RAYLEIGH LIGHT SCATTERING STUDIES OF TURBULENT MIXING

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INTRODUCTION

Turbulent combustion remains a very active research area. This interest is justified due to the pivotal role it plays in a wide variety of technologically important processes. Development of models for combusting flows is crucial for the more efficient utilization of limited hydrocarbon fuel resources, reduction in environmentally harmful pollutants, more efficient chemical processes, and for the prediction of hazard associated with unwanted fire. An important subset of turbulent combustion is turbulent jet diffusion flames because they are relatively simple to generate, and because they provide an appropriate testbed for the development of more general theories for turbulent combustion.

The most widely employed model for turbulent jet diffusion flames is the laminar flamelet concept. [1] In this approach the turbulent combustion is viewed as occurring along stoichiometric contours embedded within the turbulent flow field. Burning along these contours takes place as one-dimensional laminar flames subject to the local stretch introduced by turbulent velocity fluctuations. The scalar dissipation,

$$\chi = 2D\nabla\xi \cdot \nabla\xi, \tag{1}$$

where D is a appropriate universal molecular diffusion coefficient and ξ is the mass mixture fraction, is often employed to characterize the effects of flame stretch. Thus the two most important parameters for laminar flamelet modeling are the local mixture fraction and the scalar dissipation at the stoichiometric contour. Recently, Bilger has argued that turbulent combustion is more likely to occur in broadened reaction zones which have turbulence embedded within. [2] He suggests that such reaction behavior can be modeled if the local mixture fraction and scalar dissipation are known.

It has proven exceedingly difficult to measure mixture fraction and scalar dissipation within turbulent jet diffusion flames. For this reason it is necessary to attempt to characterize their behavior in nonreacting flows and extend the findings to turbulent combustion systems. Most advances in the understanding of turbulent combustion have occurred in this manner. Very recently researchers have even attempted to characterize the structure of chemically reacting turbulent flows by superimposing reacting fields on constant temperature images of turbulent mixing. [2]

During the past twelve years, efforts at NIST (formerly the National Bureau of Standards) have focused on the development of Rayleigh light scattering (RLS) for characterizing real-time scalar fluctuations in isothermal turbulent flows. By combining this technique with hot-wire anemometry, simultaneous real-time

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measurements of concentration and velocity have been possible. RLS measurements have been extended to many points by using linear and two-dimensional detectors and line or sheet lighting, respectively.

During this talk the various experimental approaches which have been developed will be reviewed, and some of the most important findings from earlier investigations will be summarized. Topics to be addressed include the similarity behavior of axisymmetic jets [3],[4] and the stabilization of lifted turbulent jet diffusion flames. [5],[6],[7] More recent work concerned with organized motion and scalar dissipation behavior in the these turbulent flow fields will be discussed as well. [8],[9]

RAYLEIGH LIGHT SCATTERING FOR CONCENTRATION MEASUREMENT

Rayleigh light scattering refers to the elastic scattering (i.e., no change from the incident wavelength) of light by molecular species. For a mixture of M gases, the intensity of RLS is given by

$$I(90°) = N\left(\sum_{j=1}^{M} \sigma_j(90°)X_j\right)I_o, \tag{2}$$

where N is the total number density of molecules, σ_j is the Rayleigh scattering cross section for the wavelength of incident light, X_j is the mole fraction of species j, and I_o is the incident light irradiance. This expression assumes that the molecules are isotropic. For this case the scattered light has the same polarization as the incident light. More complicated expressions are required for scattering from anisotropic molecules which depolarize the scattered light. [10]

A typical RLS experiment involves monitoring the scattered light intensity from a short length of a laser beam located within a gas flow. The information which can be determined from the signal depends on the type of gas which is located within the observation volume. RLS has been widely used to study combusting flows. Clearly, Eq. 2 does not allow the determination of individual species in such experiments. However, it has been shown that for some cases the RLS signal is proportional to density (e.g, [11]) and for carefully chosen conditions can be proportional to temperature (e.g., [12]). For the isothermal mixtures of two gases which are the focus of this paper, sufficient information is available to determine the mole fractions and hence mass fractions (i.e., mixture fractions) for the two species.

