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13. ABSTRACT *(Maximum 200 words)*

The focus of this AASERT grant was the development of two novel measurement approaches for soot property characterization: laser-induced incandescence and intrusive probe sampling of soot particles from diffusion flames. Characterization of the laser-induced incandescence technique for soot particle measurements in laminar diffusion flames resulted in the development of a new quantitative measurement technique for soot volume fraction and marked the first use of this technique for quantitative measurements. In particular, the relationship between laser fluence and the temporal character of the laser-induced incandescence signal was carefully examined and documented. Based on the laminar flame work, the technique was extended to soot particle measurements in turbulent diffusion, spray and droplet flames with similar quantitative results. Concurrently, an intrusive probe sampling system to collect soot particles from laminar diffusion flames was developed. This sampling system provided collection sample sizes up to 2 grams of soot. Large soot samples were required to allow for appropriate surface area characterization which was conducted using a BET measurement approach. The results of these measurements showed a systematic decrease in the specific soot surface area as a function of residence time in the flame. The development of data of this type is important for fundamental studies of the processes controlling soot growth and oxidation in flames.

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Final Technical Report

on

Surface Reactivity of Combustion Generated Soot Particles

(AFOSR Grant F49620-92-J-0314)

For the Period June 1, 1992 through May 31, 1995

Prepared by

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Department of Mechanical Engineering
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Submitted to

Air Force Office of Scientific Research
Bolling Air Force Base
Washington, DC

October, 1995

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Summary

The focus of this AASERT grant was the development of two novel measurement approaches for soot property characterization: laser-induced incandescence and intrusive probe sampling of soot particles from diffusion flames. Characterization of the laser-induced incandescence technique for soot particle measurements in laminar diffusion flames resulted in the development of a new quantitative measurement technique for soot volume fraction and marked the first use of this technique for quantitative measurements. In particular, the relationship between laser fluence and the temporal character of the laser-induced incandescence signal was examined and documented. Based on the laminar flame work, the technique was extended to soot particle measurements in turbulent diffusion, spray and droplet flames with similar quantitative results. Concurrently, an intrusive probe sampling system to collect soot particles from laminar diffusion flames was developed. This sampling system provided collection sample sizes up to 2 grams of soot. Large soot samples were required to allow for appropriate surface area characterization which was conducted using a BET measurement approach. The results of these measurements showed a systematic decrease in the specific soot surface area as a function of residence time in the flame. The development of data of this type is important for fundamental studies of the processes controlling soot growth and oxidation in flames.

1.0 Introduction

Over the past decade, significant progress has been made in understanding the processes which control the formation, growth and burnout of soot particles in combustion systems. Because the presence of soot particles has significant effects on radiative transfer in gas turbine engines, combustor lifetime is seriously impacted by increases in soot formation as new engine technologies are developed. Consequently, AFOSR has had a continuing effort in the study of soot particle formation aimed at understanding the fundamental processes which control its formation, growth and burnout. This program has emphasized *in situ* diagnostics to study the chemical and physical mechanisms important in the formation and oxidation processes associated with soot particles in combustion systems. These studies have led to one of the most extensive data bases available on the effects of fuel structure, species concentration, operating pressure, residence time and temperature on the processes which control soot formation. The AASERT program described in this report complemented that program by adding a study investigating the surface reactivity of the soot particles as a function of these same parameters. An additional objective emerged during the program involving the development of a novel laser-based diagnostic technique for measuring soot particle volume fraction, size and number concentration. This technique, which is based on laser heating effects to detect and characterize soot particles, is termed laser-induced incandescence (LII). Progress on the characterization of this technique is also reviewed in this report.

The report which follows summarizes the results of this Augmentation Award for Science and Engineering Research Training (AASERT).

2.0 Research Objectives

The soot formation process in combustion systems can be broadly described as: (1) a precursor chemistry stage in which the large chemical species which lead to the first particles are formed; (2) an inception stage in which a large number of small particles are formed; (3) a surface growth and coagulation stage in which most of the mass is added and particle size increases dramatically; and finally (4) an oxidation stage in which particle burnout can occur. In the present research effort, it is the third stage, the surface growth process, which is of particular interest.

Recent studies[1-4] of the surface growth process for soot particles have come to a series of differing conclusions. Although it is generally believed that acetylene (C_2H_2) is the predominant surface growth species in terms of mass addition, the specific mechanism responsible for the surface reactions is not known. Conversely, there is some evidence to indicate that possibly large polynuclear aromatic hydrogen species (PAH) can also have a significant effect.[5] The current controversy centers on the role that the particle surface area has in the growth mechanism. Some experiments observe a direct dependence on particle surface area,[3] while others show little or no dependence.[4] Furthermore, in all combustion systems, as the soot particles age in the high temperature environment, they are observed to decrease in surface reactivity. Recent papers attempting to resolve this situation have focused on the concept of active sites[1,2] on the particle surface. It is then the number of these sites which controls the growth rate. As reactions occur at the active sites, they are removed and must be regenerated.[2] Thus, the lost of surface reactivity would be a result of a decrease in the regeneration mechanism. Some success has been achieved using this approach,[1,2] but there is no direct measurement support for the details of this mechanism.

Based on the above brief review of the current controversy regarding soot particle surface growth, the present study was aimed at developing measurement capabilities which allow direct characterization of the variation in the particle surface reactivity in terms of available surface area for laminar diffusion flames. In this study, a sampling technique capable of collecting large samples of soot particles was developed. Particles extracted from the flames were analyzed to determine the relative soot mass yield, and total surface area, using a BET technique, as a function of axial position in the flame. Such measurements yield information on the chemical radical activity of the soot particles[7] which we argue is related to the number of surface active sites. More specifically, the BET surface area measurements yield information on the manner in which the surface area available for reaction is changing. These measurements can be used to compare with optical light scattering measurements of this same quantity obtained for these flames in previous studies,[6] as well as for comparison with recent aggregate interpretations of that data.[8]

Because of the importance of optical techniques for the *in situ* measurement of soot particle properties, current efforts have also emphasized the development of a novel diagnostic technique, laser-induced incandescence (LII). This technique is based on earlier work by Melton[9], Dasch[10], Eckbreth[11,12], Dec et al.[13], and Tait and Greenhalgh.[14] The present studies have focused on quantitative *in situ* measurements of soot particle properties. The objective of this work was to establish the measurement capabilities for this technique in a wide variety of steady and unsteady combustion environments.

3.0 Research Accomplishments

3.1 Sampling of Soot Particles in Laminar Diffusion Flames

The significant results of this research include the following:

1) A sampling system capable of extracting soot from laminar diffusion flames burning a wide variety of fuels, including the well studied ethene and air flame, was developed and tested.

2) This sampling approach was proven to be reproducible and robust for the flame conditions examined.

3) Comparisons with optical measurements of the soot mass showed small systematic differences between the two approaches, but revealed that the general behavior of the soot mass profiles were similar.

4) The collection system was demonstrated to be capable of collecting up to a two gram soot sample size which could then be used in further characterization studies.

The basic design for this system involves a burner, a flame quench system, a dilution probe and collection system. The sampling system is briefly described below. Further details can be found in Reference 15.

Burner System

The burner is a Burke-Schumann type burner consisting of two concentric tubes, with fuel in the center and co-flowing air in the annular region (See Figure 1). Several different burner geometries were studied during the course of the project which varied only in the dimensions of the fuel and air passages. In order to study a wide variety of fuels and fuel mixtures, a vaporizer system was also developed and incorporated into the burner system. The sampling system was found to work adequately with all the burners studied.

