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Soot particle size measurements in laminar premixed ethylene flames with laser-induced incandescence and scanning mobility particle sizer

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Soot Particle Size Measurements in Laminar Premixed Ethylene Flames with Laser-Induced Incandescence and Scanning Mobility Particle Sizer

A Thesis submitted in partial satisfaction of the requirements for the degree Master of Science in Engineering Sciences (Mechanical Engineering)

by

Chung-Yuan Yin

Committee in Charge:

Professor Steven G. Buckley, Chair
Professor Foreman Williams
Professor Robert Cattolica

2009
The Thesis of Chung-Yuan Yin is approved and it is acceptable in quality and form for publication on microfilm and electronically:

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Chair

University of California, San Diego

2009
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ABSTRACT OF THE THESIS

Soot Particle Size Measurements in Laminar Premixed Ethylene Flames with Laser-Induced Incandescence and Scanning Mobility Particle Sizer

by

Chung-Yuan Yin

Master of Science in Engineering Sciences (Mechanical Engineering)

University of California, San Diego, 2009

Professor Steven G. Buckley, Chair

Laser-induced incandescence (LII) is used to obtain soot particle size measurements in laminar premixed ethylene flames. The LII signals are calibrated by comparing with the particle diameters measured with scanning mobility particle sizer (SMPS). Issues related to the calibration of LII with SMPS are discussed, such as the effects of equivalence ratio and laser fluence. LII and SMPS have similar sensitivities to
equivalence ratio in terms of total surface area and total volume, but the equivalence ratio needs to be carefully controlled as it highly affects the results. High laser fluence affects LII results by vaporizing soot particles, which reduces particle size. The effect of high laser fluence is particularly noticeable in the flame positions near the surrounding air such as at high flame heights and the outer combustion zone of nonpremixed flames where particle oxidation takes place. Overall, the comparison between LII and SMPS data yields encouraging results as they exhibit similar trends. The correspondence of LII and SMPS implies that SMPS could be applied as a valuable tool to calibrate LII signals and avoid the necessity of developing complex LII models.
CHAPTER 1 Introduction

1.1 Background

Soot is particulate matter (PM) produced as a result of incomplete combustion of hydrocarbon fuel. Ideally, complete combustion of hydrocarbon fuel leads to only carbon dioxide and water, but in practical combustion systems, complete combustion is hard to achieve. With insufficient oxidizer to convert the fuel completely (incomplete combustion), which can be a local or a global phenomena, other products such as carbon monoxide, hydrogen, and soot exist in addition to carbon dioxide and water. Soot is also formed in the cone of a nonpremixed flame as the fuel pyrolyzes, traveling out of the core of the flame toward the oxidizer. Depending upon the residence time and local turbulence, such soot may be incompletely oxidized in the outer cone/tip of the flame.

Aerosols, including soot, classified as PM2.5 (2.5 \( \mu m \) in diameter or smaller) by the U.S. Environmental Protection Agency (EPA) pose significant environmental and health hazards [1]. Although carbon dioxide is known as the main contributor to the global warming, PM may also affect the climate due its ability to absorb and radiate solar energy in the atmosphere [2]. Because of its small size, 2.5 \( \mu m \) in diameter or smaller, soot is able to invade the body’s respiratory system and migrate into the lung. Depending on the particulate size and chemical composition, soot can cause lung cancer, asthma, cardiovascular issues, and other respiratory diseases [3]. To reduce soot emissions from combustion systems, it is important to understand the soot formation and oxidation in various stages of combustion processes, for which the appropriate diagnostic tools are required. A few soot diagnostic techniques discussed in the next sections are gravimetric
filter sampling (EPA Method 5), laser scattering-extinction, laser-induced incandescence (LII), and probe sampling using scanning mobility particle sizer (SMPS).

1.2 Soot Diagnostic Techniques

1.2.1 Gravimetric Filter Sampling (EPA Method 5)

Gravimetric filter sampling is a traditional sampling technique that measures the particles by its mass. It has been applied as EPA Test Method 5 for the determination of PM mass emission from stationary sources [4]. The basic mechanism of gravimetric filter sampling is simply the collection of particles with filters and weighing the particles. Substantial care must be taken to either measure all of the particles completely or to have a representative sample. In Method 5 and in other related methods, liquid impingers, acid solutions, and other chemical processing are involved, but it is beyond the scope of this paper to discuss these methods in detail. A primary disadvantage of gravimetric filter sampling is that these methods require a long sampling time to collect sufficient mass of particles for statistically significant mass to be determined, and thus cannot be applied for real-time measurements. Also, gravimetric filter measurements can be affected by various artifacts such as the vaporization of semi-volatile compounds from the filtered particles and the chemical reactions between the filtered particles, filter substrates, and the surrounding gas [5].

1.2.2 Laser Scattering-Extinction

Laser scattering-extinction has been widely used to determine soot volume fraction and soot particle size [6, 7, 8], and the results are often used to compare and
validate LII results [9, 10]. The evaluation of soot radiative properties from scattering-extinction measurements is based on the Rayleigh theory for isotropic spheres assuming that the particle diameters are much smaller than the wavelength of radiation and particles have a monodisperse size distribution. Derived from Rayleigh theory, the governing equations for the soot volume fraction \( f_v \) and the soot particle diameter \( d \) are [11]

\[
f_v = \frac{\lambda}{6\pi} \frac{\kappa_{ext}}{E(\tilde{m})}, \quad E(\tilde{m}) = -\text{Im}\left(\frac{\tilde{m}^2 - 1}{\tilde{m}^2 + 2}\right),
\]

(1.1)

\[
d = \lambda \left[ \frac{4}{\pi^2} \frac{E(\tilde{m})}{F(\tilde{m})} \frac{Q_{vv}}{\kappa_{ext}} \right]^{1/3}, \quad F(\tilde{m}) = \left| \frac{\tilde{m}^2 - 1}{\tilde{m}^2 + 2} \right|^2,
\]

(1.2)

where \( \lambda \) is the laser wavelength, \( \tilde{m} \) is the complex refractive index of the particle, \( \kappa_{ext} \) is the extinction coefficient, and \( Q_{vv} \) is the scattering efficiency factor. In the Rayleigh regime, scattering is negligible compared with absorption, meaning that absorption is approximately equivalent to extinction. The extinction coefficient \( \kappa_{ext} \) and scattering efficiency factor \( Q_{vv} \) can be evaluated experimentally, and the complex refractive index \( \tilde{m} \) needs to be assigned from the literature [12].