For isothermal binary mixtures it is typical to perform calibrations of the RLS scattering intensities for the two unmixed gases. The resulting expression for calculating the mole fraction of gas 2, $X_2(t)$ where the t dependence indicates that X_2 may fluctuate with time, is quite simple,

$$X_2(t) = \frac{I(t)-I_1}{I_2-I_1}, \tag{3}$$

where I(t) is the time-dependent intensity of RLS from the volume containing the gas mixture of interest, and I_2 and I_1 are observed RLS intensities from pure gases 1 and 2. The mole fraction of gas 1 is simply obtained from Eq. (3) as $X_1(t) = 1 - X_2(t)$. Once mole fractions are available, mass fractions, $Y_1(t)$ and $Y_2(t)$, can be calculated using standard formulas. An important advantage of Eq. (3) is that only relative values of RLS intensity need to be measured. It is not necessary to measure absolute intensities. This greatly simplifies the optical system and analysis requirements.

An important limitation of RLS is that resonant scattering from small particles (Mie scattering), which are generally present in laboratory environments, and surfaces (glare) can be much stronger than RLS. These light sources act as interferents and noise sources and, in worst cases, can preclude RLS measurements.

RLS MEASUREMENT OF CONCENTRATION IN TURBULENT FLOW FIELDS

By using either pulsed (e.g., [13]) or continuous wave (cw, e.g., [14]) high-powered lasers to induce RLS, it is possible to measure concentrations in turbulent flow fields of two gases with high temporal and spatial resolution. Pulsed lasers are usually employed to make multipoint (line or plane) measurements. These lasers generally generate very short pulses (in the ns to μs range) which "freeze" the turbulent motion, but have low repetition rates (typically 10 Hz) which preclude following the time behavior of the flow. cw lasers have usually been used for point measurements, but measurements along a line defined by a laser beam have also been demonstrated. [15] Data rates for such measurements are generally high enough to allow real-time recording of RLS scattering to follow the turbulent concentration fluctuations within a flow.

EXPERIMENTAL SYSTEMS DEVELOPED AT NIST

Over the past thirteen years a series of flow systems and experimental configurations have been developed at NIST for the measurement of concentration and velocity in turbulent flow fields generated by the flow of axisymmetric jets into a second gas. The focus has been on the development of systems capable of real-time measurements having high spatial and temporal resolution. Space limitations allow only brief descriptions of the experiments. Interested readers are referred to the cited references for additional details.

FLOW SYSTEMS

Figure 1 shows the initial experimental RLS system developed for single-point scalar measurements in a turbulent flow. A relatively high speed flow of one gas from a 6.35 mm diameter pipe enters a slow coflow of a second gas contained within a square glass enclosure having 10 cm sides. Both flows are filtered to remove particles. Most turbulent studies using RLS have used similar configurations to limit Mie scattering interferences since accurate measurements have proven difficult in the presence of particles. [16]

Recently a new experimental system [termed the Rayleigh Light Scattering Facility] has been developed to provide a large enough dust-free volume to allow measurements of free jets. [17] Figure 2 shows a schematic for the system. A flow of air filtered by high efficiency particle filters is used to sweep particles from within the 2.3 m long by 2.3 m diameter cylindrical test section. Experiments are performed by turning off the blowers and waiting for flow transients to dissipate. This system provides a nearly ideal environment for RLS studies of mixing since both Mie scattering and glare are barely detectable.

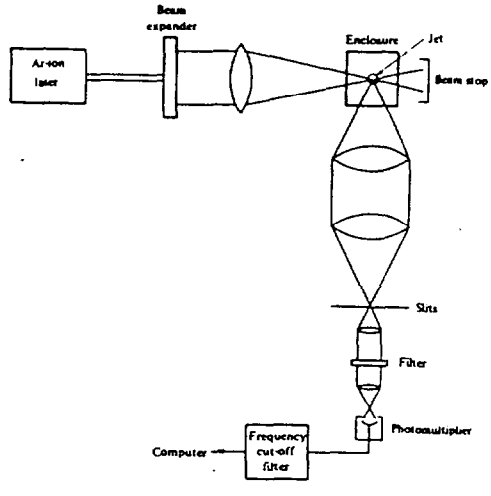

Figure 1. A schematic for real-time RLS measurement of concentration at a single point located within an axisymmetric jet of one gas entering a slow coflow of a second gas.

