On Line Measurement of Primary Fine Particulate Matter

Final Report

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ABSTRACT

The measurement of fine particulate in pulverized coal flames has several applications of importance. These include but are not limited to: 1. The detection of fine particulate in the effluent for pollution control. 2. The detection of soot and fuel burnout in real time within a boiler. and 3. The quantification of soot within coal flame for improved understanding of pulverized coal flame heat transfer and soot modeling. A method has been investigated using two-color extinction along a line of sight within the flame which provides a continuous real-time measurement of the soot concentration. The method uses two inexpensive HeNe lasers and simple light detectors. The results of testing the method on a pilot scale 0.2 MW pulverized coal reactor demonstrate the method is working well in a qualitative sense and an error analysis performed on the uncertainty of the assumed values demonstrates the method to be accurate to within $\pm 30\%$. Additional experiments designed to quantify the measurement more accurately are ongoing. Measurements at the end of the reactor just prior to the exit showed soot could not be detected until the overall equivalence ratio became greater than 1.0. The detection limit for the method was estimated to be 1×10^{-8} soot volume fraction. Peak soot concentration was found to approach a level of 0.88×10^{-6} at the sootiest condition. The method was used to obtain an axial profile of soot concentration aligned with the down-fired pulverized coal flame for three different flame swirls of 0, 0.5 and 1.5 and an overall equivalence ratio of 1.2. The axial measurements showed the soot concentration to increase initially and level off to a constant maximum value. At 0.5 swirl the soot volume fraction increased more rapidly near the burner and both the 0.5 and 1.5 swirl cases showed that soot had reached a maximum by 0.9 m, but the 0 swirl soot concentration was still increasing. Previous measurements of species and velocity in the reactor suggest that the flame is lifted at zero swirl allowing O_2 to be entrained and that the flame protrudes further down the reactor explaining the lower soot values measured. An evaluation of the potential for using this measurement technique on full scale boilers suggests that attenuation of the signal across a larger boiler distance is the largest obstacle. The beam would be expected to become completely attenuated under moderate sooting conditions; however, the longer pathlength would improve the ability of the method to measure very small amounts of soot escaping the main combustion zone.

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1 EXECUTIVE SUMMARY

The objective of this study was to investigate a new method of measuring fine particulate matter within a pulverized coal flame using two-color extinction. Theoretically, two-color extinction can measure the existence and quantity of particles less than 1 μm in diameter within a mixture of larger particles such as coal, char, and ash. Specific objects of the study are shown in a timetable (Table 1) which includes categories of: 1) An investigation to select the optimal light sources which should be used for testing and field use. 2) Experimental measurements on a pilot scale reactor to demonstrate the feasibility and accuracy of the measurements. 3) The completion of a set of particulate measurements in the pilot scale reactor. 4) An evaluation of the feasibility of full-scale application. As can be seen in the table, all of the objectives are complete with the exception of 2f. This task involves an experiment to compare the measurements of the two-color extinction method with a known quantity of soot mixed with coal, char, and ash. This final experiment is in progress.

Table 1. Table of objectives and dates completed.

Task	Month Completed											
	J	A	S	O	N	D	J	F	M	A	M	J
1. Identify Instrument Components												
1a. Laser Cost analysis	X											
1b. Sensitivity analysis	X											
2. Method Verification Process												
2a. Purchase and Assemble Components		X	X									
2b. Test components			X	X								
2c. Solid effluent samples SEM					X	X						
2d. Video Image Comparisons					X	X						
2e. Limit Analysis							X					
2f. Comparison with known particle mixture.												
3. Reactor Measurements												
3a. Fuel Air ratio effects								X	X	X	X	
3b. Axial profiles for various swirls									X	X	X	
4. Feasibility analysis											X	X

1.1 Primary Accomplishments

The primary accomplishment of this research has been the demonstration of a new technique for measuring fine particulate (primarily soot) within a pulverized coal flame in real time. For the first time, in situ soot measurements have been obtained at several operating conditions allowing a greater understanding of the quantity of soot in these flames as a function of equivalence ratio and flame swirl. The method has been presented to the coal combustion community in the form of technical presentations and technical

papers are in progress. Funding for the project was also used to support a Masters thesis graduate student who will shortly graduate. Given the short nature of this study, the results have not yet been published in an archival publication, but an abstract has been accepted for presentation as listed below and publication of the results is in progress as also outlined below. After the results are presented at the Combustion Institute meeting as listed, a paper will be submitted to an archival journal for publication.

1.2 Publications and Presentations

Tree, D.R. and Peart, J.A. (1999) "In Situ Soot Measurements in a Pulverized Coal Flame," Western States Section, The Combustion Institute, Oct. 1999.

Tree, D.R. and Peart, J.A. (1999) "On Line Measurement Of Primary Fine Particulate Matter," DOE Contractors Review Meeting, June 1999.

Tree, D.R., Peart, J.A. (1999) "In-Situ Species, Temperature, Soot and Velocity Measurements of Pulverized Coal Flames with Advanced Reburning" ACERC Annual Conference, Feb. 26, 1999.

1.3 Graduate and Undergraduate Students Supported

Jacob A. Peart, Master's Degree, December 1999.

2 INTRODUCTION

Over fifty-six percent of 1998's electricity was generated using coal (Mining Engineering, 1999). Coal is not only used to produce electric power but also steam for heating and industrial processes, and cooling using the ammonia cycle. During the coal combustion process several particles are formed: soot, ash, and char. The soot particles play an important role in chemical kinetics and heat transfer. Although temporarily useful for radiative heat transfer, it is desirable that soot particles burn completely before leaving a coal reactor. In the past, soot burnout has typically not been a problem. However, recent innovations in low-NO_x burners increase the fraction of soot and char found in the ash at the exit of the furnace (Veranth, 1999).

