Laser Raman Scattering Measurements of Differential Molecular Diffusion in Turbulent Nonpremixed Jet Flames of $\text{H}_2/\text{CO}_2$ Fuel

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ABSTRACT

In this paper we explore the effects of differential diffusion in nonpremixed turbulent jet flames. Pulsed Raman scattering spectroscopy is used to measure temperature and species concentrations in chemically reacting jets of H₂/CO₂ into air, over a range of jet Reynolds numbers from 1,000 to 30,000 based on cold jet fluid properties. Results show significant effects of differential diffusion at all jet Reynolds numbers considered. Differential diffusion between H₂ and CO₂ produces differences between the hydrogen element mixture fraction (ξ_H) and the carbon element mixture fraction (ξ_C). The greatest effects occur on the rich side of stoichiometric, where ξ_H is observed to be smaller than ξ_C at all Reynolds numbers. Differential diffusion between H₂ and H₂O creates a net flux of hydrogen element toward the stoichiometric contour and causes a local maximum in ξ_H that occurs near the stoichiometric condition. A differential diffusion variable \( z_H \) is defined as the difference between ξ_H and ξ_C. The variance of \( z_H \) conditional on ξ_C also shows that differential diffusion effects are greatest on the rich side of the flame. Conditional variances of \( z_H \) are largest at intermediate Reynolds numbers.
INTRODUCTION

Models of turbulent nonpremixed combustion frequently employ the simplifying assumption that all species diffusivities are equal and that the diffusivities of mass and enthalpy are equal (unity Lewis numbers). This leads to the definition of a single conserved scalar, the mixture fraction, that uniquely describes the transport of all species [1]. The effectiveness of the conserved scalar in collapsing the species and energy conservation equations into one general form has led to its use in many current models of turbulent combustion [2-5]. In many flows, however, differential diffusion effects can be important. In flames of hydrogen or hydrocarbons, for example, atomic and molecular hydrogen co-exist with heavier molecules that diffuse more slowly. Differential molecular diffusion occurs, and local species concentrations may differ from those predicted under the equal diffusivity assumption. Consequently, important quantities such as chemical reaction rates, local temperatures, and pollutant concentrations are different than the equal diffusivity theory predicts.

Previous researchers have found clear experimental evidence of differential diffusion in turbulent non-premixed flames. In a low Reynolds number turbulent jet flame of H₂ in air (\(Re = 1,500\)), Drake et al. [6] reported measurements of temperature and concentration which deviated markedly from predictions that assumed equal species diffusivities and unity Lewis number. They attributed these discrepancies to the unequal diffusivities of hydrogen and enthalpy in fuel rich flame zones. Bilger [7] developed a theoretical analysis of differential diffusion in a hydrogen jet flame. By relaxing the unity Lewis number assumption, while retaining the assumptions of chemical equilibrium and adiabaticity, he predicted results that coincide well with the experimental results of Drake et al. [6].

Other researchers have found differential diffusion effects appearing in the form of inconsistencies between alternative definitions of a conserved scalar. Masri, Dibble, and Barlow [8] investigated H₂/CO₂ flames at high Reynolds numbers and reported observable differences in hydrogen element and carbon element mixture fractions. Vranos et al. [9] reported differences between experimental suction probe results presented on the basis of a hydrogen element mixture fraction and those presented on the basis of a carbon element mixture fraction in H₂/CH₄ turbulent jet flames at Reynolds number 10,000. These results indicate that the state of mixing in such flames cannot be uniquely described by a single conserved scalar.
It is frequently argued that at sufficiently high Reynolds numbers the effects of differential diffusion become negligible, and a single conserved scalar may be used without penalty. Bilger and Dibble [10] proposed a model to examine this assumption in nonreacting flows. They solved a turbulent transport equation for a differential diffusion variable $z$, defined as

$$z = \xi_A - \xi_B$$

where $\xi_A$ and $\xi_B$ are the mixture fractions based on species A and B, respectively. A different type of model was developed by Kerstein [11] based on the linear-eddy concept of turbulent mixing. Kerstein [11] pointed out that both models predict that in fully developed turbulent flows the average value of $z$ will scale as $1/Re$, while fluctuations in $z$ will persist to higher Reynolds numbers. Kerstein et al. [12] presented results from a Rayleigh scattering experiment in a turbulent jet of hydrogen and Freon into air, but little other experimental work has been reported to corroborate the theoretical predictions.