Jets within the enclosure have been formed either from the pipe flow described above or by using a contoured nozzle designed to generate a top hat velocity profile at the jet exit. The enclosure includes a three-dimensional positioning system for orienting

the flow field relative to the optics which are fixed within the enclosure.

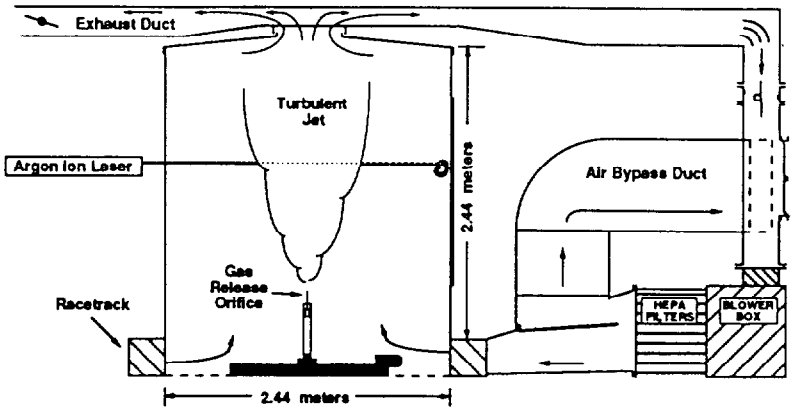

Figure 2. Schematic for the Rayleigh Light Scattering Facility.

SINGLE-POINT MEASUREMENTS OF CONCENTRATION

The use of RLS scattering for concentration measurements in turbulent flows has been discussed in detail by Pitts and Kashiwagi. [10] The optical system required for single-point RLS measurements is fairly simple (see Fig. 1). It consists of a lens to focus the laser beam from an 18 W Ar ion laser to a narrow waist (50 μm is a typical diameter) and an f/2 light collection system for imaging the scattered RLS onto a pinhole which defines the length of the observation volume. Light is then detected by a photomultiplier tube for which the current output (proportional to detected light intensity) is converted to a voltage, digitized, and stored in the memory of a computer for later analysis. Due to computer memory limitations, early measurements were limited to data records of 32,768 points. The incorporation of a new computer system now allows data sets of several hundred thousand measurements to be recorded.

Since RLS is a relatively weak optical process, the largest noise source is usually the Poisson statistics associated with photon detection, which is equal to the square root of the number of detected photons. [18] The intensity of RLS varies with the gas pairs studied so the noise level in a given experiment depends on the gases in the mixture. However, it has been shown that measurements of real-time concentration having accuracies better than 1% of full scale can be made at data rates of several thousand Hz for observation volumes as small as 0.0003 mm^3. [10] By careful design of the optical system, Dowling and Dimotakis have achieved somewhat higher temporal and spatial resolution. [19]

REAL-TIME LINE MEASUREMENTS OF CONCENTRATION

Single-point measurements are recorded by using optics to isolate a short length of scattered laser light from a laser beam. Clearly if it were possible to rapidly image several individual lengths of the laser beam onto a number of detectors it would be possible to record real-time concentration fluctuations at a number of points simultaneously. Pitts has designed a line camera system which is capable of recording propane (which has a particularly strong RLS signal) mole fraction at 128 adjacent points with a maximum data rate of 2.37 kHz. A schematic for the line camera is shown in Fig. 3. The camera design required the incorporation of an image intensifier. Due to the relatively high noise level associated with the line scanner used in the camera and the desire for high temporal response, it was necessary to use a generation I image intensifier equipped with a high-speed phosphor. An earlier version of the line camera has been described in detail. [15] It incorporated a two-stage, generation I image intensifier equipped with a P-46 phosphor. A later version of the camera having a thirteen-fold improvement in signal-to-noise ratio utilizes a three-stage image intensifier with a P-47 phosphor screen. [7]

