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14. ABSTRACT AASERT funding was used to support graduate student research in the area of spray droplet vaporization. Droplet lasing spectroscopy (DLS) was applied to the measurement of droplet size and vaporization rates in both reacting and non-reacting rectilinear droplet streams. Ethanol, methanol and a pentane/ethanol mixture were doped with Rhodamine 6G. Lasing spectra were examined in the steady state combustion regime. In the pentane/ethanol case, measurements were carried out in a sooting region of the flame. In some cases, vaporization rates were high enough to measure the rate from consecutive droplets, yielding a quasi-instantaneous measurement. In all cases, the $D^2$ law of droplet vaporization was evident. In addition, photographs of the flames yielded measurements of flame height and thickness. In the final year of support, a triggering and timing system was developed so that the DLS method could be applied to a turbulent flow in which the trajectory and location of the tagged, fluorescent droplets was uncertain.								
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## **Objectives**

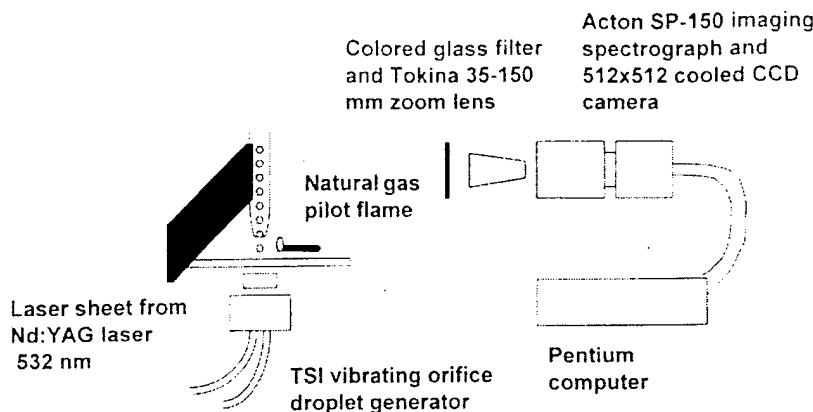
AASERT funding was used to support graduate student research in the area of spray droplet vaporization. The research aimed to develop a method for measuring droplet diameters and vaporization rates in both non-reacting and in reacting flows. The method was based on the application of droplet lasing spectroscopy.

## **Status**

Droplet lasing spectroscopy (DLS) was applied to the measurement of droplet size and vaporization rates in both reacting and non-reacting rectilinear droplet streams. Ethanol, methanol and a pentane/ethanol mixture were doped with Rhodamine 6G. Lasing spectra were examined in the steady state combustion regime. In the pentane/ethanol case, measurements were carried out in a sooting region of the flame. In some cases, vaporization rates were high enough to measure the rate from consecutive droplets, yielding a quasi-instantaneous measurement. In all cases, the  $D^2$  law of droplet vaporization was evident. An application of DLS to sprays was demonstrated at the end of the project period.

## **Accomplishments**

A TSI Model 3450 vibrating orifice aerosol generator was employed in order to form a droplet stream (Fig. 1). The volumetric fuel flow rate was set at  $2.58 \times 10^{-9} \text{ m}^3 \text{ s}^{-1}$ . Rhodamine 6G laser dye was added to the fuel at a concentration of  $2 \times 10^{-4} \text{ M}$ . The impact of the addition of the dye on the boiling point of the fuel was estimated with the assumption of an ideal solution. The elevation of the boiling point was less than  $10^{-4} \text{ K}$  at this concentration and was not expected to cause a measurable change in the vaporization rate of the fuel droplets. The fuel was pumped through a  $20 \mu\text{m}$  orifice via a syringe pump. The jet of fuel was directed upward; the orifice vibrated at a frequency of 20,000 Hz.



*Figure 1 Apparatus for droplet stream spectroscopy studies*

For the reacting cases, a small, natural gas pilot flame with a diameter of approximately 3 mm was used to ignite the flame. It was placed about 1 to 2 mm from the fuel stream. This was close enough to ignite the droplet flames, regardless of the fuel used.

