Experimental investigation of differential diffusion in turbulent mixing using planar imaging measurements

C.J. Brownell* and L.K. Su†

Applied Fluid Imaging Laboratory, Department of Mechanical Engineering
The Johns Hopkins University, Baltimore, MD 21218

Measurements of differential molecular diffusion in a propane-helium jet flowing into air are performed using planar Rayleigh scattering. A comprehensive data set has been obtained for several flow conditions, with Reynolds numbers ranging from 1000 to 3550. Observations show clear evidence of differential diffusion, particularly at low Reynolds number. The standard differential diffusion variable, $\xi$, is computed and profiles of $\xi_{rms}$ show similar features to previous 1-D measurements. In a set of separate, complementary experiments, velocity field measurements for each flow condition are obtained using PIV, and allow estimation of the local Reynolds number. The dependence of $\xi_{rms}$ on the Reynolds number is measured and results are discussed in the context of previous theoretical estimates.

I. Introduction

Simulations of turbulent combustion are complicated by the presence of many distinct chemical species, each with different physical properties. A common simplification is to use a single value of molecular diffusivity for all species involved in a system. The assumption is that in a turbulent flow, where inertial effects dominate diffusive effects, the exact nature of molecular diffusion does not change the overall flame structure. For example, it has been shown that the spreading angle of turbulent jets does not depend on the binary diffusivity of the jet and ambient fluids [4]. The obvious difficulty with this assumption is that chemical reaction requires mixing at the molecular level, where diffusive effects are significant even in turbulent flows. Because the sizes of scalar mixing structures are determined from a balance between strain and diffusion, the range of diffusivities in a reacting flow may make the structure of the mixing field different from one predicted using a uniform diffusivity assumption.

Differential diffusion, defined as the preferential mixing of a scalar with a high diffusivity relative to a scalar with a low diffusivity, has been shown to be an important factor in many reacting flows. Earlier studies involving direct measurement of differential diffusion have been helpful in identifying and measuring its presence in both laminar and turbulent flow fields [10] [12]. In the present work, we use planar imaging techniques to study differential diffusion in a nonreacting flow. We hope to gain insight into the physical mechanisms of three-species mixing, and to provide information useful for future simulations of turbulent combustion. To accomplish this, we have obtained a comprehensive set of planar measurement data to allow analysis of both statistical and structural properties of differential diffusion.

II. Experimental Method

Planar laser Rayleigh scattering yields direct measurements of differential diffusion in a turbulent axisymmetric jet. The jet fluid is a propane-helium mixture, with the species being chosen for their disparate diffusivities ($D_{C_3H_8} = 0.11\, \text{cm}^2/\text{s}$, $D_{He} = 0.72\, \text{cm}^2/\text{s}$) and for their suitability to this Rayleigh scattering

*Graduate Student, cbrownell@jhu.edu, AIAA Student Member
†Assistant Professor, lsu@jhu.edu, AIAA Senior Member

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Rayleigh scattering, a familiar diagnostic method for gas-phase flows, is the elastic scattering of light from molecules or small particles. The Rayleigh scattering signal in a fluid is a function of the intensity of incident light, the local molecular density, and the Rayleigh scattering cross-section. The Rayleigh scattering cross-section, $R$, of a given species depends on its index of refraction, $n$, as $R \sim (n-1)^2$; the scattering cross-section of a mixture is the mole fraction weighted average of the cross-sections of constituent species [5]. A combination of propane ($n = 1.00109$) and helium ($n = 1.000349$) can, given the appropriate ratio, create a mixture that has the same Rayleigh scattering cross-section as air ($n = 1.000293$). The resulting mixture has a nominal initial propane mole fraction $\chi_{C_3H_8} = 0.071$ and helium mole fraction $\chi_{He} = 0.929$.

