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# Universal Relationships in Sooting Methane-Air Diffusion Flames

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# Universal Relationships in Sooting Methane-Air Diffusion Flames

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The laminar flamelet concept is based on the premise that scalar properties in laminar diffusion flames are nearly universal functions of mixture fraction. It has been well-tested and proven for temperature and the major species, however, few studies have addressed its applicability for minor species especially the radical species. In this study, we present a direct numerical simulation of an axisymmetric, laminar, methane-air diffusion flame to examine these universal relationships, including the major and minor chemical species, and the radical species. The numerical model solves the axisymmetric, time-dependent, reactive-flow Navier-Stokes equations coupled with sub-models for soot formation and radiation transport, and includes a detailed reaction mechanism for methane-air combustion. Quantitative comparisons with existing experimental data show a slightly wider computed flame compared to the experimental flame, however, the peak values and radial locations of temperature and soot volume fraction line up well with the experimental measurements. To study the universal relationships, scatter plots are made for temperature and the major and minor species throughout the entire flame and compared with existing experimental measurements. Excellent agreement is obtained between the computations and experiments for temperature and mole fractions of CH4, O2, OH, H<sub>2</sub>O, O, H, CO<sub>2</sub> and N<sub>2</sub>, as a function of mixture fraction in the fuel lean, stoichiometric and fuel rich regions of the flame. The computations underpredict the concentration of  $H_2$  and CO in the fuel rich region, however, excellent agreement is obtained in the fuel lean and stoichiometric regions. The computations also overpredict the concentration of  $CH_3$  in the stoichiometric and fuel rich regions, however, the peak concentration occurs at the same mixture fraction for both the experiments and computations. The scatter plots indicate that many of the species studied, including the minor species and radical species, can be considered universal functions of mixture fraction, however some of the species show more scatter than others. The species which showed the least scatter were those whose production rates were fastest in the stoichiometric and fuel lean regions of the flame. Those species whose production rates were highest in the very fuel rich region showed less universality with mixture fraction.

Keywords: Sooting diffusion flames; direct numerical simulations

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#### INTRODUCTION

Over the past few years, there have been several efforts to develop timedependent simulations of axisymmetric jet diffusion flames. Due to computational cost considerations, most of these numerical models have utilized simplified or phenomenological chemical reaction sub-models. Chen et al. (1988); Davis et al. (1991) and Katta and Roquemore (1993) have studied the unsteady interactions between flames and their associated inner and outer vortex structures for  $C_3H_8$ -air flames, using flame sheet and conserved variable approximations. Ellzey et al. (1990, 1991) investigated the effects of heat release viscosity, and gravity on the development and dynamics of H<sub>2</sub>-air flames, using a finite-rate single-step reaction model. Takahashi and Katta (1993) and Hsu et al. (1993) conducted numerical and experimental investigations of flame-vortex interactions in a CH<sub>4</sub>-air jet diffusion flame, using the Shvab-Zel'dovich formulation in conjunction with a flame-sheet assumption. More recently, Katta et al. (1994) and Katta and Roquemore (1995) have incorporated a more detailed chemical reaction model for H<sub>2</sub>-air flames to study the effects of nonunity Lewis number, finite-rate chemistry, and the influence of preferential diffusion on the dynamics and structure of these jet diffusion flames.

Since many practical combustion conditions involve sooting hydrocarbon-air diffusion flames, we have focused our efforts on the development of a time-dependent, axisymmetric, reactive-flow numerical model which also includes the processes of soot formation and radiation transport. Over the past couple of years, we have used this numerical model with phenomenological chemical reaction sub-models to study various time-dependent behaviors of hydrocarbon-air jet diffusion flames. We have investigated the effects of multidimensional radiation transport in an ethylene-air jet diffusion flame (Kaplan et al., 1994a) and showed how radiative losses change the overall temperature, species concentrations, and soot volume fraction distributions, using a finite-rate single-step Arrhenius reaction rate for ethylene-air combustion. This model has also been used to investigate fluid-chemical interactions in lifted methane-air jet diffusion flames using a phenomenological Arrhenius reaction rate (Kaplan et al., 1994b), to study enhanced soot production in time-varying methane-air flames using the conserved scalar approximation (Kaplan et al., 1996a), and most recently, to study the effects of gravity on soot formation in ethylene jet diffusion flames, using a conserved scalar approximation (Kaplan et al., 1996b).

#### SOOTING METHANE DIFFUSION FLAME

In the present work, we further develop this numerical model to include a detailed multi-step, multi-species chemical reaction mechanism, not only to improve the accuracy of the chemical reaction sub-model, but also to enable us to track more species which have recently been determined to be important in the soot formation process. We conduct these simulations for a laminar methane-air flame which has been studied extensively experimentally (Shaddix et al., 1994; Shaddix and Smyth, 1995), and which has been previously studied by us numerically with a phenomenological chemical reaction model (Kaplan et al., 1996a). We present qualitative and quantitative comparisons between the simulation results and the experimental measurements in terms of radial profiles of temperature and soot volume fraction. We then present a series of scatter plots of temperature and major and minor species concentrations as a function of mixture fraction and height in the flame, to investigate whether or not these quantities are universal functions of mixture fraction throughout the entire flame. We compare these computed universal relationships with experimentallymeasured quantities (temperature and species concentrations versus mixture fraction, measured at one height in the flame) in a Wolfhard-Parker rectangular burner using existing data from Norton et al. (1993).

