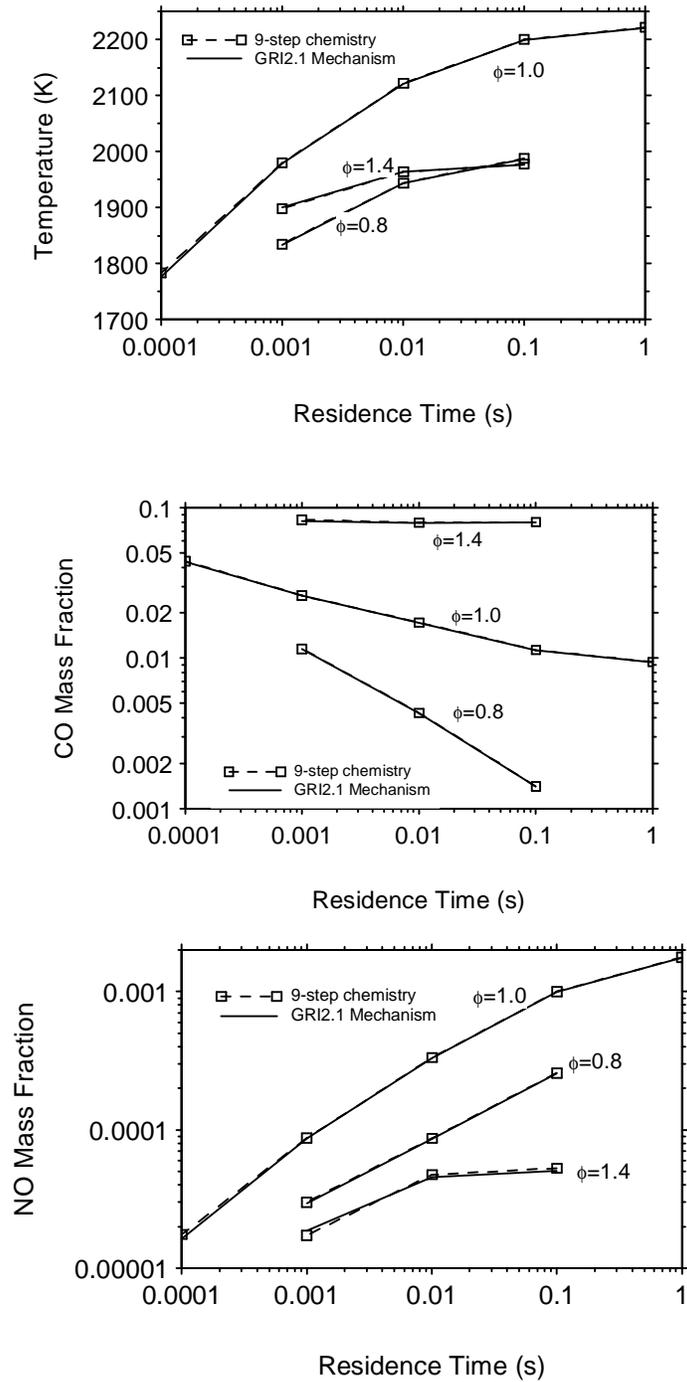Perfectly-Stirred-Reactor (PSR): CH4-Air ϕ=1,0.8,1.4, P=1atm, T_0=300K

Fig. 2 Comparison of Perfectly-Stirred-Reactor (PSR) among reduced and detailed mechanisms showing good agreement for lean, rich, and stoichiometric mixtures over a wide range of residence times
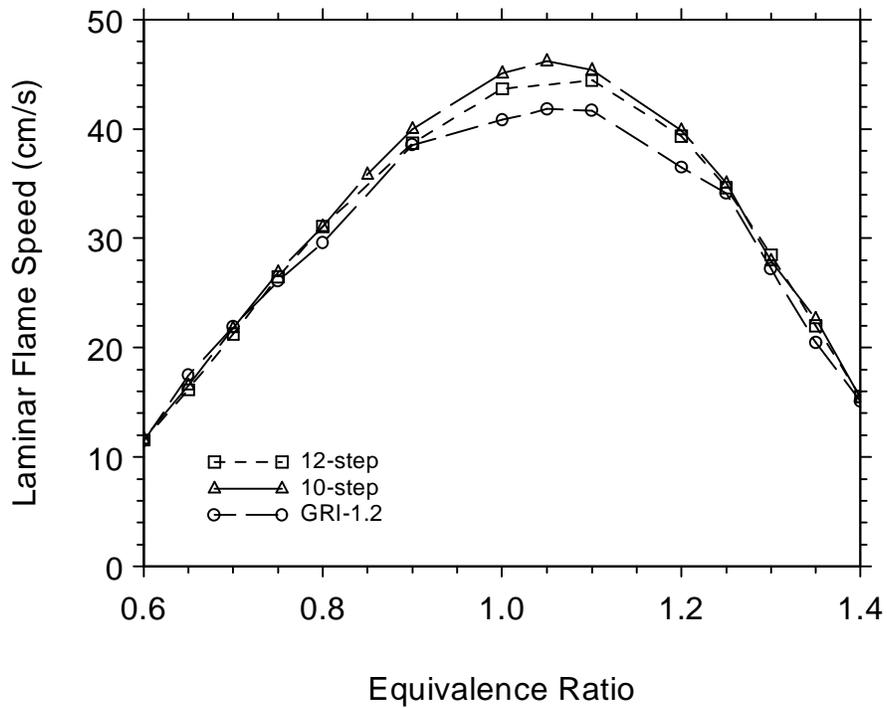
Fig. 3 One-Dimensional freely propagating flames. The flame speeds are overpredicted by about 15% by the reduced mechanisms for mixtures near stoichiometric. For rich and lean mixtures excellent agreement among the mechanism is seen.