Soot is formed from gases or volatiles evolved from the coal in the absence of sufficient oxygen. This typically occurs near the primary inlet when coal first enters the reactor. The tiny soot particles play a major role in radiating heat to the walls of the reactor (Menguc and Webb, 1993). Figure 1 shows the relationship between the quantity of soot and the soot emissivity. The figure demonstrates that soot quantities below a volume fraction of 1×10^{-8} will produce an emissivity below 0.01 while soot volume fractions above 5×10^{-6} are relatively opaque. Soot that escapes the reactor and is released into the atmosphere can be lodged in the lungs. The soot itself may be relatively harmless, but the carbon in soot acts like a sponge absorbing carcinogenic hydrocarbons (Veranth,

1999). Soot trapped with the ash by the scrubbers and bag houses increases the carbon fraction in the ash making it unmarketable. Soot is also unsightly. The presence of soot is fairly obvious within a coal flame due to the high luminosity and distinctive yellowish color, but soot is difficult to detect visually as the product gases cool. This work investigates an in-situ method for detecting and quantifying soot in a pulverized coal reactor.

Figure 1. Soot Emissivity as a function of Soot volume Fraction. Temperature = 1600 K, Mean Beam Length = 0.75 m. Adapted from Adams (1988).

3 BACKGROUND

Pulverized coal combustion involves several steps and is too complicated to describe in detail here but a simple explanation follows which is important for understanding the method used here to measure fine particulate. Coal particles enter the combustion chamber where they are heated. As the particles increase in temperature, volatiles within the coal are released and expand rapidly forming a small cloud around the coal particle. The coal particle, depleted of its volatiles, cracks and swells becoming a char particle. The devolatilized coal contains primarily carbon with small fractions of minerals. The char continues to react, burning out the carbon. Eventually, all that is left of the char are the inert minerals. Depending on the temperature, the minerals melt, vaporize, or simply maintain their initial shape and size. Ash that melts or vaporizes may later form into spherical particles. The volatiles released during the initial heating of the coal particle either oxidize or chemically combine to form tiny, *primary* soot particles that can stick together forming chains and clusters, or soot agglomerates. Primary soot particles are spherical in shape while the agglomerates they form can take on various shapes and aspect

ratios. After further congregation, soot agglomerates become soot aggregates or bigger chains and clusters of primary soot particles.

One of the major obstacles to measuring soot in a pulverized coal flame is the possible presence of four different particles during different stages of combustion. Although the particles are present concurrently, their physical properties differ, most notably, their size. Coal particles range in size from 50 to 100 μm . Canadas (1990) reports that ash particles have a bimodal size distribution with one mean at 0.1 μm and another at 10 μm . Char particles sizes are on the same order of magnitude as the coal particles initially, they then burn and fragment. The smallest char particles however will still be larger than the smallest ash particles. *Primary* soot particles have been shown to range in size from 25 nm to 60 nm (Koylu and Faeth, 1992, Ma et al. 1993). Current methods of soot detection take advantage of the differences in particle size to detect and quantify soot in the presence of coal, char, and ash particles.

There are two basic methods for soot observation, optical and sampling. Both methods have been used to observe soot in laboratory burners and scaled reactors. Soloman et al. (1988) took measurements in a laboratory scale reactor using a Fourier Transform Infrared (FT-IR) method. Soot formation was observed to correlate with the tar fraction in the parent coal. In the regions of devolatilization, where soot was expected, the shorter wavelengths of light were attenuated more than the longer wavelengths. In the regions upstream and downstream of devolatilization, all wavelengths of light were attenuated equally. This suggests that absorption by soot particles is wavelength dependent while absorption by coal and ash is not. Haneberg (1997) used two-color extinction to observe soot in a 0.2 MW cylindrical pulverized coal reactor. He showed that two-color extinction can be used to measure soot volume fraction in a pulverized coal flame. The reported results were estimated to have a total uncertainty of $\pm 50\%$ soot volume fraction. The results were not published due to insufficient data. Instantaneous, soot volume fraction data were not obtained due to high noise levels in the signal.

Ma (1996) used thermophoretic sampling to observe soot formation above a flat flame burner in an oxygen depleted environment. A particle impactor was also used to separate soot and char particles in bulk. Soot yields were again correlated to the tar content in the parent coal. Soot agglomerate and aggregate sizes were shown to increase with increased residence time. Veranth (1998) developed and tested a method of estimating quantitatively the relative mass of soot and char in coal fly ash samples using physical methods of separation. A water-cooled probe was used to collect the samples, and a cascade impactor to separate the particles according to size. The samples were further separated using gravity separation in an ethyl alcohol solution. The method is time-consuming and requires meticulous manual techniques. The data were reported as percent carbon in the fly ash. Soot volume fraction was not determined.

4 METHOD

The proposed mode of soot detection and quantification in this work is the two-color extinction method using two lasers of different wavelength in the visible spectrum. The optical method of particle separation works because the coal, char, and ash particles are orders of magnitude larger than the soot particles. The primary soot particle diameters are

much smaller than the wavelengths of light used in the extinction measurements. While the coal, char and ash particles are much larger. This allows the grouping of the coal, char and ash into one optical group and the soot into another. Mie theory of light scattering indicates that in this case soot will attenuate more light of a shorter wavelength than of a longer wavelength and that the coal, char, and ash will attenuate both the short and long wavelengths the same.