The Rayleigh scattering theory is not fully applicable to ensembles of soot particles in a flame [8]. The theory applies better in the early stages of soot formation when the particles are isolated, spherical in shape, and small. In the later stages, the individual particles grow and form aggregates with fractal shapes, at which point the assumptions of very small particle diameters and monodisperse size distribution can no longer be applied.
1.2.3 Laser-Induced Incandescence

Laser-induced incandescence (LII) involves the heating of soot particles to the temperatures above the surrounding gas temperature with laser irradiation and subsequent measurement of the emitted radiation corresponding to the elevated soot particle temperature. The temperature of the soot particles is governed by the energy balance equation [13]:

$$\frac{dT}{dt} = \frac{\Delta H_v}{M} \cdot \frac{dm}{dt} + A \cdot q_{\text{laser}} + \rho \cdot C \cdot V \cdot \frac{dT}{dt}$$  \hspace{1cm} (1.3)

The terms are, respectively from the left, laser energy absorbed (absorption efficiency \(Q_{abs}\), presented area of the particle to the laser \(A_p\), laser irradiance \(q_{laser}\)); heat transfer to the surrounding medium (generalize heat transfer coefficient \(h\)); vaporization of soot particles (heat of vaporization \(\Delta H_v\), molar mass of solid carbon \(M\)); thermal radiation (radiative heat flux \(q_{rad}\)); and the change of internal energy (density \(\rho\) and specific heat \(C\) of carbon).

LII was first considered a problem in Raman measurements conducted by Eckbreth in 1977 [14]. However, it has become a valuable soot diagnostic tool, particularly after the work of Melton published in 1984. Melton shows that in the limit of high laser power and maximum particle temperature, LII signal, or the blackbody radiation due to laser heating, is nearly proportional to the soot volume fraction at the laser focus [15]. Hence, LII can be used as a pointwise, nonintrusive, and spatially-resolved diagnostic tool for soot volume fraction. Melton also proposed that the temporal profile of the LII signal could be used to determine soot primary particle size. In 1995,
Will et al performed time-resolved LII (TIRE-LII) that allows direct measurements of the soot primary particle sizes by measuring the LII signals at two different times during the cooling of the soot particles [16]. Since the cooling behavior is a function of the surface area and volume of the particles, the signal ratio obtained can be related to the soot particle diameter. Different approaches for particle sizing based on the cooling behavior also have been conducted by other researchers, such as Roth and Filippov [17], who evaluate the full signal decay curve, and Mews and Seitzman [18], whose analysis is based on the comparison of wavelength ratios.

1.2.4 Scanning Mobility Particle Sizer

The techniques discussed in the previous two sections are laser-based and nonintrusive methods. Soot diagnostics can also be performed sample-based with a probe collecting particles directly from the flames. However, such methods are intrusive and disturb the flames. After collection, particles are analyzed by a scanning mobility particle sizer (SMPS) that determines the particle size distribution.

An SMPS consists of a condensation particle counter and an electrostatic classifier equipped with a differential mobility analyzer. As the particles are collected by the probe, they first enter the electrostatic classifier. In the electrostatic classifier, the particles pass through an aerosol neutralizer that exposes the particles to high concentration of bipolar ions. The particles collide with the ions and eventually reach to an equilibrium state with a known bipolar charge distribution. The charged particles then enter the differential mobility analyzer (DMA), which contains two concentric metal cylinders. The inner cylinder, the collector rod, is negatively charged, and the outer
cylinder is electrically grounded. This creates an electric field between the two cylinders. The particles flow down the annular space between the cylinders along with a sheath flow introduced at the top of the DMA. The electric field causes the positively charged particles to be attracted by the negatively charged collector rod. Particles with a high electrical mobility are collected on the upper portion of the rod, and particles with relatively lower electrical mobility are collected on the lower portion of the rod. Since the electrical mobility is a function of particle size, the particles are differentiated by size along the length of the collector rod. Depending on the DMA voltage, only a particular size range will be able to exit through the bottom of the DMA. Only the particles within the defined range of electrical mobility exit the DMA to enter the condensation particle counter (CPC), where the particle concentration will be determined. Figure 1.1 shows the flow schematic of an electrostatic classifier with a long DMA, which is used in this experiment [19].
1.3 Objective

LII has been shown to be a useful technique for soot diagnostic even in complex combustion systems such as diesel engines [20] due to its nonintrusive nature and high spatial/temporal resolution. A primary focus has been developing models that describe the heating and cooling mechanism of LII. These models generally solve the energy and mass balance equations for particle size and temperature (e.g. Equation 1.3). However, each model has different assumptions and parameters, which lead to a wide variability in LII predictions and experimental results [21].
In this project, a simple LII experiment was conducted with rich laminar premixed ethylene flames. A Q-switched Nd:YAG laser was used to heat the soot particles at the second harmonic wavelength (532nm), and the radiative emission of the heated particles was detected by a spectrometer at a single wavelength (400nm). No LII models were applied in this experiment. The same flames were also measured with the SMPS that determines the soot particle size distribution at the same sample points as the LII measurement. The aim of this project is to show that SMPS can be a quality tool for calibrating LII signals by comparing LII and SMPS results.
CHAPTER 2  Theoretical