Flame Quenching System

Initial work on the soot collection system lead to the observation that a screen placed at an axial cross section of a laminar diffusion flame would quench the flame at that particular

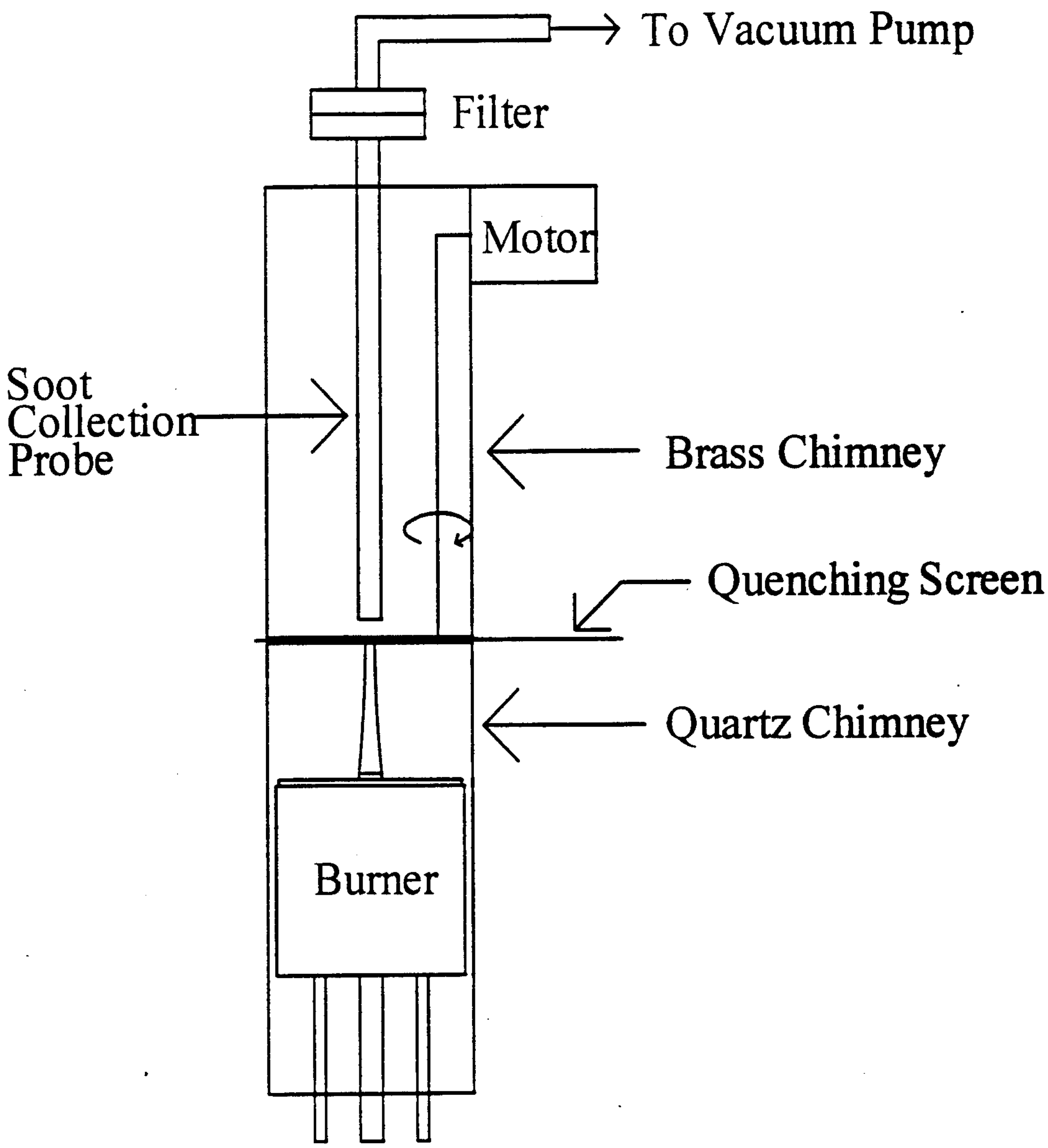

Figure 1. Burner and Soot Collection System

height. Above the screen, soot particles could be seen emitting from the quenched flame, which occurred without visually disturbing or varying the conditions of the flame below the screen. It was decided that this type of quenching method should be used for the soot collection system. The screen used in these experiments was made from type 304 stainless steel with a wire diameter of 0.017 inches and a mesh size of 0.044 inches.

Figure 1 shows a general description of how the flame quenching occurs. It was found that if the screen was left sitting over the flame for too long, the screen would become very hot at the quenching location, and soot would plug the cells. To avoid these problems, it was decided to slowly rotate the screen during the experiments. This procedure kept the soot from collecting on, and plugging one particular part of the screen. Soot collected on the rotating screen was removed using a wire brush as the screen exited the burner region.

Dilution Probe Design

The design of the dilution probe allowed for a combination of dilution, collection, and heat exchanger type cooling of the soot (See Figure 2). The probe was made of two concentric brass tubes with inner diameters of 3/8 and 1/2 inches respectively. The inner tube was connected to a filter holder and vacuum line which allowed soot and gases to be drawn into the probe. The two tubes were held in place by a brass fixture near the top of the probe along with a small ring cap at the entrance of the probe. The brass fixture allowed nitrogen to flow through the outer tube and down the probe, producing the heat exchanger effect on the soot and gases entering the probe. The nitrogen was also allowed to enter the vacuum flow of the inner tube through four small (1/16") holes in the inner tube near the bottom of the probe. The nitrogen entering the flow here would serve the purpose of diluting the particles and reactant gases, further cooling of the soot particles. The entire length of the probe was 24 inches to allow sufficient cooling of the soot and gases before entering the filter holder.

Vacuum and Collection System

The vacuum and collection system consisted of a 47 mm Belman Filter Holder, 3 meters of 3/8 inch copper tubing, a Magnehelic pressure gauge (0-100 inches H_2O), a critical orifice to control the flow rate, and a vacuum pump (See Figure 2).

The pressure gauge was used to read the pressure drop across the filter (from atmospheric pressure) which was an indication of the amount of soot collected on the filter. The 3 meters of copper tubing allowed for the gases in the vacuum line to cool to ambient conditions at the critical orifice. The vacuum pump operated at 1/10 of ambient pressure assuring a choked flow through the orifice and a constant mass flow rate through the vacuum line.

Two types of filters have been used in collecting the soot in this project. Both filters were 47 mm diameter Pallflex Teflon Coated Glass Fiber Filters. The filters used for most of