The objectives of the present work are to measure the degree of differential diffusion in turbulent jet flames and to establish a relationship between jet Reynolds number and magnitude of differential diffusion effects. A jet mixture of $H_2$ and $CO_2$ was chosen because the molecular weights, and hence the diffusion coefficients, of these species are greatly different. Measurements in nonreacting flows of $H_2/CO_2$ into air have also been made and are reported separately [13].

EXPERIMENTAL METHODS

Experiments were conducted in the Turbulent Diffusion Flame (TDF) laboratory at Sandia National Laboratories. The flow facility and optical diagnostics systems have been described previously [13-16], and only a brief summary of the measurement techniques is included here. Simultaneous point measurements of the concentrations of major species, $N_2$, $O_2$, $H_2$, $H_2O$, $CO_2$, and $CO$, were obtained by spontaneous Raman scattering of laser light from a flashlamp-pumped dye laser (750 mJ/pulse, 532 nm, 3 μs duration). Scattered light was imaged onto the entrance slit of a 3/4 m polychromator and detected using an array of photomultiplier tubes. In the present work, the ideal gas law was used to calculate temperature on each laser shot, based on the total number density of the major species. Temperature dependent Raman calibrations
were based on measurements in the equilibrium products over a nearly adiabatic laminar flat flame burner (Hencken burner).

The experimental jet flame burner was a round tube with an inner diameter of 7.7 mm and with sufficient length to ensure fully developed pipe flow. This tube was centered at the exit of a vertical wind tunnel that supplied coflowing air at 1.5 m/s. Jet exit velocities of 1.7 m/s to 50.6 m/s gave a range of jet Reynolds numbers from 1,000 to 30,000, based on fluid properties at 300 K. Higher jet velocities resulted in flame lift-off. Measurements were made at 15 and 30 nozzle diameters downstream of the jet exit. For all experiments reported here the fuel was a mixture of hydrogen and carbon dioxide with the same overall molecular weight as air (36% H₂ and 64% CO₂ by volume).

Spatial resolution is a critical consideration for investigation of differential diffusion because the smallest scalar length scales in the flame must be resolved. The spatial resolution of the measurement system as configured for these experiments was approximately 740 µm. Estimates of the smallest spatial scales for the present experimental conditions were based on an extension of scaling laws for nonreacting jets [17,18].

In fully turbulent, isothermal jets of unity Schmidt number, the smallest scalar length scale λₖ is given by the relation:

$$\lambda_B = 0.38C_B(z - z_o)Re_d^{-\frac{1}{4}}$$  \hspace{1cm} (2)

where z and z_o are the downstream distance and location of the virtual origin of the jet, respectively, and Re_d is the jet Reynolds number based on the flow parameters at the jet exit. Dowling [17] reported the constant C_B to be approximately 25, while Buch [18] found this constant to be approximately 10. If we conservatively assume that C_B = 10 and we take the virtual origin of the jet to be at the jet exit, then λₖ is 0.19 mm at z/d = 15, in an isothermal jet at Re_d = 30,000. Heat release in the flames causes the turbulence Reynolds number Reₖ to decrease and λₖ to increase. If one assumes that these changes are determined primarily by the temperature dependence of kinematic viscosity (ν ∝ T^{\frac{2}{3}}), then one would expect the local turbulence Reynolds number to decrease as Reₖ ∝ T^{-\frac{2}{3}} and the smallest scalar scale to increase as λ_B ∝ T^{\frac{2}{3}}. For the flame measurements considered here, temperatures are typically at least 1000 K. Therefore, we estimate the smallest scales at z/d = 15 in the Re_d = 30,000 flame to be at least 0.74
RESULTS AND DISCUSSION

We define a mixture fraction \( \xi_X \) based on an atomic element \( X \) as

\[
\xi_X = \frac{Y_X - Y_{X,0}}{Y_{X,f} - Y_{X,o}}
\]

where \( Y_X \) represents the mass fraction of the atomic element \( X \), and the subscripts \( f \) and \( o \) denote mass fractions in the pure fuel and oxidizer streams, respectively. Elemental mixture fractions of the elements nitrogen, oxygen, hydrogen, and carbon were calculated for each laser pulse from the measured number densities of the six major species (\( N_2, O_2, H_2, H_2O, CO_2, \) and \( CO \)). The value of all elemental mixture fractions at stoichiometric would be 0.54 if all species diffusivities were equal.

