Time-Resolved Density Measurements in Premixed Turbulent Flames

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Results of density measurements by Rayleigh scattering from an unconfined, V-shaped, methane-air flame stabilized in grid turbulence are reported and discussed. Data for mean and root-mean-square (rms) density, power spectra, and for probability density functions (pdf's) are presented along with velocity data obtained by laser velocimetry and hot-wire anemometry. Profiles of mean density and rms fluctautions show that the flame thickens with distance from the flame stabilizer but there is no significant change in rms fluctuation magnitude with distance. The pdf's are bimodal but cannot be represented adequately by two delta functions. Normalized spectra do not vary significantly across the flame and exhibit similarities to velocity spectra in the reactant flow.

Introduction

PREMIXED, turbulent flames have received considerable attention, both theoretical and experimental, in recent years and significant advances in our understanding of these flames have been made; see, for example, Refs. 1-3. Such flames are of intrinsic interest to those studying fundamental, turbulent reacting flow processes and to those interested in practical combustion devices, such as spark ignition engines and premixed, prevaporized gas turbine combustors, which burn premixed reactants.

In experiments, schlieren and shadowgraph techniques have been used for qualitative measurements and for flow visualization.[4,5] Laser velocimetry (LV) is being used currently by a number of investigators (e.g., Refs. 5-8) for velocity measurements in and around the turbulent reaction zones of these flames. However, because of various experimental difficulties, measurements of scalar quantities such as temperature and density have lagged behind other measurements. In this paper, we report the application of laser induced Rayleigh scattering to measure the gas density in premixed, methane-air flames.

Under reasonable assumptions[9]—i.e., one-step irreversible chemistry, premixed reactants, constant heat capacity, equal diffusivities and unit Lewis number—the instantaneous thermodynamic state of a reacting mixture can be related to either its temperature, density, or to another appropriate scalar. For many purposes, the preceding assumptions are justified, and, thereby, considerable simplification in experiment and theory is achieved. Certainly, time and space resolved measurements of one of these scalars can provide valuable information about turbulent flame structure and flame processes.

A number of investigators have attempted instantaneous temperature measurements using fine-wire sensors—either thermocouples or hot wires (e.g., Refs. 10-13). However, these techniques are subject to errors which are difficult to reduce or eliminate and whose magnitudes have not been fully accessed. Thermal inertia due to the finite heat capacity of the sensor limits its frequency response, and compensation techniques similar to those used in constant current hot-wire velocity measurements frequently are used to extend system response to higher frequencies. For compensation it is assumed[14] that the sensor's thermal response to temperature fluctuations is given by $d\theta/dt = (\Theta - \theta)/T$, where θ is the sensor temperature and Θ the gas temperature. T is the characteristics sensor response time, which for compensation is assumed to be a constant at each position in the flame. In reality, T is a function of gas temperature and velocity[15] and for large fluctuations in either quantity it cannot be assumed constant. Perhaps symptomatic of this variability is the observation by Yoshida and Tsuji[12] in their work that time constants determined by the step response method are unrealistic while plausible results are obtained by selecting time constants which give reasonable temperature pdf's. Overall, compensation appears to be a limited technique for temperature measurements in turbulent combustion systems, and more study is required to determine the conditions under which it can give acceptable results.

For measurements in reacting gases, the perturbing effects of catalytic reactions on thermocouple and hot-wire response must be considered. Smith and Gouldin[16] and Smith[17] report the results of temperature measurements with coated (SiO_2) and uncoated, compensated thermocouples in premixed, methane-air flames. They observe, for measurements in the hotter portion of the flame, significant differences in mean and rms temperatures between the two cases. Furthermore, they find that the coatings are easily cracked by thermal stresses and frequent recoating is necessary. Ballantyne and Moss[14] report a similar comparison for thermocouple measurements of mean temperature in a diffusion flame; they find no significant differences between results obtained with coated and uncoated wires. Apparently, the type of flame is important. Coatings increase the emissivity and, therefore, the radiant heat loss of fine-wire probes further complicating the measurements. Clearly probe techniques are limited and subject to unknown error. Alternate measurement techniques are desired.

Recently marker nephelometry[18] has been used to measure gas density in premixed flames[19] and in nonpremixed flames.[20] Although this technique is not new, its application to combustion has been limited. There are several difficulties

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in the technique associated with the fact that the measurement is indirect, i.e., marker particle density rather than gas density is measured by light scattering. For valid measurements, the marker particles must be added uniformily to the cold reactants; particle concentration must be high enough to reduce marker shot noise to acceptable levels; particle scattering properties must be constant; the particles must move with the gas; and their local number density must vary as the gas density varies which requires, among other things, that diffusophoresis and thermophoresis be negligible. These requirements generally are recognized,‡ but quantitative measures of the errors arising when they are not met have not been developed. In any case, it is doubtful that these errors are always negligible, and we expect the technique will find limited application in reacting flows.

A number of laser scattering and laser induced fluorecence techniques for finding gas temperature and density are under development.[21,22] These can provide good temporal and spatial resolution, but they are complex and difficult to apply. In certain special cases where the mixture scattering cross section is known, Rayleigh[23] scattering can be used to find molecular number density ρ. For example, in premixed flames the mixture scattering cross section does not vary appreciably with the progress of reaction and can be determined by measurements on the cold, reactant mixture. Dibble and Hollenbach[24] and Bill et al.[7,25,26] have used Rayleigh scattering for density measurements in premixed flames and in a reacting boundary layer. In nonpremixed systems Rayleigh scattering can still be used with relative ease if the separate flow streams entering the test section have equal scattering cross sections. Dibble and Hollenbach[24] use a novel fuel mixture to achieve this balance for a jet diffusion flame in air.

In addition to point measurements, instantaneous density profile measurements are possible with Rayleigh scattering by imaging scattered light from a segment (e.g., 1 cm) of a high-energy, pulsed laser beam onto an optical array detector such as a vidicon. Such measurements are reported for spark ignition engines[27] and have been performed in premixed turbulent flames.[28]§ Where applicable (the cross section is known), Rayleigh scattering appears to be a very attractive technique for density and temperature measurements.

Previously, the authors have reported on velocity and flame speed measurements in open, premixed turbulent flames of methane-air, propane-air, and ethylene-air.[8] In this paper, we report the results of Rayleigh scattering measurements in a lean, methane-air flame on the same burner. Data on mean density and rms density fluctuations, on pdf's of density, and on density fluctuation power spectra are reported and discussed along with velocity data to draw conclusions concerning the flame structure and important flame processes.

