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# Numerical Investigation of the Effect of O<sub>2</sub> Addition to Fuel on Soot Formation in Coflow Laminar C<sub>2</sub>H<sub>4</sub>/Air Diffusion Flames

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## INTRODUCTION

The effect of oxygen addition to fuel on soot formation in laminar hydrocarbon diffusion flames has received some attention in the literature [1-9]. Study of the effect of oxygen addition to fuel on soot formation in diffusion flames is of interest both fundamentally and practically. It is well established that addition of oxidative additive (such as O<sub>2</sub> and N<sub>2</sub>O) to ethylene diffusion flames significantly increases soot production [1,2,5,6,7]. Further addition of the oxidative additive to ethylene reduces the soot production or even completely suppresses soot formation as the flame becomes more like a premixed one [1,2]. The oxygen addition to fuel has a less significant effect on soot formation in other hydrocarbon diffusion flames such as propane, n-butane, and methane [1,4,5,7].

The chemical mechanism of oxygen addition to fuel on soot formation in diffusion flames has been explored by Wright [1], Schug et al. [2], Hura and Glassman [5], and recently by Hwang et al. [9]. Wright [1] made an attempt to explain the different effect of oxygen addition to ethylene and propane flames on soot formation in terms of the different catalytic effect of oxygen on the polymerization of these fuels. The positive effect of oxygen addition to ethylene on soot production was attributed to the catalytic effect of oxygen addition on the pyrolysis process of the fuel in the study of Schug et al. [2]. By experimental studies of the effect of oxygen addition to propane flames on soot loading, Wey et al. [3] found that the increased soot loading as a result of oxygen addition cannot be entirely attributed to the increase in flame temperature. Hura and Glassman [5] explained the different effect of oxygen addition to ethylene and propane flames by arguing that oxygen has a different catalytic effect on the pyrolysis process of ethylene and propane: the addition of small amount of oxygen leads no overall increase in the extent of the radical pool, which governs the fuel pyrolysis rate, in propane flames but enhances the radical pool extensively in ethylene flames. Through a series of carefully controlled experiments aiming at separating the different effects of an additive (thermal, dilution, and chemical), Gülder [8] found that oxygen addition to methane chemically suppress soot formation, but otherwise chemically enhances soot formation when added to propane and n-butane. The effect of oxygen addition to ethylene on soot formation in counterflow diffusion flames was recently studied by Hwang et al. [9] in terms of the importance of the recombination reaction of propargyl (C<sub>3</sub>H<sub>3</sub>) leading to polycyclic aromatic hydrocarbon (PAH).

In the present study, numerical calculations of axisymmetric coflow laminar ethylene flames with and without oxygen addition were conducted. The gas-phase chemistry was modeled using a rather detailed C<sub>2</sub> reaction mechanism, while a simple two-equation soot model was employed to describe the soot inception, growth, and oxidation processes. The objective of the present study is to numerically investigate the effect of oxygen addition to ethylene on fuel pyrolysis, flame structure, and soot formation in axisymmetric coflow laminar diffusion flames in order to gain a more comprehensive insight into the effect of oxygen addition in diffusion flames.

## NUMERICAL MODEL

The governing equations of mass, momentum, energy, gas-phase species, soot mass fraction, and soot number density written in axisymmetric cylindrical coordinates are solved. Details of the governing equations and the soot model are described elsewhere [10,11]. Therefore, only a brief summary of the numerical method and various physical and chemical sub-models is given here.

A modified semi-empirical two-equation formulation similar to that developed by Leung et al. [12] was used to model the effects of soot inception, growth and oxidation on the soot mass fraction and number density. The transport equations for the soot mass fraction and number density are given as

$$\rho v \frac{\partial Y_s}{\partial r} + \rho u \frac{\partial Y_s}{\partial z} = -\frac{1}{r} \frac{\partial}{\partial r} (r \rho V_{T,r} Y_s) - \frac{\partial}{\partial z} (\rho V_{T,z} Y_s) + S_m \quad (1)$$

$$\rho v \frac{\partial N}{\partial r} + \rho u \frac{\partial N}{\partial z} = -\frac{1}{r} \frac{\partial}{\partial r} (r \rho V_{T,r} N) - \frac{\partial}{\partial z} (\rho V_{T,z} N) + S_N \quad (2)$$

