

ACETYLENE AIR DIFFUSION FLAME COMPUTATIONS; COMPARISON OF STATE RELATIONS VERSUS FINITE RATE KINETICS

by

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INTRODUCTION

Fire propagation is driven by the coupling of heat and mass transfer processes between the gaseous and the condensed phases. A significant portion of the heat transfer rate is provided by radiative heat transfer mechanisms of which soot radiation contributes significantly for many flames. Although time history effects are suspected to affect the dynamics of soot evolution within heavily sooting non-premixed flames, the majority of soot chemistry calculations have been conducted for steady flame configurations.

Leung et al. (1991) detailed a four step soot formation model which was used with a detailed gas phase kinetics model to simulate ethylene and propane flames. Fairweather et al. (1992) modeled a methane air jet flame using the soot reaction mechanism of Leung and coworkers (1991) and a conserved scalar/PDF method for the gas species and temperature. Sivathanu and Gore (1994) examined a jet diffusion flame using a similar soot evolution model as Fairweather et al. (1992), but for higher sooting acetylene flames. In this study, the combustion processes for a transient spherical diffusion flame element are computed. The soot mechanism is taken from the above references. The mechanism incorporates the effects of soot nucleation, surface growth, and agglomeration processes. Two methods are used to simulate the major gas species distributions. In the first method, generalized state relations taken from Gore (1986) are used to characterize the major gas species in terms of a mixture fraction type variable. A computational formulation similar to that of Sivathanu and Gore (1994) is used to compute the unsteady combustion process. In the second method, a single-step, finite-rate, chemical reaction mechanism is used to compute the species distributions. The mechanism used is for acetylene-air flames and is taken from Westbrook and Dryer (1981). Results of the unsteady element calculations are compared using both formulations, and similarities and differences between these calculations are noted and discussed.

THEORETICAL MODEL

The thermal element is envisioned to be a spherically symmetric fuel-air element. The equations governing a thermal element reaction are

$$\frac{\partial(\rho_t r^2 \phi)}{\partial t} + \frac{\partial(u \rho_t r^2 \phi)}{\partial r} = \frac{\partial}{\partial r} \left(r^2 \frac{\mu}{\sigma_\phi} \frac{\partial \phi}{\partial r} \right) + S_\phi r^2 \quad (1)$$

with the following boundary conditions

$$\begin{aligned} \frac{\partial \phi}{\partial r} &= 0 & @ r = 0 \\ \frac{\partial \phi}{\partial r} &= 0 & @ r \rightarrow \infty \end{aligned}$$

Mathematically, initial conditions for this system were provided in terms of the dependent variables to be solved. The ideal gas assumption is used for gaseous species and the pressure is assumed to be constant in both space and time.

There exists a one to one functional relationship between gas-phase mixture fraction and species concentration in laminar flame under the laminar flamelet concept and this

relationship is independent of the location-the state relationship. The dependent variables include gas-phase mixture fraction f_g (defined as the fraction of the local material which was originally fuel species and is in gaseous phase), gas-phase total enthalpy $H = \sum c_{pi} Y_i T + \sum Y_i h_{fi}^o$, soot mass fraction Y_c , and soot particle number density N_c . μ is viscosity and is treated globally as a function of temperature only and σ_ϕ are Prandtl/Schmidt numbers. The source terms for each dependent variable are presented in Table 1.

Table 1. The source terms and Prandtl/Schmidt Numbers for state relationship formulation

Variables	S_ϕ	σ_ϕ
f_g	$-S_{cfo} + S_{cox}$	0.7
H	$Q_{rad} - \frac{W_f}{2W_c} S_{cfo} h_{ff}^o + \frac{W_{co}}{W_c} S_{cox} h_{fco}^o$	0.7
Y_c	$S_{cfo} - S_{cox} - S_{Yth}$	500.
N_c	$r_{io} - r_{v\rho_t^2} W_c^{-1/6} Y_c^{1/6} N_c^{11/6} - S_{Nth}$	500.