A 10 mm by 1 mm laser sheet of 532 nm radiation from a Nd:YAG laser was directed so that the center of the sheet was approximately 12 mm from the bottom of the flame or from the droplet generator for the non-reacting cases. The ensuing lasing emission from the droplets was imaged with a magnification of 1/1.5 via a Tokina 35-150 mm zoom lens onto the 14 mm by 50  $\mu\text{m}$  entrance slit of an Acton SP-150 imaging spectrograph. The spectrograph housed a 1200 l/mm grating. A cooled 16 bit, 512 x 512 charge coupled device (CCD) camera was attached to the spectrograph and was used to capture the spectra from consecutive droplets. Spectra from 20 to 30 droplets were typically captured per image.

The operating conditions of the Berglund-Liu droplet generator provided an initial droplet size of approximately 63  $\mu\text{m}$ , a steady state droplet spacing of 410  $\mu\text{m}$  (approximately 6.5 diameters) and a velocity of 8.2  $\text{m s}^{-1}$ . The initial droplet size could be calculated using the expression:

$$D = (6Q/\pi f)^{1/3} \quad (1)$$

where Q is the volumetric flow rate and f is the oscillation frequency of the piezo-electric crystal. Using this expression, the droplet size was predicted to be 62.68  $\mu\text{m}$ . A spectrum of lasing from an ethanol droplet

that was obtained near the exit of the Berglund-Liu generator exhibited clear resonances. The diameter that was measured from this DLS spectrum was 62.7  $\mu\text{m}$ , in very close agreement with the estimate of equation (1).

### ***Vaporization and Burning Rate Measurements***

Measurements were obtained for two cases, one with simple vaporization in room temperature air and the other with ignition of the droplet stream with subsequent combustion. Flame heights and thickness were measured from photographs of the three flames with a reference ruler in the background. The alcohol flames were cylindrical and blue with a constant thickness except for the last 5 mm where a yellow/orange ,“candle like” structure was evident. Soot production was limited to the last 5 mm of the flame. The pentane/ethanol flame was blue only for the first 5 to 7 mm and then was yellow/orange. In addition, its thickness grew approximately linearly along the flame. This may have been due to the rapid burning rate of the droplets and an accumulation of fuel vapor in the core of the flame.

### ***Comparison with $D^2$ law theory***

The  $D^2$  Law of droplet vaporization applies to spherically symmetric conditions in a quiescent environment. It predicts that the square of droplet diameter decreases linearly with time. A  $D^2$  law calculation was performed to compare with the measured vaporization rate with a theoretical value. A steady state analysis was valid because the droplet spends approximately 5-7 seconds inside the apparatus, providing sufficient time for steady state conditions to be achieved. The evaporation constant can be found using the following equation:

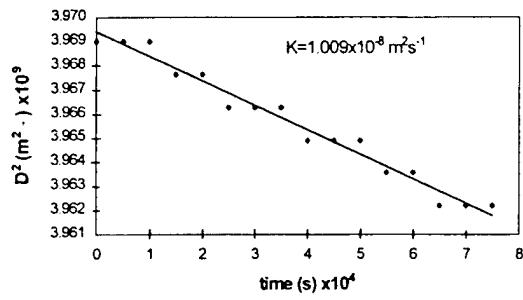
$$K = \frac{8k_g \ln(1+B)}{C_{pg} \rho_f} (1 + 0.3 \text{Re}^{1/2} \text{Pr}^{1/3}) \quad (2)$$

where  $B$ , the heat transfer number is found via iteration and  $(1+0.3\text{Re}^{1/2}\text{Pr}^{1/3})$  is the Ranz-Marshall correction for convection (Ranz and Marshall, 1952). The  $Re$  for this case is 6.3, based on a slip velocity of 2.7 m/s, and a  $Pr$  of 0.72. The calculated value of  $K$  was  $2.2 \times 10^{-8} \text{ m}^2/\text{s}$ . The corresponding constant measured for these conditions was  $4 \times 10^{-8} \text{ m}^2/\text{s} \pm 1.3 \times 10^{-8} \text{ m}^2/\text{s}$ . The similarity of the results that were obtained