As an example of the instantaneous data, we present in Fig. 1 a plot of the instantaneous values of \( \xi_C \) and \( \xi_H \) measured at \( x/d = 30 \) in the \( Re_d = 4,000 \) flame. Several features are worth noting. First, it is immediately obvious that \( \xi_C \) and \( \xi_H \) are not equal. In the absence of differential diffusion a plot of \( \xi_C \) vs. \( \xi_H \) would form a diagonal line of unity slope, passing through the origin. We shall refer to this diagonal line as the line of equal mixing. Second, it is evident that the difference between \( \xi_C \) and \( \xi_H \) is greatest in fuel rich fluid, indicating that differential diffusion is greater on the fuel rich side of the flame. Third, a maximum in \( \xi_H \) is apparent near \( \xi_C = 0.6 \). Finally, the data show a range of values of \( \xi_H \) for each value of \( \xi_C \). This "scatter" is partly due to photon shot noise in the Raman signals, but is primarily (and more importantly) due to turbulent fluctuations. Each of these features has physical significance and will be examined in more detail.

Reynolds Number Dependence of \( \xi_H \) vs. \( \xi_C \) curves

As an aid to presentation and examination of the data, and to quantify the effects of differential diffusion at different Reynolds numbers, we shall present the remainder of our results in the form of conditional averages and variances. Statistical quantities were calculated from data points which were binned together according to the measured state of mixedness, defined by the carbon element mixture fraction \( \xi_C \).
Figure 2 shows conditionally averaged values of $\xi_O$ and $\xi_C$ measured at $x/d = 15$, for several values of $Re_d$. Data from several radial locations in each flame are included. The data lie on (or very near) the line of equal mixing at all Reynolds numbers, indicating that differential diffusion is insignificant between the species which contain oxygen and carbon elements ($O_2$, $CO_2$, CO, $H_2O$). Results for $\xi_N$ and $\xi_C$ (not included here) are nearly identical at all Reynolds numbers, and fall even closer to the line of equal mixing. At $x/d = 30$, $\xi_O$, $\xi_N$, and $\xi_C$ are again very nearly equal (results not shown). Therefore, we may conclude that $N_2$, $O_2$, $CO_2$, CO, and $H_2O$ do not differentially diffuse to any significant degree at any Reynolds number.

Figures 3a and 3b plot the conditionally averaged values of $\xi_H$ and $\xi_C$, measured at $x/d = 15$ and $x/d = 30$, respectively. At all Reynolds numbers, differential diffusion between $H_2$ and the carbon species ($CO$ and $CO_2$) creates significant differences between $\xi_H$ and $\xi_C$. Furthermore, there is a Reynolds number dependence of the differential diffusion effect. At $x/d = 15$ (Fig. 3a), for example, the $\xi_H$ vs. $\xi_C$ curves are distinctly different at each Reynolds number. The differences in the curves may be understood by considering the case of a laminar jet flame. In a laminar jet flame the jet fluid becomes diluted with nitrogen and combustion products as it is convected downstream. When differential diffusion occurs, $H_2$ moves away from the jet centerline more rapidly than $CO_2$, and the $\xi_H/\xi_C$ ratio on the jet centerline decreases as the jet fluid is convected downstream. At lower jet exit velocities (lower jet Reynolds numbers) the convection time to a given axial location is larger, and the observed effect of differential diffusion is greater. As the Reynolds number increases, less time is available for $H_2$ and $CO_2$ to differentially diffuse, and the difference between $\xi_H$ and $\xi_C$ decreases, as shown by the curves in Fig. 3a for Reynolds numbers of 1,000, 2,000, 4,000, and 8,000. Figure 3b shows similar behavior for the results obtained at $x/d = 30$. In Fig. 3b the greatest difference between $\xi_H$ and $\xi_C$ occurs at the lowest Reynolds number ($Re_d = 2,000$), and differential diffusion effects become smaller as the Reynolds number increases to 4,000 and then 8,000.