The line camera is designed for 1:1 focusing of a 14.7 mm length of the laser beam onto the image intensifier. A 4:1 fiber optic taper couples the output of the image intensifier to the 3.2 mm length of the 128 pixel line scanner. The effective spatial resolution is $\approx$ 0.2 mm. The line scanner limits the maximum line read-out rate to 2.37 kHz. The output of the line scanner is digitized and stored by a computer. Early experiments using the camera were limited to 256 total line scans by computer memory limitations. The replacement of

the older computer system now allows many thousands of scans to be recorded during a single experiment.

COMBINED INSTANTANEOUS TWO-DIMENSIONAL AND REAL-TIME LINE MEASUREMENTS OF CONCENTRATION

Recently, Richards and Pitts have recorded two-dimensional images of RLS using a cooled CCD array to image RLS from a sheet generated by passing the second harmonic (532 nm wavelength) of a Nd^{+3}/YAG laser through a series of cylindrical lenses. [9] This experimental approach was first used by Escoda and Long. [13] Since the laser generates a 10 ns pulse, the flow field is "frozen" and two-dimensional images of concentration can be derived.

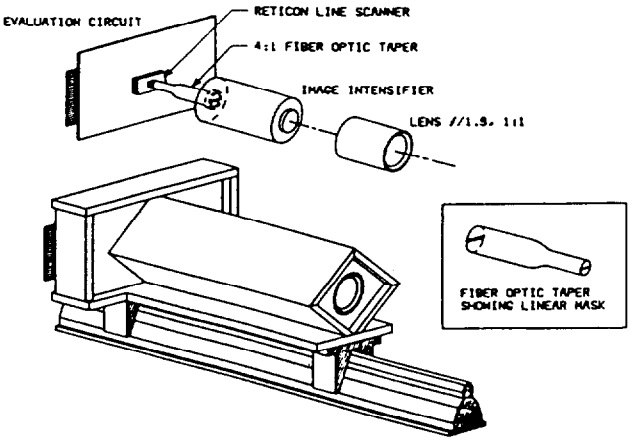

Figure 3 Schematic of the camera system used for real-time RLS measurements of concentration.

In addition to the sheet from the YAG laser, the cw Ar ion laser beam was passed through the flow field such that it was aligned parallel to and just downstream of the laser sheet. The real-time line camera was used to record measurements of concentration at this position. In this way it was possible to monitor the concentration fluctuations generated by a flow whose two-dimensional distribution was recorded just upstream an instant earlier. Figure 4 is a schematic of the optical system used for these experiments.

SIMULTANEOUS REAL-TIME MEASUREMENTS OF CONCENTRATION AND VELOCITY

Simultaneous measurements of velocity and scalar fluctuations allow details of the turbulent mixing process to be investigated. Until recently, experimental techniques were not available for such measurements, but a number of approaches have been demonstrated during the past few years. [20],[21],[22],[23] We have employed RLS and hot-wire anemometry for this purpose. [24],[25]

It is not possible to use hot-wire anemometry alone for velocity measurements in fluctuating binary mixtures since hot-wires not only respond to velocity fluctuations, but are also sensitive to variations in the gas thermal conductivity resulting from concentration fluctuations. It has been possible to overcome this limitation by using RLS to provide concentration measurements which are then used to deconvolute the velocity fluctuations from those generated by the concentration fluctuations. Calibration of the hot-wire response is required as a function of both velocity and concentration.

