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**INFRARED MEASUREMENTS OF SOOT FORMATION AND DROPLET SIZES
IN DIESEL SPRAYS**

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EXECUTIVE SUMMARY

This report describes an investigation of diesel sprays using a combination of infrared wavelength optical diagnostics to probe the high droplet number density region surrounding the injector tip. Infrared wavelengths were shown to be more effective than visible or ultraviolet wavelength light at penetrating this region of the spray. This success is easily explained by the decrease in optical cross section of small diameter drops (less than $10\text{ }\mu\text{m}$) for a wavelength shift from the visible to wavelengths near $10\text{ }\mu\text{m}$. Two types of diagnostics were implemented. First, a custom manufactured, high speed infrared spectrometer was used to monitor the spectral region between 6 and $12\text{ }\mu\text{m}$ in $0.5\text{ }\mu\text{m}$ intervals. Spectra from this instrument, for specific locations in the combusting spray were used to monitor the development of soot in the spray. The second diagnostic technique used three collinear laser beams focussed into a 0.25 mm beam to monitor the droplet size in the spray. This measurement uses a ratio of signals to remove the droplet number density as a variable and the observed signal ratio is a direct indication of the average (using an approximate radius to the fourth power weighting function) droplet size. The experiments were performed using a shock tube to simulate the conditions typical of diesel combustion (700 to 900 K in temperature and approximately 3.0 MPa in pressure) and a custom manufactured single shot fuel injection system. Optical access in the shock tube included visible and infrared transmissive windows arranged to provide orthogonal access in two directions and perpendicular to the spray axis. The fuel injection system for this work was configured to produce approximately 20 MPa of injection pressure and produced a single injection approximately 2 ms after the injection trigger. This system could therefore be controlled to inject into the quiescent reflected region of the shock tube after the incident shock reflection from the end wall.

Results from this study indicate that the wavelength region near $9.4\text{ }\mu\text{m}$ may be used to monitor soot formation in fuel sprays. Soot was observed at locations as close as 5 mm from the spray tip and out to 35 mm from the spray tip. At the 5 mm location, soot appeared after the end of the fuel spray pulse. However, at locations 10 mm from the tip

and beyond, soot was formed during the spray pulse. As the distance from the injector tip was increased the observed soot mass in the spray also increased. Induction times for soot formation were also determined and were found to decrease dramatically as the distance from the injector tip lengthened.

Measurements of droplet diameters were performed in room temperature, ambient pressure sprays, room temperature sprays into a background pressure of 1.1 MPa, room temperature and high background pressure combusting sprays. For the room ambient pressure sprays the effective droplet diameters were typically between 5 and 6 μm for locations 5, 10, and 25 mm from the injector tip. The diameter was observed to increase slightly to 8 μm range 35 mm from the injector tip. These droplet diameters are unexpectedly large. For room temperature sprays into a 1.1 MPa background pressure, the droplet size was comparable to those at atmospheric pressure but we did observe an increase in the droplet number density. The behavior of the high temperature combusting spray was markedly different than its room temperature counterpart as little as 10 mm from the injector tip the temperature fuel sprays displayed an initial period with steady droplet number density followed by ignition and significant axial variation in number density. Droplet diameters were observed to be near 3 μm and a large fraction of the fuel mass in the spray was observed to have vaporized after spray ignition. Initial temperature for the ambient environment was observed to control both the ignition delay for the spray and the rate of the spray that evaporated near the injector tip. Finally, we observed the 9.2 μm laser light scattering to be much more effective for monitoring droplet behavior than its 0.633 μm counter because the interference from soot is much smaller at the infrared wavelength. These results indicate that the region surrounding the injector tip is a primary vaporization zone and therefore will control pollutant formation in the engine. We have also found that room temperature and pressure experiments do not adequately reproduce spray conditions in the diesel engine.

1. INFRARED MEASUREMENTS OF SOOT FORMATION IN DIESEL SPRAYS

1.1 Introduction

Soot is a serious air pollutant and diesel engines, in spite of decades of research work, continue to produce soot at unacceptably high levels. In addition, soot can be a major contributor to diesel engine wear. Advances in non-intrusive optical diagnostics have increased the overall understanding of combustion processes. Unfortunately, the region surrounding a diesel spray is particularly difficult to monitor due to the large number of fuel droplets. The number densities are typically sufficient to render the spray opaque to visible wavelength light and both visible and ultraviolet based optical measurement techniques have been relatively unsuccessful at monitoring processes in the dense spray region. Our approach to this problem has been to continue to pursue optical measurement methods, but to shift the wavelength into the infrared where the optical thickness of the fuel spray is smaller relative to that in the visible. We have developed a technique that can monitor soot formation in the spray environment based upon thermal emission in the spectral region between 6 and 12 μm . The measurements are performed in a simulated diesel environment with temperatures ranging from 700 to 950 K and pressures of approximately 3 MPa. This initial effort presents results for soot formation at several positions in the spray. Presentation of the results, following this introductory section, includes a detailed description of the experiment setup and measurement techniques, the development of the data interpretation methods, and finally measurement results for our spray system.

Soot and the details of its formation have been extensively studied in several types of systems. These studies include gas phase shock tube studies to monitor soot formation as a function of temperature, total pressure, fuel type, and oxidizer content,¹⁻³ diffusion flame studies which monitor soot volume fractions as a function of position in the flame,⁴⁻⁶ and studies of soot in premixed systems^{7,8}. Results from these studies indicate that, for practical combustion devices, soot is formed primarily in pyrolysis and is a very temperature dependent process. Local fuel concentration, total pressure, and the fuel type can also be

very important parameters. Although a detailed pathway has not been identified for hydrocarbon fuels, the overall soot formation process appears to step from fuel to intermediate aromatic compounds to polycyclic aromatic hydrocarbons and finally to soot. Since soot is known to form in high temperature pyrolysis zones (1000 to 2000 K), the injector region for a diesel spray very likely is a primary source for soot in the diesel engine.

1.2 Experimental Description

The experiments were performed with a single shot, high pressure, fuel injector system synchronized to a 38 mm diam shock tube as shown in Figure 1. This fuel injector/shock tube combination is a convenient means of creating a high temperature pressure environment with the advantage of very good optical access, a well characterized temperature and pressure before injection, and a high degree of repeatability. The shock tube consists of a 3.0 m driver and a 5.7 m test section that terminates in a tube of constant cross section. The interface between the driver and test section is of the dual diaphragm variety which helps produce highly repeatable test conditions in the facility. Optical access was included on all four sides of the injector by using rectangular windows with a cross section aperture of 55 mm in the axial direction and 19 mm high. For the infrared measurements that were critical to this effort, the windows were manufactured from water-free ZnSe (tradename Cleartran) and were highly transmissive from 0.37 to 14.0 μm . Conditions in the reflected shock zone were between 700 and 950 K for pressures of approximately 10 to 20 atm. The shock tube was operated in an approximately tailored interface mode in order to maximize the test times available. The test gas was in all cases dry air (including the driver gases were mixtures of helium and argon). Test times were observed to be approximately 10 ms, which was more than adequate for this study. Boundary layer effects on the temperature in reflected shock zone for small diameter tubes such as this one have been documented to be significant⁹. However, the fuel injection/combustion process significantly perturbs the temperature in the system and this perturbation will be much greater than any produced by shock tube boundary layers. The air temperature decreases

FIGURE 1. - Schematic diagram of the shock tube and fuel injector.

to heat addition to the fuel is approximately 100 K (assuming 100 mm spray penetration) and typical adiabatic flame temperatures for a stoichiometric hydrocarbon/air system with an initial air temperature of 750 K are approximately 2500 K.¹⁰ For this study, we therefore quote the initial reflected shock temperature for the experiments and do not follow the evolution of temperature as the event progresses.

The pulsed fuel injector forms the second critical portion of the experiment system is a single stroke pump/amplifier capable of delivering pressures up to 80 MPa and an injection delay of approximately 3 ms. All experiments used pure dodecane (99% pure) as the fuel. The design of the pump follows the work of Oren¹¹ and the injector is triggered by discharging a capacitor charged with 2 kV into a standard 24V solenoid actuated pump. The pump is a positive displacement type system with an adjustable stroke length. For the experiments, we operated the system to produce approximately 20 MPa of injection pressure and a delivery volume of approximately 120 mm³. The fuel injector is connected to the pump outlet and was a Lucas Model BDLL150S6774 mounted in a standard holder. The holder was modified to have a single orifice (0.17 mm diam and 0.61 mm in length). Injection pressure was set to 7 MPa and injector pressure was monitored using a pressure transducer mounted in the fuel delivery line.

Timing is exceptionally critical for these experiments. In all cases, fuel injection was carefully controlled to occur after the incident shock reflection off the end wall. A typical experiment is illustrated in Figure 2 which shows pressure in the reflection zone, injection pressure, spray arrival via obscuration of a HeNe laser directed across the test section, and emission in the infrared from the combusting spray (the location of the spray measurements is shown in Figure 3). This experimental facility is capable of simulating conditions of temperatures and pressures typical of a diesel engine, includes high pressure pulse generation and has extensive optical access for non-intrusive measurement.