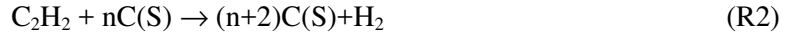
where  $Y_s$  is the soot mass fraction and  $N$  is the soot number density defined as the particle number per unit mass of mixture. Quantities  $V_{T,r}$  and  $V_{T,z}$  are the particle thermophoretic velocities in  $r$  and  $z$  directions, respectively, and are calculated as

$$V_{T,x_i} = -0.65 \frac{\mu}{\rho T} \frac{\partial T}{\partial x_i}, \quad x_i = r, z \quad (3)$$

The source term  $S_m$  in Eq.(1) accounts for the contributions of soot nucleation ( $\omega_n$ ), surface growth ( $\omega_g$ ) and oxidation ( $\omega_o$ ), i.e.

$$S_m = \omega_n + \omega_g - \omega_o \quad (4)$$

This soot model assumes that acetylene is the only species responsible for soot nucleation and growth, although it has been found that PAH also plays an important role in soot formation. The chemical reactions associated with nucleation and surface growth are assumed to be



with the reaction rates given as

$$r_1 = k_1(T)[C_2H_2] \quad (5)$$

$$r_2 = k_2(T)f(A_s)[C_2H_2] \quad (6)$$

where  $f(A_s)$  denotes the functional dependence of soot surface growth on soot surface area per unit volume. Following Leung et al. [12], we assume that the functional dependence is a simple square root relation, i.e.  $f(A_s) = A_s^{0.5}$ . Soot oxidation by the three most important oxidative agents,  $O_2$ ,  $O$  and  $OH$ , in flames was taken into account by assuming the following reactions



The rate of soot oxidation by oxygen was based on the Nagle and Strickland-Constable [13] model with the rate expressions and constants of (R3) and (R4) taken from Ref. 14. Rate constants of (R5) were based on the study of Bradley et al. [15].

The source term  $S_N$  in Eq.(2) accounts for the generation of the number of soot particles due to nucleation process and can be written as

$$S_N = \frac{2}{C_{\min}} N_A R_1 \quad (7)$$

Note that the agglomeration process of soot particles, which reduces the soot particle numbers, was neglected in the present soot model for the reasons discussed in our previous papers [10,11]. In Eq.(7)  $N_A$

is Avogadro's number ( $6.022 \times 10^{26}$  particles/kmol),  $C_{min}$  is the number of carbon atoms in the incipient carbon particle (700).

Radiation heat transfer was calculated using the discrete-ordinates method. Non-grey gas radiative properties of CO, CO<sub>2</sub>, H<sub>2</sub>O, and soot were handled using a statistical narrow-band correlated-k based band model. The gas-phase chemistry was modeled using the GRI-Mech 3.0 mechanism [16]. The governing equations were discretized using the finite-volume method. The SIMPLE-TDMA algorithm was used to solve the resultant coupled equations. However, the gas-phase species equations were solved in a fully coupled fashion at each control volume using the implicit method of Liu et al. [17].

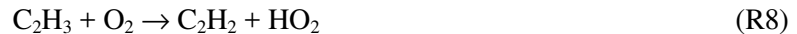
## RESULTS AND DISCUSSIONS

The axisymmetric coflow laminar ethylene diffusion flame at atmospheric pressure considered in this study was generated with a burner in which the pure ethylene or a mixture of ethylene and oxygen flows through a vertical steel tube of 10.9 mm inner diameter and the air flows from the annular region between the fuel tube and a 100 mm inner diameter concentric tube. The volume flow rates of the fuel (ethylene) and the air are kept at 194 ml/min and 284 l/min, respectively, for all the calculations. Both fuel and air are delivered at room temperature (294 K). The computational grid and boundary conditions used in the calculations are schematically shown in Fig.1. Non-uniform grids were used in both  $r$  and  $z$  directions to provide greater resolution in the large gradient regions without an excessive increase in the computing time. Very fine grids were placed between 0 and 1.2 cm in  $r$  direction. The computational domain consists of 102×60 cells. A parabolic laminar pipe flow velocity profile was assigned to the inlet velocity of the fuel stream. For the air stream, a boundary layer velocity profile was assumed inside the boundary layer (formed along the outer surface of the fuel pipe) and the uniform velocity was prescribed outside it.