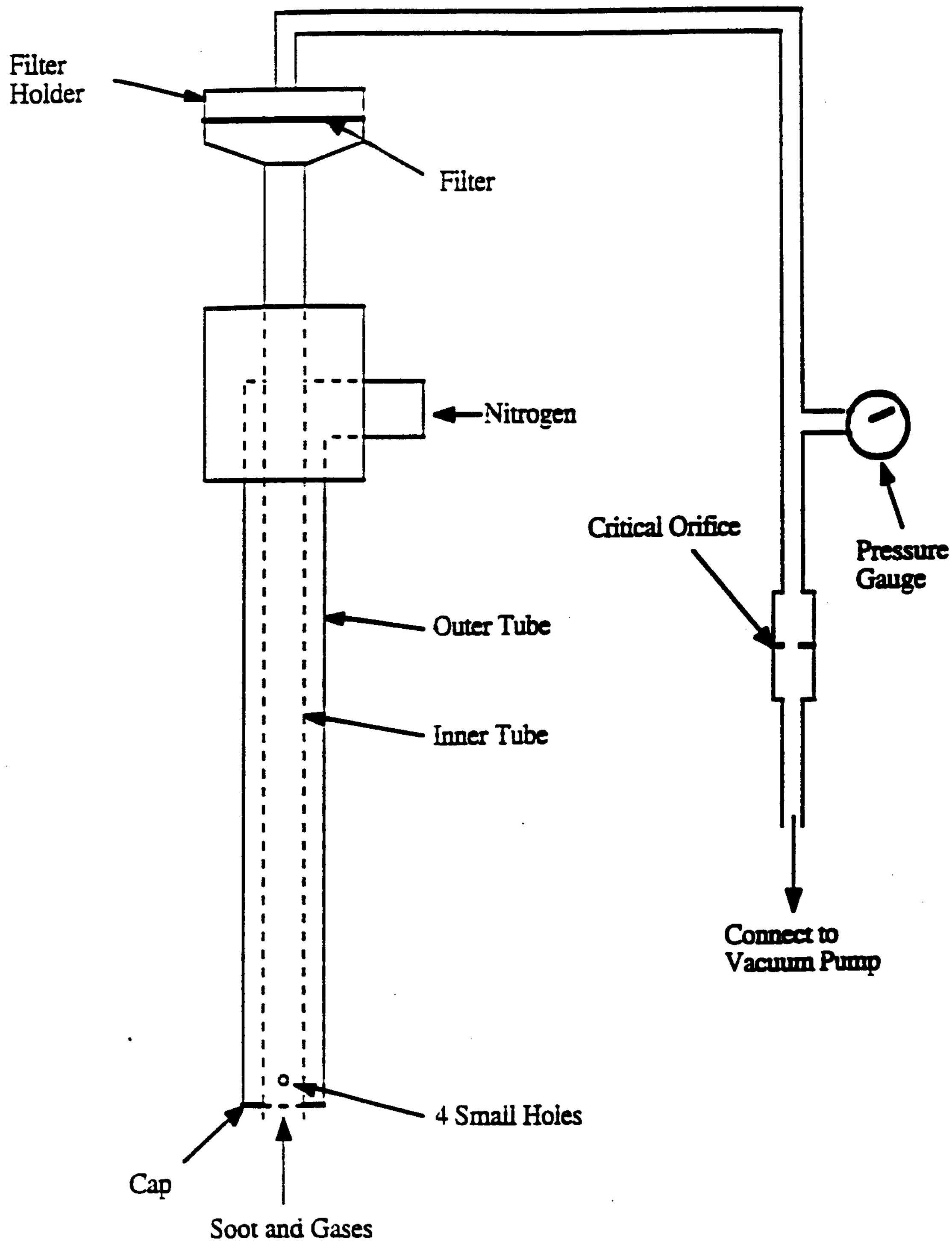

Figure 2. Dilution Probe and Collection System

the experiments were type T60A20 and have a reported collection efficiency of 70 to 80% for 0.035 μm particles and at least 95% for particles with diameters of 0.3 μm and larger. The other filters used in some of the experiments were type TX40HI20 and had a reported collection efficiency of 98 to 99% for 0.035 μm particles and 99 to 100% efficiency for particles with diameters of 0.3 μm and larger.

Surface Area Measurement Results

In order to assess the effects of fuel composition on soot surface area, samples obtained from a series of fuel mixtures were analyzed using the BET technique. Table 1 indicates the fuel mixtures studied while Table 2 indicates the resulting surface area measurements. The fuel mixtures selected produced copious amounts of soot and, thus, large soot samples could be collected easily. The two mixtures differed in the amount of aromatic content (meta-xylene). In Table 2 the average specific surface area in m^2/g is shown for the two mixture for selected heights in the flame. For mixture 1 the effect of prolonged (i.e. 24 hours) heating of the sample was evaluated and found to produce a measurable change in the specific surface area. In the case of mixture 2, several heights were sampled and a comparison of the variation of the specific surface area is plotted in Figure 3. A slight decrease in the specific surface is observed which would be consistent an annealing of the particles with increased residence time. However, the trend is slight and further measurements are warranted before firm conclusions can be drawn.

Based on the above observations, it can be stated that a suitable sampling technique has been developed for investigating surface related properties of soot particles. This sampling system provides suitable quantities of soot for application of a variety of analysis techniques. Future work should emphasize applying a variety of techniques to samples to determine systematic changes in the surface properties of soot extract from flames as a function of operating conditions and fuel composition.

Table 1 Fuel

	Liquid Volume Percent			
Mixture Number	Hexadecane	Heptamethyl-nonane	Meta-Xylene	Di-Tert-Butyl Disulfide
1	56.4	37.6	2	4
2	39.6	26.4	30	4

Table 2 Date from BET Experiments

Mixture Number	Flame Height (mm)	Average Specific Surface Area (m^2/g)	Percentage Change from Original (%)
1	40	46.82	
1*	40	34.22	-26.91%
2	15	44.44	
2	30	40.42	
2	40	39.46	
2	50	40.46	
2	60	37.94	

*Note: This sample was reheated in the vacuum oven for 24 hours prior to re-evaluation of its surface area.