Experiment

Rayleigh Scattering

For right angle scattering from a length ℓ of a laser beam the scattered power P_s as detected by a photomultiplier tube is given by

$$P_s = P_L \ell\, \Omega \eta \sigma_m \rho \tag{1}$$

where P_L is the laser power, Ω the solid angle of the collection optics, η the product of the transmission efficiency of the optical system and the quantum efficiency of the photomultiplier tube, and σ_m the scattering cross section of the gas mixture. All of these quantities with the exception of σ_m and ρ are fixed by the experimental apparatus, and their product is a constant which can be found by calibration. The mixture scattering cross section may be expressed in terms of the individual species cross section σ_α and the species mole fraction X_α; $\sigma_m = \Sigma \sigma_\alpha X_\alpha$, where the sum is over all chemical species present.

For premixed flames σ_m changes very little during combustion; this is especially true for lean methane-air mixtures. Using refractive index data[29] to find the σ_α[30] and considering only major species (N_2, O_2, CH_4, CO_2, and H_2O), we find that for a lean (equivalence ratio, $\phi = 0.8$) methane-air mixture σ_m decreases by about 2% when products are compared to reactants ($7.414 \cdot 10^{-28}$ cm^2/sr vs $7.56 \cdot 10^{-28}$ cm^2/sr). Larger variations in σ_m occur in richer CH_4-air flames and for other fuel-air mixture combinations.[26,28] However, for lean methane-air mixtures, σ_m variation with reaction can be neglected and a measurement of P_s gives ρ to good accuracy.

A 1.0 W continuous wave laser is used for these experiments, and the corresponding scattered power is low. Detection is by photomultiplier tube and photon counting. To achieve a reasonable frequency response the counting period is short, 200 μs, which provides a data rate of 5 kHz. Counting rates are low (0.25-1.7 MHz), and shot noise is systematically removed during data analyses for estimates of mean, rms, and spectral density.

The raw data recorded at a point in the flame consist of a set of photon counts taken over counting periods Δt (200 μs) in length at equally spaced times t_i—$\{n(t_i,\Delta t)\}$. $n(t_i,\Delta t)/K\Delta t$ is the average molecular number density during the period $t_i - \Delta t$ to t_i plus a random shot noise contribution. $K = (P_L \Omega \ell \eta \sigma_m)$ and is found by calibration in the cold, reactant flow. For a Poisson distribution of shot noise, the results of Penney et al.[31] and Birch et al.[32] can be used to show[33] that

$$\bar{\rho} = \bar{n}/K\Delta t \tag{2}$$

$$\mathrm{Var}(\rho) = [\mathrm{Var}(n) - E(n)]/(K\Delta t)^2 \tag{3}$$

$$R(\tau) = \overline{n(t,\Delta t)\, n(t+\tau,\Delta t)}/\overline{(K^2 \rho''^2)} - \bar{\rho}^2/\overline{\rho''^2} \tag{4}$$

$$S(f) = 4\int_0^\infty \overline{\rho''^2} R(\tau) \cos 2\pi f \tau \, \mathrm{d}\tau \tag{5}$$

$E(n)$ is the expected value of $n(t_i,\Delta t)$. Overbars denote time mean quantities, while ρ'' is the fluctuating component of ρ. $R(\tau)$ is the autocorrelation coefficient of ρ, and $S(f)$ the corresponding frequency spectrum. The density pdf $P(\rho)$ is related to the pdf for n by the following.

$$P(n) = 1/n! \int_0^\infty \overline{\rho^2} (\rho K \Delta t)^n \exp(-\rho K \Delta t) P(\rho)\, \mathrm{d}\rho \tag{6}$$

This integral equation is solved by assuming a histogram form for $P(\rho)$ and then by adjusting the height of each step in the histogram so that the $P(n)$ obtained from Eq. (6) gives the best, least-squared fit to the measured data; see Ref. 33 for details.

Apparatus

Figure 1 shows schematically the flame configuration used in this study. It is an unconfirmed V shaped flame, stabilized in grid turbulence on a rod (1.25-mm diam) mounted across

‡Becker in his extensive review[19] mentions diffusophoresis but not thermophoresis. In flows where temperature gradients are perpendicular to time-mean streamlines, we expect thermophoresis to be a significant problem.

§Preliminary instantaneous density profile measurements were performed on the burner described in this paper at the United Technologies Research Laboratories, East Hartford, Conn. in July 1981. A frequency doubled Nd:YAG laser was the scattering source, and signal detection was with an optical multichannel analyzer coupled to a minicomputer. The results support the wrinkled laminar flame model and demonstrate the feasibility of instantaneous density profile measurements by Rayleigh scattering.

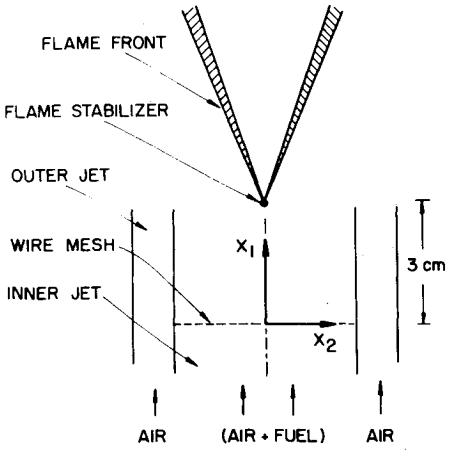

Fig. 1 Schematic of turbulent flame and burner showing location of wire mesh, stabilizer rod, and coordinate system. The shaded area indicates the turbulent flame brush.