The effect of 25% oxygen addition to the fuel (volume basis) on the predicted flame temperature is shown in Fig.2 by comparing the results of with and without oxygen addition. Although there is only about 50 K increase in the peak flame temperature when 25% oxygen is added to the fuel, the temperatures in the centerline region near the burner exit increase dramatically as a result of oxygen addition. The magnitude of the maximum temperature increase is only about half of the experimental finding of Wey et al. [3] who observed a 2% increase in the maximum temperature per 10% of oxygen added to propane in a laminar underventilated propane flame stabilized on a Wolfhard-Parker burner. Effect of oxygen addition to ethylene on the flame temperature under conditions similar to the present study, however, has not been reported in the literature. The significant increase in temperatures in the near burner exit region can be attributed to the catalytic effect of oxygen addition on the pyrolysis and oxidation processes of ethylene. It is well known that ethylene dissociation proceeds through [18,19]



The added oxygen enhances the oxidation of the C<sub>2</sub>H<sub>3</sub> radical through [19]



Subsequently, there is a significant increase in the concentrations of important radicals such as O, H, and OH and the temperature in the near burner exit region, providing a positive feedback to the pyrolysis and oxidation of ethylene and various intermediate species. Mass fractions of some important intermediate species in the near burner exit region calculated for the flame of 25% oxygen addition are shown in Fig.3 with peak mass fractions of these species indicated. Under the condition of 25% oxygen addition, some features of the double-flame (an inner premixed one and an outer diffusion one) structure start to appear.

The predicted distributions of soot volume fraction with and without oxygen addition are compared in Fig.4. As a result of oxygen addition, there is a significant increase in the soot volume fraction in the centerline region of the flame due to the combined effect of increased temperature and concentration of acetylene (shown in Fig.5 below). The peak soot volume fraction increases by about 14% with the addition of 25% oxygen. The visible flame height, however, decreases slightly with the addition of oxygen to fuel, which can be attributed to the enhanced soot oxidation. The reduced visible flame height with the addition of oxygen to ethylene also implies an accelerated combustion process.

The predicted distributions of mass fraction of acetylene, the only soot inception and growth species considered in the present soot model, are compared in Fig.5. Similar to the temperature distributions shown in Fig.2, there is a significant increase in the concentration of acetylene in the near burner exit region as a result of enhanced pyrolysis rate of ethylene by oxygen addition. Therefore, the positive effect of oxygen addition to ethylene on soot formation is through both increased flame temperature and the increased concentration of acetylene.

## CONCLUDING REMARKS

Numerical results of the present study reveal that oxygen addition to ethylene has significant effects on flame temperature, soot loading, and flame structure in general, especially in the near burner exit region. The mechanism of the influence of oxygen addition is its catalytic effect on ethylene pyrolysis and subsequent oxidation.

The effect of oxygen addition to propane on soot formation and fuel pyrolysis is currently being investigated numerically in order to understand the different effects of oxygen addition to these two fuels. Experimental studies will also be carried out in our laboratory to validate the numerical results.

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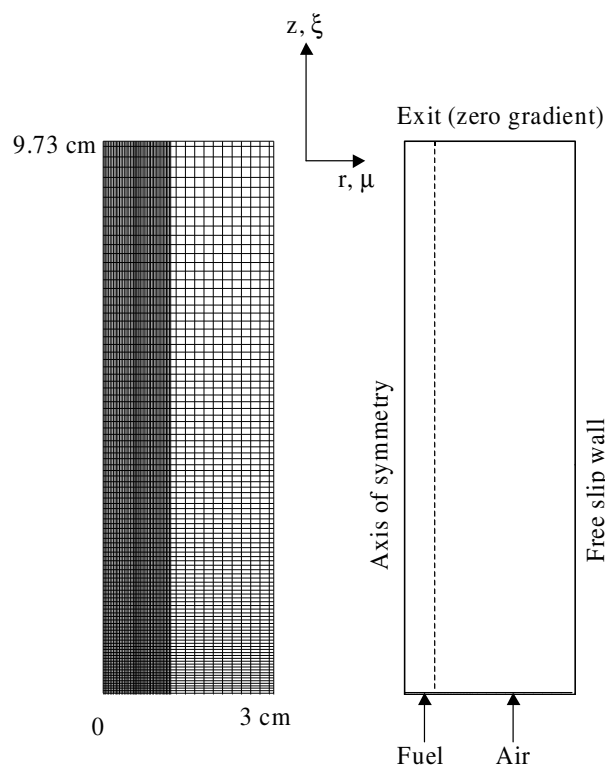