Fig. 4 Predicted peak flame temperatures for counterflow diffusion Tsuji methane-air flames at ambient conditions.

Fig. 5 Predicted structures of counterflow diffusion Tsuji flame at a strain rate of 100/s. The C_2H_2 is not included in the 9-step mechanism resulting in incorrect CO levels in rich parts of flame.

Fig. 6 Predicted intermediate and minor species of counterflow Tsuji flame at a strain rate of 100/s showing good agreement among the results obtained with reduced and detailed mechanisms.

Fig. 7 Predicted temperature and CO profiles for a transient PSR containing stoichiometric mixture of methane and air. CH_4 Transient well-mixed reactor $\phi=1.0$ ambient conditions $\tau_{ref.}=0.9$ ms, $\tau_{ref.}/\tau_{ext.}=10$, f=5 KHz.
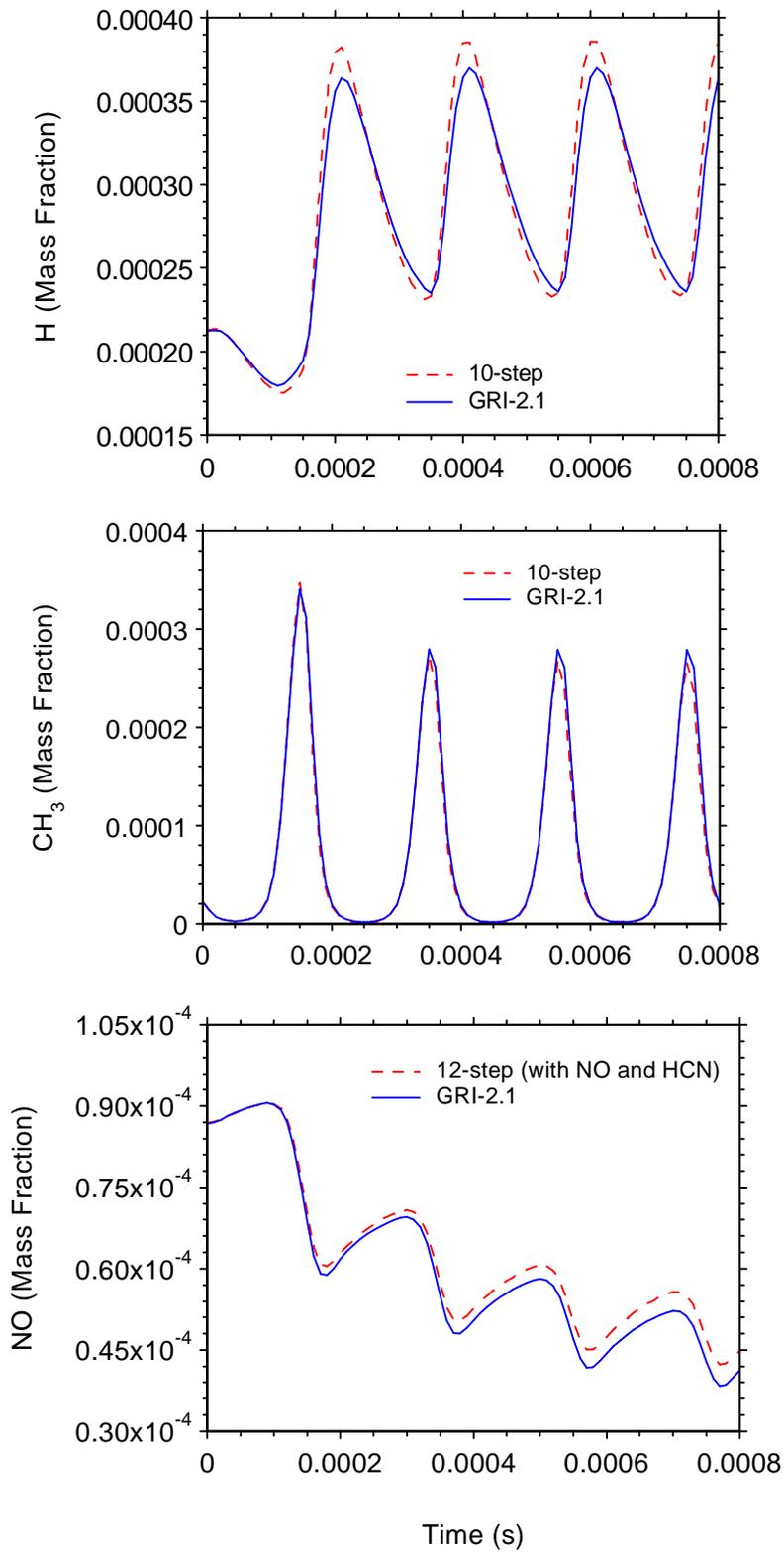
Fig. 8 Comparisons of computed CH_4 transient well-mixed reactor with GRI2.1 and the 10-step reduced chemistry at ϕ=1.0 ambient conditions $\tau_{ref.}$=0.9 ms, $\tau_{ref.}/\tau_{ext.}$=10,   f=5 KHz.