A one-step reaction mechanism of acetylene of Westbrook and Dryer(1981) is employed in this study. The reaction rate equation has the form of

$$\dot{\omega}_f = A_1 \exp\left(-\frac{E_a}{RT}\right) Y_f^{a_1} Y_{O_2}^{b_1} \rho^{a_1+b_1} W_f^{1-a_1} W_{O_2}^{-b_1}$$

The dependent variables to be solved are C_2H_2 , CO_2 , H_2O , CO , O_2 , H_2 , soot (C), soot particle number density and gas-phase total enthalpy. The source terms for soot mass fraction, soot particle number density and gas phase total enthalpy are the same as in the state relationship formulation.

The conservation equations are nondimensionlized and solved by a control volume finite-difference scheme. The velocity is obtained from the continuity. In the case of state relationship calculations, the gas-phase mixture fraction, soot mass fraction, soot particle number density, total enthalpy, continuity and equations of state are solved iteratively for each time step. The gas species mass fractions are interpolated from the experimental data of Gore(1986) at each iteration. In the one-step reaction case, nine equations plus an equation of state and continuity are solved iteratively. The total density (gas phase and soot) is obtained by a local homogeneity assumption.

RESULTS AND DISCUSSION

The discussion will first consider the global differences between the single step mechanism formulation and the state relations formulations and then will consider the detailed local similarities and differences between the two formulations.

The flame trajectory computed from state relations differs from that computed by the finite rate chemical mechanism. In particular, for low radiative-temperature cases, the differences are much greater than for high radiative-temperature conditions. The flame trajectory was computed using two criteria for the determination of the flame location. For the state relation computations, the flame location is specified in terms of the stoichiometric mixture fraction value, and also in terms of the maximum temperature. For the finite rate computations, the state flame location is calculated by finding the peak heat release rate value. In figure 1, the flame trajectory is compared for the finite rate computations and for the state relations computations. It is noted that the nondimensional time associated with fuel burnout is 14% shorter for the state relations computation as compared with the finite rate computation. It is also apparent that the significant deviation between the two trajectories occurs only after the flame has reversed its direction of propagation and intersects the soot shell which has followed the flame sheet on the fuel side. The effect of the soot shell is to reduce the flame zone temperature

sufficiently such that finite rate chemical effects begin to slow down the reaction rate below the reactant diffusion rate into the flame. Associated with this process is a broadening of the reaction zone. For large radiative environment temperatures, the heat transfer balance in the soot shell is small enough that there is not a significant disturbance to the reaction zone as the flame sheet passes through the soot shell, and for these conditions the flame sheet trajectories for the two formulations are similar.

Although state relations have been experimentally determined for a range of conditions, it was unclear what the effects of computationally determining these relations would be. State relations were extracted from the computational results and it was found that the major gas species using a single step mechanism collapsed into a global relation independent of the time at which the particular "measurement" was made. It was found that the apparent mixture fraction was the correct variable under which the global collapse would occur. The presence of any significant soot mass fractions makes both the true mixture fraction and the gas phase mixture fraction inappropriate independent variables. The species distributions for acetylene, oxygen, water, carbon dioxide as determined by the finite rate calculation are presented in figure 2.

For low radiative environment temperatures the relations for hydrogen and carbon monoxide are not universal/global. Errors are expected in the computed CO species distribution given that the chemical mechanism utilized was only one step and did not explicitly account for the fuel decomposition into CO; all CO generated in the finite rate computations is formed by soot oxidation reactions. It is well established that the soot species distribution does not follow a universal relation, as is shown in figures 3a and 3b. The magnitude of the peak soot volume fraction is nearly independent of the method of computation (i.e., finite rate or fast chemistry). The reaction rates associated with the soot mechanism of Leung et al. were compared with those generated by Gore and Sivathanu (1993). The reaction rates compare favorably, and it was found that the greatest sensitivity to these reaction rates was associated with the radiative environment temperature.