As this propane-helium jet issues into air, any nonuniformity in the scattering signal is indicative of local differential diffusion of the propane and helium, i.e. departure of the propane-helium ratio from the initial value. (This method was originally suggested by Bilger & Dibble and demonstrated by Long and by Kerstein et al.) [2] [9] [8] To quantify the measurements, we use the differential diffusion variable $\xi$, defined in terms of the propane and helium mole fractions as

$$\xi = \frac{\chi_{C_3H_8} \chi_{He}}{\chi_{C_3H_8}^0 \chi_{He}^0}$$

which can be determined directly from the Rayleigh scattering measurements [8]. The variable $\xi$ will be greater than zero in regions that have a smaller helium-to-propane ratio than at the jet exit, and will be less than zero in regions that have a larger helium-to-propane ratio. In regions where the ratio is unchanged, and in ambient air, $\xi = 0$.

The scalar measurements are supplemented by a complementary set of velocity measurements using PIV. Because the Mie scattering from the PIV tracer particles interferes with Rayleigh scattering signals, the two techniques cannot be performed simultaneously. Instead, the velocity measurements were obtained separately using the same experimental apparatus, and using identical flow conditions.

The propane-helium mixture issues from a round pipe with inner diameter $d = 4.6$ mm into a slow ($U_\infty = 0.5$ m/s) filtered air coflow. Filtering with a high-efficiency (HEPA) filter is essential to remove particulates whose presence would contaminate the Rayleigh scattering signal. The light source is a dual cavity Nd:YAG laser (Spectra-Physics PIV-400), capable of approximately 350 mJ per pulse at 532 nm. Both laser cavities are fired during each exposure, making the intensity of incident light approximately 700 mJ per image. The Rayleigh scattering signal is captured by a thermoelectrically-cooled CCD camera (Roper Scientific CoolSNAP HQ), with $1392 \times 1040$ pixel resolution. In these experiments, the camera is binned to $464 \times 346$ pixels to increase signal levels and reduce noise. The flow rates of helium and propane are each controlled by a Sierra Instruments C-100 mass flow controller.

Data was collected at a variety of jet-exit Reynolds numbers, ranging from 1000 to 3550. For each flow condition, measurements were taken at downstream positions ranging from $x = 7.0$ to $20.0$ $d$, and each condition consists of at least 1000 images. Each image spans from at least the jet centerline to the jet boundary on one side.

Preliminary experiments demonstrated that the results were very sensitive to the exact initial mixture fraction of the jet. We also found that a flow mixed at the theoretical ratio of helium-to-propane (13.02:1) would not produce a mixture with the same Rayleigh scattering cross-section as air, due to impurities in the particular jet gas samples. An optical calibration was performed to determine the appropriate helium-to-propane ratio for the gas samples used in the experiment. A series of images of a laminar propane-helium jet were obtained in the region immediately above the nozzle. Visible in each image were both the core jet fluid (with composition set by the flow rates of the gases) and ambient air. By varying the initial mixture ratio and comparing the intensity of the Rayleigh scattering signal in the core region to that in the ambient air, we determined that a helium-to-propane ratio of 13.20 to 1 results in a mixture with the same Rayleigh scattering cross-section as air.

To calculate the differential diffusion coefficient $\xi$ from Rayleigh scattering images, the images need to be processed to account for the spatial variation of intensity of incident laser light. Periodically throughout the experiment, Rayleigh scattering images of ambient air are captured that can be averaged and divided, as an effective flat field correction, from the most recent jet images. Images need to be further processed to account for pulse-to-pulse variations from the mean laser profile. The information needed to make this instantaneous correction is contained in the image itself, assuming that the jet width does not exceed the width of the imaging window. By interpolating between profiles of the Rayleigh scattering signal from the ambient air, and with geometric information on the trajectory of the laser sheet, we can create an image that represents
the instantaneous deviation of the incident laser profile from the previously defined average. Correcting for this instantaneous variation gives an image where the displayed intensity is exactly proportional to the local Rayleigh scattering cross-section of the mixture.