19

Fig. 9 Predicted auto-ignition delay times for a stoichiometric mixture of methane air at ambient pressures. The differences between reduced and detailed mechanisms are within 15%.

Fig. 10 Schematic of a transient well-stirred-reactor showing the growth of the reactor by entrainment of surrounding fluids.

Fig. 11 Predicted properties of a transient well-stirred-reactor initially containing unburned methane air mixture of equivalence ratio of 0.6. Completely burned product of the same mixture in injected into the reactor causing ignition. Good agreement between detailed and reduced chemistry is obtained. Simulation conditions: Premixed CH_4-air Combustion: $\phi_{initial}$=0.6 (unburned), $\phi_{injecting}$=0.6 (burned), T_{air}=800K, P=1atm.
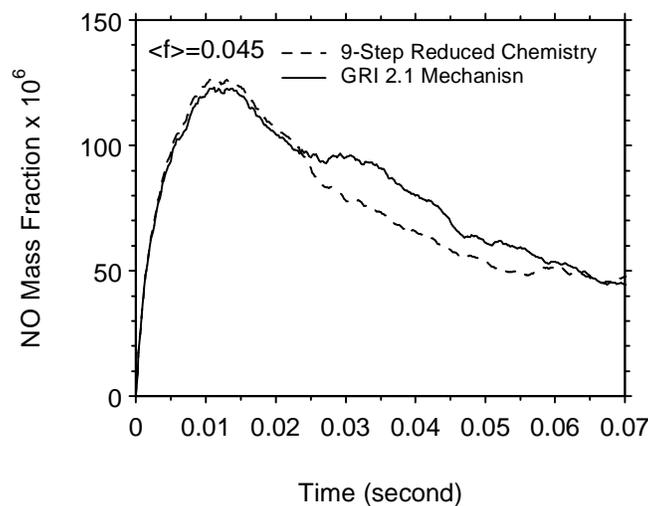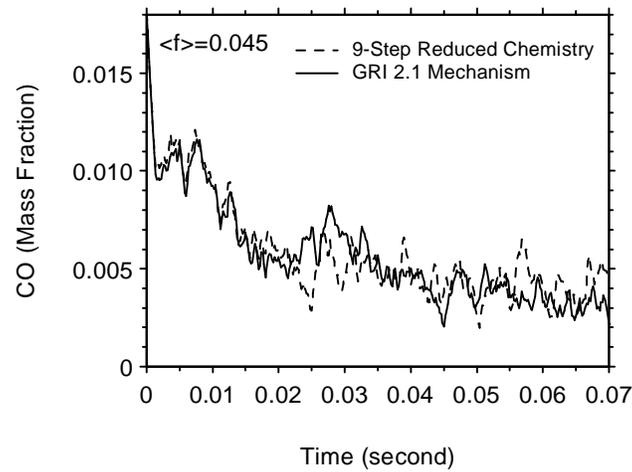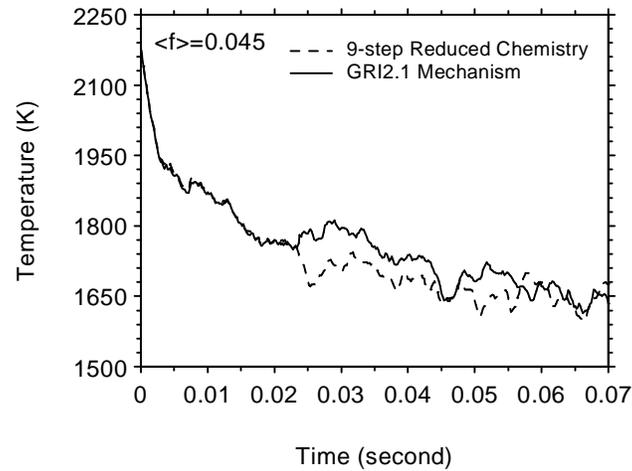
Fig. 12 Comparison of predicted mean temperatures, CO, and NO levels for a partially-stirred-reactor of non-premixed methane air of equivalence ratio of 0.8. Simulation conditions: overall equivalence ratio=0.8, mean residence time=10ms, mixing frequency=5,000 Hz, modified Curl's model.