The laminar flamelet concept (Bilger, 1977) is based on the premise that scalar properties in laminar diffusion flames are nearly universal functions of mixture fraction. The applicability of this theory is a very important assumption for computations of turbulent diffusion flames, which frequently utilize libraries of conserved scalar relationships determined from one-dimensional strained (counterflow) flame calculations (Liew et al., 1981, 1984; Peters, 1984, 1986). Laminar flamelet theory has been well-tested and proven for temperature and the major species in diffusion flames (Bilger, 1977; Gore and Faeth, 1986). However, few studies have addressed the applicability of laminar flamelet theory for minor species. especially the radical species. Also, very few comparisons have been made between experimental measurements and detailed falme computations concerning the minor and radical species (Barlow et al., 1990-a, 1990-b; Barlow and Collignon, 1991; Norton et al., 1993). In this paper, we address these issues by comparing our detailed flame computations with experimental measurements (Norton et al., 1993) for temperature and major and minor species (including the radical species), and we test the laminar flamelet theory by presenting results of these quantities as functions of mixture fraction throughout the entire flame, including the sooting region.

## NUMERICAL METHOD

The numerical model solves the time-dependent equations for conservation of mass density. momentum, energy, individual species number densities, soot number density and soot volume fraction:

$$\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho V) = 0 \tag{1}$$

$$\frac{\partial \rho V}{\partial t} + \nabla \cdot (\rho V V) = -\nabla P + \rho G - \nabla \cdot \tau$$
<sup>(2)</sup>

$$\frac{\partial E}{\partial t} + \nabla \cdot (EV) = -\nabla \cdot PV - \nabla \cdot (q_c + q_r) - \nabla \cdot \sum n_k v_k h_k + Q \quad (3)$$

$$\frac{\partial n_k}{\partial t} + \nabla \cdot (n_k V) = -\nabla \cdot (n_k \upsilon_k) + \omega_k \tag{4}$$

$$\frac{\partial n}{\partial t} + \nabla \cdot (nV) = -\nabla \cdot (v_i n) + \omega_n \tag{5}$$

$$\frac{\partial f_{\nu}}{\partial t} + \nabla \cdot (f_{\nu} V) = -\nabla \cdot (\upsilon_t f_{\nu}) + \omega_{f_{\nu}}.$$
(6)

Equations (1-6) are closed by the ideal gas relations:

$$P = n_T k T \tag{7}$$

$$de = \rho C_v \, dT. \tag{8}$$

Equations (1-4) include terms for convection, thermal conduction, molecular diffusion, viscosity, chemical reaction and energy release, gravity, and radiation transport. The soot conservation equations, Eqs. (5) and (6), include terms for convection and thermophoresis, where the thermophoretic velocity is defined by

$$v_t = -0.54 v \frac{\partial \ln T}{\partial r}.$$
 (9)

Our solution to Eqs. (1-6) includes both the radial and axial components of the convective and diffusive transport terms (thermal conduction,

molecular diffusion, and viscosity). However, only the radial component of the thermophoretic term is considered in Eqs. (5) and (6), and only the axial component of the gravitational acceleration term is included in Eq. (2). These equations are then rewritten in terms of finite-difference approximations on an Eulerian mesh and solved numerically for specified boundary and initial conditions. The model consists of separate algorithms for each of the individual processes, which are then coupled together by the method of timestep splitting (Oran and Boris, 1987).

#### **Convective and Diffusive Transport**

Fluid convection is solved with a high-order implicit algorithm, Barely Implict Correction to Flux-Corrected Transport (BIC-FCT), that was developed by Patnaik *et al.* (1987) to solve convection equations for lowvelocity flows. The Flux-Corrected Transport (FCT) algorithm (Boris and Book, 1976) itself involves an explicit, finite-difference method that is constructed to have fourth-order phase accuracy. However, because FCT is an explicit algorithm, the numerical timestep required for accuracy and stability is limited by the velocity of sound. The basic approach used in BIC-FCT is to treat only those terms whose accuracy and stability are limited by the sound speed (such as the pressure terms in the momentum and energy equations) implicity.

Thermal conduction, molecular diffusion, and viscosity are evaluated using explicit two-dimensional finite differencing (Kaplan *et al.*, 1994 a, b; 1996 a, b). The algorithm for molecular diffusion includes multi-component diffusion among all 16 species tracked, where the binary diffusion coefficients are calculated as a function of temperature for each speciespair from kinetic theory (Hirschfelder *et al.*, 1954). Similarly, the thermal conduction and viscosity coefficients are also calculated from kinetic theory for each individual species as a function of temperature. Using mixture rules (Hirschfelder *et al.*, 1954), an overall mixture thermal conductivity, mixture viscosity coefficients in each computation cell. The viscosity submodel includes all of the viscous terms in the compressible Navier-Stokes equations.

The overall timestep is determined by the convection (BIC-FCT) algorithm. In the diffusive transport submodels, however, numerical stability criteria further restrict the timestep to values less than the overall convective timestep. Therefore, the diffusive transport submodels subcycle several times within the overall convective timestep.