Fig.1 Computational grid and boundary conditions used in the calculations.

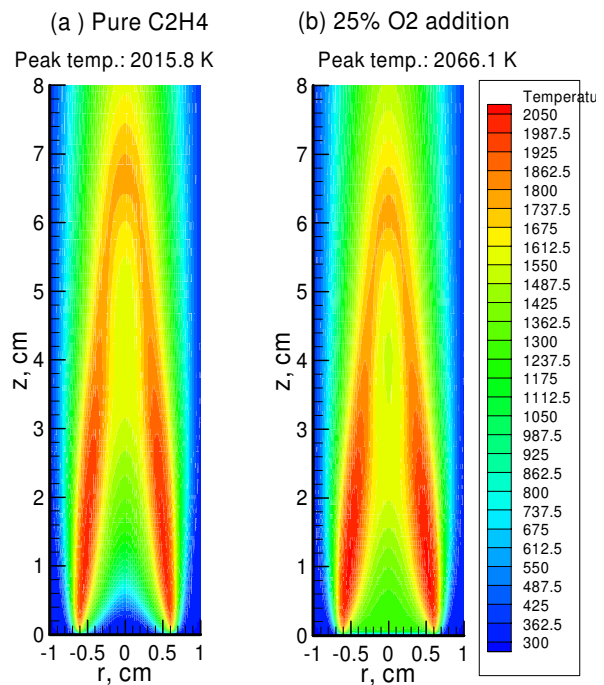


Fig.2 Predicted temperature distributions.