Optical measurements for this effort included spectrally resolved infrared emission measurements between 6 and 12 μm and a measurement of spray arrival based on obscuration observed in the HeNe laser. The infrared emission measurements use a manufactured spectrometer capable of collecting synchronous spectra (with 12-channel resolution) at 125 kHz. This device was constructed using a 12-element linear array of HgCdTe detectors mounted on a 0.35m monochromator with a 6.2 μm cut-on entrance wavelength. Wavelength resolution on a per element basis was 0.5 μm . Data acquisition used a Lecroy/Camac system with a dedicated amplifier and digitizer for each array element.

FIGURE 2. - Timing for a typical combustor spray experiment.

FIGURE 3. - Experimental layout for the spray and soot experiments.

spectrometer includes a folded optical entrance path using plane and spherical gold control the field of view. The image size was carefully adjusted to be 1.2 mm at tube centerline and the field of view expanded from 1.2 mm while at the focus point 2.1 mm when 10 mm beyond or in front of the focus point. Finally, this system is mounted on a translation and height adjustable stage so that various regions of the could be monitored easily.

In order to monitor the spray arrival at the image zone for the spectrometer laser was directed through the image zone at right angles to the line-of-sight collected the array spectrometer (the laser beam path was always moved to match the spectrometer image zone). The intensity of the beam was monitored with a red-sensitive phototube (PMT) equipped with a 1 nm notch filter centered at 633 nm. The beam was directed onto an opal glass diffuser and the PMT monitored the brightness of the spot on the diffuser. This arrangement allows the beam to wander over an approximately 10 mm diameter area with an apparent decrease in signal level. The system is therefore relatively immune to variations in beam steering in the spray. This diagnostic was used simply to verify the occurrence of the spray event and in addition proved useful in determining soot initiation times and positions in the spray. The optical setup is illustrated in Figure 3 which illustrates the injector and shock tube test section as well as both the spectrometer and the HeNe measurement system.

1.3 Diagnostic Methods

The focus of this work was to monitor soot formation in the fuel spray. However, the spray environment is exceptionally complex and may contain droplets, fuel vapor, molecular species that are either intermediate or final products of combustion, as well as soot. The soot diagnostic must be capable of differentiating between this array of components and soot. Our strategy was to use the continuum-like spectral signature of soot which is distinctly different from that of any other component in the spray, as our monitor. The array spectrometer is an ideal instrument for this task and the emissive

from the spray are easily detectable. A key to the success of this technique is to monitor a large region of the spectrum such as the 6 to 12 μm region. Figure 4 illustrates the expected "signature" of soot plotted on logarithmic scale for temperatures between 1000 and 2000 K. These curves were calculated for a 20% emissivity at 6.5 μm . The spectral shape was determined by assuming the soot was sufficiently small to be a volumetric absorber, and using the Mie scattering solutions¹² (with the complex index of refraction of soot which varies with wavelength)¹³ to calculate the absorption cross section as a function of wavelength. As shown by this family of curves, a characteristic of soot is a very smooth spectral profile. In addition, due to the relatively long wavelengths being used, the impact of temperature on the shape of the curves is small.

FIGURE 4. - Spectral signature for soot for a range of temperatures.

Other radiators in this region of the spectrum could include the fuel droplets (dodecane), fuel vapor, intermediate hydrocarbon species, and combustion products CO_2 and H_2O . None of these constituents can produce a continuum signature similar to that of the soot. Dodecane drops will not produce a continuum signature like that shown in Figure 4 since the imaginary part of the refractive index varies dramatically between 6 and 12 μm and the temperature of the drops must be less than the critical temperature of dodecane (673 K). Thin film absorption spectra for dodecane indicate strong absorption features near 7 μm with very little absorption elsewhere in the 6 to 12 μm region. Measurements in our laboratory confirmed this result and we estimate the imaginary component of the refractive index to range from 10^{-7} to 10^{-2} in this band. The radiance from dodecane drops will have an emission signal near 7 μm but will emit very weakly in the rest of the spectrum. Dodecane vapor also displays this trend and, based on atmospheric pressure, room temperature absorption measurements we estimate the absorption cross section for dodecane to be $7 \times 10^{-19} \text{ cm}^2/\text{molec}$. Carbon dioxide will radiate between 8.2 μm and 9.0 to 12.0 μm ¹⁵ while water at combustion temperatures will radiate brightly in the 6.5 μm range with a strongly decreasing radiance near 8.0 μm ¹⁶. Other molecular radiators will also display structured spectra leaving soot alone as a continuum radiator. Our measurements in this experiment will therefore be oriented to monitor structured spectra at various regions in the spray; the characteristic continuum signature of soot is used to identify its presence.

1.4. Experimental Results and Discussion

Measurements were performed with the spectrometer positioned to monitor various regions of the combustng spray for a range of initial gas temperatures. Three positions along the spray axis were monitored (5, 20, and 35 mm from the injector tip) and a position above the spray axis was also monitored (35 mm from the injector tip and 35 mm above the spray axis). As shown in Figure 3, the spectrometer is positioned so that the field of view is directed at right angles to the axis of the spray. The observed spectra

are from a region of the spray formed by the spray cone and a line of sight directed at right angles across the spray.

Figures 5 through 12 display results for each of the axial positions described above. These plots include three-dimensional representations of the spectra collected as a function of time and selected spectra at specific times for those events. The vertical axis for both plots is radiance in absolute units plotted on a logarithmic scale. The use of the logarithmic scale is convenient since the relative shapes of spectra are unaffected by the number density of radiators as long the radiating field is optically thin. For the three-dimensional plots, the x-axis represents time and the y-axis represents wavelength. These axes are plotted in a non-standard format; time increases from right to left and wavelength increases from back to front. The choice of this format for plotting results was made to enhance the view of the three-dimensional surface represented in these plots. Time in all plots is the time after the

FIGURE 5. - Three dimensional plot of radiance, 5 mm from the injector tip, as function of time and wavelength. Initial conditions are 893 K and 4.1 MPa.

FIGURE 6. - Spectra for selected times 5 mm from the injector tip.

FIGURE 7. - Three dimensional plot of radiance, 20 mm from the injector tip, function of time and wavelength. Initial conditions are 812 K and

FIGURE 8. - Spectra for selected times 20 mm from the injector tip.

FIGURE 9. - Three dimensional plot of radiance, 35 mm from the injector tip. Initial conditions 894 K and 3.1 MPa.

FIGURE 10. - Spectra for selected times 35 mm from the injector tip.

FIGURE 11. - Three dimensional plot of radiance, 35 mm from the injector tip above the spray axis. Initial conditions 949 K and 3.6 MPa.

FIGURE 12. - Spectra for selected times 35 mm from the injector tip and 5 mm above the injector axis.

incident shock arrives at the first pressure transducer. Each of the plots shows the noise level/background radiance before the arrival of the spray; the spray arrival is noted as determined by HeNe laser. Following the spray arrival the signal climbs, is relatively stable during the spray pulse, and falls at the end of the spray pulse. The utility of the three-dimensional plotting is to provide a summary of the data acquired in one experiment and to aid in the data analysis; by carefully examining these plots spectra at selected times can be examined more thoroughly.

Figure 5 illustrates results along the spray axis and 5 mm from the spray tip. The initial rise in signal out of the background starts at approximately 4500 μ s (more than 1 ms after the arrival of the spray) and rises sharply for approximately 200 μ s. Note that the radiances at shorter wavelengths appear to rise out of the background before their longer wavelength counterparts. This figure illustrates that at different times, different radiators are present. During the initial rise out of the background, the 6.5 to 8.5 μ m region of the

spectrum is much brighter than the long wavelength regions. Following this time spectra evolve to more of a continuum signature, and finally the long wavelength fall again while those at short wavelengths remain relatively bright. Each change for one wavelength relative to another indicates a change in the radiating species. An interesting observation is that the majority of the measurable radiation occurs after the spray pulse. The overall indication is that at least three different periods of the combustion event can be identified at this spray location.

Figure 6 can be used to help understand the behavior of the radiators in each region. First, during the spray pulse, we observe a radiation signal starting in the 8.0 μm region progressing to a spectrum including 6.5 to 10.5 μm at the end of the pulse. The 6.5 to 8.0 μm radiation is clearly the signature of the fuel, dodecane, which, based on its room temperature absorption properties, is expected to radiate in this region.

Unfortunately, at this time we cannot absolutely differentiate between vapor and droplets but we believe that the majority of the signal is due to vapor since the maximum temperature of a droplet is 673 K (the critical temperature for dodecane) while the vapor is at much higher temperatures. The radiances that we observe are less than the maximum radiance for 673 K so that the observed emission could be due to the droplets. However, by measuring the room temperature absorbance of dodecane vapor at 7 μm we can estimate an absorption cross section to be $7 \times 10^{-19} \text{ cm}^2/\text{molec}$; for 6 μm diameter droplets, at an absorption efficiency of one, the cross section on a per molecule basis is approximately $10^{-20} \text{ cm}^2/\text{molec}$. Direct comparison of these cross sections reveals that, for equal temperatures, dodecane vapor radiates 70 times more brightly than its droplet counterpart on a per mass basis. This implies that approximately 1% of the fuel mass would have to vaporize for radiation from the vapor to have a brightness equal to the droplets at maximum temperature, 673 K. Since the droplets at this temperature must be evaporating significantly, we conclude that the radiative signature must be due to vapor rather than droplets. To further support this conclusion, we have used the results summarized by Lefevre¹⁷ to roughly estimate the time required to vaporize 1% of a 6 μm diameter

Using a conservative estimate for the evaporative constant, $0.1 \text{ mm}^2/\text{s}$, the time required to evaporate 1% of the droplet mass is $3 \text{ } \mu\text{s}$ compared to the $50 \text{ } \mu\text{s}$ required to traverse 5 mm in the spray (the spray velocity was measured to be approximately 100 m/s by monitoring spray arrival at different points on the spray axis with a HeNe laser). Clearly the vaporization process can proceed quickly enough to produce fuel vapor sufficient to dominate the radiative signature of the fuel droplets. Therefore, the obvious presence of high temperature dodecane implies fuel vaporization only 5 mm from the tip.