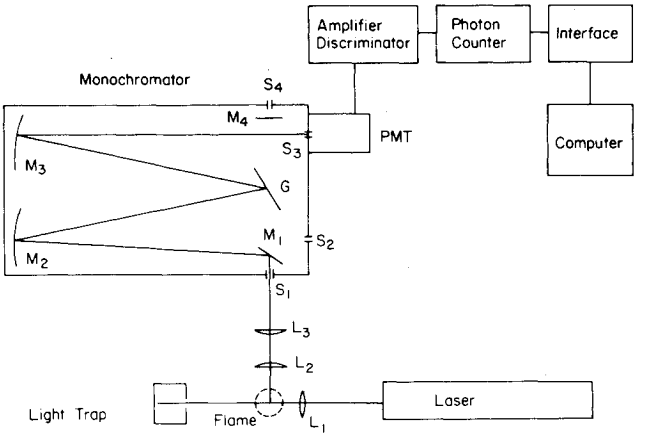

Fig. 2 Schematic of optical configuration and detection electronics for the Rayleigh scattering system. These components are held fixed, while the burner is moved for measurements at different x_1 and x_2. The letters G, M, and S denote the grating, mirrors, and slits of the monochromator; L denotes a lens.

the exit plane of a cylindrical burner (50-mm diam) and is similar to one studied previously.[8,16,17,34] Turbulence is generated by an 8 mesh, woven-wire screen (0.813-mm wire diam) mounted in the central air-fuel jet, 3.0 cm from its exit. An annular air jet (7.6-cm diam) surrounds the central air-fuel jet. The shaded region in Fig. 1 indicates the region of the time-mean turbulent reaction zone or flame brush.

A schematic of the optical apparatus used for the Rayleigh scattering measurements is shown in Fig. 2. Radiation from an argon-ion laser is focused by a biconvex lens L_1 of 200-mm focal length to a waist of approximately 0.1-mm diam at the measurement volume. A plano-convex lens L_2 of 127-mm focal length ($f/4$) collects and collimates scattered radiation from the measurement volume. In turn, another plano-convex lens L_3 images the measurement volume on the entrance slit S_1 of a monochromator filling its acceptance cone. The monochromator is equipped with a 2000 groves/mm holographic grating and provides a dispersion of 4.8 Å/mm at 2200 Å. Signal detection is by photomultiplier tube and photon counting. Count rates over a magahertz are encountered in the cold reactants. The resulting pulse pile-up error is estimated to be 1.7% and tends to cancel the error due to σ_m variation with reaction. The count data $[n(t_i,\Delta t)]$ are stored on computer for later analyses.

With this optical configuration, the measurement volume from which scattered radiation is observed has a cylindrical shape with a diameter of approximately 0.1 mm and a length of approximately 0.4 mm in a direction parallel to the flame holder. As noted earlier the data rate from the photon counter is 5.0 kHz. This temporal resolution with a flow velocity of 4.0 m/s corresponds to a spatial resolution of 0.8 mm. For comparison, the laminar flame thickness (δ) of a methane-air mixture at standard conditions and $\phi=0.8$ is 1.5 mm (Ref. 35). Density fluctuations in the flame cause random stearing of the laser beam. This effect was found to be unimportant in laser anemometry measurements[33] and is felt to be unimportant in the present experiments.

Mie scattering from particles and Rayleigh scattering from molecules are both elastic processes, and there is no shift of frequency for the scattered radiation. Since particle scattering is much stronger than molecular scattering, the former will obscure the latter when particles are present in the scattering volume. To reduce particle scattering to a negligible level the flow apparatus, which had been used previously in laser velocimetry measurements, was dismantled and carefully cleaned. Also, the gaseous flows of fuel and air are filtered to remove particles before entering the measurement region.

Measurements are made across the time-mean reaction zone for different lateral positions x_2 at two axial stations, $x_1=5.5$ and 6.5 cm (the coordinate system is defined in Fig. 1). The initial reactant flow velocity is 3.8 m/s, parallel the x_1 axis. Turbulence intensities and integral length scales (ℓ) at the two axial stations as measured in the cold, reactant flow by hot-wire anemometry[33] are 3.5 and 3.2% for intensity and 2.48 and 2.64 mm for integral scale at 5:5 and 6.5 cm. For these conditions, the turbulence Reynolds number ($R_\ell=u'\ell/\nu$) is approximately 30.

For low turbulence Reynolds number and low turbulence intensity, as is the case in these experiments, a wrinkled laminar flame structure is expected. The criterion for such flames recommended by Andrews et al.,[36] namely, $R_\ell<100$, is clearly satisfied. Furthermore, the data reported here and preliminary instantaneous density profile measurements made on this burner indicate a wrinkled flame structure. However, the Klimov-Williams criterion, expressed as $\delta/\ell_k<1$ where ℓ_k is the Kolomogrov scale in the reactant flow, it not satisfied which is surprising since it has a firm theoretical basis. For $\delta=1.5$ mm and $\ell_k=0.2$ mm (based on standard estimates for grid turbulence, $\ell_k=\ell/R_\ell^{3/4}$) the criterion is violated to a considerable degree.

A review of Williams' discussion of the criterion in Ref. 37 reveals certain quantitative problems with the δ/ℓ_k expression. The Klimov-Williams criterion is derived from a consideration of the influence of velocity field strain on the laminar flame structure. Under the assumption of constant gas properties a dimensionless group of the form $\Phi=\gamma\bar{\nu}/S_L$ appears in the governing conservation equations. $\bar{\nu}$ is an appropriate average value for ν in the flame and γ is a measure of velocity strain induced on the flame presumably by the reactant, as opposed to the product, flowfield. For $\Phi>1$ there is significant straining and, therefore, distortion of the basic, unstrained laminar flame structure and a wrinkled laminar flame cannot exist.

γ is estimated as $u'/\lambda\sim R_\ell^{3/2}\,\nu_0/\ell^2$ where u' is the turbulence intensity in the cold flow, λ the Taylor microscale, and ν_0 the kinematic viscosity in the cold flow, while δ is estimated as $\bar{\nu}/S_L$ ($\bar{\nu}/S_L$ is a diffusive length scale and is perhaps a factor of 2 or 3 smaller than δ as measured). Noting that $\ell_k\simeq\ell/R_\ell^{3/4}$, one obtains after manipulation $\Phi\simeq(\nu_0/\bar{\nu})\,(\delta/\ell_k)^2$. Normally, the distinction between ν_0 and $\bar{\nu}$ is ignored and the δ/ℓ_k form of the criterion is obtained. This step makes no qualitative difference, but obviously it is quantitatively incorrect. An estimate of $\bar{\nu}$ may be obtained from the measured value of flame thickness δ_m, e.g., $\delta_m=3\,\bar{\nu}/S_L$. Using $\delta=1.5$ mm one obtains $\bar{\nu}=1.5$ cm^2/s which is over an order of magnitude greater than ν_0. Based on the preceding discussion, we suggest a revised form for the