The extinction of light as measured by the transmittance, τ , at wavelength, λ , along a path length, L , for spherical particles of diameter, d , can be characterized by Equation 1. In Equation 1, the extinction efficiency, Q_{ext} , can be determined from Mie theory for electromagnetic radiation for particles of all sizes (Kerker, 1969). The volume fraction of the particles, f_v , along a specified path length, L , can be determined from the measurement of τ_λ , an assumed diameter, and the calculation of Q_{ext} .

$$\tau_\lambda = \exp\left(-\frac{3}{2}\left(\frac{f_v L}{d}\right)Q_{\text{ext}}\right) \quad (1)$$

According to Kerker (1969), the complete solution of Q_{ext} can be simplified for various situations according to the size parameter $\alpha = \pi d / \lambda$. The resulting simplifications are shown in Table 2 below where $m = n + ik$, the complex index of refraction.

Table 2. Summary of Important Characteristics of Q_{ext} for different size regimes.

Size Parameter	Simplified Result	Comment
$\alpha < 0.1$	$Q_{\text{ext}} = 4 \frac{\pi d}{\lambda} \left \frac{m^2 - 1}{m^2 + 2} \right ^2$	Extinction is wavelength dependent (preferential to short wavelength) but independent of diameter.
$0.1 < \alpha < 2$	Full Mie Theory, $Q_{\text{ext}} = f(\lambda, m, d)$	Extinction is preferential to shorter wavelength and also diameter dependent.
$2 < \alpha < 50$	Full Mie Theory, $Q_{\text{ext}} = f(\lambda, m, d)$	Extinction is wavelength and diameter dependent. Wavelength dependence is mixed with regard to diameter.
$50 < \alpha$	$Q_{\text{ext}} = 2$	Extinction is independent of wavelength.

Size distributions for coal, ash, and soot vary from one combustion application to another. Typical mean particle size ranges are shown below in Table 3 along with the value of the size parameter, α , for the given particle type. Literature values for agglomerated soot particles range from 0.8 to 38 μm according to Fletcher et al. (1997), but, sizes near 50 nm are typical for the primary particles. When soot agglomerations have large aspect ratios, they tend to behave as though they are a large number of individual particles at the primary soot diameter rather than one large particle the size of the

agglomerate. Thus, the optical size of soot particles may fall between 50 nm and 1 μm . The mean ash size of 10 μm was selected based on the data of Self (1988). The size parameter has been calculated based on a Helium - Neon laser wavelength of 633.5 nm.

Table 3. Size parameter α , for various particle types existing in coal combustion for a beam wavelength of 633 nm.

Particle Type	Size Parameter $\alpha = \pi d/\lambda$
Primary Soot ($d = 50 \text{ nm}$)	0.25
Soot Agglomerate ($d_{\text{mean}} = 1 \mu\text{m}$)	4.96
Ash ($d_{\text{mean}} = 10 \mu\text{m}$)	49.6
Coal ($d_{\text{mean}} = 60 \mu\text{m}$)	297.5

The table shows that soot is in the region where Q_{ext} is dependent on the wavelength and possibly the diameter of the particle, while ash and coal are independent of the wavelength. Grouping particles according to size, Equation 1 can be written to include “large” particles (coal, char, and ash) and “fine” particles (soot) particles into two optical groups. Transmittance measurements at two visible wavelengths, for example 633.5 and 543.5 nm, can be made and related to the extinction as shown in Equations 2 and 3. For the large particles, $Q_{\text{ext},543} = Q_{\text{ext},633.5}$ and can therefore be eliminated from the equations allowing for the determination of $\left(\frac{f_v L}{d}\right)_{\text{fine}}$ when the transmittance is measured and $Q_{\text{ext},543,\text{fine}}$ and $Q_{\text{ext},633,\text{fine}}$ are calculated from the Mie solution. The technique is qualitative in finding the presence of fine particulate whenever the transmittance of the two wavelengths are not equal and can also be quantitative to the extent that the diameter and refractive index of the fine particulate matter are known.

$$-\ln \tau_{543.5} = \frac{3}{2} \left[Q_{\text{ext}, \text{fine}, 543.5} \left(\frac{f_v L}{d} \right)_{\text{fine}} + Q_{\text{ext}, \text{large}, 543.5} \left(\frac{f_v L}{d} \right)_{\text{large}} \right] \quad (2)$$

$$-\ln \tau_{633} = \frac{3}{2} \left[Q_{\text{ext}, \text{fine}, 633} \left(\frac{f_v L}{d} \right)_{\text{fine}} + Q_{\text{ext}, \text{large}, 633} \left(\frac{f_v L}{d} \right)_{\text{large}} \right] \quad (3)$$

Several objectives of this research involved making the quantitative measurement of f_v more accurate. First, an attempt was made to determine the size of the soot particles more accurately than could be assumed from the literature. Second, an effort was made to improve the accuracy of the transmittance measurement, τ , by selecting two inexpensive light sources which provided enough wavelength separation to produce optimal sensitivity while being free from noise. Finally, an experiment is planned whereby a known f_v of soot is placed in a container of known length, L , and mixed with known quantities of coal, char, and ash to determine if the measurement technique can accurately produce the f_v .

4.1 Method Validation

Several experiments were planned to document and characterize the two-color extinction method's usefulness in obtaining quantitative soot measurements. The activities are listed below. The activities begin by demonstrating the technique works qualitatively and become increasingly quantitative in nature. We are unaware of any previously used calibrated method which can be used for a direct comparison.