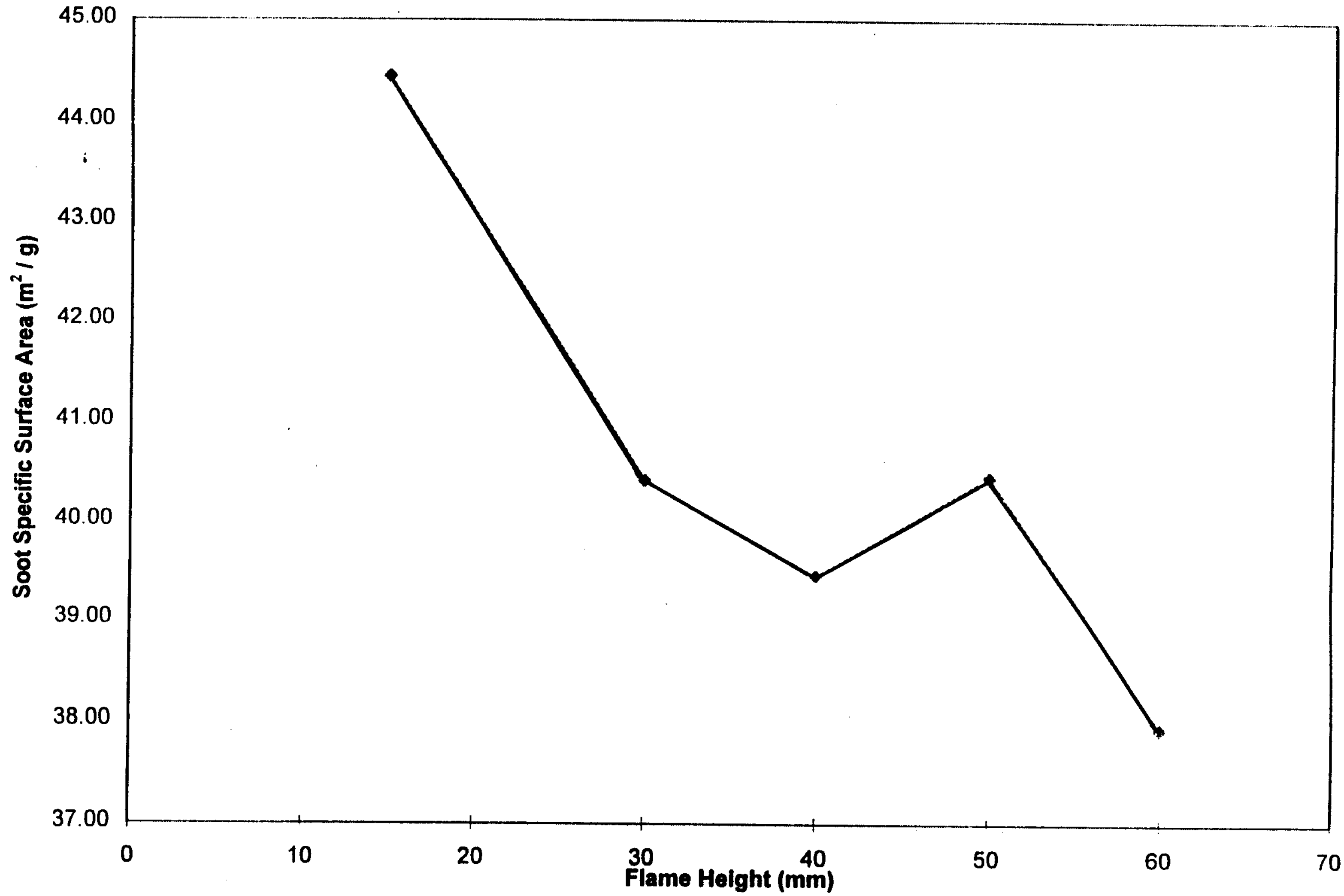

Figure 3. Soot Specific Surface Area Versus Flame Height for Mixture 2

3.2 Laser-Induced Incandescence Studies

The studies of laser-induced incandescence (LII) have focused on soot measurements in laminar diffusion flames. The results of these studies have recently appeared in an article published in *Combustion and Flame* (see Attachment A) with more recent studies of two-dimensional imaging capabilities to appear in Applied Optics[16].

Based on this study, the following conclusions concerning the LII diagnostic of soot volume fraction are made:

(1) Laser-induced incandescence has been used to obtain spatially-resolved measurements of soot volume fraction in laminar diffusion flames, in which comparisons with laser scattering/extinction data yield excellent agreement for both radial profiles and integrated volume fraction. Thus, laser-induced incandescence can be used as an instantaneous, spatially-resolved diagnostic of soot volume fraction without the need for the conventional line-of-sight laser extinction method.

(2) The temporal characteristics of the laser-induced incandescence signal is observed to involve a rapid rise in intensity followed by a relatively long (ca. 600 ns) decay period subsequent to the laser pulse, while the effect of laser fluence is manifest in non-linear and saturated response of the laser-induced incandescence signal with the transition occurring at a laser fluence of approximately 1.2×10^8 W/cm^2 for laser pulse of ca. 7 ns in duration.

(3) Spectral response of the laser-induced incandescence involves a continuous spectrum in the visible wavelength range due to the blackbody nature of the emission, where the spectral response for 300-450 nm wavelength range indicates a soot surface temperature of ca. 5000 K with the spectrum continuing at a nearly level intensity up to 750 nm wavelength due to the multiplicity of the soot particle sizes in the probe volume.

(4) Simultaneous measurements of LII and the vertically-polarized light-scattering yield encouraging results concerning the mean soot particle diameter and number concentration; thus significant applications exist in two-dimensional imaging and simultaneous measurements of laser-induced incandescence and light-scattering to generate a complete soot property characterization.

4.0 Conclusions

For this AASERT grant, "Surface Reactivity of Combustion Generated Soot Particles" the following results have been achieved:

1) A sampling system for the extraction of soot particles from laminar diffusion flames has been developed and tested.

2) A burner system has been developed which incorporates a vaporizer and heated flow system to allow studies of temperature effects and liquid fuels species on soot surface reactivity.

3) The sampling technique was demonstrated to be capable of collecting quantities up to 2 grams. These samples have been used to demonstrate that specific area measurements of the collected soot particles can be obtained.

4) The LII technique has been demonstrated to be capable of quantitative soot volume fraction measurements in laminar and turbulent diffusion flames as well for single droplet flames.

During this AASERT award Mr. Bryan Quay, a graduate student in Mechanical Engineering, has been supported under the grant. Mr. Quay has worked on both the soot sampling and LII experiments. Mr Matthew Schneider assisted Mr. Quay as part of an NSF summer research program. Mr. Chris Chandler has participated in the project under support from Lubrizol, an oil and fuels additive company. Mr. Chandler's research is complementary to the AASERT objectives and illustrates the dual-use aspects of the research.

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6.0 Publications

Quay, B., Lee T-W, Ni, T. and Santoro, R. J., "Spatially-Resolved Measurements of Soot Volume Fraction Using Laser-Induced Incandescence," *Combustion and Flame*, 97:384-392 (1994).

Ni, T., Pinson, J. A., Gupta, S. and Santoro, R. J., "Two-dimensional imaging of soot volume fraction by the use of laser-induced incandescence," *Applied Optics*, in press.

7.0 Meetings and Presentations

"Soot Formation Workshop," AFOSR Contractors Meeting on Air Breathing Combustion, Atlantic City, NJ, June 14, 1993.

"Measurements of Soot Growth Species Concentration In Diffusion Flames," 1993 Technical Meeting of the Eastern States Section of the Combustion Institute, Princeton University, Princeton, NJ, October 25-27, 1994.

"New Techniques for Quantitative, Planar Soot Measurements," 1993 Technical Meeting of the Eastern States Section of the Combustion Institute, Princeton University, Princeton, NJ, October 25-27, 1994.

"Spatially-Resolved Measurements of Soot Volume Fraction Using Laser-Induced Incandescence," 1993 Technical Meeting of the Eastern States Section of the Combustion Institute, Princeton University, Princeton, NJ, October 25-27, 1994.

"Modeling and Measurements of Soot and Species in a Laminar Diffusion Flame," The 1994 Western States Section Meeting of the Combustion Institute, University of California/Davis, Davis, CA, March 21-22, 1994.

8.0 Participating Personnel

Dr. Robert J. Santoro, Professor of Mechanical Engineering
Mr. Bryan Quay, Graduate Student, Department of Mechanical Engineering (Ph.D. expected December, 1996)
Mr. Chris Chandler, Graduate Student, Department of Mechanical Engineering (M.S. May, 1995), supported by Lubrizol - 1994-1995
Mr. Gregory Davis, Undergraduate Student, Department of Electrical Engineering
Ms. Vicki Jacobs, Undergraduate Student, Work Study Lab Assistant - 1992/93
Mr. Matthew Schneider, Undergraduate Student, supported under the NSF Undergraduate Summer Research Program in Mechanical Engineering - 1993
Mr. Jed Bailey, Undergraduate Student, supported under the NSF Undergraduate Summer Research Program in Mechanical Engineering - 1994
Dr. Tom Ni, Post-doctoral fellow
Mr. Daniel Boone, Technician, Department of Mechanical Engineering

9.0 Interactions

Lubrizol, Wickeliffe, OH - Drs. Ralph Kornbrekke and Dan Daly

A project under the support of Lubrizol was initiated as of January, 1994 to undertake a collaborative research program to examine the surface reactivity of soot particles which are formed under conditions of interest to Diesel engine combustion. The objective of this research is provide appropriate characterization of the soot particle surface to allow development of appropriate additives which would suppress the deleterious effects of soot particles on engine lubricants.

10.0 Inventions

No inventions have resulted during the second year of this program.

Attachment A

Spatially Resolved Measurements of Soot Volume Fraction Using Laser-Induced Incandescence

by

B. Quay, T.-W. Lee, T. Ni, and R. J. Santoro

Reprinted from *Combustion and Flame* 97:384-392 (1994)

Spatially Resolved Measurements of Soot Volume Fraction Using Laser-Induced Incandescence

B. QUAY, T.-W. LEE, T. NI, and R. J. SANTORO*
Department of Mechanical Engineering, The Pennsylvania State University, University Park, PA 16802

Laser-induced incandescence is used to obtain spatially resolved measurements of soot volume fraction in a laminar diffusion flame, in which comparisons with laser scattering/extinction data yield excellent agreement. In addition, the laser-induced incandescence signal is observed to involve a rapid rise in intensity followed by a relatively long (ca. 600 ns) decay period subsequent to the laser pulse, while the effect of laser fluence is manifest in nonlinear and near-saturated response of the laser-induced incandescence signal with the transition occurring at a laser fluence of approximately 1.2×10^8 W/cm^2. Spectral response of the laser-induced incandescence involves a continuous spectrum in the visible wavelength range due to the blackbody nature of the emission. Simultaneous measurements of laser-induced incandescence and light scattering yield encouraging results concerning the mean soot particle diameter and number concentration. Thus, laser-induced incandescence can be used as an instantaneous, spatially resolved diagnostic of soot volume fraction without the need for the conventional line-of-sight laser extinction method, while potential applications in two-dimensional imaging and simultaneous measurements of laser-induced incandescence and light-scattering to generate a complete soot property characterization are significant.