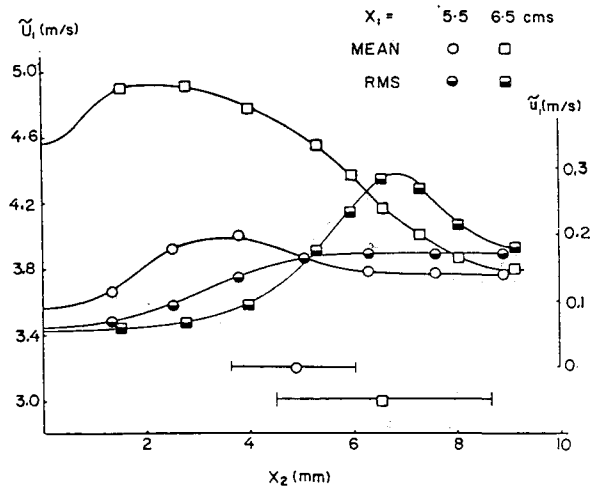

Fig. 3 Axial velocity measurements, Favre mean ($\tilde{U}_1$) and Favre rms ($\tilde{u}_1$) at two axial stations for a lean methane-air flame ($\phi = 0.8$) and an initial mean velocity of 4.0 m/s. The horizontal lines indicate the mean flame thickness determined from thermocouple mean temperature measurements.[33]

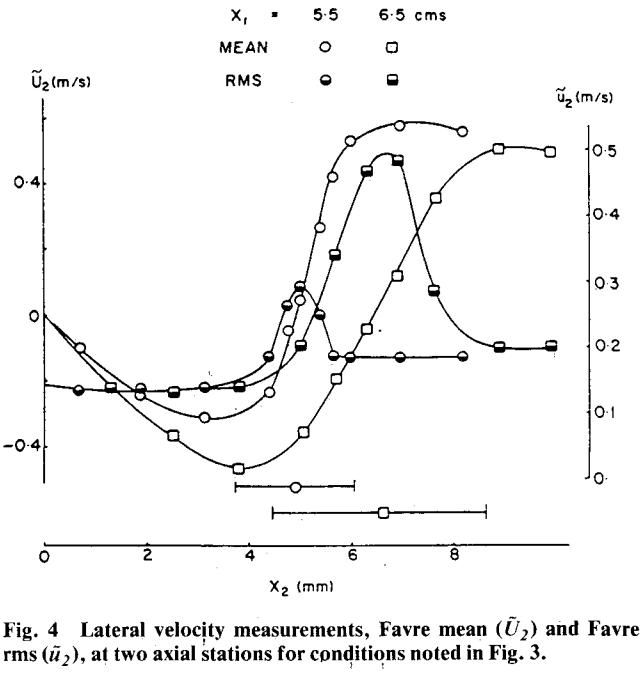

Fig. 4 Lateral velocity measurements, Favre mean ($\tilde{U}_2$) and Favre rms ($\tilde{u}_2$), at two axial stations for conditions noted in Fig. 3.

Kilmov-Williams criterion which should be more useful quantitatively than the present one

$$\Phi' = \tfrac{1}{3}\sqrt{(v_0/\bar{v})}\,\delta_m/\ell_k < 1$$

For the flames studied in the present work $\Phi' = 0.74$. The revised criterion is marginally satisfied.

Results and Discussion

Measurements were made in a single methane-air flame ($\phi = 0.8$) at two axial stations. Time series records of scattered light intensity were obtained at different points across the reaction zone at these stations, and the raw data were analyzed to determine mean densities $\bar{\rho}$, rms density fluctuations ρ', density pdf's, autocorrelations, and power spectra. In addition, at the same axial stations but during separate runs, profiles of axial and lateral velocity were measured by laser velocimetry. Velocity fluctuation power spectra characterizing the reactant flow were obtained by hot-wire anemometry measurements in an air flow, and turbulent flame speeds were measured according to the technique of Ref. 8, for details see Ref. 33.

A burst type LV processor was used for the velocity measurements, and the seed level was low. Therefore, velocity bias and the density bias are problems. Because the turbulence kinetic energy is low, velocity bias is not significant. However, the axial velocity data are corrected according to McLaughlin and Tiederman.[38] Density bias is not removed. For uniform seeding and small particles, the same arguments used to support marker nephelometry for density measurements lead one to conclude that density bias gives Favre averaged results. Therefore, both mean and rms velocities are assumed to be Favre averaged and denoted with a tilde.

Axial mean and rms velocity profiles across the flame are presented in Fig. 3, and the corresponding lateral velocity profiles are presented in Fig. 4. For both velocity components, mean and rms values are uniform in the reactants, while in the reaction zone the rms is seen to increase markedly at $x_1 = 6.5$ cm and to a lesser extent at $x_1 = 5.5$ cm. All data indicate that the flame brush thickens substantially with increasing x_1. Similar variations in velocity and flame thickness have been reported by Bill et al.[7] and Smith and Gouldin,[16] respectively. The increase in rms velocity in the reaction zone evident in Figs. 3 and 4 are attributed by Dandekar and Gouldin[8] to combustion induced mean flow

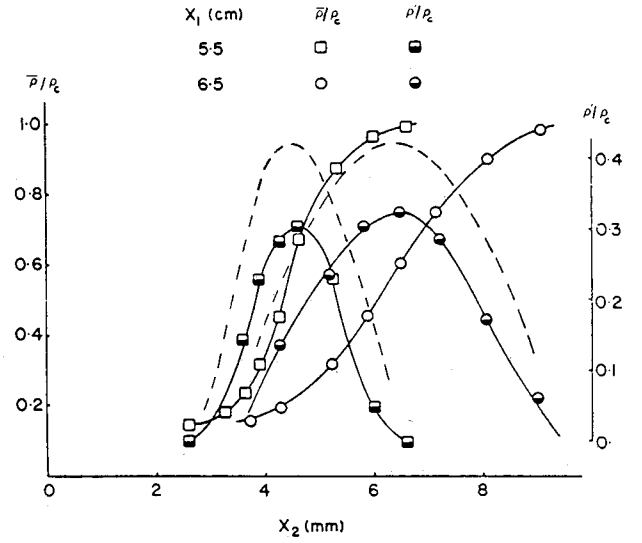

Fig. 5 Profiles of mean and rms number density normalized by the cold mixture density obtained with Rayleigh scattering at two axial stations as noted. The dashed curves are estimates of ρ'/ρ_c obtained from $\bar{\rho}/\rho_c$ by assuming a two delta function pdf form ($\gamma = 0$).

shear and to interactions between fluctuations in velocity, density, and the pressure gradient. The significance of mean flow shear is evident in Figs. 3 and 4. Both $\partial\tilde{U}_1/\partial x_2$ and $\partial\tilde{U}_2/\partial x_1$ and, therefore, the mean rate of strain $[1/2(\partial\tilde{U}_1/\partial x_2 + \partial\tilde{U}_2/\partial x_1)]$ are negative, not zero in the reaction zone. The relative importance of these terms to turbulence generation in our flames is at present unknown.