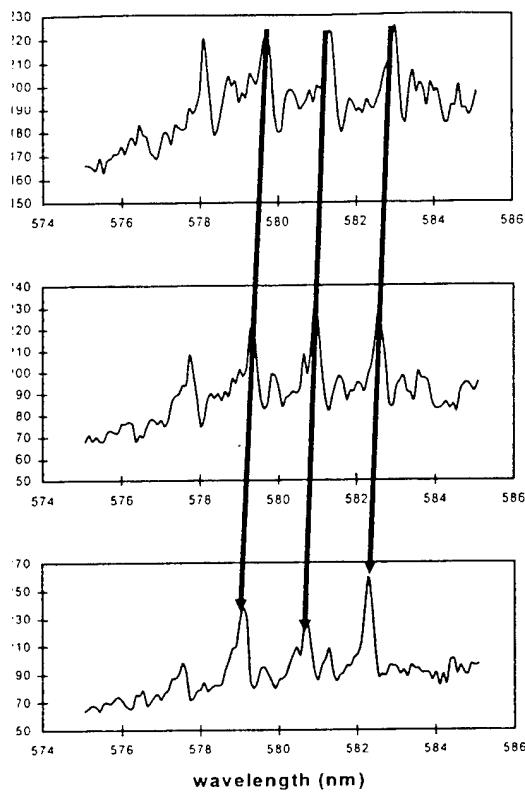
from measurement and theory indicates the potential utility of the DLS method in vaporizing sprays. It is hypothesized that the discrepancy between the measurement and the theory may arise from the acceleration of the droplet at the nozzle exit. The effect of acceleration on droplet vaporization is not incorporated in current theories and models of mass transfer in sprays.

### ***Non-Reacting Droplet Streams***

For the non-reacting cases, three fuels were used viz., ethanol, methanol and a pentane/ethanol mixture (12.5 percent by mass of ethanol). Spectra were captured from approximately 20 droplets in the steady state region of the rectilinear stream of droplets. It was easy to ascertain from the images whether the measurements occurred in the steady region because the images showed each droplet at distinct vertical locations on the CCD (the horizontal axis corresponded to wavelength). Steady droplet flow was indicated by a constant spacing of droplets on the image. Furthermore, if satellite droplets were produced, they would show up in the images as extra spectra with a different peak spacing from the primary droplets. Only ten to fifteen images were taken at each condition because the droplet streams were laminar and were practically identical. The change in droplet size was found by calculating the peak shifts. The peak shifts were obtained by calculating the wavelength distance between the intensity maxima of two peaks, assuming a blue shift. The average vaporization rates were found by plotting  $D^2$  for 5 to 10 droplets and fitting the data to a straight line (see Fig. 2 for an example).



*Figure 2  $D^2$  plot for vaporizing ethanol stream*



*Figure 3 Spectra for three consecutive vaporizing pentane/ethanol droplets with a stream*