#### **Chemical Reaction**

The reaction mechanism which was incorporated into this axisymmetric diffusion flame code is the "skeletal" mechanism for methane-air combustion (Smooke, 1991), which includes the 16 species. H, O, H<sub>2</sub>, OH, H<sub>2</sub>O, O<sub>2</sub>, HO<sub>2</sub>, H<sub>2</sub>O<sub>2</sub>, CH<sub>4</sub>, CH<sub>3</sub>, CH<sub>3</sub>O, CH<sub>2</sub>O, CHO, CO, CO<sub>2</sub> and N<sub>2</sub>. The set of 35 reactions is shown in Table I. This mechanism has been applied to various test problems for CH<sub>4</sub>-air premixed and diffusion flames, and for studies on the applications of reduced mechanisms (Smooke, 1991, chapters within).

TABLE I "Skeletal" methane-air reaction mechanism (Smooke, 1991)

No.	Reaction	A	B	$-E_a/R$
1	$H+O_2 \rightarrow O+OH$	3.320E-10	0.00	8.456E+03
2	$O + OH \rightarrow H + O_2$	2.606E-11	0.00	3.476E + 02
3	$H_2 + O \rightarrow H + OH$	2.988E-14	1.00	4.442E+03
4	$H + OH \rightarrow H_2 + O$	1.328E-14	1.00	3.403E+03
5	$H_2 + OH \rightarrow H + H_2O$	1.945E-15	1.30	1.825E+03
6	$H + H_2O \rightarrow H_2 + OH$	8.448E-15	1.30	9.355E+03
7	$OH + OH \rightarrow O + H_2O$	9.959E-16	1.30	0.000E + 00
8	$O + H_2O \rightarrow OH + OH$	9.793E-15	1.30	8.571E+03
9	$H + O_2 + M \rightarrow HO_2 + M$	6.336E-30	-0.80	0.000E + 00
10	$H + HO_2 \rightarrow OH + OH$	2.490E-10	0.00	5.052E+02
11	$H + HO_2 \rightarrow H_2 + O_2$	4.419E-11	0.00	3.524E+02
12	$OH + HO_2 \rightarrow H_2O + O_2$	3.320E-11	0.00	5.028E+02
13	$HO_2 + HO_2 \rightarrow H_2O_2 + O_2$	3.320E-12	0.00	0.000E + 00
14	$H_2O_2 + M \rightarrow OH + OH + M$	2.158E-07	0.00	2.290E+04
15	$OH + OH + M \rightarrow H_2O_2 + M$	2.716E-33	0.00	-2.552E+03
16	$H_2O_2 + OH \rightarrow H_2O + HO_2$	1.660E-11	0.00	9.057E+02
17	$H_2O + HO_2 \rightarrow H_2O_2 + OH$	4.747E-11	0.00	1.650E+04
18	$H + OH + M \rightarrow H_2O + M$	6.060E-26	-2.00	0.000E + 00
19	$H + H + M \rightarrow H_2 + M$	4.959E-30	-1.00	0.000E + 00
20	$CO + OH \rightarrow CO_2 + H$	2.506E-17	1.30	-3.813E+02
21	$CO_2 + H \rightarrow CO + OH$	2.606E-15	1.30	1.127E+04
22	$CH_4 \rightarrow CH_3 + H$	2.300E + 38	-7.00	5.756E+04
23	$CH_3 + H \rightarrow CH_4$	3.155E+12	-7.00	4.555E+03
24	$CH_4 + H \rightarrow CH_3 + H_2$	3.651E-20	3.00	4.403E+03
25	$CH_3 + H_2 \rightarrow CH_4 + H$	1.588E-21	3.00	4.403E+03
26	$CH_4 + OH \rightarrow CH_3 + H_2O$	2.656E-18	2.10	1.238E+03
27	$CH_3 + H_2O \rightarrow CH_4 + OH$	5.012E-19	2.10	8.768E+03
28	$CH_3 + O \rightarrow CH_2O + H$	1.129E-10	0.00	0.000E + 00
29	$CH_2O + H \rightarrow CHO + H_2$	4.149E-11	0.00	2.009E+03
30	$CH_2O + OH \rightarrow CHO + H_2O$	4.979E-11	0.00	6.014E+02
31	$\overline{CHO} + H \rightarrow CO + H_2$	6.639E-11	0.00	0.000E + 00
32	$CHO + M \rightarrow CO + H + M$	2.656E-10	0.00	7.398E+03
33	$CH_3 + O_2 \rightarrow CH_3O + O$	1.162E-11	0.00	1.291E+04
34	$CH_3O + H \rightarrow CH_2O + H_2$	3.320E-11	0.00	0.000E + 00
35	$CH_3O + M \rightarrow CH_2O + H + M$	3.983E-11	0.00	1.450E+04

Units are in molecules, cubic centimeters, seconds, K, cal/mole.

Third body efficiencies:  $CH_4 = 6.5$ ,  $H_2O = 6.5$ ,  $CO_2 = 1.5$ ,  $H_2 = 1.0$ , CO = 0.75,  $O_2 = 0.4$ ,  $N_2 = 0.4$ .

#### Soot Formation and Radiation Transport

The formulation of integrated soot models (including soot inception, surface growth, agglomeration and oxidation processes) is an active area of combustion research (Moss *et al.*, 1988; Syed *et al.*, 1990; Stewart *et al.*, 1991; Kennedy *et al.*, 1990, 1991; Villasenor and Kennedy, 1992; Leung *et al.*, 1991; Lindstedt, 1994; Honnery *et al.*, 1992; Kent and Honnery, 1994; Honnery and Kent, 1992). In this work, as in some previous studies (Kaplan *et al.*, 1994b, 1996), we use the integrated soot model of Syed *et al.* (1990), as it is based on local gas properties and is easily incorporated into a multidimensional finite-difference code. In future studies, as we use more detailed chemical reaction models which include the more-relevant soot formation species (such as the  $C_2$  species), we will be able to use more detailed integrated soot models.