III. Results

Figures 1 and 2 show sample results for the $\xi$-field measurements. The jet flows upward in the imaging windows, in the positive $x$ direction. Because the jet fluid is predominantly helium, buoyancy has a minor effect on the evolution of the mean velocity, resulting in a local Reynolds number that increases slightly with downstream distance. In the case with jet Reynolds number 1000, shown in Fig. 1, the jet is initially laminar and undergoes the transition to turbulence immediately below the bottom of image (a). Previous results have found clear divisions within the cross-section of a differentially diffusing laminar jet. Specifically, in the laminar core there is a region of jet fluid that has not seen any substantial changes in its relative propane-helium concentration. The outermost radial layer is a wide ring of helium-rich fluid, which has diffused radially farther than the propane. Immediately outside the core there is a region of propane-rich fluid, created primarily by an absence of the fast-diffusing helium. In the current images, large coherent regions of both helium- and propane-rich fluid are visible in the turbulent jet. It is likely that many of these regions were created from differential diffusion during the laminar phase, and have been clipped off and advected downstream into the turbulent region. Image (b) in Fig. 1 is another instantaneous image at Re = 1000, but looking farther downstream than image (a). Typical of images from this location, the regions of helium- and propane-rich fluid are still obvious, but have diminished somewhat in their overall size and intensity. Unlike the laminar portion of the jet, there are no clearly identifiable regions that, averaged over time, are consistently rich in either helium or propane.

The images in Fig. 2 are from a jet with Re = 2500, at two different downstream locations. This set is inherently different from the low Reynolds number jet due to an absence of any significant laminar characteristics near the jet exit. Still, there is significant evidence of differential diffusion throughout both images. Regions of non-zero $\xi$ appear at various points in the flow, although the structures are on average smaller than those from the Re = 1000 jet. In this flow, all deviations from the original helium-propane ratio must have formed in flow regions characterized by at least moderate turbulence.
Figure 2. Sample images from a moderate Reynolds number jet (Re = 2500) show many of the same features as seen in Fig. 1 for both the near-field (a) and far-field (b), although the average magnitude of $\xi$ is significantly lower due to the increased turbulent mixing in this flow.

Figure 3. The location of the imaging areas relative to the nozzle, and the relative sizes of the imaging windows. Probability density functions of the $\xi$-field were computed in the regions designated A to G.

A. Statistical Analysis

For each data set, we have computed probability density functions for $\xi$ at different radial positions throughout the jet. In the near-field regions, statistics were obtained from points in regions A, B, and C, shown in Fig. 3. Region A is along the jet centerline, and regions B and C are centered at $r/d = 1$ and $r/d = 2$, respectively. In the far-field images four different regions were used, as shown in Fig. 3, starting with region D on the centerline and then moving outward one nozzle diameter each for regions E, F, and G.

In the near-field statistics for the Re = 1000 case (Fig. 4), we can see a dramatic difference between the PDF from region C, an area that typically sees no jet fluid, and those from regions A and B that are within the jet boundaries. While the variations seen in region C are attributed to noise, the difference between it and the other two regions is the result of fluctuations in $\xi$ caused by differential diffusion between the helium and the propane. It is also noteworthy that the peak for region A lies slightly above zero, while the peak for region B is below zero. This shows that the helium has spread farther from the jet centerline than the propane, an effect that is likely exaggerated by the pre-existing laminar diffusion. In the far-field these same features are observed, with the primary difference being the convergence of the statistics as all species are given more time to mix. The near-field statistics for the Re = 2500 case, shown in Fig. 5(a), are qualitatively different from those in the lower Reynolds number case. The three PDFs are more consistent, due to the increased turbulent mixing in this flow. We also observe that the PDF that peaks at the highest value of $\xi$ is in region C, on the outside of the jet. This implies a greater prevalence of the slower-diffusing propane on the outside of the jet, a counter-intuitive observation but one that was also seen in experiments by Kerstein.
Figure 4. Cross-stream probability density functions from a Re = 1000 jet show a clear difference between the statistics obtained from the in-jet regions (A, B, D, E, and F) and those obtained outside the jet boundaries (C and G). Within the jet, propane-rich fluid is more common along the centerline and helium-rich fluid is more common on the perimeter.