This fuel vapor can either be convected along with the spray or conversely, could move much more slowly than the high velocity droplets which comprise the spray. We observe an increasing radiance level from dodecane during the spray pulse which implies that fuel vapor is not convecting along with the spray (if the vapor were moving with the droplets, a steady-state signal would be achieved). This implies that a slower moving region surrounding the spray contains the fuel vapor.

Spectra taken at times 5, 5.5, and 6.0 ms contain a continuum component indicative of soot. Figure 6 includes a soot radiative signature and shows that the spectra from 5.0, 5.5, and 6.0 ms contain molecular radiative components on top of a broad, soot-like continuum. This is in sharp contrast to the spectra from 4.2, 4.75, and 7.0 ms, which exhibit no radiation beyond $8 \text{ } \mu\text{m}$.

The end of the spray pulse roughly corresponds to a change in slope in the radiation signals. The remainder of the event is combustion of the fuel that has previously been injected. As shown in Figure 6, this includes two distinct time periods. First, for approximately 1.25 ms after the end of the spray pulse, the spectra are quite bright and include distinct features of the soot continuum. There are clearly molecular radiators in addition to soot but the underlying continuum is distinct. Following this 1.25 ms time period, the continuum radiation disappears. Spectra from this time resemble those of the dodecane fuel seen during the spray; these spectra could also be due to the combustion product water. This implies that the soot which was responsible for the continuum signature

has either left the field of view or has been consumed. Although the nature of this experiment does not allow an exact specification of the stoichiometry, we can estimate the length of shock-heated gas slug at the initial pressure and temperature required to heat the 120 mm³ injected into the system. This estimate is 100 mm, comparable to the spray penetration, which implies that the event was near stoichiometric.

We therefore expect that nearly all of the fuel participated in the combustion and a negligible fraction of the initial fuel remains as dodecane. Even for fuel rich conditions of the system, we expect pyrolytic reactions, at the combustion temperatures, to transform fuel into intermediate hydrocarbon products or soot.

We therefore expect a negligible fraction of the fuel to remain as n-dodecane. As a product of combustion, water will definitely be present in relatively high concentrations ($\sim 4 \times 10^{19}$ molec/cm² if the jet reacts in a 100 mm length). Our measurements of vapor emission¹⁶ indicate that the intensity of the signal we observe is consistent with concentrations of approximately 10^{19} molec/cm². The late time emission signal can therefore be attributed to water vapor.

Figures 5 and 6 have illustrated the behavior of the spray 5 mm from the injector. We observed fuel vapor during the spray event; after the spray pulse, we saw a radiation signature with an underlying continuum followed by a signature typical of water vapor.

Figures 7 and 8 show results for a position 20 mm from the spray injector. In Figure 5, the arrival and end of the spray pulse are noted. Within 200 μ s of the spray arrival, the characteristic signature of the fuel can be noted. This signature is shown in the early time spectrum of Figure 8. Approximately 200 μ s after the appearance of fuel vapor, the radiative signals increase dramatically and evolve into continuum-like signals as shown in Figure 8. Once again these spectra appear to be an underlying soot continuum with molecular radiators contributing in the 6.5 to 8.5 μ m range (presumably from dodecane), in the 10 μ m range and near 11.5 μ m. At the end of the spray pulse,

vapor signature is apparent as shown by the late time spectra in Figure 8. This figure includes a reference water spectrum for comparison.¹⁶ Our measurements at this location indicate substantially different behavior than at 5 mm from the spray tip. As before we observe the signature of the fuel after the arrival of the spray. However, ignition and combustion appear to occur at this point during the spray pulse after 200 μ s and we observe a soot-like radiative signature for the remainder of the spray pulse.

In Figure 7, the end of the spray pulse as indicated by the laser extinction measurement and the end of the soot-like signature are exceptionally well correlated in time. Our companion work (see Chapter 2) which has probed the fuel spray with visible and infrared lasers indicated that, for high soot loadings, visible wavelength lasers were not capable of distinguishing between the fuel spray and soot. The length of the spray pulse, as determined by the HeNe laser measurements in Figure 7 is 2.7 ms versus 2 ms in Figure 5. Careful examination of the injection pressure records for these two events indicates near-identical injection pulses. We therefore conclude that the laser extinction measurement does not accurately indicate the end of the spray pulse, and that soot is formed during the spray pulse and persists, at this location, after the spray. Careful examination of the intensities in Figure 7 provides further insight into the spray combustion process. Figure 7 includes an estimated spray pulse end time based on the observed 2 ms spray period in Figure 5. The estimated end of the pulse correlates well with a near-steady radiating time period. After the end of the spray, the intensity at all wavelength increases indicating increased temperatures, soot number densities, or both. At this time we cannot determine the radial location of the soot formation zone and cannot specifically differentiate between soot on the periphery and in the core of the spray. However, if the soot were primarily at the core of the fuel spray we would expect the disappearance of soot to coincide with the end of the spray, which it does not. The implication, therefore, is that soot is formed on the outer, low momentum, regions of the spray and the data of Figure 7 clearly indicate that soot does not follow the spray.

The increased radiance after the spray pulse end is interesting since it implies continued chemical reaction/combustion at this location after the end of the spray pulse. This

result is supported by the observation in our companion work (see Chapter 2) which examined the number density/cross section product using an infrared laser. Our results indicate substantial vaporization (up to 1/2 of the spray mass) occurs at locations 10 to 20 mm from the injector outlet. What we observe in Figure 7 is an indication that fuel, created during the spray pulse, is not immediately consumed but rather is present at a rate exceeding the local fuel consumption rate. This implies that at the end of the spray pulse a fuel rich zone exists which subsequently reacts to form soot or combustion products.

The data of Figure 7 indicate several distinct periods during the fuel spray pulse at a location 20 mm from the injector tip. The periods are: an initial period when the spray contains no apparent soot, but does include fuel vapor, a second period where the soot loading appears to be relatively steady, a period after the end of the spray where the soot loading and/or temperature increase, and, finally, a period in which H_2O is the dominant spectral signature.

Figures 9 and 10 illustrate results at a location 35 mm downstream of the injector tip. An initial period dominated by the fuel spectral signature is not evident in this plot. The early time spectrum included in Figure 10 does show a signature typical of dodecane, but another radiator in the $10\ \mu m$ region is clearly present. This is probably due to an intermediate product and it is interesting to note that carbon double bonds in straight chain hydrocarbons will produce absorption features near $10\ \mu m$ ¹⁴. The radiative signature quickly evolves to the continuum signature of soot and is constant within a factor of two after the estimated end of the spray pulse.

Figure 9 includes both the end of the spray pulse indicated by the HeNe laser measurement and the end estimated from the observed pulse in Figure 5. Note that the estimated end of the spray pulse correlates with a downturn in the radiance levels, indicating lower concentration or temperatures of soot. This is in direct contrast to the results from the spray tip where soot loading and or temperature were observed to increase at the end of the spray pulse. It appears that for this location, 35 mm from the spray tip

initial temperature, 894 K, that the local fuel consumption rate (via combustion reactions) was as large as the local fuel vaporization rate at this location. However, after the estimated end of the spray pulse, we still observe the characteristic spectral signature of soot.

The data of Figure 9 and 10 therefore indicate several distinct periods: a very short initial time period that includes a fuel vapor signature augmented with a $10\text{ }\mu\text{m}$ radiation feature, the remainder of the spray pulse with a relatively steady continuum radiance typical of soot, and a period after the end of the spray pulse with decreasing continuum radiance levels.

Finally, Figures 11 and 12 illustrate results 35 mm from the injector tip and 5 mm above the injector axis. These results are quite similar to those in Figures 9 and 10. The induction period is quite short, initially the signature of dodecane is apparent in addition to the radiator centered near $10\text{ }\mu\text{m}$. During the spray event the continuum signature of soot is quite evident and at times significantly after the end of the spray, the signature is primarily due to water. The results from the two locations 35 mm from the spray tip are interesting because the time duration that soot is present is substantially greater than that observed close to the spray tip. We observe "soot duration times" of approximately 1.3 ms 5 mm from the top, 2.3 ms 20 mm from the top, and greater than 3 ms 35 mm from the top. The increase in soot duration times as a function of distance from the injector indicates that soot may be transported away from the tip.