2.1 Soot Formation

During combustion, the hydrocarbon fuel in premixed flames is degraded by heat and oxidizers into small hydrocarbon radicals, forming soot precursors such as acetylene and polycyclic aromatic hydrocarbons (PAH). The precursors then coalesce into soot nuclei. These primary particles undergo surface growth due to direct addition of molecules from the gas phase or grow through coagulation with each other. Surface growth contributes to the major part of the final soot mass as it increases soot volume fraction [22]. Particle coagulation does not alter soot volume fraction but reduces the particle number density and form irregular aggregate structures as shown in Figure 2.1. The oxidation of soot particles occurs as the particles react with abundant oxidants in the atmosphere. Figure 2.2 shows a rough schematic of soot formation in premixed flames [22]. It can be seen that the primary particles formed in the early stages of soot formation are generally small and spherical. At later stages, soot particles begin to coagulate and form aggregates with fractal shapes. The different morphologies at various stages of soot formation pose complex problems for soot diagnostics. To simplify the problem, it is assumed in this paper that soot particles detected are the primary particles, which are small and spherical.
Figure 2.1: TEM photograph of soot aggregate sampled from a laminar acetylene flame [23]

Figure 2.2: A rough schematic of soot formation in premixed flame [22]
2.2 Soot Characterization

Soot particles generated from combustion systems are very small and generally characterized by the total mass of the condensed phase. With the assumption that the particles are monodisperse and spherical, soot particles could be characterized with the knowledge of the particle volume fraction $f_v$, particle number density $N$, and particle size (diameter) $d$. In this simple case, these three quantities are mutually dependent and can be related with the following equation [11]:

$$f_v = \frac{\pi}{6} \cdot N \cdot d^3$$

(2.1)

2.3 Radiative Properties

When a photon or an electromagnetic wave (e.g. laser pulse) interacts with a medium containing small particles (e.g. soot), the radiative intensity may be changed by absorption and/or scattering of the particles. The amount of absorption and scattering is determined by (i) the shape of the particle, (ii) the material of the particle (i.e. the complex index of refraction, $\tilde{m} = n - ik$), (iii) particle size, and (iv) the space between particles [24].

The absorption cross-section $C_{abs}$ and the scattering cross-section $C_{sca}$ are used to express the amount of absorption and scattering. However, an efficiency factor $Q$ is often used instead of cross-sections $C$ as $Q$ is nondimensional, obtained by dividing the cross-section $C$ with the projected area $\pi \cdot a^2$. The total amount of absorption and scattering is expressed in terms of the extinction cross-section and the extinction efficiency factor:
\[ C_{\text{ext}} = C_{\text{abs}} + C_{\text{sca}} \]  \hspace{1cm} (2.2)

\[ Q_{\text{ext}} = Q_{\text{abs}} + Q_{\text{sca}} \]  \hspace{1cm} (2.3)

2.4 Physics of LII Measurements

The physics behind LII measurements is governed by a simple energy balance of an irradiated particle (i.e. Eq. 1.3). A schematic representation of a laser-heated soot particle is shown in Figure 2.3. As a pulsed laser irradiate on a soot particle, the particle changes the laser intensity by absorption and/or scattering. Absorbed radiation increases the particle temperature above the surrounding gas temperature. The heating time is the duration of the pulsed laser and is in nanoseconds. After the particle is heated, there are three energy loss paths, heat conduction, particle vaporization, and heat radiation. A spectrometer may be used to observe the radiative intensity emitted from the particles, and different sizes of particles emit different time-dependent radiative intensities or LII signals; thus, LII signals could be used to distinguish particle sizes.

![Figure 2.3: Schematic representation of a laser-heated soot particle](image)
2.5 Previous LII Work

2.5.1 Soot Volume Fraction

According to Melton [15], the dominant energy loss path for particles at temperatures below 3300 K is heat transfer to the surrounding medium, and at the temperatures above 3700 K, vaporization becomes the dominant heat loss path. In between the temperatures of 3300 K and 3700 K, neither heat conduction nor particle vaporization is dominant. Thermal radiation is important only at extremely high temperature (e.g. above 10,000 K), but it is not attainable with laser irradiation. Therefore, in the limit of high laser power and maximum temperature, vaporization dominates and Eq. (1.3) becomes

$$\frac{dt}{dm} \cdot \frac{Q_{abs} \cdot A_p \cdot q_{laser}}{M} \cdot \Delta V \approx \cdot \Delta V \cdot \Delta T$$

(2.4)

where the left-hand side of the equation is the laser energy absorption, and the right-hand side is the vaporization energy. Soot particles in the Rayleigh limit exhibit a volumetric radiative absorption and emission where the absorption coefficient $Q_{abs}$ is linear with particle size $a$ in this regime. With $Q_{abs}$ dependent on radius $a$ and vaporization energy dependent on the particle temperature $T$, Eq. (2.4) relates the particle temperature to the particle size. From this relationship, Melton [15] derives the following equation that shows the dependency of the LII signal $J$ on the particle radius $a$:

$$J = C_1 \cdot \int_0^a N \cdot P(a) \cdot a^{3+0.154} \cdot \lambda_{em} \cdot da$$

(2.5)

where $C_1$ is a constant, $N$ is the total number density, $P(a)$ is the normalized probability density for a particle of radius $a$, and $\lambda_{em}$ is the detection wavelength in micrometers.
long detection wavelengths, the second power term in Eq. (2.5) can be neglected and the particle radius $a$ is raised simply to the power of 3, which can be related to the volume fraction according to Eq. (2.1). In his analysis, Melton has shown that the LII signal is proportional to the soot volume fraction at particle temperature near the vaporization point.

### 2.5.2 In-Situ Particle Sizing

Melton [15] states that smaller particles cool down faster than larger ones since the cooling rate of a soot particle is controlled by the ratio of surface area to particle volume and thus the primary particle size. There are various approaches that determine soot particle size based on the cooling behavior, including a comparison of signal ratios at different delay times (time-resolved LII) [16], an evaluation of the full decay curve [17], and a pyrometric measurement based on the LII signal ratios at two different wavelengths [18].