The thermophoretic term which appears in the conservation equations (Eqs. 5 and 6) is solved with explicit finite differencing. The source terms in Eqs. (5) and (6) are represented by a set of two coupled ordinary differential equations in terms of the soot number density and the soot volume fraction. These quantities are computed as a function of the local gas properties based upon the simplified rate expressions of Syed *et al.* (1990), which were developed from measurements in a steady  $CH_4$ /air flame:

$$\omega_n = \frac{dn}{dt} = C_{\alpha} N_o \rho^2 T^{1/2} X_{\text{fuel}} e^{-T_{\alpha}/T} - \frac{C_{\beta}}{N_o} T^{1/2} n^2 - \frac{(36\pi)^{1/3} W_{\text{NSC}} n^{4/3}}{\rho_{\text{sout}} f_v^{1/3}} \quad (10)$$

$$\omega_{f_{v}} = \frac{df_{v}}{dt} = \frac{C_{\delta}}{\rho_{\text{soot}}} C_{\alpha} \rho^{2} T^{1/2} X_{\text{fuel}} e^{-T_{\alpha}/T} + \frac{C_{\gamma}}{\rho_{\text{soot}}^{1/3}} \rho T^{1/2} X_{\text{fuel}} e^{-T_{\gamma}/T} n^{1/3} f_{v}^{2/3} - \frac{(36\pi)^{1/3} W_{\text{NSC}} n^{1/3} f_{v}^{2/3}}{\rho_{\text{soot}}},$$
(11)

where the soot particle density,  $\rho_{soot}$ , is assumed to be 1.8 g/cm<sup>3</sup>, and the coefficients and activation temperatures are

$$C_{\alpha} = 6.54 \times 10^{4} \text{ cm}^{3}/g^{2} \cdot \text{K}^{1/2} \cdot \text{s}$$
  

$$C_{\beta} = 1.3 \times 10^{13} \text{ cm}^{3}/\text{K}^{1/2} \cdot \text{s}$$
  

$$C_{\gamma} = 1.0 \times 10^{3} \text{ cm}^{3}/g^{2/3} \cdot \text{K}^{1/2} \cdot \text{s}$$
  

$$C_{\delta} = 1.44 \times 10^{5} \text{ g}$$
  

$$T_{\alpha} = 4.61 \times 10^{4} \text{ K}$$
  

$$T_{\gamma} = 1.26 \times 10^{4} \text{ K}.$$

The first and second terms on the right hand side of Eq. (10) correspond to nucleation and coagulation, while the first and second terms in Eq. (11) represent nucleation and surface growth. The third term on the right-hand side of Eqs. (10) and (11) represents soot oxidation, which includes the Nagle and Strickland-Constable (NSC) (1962) rate for oxidation by  $O_2$  (g/cm<sup>2</sup>·s),

$$W_{\rm NSC} = 12 \left\{ \frac{k_A P_{\rm O_2}}{1 + k_z P_{\rm O_2}} \chi + k_B P_{\rm O_2} (1 - \chi) \right\},\tag{12}$$

where

$$\chi = \frac{1}{1 + \frac{k_T}{k_B P_{0_2}}} \tag{13}$$

and

$$k_A = 20 \exp(-30000/RT) \text{ g/cm}^2 \cdot \text{s} \cdot \text{atm}$$
  
 $k_B = 4.46 \times 10^{-3} \exp(-15200/RT) \text{ g/cm}^2 \cdot \text{s} \cdot \text{atm}$   
 $k_T = 1.51 \times 10^5 \exp(-97000/RT) \text{ g/cm}^2 \cdot \text{s}$   
 $k_Z = 2.13 \exp(4100/RT) 1/\text{atm}.$ 

The other variables and factors in the oxidation terms in Eqs. (10) and (11) are used to convert the NSC oxidation rate into appropriate units for the set of ordinary differential equations, and have been described in detail previously (Kaplan *et al.*, 1996a).

Because methane is a relatively low sooting fuel, the radiation transport sub-model uses an optically-thin assumption,

$$-\nabla \cdot q_r = 4a_{\text{overall}} \sigma \left(T^4 - T^4_{\infty}\right), \tag{14}$$

where  $\sigma$  is the Stefan-Boltzman constant. The absorption coefficient for soot (cm<sup>-1</sup>) is based on an expression from Kent and Honnery (1990), while that for CO<sub>2</sub> and H<sub>2</sub>O is derived from an expression from Magnussen and Hjertager (1977). These are then combined to form an overall Planck mean absorption coefficient:

$$a_{\text{overall}} = 2.66 \cdot \text{const} \cdot f_{\nu} \cdot T + 0.001 (X_{\text{CO}_2} + X_{\text{H}_2\text{O}}).$$
(15)

### **RESULTS AND DISCUSSION**

The flow conditions for the simulations were specifically chosen to reproduce the conditions used in the experiments of Shaddix et al. (1994)

and Shaddix and Smyth (1995). The fuel and air cold flow velocities are both 7.9 cm/s; the fuel tube radius is 0.55 cm and the air annululs radius is 5.1 cm. The full computational domain is extended out to 10 cm radially (and contains quiescent air from 5.1 cm to 10 cm radially), 20 cm axially, and contains  $128 \times 152$  cells. Cells of 0.02 cm  $\times 0.02$  cm are concentrated around the lower and left-hand boundaries (where the flame is located), and the grid is then stretched both radially and axially. The fuel and air are preheated to 550 K and 330 K, respectively, based upon experimental thermocouple measurements for a series of heights just above the burner (Kaplan *et al.*, 1996). The conditions modeled include parabolic pipe flow for the fuel and a flat velocity profile for the coflowing air stream at the inflow boundary, flat thermal profiles for the fuel and air streams, symmetry at the left-hand boundary, freeslip wall at the right-hand boundary, and zero-gradient (simple continuative) outflow condition at the top of the computational domain.