1. Simultaneous measurement of soot using the proposed two-color extinction method and visual observations recorded with digital camera images.
2. Simultaneous measurement of the soot using the two-color extinction method and solid particulate sampling of the effluent. Solid samples were then examined using a Scanning Electron Microscope (SEM) and visual inspection of the samples were compared to the optical measurement results.
3. High speed video images of transient soot plumes in the coal flame were taken simultaneously with the two-color extinction images to compare visual and two-color results.
4. A study of soot concentration vs. equivalence ratio was obtained to determine if measured two-color soot concentrations reached levels consistent with tar yield for the coal flame.
5. A study of soot concentration vs reactor swirl was completed to characterize the location of soot in the reactor at different operating conditions.
6. An experiment is planned whereby known concentrations of soot and ash will be mixed and the measured two-color soot concentration will be compared to the known value.

5 EXPERIMENTAL APPARATUS

A down-fired, 0.2 MW, variable swirl, pulverized coal reactor was used to produce a flame for testing the two-color extinction technique. A schematic diagram of the reactor is shown in Figure 2. The reactor is 2.7 m in length and 0.76 m in diameter. Pulverized coal was fed using a variable speed auger into a primary air tube with 2.54 cm inside diameter. Secondary air was passed through a movable block swirl generator before exiting the burner concentric to the primary tube. Below the swirl section, six identical sections, each 40 cm in length were stacked axially followed by an outlet section. The six reactor sections each contained four windows located 90 degrees apart, which provided optical access. A blower on the exhaust end of the reactor pulled the effluent gas through the scrubber. Opening a valve at the blower inlet caused room air to be drawn through the blower rather than the reactor exhaust which produced a positive pressure in the reactor. Slightly positive pressures of 0.2 mm Hg were used during reactor testing in order to keep O₂ from leaking into the reactor.

Baseline operating conditions for the reactor are shown in Table 4. For studies investigating a change in fuel equivalence ratio, the fuel flow rate was held constant while the mass flow of secondary air was increased. Air temperature was held constant for all of the tests. Coal used in the tests was a Wyodak, sub-bituminous rank. Proximate and

ultimate analysis for the coal are shown in Table 5. The coal was fairly high in moisture, low in ash and sulfur. The volatile fraction (34%) is of particular interest in this study because it is the source of soot precursors.

Figure 2. Schematic diagram of pulverized coal reactor, fuel feed, and exhaust system.

Table 4. Operating conditions for the detailed data map.

Parameter	Flow Rate (kg/hr)	Temperature (K)
Primary Air	16.2	310
Secondary Air	176.5	600
Coal	25.6	300

Table 5. Ultimate and proximate analysis of the Wyodak sub-bituminous coal.

Proximate					
Description	<u>Moisture</u>	<u>Ash</u>	<u>Volatiles</u>	<u>Fixed Carbon</u>	<u>Heating Value (kJ/kg)</u>
As Received	25.3	5.1	34.0	35.6	20,830
Ultimate					
Description	<u>C</u>	<u>H</u>	<u>N</u>	<u>S</u>	<u>O By Difference</u>
As Received	53.32	3.47	0.74	0.36	11.74

The optical set-up for the two-color transmittance measurements is shown schematically in Figure 3. The selection of the laser wavelengths and the optical detector set-up were principal objectives of this study, therefore, details will follow in the next section concerning the reasons for the wavelengths used. Two helium-neon (HeNe) lasers at wavelengths of 633.5 nm (red) and 543 nm (green) were transmitted through open window ports in the coal reactor from a transmitting optics table. On the transmitting side, the beams were combined into a single coincident beam using a low-pass filter which reflected light at the longer red wavelength (633.5 nm) and transmitted light at the shorter green wavelength (543 nm). Both beams then passed coincidentally through the flame zone in the reactor. On the receiving side, the beams were split at a second low pass filter which transmitted the green beam while reflecting the red wavelength. Once split apart, the beams were reflected onto photodetectors which produced a voltage proportional to the beam's intensity.

The detectors were tested using various neutral density filters of known transmittance and shown to be linear in previous experiments (Haneberg, 1997). Two optical components were placed in the path of the beams to reduce unwanted radiation from reaching the detectors. Most importantly, a narrow band pass filter was placed in the path

Label	Element	Information	
A	He-Ne laser	543.5nm	0.75mW
B	Low-pass filter	600nm cutoff	
C	Mirror		
D	Collimator	TSI 9108-2	
E	He-Ne laser	633nm	1.5mW
F	7/16 inch orifice		
G	Polarizer		
H	Low-pass filter	600nm cutoff	
I	Band-pass filter	633nm center	1nm
J	Opal glass diffuser		
K	Photodiode	UDT Sensors	PIN-25D
L	Mirror		
M	Band-pass filter	544nm center	10nm
N	Opal glass diffuser		
O	Photodiode	UDT Sensors	PIN-25D

Figure 3. Two-color extinction measurement system as viewed from the top of the reactor.

of light just prior to entering the detector. This reduced broadband radiation from the flame to the bandwidth of the optical filter. Secondly, a polarization lens was placed in the path of the beam. Light from the laser is theoretically completely polarized while light from the flame, reactor wall, or from multiple scattering would tend to be depolarized. The polarizer therefore helps to eliminate light which did not originate from the laser or laser light which has been depolarized through multiple scattering off of coal and char particles.

