

IMPLEMENTATION OF REDUCED CHEMICAL KINETIC MECHANISMS IN THE PDF CALCULATION OF TURBULENT NON-PREMIXED H₂-CO₂ FLAMES

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ABSTRACT

The transport equation for the joint velocity-composition probability density function (pdf) has been solved for pilot-stabilised turbulent jet nonpremixed flames of H₂-CO₂ fuel mixture using Monte Carlo methods. A three-step chemical kinetic mechanism, reduced systematically from a more detailed scheme is used in the calculations to represent the chemistry. To reduce computational time, look-up tables are generated for density and other dependent properties as well as composition increments due to chemical reaction. The paper outlines the methodology of generating these tables and incorporating them in the Monte Carlo computations.

Solutions are obtained on a Sun-Sparc1 station using 30000 Monte Carlo particles. The initial and boundary conditions for the modelled flames are similar to those investigated experimentally. The results from the calculations are compared with data obtained using the joint Raman-Rayleigh-Fluorescence technique. Good agreement is obtained with the measured temperature and stable species mass fractions for flames where the chemical kinetic effects are not so significant.

INTRODUCTION

The systematic reduction of detailed chemical kinetic mechanisms to much shorter schemes is a significant advancement of combustion science. The use of these reduced mechanisms with the probability density function (pdf) methods enables, in principle, the computation of finite-rate chemistry effects in flames. This is an important objective in combustion research. The full chemical kinetic mechanisms describing the oxidation of methanol (Chen,1991), methane (Bilger et al.,1990) and CO-H₂-N₂ (Rogg and Williams,1989) fuels have been reduced, respectively, to five-, four-, and three-step schemes which are still capable of predicting, reasonably well, the detailed laminar flame structure as well as flame extinction.

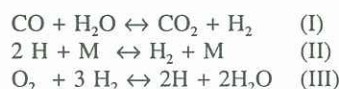
Chen et al.(1988-1990) have used the Monte Carlo method to solve the transport equation for the joint pdf of composition, $f(\psi;x,t)$ for jet flames of various fuels. Reduced mechanisms have been used for CO-H₂-N₂ (Chen et al.,1990), methane (Chen et al.,1989) and methanol (Chen,1988) fuels to represent the chemistry. Nguyen and Pope (1984) and Masri and Pope (1990) have also used the Monte Carlo method to solve the transport equation for the joint velocity-composition pdf for jet flames. The

use of reduced chemical kinetic mechanisms with this approach has not yet been attempted.

This paper illustrates the use of reduced chemistry with the joint velocity-composition pdf method to calculate the structure of pilot-stabilised turbulent nonpremixed flames of H₂-CO₂ fuels. A three-step reduced mechanism (Rogg and Williams,1988) is adopted and the Monte Carlo method is used to solve the pdf transport equation. To reduce the computational time, multi-dimensional look-up tables are generated for the density, dependent properties and composition increments due to chemical reaction. Results are compared with experimental measurements obtained in similar flame.

TABULATION METHODS:

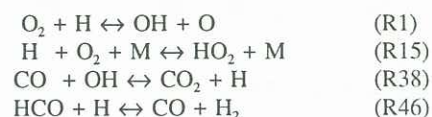
The three-step reduced mechanism used in this paper is adopted from Rogg and Williams (1988):



The global reaction rates are expressed in terms of elementary reaction rates as follows:

$$\begin{aligned} \omega_1 &\approx \omega_{38} & (f,b) \\ \omega_{11} &\approx \omega_{15} + \omega_{46} & (f,f) \\ \omega_{11} &\approx \omega_1 & (f,b) \end{aligned}$$

Where ω_i is the net forward rate for reaction i and f and b are the forward and backward rates respectively. The reaction rates ω_1 , ω_{15} , ω_{38} and ω_{46} correspond to R1, R15, R38 and R46 which are in the full mechanism (Rogg and Williams,1988) and are listed here only for convenience