Mean and rms density profiles for the two axial measurement stations are presented in Fig. 5. A thickening of the flame with downstream distance is quite evident in these data as it was in the velocity data. Note, however, that there is no increase in ρ' with downstream distance and that the maximum ρ' occurs at about the same $\bar{\rho}$ at the two stations.

Density pdf estimates in the form of histograms derived from raw photon count data obtained at several horizontal locations which are representative of different regions in the flame ($\xi = 0.32$, 0.62, and 0.84) are presented in Figs. 7 and 8; $\xi \equiv \rho/\rho_c$. The pdf's are distinctly bimodel as hypothesized in

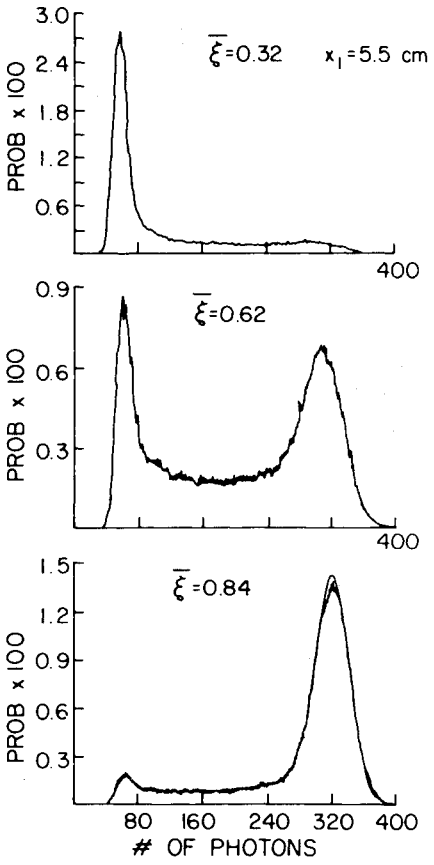

Fig. 6 The pdf's of photon counts, $n(t_i, \Delta t)$, as measured at three lateral locations characteristic of the cold and hot sides of the flame and of the middle of the flame. These are raw data and contain significant shot noise. The smooth curve for $\bar{\xi}=0.84$ is obtained from the histogram at $\bar{\xi}=0.84$ (see Fig. 7) by Eq. (6) to demonstrate the goodness of the solution for $P(\rho)$ from Eq. (6).

most current turbulent flame models including wrinkled laminar flame models and as implied by the temperature data of other investigators (e.g., Refs. 7, 12, 39, and 40).

Although the histogams appear crude, they give good fits through Eq. (6) to the experimental data for $P(n)$ as can be seen in Fig. 6. Overall, the accuracy of the $P(\rho)$ results is determined by the spatial and temporal resolution of the measurement systems and the accuracy of our solution to Eq. (6). Although we have not performed a detailed sensitivity analysis, we believe that the temporal resolution is the limiting factor.

Bray and Moss[39] suggest a pdf form for premixed flames: $P(\xi)=\alpha\delta(\xi-\xi_H)+\beta\delta(\xi-\xi_C)+\gamma f(\xi)\,[H(\xi-\xi_H)-H(\xi-\xi_C)]$,¶ where δ and H are the delta and Heaviside functions, and α, β and γ are functions of position and must sum to one. $f(\xi)$ represents the variation of the pdf between the peaks contributed by the cold reactants and hot products. With $f(\xi)$ normalized to 1, γ is the probability of partially reacted mixture being present. Generally, γ is expected to be small since the chemical reaction rates converting reactants to products are expected to be large and the probability of finding partially burned gases correspondingly low. This pdf form seems quite plausible and is supported by our results and those of others as noted earlier. It has been used extensively by Bray, Libby, and co-workers (e.g., Refs. 41-43) and by Borghi,[40] among others, for combustion modeling purposes.

In wrinkled laminar flames, contributions to $\gamma f(\xi)$ come from the instantaneous flame zone. For these flames, a physically appealing criterion for γ vanishing small is for the ratio of laminar to turbulent flame thicknesses to be small $\delta/\delta_T \ll 1$. In our studies, $\delta \simeq 1.5$ mm as noted, while at the measurement locations $\delta_T = 2$ and 4 mm. By this reasoning one does not expect γ to be vanishingly small as is confirmed by the pdf data.

If γ is vanishingly small, values of α and β at any location can be determined from $\rho_c\alpha+\rho_H\beta=\bar{\rho}$ and $\alpha+\beta=1$ and, in turn, used to find $\rho'([\rho_c-\bar{\rho})^2\alpha+(\rho_H-\bar{\rho})^2\beta]^{1/2})$. The dashed curves in Fig. 5 were obtained in this manner. One sees that ρ' is overpredicted by this procedure, another indication that γ is not vanishingly small in our flame.

The pdf data can be used to estimate S_T which, in turn, can be compared to measured values. With some assumptions[9] which allow the thermodynamic state of the gas to be completely specified by one scalar, ξ in this case, these pdf data can be used to estimate the mean fuel consumption rate $\bar{\omega}$. With the Bray-Moss pdf form

$$\bar{\omega}=\gamma\int_{\xi_H}^{\xi_c} f(\xi)\,\omega(\xi)\,d\xi \tag{7}$$

Furthermore, if we assume a normal flame[42] (no variation, in the mean, along the flame surface) and stationary turbulence, a time-mean equation for the fuel mass fraction Y may be driven[44]

$$\frac{d}{d\eta}(\overline{\rho u Y})-\frac{d}{d\eta}\left(\overline{\rho D\frac{dY}{d\eta}}\right)=\bar{\omega} \tag{8}$$