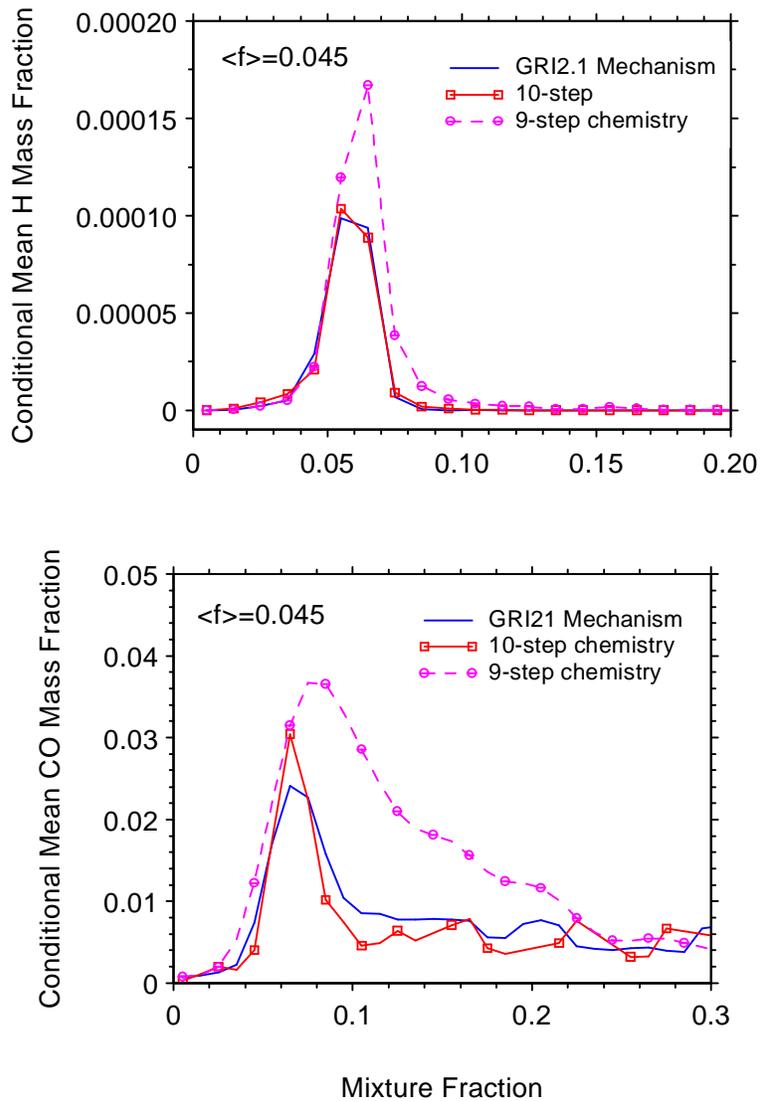
Fig. 13 Comparison of predicted conditional mean statistics of H and CO for a partially-stirred-reactor of non-premixed methane air of equivalence ratio of 0.8. Simulation conditions: overall equivalence ratio=0.8, mean residence time=10ms, mixing frequency=5,000 Hz, modified Curl's model.

Figure Captions:

Fig. 1 Flow chart of using CARM with Chemkin and flame codes for automatic development of reduced mechanisms.

Fig. 2 Comparison of Perfectly-Stirred-Reactor (PSR) among reduced and detailed mechanisms showing good agreement for lean, rich, and stoichiometric mixtures over a wide range of residence times

Fig. 3 One-Dimensional freely propagating flames. The flame speeds are overpredicted by about 15% by the reduced mechanisms for mixtures near stoichiometric. For rich and lean mixtures excellent agreement among the mechanism is seen.

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Fig. 10 Schematic of a transient well-stirred-reactor showing the growth of the reactor by entrainment of surrounding fluids.

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