6 SELECTION OF OPTICAL COMPONENTS

The most important decision regarding optical components involves the selection of the wavelengths and light sources used for the transmittance measurements. The following list of information was considered in making this decision.

- The ideal wavelength for the transmittance measurement would produce an α of 0.1 or less for the soot particles and an α greater than 50 for the coal, char, and ash particles. As is discussed in section 4 and demonstrated in Tables 2 and 3, this would allow the transmittance measurements to produce different optical characteristics for the two groups of particles.
- Fluctuations in the light source must be minimized. Light source fluctuations produce fluctuations in the transmittance which produce uncertainty in the measurement. Two different types of fluctuations are prevalent in most light sources, RMS noise and drift. Both should be minimized for accurate measurements.
- Broadband radiation from the flame peaks at approximately a wavelength of 2000 nm (assuming a flame temperature of 1600 K). Selecting shorter wavelengths for the transmittance measurements can reduce the unwanted light from radiation.
- The light source should be as inexpensive and durable as possible for full scale application.
- While not necessary, polarized light provides a better opportunity to filter out light produced by combustion and light generated through multiple scattering.
- Transmittance wavelengths should avoid the absorption bands of gases involved in the combustion process such as CO₂, CO and H₂O.
- Lasers are available at only a limited number of discrete wavelengths.
- Visible light is preferable to infrared and ultraviolet because it is easier to work with when aligning optics and because detectors, mirrors, lenses and other optical components are not as efficient and are more expensive outside of the visible range.
- A beam of light must traverse the measurement volume and not expand beyond the area of the receiving optics.

Given the above mentioned information and criteria, a survey of available laser and diode light sources was completed. A list of potential light sources is shown in Table 6 listing costs and characteristics. Only one example of each light source is represented, and only light sources costing less than \$10,000 are represented. Argon ion lasers are capable of producing light at several wavelengths and can be used for multi-wavelength or single wavelength applications. Multi-wavelength operation is desirable because the two beams are already coincident.

Table 6. Commercially available lasers and their characteristics.

Type	Wavelength	Power (mW)	Beam Divergence (mrad)	Beam Diameter (mm)	Pointing Stability (mrad)	Noise RMS	Drift RMS	Cost \$
Ar ⁺	457-514	25	1.0	0.66	0.01	3%	± 1%	4,000
Ar ⁺	488	10	0.95	.67	± 0.1	3%	± 1%	4,000
Ar ⁺	514.5	2	1.26	0.63	0.03	NA	NA	4,600
He Ne	543.5	0.25	0.98	0.70	0.02	0.25%	0.25%	850
He Ne	633	2	1.3	0.63	0.02	0.1%	2.5%	650
Diode	633	.9	.5	.1	0.1	NA	NA	500
Diode	635	1	1	1	NA	NA	NA	850

In the methods section, a discussion was given concerning the importance of the size parameter, α , in using transmittance measurements to optically separate the soot from the coal, char, and ash. Ideally, the transmittance wavelengths should be selected to make α less than 0.1 for the soot and greater than 50 for the coal, char, and ash. Table 3 demonstrates that the particle diameters are not different enough to produce the desired separation, therefore a compromise was sought. The example shown in Table 3 shows that a transmittance source at 633.5 nm is reasonable. At 633.5, α for soot is 0.25 which is larger than the desired value of 0.1, and α is 49.6 for 10 μ m ash particles which is close to the desired value of $\alpha > 50$. The experiments proved that moving to longer wavelengths was not desirable because of increased background radiation from the flame.

The two-color method requires that two light sources be selected. While both need to meet the above stated criteria for α , the greater the separation of the two wavelengths, the greater the difference in the transmittance of the beams through small particles. Thus a greater separation in beam wavelength produces a greater sensitivity to the presence of soot. While tunable lasers are available, they are costly, difficult to maintain and potentially dangerous when for use in industrial applications. Therefore, only inexpensive, low-energy lasers were considered. The existence of only a few wavelengths among these laser sources allows only a limited number of combinations. The available wavelengths from argon ion, helium neon, and diode lasers are given in the Table 7. The shortest wavelength is from an argon ion laser at 454.5 nm and the longest is from a diode at 635 nm. The diode laser did not have a noise level listed by the manufacturer and its wavelength was only slightly higher than the wavelength available from a HeNe laser at 633.5 nm, therefore, the HeNe laser at 633.5 nm was selected.

The shortest wavelength available was from a multi-wavelength argon ion laser. The multi wavelength, argon ion laser introduced two problems. The first was that the power was controlled for the sum of all wavelengths but not for the individual wavelengths. This meant the beam power of individual wavelengths could possibly drift. The second was that initial testing of a water cooled argon ion laser showed 6% RMS noise caused by vibrations from the water jacket surrounding the plasma tube. An air cooled argon ion laser at a single wavelength produced less RMS noise but was only available at 488 and 514.5 nm. This meant that the best possible readily available combinations were a 633 nm HeNe laser combined with either a 488 nm air-cooled, argon ion or a 543.5 nm HeNe laser.

An error analysis showed both combinations would produce similar error with the assumed RMS noise of 3 and 0.25% for the argon ion and HeNe lasers respectively. Since the HeNe laser was less expensive, it was selected. If the argon ion laser produces less than 3% RMS noise, it would be preferable to the HeNe laser used in this study.

Table 7. Source laser wavelength options.