Radiative quenching becomes a possible when the soot shell begins to interact with the flame sheet. The net radiative losses and the chemical heat release are shown in figures 4a and 4b as a function of time. For finite rate chemistry and the radiative temperature case of $T = 1000\text{K}$ the net losses never exceed the chemical heat release rate (fig. 4a), while for the state relations calculations, the crossover occurs for both radiative temperature cases of $T = 500\text{K}$ and 1000K (fig. 4b). These crossover occur at approximately 4 to 5 nondimensional time. The criteria for transient quenching is somewhat more complicated than the a simple balance of total losses to net release; such a criteria has been established by Wichman (1994) and was shown applicable to the transient quenching phenomena in Ezekoye and Zhang (1994).

SUMMARY

In this study, the combustion processes for a spherical acetylene-air diffusion flame element using computed using two different approaches. In the first case, generalized state relations are used to specify the major gas species distributions, while in the second case, a finite rate reaction mechanism is used. A simplified soot mechanism which incorporates the effects of soot nucleation, surface growth, and agglomeration processes was used to specify the soot species evolution. It was found that as the net radiative losses for the diffusion flame element approaches zero that the predictions of the state relations match those of the finite rate chemistry calculations. Computationally derived generalized state relation were found to collapse the major species within the flame, although minor species deviate at large net radiative loss fractions.

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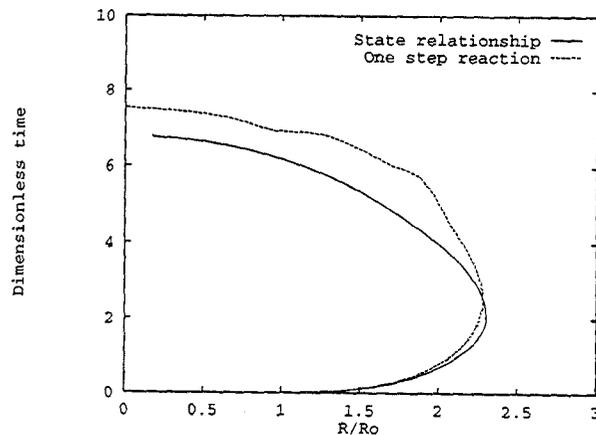


Figure 1. Flame trajectory for finite rate as compared with state relation.
 $R_{\text{fuel}} = 5\text{mm}$, $T_{\infty} = 1000\text{K}$

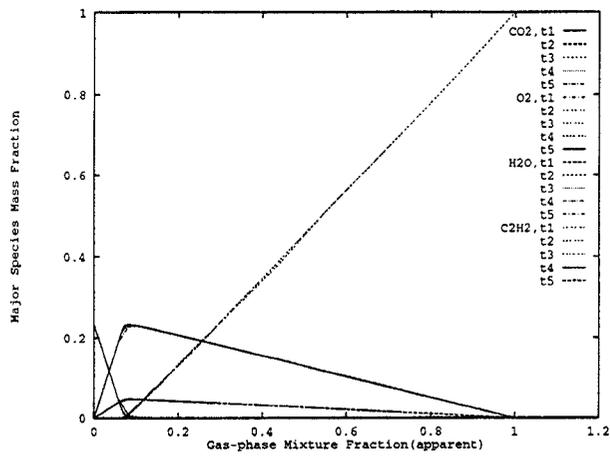


Figure 2. Major gas species state relationship computed using one step mechanism. $T_{\infty}=1000\text{K}$