INTRODUCTION

Formation, growth, and oxidation of soot particles in diffusion flames involve a complex interaction between chemistry and fluid mechanics. The understanding of these chemical and physical processes is important not only from a fundamental scientific standpoint, but also due to their applications in practical combustion devices. For example, soot emission from automotive and gas turbine engines constitutes one of the major pollutants that needs to be minimized, while excessive soot formation and radiation in propulsion devices have adverse effects on combustor and flow components. In this regard, sooting characteristics of both turbulent and laminar flames have been investigated by numerous researchers, while in this laboratory attention has been focused on axisymmetric laminar diffusion flames. The soot property measurements made in this flame, thus far, involve the laser scattering/extinction method. This technique yields measurements of soot volume fraction, mean soot particle diameter, and number density after tomographic inversion of the laser extinction data due to the line-of-sight nature of these measurements.

However, recent studies of a process involving laser-induced incandescence (LII), in which the soot particles are heated up by the laser energy and emit blackbody radiation or incandescence at elevated temperatures, have shown that LII can be used as a nonintrusive spatially resolved soot diagnostic [1–6]. In particular, it has been pointed out by Melton [1] that the LII signal is nearly proportional to the local soot volume fraction for sufficiently large laser fluence; thus, a pointwise measurement of soot volume fraction can be made without the need for the line-of-sight laser extinction and time-consuming tomographic inversion method. While other applications of LII in soot diagnostics, for measurements of soot particle size distribution for example, have been suggested [1], the most direct and significant application of LII may be in obtaining point measurements of soot volume fraction since the line-integral nature of laser extinction and subsequent tomographic inversion technique have deficiencies in some laminar flame and most

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turbulent flame configurations. For example, instantaneous measurements of local soot volume fraction can be made in turbulent diffusion flames using LII without being limited to time-averaged data or an axisymmetric burner geometry. Furthermore, applications of LII in investigations of soot properties include two-dimensional imaging of soot volume fraction distributions and simultaneous LII and light-scattering measurements to construct a complete soot property characterization. Recently, qualitative information on soot formation under Diesel engine conditions have been reported in which simultaneous two-dimensional LII and light-scattering images were obtained [5, 6].

In spite of these potentially significant applications of LII in soot diagnostics, no experimental verification of the LII technique for determining the local soot volume fraction has been reported to date. The objectives of this investigation, therefore, were to experimentally determine the applicability of the LII method to obtain spatially resolved measurements of soot volume fraction, to study the feasibility of making simultaneous LII and light-scattering measurements to obtain a complete soot property characterization, as well as to investigate the detailed characteristics of LII in laminar diffusion flames.

LASER-INDUCED INCANDESCENCE

Laser-induced incandescence involves the heating of soot particles to temperatures above the surrounding gas temperature due to the absorption of laser energy, and subsequent detection of the blackbody radiation corresponding to the elevated soot particle temperature. The temperature of the soot particle is determined by the rate of laser energy absorption, conductive heat transfer to the surrounding gas, soot vaporization, and radiative heat loss through blackbody radiation [1, 2]. For example, a Nd:YAG pulsed laser beam of ca. 7 ns duration used in the present LII measurements represents an energy source in the energy balance equation, and the soot particle temperature rapidly rises during the duration of the laser pulse as the soot particles absorb the laser energy. The heat sink terms in this phase are the conductive and radiative heat losses to the surrounding gas, which are much smaller than the laser energy absorption rate for laser fluence levels relevant to LII. Near a soot particle temperature of ca. 4000 K, which is close to the soot vaporization point, the temperature rise is severely curtailed by the energy expended in the vaporization of soot particles [1], although soot surface temperatures as high as 5000 K have been observed for sufficiently large laser fluence [7]. Subsequent to the laser pulse, the temperature of the soot particles gradually decreases due to conductive and radiative heat losses.

The intensity of the LII, or the blackbody radiation due to laser heating, for a single soot particle has a dependence on the soot particle temperature, detection wavelength, and the laser fluence. The total incandescence emitted from the soot particle surface has a fourth-order dependence on the soot particle temperature. The spectral shape of the incandescence is determined by Planck's law with the maximum in the blackbody radiation occurring at a wavelength inversely proportional to the soot particle temperature according to Wien's displacement law. Thus, the temporal variation in the LII signal at a given detection wavelength qualitatively follows the soot particle temperature in time, with the exact functional relationship determined by the processes described above.

Computations of the LII in response to an idealized laser pulse based on the above blackbody radiation laws and the soot particle energy balance have been performed by Melton [1] and Tait and Greenhalgh [4]. In particular, in the limit of high laser power and maximum soot particle temperature near its vaporization point, Melton [1] has shown that the intensity of the LII signal for a group of soot particles has a dependence on mean soot particle diameter raised to the power of $(3 + 0.154\lambda_{det}^{-1})$, where λ_{det} is the detection wavelength expressed in microns. For λ_{det} between 0.7 and 0.4 μm, for example, the LII signal is proportional to the mean soot diameter raised to the power of 3.22 to 3.38, or approximately to the soot volume fraction. This forms the basis for the current approach of using LII for pointwise measurement of soot volume fraction.

EXPERIMENTAL METHODS

The experimental apparatus involved a coannular laminar diffusion flame burner that was identical to the burner employed in this laboratory in previous studies of soot properties [8–10]; thus, only a brief description will be given here. The overventilated laminar flame burner consisted of two concentric brass tubes with fuel and air flowing through the inner (11.1 mm i.d.) and outer (101.6 mm i.d.) tubes, respectively, where the fuel tube extended 4 mm beyond the exit plane of the air tube. Flow conditioning for the air was achieved via a layer of 3.0-mm glass beads, a series of wire screens and ceramic honeycomb section, while the fuel passage contained a layer of 3.0-mm glass beads and a single wire screen. A 405-mm long brass cylinder that fit over the outer tube was used as a chimney to shield the flame from laboratory air disturbances; and optical access was obtained through machined slots on the brass cylinder which traversed with the burner assembly. In addition, screens and a flow restrictor were placed at the chimney exit to achieve a highly stable flame similar to previous studies [8–10]. The traverse system involved a stepper motor and controller (Daedal PC-410-288) that provided positioning capability with a resolution of 0.0127 mm.

