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LASER-INDUCED SOOT VAPORIZATION CHARACTERISTICS IN THE LAMINAR DIFFUSION FLAMES

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ABSTRACT–The characteristics of soot vaporization induced by a high-energy pulsed laser were studied in an ethylene-air laminar flame. A system consisting of two pulsed lasers was used for the experiments. The pulse from the first laser was used to vaporize the soot particles, and the delayed pulse from the second laser was used to measure the residual soot volume fraction. Laser-induced soot vaporization was characterized according to the initial particle size distribution. The results indicated that soot particles could not be completely vaporized simply by introducing a high intensity laser pulse. Residual soot volume fractions present after vaporization appeared to be insensitive to the initial soot particle size distribution. Since the soot vaporization effect is more pronounced in the region of high soot concentrations, this laser-induced soot vaporization technique may be a very useful tool for measuring major species in highly sooting flame.

KEY WORDS : Soot vaporization, Laser-induced incandescence, Soot volume fraction, Particle size, Diffusion flame

1. INTRODUCTION

The presence of soot particles often hinders the measurement of major species (O_2, N_2, CO_2, H_2O, etc.) that are required to investigate the flame structure in most combustion environments (Eckbreth, 1977). Modeling the soot formation process in sooting flames has been limitedly developed to predict soot concentrations based on the experiments (Kennedy, 1997). Previous work has shown that soot particles in a flame can distort mixture fraction measurements by biasing the temperature due to the radiant heat loss effect from soot particles existing in the flame, particularly in the fuel-rich side (Kent *et al.*, 1987).

Soot volume fractions have been non-intrusively measured using light scattering, light extinction and the laser-induced incandescence (LII) techniques with great success (Santoro *et al.*, 1983; Quay *et al.*, 1994). However, very little data for major/intermediate species in sooting flames is available from non-intrusive measurements because of the difficulties caused by laser-modulated particulate incandescence (Eckbreth, 1977). One possible method for measuring major species in sooting flames is to use a high-energy laser pulse for vaporizing soot particles in the region of interest, followed by a second laser pulse for measuring species concentrations *via* scattering. In order to produce a meaningful species measurement, the first laser pulse must destroy soot particles without disturbing the flame condition or structure.

Dasch (1984) measured the variation in the transmission and scattering associated with vaporization and determined that vaporization does not affect the number of soot particles; it only affects the size of the particles. This indicates that the particles are shrinking due to vaporization but are not being completely destroyed. When a high intense laser is to vaporize soot particles, a certain limitation exists in which soot could not be completely vaporized. In other words, the measurement of major species, especially using Raman scattering technique that has inherently small signal cross-section over most species is still bothered by the presence of residual soot particles after vaporization.

The objective of this study is to characterize soot vaporization induced by the intense laser beam. Also, the limitation of soot vaporization is discussed regarding to initial soot particle sizes and thus, the feasibility of the Raman scattering measurement is discussed.

2. EXPERIMENTAL SETUP

The experimental apparatus involved a coannular laminar

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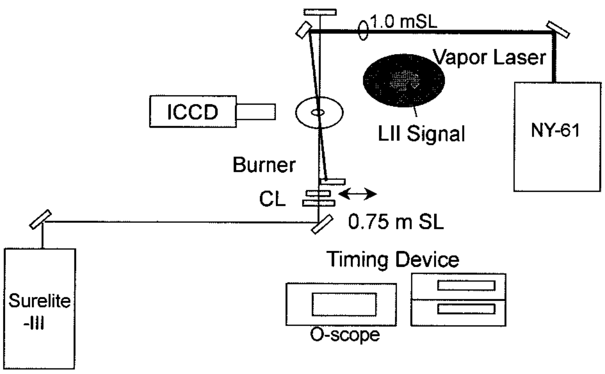

Figure 1. Optical arrangement for soot vaporization measurements.

diffusion flame burner that was identical to the burner employed in the previous study of soot properties (Santoro *et al.*, 1983). The fuel was chosen as ethylene with sufficient air environment in that the ethylene and airflow rates were 3.85 cm^3/s and 700 cm^3/s, respectively.

The schematic diagram of the optical arrangement for the soot vaporization is shown in Figure 1 using the two laser systems. The first laser system include Nd:YAG pulse laser (Continuum Model NY61-10), the output beam of which was *ca.* 7ns in duration and approximately 9 mm in diameter. The second laser system utilizes a frequency-doubled Nd:YAG pulsed laser (Surelite III, Continuum) operating at 10 Hz with a pulse width of 7 ns FWHM. A 285-mJ/pulse, 532-nm laser beam (Gaussian profile at far field) from the YAG laser was shaped into a 350-μm-thick (FWHM) laser sheet by a combination of a diverging cylindrical lens and a spherical lens. Laser (NY61-10) is first brought to vaporize soot particles and subsequently the second laser (Surelite-III) is fired with a certain delay time regarding to the first laser firing sequence. The timing sequence of the two laser systems was accurately controlled by the two pulse generators and continuously monitored by the oscilloscope. Intensified CCD camera (IMAX) was triggered from a synchronous trigger output from the second pulse laser.