In a turbulent jet, the Reynolds number dependence of the ratio $\xi_H/\xi_C$ cannot be described simply by the ratio of a radial diffusion time to an axial convection time, since radial transport by turbulent convection also occurs. The flame length was observed to be constant and independent of Reynolds number above $Re_d = 8,000$, suggesting the dominance of radial transport by turbulent convection rather than molecular
diffusion at the higher Reynolds numbers. In Figs. 3a and 3b the differences between the $\xi_H$ vs. $\xi_C$ curves are negligible for Reynolds numbers over 8,000. This is also attributed to the dominance of turbulent convection over molecular diffusion for bulk transport of species in the radial direction. It is important to note that the $\xi_H$ vs. $\xi_C$ curves do not lie on the line of equal mixing above $Re_d = 8,000$. Thus, while turbulent convection drives bulk transport in the higher Reynolds number flames, differential diffusion remains important at the smaller scales. In general, turbulent flames of multicomponent fuels may exhibit differential diffusion effects at all Reynolds numbers. This is especially important in flames based on fuels containing $H_2$ and heavier species. Equally important are hydrocarbon fuels which pyrolyze, generating unsaturated hydrocarbons and molecular hydrogen. Should conditions lead to soot formation, large differences between elemental mixture fractions can be expected.

**Effects of Chemical Reaction on Differential Diffusion**

In contrast to the results shown in Figs. 3a and 3b, measurements in nonreacting $H_2/CO_2$ jets [13] showed no significant effect of differential diffusion on average species concentrations for jet Reynolds numbers greater than 1,000. The different behavior of reacting versus nonreacting jets can be attributed in part to the fact that the local turbulent Reynolds number $Re_t$ is much lower in a reacting jet than in a nonreacting jet of the same jet exit Reynolds number. If we assume that $Re_t$ scales as $T^{-\frac{5}{3}}$, then $Re_t$ in an $H_2/CO_2$ jet flame may be ten times smaller than in a nonreacting jet of the same $Re_d$. If this were the only difference between reacting and nonreacting jets, however, one would not expect to observe average differential diffusion effects in flames at a jet Reynolds number of 30,000.

Some important differences between the differential diffusion effects measured in flames and nonreacting jets result from the fact that the rapidly diffusing species $H_2$ is converted to $H_2O$ in the reaction zone. The existence of a sink for $H_2$ at the flame front accentuates differential diffusion of $H_2$ and $CO_2$. $H_2O$ does not diffuse significantly faster than $N_2$, $O_2$, $CO_2$, or $CO$. Therefore, differential diffusion drives up $\xi_H$ in the vicinity of the reaction zone. If the $\xi_H/\xi_C$ ratio in the fuel rich core of the jet remained at its initial fuel stream value of unity, then $\xi_H$ at the flame would be greater than $\xi_C$. This is observed in laminar flame calculations presented later. However, the $\xi_H/\xi_C$ ratio on the centerline of the jet flames is less than unity, and this forces the measured curves of $\xi_H$ vs. $\xi_C$ to move downward.
By the mechanism described above, differential diffusion of \( \text{H}_2 \) and \( \text{H}_2\text{O} \) can cause a local maximum in \( \xi_H \) to occur near the flame. This is evident in Figs. 3a and 3b, where local maxima in \( \xi_H \) near \( \xi_C = 0.6 \) are observed. In Fig. 3a, the data from \( x/d = 15 \) show a local maximum in \( \xi_H \) for Reynolds numbers up to 4,000. At \( x/d = 30 \), a local maximum in \( \xi_H \) is apparent at all Reynolds numbers, as shown in Fig. 3b.

At all Reynolds numbers the difference between \( \xi_H \) and \( \xi_C \) in Figs. 3a and 3b is greatest when \( \xi_C \) is greater than 0.54. Differential diffusion effects are greatest on the fuel rich side of the flame because \( \text{H}_2 \) exists only on the rich side of the flame. Differences between \( \xi_H \) and \( \xi_C \) on the lean side of the flame are due to the fact that differential diffusion on the rich side of the flame affects the value of \( \xi_H \) at the stoichiometric contour.