Light Source	Wavelength (nm)		
Argon Ion Laser	454.5	458	466
	473	477	488
	497	502	514.5
Helium Neon	543.5	594	612
	633	1523	
Diode	635	670	

7 RESULTS

7.1 Transmittance vs. Equivalence Ratio

Transmittance measurements were taken near the exit of the reactor 232 cm below the primary inlet tube, then used to calculate soot volume fraction. In order to obtain soot volume fraction from the extinction measurement the following assumptions were made: 1) The soot was assumed spherical with a diameter of 30 nm. 2) The refractive index used for soot was that of Charalampopoulos and Chang (1998) ($n = 1.43$, $k = 0.30$ at 543 nm). 3) The extinction efficiency of the ash, char, and coal at both wavelengths are equal. 4) The particles are confined to the path length of the reactor (.75 m) and the soot volume fraction obtained is an average for the entire path length. The equivalence ratio was varied by changing the air flow rate while holding the coal mass flow rate constant.

The results, comparing soot volume fraction to equivalence ratio are plotted in Figure 4. The values shown in Figure 4 were taken on two days. All of the conditions except an equivalence ratio of 1.0 and 1.5 were repeated to assure the repeatability of the measurement system. The measurement showed no soot under lean conditions with a small amount of soot appearing (2×10^{-8}) when the reactor became overall rich at $\phi = 1.1$. The soot concentration appears to have a slightly "S" shaped curve increasing in soot concentration rapidly above $\phi = 1.1$ and increasing more slowly after $\phi = 1.4$. At $\phi = 1.4$, the transmittance of the beam was very low, often appearing completely opaque. It is uncertain whether the decrease in the rate of soot concentration is caused by less soot formation or by error in the measurement caused by a completely attenuated signal. The maximum soot volume fraction measured was 88×10^{-8} . An upper limit on soot volume fraction can be estimated by assuming all of the volatiles in the coal form soot. Assuming soot has a density of 2 g/cm^3 and the combustion products are at a temperature of 1350 K at the measurement location, the maximum soot volume fraction for the measured operating

condition is 5.0×10^{-6} . Considering the fact that not all of the volatiles are tar and prone to produce soot, the measured values seem reasonable. In order to determine if higher concentration of soot exists at higher equivalence ratios, a smaller path length would be needed so that the beam would not be completely attenuated.

Figure 4. Soot volume fraction plotted as a function of equivalence ratio.

7.1.1 Comparison with video

When soot is hot, its radiation in the visible spectrum can be seen as yellow-white flashes in a pulverized coal flame. To help qualitatively determine the usefulness of the method, transmittance data were taken concurrently with video images. Simultaneous imaging and extinction measurements of soot were obtained by allowing the laser light to pass through a pinhole in a mirror which was imaging the same path length that the beam was traversing. A sequence of video images taken 1 ms apart were obtained while the transmittance was measured continuously and sampled at 1 ms intervals by the data acquisition program on the PC. Bright flashes of light are an indication of soot at lean equivalence ratio while under richer conditions the soot turns more yellowish and does not appear as bright. The camera records only black and white images and gray scales in between. Soot is more luminous or whiter than the wall under lean conditions but becomes cooler because of lower flame temperature under rich conditions and begins to approach the same color as the wall. Another indication of soot to be observed in the video images is the opacity of the picture. Soot is a very good absorber of light and therefore when an image becomes opaque it is a good indication of soot.

The bright flashes seen under lean conditions were compared to changes in the transmittance ratio obtained from the optical measurement. For unknown reasons the visual flashes and changes in transmittance ratio did not always occur simultaneously. However, the occurrences of soot and flashes of light did have approximately the same

frequency. Also, when conditions of no soot were recorded visually, transmittance ratios agreed.

An example of the observed behavior is shown in Figure 5. which shows several images recorded by a high speed video camera under lean or no soot conditions, slightly rich or intermittently sooty conditions, and very rich continuously sooting conditions. Figures 6-8 show the corresponding transmittance data for the equivalence ratios shown in Figure 5. Figures 6-8 record 1 second of transmittance signals or 1000 data points. It is not practical to show 1000 video images so sampling of images every 0.2 seconds has been displayed. Three equivalence ratios are represented. The first two rows of images are circular in shape because the camera is looking into a circular port with the laser passing through the center of the port. The third row of images was not taken at the same time as the transmittance measurement and was taken through a larger rectangular window. The size of the circular image is approximately 5 cm while the rectangular image is larger at about 33 cm long and 10 cm wide.

In the first row of images there is no change in the luminosity at any recorded time. The image shows an octagonal shape which is the aperture of the window on the near inside wall. The small black hole is a smaller aperture on the opposite side of the reactor approximately 1 cm in diameter through which the laser beam exits. The light shading surrounding the dark hole on the opposite side of the reactor is luminosity from the hot wall surface.

Images for an equivalence ratio of 1.0 shown in the second row demonstrate the periodic soot excursions. At 0.0 seconds there is a bright flash of soot over a large portion of the imaged area. The black hole on the opposite side of the reactor is visible but not easily defined suggesting soot obscuring the view. In the second image, at 0.2 seconds, a bright flame of soot totally obscures the opposite wall. At 0.8 seconds, the image contains no soot and looks just as the image in the frame above. In the final row of images a rectangular window can be seen with cloud like whitish images throughout the window area. A dark image of the rectangular window on the opposite side of the reactor is not visible in any of the images. At this condition soot was present at all times and the rich condition produced a lower flame temperature and much lower luminosity. The flame was orange yellow in color indicating large amounts of soot.

