

Impacts Of Unburned Carbons And Activated Carbon Injection On Particulate Control Devices

Cameron E. Martin

ADA-ES, Inc. 8100 SouthPark Way, Unit B, Littleton, CO 80120

Victor H. Belba

Belba and Associates, P.O. Box 11315, Boulder, CO 80301

Stephan A. Johnson

Quinapoxet Solutions, 50 Nashua Road, Suite 112, Londonderry, NH 03053

Ralph Altman

Electric Power Research Institute, 5726 Marlin Rd., Chattanooga, TN 37411

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ABSTRACT

Predicting the performance of particulate control devices (PCD) such as electrostatic precipitators (ESP) and fabric filters (FF) has become an established exercise for pulverized coal-fired applications. However, uncertainties have been introduced to this task with the recent application of low-NO_x burners (LNB) and future use of activated carbon injection (ACI) for mercury control.

The purpose of this study is to determine how the increase in particulate carbon loading from low-NO_x combustion technologies and ACI might affect the performance of PCDs. The work is concentrated on determining the properties of unburned carbons and activated carbons that enter and escape PCDs. A literature search was conducted to determine the existing knowledge base and to identify gaps in the knowledge.

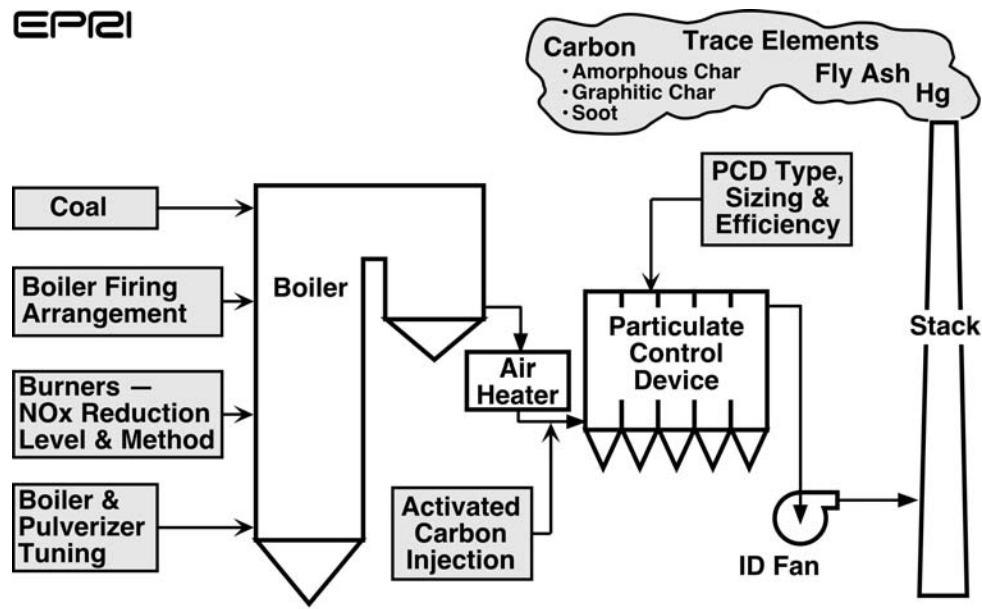
An additional goal is the development of a test program, to be conducted under a separate phase of this project, so that the gaps in the knowledge base can be closed. This new knowledge will allow PCD performance models to be improved while characterizing the properties of the carbon emissions resulting from the implementation of low-NO_x combustion and activated carbon injection technologies.

INTRODUCTION

Predicting the performance of particulate control devices (PCD) such as electrostatic precipitators (ESP) and fabric filters (FF) has become an established exercise for pulverized coal-fired applications. However, uncertainties have been introduced to this task with the recent application of low-NO_x burners (LNB) and future use of activated carbon injection (ACI) for mercury control.

The fact that unburned carbon content has increased in the fly ash from low-NO_x burner installations is well documented. There have also been numerous cases in which ESP performance and stack opacity have been negatively impacted by the retrofit of low-NO_x combustion technologies such as low-NO_x burners. It is generally believed that due to its low electrical resistivity, carbon particulate is subject to reentrainment and is difficult to collect in ESPs. Despite these general observations, there has been very little research toward understanding the problem.