**Variance of the Differential Diffusion Variable \( Z_H \)**

The conditionally averaged mixture fractions presented in Figs. 2 and 3 are useful for describing the average effects of differential diffusion. Instantaneous effects of differential diffusion are also important and can be quantified by the variance of a differential diffusion variable \( Z_H \) [7], defined as

\[
Z_H = \xi_H - \xi_C
\]

The variable \( Z_H \) takes on non-zero values only when \( \xi_H \) and \( \xi_C \) are unequal, that is, only when differential diffusion of \( \text{H} \) and \( \text{C} \) elements has occurred. The photon shot noise contribution to the variance of \( Z_H \) was determined by the method described in our companion paper [13] and subtracted to yield the variance due to turbulence fluctuations of \( Z_H \).

Figure 4 shows results for \( Z_{H\text{rms}} \), the standard deviation of \( Z_H \) conditional on \( \xi_C \), for \( x/d = 15 \). While average differential diffusion effects were observed in Figs. 3a and 3b to be largest at the smallest Reynolds numbers, Fig. 4 shows that fluctuating differential diffusion effects are smallest at the lowest Reynolds numbers and largest at intermediate Reynolds numbers. Changes in \( Z_{H\text{rms}} \) with increasing Reynolds number reflect the development of the jet (from laminar, to transitional, to turbulent) with increasing Reynolds number. At the lowest Reynolds number in Fig. 4 (\( Re_d = 1,000 \)) the jet is laminar, and fluctuations in \( \xi_H \) at a given value of \( \xi_C \) are small. As the Reynolds number increases the jet goes through transition, and \( Z_{H\text{rms}} \) increases, reaching a maximum at \( Re_d = 8,000 \), both at \( x/d = 15 \) and at \( x/d = 30 \) (results not shown). At
Reynolds numbers greater than 8,000 the flow is turbulent, as indicated by the Reynolds number invariance of the flame length and by the similarity of the $\xi_H$ vs. $\xi_C$ curves in Figs. 3a and 3b. $z_{HRMS}$ becomes smaller as the Reynolds number rises above 8,000.

Figure 4 shows that $z_{HRMS}$ is largest on the fuel rich side of the flame ($\xi_C > 0.54$). The fact that fluctuations in differential diffusion effects are largest on the rich side of the flame is consistent with our earlier observation that average differential diffusion effects are largest on the rich side of the flame, and may be attributed to the fact that the highly diffusive species $H_2$ exists only in fuel rich mixtures.

Comparison to Laminar Flame Calculations

Flamelet models of nonpremixed turbulent combustion treat a reacting flow as an ensemble of strained laminar flames. Therefore, it is of interest to compare differential diffusion effects measured in jet flames to those predicted for laminar opposed flow flames. The opposed flow flame results were calculated using the Chemkin [19] based numerical model of the Tsuji geometry [20] described by Miller et al. [21]. The Sandia transport property code [22] was used to account for diffusion of all species, using the full multicomponent formulation for transport properties.

Numerical results for strain rates of 50 s$^{-1}$, 500 s$^{-1}$, and 4500 s$^{-1}$, applied to flows with 36% $H_2$ and 64% $CO_2$, are presented in Fig. 5 as plots of $\xi_H$ vs. $\xi_C$. Flame extinction occurs at a strain rate of 5000 s$^{-1}$. Conditionally averaged values of $\xi_H$ and $\xi_C$ from measurements at $x/d = 30$ in the $Re_d = 8,000$ jet flame are also included in Fig. 5. The results show that $\xi_H$ is much lower in the jet flames than in the calculated laminar flames. This is because differential diffusion of $H_2$ and $CO_2$ causes the ratio $\xi_H/\xi_C$ on the jet centerline to decrease with downstream distance, as discussed earlier. Consequently, the constant fuel stream boundary condition for the laminar flame calculations is inconsistent with the changing centerline condition in the jet flame. The comparison between measurement and prediction is much improved when the fuel side boundary condition for the flamelet calculations are adjusted to be consistent with measured centerline species concentrations. This is shown by the dotted curve in Fig. 5. Because the $\xi_H/\xi_C$ ratio decreases along the jet centerline, thermochemical states in these jet flames cannot be represented by an ensemble of strained laminar flame calculations that use a fixed $H_2/CO_2$ ratio for the fuel side boundary condition. Moss and Roberts [23] commented on the boundary condition problem in laminar flamelet modeling of
turbulent flames, pointing out that changing flamelet boundary conditions may result in varying flamelet species profiles. More recently, Bish et al. [24] addressed the boundary condition issue by formulating a model of turbulent nonpremixed combustion which allows local boundary conditions for both the fuel rich and fuel lean sides of strained dissipation layers to vary throughout the flow.