#### 2.5.2.1 Signal Ratios

Although thermal radiation, which is a function of the spectral radiant exitance $M_\lambda$, is insignificant in the particle energy balance, it represents the electromagnetic energy that a spectrometer would see. Thus, the LII signal $J$ acquired with a spectrometer could be shown as [25]

$$J \propto d^2 \int R(\lambda) \cdot \epsilon(d_p, \lambda) \cdot M^b_\lambda(T, \lambda) \cdot d\lambda$$  \hspace{1cm} (2.6)
where $R$ is spectral characteristics of the detection path, $\varepsilon$ is the emissivity of the soot particles, and $M^b_\lambda$ is the blackbody spectral radiant exitance. The blackbody spectral radiant exitance of a soot particle is given by Planck’s law [24],

$$M^b_\lambda(T, \lambda) = \frac{2\pihc^2}{\lambda^5 \cdot [\exp(hc / \lambda kT) - 1]}$$  \hspace{1cm} (2.7)

where $h$ and $k$ are Planck and Boltzmann’s constants respectively. Since the soot particles are much smaller than the irradiation wavelength, the emission coefficient $\varepsilon$ can be described by the Rayleigh approximation [25],

$$\varepsilon(d, \lambda) = \frac{4\pi \cdot d}{\lambda} \cdot E(\tilde{m}), \quad E(\tilde{m}) = -\text{Im}\left(\frac{\tilde{m}^2 - 1}{\tilde{m}^2 + 2}\right)$$  \hspace{1cm} (2.8)

The basic idea of time-resolved LII (TIRE-LII) is to measure the LII signals at two different times during the cooling period of the particles and to calculate the signal ratios. As particles of different sizes exhibit different cooling behaviors, soot primary particle sizes could be deduced from the calculated signal ratios. It is important to pick the optimum times that best distinguish particle sizes. For example, as shown by Will et al [13], the cooling behaviors of particles with various sizes are similar during the first 100 ns, which would be an ideal time for the first observation, but if the second observation time were also selected at a delay time before 100 ns, different particle sizes could not be distinguished. Therefore, the first observation time should be set after the initial decay of the curve (e.g. 100 ns), and the second observation times should be chosen late enough to ensure a significant signal difference for different particle sizes yet the signal is still detectable.
2.5.2.2 Full Decay Curve Evaluation

Unlike Will et al’s [16] approach that calculates the signal ratios at different times during the cooling period of the particles, Roth and Filippov [17] evaluates the full signal decay curve with a pointwise detection by a photomultiplier tube. From the analysis of particle absorption and emission, Roth and Filippov [17] developed the following equation that describes the detected radiation intensity $J$ in terms of time,

$$J_{\lambda}(t) = n_p V_m \int_{a_l}^{a_u} S_{\lambda}(a, I, t) \cdot f(a) da$$  

(2.9)

where $n_p$ is the particle number concentration, $V_m$ is the irradiated test volume, $S_{\lambda}$ is the radiation intensity, and $f(a)$ is the particle size distribution. By analyzing the full decay curve of the measured LII signals numerically with Eq. (2.9), particle size distribution could be reconstructed.

2.5.2.3 Wavelength Ratios

Another approach of particle sizing, proposed by Mews and Seitzman [18], is to compare the LII signals at the same defined time or time interval, but at different detection wavelengths. This approach allows the signal ratios or the particle sizes to be determined with a single laser shot. The advantage of this approach is that signal ratio is almost independent of local gas temperature for optimum laser intensity and time gate [18]. However, some argue that the wavelength ratio may not depend on particle size either [13].
2.6 Electrical Mobility Diameter Measurements with SMPS

The electrical mobility diameter \( d_m \), is the diameter of a sphere with the same migration velocity in a constant electric field as the particle of interest [26]. Instruments such as SMPS measure the electrical mobility diameter by balancing the electrical force and the drag force experienced by the particle. As mentioned in Section 1.2.4, soot particles are electrically charged in the electrostatic classifier and enter an electric field created in the DMA. An aerosol particle in an electric field carrying electrical charges experiences an electrical force, which is defined as [27]:

\[
F_{\text{elec}} = n e E
\]  

(2.10)

where \( n \) is the number of charges on the particle, \( e \) is the elementary unit of charge, and \( E \) is the strength of the electric field.

The drag force of an aerosol particle is governed by Stokes Law defined as [27]

\[
F_{\text{drag}} = -\frac{3 \pi \eta v d_p}{C}
\]  

(2.11)

where \( \eta \) is the gas dynamic viscosity, \( v \) is the velocity of the particle relative to the gas, and \( d_p \) is the physical diameter. A correction to the Stokes Law was introduced to account for the slip boundary condition for the fluid at the particle surface [27]. This correction is implemented as the Cunningham Slip Correction Factor \( C \), which was parameterized by Allen and Raabe [28] as:

\[
C = 1 + Kn \left[ \alpha + \beta \cdot \exp \left( -\frac{\gamma}{Kn} \right) \right]
\]  

(2.12)

where \( \alpha \), \( \beta \), and \( \gamma \) are empirically determined constants specific to the system under analysis. For solid particles (e.g. soot), \( \alpha \) is 1.142, \( \beta \) is 0.558, and \( \gamma \) is 0.9999 [28]. \( Kn \)
is the Knudsen number, which determines the flow regime of the gas around a particle. Knudsen number is defined as the ratio of the gas molecule mean free path $\lambda$ to the particle radius $r$,

$$Kn = \frac{\lambda}{r} = \frac{2\lambda}{d} \hspace{1cm} (2.13)$$

When a particle reaches a terminal migration velocity, the electrical and drag forces are equal and opposite [27]. Thus, the electrical mobility per unit electrical strength of a particle $Z_p$, could be determined by equating Eq. (2.10) and (2.11) as:

$$Z_p = \frac{neC}{3\pi\eta d_m} \hspace{1cm} (2.14)$$

For spherical particles, the electrical mobility diameter is equal to the physical diameter (i.e. $d_m = d_p$). If the particle is not spherical (e.g. soot aggregates), $d_m$ is greater than $d_p$ and a dynamic shape factor needs to be accounted for the increased drag on a particle due to the nonspherical shape [27].
CHAPTER 3  Apparatus and Procedure

3.1 Burner

The laminar premixed ethylene flames were produced with a laboratory burner with a diameter of 2.54 cm and a length of 61 cm. A short ceramic honeycomb section was inserted at the top of the burner tube to stabilize the premixed flames. To further enhance the flame stability at the sooting region, a chimney was introduced on top of the flames at the height of 10 cm above the burner. The burner rested on a small lab jack that was mounted on a horizontal translator to provide a 3-dimensional motion. The lab jack enables vertical movement for the burner, while the horizontal translator provides the horizontal movement in two directions.