Unburned carbon has been the subject of a considerable amount of research to improve combustion performance and fly ash utilization options. However, since most information in the literature appears to have been gained by analyzing fly ash samples from economizer and PCD hoppers, the actual magnitude of the increase in carbon in the flue gas entering and leaving the PCD is not well defined. Also, the properties of the increased carbon, such as particle size, shape, density, surface characteristics, resistivity, cohesivity, and chemical composition and its interaction with the mineral fly ash components are not necessarily known in sufficient detail to allow prediction of the effects on PCD performance. Additionally, different combinations of boiler/furnace and low-NO_x burner designs and arrangements, as well as operating settings, may have differing affects on unburned carbon quantities and properties. Figure 1 summarizes these suspected causes and effects of increased carbon carryover.



Factors Affecting Carbon Emissions

Figure 1. Factors Affecting Carbon Emissions

The purpose of this study is to determine what is known about, and to establish the gaps in the knowledge base regarding, particulate carbon loading to and from PCDs and the carbon's properties. This investigation includes the effects of low-NO_x combustion technologies and ACI. An additional goal is the development of a test program, to be conducted under a separate phase of this project, so that the gaps in the knowledge base can be closed. This new knowledge will allow PCD performance models to be improved while characterizing the properties of the carbon emissions resulting from the implementation of low-NO_x combustion and ACI technologies.

INFORMATION REQUIRED TO PREDICT PCD PERFORMANCE

Electrostatic Precipitator Performance Prediction

Various proven mathematical and computerized models can reasonably predict the performance of ESPs collecting fly ash from coal-fired boilers using conventional burner technology in use prior to the implementation of low-NO_x burners. However, the increased unburned carbon due to low-NO_x burners and other combustion modifications has added uncertainty to such predictions.

Typical information needed to evaluate or predict an ESP's performance includes:

- ESP size (SCA);
- ESP Design – e.g., Plate Ht, Spacing, etc.;
- ESP sectionalization, TR Ratings, etc.;
- ESP flow distribution, hopper sweepage, etc.;
- Coal and Ash Analyses;
- PM (fly ash) inlet loading;
- Particle Density (skeletal or true);
- Aerodynamic Particle Size Distribution (PSD);
- ESP Electrical Performance Parameters; and
- Rapping Reentrainment.

For most ESP installations, many factors such as ESP size, design parameters and coal and ash analyses are known. For a traditional fly ash, the remaining performance parameters typically can be calculated based on empirical models. However, it is likely that an increase in carbon carryover with fly ash will change the nature and properties of the dust significantly. The concern is that these changes may limit the ability of current performance models to effectively predict ESP performance.

Also, there is concern that increased carbon, with significantly different parameters than traditionally encountered with normal fly ash (e.g., particle density, shape, PSD, resistivity), can significantly affect such ESP performance parameters as rapping reentrainment and hopper sweepage. These empirical correction factors to the models may have to change to represent ESP performance in high carbon loading conditions.

Research done by an EPRI member¹ on several pulverized coal (PC)-fired units suggests that previously unexpected phenomena may contribute to reentrainment of carbon from ESP hoppers. This same work suggests that carbon, in this case measured as loss on ignition (LOI), is enriched by up to 600% in particulate matter (PM) samples taken from the ESP outlet versus samples taken from the inlet. PM samples taken from the first ESP hoppers from these units reflect similar, if not slightly lower, LOI contents as compared to PM samples taken from the ESP inlet. These data suggest that carbon is selectively migrating through the ESP. Rapping reentrainment is the suspected mechanism.

Empirical ESP performance models were originally developed under the assumption that mineral fly ash is overwhelmingly the dominant PM being collected. Such drastic increases in carbon content and changes in PM properties from low NO_x combustion technologies require that the empirical performance models be reevaluated and/or revised.

Fabric Filter Performance Prediction

Fabric filter performance models have not reached the level of sophistication of ESP models. FF performance in terms of PM emissions and pressure drop is expressed more in terms of general design guidelines.