In our companion paper [13], we showed that differential diffusion effects are independent of strain rate in nonreacting laminar opposed flows. Laminar opposed flow flame calculations with very fast chemical reaction rates (near equilibrium chemistry) also show that differential diffusion effects are independent of strain rate. When chemical reactions occur at finite rates, however, the effects of differential diffusion on relative elemental mixture fractions vary with strain rate, as shown in Fig. 5. Between the two extremes of nonreacting flow and equilibrium chemistry there is a continuum of solutions to the species and energy conservation equations. The ratio of chemical reaction rate to strain rate (a Damköhler number) is the dimensionless parameter which determines whether the solutions lie closer to the nonreacting or the equilibrium limit.

CONCLUSIONS

Raman scattering measurements of major species concentrations were used to investigate the effects of differential molecular diffusion in jet flames of H\textsubscript{2}/CO\textsubscript{2} in air, for Reynolds numbers from 1,000 to 30,000. These measurements lead to the following conclusions:

1. Differential diffusion between H\textsubscript{2} and CO\textsubscript{2} creates differences between conditionally averaged values of the elemental mixture fractions ξ\textsubscript{H} and ξ\textsubscript{C} for all Reynolds numbers and downstream locations.

2. Differential diffusion between H\textsubscript{2} and H\textsubscript{2}O creates local maxima in ξ\textsubscript{H}, at x/d = 30 for all Reynolds numbers, and at x/d = 15 for jet Reynolds numbers of 1,000, 2,000, and 4,000.

3. Average and fluctuating effects of differential diffusion are greatest on the fuel rich side of the flame, where the highly diffusive species H\textsubscript{2} exists.

4. Differential diffusion of H\textsubscript{2} and CO\textsubscript{2} causes the ratio ξ\textsubscript{H}/ξ\textsubscript{C} on the jet centerline to decrease with downstream distance. This effect should be considered whenever fuel mixtures include gases with widely different molecular weights, particularly combinations of H\textsubscript{2} with heavier
species. Furthermore, because the $\xi_H/\xi_C$ ratio decreases along the jet centerline, thermochemical states in these jet flames cannot be represented by an ensemble of strained laminar flame calculations that use a fixed $H_2/CO_2$ ratio for the fuel side boundary condition.

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FIGURE CAPTIONS

Figure 1. Scatter plot of $\xi_H$ vs. $\xi_C$ for instantaneous data obtained at $\xi = 30$ at $Re_d = 4,000$. Each point is a single laser shot.

Figure 2. Comparison of conditionally averaged values of $\xi_O$ and $\xi_C$ for data obtained at $x/d = 15$ over a range of Reynolds numbers from 1,000 to 30,000. The diagonal line is the line of equal mixing.

Figure 3. Comparison of conditionally averaged values of $\xi_H$ and $\xi_C$, for data obtained at: (a) $x/d = 15$ over a range of Reynolds numbers from 1,000 to 30,000 and (b) $x/d = 30$ over a range of Reynolds numbers from 2,000 to 30,000. The diagonal line is the line of equal mixing.

Figure 4. Standard deviations of $z_H$ conditional on $\xi_C$ for data obtained at $x/d = 15$ over a range of Reynolds numbers from 1,000 to 30,000.

Figure 5. Comparison of $\xi_H$ vs. $\xi_C$ curves from calculated laminar opposed flow flames and from experimental jet flames. Measurements are from $Re_d = 8,000$ at $\xi = 30$. Calculations are for three different strain rates ($50 \text{ s}^{-1}$, $500 \text{ s}^{-1}$, and $4500 \text{ s}^{-1}$) with a fuel stream boundary condition of 36% $\text{H}_2$ and 64% $\text{CO}_2$, and for one strain rate ($10 \text{ s}^{-1}$) with a fuel stream boundary condition taken from measurements on the jet centerline.
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