The optical setup for the LII included an Nd:YAG pulse laser (Continuum Model NY61-10), the output beam of which was ca. 7 ns in duration and was focused to approximately 0.38 mm in diameter using a bi-convex lens of 400 mm focal length. The probe volume location was displaced ~ 25 mm from the focal point of the lens in order to achieve this 0.38 mm diameter. A schematic of the optical arrangement is shown in Fig. 1. Both the 1064-nm and frequency-doubled 532-nm beams from the Nd:YAG laser were used, the diameter of which prior to the focusing lens was approximately 9 mm with a nearly Gaussian profile. In order to observe the effect of varying the laser fluence for the 532 nm wavelength, a combination of a half-wave plate, mounted in a rotation stage, in series with a polarizing beam splitter was used to attenuate the laser energy by a varying amount. A neutral density filter (0.7 N.D.) preceded the half-wave plate to reduce the laser power to an acceptable value. By rotating the half-wave plate to vary the ratio of vertically polarized to horizontally polarized light, in conjunction with aligning the beamsplitter to transmit only the vertically polarized portion of the beam, laser energies ranging from 0.2 to 3.3 mJ were obtained corresponding to laser fluences between 2.5×10^7 and 4.2×10^8 W/cm^2. The laser energy was monitored during the experiment using a pyrometer (Molectron J1000), and was maintained during the actual measurements of the soot volume fraction at 2.4 mJ for a laser fluence of 3.0×10^8 W/cm^2 in order to minimize the effect of laser beam attenuation across the flame (see discussion below). The LII signal was collected at a 90° angle using a focusing lens with an *f*-number of 3 at unit magnification. Except for the spectral scans, an inference filter (400 ± 10 nm) was placed in front of the monochromator to minimize interference from light scattering. For the spectral scans, the interference filter was removed from in front of the monochromator and the 1064-nm wavelength laser probe beam was used to produce the LII signals. For these measurements, a series of neutral density filters were used to vary the laser fluence rather than the half-wave plate/beam splitter arrangement used with the 532-nm wavelength laser probe beam.

Since the LII signal had a continuous spectrum in the visible wavelength range, while

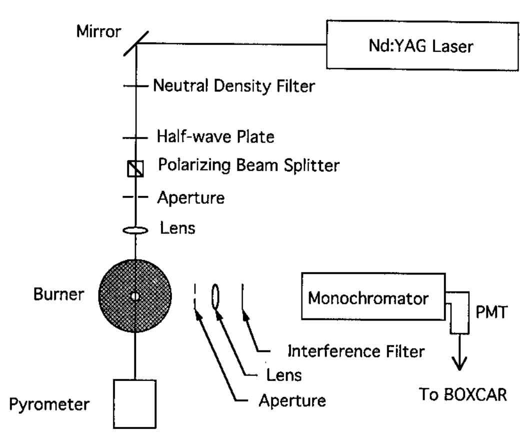

Fig. 1. Optical arrangement for laser-induced incandescence measurements.

interference from light-scattering and PAH fluorescence was expected near wavelengths of 532 nm and above [10, 11], the detection of the LII signal was made at a wavelength of 400 nm for both 532- and 1064-nm wavelength probe laser beams. Measurements made at 500- and 700-nm detection wavelengths for the 1064 nm wavelength probe laser yielded identical results due to the continuous nature of the LII spectrum in the visible wavelength range. The detection wavelength was set by using a 0.25-m monochromator (Instruments SA H20) with a grating blazed at 330 nm with 1 mm × 1 mm slits, which also defined the length on the probe volume. The bandpass of the monochromator was estimated to be 4 nm FWHM, while the spectral response of the monochromator was calibrated using an incandescent lamp (Eppley T24). A photomultiplier tube (Hammamatsu R928) was connected to the exit slit of the monochromator, the signal from which was conditioned using a boxcar integrator with a gate width of 10 ns and averaged over 100 laser shots.

Temporal variations of the LII signal were observable by moving the boxcar gate in 2–10-ns increments. LII profiles across the flame and spectral characteristics of the LII signal were observed by using the burner traverse system described above and by the scanning of the detection wavelength via the monochromator, respectively. The measurements were made in nonsmoking ethylene/air diffusion flames where the ethylene and air flow rates were 3.85 cm^3/s and 1060 cm^3/s, respectively.

RESULTS AND DISCUSSION

Figure 2 shows a typical temporal variation of the LII and vertically polarized light-scattering signals taken at a height of 40 mm above the fuel tube exit for an ethylene laminar diffusion flame at the radial location where the peak soot volume fraction is observed ($r = 2.5$ mm). The variation of the LII signal in time has been obtained by increasing the boxcar gate delay in 2–10-ns increments with respect to the laser pulse while averaging over 100 laser shots, as described above. It can be observed in Fig. 2 that the initial phase of the signal involves a rapid rise in the LII signal intensity due to the increase in the soot particle temperature during the laser pulse of ca. 7 ns in duration. Subsequent to the peak in the LII signal, the soot particles undergo conductive and radiative heat loss to the ambient gas and the LII signal gradually decreases, although a sensible LII signal is still observed at approximately 600 ns after the laser pulse. The temporal variation of the LII signal shown in Fig. 2 is qualitatively very similar to the LII response function for an idealized laser pulse computed by Melton [1]. A characteristic time constant for the LII process for soot particles has been shown to be linearly proportional to the soot particle diameter [3], and is estimated to be approximately 700 ns for a diameter of 100 nm. The decay time observed in Fig. 2 is approximately 600 ns for the signal to decrease to 5% of the peak value, while the mean soot particle diameter (D_{63}) at this location is approximately 135 nm (see Fig. 7b). Thus, in contrast to the light-scattering signal, which is observable in principle only during the duration of the laser pulse due to its elastic scattering nature, the LII signal exhibits a much longer temporal characteristic, as shown in Fig. 2. Melton [1] has discussed the potential for obtaining particle size distribution parameters from the temporal behavior of the LII signal. Such information is better obtained using a shorter probe laser wavelength than that used in the present study and may also suffer from interference from laser-influenced fluorescence from PAH species [1].

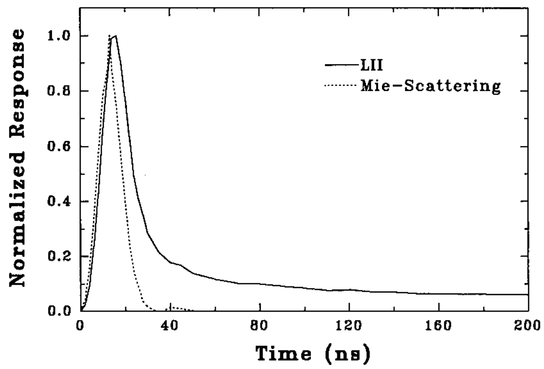

Fig. 2. Temporal response of the laser-induced incandescence.

A comparison between the soot volume fraction measured by LII and the laser scattering/extinction technique is shown in Figs. 3a–3c, where the soot volume fraction is plotted as a function of radial location at selected heights ranging from 10 to 70 mm above the fuel tube exit. The open and dark symbols represents laser scattering/extinction and LII data, respectively. The laser scattering/extinction data for soot volume fraction in this flame has been taken from Santoro et al. [8, 9], and involves the well-known method of measuring the line-of-sight extinction of the laser beam followed by a tomographic inversion in order to reconstruct the local soot volume fraction. Further details of this technique and the data can be found in Santoro et al. [8, 9]. In order to calibrate the observed LII signal, the LII signal at a single spatial point corresponding to the radial location where the peak soot volume fraction occurs ($r = 2.5$ mm) at the 40 mm height has been equated with the known value of soot volume fraction at this location from the laser scattering/extinction measurements. All other LII data can then be converted to absolute soot volume fraction based on this single-point calibration procedure.

The radial profiles of soot volume fraction obtained in this manner, as shown in Figs. 3a–3c, exhibit the familiar physics of soot growth and oxidization in this flame. At low heights, soot particles are observed in the annular region on the fuel-rich side of the flame. Soot formed in this region undergoes net growth with increasing height up to $H = 40$ mm where the peak soot volume fraction is observed at a radial location of 2.5 mm from the centerline, while soot is observable at the centerline at a height of 30 mm in Fig. 3a. Subsequent development involves a net destruction of soot particles through soot oxidation with soot volume fraction in the annular region diminishing more rapidly than in the central region.