Air and fuel were introduced at the bottom of the burner through two separate lines. Air was supplied by the building supply of pressurized air and fuel was supplied by a pressurized ethylene gas bottle purchased for this experiment. A Dwyer Instruments™ rotameter (range 0-100 SCFH air, RMC Series) was used to regulate air flow. Fuel flow was regulated with a Brooks Instruments™ flow meter (0-150mm, tube size R-2-15-B), which does not have an upstream pressure regulator, so it was connected to a Swagelok™ needle valve (stainless, straight 10-turn needle valve) that precisely controls the fuel flow going into the flow meter. Before the fuel enters the burner, it passes through a Swagelok™ flashback-arresting filter (stainless steel, 15 μm filter) that prevents the possibility of flames propagating upstream into the fuel supply. By adjusting the air and fuel flow meters, different equivalence ratios could be achieved. The next section discusses the flow meter calibration process.
3.1.1 Flow Calibration

A DryCal™ DC-Lite digital flow meter was used to calibrate both air and fuel flow meters. The DC-Lite flow meter employs patented near-frictionless piston technology and photo optic sensors to determine volumetric flow rate quickly and accurately. To calibrate the air flow meter, the DC-Lite flow sensor was connected downstream of the air flow meter. The upstream air supply into the air flow meter was kept at the pressure of 20 psi. By adjusting the control valve of the air flow meter, different amounts of air passed through the flow meter, and the DC-Lite flow meter would immediately show the volumetric flow rates. Twelve readings were obtained for the air flow meter, and the results were plotted to find a least-squares relationship between air flow meter readings and the volumetric flow rates. The least-squares relation is then used to determine the flow rates at each air flow meter reading. The same calibration method was carried out for the fuel flow meter with the upstream pressure (i.e. ethylene gas bottle pressure) set at 30 psi. Twelve fuel flow meter readings corresponding to the volumetric flow rates measured by the DC-Lite flow meter were obtained to find the least-square relationship between the fuel flow meter readings and its volumetric flow rates. The fuel flow meter calibration plot is shown in Figure 3.1, and the $R^2$ value of the linear fit is 0.9912.
3.2 LII Setup

The schematic of the LII experimental setup is shown in Figure 3.2 below. This setup mainly consisted of a Q-switched Nd:YAG laser, a spectrometer equipped with an intensified charge-coupled device (ICCD) camera, and a computer. There was also a chimney introduced at the top of the flame that is not shown in this schematic.
3.2.1 Laser

Laser pulses were generated from a Q-switched Nd:YAG laser operating at the second harmonic wavelength (532 nm) and the frequency of 10 Hz. The pulse duration of the Big Sky CFR-400 laser is approximately 10 ns and the average laser energy used was approximately 138 mJ. To examine the effects of varying the laser fluence, which will be discussed in Chapter 4, neutral density filters were placed in front of the flames (see Figure 3.2) to reduce the laser energy. Laser energy was not varied by changing the
voltage of the flashlamps or the timing of the Q-switch because both strategies change the laser beam profile. The laser beam has a diameter of 10 mm.

### 3.2.2 Spectrometer

LII signals were observed with a spectrometer (Acton Research™, SpectraPro 300i) mated to a time-gated ICCD camera (Princeton Instruments™, PI-MAX). Two bi-convex focus lenses (f = 75mm, f = 125mm) were used to focus LII signals into a fiber optic bundle that transmits the signals to the spectrometer. The signals were collected immediately after each laser pulse. To synchronize camera timing with laser pulses, the ICCD camera controller was connected to the Q-switch output of the laser with a triggering cable. Spectrometer and camera settings were established with spectroscopic software (Princeton Instruments™, WinSpec/32) that was installed on a computer in the laboratory.

### 3.3 SMPS Setup

The experimental setup of SMPS measurements is shown in Figure 3.3. The sampling probe was made out of a stainless steel tube (OD = 3.175 mm, ID = 1.588 mm) with an orifice (d = 0.681 mm) drilled in the center. The probe was placed horizontally with the orifice facing downward in the flames. Soot particles were drawn through the orifice and diluted immediately with a nitrogen flow to minimize particle coagulation in the sampling line. The diluted flow then entered the SMPS, which consists of a CPC and an electrostatic classifier equipped with a long DMA, at the flow rate of 1.05 L/min. Details of the measurement physics of the SMPS were discussed in Chapter 1 and 2. The
vacuum pump connected to the CPC provides the suction required to extract the soot particles from the flame, and its exhaust was connected to the vacuum exhaust of the laboratory. The Aerosol Instrument Manager software, which was installed on the computer connected to the CPC, analyzes the data acquired and is used to plot the particle size distribution of the flow.

![Diagram of SMPS experimental setup](image)

**Figure 3.3**: Schematic of SMPS experimental setup

3.4 **Experimental Procedure**

Air and fuel flow rates were set at 8.7 L/min and 1.4 L/min, respectively, to establish an equivalence ratio of 2.3. A lighter was used to ignite the flames. For LII
measurements, the pulsed laser was fired at the flames at the frequency of 10 Hz. To reduce the laser energy, neutral density filters with different transmittances were put in front of the flames. Spectrometer and camera settings were established with WinSpec/32 software as the following:

- Spectrometer grating type: 600 BLZ = 300nm
- Center wavelength: 400 nm
- Number of spectra: 1000
- Gate Width: 20 ns
- Gate Delay: 1270 ns

With Gate Delay set at 1270 ns, WinSpec/32 software would acquire signals at the delay time of 1.27 μs after the camera had been triggered by Q-switch. The Gate Width and Gate Delay settings were chosen to deliver the highest signals. The same LII procedure was repeated at different positions in the flames, as measured by the height above the burner.