The species involved in the reduced mechanism are CO, CO₂, H₂, H₂O, O₂, H. The mixture fraction, ξ is defined as

$$\xi = \frac{\beta_i - \beta_{i,0}}{\beta_{i,j} - \beta_{i,0}} \quad (1)$$

where the conserved scalar, β_i is the mass fraction of the element i . Subscripts j and 0 refer to the fuel jet stream and the coflow air stream respectively. Mixture fraction is zero in the oxidant stream and unity in the fuel stream at the jet exit plane. The fuel used is a mixture of 53.5% of H_2 and 46.5% CO_2 (by volume). This corresponds to mass fractions of 5% H_2 and 95% CO_2 giving a stoichiometric mixture fraction, $\xi_s = 0.37$. The mass fractions of carbon, oxygen and hydrogen atoms are given respectively:

$$\begin{aligned} \beta_C &= W_C (\Gamma_{CO} + \Gamma_{CO_2}) \\ \beta_O &= W_O (\Gamma_{CO} + 2\Gamma_{CO_2} + \Gamma_{H_2O} + 2\Gamma_{O_2}) \\ \beta_H &= W_H (\Gamma_H + 2(\Gamma_{H_2} + \Gamma_{H_2O})) \end{aligned}$$

Where $\Gamma_i = Y_i/W_i$, Y_i is the mass fraction of species i and W_i is the molecular weight. The H_2 - CO_2 fuel mixture used in this paper have $\Gamma_{CO_2,J} = 0.0216$, $\Gamma_{H_2,J} = 0.025$ and

$$\begin{aligned} \beta_{C,0} &= 0 \\ \beta_{O,0} &= \alpha, \text{ where } \alpha = Y_{O_2,0} = 0.233 \\ \beta_{H,0} &= 0 \\ \beta_{C,J} &= W_C \Gamma_{CO_2,J} \\ \beta_{H,J} &= 2 W_H \Gamma_{H_2,J} \\ \beta_{O,J} &= 2 W_O \Gamma_{CO_2,J} \end{aligned}$$

The mixture fraction calculated with respect to conserved scalars β_C , β_O and β_H is represented as:

$$\xi_C = \frac{\beta_C - \beta_{C,0}}{\beta_{C,J} - \beta_{C,0}} = \frac{\beta_C}{W_C \Gamma_{CO_2,J}} = \frac{W_C}{W_C \Gamma_{CO_2,J}} (\Gamma_{CO} + \Gamma_{CO_2}) \quad (2)$$

$$\xi_O = \frac{W_O (\Gamma_{CO} + 2\Gamma_{CO_2} + \Gamma_{H_2O} + 2\Gamma_{O_2}) - \alpha}{2W_O \Gamma_{CO_2,J} - \alpha} \quad (3)$$

$$\xi_H = \frac{W_H}{2W_H \Gamma_{H_2,J}} (\Gamma_H + 2\Gamma_{H_2} + 2\Gamma_{H_2O}) \quad (4)$$

Disregarding any differential diffusion effects, the mixture fraction ξ calculated from β_C , β_O and β_H should be identical and $\xi = \xi_C = \xi_O = \xi_H$. Equations 5, 6 and 7 are therefore generated from Eqs. 2, 3 and 4. Since a total of six species are used, then, three other independent variables remain and these must be used in further calculations. The final selected variables are mixture fraction, ξ and three reactive scalars: Γ_{CO_2} , Γ_{H_2O} and Γ_H .

$$\xi \Gamma_{CO_2,J} = \Gamma_{CO} + \Gamma_{CO_2} \quad (5)$$

$$\xi \Gamma_{CO_2,J} + \frac{\alpha}{W_O} (1 - \xi) = \Gamma_{CO} + 2\Gamma_{CO_2} + 2\Gamma_{O_2} \quad (6)$$

$$2 \xi \Gamma_{H_2,J} = 2 \Gamma_{H_2} + 2 \Gamma_{H_2O} + \Gamma_H \quad (7)$$

Upper and Lower Limits

To reduce computational times, three look-up tables are used instead of direct calculations for density, mass densities of dependent variables and composition

increments due to chemical reaction. The density and properties tables are produced for the full allowed space while the table for the composition increments due to reaction is produced only for the reactive space which covers a narrower mixture fraction range bounded by the rich and lean reactive limits, ξ_R , ξ_L respectively. This is useful since it leads to a reduction in computer storage space. Values are stored in the tables for a selected number of nodes which divide the valid domain of each independent variable. This valid domain is bounded by upper and lower limits determined sequentially as follows:

(1) Mixture fraction ξ :

$$\begin{aligned} \text{In reactive space} \quad & \xi^{\min} = \xi_L = 0.1 \\ & \xi^{\max} = \xi_R = 0.6 \\ \text{Allowed space} \quad & \xi^{\min} = 0.0 \\ & \xi^{\max} = 1.0 \end{aligned}$$

(2) The reactive and allowed mixture fraction spaces are divided into a number of nodes and entries in the tables are then generated for each node. Given a value for ξ at any node within the valid domain, upper and lower limits for Γ_{CO_2} are determined. The same formulation is used for Γ_{CO_2} limits in the reactive and allowed spaces.

$$\begin{aligned} \Gamma_{CO_2}^{\max} &= \xi \Gamma_{CO_2,J} \\ \Gamma_{CO_2}^{\min} &= 0.9 \xi \Gamma_{CO_2,J} \end{aligned}$$

For the lower limit, $\Gamma_{CO_2}^{\min}$, it is assumed that a maximum of 10% of the CO_2 available may be converted to CO . This is a realistic limit which is not exceeded neither in the experiments nor in the laminar flame calculations for H_2 - CO_2 fuel mixtures.

(3) The range ($\Gamma_{CO_2}^{\max} - \Gamma_{CO_2}^{\min}$) is then divided into intervals. Given a value of ξ and Γ_{CO_2} , upper and lower limits for Γ_{H_2O} are determined as follows:

In the reactive space

$$\Gamma_{H_2O}^{\max I} = \Gamma_{CO_2,J} \xi - \Gamma_{CO_2} + \frac{\alpha}{W_O} (1 - \xi)$$

$$\Gamma_{H_2O}^{\max II} = \xi \Gamma_{H_2,J}$$

$$\Gamma_{H_2O}^{\max} = \min(\Gamma_{H_2O}^{\max I}, \Gamma_{H_2O}^{\max II})$$

$$\Gamma_{H_2O}^{\min} = 0.3 \xi \Gamma_{H_2,J}$$

In order to maintain adequate resolution in the reactive domain for Γ_{H_2O} we assume that a minimum of 30% of the available H_2 is converted to H_2O . This, of course, is realistic only in the reactive space and is supported by experimental data and laminar flame calculations.

In the allowed space

$$\Gamma_{H_2O}^{\max} = \min(\Gamma_{H_2O}^{\max I}, \Gamma_{H_2O}^{\max II})$$

$$\Gamma_{H_2O}^{\min} = 0.0$$

(4) The range ($\Gamma_{H_2O}^{max} - \Gamma_{H_2O}^{min}$) is divided into a number of nodes each of which is taken separately. Given a value of ξ , Γ_{CO_2} and Γ_{H_2O} , upper and lower limits for Γ_H are determined for both *reactive* and *allowed spaces* using the same formulation. It is assumed, realistically, that a maximum of 5% of the H_2 available may be converted to H atoms to give Γ_H^{max} .

$$\Gamma_H^{max} = 0.05 \xi \Gamma_{H_2, J}$$

$$\Gamma_H^{min} = 0.0$$

To illustrate the tabulation procedure further, Fig. 1 shows the valid domain as well as the upper and lower limits for both Γ_{H_2O} , Γ_H given $\xi=0.5$ and two different values of Γ_{CO_2} . At every node in the valid domain for each variable ($\xi, \Gamma_{CO_2}, \Gamma_{H_2O}, \Gamma_H$) values for density and other properties as well as incremental change in composition due to chemical reaction over a time interval, t are generated and stored. The reaction table is five dimensional in ($\xi, \Gamma_{CO_2}, \Gamma_{H_2O}, \Gamma_H, t$) with dimensions of (20,8,8,8,10), where t represents reaction time intervals. The density and composition tables are four dimensional in ($\xi, \Gamma_{CO_2}, \Gamma_{H_2O}, \Gamma_H$) with dimensions of (20,8,8,8). During the Monte Carlo simulations, the scalar changes due to chemical reaction for each particle in the reactive zone are obtained from the reaction table by an efficient multi-linear interpolation scheme. Density and the mass density of other species in the allowed spaces are interpolated similarly.