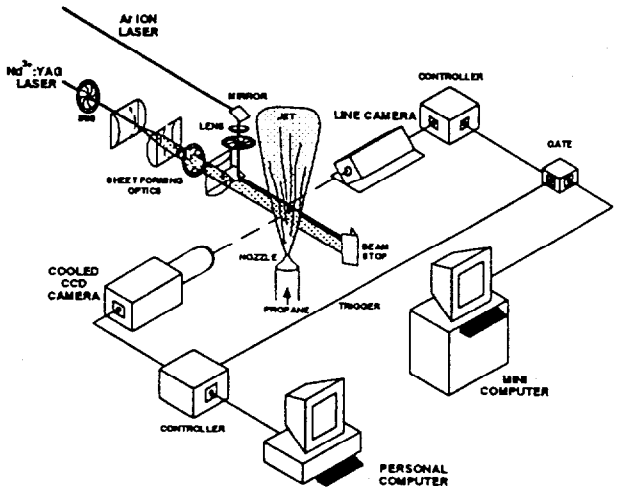

Figure 4. Schematic of the optical arrangement of joint instantaneous two-dimensional and real-time line measurements of concentration.

Pitts et al. have demonstrated that accurate velocity and concentration measurements are feasible at a single point. [24] Muck et al. have extended the technique to a line by

utilizing a linear rake of seven hot-wires and the line camera to provide real-time (maximum of 2.37 kHz) simultaneous measurements of concentration at 128 points and velocity at seven positions spaced along the RLS observation line. [25]

RESULTS

SIMILARITY BEHAVIOR OF VARIABLE-DENSITY FLOWS

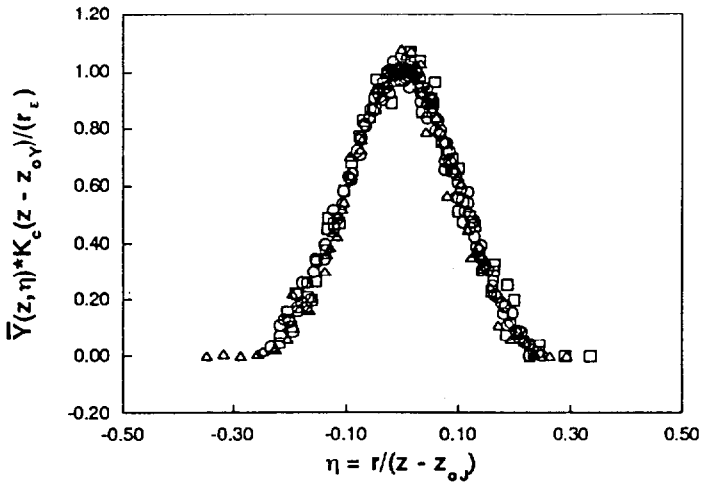

Figure 5. Similarity plots of time-averaged mass fraction for axisymmetric jets of helium (○), methane (□), and propane (△).

It has been known for some time that constant density turbulent axisymmetric jets develop a self-similar behavior in which only a single length scale is necessary to characterize the time-averaged and fluctuation profiles. (e.g., [26]) An important question was whether or not jets having global density variations due to density differences between the jet and surroundings would also develop self-similar behavior with increasing downstream distance as their densities approached that for the ambient gas due to continual entrainment of fluid into the jet.

Earlier experiments (including work from our laboratory) indicated that such flows did not achieve self-similar behavior. [3],[27] The Rayleigh light scattering facility allowed careful measurements of the scalar and fluctuation fields for jets of helium, methane, and propane entering ambient air for which buoyancy effects were unimportant. [4] Figure 5 shows the experimental results for time-averaged concentration plotted in terms of appropriate similarity parameters. The collapse of the data is excellent, indicating that these flow fields do achieve fully self-similar behavior. Profiles for concentration fluctuations also obey similarity relationships. The similarity profiles agree well with those measured for constant density jets by Dowling and Dimotakis. [19]

Propane jet measurements were recorded for both the pipe and contoured-nozzle flows, which are expected to have quite different initial velocity profiles and turbulence levels. The results indicate that the final similarity state achieved by the variable density flows is independent of the initial density ratio as well as the velocity distribution. Interestingly, it was found that two virtual origins were required to provide the collapse of the data shown in Fig. 5. This observation is attributed to differences in the development of the scalar and velocity fields in the near field of the jet.

