**Title and Subtitle**

(U) Measurements of Droplet Vaporization in Sprays

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**Supplementary Notes**

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**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. A Berglund-Liu droplet generator was used to generate a stream of droplets, approximately 63 microns in diameter and 6.5 droplet diameters apart. 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 D² law of droplet vaporization was evident. In addition, photographs of the flames yielded measurements of flame height and thickness.
Progress Report

A TSI Model 3450 vibrating orifice aerosol generator was employed in order to form the droplet stream. The volumetric fuel flow rate was set at 2.58x10^9 m^3 s^-1. Rhodamine 6G laser dye was added to the fuel at a concentration of 2 x 10^{-4} 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} 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 μm orifice via a syringe pump. The jet of fuel was directed upward; the orifice vibrated at a frequency of 20,000 Hz.

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 μm 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 μm, a steady state droplet spacing of 410 μm (approximately 6.5 diameters) and a velocity of 8.2 m s^-1. The initial droplet size could be calculated using the expression:

\[ D = \left( \frac{6Q}{\pi f} \right)^{1/3} \]

(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 μm. 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 μm, 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 measurements of flame height and thickness are summarized in Table 1. 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.

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.

The measured vaporization rate constants were $1.01 \times 10^8$, $1.42 \times 10^8$, and $8.13 \times 10^8$ m$^2$ 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 [1], the vaporization rate of an isolated methanol droplet at an ambient temperature of approximately 1000K was $4.3 \times 10^8$ m$^2$ s$^{-1}$. As expected, this vaporization rate was 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 [2], 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 from droplet to droplet permitted instantaneous vaporization rates to be measured.
Reacting Droplet Streams

The experiments were repeated for the 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 $7.28 \times 10^7$ m$^2$ 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 [2] in hexane with a range of droplet spacings. Extrapolation from their data suggested a burning rate constant of approximately $6.5 \times 10^7$ m$^2$ 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 [2]. 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 μs 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 $2.42 \times 10^7$ m$^2$ 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 μm.

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 [3] were undertaken at high ambient temperatures, greater than 500 C. Silverman and Dunn-Rankin [2] looked at relatively large 100 μm droplets and jets with larger momentum; Sankar et al. [4] 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 [1] (at an ambient
temperature of 1000 K) and Yang et al. [5] (at 298 K) show a 30 percent difference in methanol burning rates due to ambient temperature differences. Consequently, it was not possible to attempt a direct comparison of the current vaporization rates in the droplet stream with literature values, other than to note a qualitative agreement with similar investigations.

References


Publications