Computations

In turbulent reactive flows, the joint pdf, $f(V, \psi; x, t)$ contains a complete one-point statistical description of velocity and composition. The transport equation for the joint pdf $f(V, \psi; x, t)$ is given by:

$$\begin{aligned} \rho(\psi) \frac{\partial f}{\partial t} + \rho(\psi) \frac{\partial f}{\partial x_i} \left(\rho(\psi) g_j - \frac{\partial \bar{p}}{\partial X_j} \right) + \frac{\partial}{\partial \psi_\alpha} \left[\rho(\psi) S_\alpha(\psi) f \right] \\ = \frac{\partial}{\partial V_j} \left[\left(\frac{\partial \tau_{ij}}{\partial x_j} + \frac{\partial p'}{\partial x_j} \right) V_j \psi \right] + \frac{\partial}{\partial \psi_\alpha} \left[\left(\frac{\partial J_i^\alpha}{\partial X_i} \right) V_j \psi \right] \end{aligned}$$

In the above equation, \bar{p} and p' are the mean and fluctuating pressure; g_j is the body force (per unit mass) in the x_j direction; S_α is the mass rate of addition (per unit mass) of species α due to reaction; τ_{ij} is the sum of the viscous and viscous-diffusive stress tensors; J_i^α is the diffusive mass flux of species α in x_i -direction. All the terms on the left-hand side are known, while the terms τ_{ij} , p' and J_i^α on the right-hand side need to be modelled. Details about the modelling and associated constants used in the computations may be found in another reference (Masri and Pope, 1990).

The joint pdf transport equation is represented by a large number of stochastic particles, N . Initial conditions for velocity, ξ , Γ_{CO_2} , Γ_{H_2O} , and Γ_H are assigned to the particles according to their radial location at the jet exit

plane as follows,

	jet	pilot	air
ξ	1.0	0.37	10^{-8}
Γ_{CO_2}	0.0216	0.00735	10^{-8}
Γ_{H_2O}	10^{-8}	0.009	10^{-8}
Γ_H	10^{-8}	0.0005	10^{-8}

Given ξ , Γ_{CO_2} , Γ_{H_2O} , and Γ_H , values for density and other properties are looked up in the tables for each particle and at each step. Also, given a reaction time, t the composition increments due to chemical reaction are looked up from the table. All calculations are done on the Sun-spar1 station and are limited to a downstream axial location $x/R_j \leq 200$. The CPU times required to calculate the flame with a jet velocity $u_j = 130 m/s$ is approximately 3000 minutes.

Results are shown for only one flame with a jet velocity of 130 m/s. This flame is far from extinction but significantly turbulent with a jet Reynolds number of 15000. Experimental data are available for this flame (Masri et al., in press) and comparisons are made in Fig. 2 between computations and measurements of instantaneous temperature and species mass fractions at $X/D_j = 39$ in the flame. The calculations agree reasonably well with the data. The peak temperature and mass fraction of H_2O are within about 10% of the measured values. The peak mass fraction of CO is lower than that measured by about 30% but occurs at the same mixture fraction. On the lean side of the flame at, $\xi < 0.37$, the mass fraction of CO is over predicted with the calculation showing a conversion of CO_2 to CO for very lean mixtures.