For all of these experiments, once the continuum signature is established, the radiation at $9.4\text{ }\mu\text{m}$ appears to be the least contaminated by other radiators. This wavelength can therefore be used to estimate the soot content in the spray as a function of time for the locations that were probed. This calculation will be an upper bound on the soot content since we cannot be certain that other radiators do not contribute to the signal at $9.4\text{ }\mu\text{m}$. For a line-of-sight emission measurement, the emissivity is defined as the ratio of the observed radiance to the radiance for a black body at the same temperature. Emissivity due to particulates is defined in Eq. (1).

$$\epsilon = 1 - e^{-N\sigma\ell}$$

where:

ϵ	=	emissivity
N	=	particle number density
σ	=	particle cross section
ℓ	=	path length.

The mass of soot along the line of sight is given by Eq. (2).

$$m = NA\ell V_p \rho$$

where:

m	=	soot mass
A	=	radiating area
V_p	=	particle volume
ρ	=	soot density (1.86 g/cm ³).

Combining these equations and solving for the mass of soot per unit area yields E

$$\frac{m}{A} = -\ln(1-\epsilon) \frac{\rho}{(\sigma/V_p)}$$

The term σ/V_p can be reduced to the ratio of absorption efficiency to particle radi shown in Eq. (4),

$$\frac{\sigma}{V_p} = \frac{Q_a}{\frac{4}{3} r} \quad (4)$$

where r is particle radius and Q_a is the absorption efficiency.¹² Figure 13 illustrates this ratio as a function of particle size as calculated using the complex index of refraction for soot and the Mie scattering solutions. At $9.4 \mu\text{m}$ the ratio is nearly constant for particle sizes up to $0.3 \mu\text{m}$ radii. Therefore, for soot with diameters less than $0.6 \mu\text{m}$, Eq. (6) provides a straightforward means of calculating the mass loading within the spray. However, to perform this calculation, the temperature of the spray must be known in order to determine the emissivity. For these experiments we did not make temperature measurements; however, a reasonable estimate for combustion systems is that the temperature lies between 1400 and 2000 K. Assuming that the temperature lies within these temperature bounds, we can determine the emissivity with an accuracy of $\pm 25\%$. The relative insensitivity of the radiance to temperature is a well established phenomena. Radiance is approximately proportional to temperature for wavelengths longer than the blackbody function maxima.¹⁸ The error in our calculation of soot mass, assuming no other radiators contribute to the $9.4 \mu\text{m}$ spectrometer channel, is therefore $\pm 25\%$.

Figure 14 illustrates results from this calculation for three different positions in the spray (for initial temperature of $875 \pm 20 \text{ K}$). The curves in this figure have been shifted to have the same spray arrival times. As one would expect, at positions further from the spray tip the soot mass loading is greater. In addition, the length of time that measurable soot is present is longer for positions further from the spray tip with significantly longer soot duration times at the location 35 mm from the spray tip.

Finally Figure 15 illustrates the soot appearance time which is defined as the time interval starting with the arrival of the spray and ending with soot mass climbing to 0.1 gm/m^2 . This appearance time was clearly shown in Figure 14 by the different initial times required for soot concentrations to rise above 0.1 gm/m^2 . These times are plotted on logarithmic scale versus the inverse of the temperature before injection. In all cases, the

FIGURE 13. - Ratio of absorption efficiency and particle radius as a function of particle radius.

FIGURE 14. - Soot mass in the spray as a function of time and position

FIGURE 15. - Soot appearance times in the spray.

induction periods (for each position in the spray) shorten with increasing temperature. As expected, the times decrease as the distance from the spray tip grows longer. Of particular interest is that this trend includes both the points moving away from the spray and the points that are 5 mm above the spray axis. Since soot is a product of high temperature pyrolytic chemistry, the obvious implication is that induction times for locations further from the spray tip are shorter simply because the temperatures are greater further from the tip, the fuel concentrations are greater, and/or the significant reaction occurs during the convection time between the various measurement points. The formation of soot requires combustion to generate the high temperature, fuel rich zones which are soot sources. Our observation therefore shows that high temperature fuel rich zones are first formed away from the spray tip with reaction/hot zones gradually approaching the injector outlet.

1.5. Conclusions and Recommendations

The results of this initial effort have demonstrated the use of infrared-based diagnostics in the region near the outlet of a diesel spray injector for combustion. We have developed the capability to monitor soot in the fuel spray, and have also demonstrated an ability to detect the fuel vapor in the spray. We did not monitor temperature of the various locations of the spray which limited the accuracy of our measurements to $\pm 25\%$. An obvious next step is to include a radiometric temperature monitor. At the present time we cannot conclusively differentiate between hot fuel and fuel vapor; however, our calculations indicate that the signals from the radiation must be for the vapor phase.

Soot was observed at all locations that were probed in the spray. However, the point closest to the spray tip, 5 mm from the outlet, soot did not appear until after the spray pulse. The soot appearance times were observed to depend both upon the temperature and position in the spray and the radiative behavior at the different locations. The spray indicated different behaviors for the different spray locations. At the location 5 mm from the spray tip, soot apparently was formed in a post spray pulse time. At late times in this event the radiative signature was replaced by that of water. At 20 mm from the axis, we observed an initial period in which fuel vapor is the dominant radiator, followed by a period in which soot was the dominant radiator. After the spray pulse, soot concentrations or temperatures increase followed by a time period with fuel vapor as the dominant radiator. For locations 35 mm from the spray tip, the initial period for soot formation was exceptionally short, followed by a steady soot loading. After the spray pulse, soot loadings appeared to decrease. In all cases, we have observed increased soot loadings and residence times with increased distance from the injector. An obvious extension of this work would be to use these techniques to make detailed comparisons of soot formation zones for different injectors and initial conditions.

2. A COMPARATIVE STUDY OF ROOM TEMPERATURE AND COMBUSTING FUEL SPRAYS NEAR THE INJECTOR TIP USING INFRARED LASER DIAGNOSTICS

2.1 Introduction

The diesel engine is a critical component of the U.S. transportation industry and its operation is dependent on the auto-ignition and combustion of a high pressure fuel spray that is injected into the combustion chamber. With increasingly stringent emission standards for diesel engine operation, an understanding of the fuel spray and its ultimate effect on the combustion process is necessary. Unfortunately, detailed studies of the fuel spray near the injector tip are exceptionally difficult using conventional optical diagnostics because of the large optical thickness for visible and ultraviolet wavelength light. In addition, studies of the fuel sprays must be performed at temperatures and pressures representative of diesel operation.

This paper reports results from an initial investigation of diesel sprays near the injector outlet using extinction measurements in both the infrared and visible wavelength regions. Injection pressures were similar to that used for diesel engines and the spray environment included room temperature and pressure air, room temperature and high pressure argon, and finally high temperature and pressure air. This study therefore spans a range of commonly used experimental conditions (i.e., room ambient conditions) and the conditions typical of a diesel at the time of injection. The comparison of these results is exceptionally illuminating in terms of the usefulness of room temperature and pressure experiments. In addition, we have successfully quantified the average droplet size in the fuel sprays at distances ranging from 5 mm to 35 mm from the injector tip.

The remainder of this paper presents the details of this study. We first include a brief summary of relevant literature regarding measurements methods and results for high pressure sprays. This is followed by a concise description of the experimental apparatus and its operation. The multiple color extinction measurement which was used to produce droplet

size information is discussed along with the implications of a droplet size distribution measurement. Experimental results are presented next for room temperature and sprays, sprays into a pressurized argon environment, and finally sprays into high (700 to 900 K) and pressure (2.5 to 4.0 MPa) air. The results are followed by a discussion of their implications and finally a set of conclusions for this initial study.

The study of fuel sprays, diesel engines, and measurement methods for fuel sprays has been an active area of research for at least two decades. A brief summary of the available literature on the subject must include diagnostic methods for fuel sprays and experimental results for this type of spray. The development of optical diagnostic combustion systems has included the use of methods for monitoring performance of fuel systems. Typically these measurements are concerned with quantifying the droplets produced in a spray and several techniques have evolved. These techniques can be categorized as single droplet techniques, ensemble averaging techniques, and image analysis techniques. Each has a range of applicability which produces very high quality results without imposing empirical corrections. The single droplet techniques are most commonly characterized by the phase Doppler type of measurements.¹⁹ This measurement technique uses the scattering response of spherical particles and uses sophisticated measurement techniques to ensure that measurements are only recorded when one droplet occupies the probe volume. Limitations for this measurement technique are a requirement for spherical shape, a dilute system that is dilute enough that only one droplet occupies the probe volume, a large dynamic range for the instrument, and a lower bound on the droplet size that can be measured (approximately 1 μm). For experimental environments that meet all of these restrictions, this instrument provides an excellent means of measuring droplet size distributions and velocities. For the dense regions of sprays the restriction on droplet sphericity and low number density makes this instrument of questionable utility. Martin²⁰ have performed phase Doppler measurements in the dense regions of room temperature and pressure sprays at high injection pressure and found that as many as 50% of the discrete measurements had to be discarded due to high number density or lack of sphericity. Very large numbers of droplets were observed near the measurement

size (in this case, 8.6 μm diameter) which suggests that a substantial portion of the droplet size distribution was omitted.