For SMPS measurements, the same flames were used. The probe was placed horizontally in the flames to collect soot particles (see Figure 3.3) with a nitrogen dilution flow set at the rate of 0.8 L/min. The diluted flow then entered SMPS at the rate of 1.05 L/min. The sheath flow rate in long DMA was set at 10.5 L/min due to the requirement that sheath flow rate has to be at least 10 times the particle-laden air flow rate for accurate measurements. The Aerosol Instrument Manager software showed the particle size distribution in the range from 7 nm to 280 nm. This range was determined by the particle flow rate and the sheath flow rate. Clogging is a continuous problem when
sampling soot particles with a probe. Thus, between experiments the probe needed to be flushed with pressurized air to remove soot particles stuck to the inner wall and in the orifice. Also, the inlet impactor (see Figure 1.1) needed to be cleaned occasionally with a soft cloth soaked with alcohol to remove soot build-up.

3.5 Data Analysis

LII signals were from the average of 1000 spectra (1000 signals from 1000 laser shots) to minimize the effect of flame instabilities. The average signal (mean) and the standard deviation of mean (SDOM) were calculated with the following equations [29]:

\[
\bar{x} = \frac{1}{N} \sum_{i=1}^{N} x_i = mean
\]

(3.1)

\[
\sigma_{\bar{x}} = \frac{1}{\sqrt{N-1}} \sqrt{\frac{1}{N-1} \sum_{i=1}^{N} (x_i - \bar{x})^2} = SDOM
\]

(3.2)

where \(x_i\) is the signal at each laser shot and \(N\) is the total number of shots.

SMPS data were interpreted in four ways, through the determination of the peak diameter, number averaged diameter, surface area averaged diameter, and volume averaged diameter. Peak diameter is simply the particle diameter that has the highest number density in the particle size distribution. The number, surface area, and volume averaged diameters were obtained with the following equations:

\[
\text{number\_avg\_diameter} = \frac{\sum_{i=1}^{a}(\#_i)(d_i)}{\sum_{j=1}^{a}(\#_j)}
\]

(3.3)
\[
\text{surface\_area\_avg\_diameter} = \frac{\sum_{i=1}^{n} (\#_{i}) \left[ 4\pi \cdot \left( \frac{d_{i}}{2} \right)^{2} \right] (d_{i})}{\sum_{j=1}^{n} (\#_{j}) \left[ 4\pi \cdot \left( \frac{d_{j}}{2} \right)^{2} \right]}
\]

(3.4)

\[
\text{volume\_avg\_diameter} = \frac{\sum_{i=1}^{n} (\#_{i}) \left[ \frac{4}{3} \pi \cdot \left( \frac{d_{i}}{2} \right)^{3} \right] (d_{i})}{\sum_{j=1}^{n} (\#_{j}) \left[ \frac{4}{3} \pi \cdot \left( \frac{d_{j}}{2} \right)^{3} \right]}
\]

(3.5)

where \( d_{i} \) is the diameter of particles in bin \( i \) and \( \#_{i} \) is the number of particles in bin \( i \).

The analysis was based on the assumption that the particle shape is spherical instead of fractal.
CHAPTER 4     Results and Discussion

4.1     Temporal Response of LII

Figure 4.1 shows the LII signals corresponding to the temporal variation of the laser-heated soot particles. When a particle increases in temperature by absorbing laser energy, its thermal radiation or LII signal increases, according to the energy balance equation (Eq. 1.3). Observing the temporal variation in LII signal in Figure 4.1, the initial phase involves a rapid rise in the LII signal due to the increase of soot particle temperature during the laser pulse and shortly after the laser pulse. After the laser pulse, particle temperature quickly starts to decrease due to the heat loss to the surrounding medium and thermal radiation. The LII signals decrease rapidly subsequent to the maximum signal but, as expected from heat transfer arguments, the cooling rate slows down at lower particle temperatures. LII signals can still be detected at approximately 100 ns after the laser pulse. As discussed in Section 3.2.2, the camera is connected to and triggered by the Q-switch output of the laser. Delays between the trigger and the laser firing amounted to approximately 1240 ns, in fact, no LII signal was detected before 1240 ns following triggering. Hence in Figure 4.1 the time 0 ns corresponds to the delay time of 1240 ns. To assure the highest signal intensity, the signals obtained in LII measurements were acquired at the time of 30 ns (Time Delay = 1270 ns) as shown in Figure 4.1.
4.2 Typical SMPS Particle Size Distribution

The DMA distinguishes particle sizes by the electrical mobility of particles, and the CPC counts the number of particles at each size; hence, SMPS measures the soot particle size distribution based on the “electrical mobility diameter.” Figures 4.2 and 4.3 show the typical particle size distributions measured with SMPS in this experiment at the center of the flame and at different heights above the burner (HAB). Soot particles measured with SMPS have extended reaction times in the sampling line that could allow particles to further coagulate with each other, potentially resulting in a decrease in particle number density and an increase in particle size. A nitrogen dilution flow was introduced in the probe to reduce particle coagulation in the sampling line, but coagulation still affects the particle size distribution as it can be demonstrated by comparing Figures 4.2 and 4.3. In Figure 4.2, the particle size distribution is measured at 25 mm HAB, and the soot particles at this flame height are generally small and isolated.
because they are in the early stage of soot formation (see Figure 2.2). These small and isolated particles are less likely to coagulate in the sampling line. It can be seen from Figure 4.2 that the particle size distribution is nearly a Gaussian distribution with the maximum number density located at the diameter of 15.1 nm with slight effect of particle coagulation. In Figure 4.3, which shows the particle size distribution obtained at 45 mm HAB, the peak number density is achieved at the particle diameter of 34.6 nm. At particle diameters greater than 50 nm, the number density decrease steadily and slowly until a diameter of 150 nm is reached. The expanded distribution in Figure 4.3 compared with the Gaussian distribution in Figure 4.2 is due to the effect of particle coagulation. Notice also that the particle numbers in Figure 4.3 are two orders of magnitude higher than the particle numbers in Figure 4.2. This means that more small particles had formed, as well as additional coagulation. Coagulation naturally happens in the flame and is a recognized, important step in soot growth [30, 31]. Particle coagulation is generally most important at the later stage of soot formation, which can be presumed as at higher flame heights (i.e. 45 mm). Coagulation continues to an unknown extent in the sampling line as it spreads out the particle size distribution by reducing the particle number density and increasing the particle diameter. As stated in Section 3.4, nitrogen dilution flow was set at the rate of 0.8 L/min. At higher flow rates, SMPS would be overwhelmed by nitrogen and could not show variations in soot particle size distributions at different flame positions.
Figure 4.2: SMPS particle size distribution at $R = 0$ mm and $HAB = 25$ mm