A quantitative comparison is made between the computations and experimental measurements (Shaddix *et al.*, 1994; Shaddix and Smyth, 1995; Kaplan *et al.*, 1996) of the CH<sub>4</sub>-air flame under the same conditions. These comparisons are shown in Figure 1 for temperature, and in Figure 2 for soot volume fraction. Although the computations result in a wider flame thickness at mid-flame heights compared to the experiments (as shown in both the temperature and soot volume fraction comparisons), the overall flame height and soot burnout height are well predicted. Quantitative comparisons of soot volume fraction show that the computations underpredict peak soot volume fraction at the lower and mid-flame heights, but are within 10% of the peak soot volume fraction and the radial location of the soot profiles in general, are well matched between the experiments and the computations.

The remaining results address the universality of temperature and major and minor species as a function of mixture fraction throughout the entire flame. The mixture fraction ( $\xi$ ) at any location can be physically interpreted as the mass fraction of fluid at that location which originated in the fuel stream, and is defined as

$$\xi = \frac{\beta - \beta_2}{\beta_1 - \beta_2} \tag{16}$$

where the subscripts 1 and 2 refer to the fuel and oxidizer streams, respectively, and  $\beta$  is a weighted summation of atomic fractions (Bilger, 1988)

$$\beta = 2\frac{Z_C}{W_C} + 0.5\frac{Z_H}{W_H} - \frac{Z_O}{W_O}.$$
 (17)



FIGURE 1 Comparison between computed and experimentally-measured (Shaddix *et al.*, 1994; Shaddix and Smyth, 1995; Kaplan *et al.*, 1996) radial profiles of temperature at various heights.

Figure 3 shows the mixture fraction and chemical heat release rate for the computed axisymmetric flame. The solid line in the mixture fraction image corresponds to the location of the stoichiometric contour,  $\xi = 0.054$ . As expected, the maximum chemical heat release rate occurs along this stoichiometric contour. The computations also show that there is a thin region of negative chemical heat release along the slightly fuel rich contour, that is for  $0.054 < \xi < 0.15$ . This corresponds to the very light region immediately to the left of the stoichiometric heat release rate. Although there is a negative heat release rate at this location, the local temperature (as shown in Fig. 1) does not decrease due to thermal conduction and convection.

The species concentrations shown below are for the 16 species which are included in the "skeletal" mechanism (Smooke, 1991). The data from the computations are shown at six different heights (1, 2, 3, 4, 5 and 6 cm) in the flame, to illustrate whether or not these species are universal functions of mixture fraction throughout the entire flame, or only in certain locations. The results from the computations are obtained by taking radial traverses at



FIGURE 2 Comparison between computed and experimentally-measured (Shaddix et al., 1994; Shaddix and Smyth, 1995; Kaplan et al., 1996) radial profiles of soot volume fraction at various heights.

the indicated heights and creating scatter plots of the computed quantity (temperature or species mole fraction) versus the mixture fraction at each point along the radial traverse; hence, each scatter plot contains approximately 300 data points. These figures also include experimental data in terms of temperature and species concentrations as a function of mixture fraction taken at one axial location (9 mm) in a rectangular laminar methane-air diffusion flame. The experimental data are taken from Norton *et al.* (1993) and are available for  $0 < \xi < 2$ . Although the experimental



FIGURE 3 Mixture fraction and chemical heat release rate for the computed flame. The solid line corresponds to the stoichiometric mixture fraction.  $\xi = 0.054$ .

configuration (rectangular Wolfhard-Parker burner) is very different than the conditions of our simulations (axisymmetric co-flow burner), one would expect that the universal relationships with mixture fraction would still be valid, regardless of the flame geometry. The legend, shown only in Figure 4, is applicable for all of the scatter plots in Figures 4-12. Each figure below also contains a grayscale image of the quantity in the scatter plot to better understand the flame structure and how that affects universality.

Figure 4 shows images and scatter plots for temperature and CH<sub>4</sub> mole fraction. The scatter plots show that both temperature and CH<sub>4</sub> are strong universal functions of mixture fraction at all six heights. It is noteworthy that the universality is maintained even in the sooting region (slightly fuelrich region) of the flame, where the effects of radiative heat losses are the most significant. In the fuel-lean region, temperatures predicted by the computations are in excellent agreement with those measured experimentally. However, in the fuel-rich region,  $0.054 < \xi < 0.2$ , the computations predict temperatures which are 50-100 K higher than those measured



FIGURE 4 Images and scatter plots of temperature (K) and  $CH_4$  from the computations at six heights in the flame and from the experiments of Norton *et al.* (1993) at one height. The legend shown with Figure 4 is applicable for Figures 4–11.

experimentally. At each of the six heights, the computations show that the temperature peaks at 2100 K at  $\xi = 0.054$ ; the experiments show a peak temperature of 2080 K at  $\xi = 0.0485$  for the one height represented in the experiments. The predicted CH<sub>4</sub> mole fraction is in excellent agreement with



FIGURE 5 Images and scatter plots of  $O_2$  and OH from the computations at six heights in the flame and from the experiments of Norton *et al.* (1993) at one height.

the experimental measurements in the fuel-rich, stoichiometric, and fuel-lean regions.