APPLICATION OF EXPERIMENTAL FINDINGS TO THE UNDERSTANDING OF STABILIZATION MECHANISMS AND BLOW OUT OF LIFTED TURBULENT JET DIFFUSION FLAMES

The stabilization mechanisms of lifted turbulent jet flames have been the subject of a great deal of recent interest. The experimental behavior is well characterized. As the velocity of a turbulent jet flame is increased, the jet first detaches from the burner lip, and the base of the flame fluctuates around a well characterized position downstream of the jet exit. The lift-off height is found to increase linearly with jet exit velocity. Eventually a velocity is reached where the flame suddenly moves rapidly downstream and disappears (i.e, the flame blows out).

Many theories have been proposed to explain lift-off and blow out behavior. However, Pitts has argued that all of these theories have important limitations. [7] An understanding of the mechanisms for these fairly well characterized processes is necessary for improved modeling of chemically reacting turbulent flow and extinction.

Simultaneous line measurements of concentration and velocity have been used to provide insights into the velocity and concentration behaviors which are characteristic of the flow positions where flame stabilization takes place. [5],[6],[7] Stabilization is found to occur in the intermittent region of axisymmetric jets where both mixed jet fluid and air are observed. In this region the turbulence is characterized by organized motion of large-scale turbulent structures (LSTS) which result in high concentration gradients at the downstream edges of the structures. The velocity measurements show that these downstream locations are also regions of high strain rate. Locations within the LSTS further upstream tend to have much smaller concentration gradients and strain rates. These observations suggest that LSTS play a fundamental role in flame stabilization and that extinction is likely to take place on downstream edges of LSTS, while flame stabilization is likely in upstream regions.

ORGANIZED MOTION AND SCALAR DISSIPATION IN AXISYMMETRIC JETS

The latest version of the line camera has been used to record real-time line images of concentration for a propane jet flowing from a nozzle into quiescent air. Measurements were made for various radial sections located 40 radii downstream from the jet exit. Three thousand line scans (384,000 individual concentration measurements) were recorded during a single experiment. The availability of such large data sets allows a number of statistical properties for the concentration field to be calculated. The time-averaged radial mass fraction profile is found to be in excellent agreement with earlier measurements in the same flow system. [4]

Actual time series for the data are difficult to reproduce here, but will be shown during the presentation. These real-time line measurements reveal the same types of structures deduced from the short time records recorded earlier. [5],[6],[7] Strong mixing occurs near the centerline. As one moves further from the centerline, the flow becomes intermittent (both mixed jet fluid and ambient air are observed). The time structure found for the turbulent fluid in this region is quite distinctive. For a particular radial location it is observed that when the turbulent fluid first appears (i.e., at the end of a time period during which only air is present) there is a very rapid increase in jet fluid concentration. This rapid increase is followed by much slower fluctuations in concentration which gradually drop off until ambient air is once again observed. Such a behavior has often been noted in single-point scalar measurements and has been referred to as "ramp-like" structures. The line images demonstrate that ramp-like structures extend across a large radial extent of the flow. This is a clear indication that the motions which generate large-scale turbulent structures are organized. Previous measurements suggest that the downstream edges of the structures are the result of strong ejections of fluid from the central region of the jet, and that air entrainment occurs in upstream regions of the LSTS. [28]

Additional insights into the degree of organization can be obtained from temporal correlation coefficient measurements recorded as a function of position along the radial direction. The temporal correlation, $\bar{R}_{ij}$, is defined as

$$\overline{R_{ij}} = \frac{\overline{(Y_i(t)-\bar{Y}_i)(Y_j(t)-\bar{Y}_j)}}{Y_i' Y_j'} \qquad (4)$$

where $Y_i(t)$ and $Y_j(t)$ are instantaneous mass fractions at pixel i and j, respectively, and $\bar{Y}$ and Y' are time-averaged mass fraction and rms values. A plot of such a correlation is shown in Fig. 6. Included on the same plot is the result when the correlations are obtained over 100 scan segments during which there is visual evidence of organized structure. The higher values of $\bar{R}_{ij}$ in the conditional case is indicative of organized LSTS in the flow field.