Figure 4.3: SMPS particle size distribution at $R = 0$ mm and $HAB = 45$ mm

4.3 Equivalence Ratio

The number concentration of soot particles generated from the flames is highly dependent on the equivalence ratio. Higher equivalence ratio means the flame is more
fuel-rich and thus fewer particles are completely oxidized, leading to a higher number distribution of particles. In Figure 4.4, LII signals at equivalence ratios of 2.2, 2.3, and 2.4 were plotted. The data are collected at the flame height of 50 mm and at the center of the flame. It can be seen that LII signal increases as the equivalence ratio increases, and the LII signals are very sensitive to the equivalence ratios. At the equivalence ratio of 2.2, LII signal is hardly detectable because the incandescence from the soot particles is not strong enough to be seen by the spectrometer. On the other hand, the LII signal is very strong at the equivalence ratio of 2.4. Unfortunately, at the equivalence ratio of 2.4, too many soot particles were generated and often clogged the SMPS. In order to compare data between LII and SMPS measurements with high soot loading, the equivalence ratio of 2.3 was used throughout this experiment.

A comparison of sensitivities to equivalence ratios of LII and SMPS measurements is demonstrated by comparing Figures 4.4 and 4.5. Figure 4.5 shows the peak diameters, surface area average diameters, and volume average diameters deduced from the particle size distributions measured with SMPS at the same equivalence ratios as LII measurements. It can be seen that the surface area average diameters and volume average diameters follow the same trend as the LII signals as there is an accelerating growth when the equivalence ratio changes from 2.3 to 2.4. Peak diameters do not show the same growth rate. This result is expected as an increase of equivalence ratio increases the amount of unoxidized carbonaceous material, which in turn increases the particle size and concentration when coupled with particle coagulation and surface growth. LII measurements are highly affected by both particle size and concentration because they together determine the total amount of incandescence. SMPS measurements measure the
particle size distribution, and any increase in particle size would lift the particle size distribution curve to higher number densities but the shape of the curve remains approximately the same, to first order. In other words, the SMPS measured peak diameter should not vary much with number concentration. However, the combined effects of particle coagulation and surface growth increase the total surface area and volume of the soot particles, which results in an accelerating growth of surface area average diameters and volume average diameters in Figure 4.5. The correlation of LII signals to SMPS measured surface-area/volume average diameters demonstrates that an increase of equivalence ratio mainly increases the soot particle concentration but not the particle diameter.

Figure 4.4: LII signals at different equivalence ratios
4.4 Laser Fluence

The effect of laser fluence on the LII signal is shown in Figure 4.6. It shows that LII signal increases linearly at lower laser energy, but at the laser energy of 138 mJ, the LII signal has begun to level. The leveling of LII signal at higher laser energies is due to particle vaporization [32, 33]. During laser irradiation, a soot particle absorbs laser energy and increases in temperature, which corresponds to higher LII signals. Hence, LII signal increases with increasing laser energy due to increased peak temperature of the particles. But if the laser energy becomes too high, LII signals reach a “saturation” regime. At the “saturation” regime, particle vaporization becomes a dominant heat loss path and reduces soot particle size. The increased LII signals resulting from the elevated particle temperature are offset by the reduced particle size. Two laser fluences, 0.065 J/cm² and 0.1725 J/cm² corresponding to laser energies of 52 mJ and 138 mJ, were applied and compared with SMPS results in this project.
4.5 Soot Dependence on Flame Height

Soot particles at the center of the flames but at different heights above the burner (HAB) were measured with LII and SMPS. The results are shown in Figures 4.7 and 4.8. In Figure 4.7, LII measurements were performed at two laser fluences, which are at laser energy of 52 mJ and 138 mJ. At both laser fluences, LII signals increase exponentially with flame heights at HAB lower than 50 mm. This indicates that at these lower flame heights, the small particles were incepted and then rapidly underwent surface growth and coagulation. At the laser energy of 52 mJ, the exponential increase of LII signals is leveled after 50-mm HAB, and it is due to the effect of particle oxidation that decreases the particle size and concentration. At the laser energy of 138 mJ, the result follows the same trend as at the laser energy of 52 mJ, but rather than remaining level, the LII signals start to decrease at flame heights higher than 60 mm HAB. This is due to the combined
effects of particle vaporization and oxidation. As suggested above, particle vaporization is important at laser energy of 138 mJ. When soot particles reach the stage of oxidation at higher flame heights, the combined effects of particle vaporization and oxidation significantly reduce particle mass and concentration, which results in a decrease of LII signals as it can be seen in Figure 4.7. The effect of high laser fluence at lower flame heights is not that prominent.

Soot particle size distributions were measured with SMPS at each flame height, and the following diameters were obtained: peak diameter; number averaged diameter; surface area averaged diameter; and volume averaged diameter. Figure 4.8 shows the plots of particle diameters at different flame heights, and it can be seen that the peak diameter is the lowest diameter and volume averaged diameter is the highest diameter although all the diameters exhibit similar trends. One obvious point of interest for the volume averaged diameter occurs at flame heights between 30 mm and 35 mm HAB, where the volume-averaged particle diameter seemingly remains constant while the other diameters continue to increase. If this is not an anomaly in the data, it would indicate that perhaps the net volume growth stopped at this point but the surface area was increasing, perhaps through a burst of nucleation of small particles. The trend of each diameter in Figure 4.8 is similar to the LII signals at the laser energy of 52 mJ in Figure 4.7, at which particle vaporization is relatively much less important than at higher energies.
**Figure 4.7:** LII signals at different heights above burner (HAB)

**Figure 4.8:** Diameters measured with SMPS at different height above burner (HAB)
4.6 Soot Dependence on Flame Radial Position

Premixed flames consist of an inner and an outer combustion zone, which are the center and the annular regions of the flames respectively. Figure 4.9 shows the radial profile of the LII signals at HAB = 25mm. As observed in Figure 4.9, the LII signals increase as the distance from the center of the flame increases until the radius of 8.5 mm is reached. The radius of 8.5 mm is the boundary between the inner and the outer combustion zones. At the outer combustion zone, soot particles are exposed to the surrounding air, and particle oxidation reduces the particle size and concentration; hence, the LII signal also decreases. Both laser fluences at 52 mJ and 138 mJ in Figure 4.9 follow the same trend, which is an increase with increasing radius in the inner combustion zone and a decrease with increasing radius in the outer combustion zone. When the laser energy is at 138 mJ, the LII signals decrease more rapidly in the outer
combustion zone compared to the LII signals at the laser energy of 52 mJ. Again, this is due to the effects of particle vaporization of the oxidizing particles at higher laser fluences.