The scatter plots for O<sub>2</sub> and OH mole fraction (Fig. 5) show excellent universality throughout the entire flame, and show excellent agreement with the experimental data (available for  $0 < \xi < 2$ ). The O<sub>2</sub> plot shows oxygen leakage into the fuel rich region of the flame. The scatter plot for OH shows that OH peaks at  $\xi = 0.048$  at all six heights, and the peak value varies from  $5.4 \times 10^{-3}$  to  $6.2 \times 10^{-3}$  among the different heights. This is in good agreement with the experiments, which show a peak OH mole fraction of  $5.0 \times 10^{-3}$  at  $\xi = 0.0477$  for the one height covered in the experiment. The maximum production rate of OH occurs in a fuel lean region ( $\xi = 0.048$ ), where the heat release rate is moderate ( $\sim 800 \text{ erg/cm}^3$ -s).



FIGURE 6 Images and scatter plots of  $H_2O$  and  $H_2$  from the computations at six heights in the flame and from the experiments of Norton *et al.* (1993) at one height.

Scatter plots for H<sub>2</sub>O and H<sub>2</sub> mole fraction are shown in Figure 6. The computations show that H<sub>2</sub>O peaks at 0.163 to 0.167, for  $\xi$  ranging from 0.054 to 0.058, at the six different heights in the flame. The experiments show a peak H<sub>2</sub>O mole fraction of 0.165 at  $\xi = 0.0577$  at the 9 mm height represented. The predicted H<sub>2</sub>O mole fraction is in good agreement with the experimental measurements in the fuel-rich, stoichiometric, and fuel-lean regions. However, for a given mixture fraction in the fuel rich region, there is ~10-15% scatter in the corresponding H<sub>2</sub>O mole fraction. The maximum production rate of H<sub>2</sub>O occurs in the stoichiometric region, which is the area of maximum heat release, ~ 3000 erg/cm<sup>3</sup>-s.

The computations also show that H<sub>2</sub> peaks at 0.0189 to 0.021, for  $\xi$  ranging from 0.09 to 0.1, while the experiments show a peak H<sub>2</sub> mole



FIGURE 7 Images and scatter plots of  $HO_2$  and  $H_2O_2$  from the computations at six heights in the flame. There are no experimental data for these species.

fraction of 0.0348 at  $\xi = 0.108$ . The computations significantly underpredict the peak H<sub>2</sub> concentration compared to the experimental measurements in the rectangular burner. Although the mechanism was tested extensively and calibrated for laminar premixed and laminar counterflow diffusion flames (Smooke, 1991), it seems to underpredict H<sub>2</sub> concentration in this coflow diffusion flame configuration. The H<sub>2</sub> plot also shows some scatter in the fuel rich region: that is, for a given mixture fraction in the fuel rich region, there is ~ 15% scatter in the corresponding H<sub>2</sub> mole fraction. The fastest reaction rate for the production of H<sub>2</sub> occurs in a fuel-rich region,  $\xi \sim 0.1$ , where the heat release rate is negative, ~ -90 to -100 erg/cm<sup>3</sup>-s.

Figure 7 shows images and scatter plots for  $HO_2$  and  $H_2O_2$ . Considering the relatively small quantities of these species present in the flame, these



FIGURE 8 Images and scatter plots of O and H from the computations at six heights in the flame and from the experiments of Norton *et al.* (1993) at one height.

scatter plots show remarkable universality. The images show that HO<sub>2</sub> and H<sub>2</sub>O<sub>2</sub> appear as an outer envelope around the flame. In the lean region of the flame, HO<sub>2</sub> peaks at  $1.7 \times 10^{-5}$  at  $\xi = 0.02$ , and in the fuel rich region of the flame, HO<sub>2</sub> peaks at  $1.2 \times 10^{-5}$  at  $\xi \sim 0.4$ . There is a very small degree of scatter in the fuel lean region; e.g., HO<sub>2</sub> mole fraction varies from  $1.55 \times 10^{-5}$  to  $1.7 \times 10^{-5}$ , at  $\xi = 0.02$ , depending on height. The fastest production rate of HO<sub>2</sub> occurs in the fuel lean region, where the heat release rate is very small,  $\sim 90$  erg/cm<sup>3</sup>-s.

In the lean region,  $H_2O_2$  peaks at  $5.5 \times 10^{-6}$  at  $\xi = 0.006$ . The peak value varies from  $4.9 \times 10^{-6}$  to  $5.5 \times 10^{-6}$ , and occurs at  $\xi$  ranging from 0.006 to 0.0072. There are no experimental data from Norton *et al.* (1993) for these species for comparative purposes.  $H_2O_2$  is produced in the very fuel lean



FIGURE 9 Images and scatter plots of  $CO_2$  and CO from the computations at six heights in the flame and from the experiments of Norton *et al.* (1993) at one height.

region, where the heat release rate is  $\sim 1-2$  erg/cm<sup>3</sup>-s. There is also significant production of H<sub>2</sub>O<sub>2</sub> and HO<sub>2</sub> at very low heights (1-2 cm) in the fuel rich region, primarily due to the oxygen leakage into that region; however, because these species are only produced at low heights in this fuel rich region, universality at all heights cannot be addressed.