Figures 3a–3c show an excellent agreement between the LII and laser extinction/scattering data for the soot volume fraction with data being within 5%–10% of one another at most of the heights where measurements have been obtained. However, there is a tendency for the LII data to underestimate the soot volume

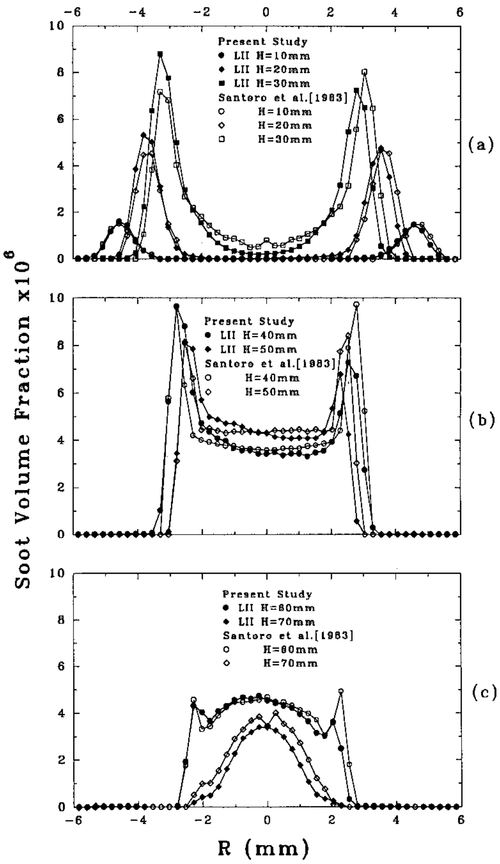

Fig. 3. Radial profiles of the soot volume fraction obtained via laser-induced incandescence and laser scattering/extinction at several heights (H) above the fuel tube exit of the burner (a) $H = 10$, 20, and 30 mm; (b) $H = 40$ and 50 mm; and (c) $H = 60$ and 70 mm.

fraction on one side of the flame, resulting in slightly asymmetric soot volume fraction profiles in Fig. 3b. This effect is more pronounced at the height of 40 mm than elsewhere and is attributable largely to the fact that the LII signal from the far soot peak traverses the flame in order to arrive at the signal detection site and, thus, is subject to increased path length and correspondingly increased absorption of the signal by the soot and PAH species in the flame. This effect may be correctable by estimating and integrating the local extinction of the signal across the flame. Figure 4 similarly shows the integrated soot volume fraction as a function of height. The comparison between the LII and laser extinction/scattering data [8, 9] yields reasonably good agreement

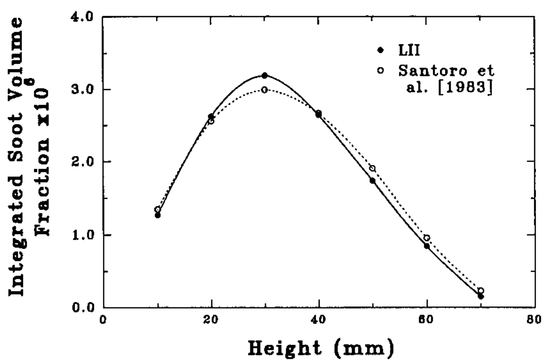

Fig. 4. Integrated soot volume fraction plotted as a function of height above the fuel tube exit of the burner.

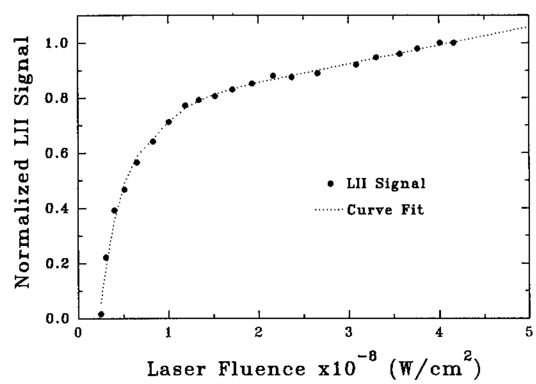

Fig. 5. Effect of laser fluence on the laser-induced incandescence signal.

with the maximum discrepancy being approximately 10% at a height of 30 mm.

The effect of laser fluence on the LII signal is shown in Fig. 5. The laser fluence has been varied from 2.5×10^7 to 4.2×10^8 W/cm^2 using the combination of the half-wave plate and beamsplitter, as described earlier. For a laser fluence from 2.5×10^7 to 1.2×10^8 W/cm^2, it can be seen in Fig. 5 that the LII signal increases rapidly as the laser fluence increases. This effect is due to the fact that the soot particle temperature increases as a function of the laser fluence, which causes a corresponding increase in the LII signal. In contrast, the LII signal for laser fluence beyond ca. 1.2×10^8 W/cm^2 exhibits a small increase with respect to an increase in the laser fluence. The influx of laser energy on the soot particles can cause vaporization of small carbon fragments, such as C_2 and C_3, from the soot particle surface. For sufficiently large laser fluence this vaporization mechanism and corresponding mass loss can become the dominant effect which limits the increase in soot particle temperature and, thus, causes a leveling of the LII signal as shown in Fig. 5. However, the LII signal intensity in this "saturation" regime increases as a weak function of the laser fluence in Fig. 5 similar to the results of Eckbreth [7] in which soot surface temperatures as high as 5000 K were observed with increasing laser fluence. From the data shown in Fig. 5, it is estimated that the "saturation" regime for the LII signal occurs for laser fluences above 1.2×10^8 W/cm^2 in the present studies. During the actual measurements of soot volume fraction, large laser fluence levels in the "saturation" regime, as were used in the present study, have the advantage of being least affected by the effects of the laser beam attenuation across the flame, since the LII signal is a weak function of the laser fluence in this regime. For the results reported here, the LII response varied by less than 5% for the range of laser fluence attenuations (~ 30% maximum attenuation) encountered in the present study. The observed response of the LII signal to the laser fluence is sensitive to the laser beam intensity profile, the specific focusing arrangement employed for the incident beam and the timing of the boxcar gate pulse with respect to the laser pulse. Other researchers have observed more significant effects of the laser fluence on the LII signal [12] and further research is needed in this area. However, for the conditions described in the present work, reproducible results were always obtained as long as care was taken in the alignment of the optics and achievement of a near-gaussian beam intensity profile.

The spectral response of the LII signal is shown in Fig. 6, where the LII signal is plotted as a function of the detection wavelength. The spectral scan of the LII signal has been obtained by rotating the grating in the monochromator so that the detection wavelength is varied in 10-nm increments. Measurements have been taken at the radial location where the peak soot volume fraction occurs at the 40 mm height for a probe laser beam at the 1064-nm wavelength with a beam diameter of 0.5 mm

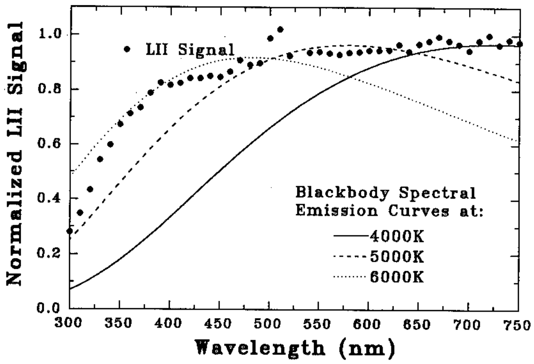

Fig. 6. Spectral response of the laser-induced incandescence where incident laser light at a wavelength of 1064 nm was used to obtain the spectrum shown.

and an energy of 1.5 mJ/pulse or laser fluence of 9.5×10^7 W/cm^2. For the 1064-nm wavelength laser probe beam, interference from light-scattering and PAH fluorescence is minimal in the visible wavelength range, although a fortuitous peak at 532 nm is seen due to the leakage of the 532-nm beam from the laser and corresponding light-scattering at this wavelength. It can be observed that as expected the LII signal exhibits a continuous spectra in the visible wavelength range with the signals decreasing to small levels below 300 nm. The LII spectrum also shows no distinct peaks and continues up to 750 nm at a nearly constant level. Comparison with computed blackbody radiation curves, as shown in Fig. 6, indicates that the LII spectrum in the 300–450-nm range corresponds to a soot temperature between 5000 and 6000 K, which is higher than the estimated soot vaporization temperature of ca. 4000 K. That the LII spectrum continues at a nearly constant level up to 750 nm and beyond, while the computed radiation curves begin to decline, is attributed to the fact that a large soot number density, and corresponding multiplicity of soot particle size, is present in the probe volume with different soot surface temperatures induced by the laser fluence. Hence, a more continuous and level spectral response is expected, as shown in Fig. 6 for a group of soot particles in comparison to a radiation curve computed for a single soot particle surface temperature. From Fig. 6, it can also be observed that by using 1064-nm wavelength probe laser, LII measurements can be taken at nearly any wavelength in the visible range, except near 532 nm for our optical arrangement, due to the absence of interference from PAH fluorescence and light-scattering as noted above, and measurements taken at detection wavelengths of 400 and 700 nm have yielded identical soot volume fraction results.