Figure 5. Cross-stream probability density functions from a Re = 2500 jet are somewhat different from their low Reynolds number counterparts. As this jet was wider than the jet in Fig. 4, regions C and G cannot be assumed to be outside of the jet flow. Also contrary to the lower Reynolds number results, these statistics show a preference for propane-rich fluid on the perimeter and helium-rich fluid near the center of the jet.

et al. (1989). In the far-field, Fig. 5(b), there is further convergence of the different PDFs, although the greater prevalence of propane on the outside of the jet appears to persist.

B. Spatial Autocorrelations and Isotropy

Spatial autocorrelations are useful for studying relevant length scales and isotropy in a flow. In Fig. 6, we see the autocorrelation function of $\xi$ in both the $r$ (radial) and $x$ (axial) directions in a Re = 2500 flow. The autocorrelations were taken along the jet centerline at (a) 13 and (b) 19 diameters from the nozzle.

Figure 6(a) shows similarities in the $r$- and $x$-directions over small scales, but notable differences at scales larger than about $1/4$ d. This suggests anisotropy in this region, and is possibly due to the flow not being fully developed. It also shows that fluctuations in $\xi$ over length scales approaching the integral scale may be significant in certain regions of the flow. In Fig. 6(b), there is convergence in the autocorrelations in the $r$- and $x$-directions, implying increased isotropy with increased downstream distance and flow development. Although we can see that the small length scales are important, the autocorrelation is still greater than zero even as it approaches the integral scale. This observation is important for explaining later results on the Reynolds number dependence of this flow.

C. RMS Fluctuations

In a fully turbulent jet, the mean value of the differential diffusion coefficient $\xi$ at any point in the flow should be zero, or near zero. The appropriate parameter for estimating the magnitude of differential diffusion effects is therefore either the variance of $\xi$ or, as used here, the RMS magnitude of fluctuations in $\xi$, defined as

$$\xi_{rms} = (\xi_n - \bar{\xi})^{1/2}$$

(2)
This quantity has been computed for each data set, and results for Re = 1000 (Fig. 7) and Re = 2500 (Fig. 8) are shown. In the near field at the lowest Reynolds number, we can again see residual features from the upstream laminar region, this time in a bimodal form of the $\zeta_{rms}$ profile. The two peaks are present at the bottom of the image and extend to approximately 8.5 diameters downstream. It is over this region that we expect to see the largest fluctuations in $\zeta$ due to coherent regions of helium- or propane-rich fluid being advected through. As the flow field becomes more turbulent, these peaks are quickly smoothed over, and by $x = 9 \, d$, the $\zeta_{rms}$ profile is very uniform across the entire jet width. From the far-field images, we can see that as the flow evolves further this uniform profile gives way to a second bimodal structure. Here, peak values of $\zeta_{rms}$ are located on the edges of the jet, while the jet centerline sees consistent but smaller fluctuations.

Measurements of $\zeta_{rms}$ from the Re = 2500 jet (Fig. 8) confirm the bimodal structure observed downstream in the lower Reynolds number case. As this flow evolves, we see that the distance between the two peaks increases with the width of the jet, and there is a corresponding decrease in the magnitude of each peak (Fig. 9). The magnitude of $\zeta_{rms}$ at the centerline can also be seen to decrease with increasing downstream distance, but this appears to occur more slowly. The decay can be partially explained by the increased entrained at greater downstream distances, which dilutes the jet species and therefore the Rayleigh scattering signals. These observations of the structure of the $\zeta_{rms}$-field are qualitatively similar to previous results of Kerstein et al. (1989) and Bilger and Dibble (1982).