The measured vaporization rate constants were  $1.01 \times 10^{-8}$ ,  $1.42 \times 10^{-8}$ , and  $8.13 \times 10^{-8} \text{ m}^2 \text{ s}^{-1}$  for ethanol, methanol, and the pentane/ethanol mixture respectively. The vaporization rates of a methanol droplet stream and an isolated methanol droplet can be compared as follows. From Lee and Law (1992), the vaporization rate of an isolated methanol droplet at an ambient temperature of approximately 1000K was  $4.3 \times 10^{-8} \text{ m}^2 \text{ s}^{-1}$ . As expected, this vaporization rate is somewhat greater than the value that was measured with DLS in the droplet stream. The value measured in a droplet stream was consistent with the results of Silverman and Dunn-Rankin (1994), where the vaporization rates for the droplet stream were found to be 2 to 3 times lower than for the isolated droplet. In the pentane/ethanol case, the significant shift in spectra (see Fig. 3) from droplet to droplet permitted instantaneous vaporization rates to be measured. The vaporization rate calculated from selected droplet pairs was  $7.16 \times 10^{-8} \text{ m}^2 \text{ s}^{-1}$ , while for other pairs it was  $9.52 \times 10^{-8} \text{ m}^2 \text{ s}^{-1}$ . Hence, an apparent variation in the vaporization rate along the stream was evident. However, this variation may have arisen from insufficient

resolution of the spectrum in view of the fact that the small vaporization rate gave rise to a shift in the spectrum of only one or two pixels on the CCD detector.

### ***Reacting Droplet Streams***

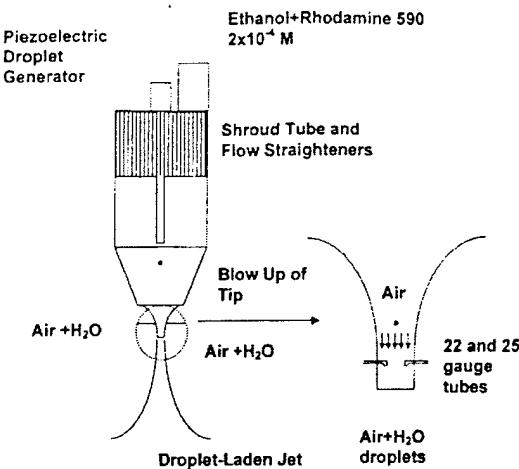
The experiments were repeated for burning droplet streams. To the best of our knowledge, lasing spectra from burning droplets have not been reported previously. The lasing measurements were taken approximately 12 mm from the point of ignition except for the pentane/ethanol mixture where they were made at 10 mm. At approximately 15 mm from the point of ignition, the pentane/ethanol stream became unstable. Hence, the measurements were made far from this region. The burning rate constants were found to be  $1.62 \times 10^{-7}$ ,  $2.08 \times 10^{-7}$ , and  $7.28 \times 10^{-7} \text{ m}^2 \text{ s}^{-1}$  for ethanol, methanol, and the pentane/ethanol mixture respectively.

The pentane/ethanol burning rate compared very well to the measurements of Silverman and Dunn-Rankin (1994) in hexane with a range of droplet spacings. Extrapolation from their data suggested a burning rate constant of approximately  $6.5 \times 10^{-7} \text{ m}^2 \text{ s}^{-1}$  for hexane droplets with a separation of 6.5 diameters. It was expected that the pentane/ethanol mixture, which was predominately pentane, would burn slightly faster than hexane, given the relative burning constants for isolated droplets. Instantaneous burning rates of methanol and ethanol could be measured as a result of the significant shift in the spectra from droplet to droplet.

It should be noted that the measurement of the burning rate for the pentane/ethanol mixture was not as straightforward as in the pure alcohol cases. During the 50 ms between the passage of the droplets, the amount of burning was so great that the peaks shifted more than the peak spacing. This added an ambiguity to the data processing. A burning rate greater than  $2.42 \times 10^{-7} \text{ m}^2 \text{ s}^{-1}$  was found to cause a shift in the spectrum greater than the peak spacing. Consequently, the data analysis used a change in peak spacing to measure the burning rate. This was not difficult because 20 to 30 droplets were imaged at a time and enough droplets were captured so that a reasonable change in peak spacing of 4 to 5 pixels was observed. Fifteen images were collected and all were very repeatable. The average droplet diameter was about 43  $\mu\text{m}$ .