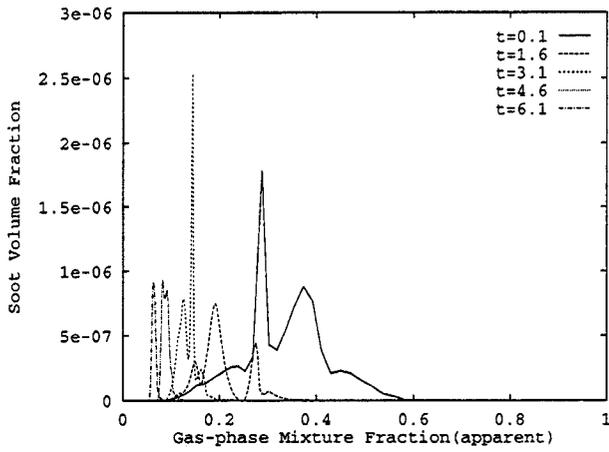


Figure 3a. Soot volume fraction computed using state relationship.

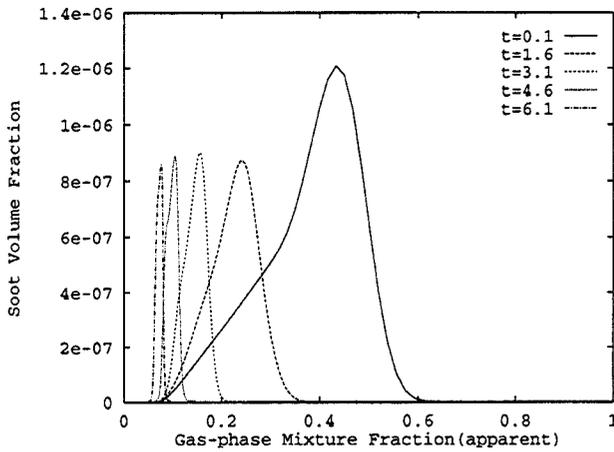


Figure 3a. Soot volume fraction computed using single step mechanism.

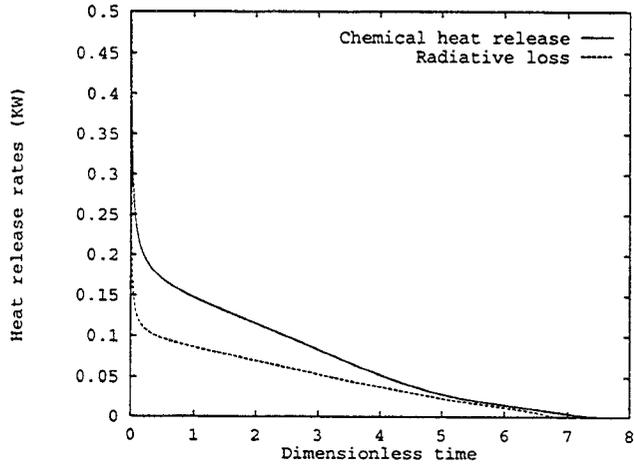


Figure 4a. Heat release rate and net radiative losses computed using one step mechanism; $T_{\infty} = 1000\text{K}$.

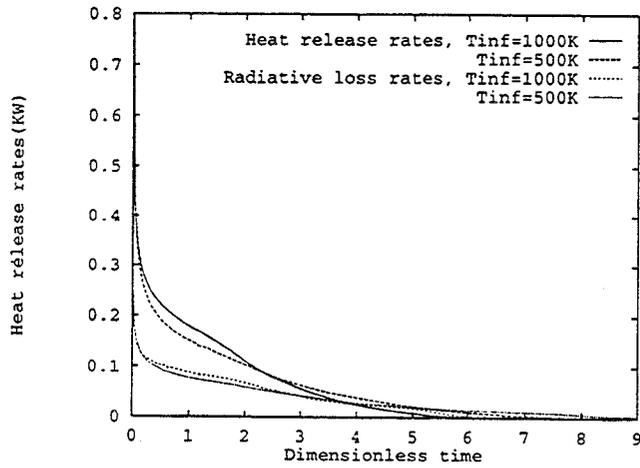


Figure 4a. Heat release rate and net radiative losses computed using state relationships.