Droplet measurement techniques that are applied to a probe volume containing many particles are known as ensemble scattering methods. A common type of measurement is the diffraction based measurement which collects forward scattered light from the probe volume. This type of system is typically used for droplet diameters from 2 to 1000 μm ²¹ and it has been implemented on spray systems with very high obscuration levels.²² The instrument uses a series of detector rings to capture the forward scattering pattern and determines the size distribution of particles which best describes this scattering pattern. For sprays with high droplet number densities (such as diesel sprays), multiple scattering effects can become important and, for obscuration levels greater than 50%, empirical²² or theoretical²³ corrections must be applied to the results. Results from this work have indicated optically dense diesel sprays to have Sauter mean diameters between 5 and 15 μm . In addition to uncertainties introduced by optical thickness, diffraction based instruments are unable to resolve droplet diameters below 1 or 2 μm . For distributions including very small droplets, this will, as with the phase Doppler measurement, omit some of the droplets from the measurement which introduces a bias into the derived size distribution.

Other measurements which have been performed in the dense region near the injector tip include imaging measurements,²⁴ holography,²⁵ and imaging of pulsed laser sheets in a spray.²⁶ The imaging measurements were performed at high magnification ratio at the edge of the spray and droplets that were well separated from the spray cone could be resolved and their size determined.²⁵ The smallest droplet observed in this work was 3 μm in diameter and the theoretical measurement minimum was 1.7 μm . Sauter mean diameters for this system over a range of parameters ranged from 7 to 12 μm although the modeling effort for this study predicted much smaller droplets in the spray core.²⁵

Measurements of the dense region of diesel sprays are clearly difficult; however, the previously described methods and results indicate that droplet sizes may be relatively small,

i.e., less than $10\text{ }\mu\text{m}$ in diameter. Droplet size measurements are exceptionally in modeling predictions since vaporization rates and droplet lifetimes are strong function of droplet diameter. However, instrumentation used to determine the droplet size and distribution clearly can bias the observed sizes by simply omitting the small size class droplets that may be present. This result has not been completely resolved; in an excellent review of diesel engine technology,²⁷ droplet size data is presented that shows Sauter mean diameter less than $10\text{ }\mu\text{m}$ while the numerical simulations of sprays indicate droplet diameters of $50\text{ }\mu\text{m}$ or more. These large droplet sizes have been observed far from the injector tip in diesel like sprays.²⁸ However, regions far from the tip are of marginal importance for engine operation since injector to wall distances are small and, as our results show, the combustion zone is relatively close to the injector outlet. Clearly, considerable uncertainty still exists regarding the size of droplets in the diesel sprays, particularly in the dense region surrounding the injector tip.

2.2 Experimental Apparatus

The experiments were performed using a pulsed, high pressure, metering fuel injection system, a three laser extinction measurement system, and a shock tube to provide a high temperature and pressure environment for the combustor. The fuel injection system is based on an amplifying pump system designed by Oren.¹¹ The pump system is capable of high pressure injection (up to 80 MPa), produces a single pressure pulse when triggered, and when triggered produces the pressure pulse after approximately 100 μs . Injection pressures for this initial study were approximately 20 MPa for all data that is presented and the volume of fuel injected was approximately 120 mm^3 . The high pressure fuel pump was connected via stainless steel tubing to a Lucas injector and injector driver. The fuel injector was Lucas model BDLL150S6774 modified to have a single orifice (0.17 mm diam and 0.61 mm in length). Injector needle lift was set to 7 MPa and the injection pressure was monitored with a high frequency pressure transducer mounted on the delivery line between the pump and injector.

Optical diagnostics for this study were extinction measurements at three separate wavelengths: 0.633, 1.06, and 9.2 μm . These wavelengths were chosen to span a large wavelength range, to avoid known vapor absorption features in the fuel spray, and to coincide with readily available lasers. In addition the use of infrared wavelengths is particularly important since the optical depth of the spray for long wavelengths is smaller than that observed in the visible. Infrared lasers can therefore more effectively probe the dense regions of the spray than their visible and ultraviolet counter parts. The optical setup is shown in Figure 16. The lasers are co-aligned using a series of beam splitters and are carefully focussed to produce a 0.25 mm beam which passes through the spray. Lasers used for this study were a polarized 7 mW HeNe, a 2.5W Nd:YAG operated in single mode, and a 5W grating-tunable CO₂ laser operated at 9.2 μm . Beam splitting optics were carefully

FIGURE 16. - Schematic diagram of the three color extinction measurement setup.

arranged to exclude reflections from secondary interfaces and each beam was directed through the spray and onto a diffuser/detector system. The purpose of the diffuser was to mitigate the need for precise alignment required to make accurate measurements with unshielded detectors and to avoid beam steering effects from the spray. Detectors for the three lasers were a photomultiplier tube for $0.633\text{ }\mu\text{m}$, a silicon photodiode for $1.06\text{ }\mu\text{m}$, and a liquid nitrogen cooled HgCdTe detector for $9.2\text{ }\mu\text{m}$. Each detector included a bandpass filter to exclude radiation other than that from the laser. In addition, a fraction of the output from the CO_2 laser was directed onto a second detector before the spray. The CO_2 laser power level fluctuated gradually with time and signals from the HgCdTe detectors include a DC component. The zero level for the CO_2 laser measurement was determined by first chopping the laser beam and determining the relative signal level from the two detectors. By subsequently monitoring the power level in the CO_2 laser before the spray the zero level could always be calculated. Data acquisition for this experiment used a LeCroy/CAMAC system that included eight channels capable of up to 1 M operation. A personal computer was interfaced to this system for subsequent data storage and analysis.

The fuel injector was mounted in the endwall of a 38 mm diameter shock tube. For Room ambient tests were performed with the windows removed from the test section. For high temperature, high ambient pressure tests (1.1 MPa) were performed with window removed and the test section pressurized with argon. Combusting spray measurements were performed using the shock tube to produce a high temperature and pressure air environment similar to that produced in a diesel engine. The shock tube test section is square section (22.5 mm in width and height) and is specially constructed to include windows (19 mm high and 55 mm long). These windows are made of ZnS which is transparent from 0.37 to $14.0\text{ }\mu\text{m}$ and are positioned to provide full view of the injector tip. The spray tests are carefully controlled to produce injection into the quiescent, post-re shock region and test times for the temperatures used in this study ranged from 5 to 1000 K. Injection timing is carefully monitored to insure that injection occurs approximately 100 μs after the arrival of the incident shock at the end wall. Temperatures and pressure

for this study ranged from 700 to 900 K and 2.5 to 4.0 MPa. These conditions are similar to those in a diesel at top dead center of the compression stroke.

2.3 Extinction Measurement Theory

The focus for this study was to probe the dense region of the fuel spray in a non-traditional fashion. Our approach was to use infrared wavelengths which will effectively penetrate the spray if the droplet sizes are not large. Laser extinction measurements are relatively simple to implement; the attenuation of the beam by the spray is determined by measuring the propagated laser intensity with and without the spray. Equation (5) defines the measurement quantities and their relationship to the droplet cross sections in the fuel spray.

$$\ln I_0/I = N\ell \int_0^{\infty} n(D_p) \sigma(D_p) dD_p \quad (5)$$

where:

I_0, I	=	beam intensities before and after traversing the spray
$\sigma(D_p)$	=	extinction cross section for a particle with diameter D_p
D_p	=	particle diameter
ℓ	=	path length through the spray
N	=	droplet number density
$n(D_p)$	=	size distribution function for the droplets.

Fundamentally, extinction measurements determine the product of number density and droplet cross section over a known optical path. This equation does not include variations in droplet distribution along the path through the spray since we have not attempted to deconvolve the structure and droplet size as a function of radial position in the spray.

The simplicity of Eq. (5) allows the accuracy of the number density, cross product to be determined, as a function of transmission level, from the known m errors in I and I_0 . We have determined the measurement accuracy of I and I_0 and chosen $\pm 25\%$ as the maximum acceptable error level in the measurement of the number density, cross section product (i.e. $\ln(I_0/I)$). The actual measured quantities are limited by the error in each of these quantities limits the range of $\ln I_0/I$ which can accurately be determined. At very low extinction levels, I is indistinguishable from I_0 ; at high extinction levels, I is indistinguishable from zero. The focus for this work is the optically transparent environment. We have therefore examined the measurement accuracy for I and I_0 and determined maximum values for $\ln(I_0/I)$ consistent with our $\pm 25\%$ maximum error. Measured extinction levels greater than these maximum values will include an error greater than 25%. The values are 5.0, 3.7 and 3.0 for the HeNe, Nd:YAG, and Ar-ion laser measurements respectively.

The fuel spray is a very complex environment and may include droplets, vapor, and combustion intermediates and products. The extinction/absorption of each of these constituents can be important and must be accounted for in the interpretation of the measurements. For room temperature sprays, fuel vapor is the only other concern. The laser wavelengths have been carefully chosen to avoid absorbing fuel vapor. Dodecane was used as the fuel. We have measured the imaginary index of refraction for liquid fuel using visible and infrared spectrophotometers. The results were 1.0×10^{-7} , 4.4×10^{-7} , and 0.0012 at 0.633, 1.06, and 9.2 μm respectively. For the shorter wavelengths, we can crudely estimate the quantity of dodecane required to produce an absorption over a 20 mm path length to be greater than 1 MPa of dodecane vapor. At 9.2 μm this value is approximately 0.02 MPa but we believe the inferred value of the absorption cross section to be greatly overestimated due to the broadened absorption spectrum in the liquid state. In addition our results, as will be discussed, indicate small droplet sizes. Certainly for room temperature sprays the effect of fuel vapor absorption on the extinction measurement is small. At combustion temperatures we expect this observation to be true since (1) for the shorter wavelengths we did not observe an absorption band

would become important at higher temperatures, (2) spectrally resolved emission measurements found the region near $9.2\ \mu\text{m}$ in the combusting spray to be relatively free of molecular absorption features. Our conclusion is that for room temperature and combusting sprays, the effect of vapor absorption on the measured properties of the spray is secondary at the most.