In the measurement of residual soot volume fraction, the high intensity laser beam (vapor laser beam) intends to vaporize soot particles at the region of the focused beam in the flame and subsequently, the second laser (probe laser beam) produces the two-dimensional sheet to measure the soot field. The two-dimensional laser sheet for the soot volume fraction measurement was formed with combination of 0.75 m spherical and 5 cm cylindrical lenses in which the sheet beam is focused above the fuel tip. The preceding laser energy for soot vaporization is high enough to burn a hole through the soot field, when the laser crosses at 45° with respect to the second two-dimensional laser beam.

3. RESULTS AND DISCUSSION

3.1. Typical Image of Soot Vaporization

In order to observe soot particle vaporization in the flame, two-dimensional LII signal by a probe laser beam was obtained after a certain pulse delay of vapor laser beam. Soot vaporization zone are clearly observed as the hole-marker in the soot field as shown in Figure 2. The left side image demonstrates the original soot field collected without the vapor laser beam. The vaporization zone at the flame height of HAB 40 mm where high soot concentrations occurs can be seen in the middle of the figure. This soot vaporized zone in the middle of the flame is marked as the shape of the vapor laser beam profile crossing at 45° with respect to the second two-dimensional laser beam. Note that the vapor laser fluence to produce soot vaporization can be easily achieved once it is beyond a critical threshold value in which the LII signal starts to reach a saturation regime demonstrated by elsewhere (Dasch, 1984). It found the LII signal is linearly proportional to the laser fluence below certain threshold fluence while above this threshold the signal becomes saturated and maintains a nearly constant value (Dasch, 1984). From the images, it appears that soot particles apparently tend to be fully vaporized at both regions.

Since the LII signal tends to be often persistent for longer time, approximately an order of microsecond (Melton, 1984; Quay *et al.*, 1994), the signal affected by the vapor laser beam is here characterized, as does the initial particle size. Residual soot volume fractions as a function of pulse separation time are plotted in Figure 3 at the flame height of HAB 30 mm. Undisturbed soot volume fractions at the annulus and middle are 9.02 and 0.55 ppm, respectively. The pulse separation time below 5 μsec influences residual soot volume fraction since the signal by the vapor laser beam persists as background noise. The pulse separation time between 10 and 150 μsec indicates the minimum variations of the signal,

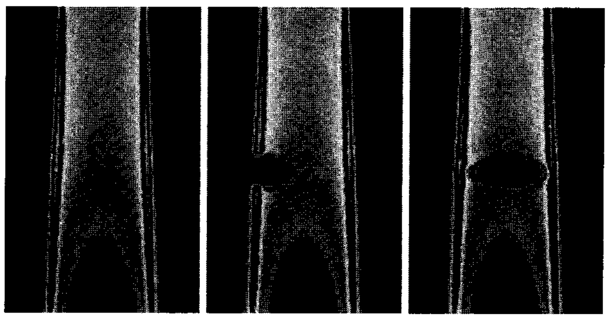

Figure 2. Typical images of soot particle vaporization. The height at the center of the "hole" produced by the high intense laser beam was 40 mm above a burner tip.

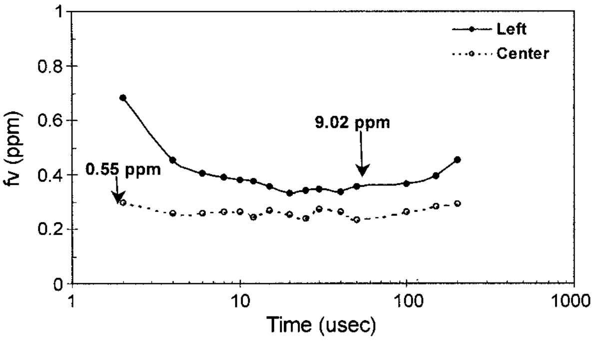

Figure 3. Residual soot volume fractions as a function of the pulse separation time along soot annulus and center of the flame at HAB 30 mm.

while the separation time beyond 200 μsec shows that the vaporized region disappears by the flow. Since the flow velocity is within an order of 1 m/sec, it takes time for the vaporized volume to be swept through the probe laser by the gas flow.