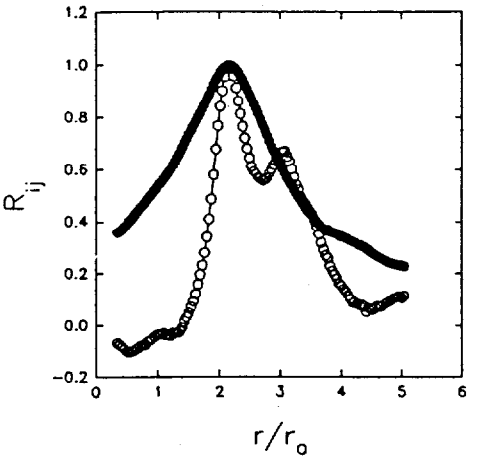

Figure 6. Long time temporal correlations (•) are shown for propane mass fraction measurements taken using the line camera. Short time interval values of R_{ij} conditioned on the presence of turbulent fluid (o) are also plotted.

The line camera results have also been used to estimate values for the time-averaged component of scalar dissipation in the radial direction ($\bar{\chi}_r$). Values of $\bar{\chi}_r$ are shown as a function of r in Fig. 7. By using Taylor's hypothesis, it is also possible to estimate the time-averaged component of scalar dissipation ($\bar{\chi}_z$) in the jet downstream direction. By assuming the azimuthal component equals the radial component, the following relationship is obtained for $\bar{\chi}_e$,

$$\chi_e = 2D[2(\overline{\partial Y/\partial r})^2 + 1/(\bar{U})^2(\overline{\partial Y/\partial t})^2], \qquad (5)$$

where the e subscript indicates that the time-averaged scalar dissipation is an estimated value and $\bar{U}$ is the average velocity for the spatial location which is calculated assuming self-similar behavior for the flow field. During the presentation measured values of χ_e will be compared with available literature data, and a discussion of the appropriateness of using single component data to estimate a three-dimensional scalar dissipation will be addressed.

The last set of results to be discussed will be measurements in which instantaneous two-dimensional images of propane mole fraction have been recorded simultaneously with real-time line measurements at a location just downstream of the sheet location. The purpose of these measurements is to gain insight as to how the structures observed in the line measurements are related to the turbulent structures observed in instantaneous two-dimensional images and how LSTS are modified as they convect downstream. These images are currently being analyzed, and it is not possible to summarize the findings at the time this abstract is being prepared. Results of the analysis will be presented during the talk.

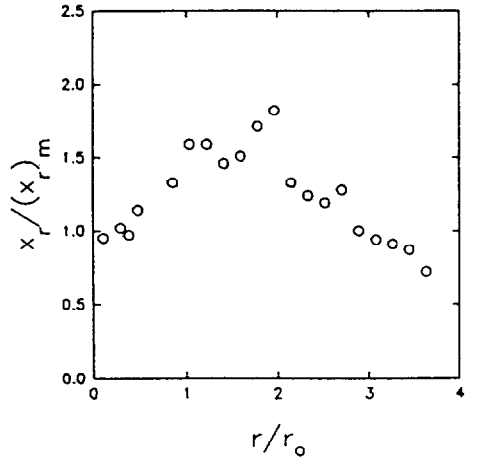

Figure 7. Values of the time-averaged radial component of scalar dissipation (normalized by the centerline value) for a propane jet (Reynolds number = 4000) as a function of radial position twenty diameters from the jet exit.

CONCLUSIONS

A number of new experimental diagnostics for characterizing the scalar and velocity fields in turbulent flows of two gases have been developed. These techniques have been used to demonstrate that LSTS play a fundamental role in the mixing behavior of axisymmetric jets. Measurements of scalar dissipation have contributed to an improved understanding of the behavior of this important turbulence property. The findings of the these studies have indicated the important role of LSTS in the stabilization behavior of lifted turbulent jet diffusion flames. Results of these studies along with those from other laboratories will ultimately provide the understanding necessary to develop effective models for predicting the behaviors of complicated turbulent combustion systems.

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