Where η is the coordinate perpendicular to the time-mean flame surface. Far from the flame, the gradients of mass fraction are small and $Y\to 0$ as $\eta\to\infty$ (a lean mixture). Integration of Eq. (8) across the flame, $\eta=-\infty$ to ∞, gives

$$\rho_c S_T Y_c=\int_{-\infty}^{\infty}\bar{\omega}\,d\eta \tag{9}$$

an expression for the turbulent flame speed which may be evaluated from the pdf data, provided an expression for ω is available. Several such expressions for methane-air are available in the literature. Borghi et al.[40] report an expression obtained from stirred reactor data; while Westbrook and Dryer[45] recommend a different expression which they find gives correct laminar flame speeds and other flame properties for a range of conditions. These two rate expressions are used to estimate $\bar{\omega}$ at $x_l=5.5$ and 6.5 cm. For the calculations $\gamma f(\xi)$ is assumed to be constant as suggested by the data (Figs. 7 and 8), and the value for γf obtained from the minimum of the pdf in the interval $[\xi_H,\xi_C]$. In view of the limited resolution of our density measurements, a more complex form for $\gamma f(\xi)$ is not warranted. Flame speeds estimated from Eq. (9) are similar for both kinetic expressions, about 2 cm/s at $x_l=5.5$ cm and slightly higher at 6.5 cm, and are over an order of magnitude below the measured values of 50 and 58 cm/s, respectively.

The assumptions leading to these estimates of S_T are questionable. However, they are frequently made in modeling; and, therefore, the large discrepancy is disturbing. There are three points which appear most critical to us. First, the measured mean velocity normal to the flame increases by a factor of 3 across the flame rather than the 6.7 expected with the given fuel-air mixture for a normal flame indicating that there is significant streamtube expansion across the flame and that the normal flame assumption may not be valid. Also, the observed rapid flame thickening with increasing x_l is inconsistent with a normal flame assumption. We believe the normal flame assumption is a major weakness in the estimate of S_T.

Second, because of the limited resolution of our measurements, the instantaneous reaction zone may not be

¶This form may be criticized because delta functions are too peaked at ξ_H and ξ_C. As we shall see, the form of the pdf for ξ near ξ_H is important.

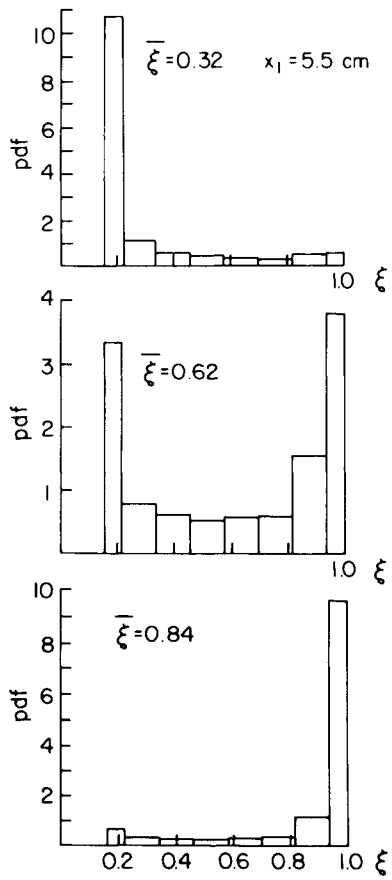

Fig. 7 Histogram representations of density pdf's obtained from the raw data in Fig. 6 ($x_1 = 5.5$ cm) by inversion of Eq. (6).

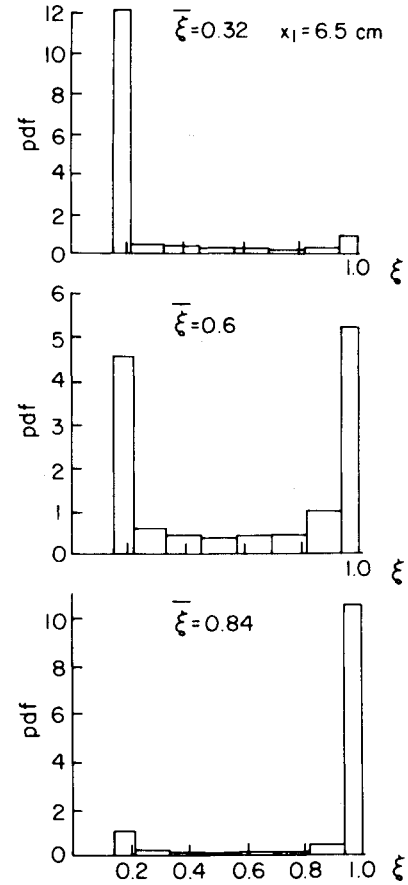

Fig. 8 Histogram representations of density pdf's at the second axial measurement station, $x_1 = 6.5$ cm.

fully resolved; and, hence, the measurement of $\gamma f(\xi)$ may not be sufficiently accurate for this calculation. However, if $f(\xi)$ is slowly varying or constant in ξ, a large, measureable increase in γ would be required for the estimated S_T to approach measured values. Furthermore, inadequate resolution would result in overestimates of γ, not underestimates. On the other hand, should $f(\xi)$ become peaked near ξ_H or rise sharply as ξ approaches ξ_H much larger values of S_T would be estimated. It is quite possible that our system does not have sufficient resolution to discern such trends in $f(\xi)$. Higher resolution measurements are needed to determine accurately the form of $f(\xi)$ in this flame and other flames having a wrinkled laminar flame structure.

Third, the accuracy of the kinetic rate expressions is open to question. Two overall rate expressions from different sources and based on different rate data are used, yet they gave nearly the same results. Thus, one suspects the assumption of one-step, irreversible chemistry rather than the particular choice of rate expression. Westbrook and Dryer[45] discuss more complex mechanisms than a single-step reaction and point out shortcomings in single-step models. Borghi et al.[40] suggest that a more complex rate mechanism may be more appropriate. We believe that the effects of turbulence on kinetic mechanisms require further study.

Spectral density and autocorrelation functions of density fluctuations carry information about the frequency content of these fluctuations. By invoking Taylor's hypothesis,[46] one obtains from such data information about the spatial scales of density inhomogeneities in the reaction zone. Velocity autocorrelation measurements[8] reveal an increase of scale across the flame, and it is of interest to see if similar changes occur in density fluctuation scales. Several investigators have suggested that laminar flame instabilities which occur at discrete frequencies are a major source of density fluctuations in premixed turbulent flames, especially for small R_ℓ. Spectral measurements will reveal the presence of such instabilities and will help identify their mechanisms of formation if they are present.