Figure 4 shows radial profiles of soot volume fraction after 12 μsec pulse delay at the flame height of HAB 40 mm. The two vaporized regions are located at the left side of the annulus and the middle in the soot field as illustrated in Figure 2. Relatively large amount of vaporization occurred in the annulus region while relatively small amount of vaporization occurs at the center region. The vaporization effect is significantly pronounced in the region of high soot concentrations. It can be expected that the degree of soot vaporization may be dependent of initial soot particle size since heat transfer process around soot particles exposed to the high intense laser beam is significantly dependent of initial soot particles. For smaller particles, the absorption is proportional to the volume, while the heat loss scales as surface area. Hence smaller particle is hard to vaporize. A limitation for vaporization of soot particles exists

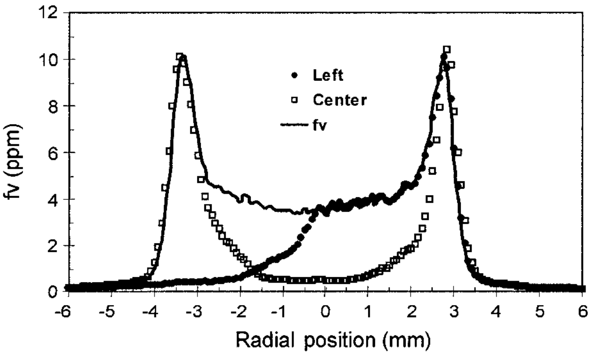

Figure 4. Residual soot volume fraction profiles across the flame at HAB 40 mm.

depending on the initial particle size distribution. This is very consistent with the measurement at different flame locations such that residual soot volume fractions were observed approximately 0.3 to 0.5 ppm when the original soot volume fractions are greater than 0.8 ppm.

The particle size effect on the vaporization is shown in Figure 5 with the radial profiles of soot volume fraction at various flame heights. The initial particle distribution can be estimated for this particular flame if the flame position is specified (Vander Wall, 1996; Ni *et al.*, 1995). If the vaporization region is located in the left annulus region as shown in the middle of Figure 2, the right side of soot annulus remains as the undisturbed soot field. As pointed out in Figure 3, residual soot volume fractions remain as the almost same levels whether undisturbed soot volume fractions are in high concentrations or not. According to the LII theory developed by some research groups (Melton, 1984; Tait *et al.*, 1992; Mews *et al.*, 1997), soot particles can be completely vaporized depending on the energy level of the laser regardless of the initial particle sizes. However, in the present study, the change of vapor laser energy does not affect much on the residual soot volume fraction and this is consistent with others (Dasch, 1984). In other words, residual soot volume fractions remain the same values although the laser power is increased. The published theory has lack of the detail explanation for rapid vaporization of soot particles as well as soot properties were treated as invariable.

Here a question arises whether residual signals come from the presence of tiny soot particles which survive over the vaporization or complex carbon molecules generated during the process of soot particle vaporization such as C_2 molecules. Supposed soot particles are vaporized completely, carbon compounds such as C_2 molecules are concurrently generated (Bengtsson *et al.*,

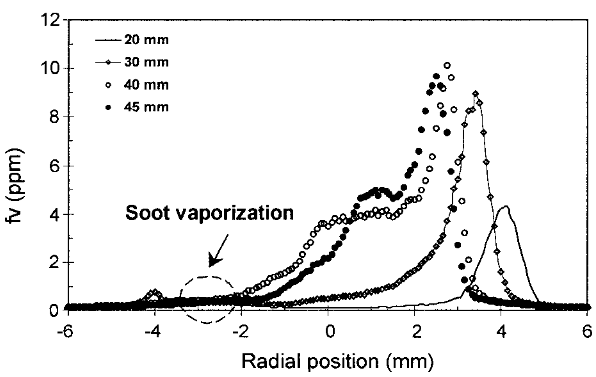

Figure 5. Radial profiles of residual soot volume fractions (left soot annulus) and undisturbed soot volume fraction (right soot annulus) as a function of initial soot particle size distribution.

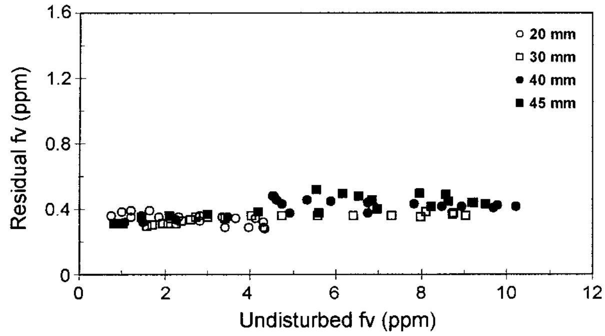

Figure 6. Undisturbed and residual soot volume fractions over the various flame heights.