Corresponding transmittance measurements agree well with the video images. In the first frame, of Figure 6, the signal from the two lasers are on top of each other. The transmittance changes but equally at both wavelengths. This indicates that coal and ash are fluctuating in the turbulent quasi-steady flow of the reactor but there is not soot. In Figure 7, transmittance is again fluctuating but during the dips in the transmittance signal, the shorter green wavelength is attenuated more indicating the presence of soot. Note that when soot is intermittent, visually it appears intermittent in the transmittance as well, but the transmittance images showing soot do not always coincide with a dip in the transmittance. The intermittent transmittance signal suggests that the flame is unsteady and turbulent, probably due to variation in the coal feed rate and turbulent mixing of the coal with the air but the presence of soot does not always coincide with these bursts of fuel and flame passing across the field of view. In Figure 8, the green beam is seen to be continuously lower than the red beam, indicating the continuous presence of soot as seen in the video images.

Figure 5. Sequences of video images for the pulverized coal flame under non sooting ($\phi = 0.9$), intermittent sooting ($\phi = 1.0$) and continuously sooting ($\phi = 1.2$) conditions.

Figure 6. Transmittance for the red and green beams through a pulverized coal flame at $\phi = 0.9$, a non sooting condition.

Figure 7 Transmittance for the red and green beams at an intermittent sooting condition ($\phi = 1.0$).

Figure 8. Transmittance measurements for the red and green beams for a continuously sooting condition ($\phi = 1.2$).

7.1.2 Comparison with effluent solid samples

Solid samples were taken from the exhaust of the reactor for comparison with transmittance ratio measurements taken at the reactor exit. At the exit, the combustion gases were quenched with a water spray. The spray cooled the product gasses and removed particles from the gas stream. The particles, in solution with water, were collected using a gravity driven water trap. The resulting slurry was studied and photographed then filtered to remove the particles. SEM images were taken of the filtered particles.

Figure 9 shows particles taken in solution from the exhaust line of the coal reactor at various equivalence ratios. Dark particles are char or soot particles while lighter gray colored particles are ash. The particle solutions become increasingly dark as the equivalence ratio increases from 0.9 to 1.5. In solution, smaller particles like soot remain suspended longer than larger particles. These samples were shaken just prior to the photograph, yet settling in the 0.9 through 1.2 equivalence ratio solutions was already significant as evidenced by a layer of gray particles on the bottom of the containers. This indicated the presence of a significant fraction of char and ash particles. Although it is difficult if not impossible to quantify what is shown in Figure 9, it can easily be seen that there exists very few if any soot and char particles in the solutions of 0.9 and 1.0 equivalence ratio. This conclusion is in agreement with the measured values shown in Figure 4. At 1.0 equivalence ratio, a small number of dark particles can be seen. An increasing number of dark particles can be seen as the equivalence ratio increases from 1.2 to 1.5. The equivalence ratio at which soot begins to survive the combustion process is in quantitative agreement and the color of the particulate samples is in qualitative agreement with the two-color extinction data shown in Figure 4.

Figure 9. Particle samples in solution labeled according to their equivalence ratios.

7.1.3 Comparison with SEM

Effluent particles were examined using a Scanning Electron Microscope (SEM) in an attempt to determine primary soot particle size and confirm qualitatively the presence of soot. The results of the SEM images were however difficult to interpret. Particles of all sizes and shape could be seen at all operating conditions sampled making the selection of a representative image subjective. One complication of the SEM images was the poor depth of focus available at high magnification. A representative picture of a lean operating condition at $\phi = 0.9$ is shown in Figure 10. These images show very little evidence of small particles in the range of 50 nm or smaller (This image is at the same scale as Figure 11). The large particles consisting of a fabric of interconnected strands is an unknown material. The circled particles (150-200 nm) are spherical in shape but are larger than the 30 – 50 nm expected for primary soot particles. If the circled particles are soot, there are very few of them and as it was difficult to find any particles of this size within the sample.

Figure 10. SEM image of exhaust particles taken at an equivalence ratio of 0.9.

In Figure 11, three of many small particles are circled in an image magnified 40,000 times. In this photograph, large particles are also present which may be agglomerated soot, char, or ash. Because of the high magnification, the surface of the large particles can not be seen clearly making it difficult to determine if they are made up of primary soot. The circled particles are spherical and close to 50nm in diameter, the approximate size of primary soot particles. The figures suggest that soot is not present at low or lean equivalence ratios while it is present under rich conditions. This matches qualitatively the

optical extinction measurements obtained. The size of the small particles is too difficult to accurately determine. Statistical processing of the images for size distribution was not done and would be difficult given the broad size distribution in the sample and the difficulty in focusing particles of various size.

Figure 11. SEM image of exhaust particles taken at an equivalence ratio of 1.5.

7.2 Axial Profile

The soot volume fraction changed according to axial position in the reactor as shown in Figure 12. The axial values are referenced to the primary fuel inlet tube. The equivalence ratio used for the axial profiles was 1.2. At the first measurement, 30 centimeters in the axial direction, a significant amount of soot has already formed, particularly at 1.5 swirl. At all three swirl conditions, the soot volume fraction increases as the flow moves down the reactor. The soot volume fraction measured with a swirl of 0.5 and 1.5 appears to approach an asymptote at an axial position somewhere between 80 and 112 cm. This suggests that the soot volume fraction reaches a steady state condition. The soot volume fraction appears to still be climbing at 112 cm for the swirl of 0 case. The results agree well with previous measurements of species, temperature, and velocity obtained in the reactor (Nazeer et al. (1999)). At 0 swirl, the flame is long, and lifted, penetrating well into the reactor. Air is entrained into the primary fuel air mixture during devolatilization oxidizing soot precursors. At 0.5 swirl the flame has been shown to attach to the quarl and shorten creating a fuel rich zone conducive to soot formation just below the primary fuel inlet tube. As swirl is further increased to 1.5, combustion products and

secondary air mix more rapidly with the primary air, further shortening the devolatilization zone while also increasing the oxidation of soot precursors. While understandable from the previous species, velocity, and temperature data it is somewhat surprising that the 0.5 swirl soot volume fraction is twice as large as the 1.5 swirl data, but this result was repeatable. The data suggest that region where the tar is evolved in the 0.5 swirl case is much richer than the 0 and 1.5 swirl cases.