Spectral measurements were carried out at the positions where pdf measurements are reported. Figure 9 shows a comparison of one-dimensional normalized spectra of density and velocity fluctuations. The zigzag curve shows a density spectrum obtained on the cold side of the flame near the surface of initial mean temperature rise at $x_1 = 5.5$ cm (see Fig. 7 for pdf). The dotted curve shows the velocity spectrum measured by hot-film anemometry at the same location but without combustion. Under Taylor's hypothesis these two spectra can be compared for spatial scales because the total mean velocities in the two cases are the same. The spectra are quite similar with the exception of a peak in the density spectra at 30 Hz. The similarity of the two spectra is easily explained. The different spatial scales of motion in the cold reactant flow contort the reaction sheet on the cold side of the flame according to their sizes and, therefore, the scales of density inhomogeneities on cold side of the flame and velocity eddies in the approach flow are similar.

Figure 10 shows density spectra at $x_1 = 5.5$ cm and at the same x_2 locations as in Fig. 7. Velocity measurements show that the total mean velocities are approximately the same at these positions and, therefore, unnormalized (by the local mean velocity) frequency spectra may be compared in terms of space scales. The spectra do not vary significantly across the flame, indicating that the spatial scales of density inhomogeneities in the mean flow direction do not change across the flame. In particular, there is no indication that these scales increase across the flame as velocity scales do. The spectra of density fluctuations at $\bar{\xi} = 0.12$ and 0.84 show slight local maxima near 250 Hz. These local maxima indicate

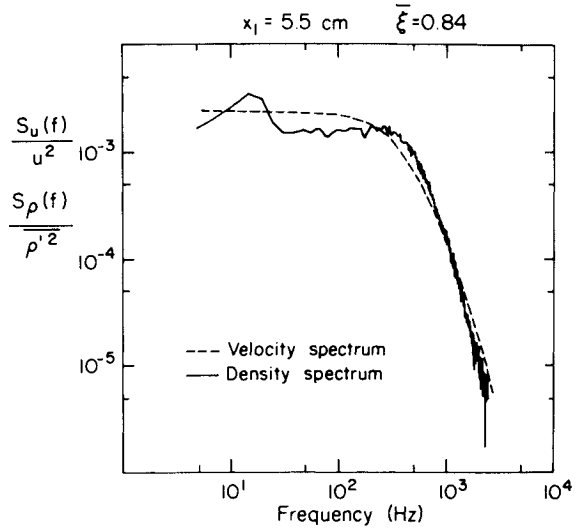

Fig. 9 Comparison of velocity and density fluctuation spectra obtained at the same point on the reactant side of the flame near the point of initial temperature rise. Velocity is measured by hot wire in the absence of combustion; see Ref. 33 for details.

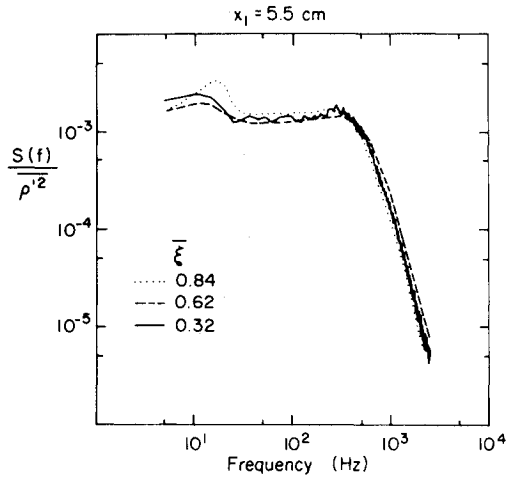

Fig. 10 Comparison of density fluctuation spectra obtained across the flame at the same axial station ($x_1 = 5.5$ cm). Note the spectra are nearly identical above and about 50 Hz.

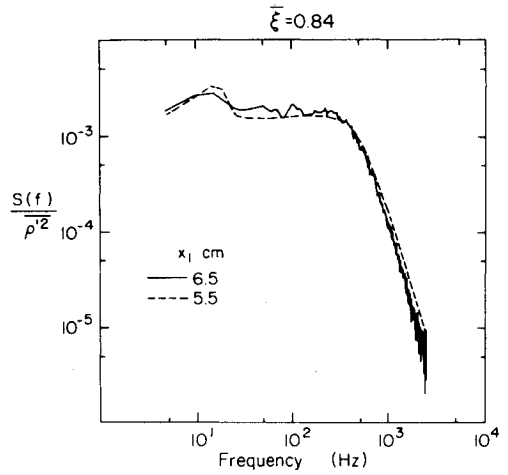

Fig. 11 Comparison of density spectra obtained on the cold side of the flame at the two axial stations showing the great similarity between the two.

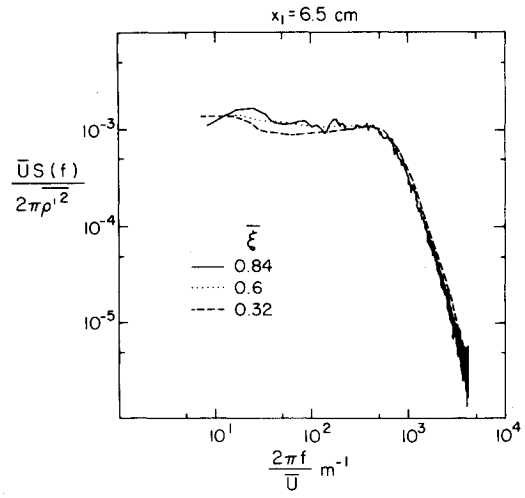

Fig. 12 Comparison of density spectra at $x_1 = 6.5$ cm after conversion to wavenumber space under Taylor's hypothesis. As is the case at $x_1 = 5.5$ cm, there is great similarity between the spectra.

discrete frequency oscillations are present, possibly as a result of flame instability. However, in all cases these oscillations make only minor contributions to measured rms fluctuations.

In constant density, free shear flow (e.g. jets, wakes, and shear layers) the length scales of velocity fluctuations scale with the width of the shear layer, which grows with downstream distance. In the present case, the flame width grows with downstream distance, and it is of interest to see if density fluctuation length scales increase as well. Spectral measurements were made at $x_1 = 6.5$ cm, and results are presented in Figs. 11 and 12. Spectra for $\bar{\xi} = 0.84$ at $x_1 = 5.5$. and 6.5 cm are compared in Fig. 11. The total mean velocities at these measurement points are the same; and, therefore, the frequency spectra can be compared directly. The velocity integral scale of turbulence in the cold reactant flow changes by only 19% from 5.5 to 6.5 cm. The magnitude of the density spectra at the two locations in the frequency range 30 to 300 Hz (where the spectra are comparatively flat) differ by less than 20%. Surprisingly, the large growth in flame thickness between these two axial stations is not reflected in the spectral data.