The scatter plots for O and H atoms (Fig. 8) show excellent universality considering the relatively small quantity of these species present in the flame. O peaks at  $\xi = 0.0455$  at all six heights in the flame, and the peak value varies from  $1.28 \times 10^{-3}$  to  $1.34 \times 10^{-3}$  depending upon height. Experimental measurements show a peak value of O at  $2.01 \times 10^{-3}$  for  $\xi = 0.0376$ . The maximum production rate of O atom occurs in the slightly fuel lean region, where the heat release rate is  $\sim 900-1000$  ergs/cm<sup>3</sup>-s. H atom peaks at



FIGURE 10 Images and scatter plots of  $CH_3$  and HCO from the computations at six heights in the flame. Experimental data are available for  $CH_3$  only.

 $\xi = 0.052$  at all heights, and the value of the peak varies from  $1.61 \times 10^{-3}$  to  $1.63 \times 10^{-3}$ . Experiments show a peak value of H at  $2.5 \times 10^{-3}$  for  $\xi = 0.0580$ . The maximum production rate of H atom also occurs in the slightly fuel lean region, where the heat release rate is 1100 - 1200 ergs/cm<sup>3</sup>-s. Although the computations underpredict the peak mole fraction for O and H atom, the location of the peak in the mixture fraction space and the general relationship between these species mole fractions and mixture fraction is well predicted.

Scatter plots for CO<sub>2</sub> and CO are shown in Figure 9. The scatter plot shows that CO<sub>2</sub> mole fraction peaks at  $\xi = 0.0532$  at all six heights in the flame, and peak mole fraction varies from  $8.84 \times 10^{-2}$  to  $8.99 \times 10^{-2}$ . The experimental peak CO<sub>2</sub> is  $8.30 \times 10^{-3}$  and occurs at  $\xi = 0.0709$ . The



FIGURE 11 Images and scatter plots of  $CH_2O$  and  $CH_3O$  from the computations at six heights in the flame. There are no experimental data for these species.

computations show that the maximum production rate of  $CO_2$  occurs in the slightly fuel lean region, where the heat release rate is ~1500-2000 erg/cm<sup>3</sup>-s. The predicted  $CO_2$  mole fraction is in good agreement with the experimental measurements in the stoichiometric and fuel-lean regions, however, the computations slightly overpredict  $CO_2$  in the fuel rich region.

CO mole fraction peaks at values ranging from 0.0306 to 0.0318, and at values of  $\xi$  which vary from 0.0647 to 0.0731, while the experimental peak mole fraction CO is 0.0335 and occurs at  $\xi = 0.108$ . The computations significantly underpredict the peak CO concentration in the fuel rich region, compared to the experimental measurements in the rectangular burner. Again, this skeletal mechanism was tested extensively for laminar premixed and laminar counterflow diffusion flames (Smooke, 1991), but seems to



FIGURE 12 Images and scatter plots of  $N_2$  from the computations at six heights in the flame and from the experiments of Norton *et al.* (1993) at one height.

underpredict CO concentration in this coflowing diffusion flame. The computations show that the maximum production rate of CO occurs in the fuel rich region, where the heat release rate is negative,  $\sim -225$  erg/cm<sup>3</sup>-s.

Figure 10 shows images and scatter plots for HCO and CH<sub>3</sub> mole fraction. Peak HCO mole fraction varies from  $1.03 \times 10^{-6}$  to  $1.13 \times 10^{-6}$ , over values of  $\xi$  ranging from 0.0547 to 0.0538. There are no experimental measurements for HCO for comparative purposes. The maximum production rate of HCO occurs in the stoichiometric region, which is the area of maximum heat release, ~ 3000 erg/cm<sup>3</sup>-s. The computations predict that CH<sub>3</sub> mole fraction peaks at  $7.72 \times 10^{-3}$  to  $8.37 \times 10^{-3}$ , for values of  $\xi$  ranging from 0.0757 to 0.0815. The experiments show that CH<sub>3</sub> peaks at a similar mixture fraction,  $\xi = 0.0820$ , however, the maximum measured mole fraction of CH<sub>3</sub> is  $2.17 \times 10^{-3}$ . The CH<sub>3</sub> plot also shows significant scatter in the fuel rich region. The fastest reaction rate for the production of CH<sub>3</sub> occurs in a fuel-rich region,  $\xi \sim 0.07 - 0.08$ , where the release rate is negative, ~ -150 erg/cm<sup>3</sup>-s.

Images and scatter plots for CH<sub>3</sub>O and CH<sub>2</sub>O are shown in Figure 11. These plots clearly show significant scatter, especially in the fuel rich region. The fastest reaction rate for the production of CH<sub>3</sub>O occurs in the fuel rich region,  $\xi \sim 0.2$ , where the heat release is small,  $\sim 40-50$  erg/cm<sup>3</sup>-s. Similarly, the fastest reaction rate for the production of CH<sub>2</sub>O occurs in an even richer region,  $\xi \sim 0.4$ , where the heat release rate is  $\sim 4-5$  erg/cm<sup>3</sup>-s. The production of these two species is greatest in the area where oxygen has leaked into the fuel rich region. Lastly, Figure 12 shows scatter plot for  $N_2$ , which is considered to be chemically inert in this "skeletal" mechanism. As expected, the computations predict that  $N_2$  is a strong universal function of mixture fraction at all heights in the flame. Predictions from the computations compare very well with the experimental measurements over the fuel lean, rich and stoichiometric regions.