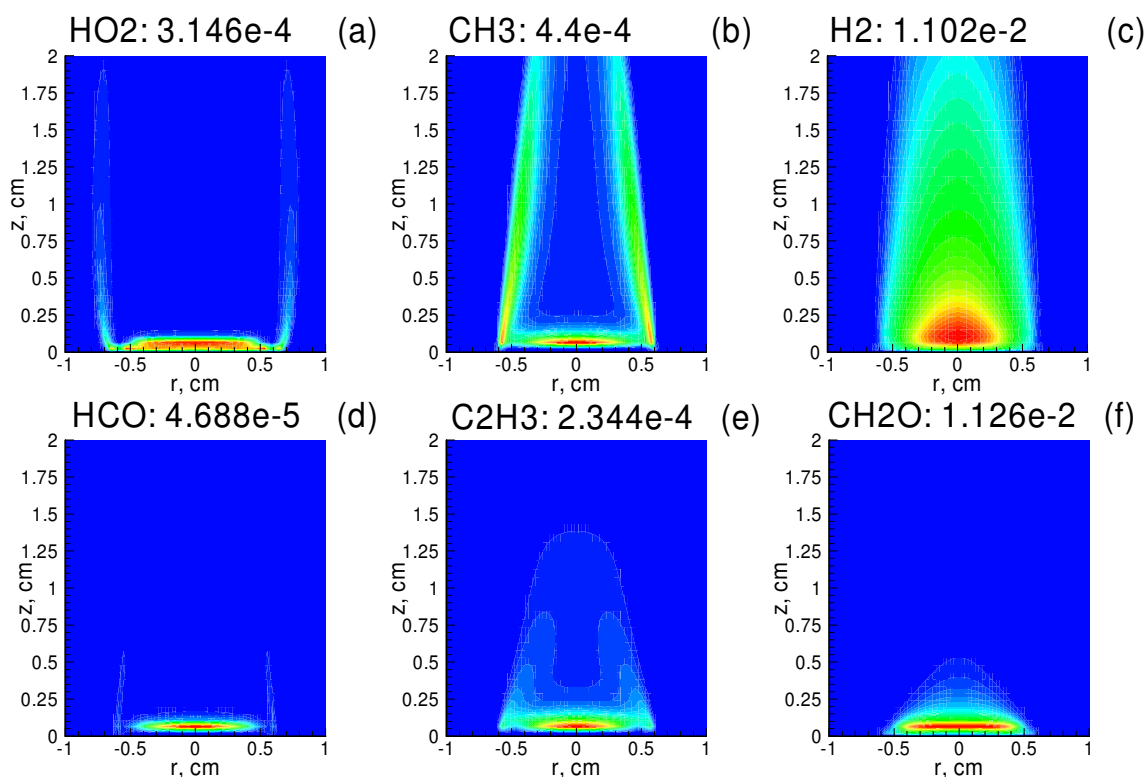


Fig.3 Predicted mass fractions of some important intermediate species in the near burner exit region for the flame of 25% oxygen addition.

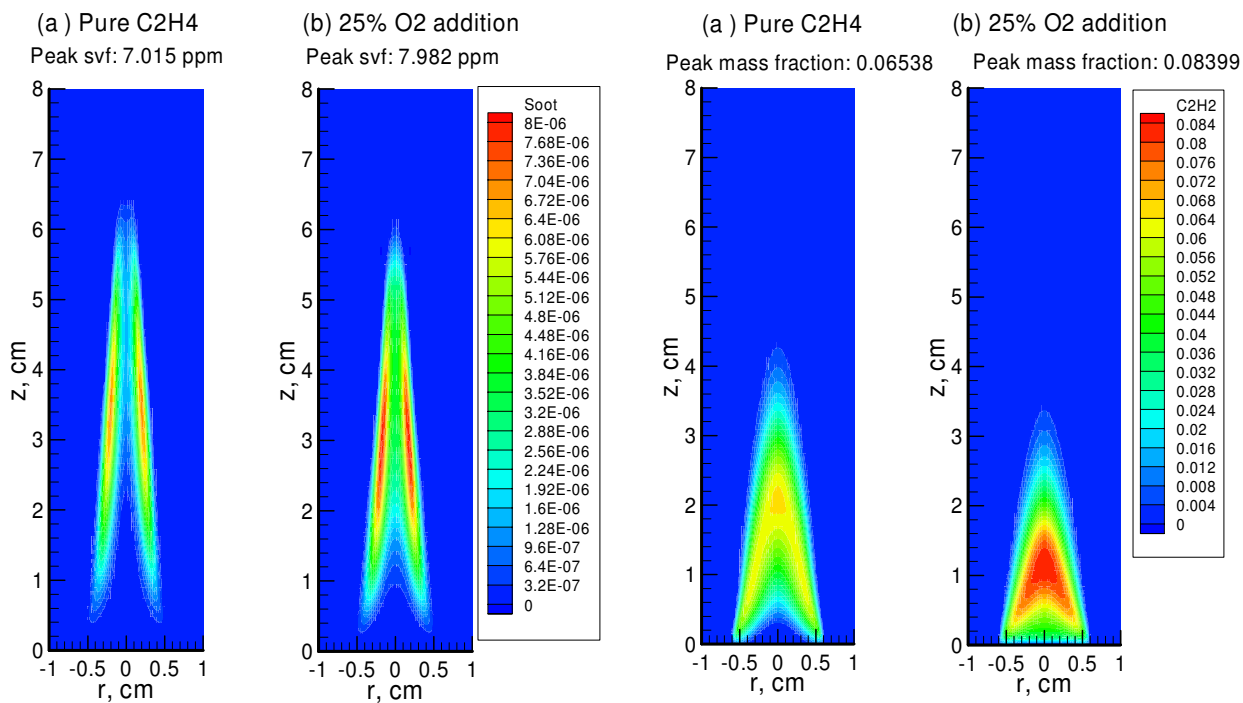


Fig.4 Comparison of the predicted distributions of soot volume fraction.

Fig.5 Comparison of the predicted mass fractions of acetylene.