Within the combusting spray, soot and combustion products and intermediates must also be considered as potential absorbers for the three lasers. Soot can definitely be an absorbing constituent for all three laser measurements and a discussion of its effect will be included in the results section. The primary products of combustion, H_2O and CO_2 , do not contain significant absorption bands at 0.633 , 1.06 (both CO_2 and H_2O have very weak overtone absorption systems in this region), and $9.2\ \mu\text{m}$ (a weak absorption system is present for CO_2 near $9.2\ \mu\text{m}$ but its effect would be negligible in the present system).²⁹ Intermediate molecular species are more troublesome to quantify. However, the region near $1.06\ \mu\text{m}$ lies conveniently between typical vibrational and electronic features. Although overtone absorption features will be present in this region, the absorption strength will not be significant for the spray system. At $0.633\ \mu\text{m}$ polycyclic aromatic hydrocarbons will absorb and we will, as with soot, address the possibility of molecular absorption in the results section. At $9.2\ \mu\text{m}$, spectral measurements performed in our laboratory have shown this wavelength region to be relatively free from molecular absorption effects. The extinction measurements in the combusting spray therefore must be interpreted carefully to insure that the possible effects from soot or polycyclic aromatics are taken into account.

The extinction response of fuel droplets to the three wavelengths may be used, in the absence of other absorption features, to determine the size of the droplets in the system. This measurement uses a ratio of the extinction measurements at two wavelengths to produce a quantity that is uniquely related to droplet size. Equation (6) illustrates that this reduces to a ratio of the size distribution/droplet cross section integrals.

$$\frac{(\ln I_0/I)_{\lambda_2}}{(\ln I_0/I)_{\lambda_1}} = \frac{\int_0^{\infty} n(D_p) \sigma_{\lambda_2}(D_p) dD_p}{\int_0^{\infty} n(D_p) \sigma_{\lambda_1}(D_p) dD_p}$$

where subscripts λ_1 and λ_2 indicate the probe wavelength. Number density and wavelength do not appear in this equation. For monodisperse systems, Eq. (6) immediately reduces to the ratio of droplet cross sections as a function of wavelength. Unfortunately, size distribution for the droplets must be considered since different measurement techniques will apply different weighting functions to the size distributions. In this case the weighting function at each wavelength is simply the optical cross section which varies with wavelength. To examine the optical response for the spray, we have used the log-normal distribution which is commonly used in fuel sprays¹⁷ and is described in detail elsewhere.³⁰ The log-normal distribution can be described with two parameters: the median diameter and the geometric standard deviation. Equation (7) defines the log-normal distribution in terms of particle diameter, D_p , median diameter, $\bar{D}_{pg}$, and geometric standard deviation, σ_g .

$$\frac{dn}{dD_p} = \frac{1}{(2\pi)^{1/2} \ln \sigma_g} \exp \left[-\frac{(\ln D_p - \ln \bar{D}_{pg})^2}{2 \ln^2 \sigma_g} \right]$$

Figures 17 and 18 illustrate this distribution for a number weighted and radius to the fourth power weighted mean diameter equal to 1 μm for different geometric standard deviations. The effect of the weighting function is clear: the radius to the fourth power weighted distribution includes a large number of sub-micron particles.

FIGURE 17. - Log normal distributions with a number weighted mean of $1 \mu\text{m}$.

FIGURE 18. Log normal distributions with a radius⁴ weighted mean of $1 \mu\text{m}$.

The optical cross section of the droplets as a function of size can be calculated using the well documented Mie scattering solutions¹¹ and the complex index of refraction of dodecane. As previously described we determined the imaginary index of refraction of dodecane at the three wavelengths. The real index is tabulated in the literature to visible wavelengths,³¹ and based on reflectance measurements at 10.6 μm we found the index in the infrared to 1.42. We therefore used 1.42 as the real index for all wavelengths. Using the log normal size distribution function and the calculated extinction cross section as a function of particle size, Eq. (6) can be evaluated numerically. Results are given in Figures 19 and 20 for the ratio of the 9.2 μm signal to the 0.633 μm signal. We used the ratioed measurement to approximately monitor the radius to the fourth power mean diameters up to 10 μm , the long wavelength extinction cross section increases approximately linearly with the sixth power of radius and the short wavelength extinction cross section increases linearly with the second power of radius. As shown in Figures 19 and 20, the diameter range for the mean diameter calculated with an r^4 weighted function is narrower than for the number weighted mean. The narrower range in mean drop diameter for a given signal ratio simply implies decreased sensitivity to the geometric standard deviation. This in turn indicates that, for an unknown size distribution, the indicated size corresponds roughly to a size based on an r^4 weighting function.

The computed signal ratio for 1.06 to 0.633 μm is given in Figure 21. The difference between the two signal ratios is exceptionally important. The ratio of 9.2 to 0.633 μm produces a steep droplet sizing curve. This implies that indicated droplet sizes will be relatively insensitive to measurement error. In contrast, the measurement ratio for 1.06 to 0.633 μm is much less steep and more sensitive to the geometric standard deviation of the distribution function. Clearly, the ratio for 1.06 to 0.633 μm signals will be less useful for determining droplet sizes.

This discussion has been based on the assumption that droplets in the near field are spherical. However, recent measurements of the depolarization ratio indicate that droplets may not be spherical and the assumed droplet shape is a prolate spheroid. A prolate spheroid is oriented to minimize drag, i.e., with the major axis approximately parallel to the flow direction.

FIGURE 19. - Ratio of extinction at $9.2 \mu\text{m}$ to $0.633 \mu\text{m}$ as a function of number weighted near diameter.

FIGURE 20. - Ratio of extinction of $9.2 \mu\text{m}$ to $0.633 \mu\text{m}$ as a function of radius⁴ weighted diameter.

FIGURE 21. - Ratio of extinction measurement for 1.06 μm to 0.633 μm versus radius⁴ weighted mean diameter.

spray axis. Solutions for prolate spheroid extinction are available³³ and in the near geometric limit (i.e., the 0.633 μm measurement) the cross section is similar to the sphere with an equivalent volume. For smaller size parameters, the extinction efficiency at oblique incidence does not display the large oscillations typical of head-on incidence and can be approximated with the volume equivalent sphere. Extinction measurements should be interpreted using spherical results in the presence of non-spherical droplets.

We also expect soot to appear in regions of the combusting fuel spray. The calculation procedure for fuel droplets has been repeated using the complex index of refraction for soot¹³ and the results are given in Figures 22 and 23. For particle diameters below 0.1 μm , the signal ratios are fixed at a constant value and the range of observation for the two shorter wavelengths is quite small. The clearest indicator of a droplet system is a small signal ratio for the two short wavelengths which cannot be produced by soot alone.

FIGURE 22. - Signal ratio, 9.2 μm to 0.633 μm , for a radius⁴ weighted diameter.

FIGURE 23. - Signal ratio, 1.06 μm to 0.633 μm , for a radius⁴ weighted diameter.

soot. In other cases, the signal ratios must be considered to determine if soot, or combination of both is being monitored.

2.4. Experimental results: Room temperature sprays

The previous discussion has focussed on interpretation of signal ratios for combustor spray. A much simpler system is the room temperature fuel spray; see the particle sizing curves in Figures 20 and 21 without the complication of soot. monitored extinction as a function of time in the spray for positions on the spray 10, 15, 25, and 35 mm from the injector tip. Sample results for the $9.2\ \mu\text{m}$ extinction measurement are shown in Figure 24. The results for 5, 10, and 15 mm from the little structure in the spray; we presume that for these distances a spray structure the time to develop and be discernable to the measurement system. However, 25

FIGURE 24. - Extinction ($9.2\ \mu\text{m}$) as a function of time and position for a room temperature spray.

from the injector tip, factor-of-three variations in the number density, cross section product are apparent. In all cases the spray is very optically dense with obscuration levels of 95% or more.

The ratio of extinction signals from the 9.2 μm laser to that from the 0.633 μm system can be used to monitor the mean droplet diameter, averaged over the width of the spray, at each of the axial locations. Results from this measurement are given in Tables 1 and 2. The tables include results 5, 10, 15, 25, and 35 mm from the injector tip and list the mean ratio for different time periods in the spray (initial or the start of the spray, the steady state portion of the spray, and the end or tail of the spray). In all cases the optical depth of the spray at the three wavelengths was monitored and time periods that included optical depths that implied errors in excess of $\pm 25\%$ were excluded. Tabulated values for the ratios are averages from several hundred microseconds during the spray. In theory, the combination of three measurements would allow the determination of the mean diameter and the width of the distribution. Unfortunately, this requires that the log normal distribution be a very accurate representation the actual distribution, and that the absolute accuracy of the two measurements be better than the 25% value which we have used. In general, the 9.2 to 0.633 μm ratios indicate droplets that are small, in most cases less than 8 μm in diameter. Qualitatively, this conclusion is supported by the 1.06 to 0.633 μm ratio.