### SUMMARY AND CONCLUSIONS

Direct numerical simulations of an axisymmetric, laminar, methane-air diffusion flame were conducted to study universal relationships of the major and minor chemical species with mixture fraction. The numerical model solves the axisymmetric, time-dependent, reactive-flow Navier-Stokes equations coupled with sub-models for soot formation and radiation transport. The computations include detailed convective and diffusive transport processes, with a 16-species "skeletal" reaction mechanism for methane-air combustion, while soot formation and radiation transport are treated more phenomenologically. Qualitative and quantitative comparisons are made between the simulation results and existing experimental data for a methane-air flame with the same flow configuration. Qualitative comparisons show excellent agreement in overall flame shape, soot burnout height, and peak temperatures and soot volume fractions. More quantitative comparisons between radial profiles of temperature and soot volume fraction at selected heights show a slightly wider computed flame compared to the experimental flame, however, the radial locations of temperature and soot volume fraction line up well with the experimental measurements.

Results from the computations are used to examine the universality of temperature and major and minor species throughout the entire flame (including the sooting region). Scatter plots are made by taking radial traverses at six different heights throughout the flame (covering 1-6 cm height) and plotting the indicated quantity (temperature or species concentration) versus mixture fraction at each computational cell along the radial traverse. The results also include experimental data in terms of temperature and species concentrations as a function of mixture fraction taken at one axial location (9 mm) in a rectangular methane-air diffusion flame (Norton *et al.*, 1993). For CH<sub>4</sub>, O<sub>2</sub>, OH, H<sub>2</sub>O, O, H, CO<sub>2</sub> and N<sub>2</sub>, excellent agreement is obtained between the computations and experimental results in terms of species mole fraction as a function of mixture fraction in the fuel lean, stoichiometric and fuel rich regions of the flame. Likewise,

good agreement is also obtained between the experiments and computations for temperature as a function of mixture fraction in the fuel lean, rich and stoichiometric regions. The computations underpredict the concentration of  $H_2$  and CO in the fuel rich region compared with the experimentally measured values, however, excellent agreement is obtained in the fuel lean and stoichiometric regions. The computations also overpredict the concentration of CH<sub>3</sub> in the stoichiometric and fuel rich regions, however, the peak concentration occurs at the same mixture fraction for both the experiments and computations. Experimental data were not available for species HO<sub>2</sub>, H<sub>2</sub>O<sub>2</sub>, HCO, CH<sub>3</sub>O and CH<sub>2</sub>O.

The scatter plots indicate that many of the species can be considered universal functions of mixture fraction, however, some of the species show more scatter than others. CH<sub>4</sub>, O<sub>2</sub> and N<sub>2</sub> are very strong universal functions of mixture fraction, and showed no scatter. The species which showed very little scatter are those whose production rates are fastest in the fuel lean region (OH, O, H, HO<sub>2</sub> and H<sub>2</sub>O<sub>2</sub>) and in the stoichiometric region (H<sub>2</sub>O, CO<sub>2</sub> and HCO). H<sub>2</sub>O showed more scatter than CO<sub>2</sub> or HCO, but may have been due to the higher degree of scatter in H<sub>2</sub> mole fraction, some of which subsequently converts to H<sub>2</sub>O. Species which showed more scatter are those whose production rates are fastest in the slightly fuel rich regions of the flame  $(H_2, CO \text{ and } CH_3)$  where there is a small negative chemical heat release rate (although not a temperature decrease). Finally, the species which form in the very fuel rich region of the flame (CH<sub>3</sub>O, CH<sub>2</sub>O) cannot be considered universal functions of mixture fraction due to the very high degree of scatter. These species only form in the lower heights of the flame, in the region where oxygen has leaked into the very fuel rich region.

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#### NOMENCLATURE

 $a_{overall}$ overall Planck mean absorption coefficient $C_v$ heat capacity at constant volume

internal energy density
fluid energy density
soot volume fraction
gravitational acceleration
enthalpy of species k
Blotzmann constant
constant in Nagle and Strickland-Constable model
soot number density
species number density for species k
total number density
Avogadro's number
partial pressure of species $k$
pressure
chemical heat release
thermal conductive and radiative heat flux
radial distance
universal gas constant
fluid temperature
temperature of background gas
diffusion velocity of species k
thermophoretic velocity
time
fluid velocity
molecular weight of element k
oxidation rate, Nagle and Strickland-Constable model
mole fraction of species k
mass fraction of species k
axial distance
mass fraction of element k

.

# Greek

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$\beta$	weighted summation of atomic fractions
$C_{\alpha}, C_{\beta}, C_{\gamma}, C_{\delta}, T_{\alpha}, T_{\gamma}$	constants in Syed et al., soot formation model
ξ	mixture fraction
$\xi_{st}$	stoichiometric mixture fraction
ρ	fluid density
$\phi$	equivalence ratio
$ ho_{soot}$	density of a soot particle
$\sigma$	Stefan-Boltzmann constant

- au viscous stress tensor
- v kinematic viscosity
- $\omega_k$  source term for chemical reaction for species k
- $\omega_n$  source term for soot number density
- $\omega_{f_v}$  source term for soot volume fraction

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