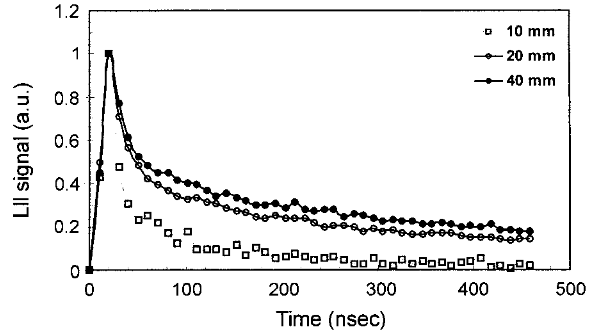

Figure 7. The time-resolved LII signal as a function of the particle size distribution.

1991; Hofeldt, 1993). Since the fluorescence signal from C_2 molecules was observed to be proportional to the LII signal, observed residual signal must be proportional to the undisturbed soot volume fraction if the soot particles are vaporized completely which is not the case in the present study as shown in Fig. 6. Residual signal appears to be independent of the undisturbed soot volume fraction. Thus, it is obvious that the residual signal results from the presence of tiny soot particles survival over the vaporization. More detailed explanation of soot vaporization should be provided with the theory supported by the experiment.

3.2. Time Evolution of LII Signal

The measurement of LII signal according to time evolution is made in Figure 7 as a function of the flame height and thus, the different particle size. Primary particle sizes at HAB 10, 20 and 40 mm are 20 nm, 40 nm and 80 nm respectively. For single spherical particle exposed to an intense laser energy filed, the energy gained by absorption of laser radiation, is balanced by heat loss through cooling, vaporization and radiation. The time evolution associated with the particle temperature history can therefore be separated into the time corresponding to the laser pulse duration and the subsequent time after the laser pulse (Shaddix, 1996; Smyth, 1997).

From a phenomenological point of view, when a single particle is exposed to laser radiation, the particle undergoes initially a rapid heat-up process until it reaches its vaporization temperature. Upon reaching the vaporization temperature, additional energy fluence causes full or partial particle vaporization. The final particle size is determined by the extent of vaporization. If the energy fluence is not high enough to achieve the vaporization temperature, rapid vaporization cannot take place and thus, the particle size changes very little. If the particle lies still under the Rayleigh limit during this period, it undergoes a transition period during which the temperature drops rapidly just before the end of the laser pulse. This occurs because energy fluence during this portion of the laser pulse is not sufficient to generate the LII signal. Shortly after the laser pulse, the temperature begins a more gradual decrease due to heat transfer to the surroundings. Melton (1984) pointed out that collisional cooling is a dominant energy loss path for soot particles below 3300 K, while above 3700 K, vaporization is dominant factor in the energy loss. Since the heat convection cooling term is much smaller than the vaporization term during the laser pulse, the evaporating particle mass depends only on the absorbed energy. The increase in LII signal during the laser pulse does not appears to be sensitive to the particle size while the signal after the laser pulse decreases exponentially and also depends on the initial particle size.

3.3. Raman Signal

Residual signal from the presence of soot particles survival over the vaporization is significant in terms of the measurement of Raman signal due to a very weak process, which typically limits its Raman scattering technique is not sensitive to quenching effects, scales linearly with pressure, species specific, and furthermore, dependence of the polarized incident light (Eckbreth, 1988). Since the LII signal produces the unpolarized signal of black body emission by the absorption of intense laser fluence, the Raman signal could be measured at the two different polarization angles by subtracting the horizontally polarized signal, *i.e.*, residual LII signal from the vertically polarized signal, *i.e.*, combination of the Raman scattering and residual LII signals. This laser-induced soot vaporization technique may be very useful tools for measuring major species in highly sooting flame since soot vaporization effect is more pronounced in the high concentrations of soot particle regions.

4. CONCLUSION

The soot vaporization characteristics rates were experimentally studied in laminar ethylene flames. The double pulse laser systems were fabricated to produce vaporization for soot particles using the first laser pulse, and subsequently measure soot volume fraction preceded by the second laser pulse, delaying the second laser beam. Laser-induced soot vaporization was characterized by the initial particle size distributions. It is found that soot particles could not be completely vaporized, simply by introducing the extremely high laser intensity. Residual soot volume fractions surviving over the vaporization tend to be insensitive to the initial particle size, which is approximately 0.3 to 0.5 ppm over the sooting flame. Vaporization effect is more pronounced in the large particle size. Laser vaporization of soot has possible applications to the measurement of major species in the highly sooting flame.

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