The results from researchers in the area of droplet stream flames are qualitatively very similar, but making quantitative comparisons of the data is difficult because of the varying conditions under which the experiments were done. The computations of Leiroz and Rangel (1997) were undertaken at high ambient temperatures, greater than 500 C. Silverman and Dunn-Rankin (1994) looked at relatively large 100  $\mu\text{m}$  droplets and jets with larger momentum; Sankar et al. (1997) did not quote the droplet spacing. In addition, comparisons with isolated droplet experiments are also difficult because in most of those experiments, the initial droplet size was much larger than those used in droplet stream flames and the ambient temperatures were quite high. Initial droplet size and ambient temperature may have potentially important effects on vaporization rates. For example, the work of Lee and Law (1992) (at an ambient temperature of 1000 K) and Yang et al. (1990) (at 298 K) show a 30 percent difference in methanol burning rates due to ambient temperature differences. Consequently, it has not been possible to attempt a direct comparison of the present vaporization rates in the droplet stream with literature values, other than to note a qualitative agreement with similar investigations.

In the final year of the project, an undergraduate student worked with the Principal Investigator on a method for triggering the YAG laser when a droplet passed through an Ar ion beam that was used for monitoring. Fluorescence from a droplet was imaged onto a pinhole in front of a photomultiplier tube. The segment of the Ar ion beam that was defined by the pinhole determined when the YAG laser should be triggered. Electronic circuits were designed and built to provide the logic and timing signals to synchronize the YAG laser with the CCD camera and spectrometer. The system was implemented successfully and was able to yield images of vaporizing droplets in a turbulent spray exiting a round nozzle using the apparatus shown in Fig. 4. The scheme can be used in the future to make vaporization measurements in sprays.



*Figure 4 Spray apparatus for single droplet spectroscopy studies*

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### **Personnel supported:**

- Philip Santangelo (PhD 1998): Research Staff, Sandia National Laboratory, Livermore
- Daniel Flowers (MS 1997): Research Staff, Lawrence Livermore National Laboratory

## **Publications**

Philip J. Santangelo, Daniel Flowers and Ian M. Kennedy, Demonstration of Droplet Size and Vaporization Rate Measurements in the Near Field of a Two Phase Jet using Droplet Lasing Spectroscopy, *Applied Optics* **37**, 5573-5578 (1998).

Philip J. Santangelo and Ian M. Kennedy , Droplet Lasing Spectroscopy Applied to Droplet Stream Flames, *Combust. Flame* **117**, 413-421 (1999).

## **Interactions**

(a) The results of this research were presented at meetings of the Eastern States Section and the Western States Section of the Combustion Institute, as well as the AIAA Aerospace Sciences Meeting.

P. Santangelo and I. M. Kennedy, An Experimental Study of Droplet Dynamics in a Turbulent Droplet Laden Round Jet, Paper 96F-055, Fall Meeting, Western States Section of the Combustion Institute, University of Southern California CA 1996.

P. J. Santangelo and I. M. Kennedy, An Experimental Study of Droplet Dynamics in a Turbulent Droplet-laden Round Jet, Paper AIAA 97-0124, 35<sup>th</sup> Aerospace Sciences Meeting, Reno NV 1997.

P. Santangelo and I. M. Kennedy, Measurements of Droplet Size in the Near Field of a Droplet Laden Jet using MDR Spectroscopy , Proceedings of Eastern States Section of the Combustion Institute, pp. 265 – 268, Hartford CT 1997.

P. Santangelo and I. M. Kennedy, Droplet Lasing Spectroscopy in Burning Droplet Streams, Paper 98S-05, Spring Meeting, Western States Section of the Combustion Institute, UC Berkeley CA 1998.

Philip Santangelo presented his work at a seminar in the Combustion Research Facility of Sandia National Laboratory.

- (b) None
- (c) None

**New Discoveries, patents etc.**

None

**Honors/Awards**

None

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Title of Article: Demonstration of Droplet Size and Vaporization Rate Measurements in the Near Field of a Two Phase Jet.

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