In the 0.5 and 1.5 swirl cases, the soot volume fraction reaches a steady value and does not increase further after 90 cm. This is an expected result signifying the point where all of the gas phase precursors have been converted to solid carbon leaving no source for an increase in soot mass with no oxygen available for soot oxidation. Even more significant to this study however is the fact that the steady soot volume indicates the soot size is not increasing. When calculating the soot volume fraction from the extinction data, the soot diameter was assumed to remain constant. If the diameter were changing, the assumption of a constant diameter would result in a change in calculated soot volume fraction. The change in maximum soot volume fraction with swirl also indicates that although the tar fraction in the coal controls the maximum amount of soot possibly formed, oxygen in the devolatilization zone can significantly reduce the soot formed.

Figure 12. Soot volume fraction plotted against axial position at three different swirls and an equivalence ratio of 1.2.

8 FULL SCALE APPLICATION

One of the great advantages of soot detection in a coal flame using two-color extinction is its simplicity. The lasers, detectors, and other optical equipment are inexpensive, rugged and readily available. Implementing the method requires only a hole in the reactor through which the beams can pass, and something to secure the optics. Another significant advantage is that the data can be received in real time and possibly used to control fuel air ratio or the fraction of air being injected in different ports of a

reactor. Nevertheless, significant obstacles exist before implementation in a full scale reactor can be realized.

First it must be realized that the path length in a full-scale reactor is 10-20 times longer than was used in this study. Soot concentrations equivalent to those measured in the test facility would completely attenuate the beam before it could cross the reactor. The longer path length would however make small concentrations of soot more easily detected. Obscuration of the beam is a fundamental limitation of the method which limits the range of usefulness but if the limitation is understood it does not effect the validity of the results.

A second significant draw back to the method is the tendency of the beam to drift in power and the detectors to drift in sensitivity. Beam power fluctuations can be monitored and corrected by splitting the beam and placing a detector on both sides of the measurement volume. This technology is used in the laser itself to measure and control beam power. Part of the beam is reflected to a detector and the power of the reflected portion is continuously monitored and changed to obtain a steady value. Similarly, the beam power entering the measurement volume could be continuously monitored and the unattenuated value corrected accordingly. Changes in detector sensitivity are more difficult to control. The largest cause of detector drift is normally temperature change. Detectors could be temperature controlled and multiple detectors could be used in order to reduce this problem. Similar to beam power fluctuations, window fouling can reduce transmittance with time. Recessed windows with air cleaning could be used but this problem introduces an additional source error. In this project, the flame was open to the atmosphere and windows were not used. Developing a system capable of automatically adjusting for beam power drift, detector drift, and window fouling was beyond the scope of this project.

Although significant, neither of these problems would appear to eliminate the usefulness of this technique in full scale boilers. The technology and hardware exist for the development of this method.

9 CONCLUSIONS AND RECOMMENDATIONS

A method has been developed and tested for the measurement of integrated soot volume fraction along a line of sight in a pulverized coal flame. The method has been demonstrated in a 0.2 MW, down-fired, variable swirl reactor. The results showed that coal, char, and ash could be treated optically as a grouped of particles separate from soot where the coal char and ash attenuate visible light equally at all wavelengths but the soot preferentially absorbs the shorter wavelengths. Soot was found to form intermittently at all equivalence ratios and swirl numbers but was readily oxidized at equivalence ratios 1.0 and below. The soot volume fraction measured from the optical extinction technique agreed qualitatively with visual measurements of soot captured on high speed video and visual and SEM photographs of solid soot samples. The detection limit of soot volume fraction is approximately 1×10^{-8} . The maximum soot volume fraction measured at $\phi = 1.5$ and 0.5 swirl was 0.88×10^{-6} . Soot volume fraction increased with increasing equivalence ratio. A maximum soot volume fraction was not observed but the rate of increase in soot volume fraction with increasing equivalence ratio slowed above $\phi = 1.4$. Spatially, soot was formed in the upper portion of the reactor and soot volume fraction increased to a constant

value as the fuel proceeded downstream. The steady value of soot volume fraction reached in the reactor was dependent on the swirl in the burner. A lifted flame produced the lowest soot volume fractions while a low swirl, attached flame produced the highest soot concentrations. The constant soot volume fraction in the lower half of the reactor indicate that the effective optical soot diameter is not changing with time as the soot flows downstream.

The two-color extinction technique appears to be a viable method for measurement and control of soot in full-scale industrial furnaces. The light sources are relatively inexpensive, durable, easily obtained and easily maintained. There are no unusually power requirements and little training would be needed for system operation. It is recommended that the method be further investigated by demonstrating its usefulness in a full-scale facility. Use of the technique is also recommended for the measurement of soot production trends in pulverized coal flames as a function of operating condition and coal type.

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