SMPS data at different radial positions were interpreted the same way as at different flame heights (i.e. Figure 4.8). Peak, number averaged, surface area averaged, and volume averaged diameters are plotted in Figure 4.10. The plots follow similar trends as the LII signals in Figure 4.9 except near the center of the flames, which is at the radial positions less than 5 mm. The volume and surface area averaged diameter decreases with increasing radius at the center of the flames and then starts increasing at R = 3.4 mm. This is different from the trends of peak diameter, number averaged diameter, and LII signals. It also contradicts the result that LII signals match best with volume averaged diameters at different flame heights. The effects of particle oxidation can again be observed with SMPS data such that after the radial position surpassed the inner combustion zone boundary at R = 8.5 mm, all the diameters start to decrease.
Figure 4.10: LII signals at different radial positions of flames at HAB = 25 mm

Figure 4.11: Diameters measured with SMPS at different radial positions of flames at HAB = 25 mm
4.7 Comparison of LII to SMPS

Figures 4.4 and 4.5 show a good correlation between LII signals and SMPS measured surface-area/volume average diameters at different equivalence ratios. The high sensitivities of LII and SMPS to equivalence ratio required precise control of fuel and air flow rates for these experiments because equivalence ratio greatly affects the results. The figures also show the limitation of LII and SMPS measurements. For example, at the equivalence ratio of 2.2, the LII signal is too weak to be detected by the spectrometer, and at the equivalence ratio of 2.4, the abundant amount of soot particles would clog the probe for SMPS measurements. However, comparisons of LII and SMPS can still be made by keeping the equivalence ratio constant at 2.3 throughout the experiment. Figures 4.7 and 4.8 show the LII and SMPS results at different flame heights, and they generally follow the same trend, which is that LII signals and particle diameters increase exponentially at lower flame heights and eventually become constant or start to decrease at the tip of the flames. Figures 4.9 and 4.10 show the LII and SMPS results at different radial positions of the flames. The results also exhibit a similar M-shaped trend although some differences were observed near the center of the flame. In general, both LII and SMPS results agree with soot formation theory and display similar trends. The LII and SMPS results were correlated directly by plotting the LII signals against the particle diameters measured by SMPS. The plots exhibit poor correlation and are shown in the Appendix for future consideration.
CHAPTER 5 Conclusions

5.1 Summary

In the present paper, LII has been shown to be a useful tool to describe soot formation in premixed flames. However, there are factors such as equivalence ratio and laser fluence that would affect the results and should be treated carefully. For example, when the equivalence ratio is increased, the LII signal would increase significantly due to the higher incandescence emitted from the increased particle size and concentration. Although SMPS shows similar sensitivity as LII in terms of surface area average diameters and volume average diameter, it is still necessary to calibrate LII signals at each equivalence ratio with SMPS when different equivalence ratios are applied. Laser fluence also varies LII results. At higher laser energies, the laser intensity can cause vaporization of small carbon particles from the soot particle surface that would decrease particle size. At the surface growth and coagulation stages of soot formation, the effect of particle vaporization is negligible; but at the oxidation stage, particle vaporization significantly reduces the particle size, resulting in a reduction of LII signals. Although employing SMPS as a calibration tool for LII measurements requires careful calibration steps, the LII and SMPS results obtained in this project show a nice agreement. LII is an effective nonintrusive diagnostic tool that provides in-situ particle size measurements, and with the assistance of SMPS, it is possible to deduce particle sizes from LII signals without applying any LII models. Further works and studies still need to be conducted as the plots in the Appendix show that precise correlations between LII signals and SMPS measured diameters were not achieved in this project.
5.2 Future Works

SMPS was used as a calibration tool in the present experiment yet there are still some uncertainties regarding the interpretation of soot data. One of the common issues that often affect SMPS results is soot aggregates. In this experiment, it was assumed that the soot particles detected or measured are primary particles that are spherical in shape. This assumption is faulty in practical combustion systems because soot particles inevitably coagulate into fractal aggregates during soot formation. Soot aggregates affect the SMPS results because the diameter measured by SMPS is the electrical mobility diameter, which is only equivalent to the physical particle diameter if the particle shape is spherical. To account for the effects of soot aggregates and deliver more accurate results, fractal morphology analysis should be incorporated in the future experiments.

Soot aggregates not only affect the SMPS results with their shapes; they also change the particle size distribution during the extended coagulation time in the sampling line. In this project, a nitrogen dilution flow was used to minimize particle coagulation in the sampling line, but the dilution ratio was limited by the CPC. The CPC restricts the flow rate of the particle-laden gas to be approximately 1 L/min. The low particle flow rate allows a small room to operate to achieve the optimum dilution ratio. From example, if the nitrogen flow rate were too high, the probe would have the nitrogen gas flowing out of the orifice instead of extracting soot particles into the orifice. On the contrary, if the nitrogen flow rate were too low, the amount of nitrogen gas was insufficient to dilute the flow. In the future experiments with SMPS, the mentioned problem should be solved to obtain the optimum dilution ratio that effectively prevents the soot particles from coagulating in the sampling line.
APPENDIX

LII signal vs. SMPS diameter:

Figure A: At different flame heights, laser energy = 52 mJ

Figure B: At different flame heights, laser energy = 138 mJ
Figure C: At different flame radius, laser energy = 52 mJ

Figure D: At different flame radius, laser energy = 138 mJ
REFERENCES