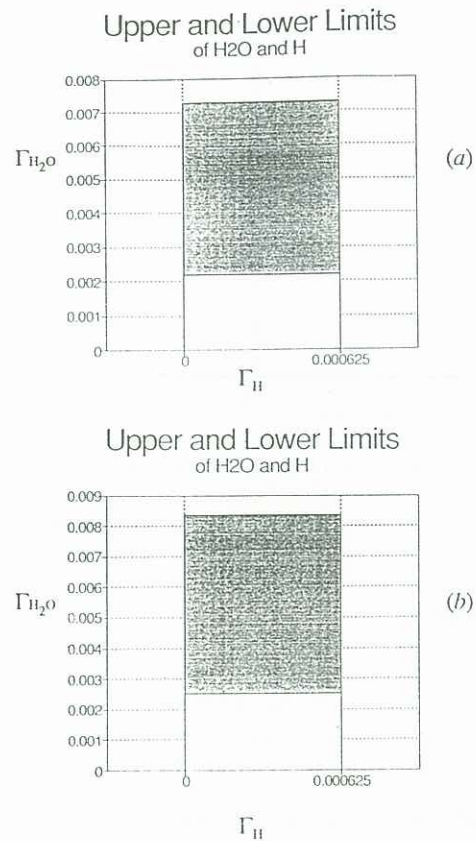


Fig. 1 Valid reactive space for Γ_{H_2O} , Γ_H (shaded region) given $\xi=0.5$ and (a) $\Gamma_{CO_2} = \Gamma_{CO_2}^{max} = 0.018$. (b) $\Gamma_{CO_2} = \Gamma_{CO_2}^{min} = 0.00972$

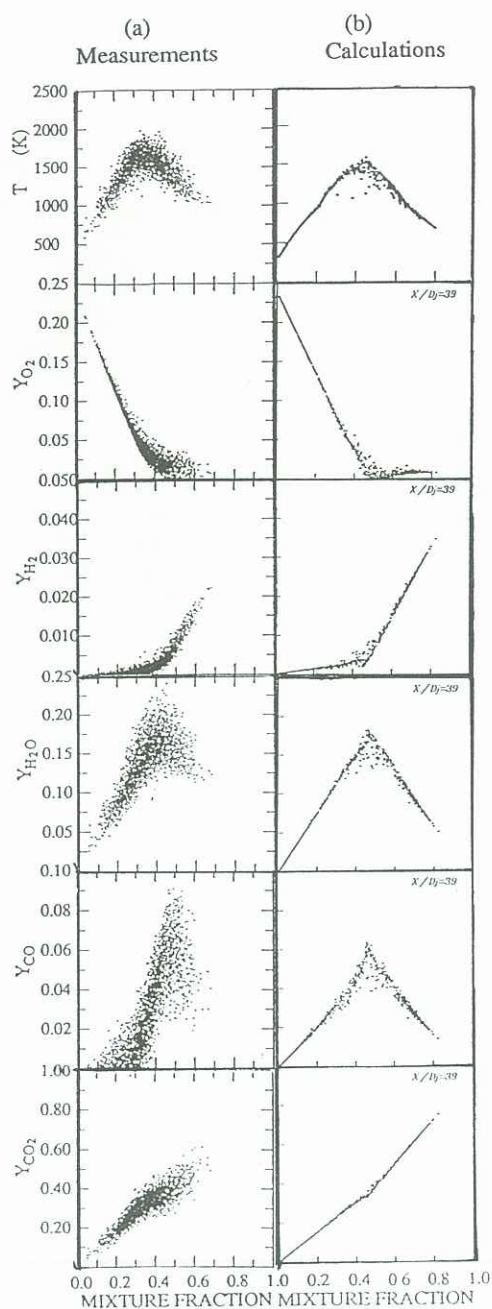


Fig. 2. Comparison of (a) measured and (b) calculated scatter plots for temperature and species mass fractions at $X/D_j=39$ in a flame of H_2-CO_2 fuel with $u_j=130m/s$.

CONCLUSION

A three-step reduced chemical mechanism for H_2-CO_2 fuel mixture is incorporated in the calculation of the transport equation of the joint velocity-composition pdf using Monte Carlo methods. Density, other dependent properties and composition increments due to chemical reaction are determined using look-up tables which are generated only once and used in all subsequent computations. Results for temperature and the concentration of all stable species compare well with experimental data for flames which are reasonably far from extinction.

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