The heating of the soot particles to such high temperatures may lead one to be concerned about altering the properties of the soot particles. In the present experiments we assume that the soot properties are not seriously affected by the large laser fluence before the LII measurements are made. Furthermore, since the LII signals are calibrated against independently measured soot volume fraction values, any changes introduced, as long as they are the same throughout the soot field, are compensated for in the calibration procedure. These assumptions will need to be verified in future studies, but are not anticipated to introduce errors any more significant than the present knowledge of soot properties necessarily include.

Figure 7a shows the soot volume fraction measured by LII and the vertically polarized component of the light-scattering signal (Q_{vv}), while Fig. 7b contains the data for the mean soot particle diameter and the soot number density obtained from these measurements. To obtain the results shown in Fig. 7b, the Q_{vv} and soot volume fraction measurements were analyzed in a manner similar to that described by Santoro et al. [8]. The vertically polarized light-scattering signal has been obtained for the 532-nm wavelength laser probe beam, and

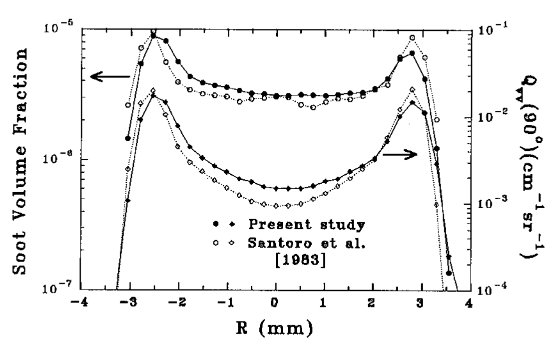

Fig. 7. (a) Radial profiles of the soot volume fraction and vertically polarized light-scattering signal, Q_{vv}, at a height, H, of 40 mm above the fuel tube exit of the burner.

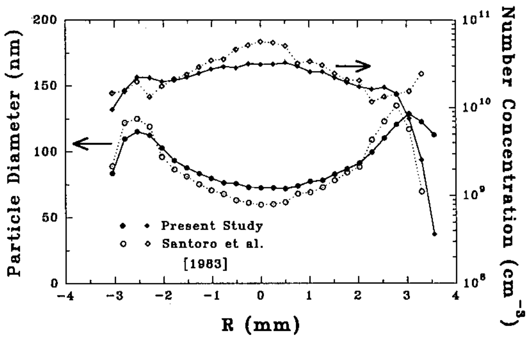

Fig. 7. (b) Radial profiles of the mean soot particle diameter, D_{63}, and soot number concentration, N, at a height, H, of 40 mm above the fuel tube exit of the burner.

the calibration for the absolute value of Q_{vv} has been obtained by matching with the known value of this signal at a single radial position using measurements by Santoro et al. [8] similar to the calibration procedure for the soot volume fraction. From the soot volume fraction and light-scattering data, which is proportional to the sixth moment of the soot particle diameter distribution, the mean soot particle diameter (D_{63}) and the soot number density (N) can be obtained as follows [8]:

$$D_{63} = \lambda^{4/3}\left(\frac{2Q_{vv}}{3\pi^3 F(\tilde{m}) f_v}\right)^{1/3}, \tag{1}$$

where

$$F(\tilde{m}) = \left|\frac{\tilde{m}^2 - 1}{\tilde{m}^2 + 2}\right|^2, \tag{2}$$

$$N = \frac{12 f_v}{\pi D_{63}^3}. \tag{3}$$

Here, f_v denotes the soot volume fraction while the complex index of refraction, $\tilde{m}$, is taken as $(1.57 - 0.56i)$ following Dalzell and Sarofim [13]. Similar to light scattering/extinction, the LII/light scattering technique yields the ratio of the sixth to third moment (D_{63}) of the particle diameter. The resultant mean soot particle diameter and the soot number density are again compared with the previous data obtained for this flame using the laser scattering/extinction method [8]. Figure 7b shows that the mean soot particle diameters are in very good agreement, with the discrepancy being mostly limited to the central region where due to the relatively low signal levels there is the largest uncertainty in both the LII and light-scattering data. The mean soot particle diameter from LII data in the present study ranges from 75 to 130 nm, while diameters of 60–135 nm have been observed by Santoro et al. [8].

It should be noted that the particle diameters reported, in some cases, exceed the size normally considered to be appropriate for a Rayleigh theory analysis. In fact, the soot particles in this flame are aggregates composed of primary particles that do satisfy the Rayleigh theory limit [14]. Thus, strictly, D and N refer to the volume equivalent diameter and number density, respectively, of these aggregates. Particle morphology effects are not included explicitly in the current study, which would require multiple-angle light scattering measurements. However, such measurements are feasible, in principle, and represent another area where the LII technique can contribute to research related to soot particle property determination.

The soot number density profiles are similarly in reasonably good agreement with the discrepancy again occurring near the centerline. Since the number density is inversely proportional to the cube of the mean soot particle diameter, the factor of 2 difference in the soot number density near the centerline is caused by the corresponding difference in the mean soot particle diameter shown in Fig. 7b. Thus, Fig. 7b shows that the single-point or two-dimensional measurements of the LII and light scattering, which can be set up with relative ease, a complete characterization of soot particle properties may be directly obtained.

CONCLUSIONS

From the discussion above, the following conclusions concerning the LII diagnostic of soot volume fraction are made:

1. Laser-induced incandescence has been used to obtain spatially resolved measurements of soot volume fraction in laminar diffusion flames, in which comparisons with laser scattering/extinction data yield excellent

agreement for both radial profiles and integrated volume fraction. Thus, LII can be used as an instantaneous, spatially resolved diagnostic of soot volume fraction without the need for the conventional line-of-sight laser extinction method.

2. The temporal characteristics of the LII signal are observed to involve a rapid rise in intensity followed by a relatively long (ca. 600 ns) decay period subsequent to the laser pulse, while the effect of laser fluence is manifest in nonlinear and saturated response of the LII signal with the transition occurring at a laser fluence of approximately 1.2×10^8 W/cm^2 for a laser pulse of ca. 7 ns in duration.
3. Spectral response of the LII involves a continuous spectrum in the visible wavelength range due to the blackbody nature of the emission, where the spectral response for 300–450-nm wavelength range indicates a soot surface temperature of ca. 5000 K with the spectrum continuing at a nearly level intensity up to 750-nm wavelength due to the multiplicity of the soot particle sizes in the probe volume.
4. Simultaneous measurements of LII and the vertically polarized light-scattering yield encouraging results concerning the mean soot particle diameter and number concentration; thus, significant applications exist in two-dimensional imaging and simultaneous measurements of LII and light scattering to generate a complete soot property characterization.

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