The values tabulated in Tables 1 and 2 include mean ratio values for different time periods during the spray. In addition, we have assumed a geometric standard deviation of 1.5 and used the sizing function given in Figure 19 to determine the average droplet diameter. This average diameter, as previously discussed, is an average based on an r^4 weighting function. The indicated droplet diameters are for most cases less than 8 μm . The table also includes a maximum possible diameter for the first two positions in the spray. This diameter represents the maximum value for the diameter that the measurements could imply. The sizing curve used was for a monodisperse system (which indicates the maximum diameter for a given ratio) and an upper bound for the signal ratio was determined by taking the rms sum of the error in each of the extinction measurements. This error is

TABLE 1. - Room temperature and pressure spray results, injection pressure -

Run No.	$\frac{\ln(I_o/I)_{9.2\mu m}}{\ln(I_o/I)_{0.633\mu m}}$	Spray Time Period	Indicated Mean Diameter (μm) ($\sigma_g = 1.5$)	Maximum Possible Diameter (μm)
5 mm from injection tip				
1	0.7	Initial	5.8	
2	0.5	Initial	4.8	
2	0.5	Tail	4.9	
3	0.8	Initial	6.6	> 1
3	0.8	Steady state	7.0	> 1
4	0.5	Initial	5.0	
10 mm from the injector tip				
1	0.7	Initial	6.0	
1	0.7	Steady state	6.1	
2	0.6	Initial	5.4	
2	0.5	Tail	5.3	
3	0.6	Initial	5.5	
3	0.6	Tail	5.7	
4	0.6	Initial	5.7	
4	0.7	Steady state	5.8	

TABLE 2.- Room temperature and pressure spray results. injection pressure ~ 20 MPa

Run No.	$\frac{\ln(I_o/I)}{\ln(I_o/I)_{0.633\mu m}}$	Spray Time Period	Indicated Mean Diameter (μm) ($\sigma_g = 1.5$)	Minimum Diameter (μm)	Maximum Diameter (μm)
15 mm from the injector tip					
1	0.6	Initial	5.5	5.2	5.8
1	0.7	Steady state	5.7	5.5	5.9
2	0.7	Initial	5.8	5.6	6.1
2	0.7	Steady state	6.1	5.7	6.6
3	0.6	Initial	5.4	4.9	5.7
3	0.6	Steady state	5.7	5.4	6.0
25 mm from the injector tip					
1	0.6	Steady state	5.6	5.0	6.2
2	0.6	Steady state	5.5	5.0	6.0
3	0.5	Initial	5.1	4.8	5.5
3	0.7	Steady state	6.0	6.5	5.5
35 mm from injector tip					
1	1.1	Initial	> 9	6.5	> 9
1	0.7	Steady state	6	5.6	6.4
1	0.8	Tail	6.5	6.0	6.9
2	1.0	Initial	7.8	> 8.5	7.0
2	0.7	Steady state	6.3	5.6	7.0
2	1.0	Tail	7.5	6.8	> 8.5
3	0.7	Initial	6.0	5.4	6.8
3	0.6	Steady state	5.7	5.5	6.0
3	0.8	Tail	6.6	6.2	7.0

25% on a sample by sample basis, not on an average of samples and the error in a strong function of optical thickness. In many cases, a 25% error range is very conservative. The results indicate that for 2 out of 14 measurements the maximum diameter was above the effective measurement range. In all other cases, the maximum possible droplet diameter was less than $8.0\text{ }\mu\text{m}$.

The final three axial measurement locations, 15, 25, and 35 mm from the injector tip include a range in droplet diameters and optical thickness. The structure that is a function of time. For each of the measurements the range of droplet diameters in addition to the mean droplet diameters. The diameter range corresponding to the sizes is indicated by the standard deviation in the mean signal ratio over the averaging period. As before, the assumed geometric standard deviation for the log normal distribution was 1.5. The range of droplet diameters 15 and 25 mm from the injector tip was $\pm 10\%$ of the mean droplet diameter (typically between 5 and $6.5\text{ }\mu\text{m}$). At the measurement location 35 mm from the injector tip, the range of droplet sizes continued to be approximately $\pm 10\%$ of the mean diameter. However, the droplet diameters were larger, and for three of the data sets, the droplet size was too large to determine.

Our results for room temperature sprays therefore indicate that the average droplet diameter, assuming a weighting function proportional to the fourth power of radius, is in all cases less than $8\text{ }\mu\text{m}$. At the measurement location furthest from the injector tip the droplet sizes appear larger than in the locations closer to the tip. This trend implies that droplet coalescence is increasing the mean size, or that the very small droplets on the periphery of the spray do not have the momentum to propagate downstream with the spray.

Room temperature results also include two injection events with measurements 20 mm from the spray tip into a 1.1 MPa ambient pressure of argon. Results from these tests are included in Table 3. Unfortunately, the optical thickness of the spray was

TABLE 3. - Room temperature, 1.1 MPa background pressure injection pressure ~ 20 MPa

$\frac{[\ln(I_0/I)]_{9.2 \mu\text{m}}}{[\ln(I_0/I)]_{0.633 \mu\text{m}}}$	Spray Time Period	Mean Diameter	Minimum Diameter	Maximum Diameter
10 mm from injector tip				
0.6	Tail	5.8	4.9	6.8
20 mm from injector tip				
0.7	Tail	6.1	5.8	6.4
0.6*	Steady state	5.5*	3.9*	
*Upper bound values.				

noticeably larger in the pressurized environment and accurate measurements were only possible at the end of the spray event. As before we have assumed a geometric standard deviation for the distribution of $\sigma_g = 1.5$ and the measured diameters span a range from 4.9 to 6.8 μm . This table also includes data from a region of the spray where the thickness of the spray was above the limit set for the HeNe extinction measurement to be within 25%. We have used this region to place an upper bound on the droplet size in this spray region. We did not observe a systematic difference in the droplet sizes for the spray into atmospheric pressure gas versus the spray into a high pressure. Sizes for both systems were very similar.

2.5 Experimental Results: Combusting Sprays

A primary motivation for this work was to probe high pressure sprays in a diesel-like environment. Our spray injection system is integrated with a shock tube which can produce 700 to 900 K temperatures and pressures between 2.5 and 4.0 MPa. We have produced a data set, similar to the room temperature data which was previously presented, for sprays in a high temperature and pressure environment. These measurements used air as the test gas in the shock tube so that the spray will autoignite in the same fashion as the spray in a diesel engine.

Figure 25 shows results from a spray injection into 750 K, 2.5 MPa air with extinction set to probe 10 mm from the injector tip on the injection centerline. The difference between the 9.2 μm extinction at room temperature, Figure 24, and the extinction in this combusting spray is striking. At the start of the injection event (for approximately 0.2 ms) the two sprays are qualitatively similar but the spray in the high temperature environment is a factor of two less optically dense. The signal ratio for this period is 0.20 to 0.25 which implies droplet diameters of 3 to 4 μm (assuming a geometric standard deviation of 1.5 as we did for the room temperature spray). This implies a decrease from approximately 6 μm in diameter at room temperature to 4 μm for combustion conditions. However, as previously discussed, the interpretation of droplet sizing information is considerably more complex for the combusting spray environment since fuel vapor, molecular soot precursors, or soot particles may contribute to the extinction. For this initial work, we can infer the relative role of each of the possible interfering species. Fuel vapor may interfere with the 9.2 μm extinction signal. However, the spray is less dense and the apparent droplet sizes are smaller than observed at room ambient conditions. Additional absorption by fuel vapor at 9.2 μm would have the opposite effect i.e., signals indicating larger droplets and number densities. This observation, insignificant interference at 9.2 μm , is supported by the low emissivities (<0.017) observed in sooting periods for the fuel spray at 9.4 μm (see Chapter 1). These observations are consistent with those on the same spray and for similar conditions; the measurements for this work were emission measurements. Our preliminary conclusion is that fuel vapor does not significantly affect the 9.2 μm extinction in the initial portion of the spray.

Other possible interfering species are soot and soot precursors. We have previously measured soot initiation in the same spray and, for the initial spray regions soot concentrations were less than 0.1 gm/m^3 . Equation (8) relates mass per area in the spray to the product of number density, extinction cross section and path length and we find that the maximum contribution of soot to the extinction signals was 0.017 at 9.2 μm and 0.0633 μm .

FIGURE 25. - Combustion results 10 mm from the spray tip.

$$N \sigma_e \ell = \frac{m}{A} \frac{(\sigma_e/V_p)}{\rho}$$

where:

N	=	number density
σ_e	=	extinction cross section
ℓ	=	path length
m/A	=	mass per area for cylindrical probe volume $A\ell$
(σ_e/V_p)	=	extinction cross section to particle volume ratio, constant for small particles
ρ	=	density of soot, 1.86 gm/cm^3 .