Figure 12 presents a comparison of normalized wavenumber spectra in the flame at $x_1 = 6.5$ cm and at $\bar{\xi} = 0.84$, 0.62, and 0.32. At these positions, total mean velocities are not equal, and the spectra are scaled with the local total mean velocity to obtain wavenumber spectra. As the results at $x_1 = 5.5$ cm indicate, these data show that normalized wavenumber spectra do not vary appreciably across the flame. Overall, the spectral data at the two axial locations suggest that the axial length scales of density inhomogeneities in the reaction zone are affected primarily by the length scales of the approach flow turbulence and to a much lesser extent by the thickness of the reaction zone and by combustion related processes such as dilatation.

Since the direction of flow is nearly parallel to the flame, these spectral measurements yield length scales along the flame front, and although these scales appear nearly constant, the observed growth in flame thickness implies that lateral scales do grow. Recent space correlation measurements by Rajan et al.[28] in propane-air V-flames confirm that lateral space scales increase with downstream distance. Their measurements and measurements on our burner show that the lateral distance over which wrinkled laminar flame motion occurs increases with distance with a resulting increase in lateral space scales.

Two explanations, both employing the wrinkled laminar flame picture, for these space scale observations are suggested—laminar flame instabilities and turbulent

dispersion. The first explanation assumes that flame instabilities are triggered by turbulence in the reactant flow and that these instabilities are manifest as weaves in the laminar flame which grow in amplitude as they propagate downstream along the flame. Density fluctuations occur as the instantaneous laminar flame oscillates back and forth across the measurement volume. Scales in the flow direction are determined by the wavelengths of these oscillations, while lateral scales are related to wave amplitude. The absence of energetic discrete frequency oscillations in the measured density spectra suggest that, if flame waves are present, they must be present at all frequencies and there must be many waves with random phase relationships. There are two obvious difficulties with this explanation. First, while laminar flames are known to be unstable under certain conditions, it seems unlikely that our flames are unstable to a broad range of excitation frequencies in the reactant flow. Second, an increase in wave amplitude with distance means an increase in average laminar flame surface area per unit mean turbulent flame surface area. According to the wrinkled laminar flame model such an increase will cause a corresponding increase in the turbulent flame speed. No such large increase in S_T with distance is observed.

Alternately, one can argue that the lateral distance over which the laminar flame can be found will increase with downstream distance solely by turbulence induced random motion, a random walk process, or turbulent dispersion. In this picture the scales of instantaneous flame wrinkling are determined by the approach flow turbulence, and there is no increase in laminar flame area per unit turbulent flame surface with increasing x_1. However, the lateral distance over which the instantaneous flame travels increases with axial distance due to the cumulative effect of all turbulence induced random displacements of the flame upstream of the plane of observation. Thus, as x_1 increases, the turbulent flame thickens, the lateral space scales increase, but there is no corresponding significant change in S_T.

A similar process of turbulence dispersion is observed behind heated wires in turbulent flow produced by grids.[47] With a small wire there is no momentum wake and the wire is a line source of temperature excess. Near the wire, the thermal wake is found to consist of a wrinkled, but contiguous sheet of temperature excess analogous to a wrinkled laminar flame. Further downstream the sheet becomes thick and fragmented due to molecular and turbulent diffusion. In the near field where the sheet is present, the mean thermal wake is found to grow linearly with downstream distance. Smith and Gouldin[16] also find a linear growth rate for flame thickness in V-flames. Although at this time conclusive evidence is not available, we believe that the latter explanation, turbulent dispersion, for the observed flame thickening is the correct one and that its validity should be tested further by experiment and analysis.

Some measure of system resolution and shot noise effects can be obtained from the power spectral data. Shot noise contributes a white spectrum, and from Figs. 10-13 it is evident that this contribution has been effectively removed by analysis. Also, it is seen that, at frequencies of approximately 2 kHz and above, the power spectra drop by over two orders of magnitude from their maximum values, indicating that the system has sufficient response to provide good estimates of mean values such as rms and of the large scales of fluctuation.

In conclusion, it is interesting to note that Yoshida and Gunther[13] on a bunsen burner and Parker and Guillon[10] on a burner similar to ours measure temperature fluctuation spectra and find the same similarity between temperature spectra in the flame, properly normalized, and velocity spectra in the approach flow. Other similarities with the Yoshida and Gunther data are observed: the flame thickens with downstream distance while local maxima in rms temperature fluctuations are relatively constant along the flame. These similarities suggest a similar instantaneous wrinkled laminar flame structure for the two experiments despite differences in burner geometry and flow conditions. On the other hand, previous measurements in our burner at higher flow velocities[16] give different trends in rms temperature fluctuations, while Yoshida and Tsuji[12] on a burner similar to that of Yoshida and Gunther[13] find that rms temperature fluctuations grow in the streamwise direction. Thus, we are forced to conclude that these statistical characteristics of scalar properties are dependent on the burner configuration and on the flow conditions of the experiment.

Summary

We have reported the results of density and velocity measurements in an open, lean, premixed methane-air flame stabilized in grid turbulence of low Reynolds number. The following conclusions summarize our results.

1) Where applicable, Rayleigh scattering can be used to good advantage to measure molecular number density. Although raw data may be contaminated by shot noise, there are effective means for reducing or eliminating this noise in finding statistical quantities such as pdf's, moments, autocorrelations, and spectra.

2) Mean and rms density results show that the mean flame thickens with axial distance but that the local maximum in rms does not change appreciably.

3) Estimates of the mean volumetric fuel consumption rate are made across the flame using measured pdf's and then are used to predict turbulent flame speeds. The predicted speeds are over an order of magnitude low, and doubt is cast a) on the validity of the normal flame assumption for open, oblique flames, b) on the accuracy of pdf forms currently used for modeling, and c) on the accuracy of one-step reaction rate expressions.

4) Density spectra when appropriately normalized are similar, at all locations, to each other and to velocity spectra in the reactant flow implying that density fluctuation length scales in the flow direction are determined by velocity fluctuations in the reactant flow. Fluctuation scales in the lateral direction increase in the flow direction. This thickening can be explained by a turbulence induced random walk process.

5) Our results are consistant with a wrinkled laminar flame picture, as is expected, in view of the low turbulence intensity in the reactant flow.

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