D. Reynolds Number Dependence

Observing the evolution of $\zeta_{rms}$ and the qualitative changes between the Re = 1000 and the Re = 2500 cases shown above, it is clear that there is a dependence of $\zeta_{rms}$ on the Reynolds number in turbulent flows. Kerstein et al. derived a $\zeta_{rms} \sim Re^{-1/4}$ scaling by assuming that fluctuations in $\zeta$ occur mainly at scales smaller than the smallest scalar length scale of the more diffusive species [7]. This result was in contrast to an earlier result by Bilger and Dibble that resulted in a Re$^{-1/2}$ scaling by assuming a balance of production and dissipation of $\zeta$ based on large-eddy values [2].

To determine the Reynolds number dependence of the differential diffusion in the turbulent jet, it was necessary to determine the local Reynolds number of each flow based on the jet width and centerline velocity. As this jet is predominantly helium, a jet like $x^{-1}$ scaling of the velocity with downstream position cannot be assumed. However, from our velocity measurements we found buoyancy effects to be minor with respect to the scaling properties, and the assumptions of a linear spreading rate and a $x^{-1}$ dependence of centerline velocity to be valid throughout the imaging area. Based on this, the Reynolds number can also be assumed to be relatively constant throughout the imaging area, as in a nonbuoyant jet.

Because the variance calculations are very sensitive to experimental noise, the data used for the following comparisons was collected in a single session to minimize subtle changes in the intensity of the laser sheet or the composition of the gases from one Reynolds number to another. Figure 10 shows profiles of $\zeta_{rms}$ for...
five jets with Reynolds numbers between 2350 and 3550, each taken approximately 15 diameters from the jet exit. Each profile consists of data from 1000 images, and then averaged in the axial direction over a 2 diameter region. The convergence is very slow, but these results are smooth enough so that qualitative conclusions can be drawn. Future experiments will expand this data set over more Reynolds numbers, and assure more complete convergence for each flow condition.

From these five points it was found that $\xi_{\text{rms}}$ varies as $Re^{-0.47}$ in this flow. The observation of a decay in the variance of $\xi$ that is significantly faster than the result derived by Kerstein et al. is likely due to the low levels of turbulence in this flow. The assumption that variations in $\xi$ are completely absent from the large scales is in this case inaccurate, as seen in the spatial autocorrelations earlier. It is likely that a different dependence would be found from repeating this experiment at Reynolds numbers above some threshold where the large scales are no longer important.

IV. Conclusions

Results from differential diffusion imaging of a propane-helium jet issuing into air confirm that differential diffusion is significant in turbulent shear flows. Once present, differential diffusion structures are advected and rapidly dissipated by the turbulent velocity field. The degree to which differential diffusion occurs is affected by the Reynolds number of the flow. In the low Reynolds number turbulent jet studied here, the relationship $\xi'_{\text{rms}} \sim Re^{-0.47}$ is found, which is a faster decay than the prediction for a fully turbulent flow. Differences are explained by the presence of statistically significant variations in $\xi$ over larger length scales than what is expected in a higher Reynolds number flow.

Acknowledgments

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References

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Figure 8. These images show the $\xi'_{rms}$ field for Re = 2500 at different positions within the flow. Here, the images capture slightly more than the jet half-width, to increase spatial resolution while maintaining visibility of all large-scale flow structures. The same bimodal structure seen downstream in Fig. 7 is present here, even though this jet has no obvious laminar regions.

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Figure 9. Profiles from Fig. 8 above give more insight to the evolution of the flow. We observe a decay in the magnitude of the fluctuations in $\xi$ at both the peaks and along the centerline, with a significantly faster decay at the peaks. This decay can be partially explained by the entrainment of air into the flow, diluting the jet species and the Rayleigh scattering signals.

Figure 10. This figure shows profiles of $\xi_{rms}$ for various Reynolds numbers at a fixed point in the jet (approximately 15 diameters). The small fluctuations in each line are due to incomplete convergence of the statistics. When the peak magnitudes here are plotted as a function of the local Reynolds number, we find that $\xi_{rms} \sim Re^{-0.47}$. This is a faster decay than the theoretical prediction by Kerstein et al. but can be justified by the presence of larger scalar structures due to the relatively low Reynolds number.