These values were calculated using the complex index of refraction for soot¹³ and scattering solutions¹² to determine the extinction cross section to particle volume ratio for small particulate ($3.3 \times 10^3 \text{ cm}^{-1}$ at $9.2 \text{ }\mu\text{m}$ and $7.6 \times 10^4 \text{ cm}^{-1}$ at $0.633 \text{ }\mu\text{m}$). None of these soot extinction values is significant compared to the initial optical density in the spray.

Finally, the effect of soot precursors must be determined for the $0.633 \text{ }\mu\text{m}$ measurement. A primary means of identifying absorption by polycyclic aromatic hydrocarbons is to compare the ratio of extinction signals between 1.06 and $0.633 \text{ }\mu\text{m}$. As shown in Figure 21, if the signals are due to droplets, the signal ratio will remain relatively constant. Unfortunately, the time response of the $1.06 \text{ }\mu\text{m}$ system was significantly longer than that exhibited by the two laser/detector systems. However, in the early portions of the spray, the ratio of the signals is constant and since we would expect signals from soot precursors to be present from the beginning with time, we conclude that the $0.633 \text{ }\mu\text{m}$ extinction measurement is sensing primary droplets early in the spray. Based on this examination of the absorbing species in the spray system, we conclude that we can perform size determinations in the early portions of the spray by using the ratio of the $9.2 \text{ }\mu\text{m}$ extinction to the $0.633 \text{ }\mu\text{m}$ extinction.

After the initial steady spray period, the extinction level in the spray decreases and exhibits a time varying behavior. This "structure" in the spray was not apparent in the room temperature sprays at locations close to the injector tip. We attribute this structure to the combustion of the spray and have defined an ignition point as the time at which there is a discernable decrease in the $9.2\ \mu\text{m}$ extinction signal. Immediately after ignition the extinction signal at $9.2\ \mu\text{m}$ decreases to approximately 50% of its original value and varies by a factor of six or more for the remainder of the spray pulse. Since extinction at this wavelength is weighted by the sixth power of radius, the mass of droplets at this location in the spray has decreased to approximately 70% of the initial value and the large variation in the spray optical depth implies an axial structure in the spray. The signal ratio has also dropped by approximately 50% which implies droplet sizes of approximately $3\ \mu\text{m}$. Once again the effect of other absorbing species must be evaluated. For the post-ignition environment, we cannot rule out absorbing interference by soot precursors at $0.633\ \mu\text{m}$ and fuel vapor at $9.2\ \mu\text{m}$. The data given in Figure 25 show the soot appearance delay which we observe for this temperature in the spray and soot clearly is not an interfering absorber for most of the spray period. The ratio of signals is relatively stable which implies that the two signals are both responding primarily to droplets. However, we simply cannot rule out significant absorption by fuel vapor at $9.2\ \mu\text{m}$ in the post ignition environment and, for some of our higher temperature data, the extinction measurements at $1.06\ \mu\text{m}$ do not agree well with those at $0.633\ \mu\text{m}$. An obvious source of this disagreement would be absorption by soot precursors. Since both 9.2 and $0.633\ \mu\text{m}$ signals could be influenced by species other than droplets, the indicated droplet diameter of $3\ \mu\text{m}$, after ignition, must be considered to be approximate.

Figure 26 is an Arrhenius plot of the ignition delay times observed in the spray using the $9.2\ \mu\text{m}$ extinction measurement. This plot includes data for locations both 10 and 20 mm from the injector tip. The data show modest scatter typical of ignition delay data but do not display a location-specific bias. We defined ignition delay as the time period bracketed by the arrival of the spray and the first significant decrease in optical thickness in the spray.

FIGURE 26. - Ignition Delays in the Diesel Spray

We have also included soot initiation times for the location 20 mm from the injector which were determined by monitoring radiance at $9.4 \mu\text{m}$. The initial ignition and appearance of soot are well separated in time over the entire range for this experiment observation is important since soot can complicate droplet sizing measurements.

Figures 27 and 28 show the extinction measurements from two injector events at 860 K and 10 mm from the injector tip and at 760 K and 20 mm from the injector tip. These results show the same global trends indicated in Figure 25. The extinction at $9.2 \mu\text{m}$ is smaller than it was for the corresponding room temperature cases while the extinction level at $0.633 \mu\text{m}$ is still very high. Since approximately the same mass of fuel was injected, the droplet size must be smaller than observed for the room temperature experiments. Both data sets show signal ratios in the region before the onset of a

FIGURE 27. - Extinction results 10 mm from the spray tip for temperature = 860 K.

FIGURE 28. - Results 20 mm from the injector tip at 760 K initial temper:

spray that indicate droplet diameters near $3\text{ }\mu\text{m}$ which is smaller than observed in the room temperature sprays.

Figure 27 is at the same sampling location as Figure 25; the difference in these data sets is the initial temperature. Figure 27 is for an initial temperature 860 K versus 750 K for Figure 25. The higher temperature data set exhibits a shorter initiation time for the structured portion of the spray. As before, the $9.2\text{ }\mu\text{m}$ extinction is much more sensitive to changes in the spray and shows structures in the spray that are not apparent in the $0.633\text{ }\mu\text{m}$ extinction signal. The extinction signal at $9.2\text{ }\mu\text{m}$ can be grouped into three regions for this spray: (1) the relatively thick precombustion region of the spray, (2) an intermediate region starting near 0.5 ms and ending at 0.8 ms, and (3) a subsequent region of very low extinction levels. The intermediate region contains two distinct pulses and the onset of this region coincides approximately with the observed soot onset time. However, based on our observations of soot using infrared emission, the maximum soot level observed for this region of the spray was approximately 1 gm/m^2 which would produce an extinction level of 17% (at $9.2\text{ }\mu\text{m}$). The observed extinction levels are too large for these signals to be exclusively soot. The signal ratio for this region of the spray indicates droplet diameters of approximately $2.5\text{ }\mu\text{m}$ assuming a geometric standard deviation of 1.5. As before this result is somewhat qualitative due to the possible interferences from vapor and soot precursors. Following the intermediate region is a region of very low extinction level. This region does not correspond to the end of the spray pulse since the pressure signal for the spray indicates a typical pulse length between 1.5 and 2.0 ms. Extinction levels for the $9.2\text{ }\mu\text{m}$ measurement in the second time period indicate that over 60% of the spray mass (based on the initial spray extinction level) has been converted out of the liquid phase. This implies that for this high temperature spray event, a primary evaporation zone for the spray must be between the injector tip and 10 mm from the tip on the injector axis. The low extinction level in the $9.2\text{ }\mu\text{m}$ signal is accompanied by a precipitous drop in the $0.633\text{ }\mu\text{m}$ signal. For this region, the extinction levels are small enough for each of the signals that soot could be a dominant component for the measurement. We have therefore not attempted to interpret the droplet size in this region.

Figure 28 illustrates results 20 mm from the injector tip and for an initial temperature of 760 K. As before the data show an initially thick and non-reactive spray followed by a highly structured spray that has a significant mass of fuel in the vapor phase. Individual droplet sizes are similar to those of Figures 27 and 25: 3 μm diameters during the initial portion of the spray and approximately 2.5 μm after the spray develops structure. Compared to the results in Figure 27, the extinction level never falls to the 0.2 range or below at the sampling location being twice as far from the injection tip. This is an extremely interesting observation since, if evaporation of the spray in the near injector region is caused by the heat release from the combustion event, we would expect that, after ignition, the fraction of fuel in the liquid phase would decrease dramatically with increasing distance from the injector tip. We have observed something substantially different: the fraction of fuel in the liquid phase seems to decrease more strongly with increasing initial temperature. We can speculate that the location of the spray's high extinction zone is different for the two initial temperatures.

2.6 Conclusions

Several conclusions can be drawn from this initial study of room temperature combustor sprays. First, infrared wavelengths have been shown to be more effective than visible wavelengths at probing the near injector region of the fuel spray. At injection pressures of approximately 20 MPa into room ambient conditions, r^4 weighted droplet diameters (assuming a lognormal distribution with a geometric standard deviation of 1.5) were typically between 5 and 6 μm for positions 5, 10, 15, and 25 mm from the injector tip. The diameter was observed to increase slightly to the 7 to 8 μm range 35 mm from the injector tip. For room temperature sprays into a 1.1 MPa backpressure environment, we did not see a systematic change in the droplet diameter, but we did observe an increase in the product of droplet number density and optical cross sections.

Results for the combustor spray were very revealing. First, we found that the spray characteristics of the high temperature combustor spray to be significantly different than that of

temperature counterpart even for locations 10 mm from the injector tip. High temperature fuel sprays displayed a steady initiation period followed by a region of significant axial structure. Droplet diameters were observed to be near $3\text{ }\mu\text{m}$ which is slightly smaller than those in the room temperature case. A large fraction of the fuel mass in the spray, typically 50% or more, was observed to have left the liquid phase after spray ignition for a measurement location 10 mm and further from the injector tip. Initial temperature for the ambient environment was observed to control both the ignition delay for the spray and the fraction of the spray that evaporated near the injector tip. Finally, our results indicate that the region immediately surrounding the injector tip is very important to diesel operation. The fuel rich, pyrolysis zone has been known to be a source of soot for some time; our measurements show that the fuel vapor rich zone is within 10 nm of the injector tip.

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