The typical information needed to evaluate or predict a FF's performance includes:

- FF size (G/C Ratio);
- FF cleaning method – e.g., RGFF, PJFF, S/D;
- Bag fabric material and details;
- FF/bag configuration and dimensions;
- FF empirical parameters;
- PM (fly ash) inlet loading;
- Coal and ash analyses;
- Dust cohesivity;
- Particle density (skeletal or true);
- Aerodynamic particle size distribution (PSD);
- PM charge and resistivity; and
- Operational history on filter bag.

In general, a FF that is designed well for its operating conditions can be expected to achieve very low PM emission levels. The factor having the greatest effect on PM emissions from a fabric filter is its cleaning cycle. Emissions peak during a cleaning cycle.

The parameter of primary concern has been the pressure drop that develops across the fabric filter over time. A fabric filter's pressure drop is affected by its gas-to-cloth (G/C) ratio, PM inlet loading, particle size distribution, other PM properties, operational history of the FF's bag set and its cleaning cycle frequency. Pressure drop (ΔP) generally increases over time and the cleaning cycle frequency will be increased to maintain ΔP within reasonable levels that can be accommodated by the unit's fans and design limits. In this way, ΔP indirectly affects PM emissions from a FF.

Clearly, any process that increases inlet PM loading or changes the PM's particle size distribution (PSD) or properties can affect the ΔP across the FF and/or its emissions. Therefore, fabric filters have been included in this research as well.

COHPACTM Performance Prediction

Compact hybrid particulate collectors (COHPACTM) typically involve the retrofit of a small, pulse-jet fabric filter (PJFF) downstream of an undersized ESP to reduce PM emissions and opacity. Due to the reduced PM loading from the upstream ESP, the PJFF in a COHPACTM installation typically needs to be only one-half the size of a conventional PJFF sized to treat the full ash loading from a coal-fired boiler. However, its reduced size makes the COHPAC's PJFF pressure drop and emission performance very sensitive to changes in PM properties or increases in inlet loading resulting from ACI or low-NO_x combustion modifications.

These factors need to be incorporated into the COHPACTM performance prediction models² currently in use.

METHODS TO GATHER INFORMATION

The first step in gathering the desired information was conducting a literature search to determine what information was already available regarding the parameters of interest and the test and analytical methods used to obtain the information. The literature search included a review of publications as well as interviews of key individuals with experience in these fields. Then, knowing what is available, the information gaps can be determined and a test program devised to allow the missing data to be developed.

Literature Search

Since the properties of activated carbons are well documented³, the literature search was focused on unburned carbons. While there has been limited work done to address unburned carbon for the specific objectives of this study, much work has been directed toward characterizing unburned carbon for other purposes including low-NO_x combustion optimization and ash utilization. There is also useful information regarding sampling and analytical techniques for similar end uses. This section summarizes some of the current knowledge about carbon properties, measurement and analytical techniques.

A literature search was conducted to determine the current level of understanding of unburned carbon from coal-fired boilers as well as sampling and analytical techniques used to characterize the carbon. The following major categories were considered.

- Sources and root causes of unburned carbon from low-NO_x coal combustion-the fundamentals
- Physical and chemical characteristics of unburned carbons
- Sampling techniques to collect adequate quantities of carbon rich particulate
- Analytical techniques to evaluate carbon properties

RESULTS AND DISCUSSION

Summary of Carbon Properties Knowledge Base

Limitations

The majority of the research conducted on unburned carbons has been directed toward determining the reasons for incomplete combustion and for characterizing the impacts on fly ash utilization^{4,5,6,7}. There has been little work on characterizing the “in-flight” carbon that enters or leaves the PCD. Most of the physical and chemical analyses have been conducted on fly ash samples taken from the hoppers of the particulate collector (ESP, fabric filter, mechanical collector). Only a few studies have been conducted on samples taken from the stack. Therefore, some of the carbon particles, which are of interest to this project, may not have been included in most studies.

Because unburned carbon represents only a fraction of the particulate emissions from a typical coal-fired plant, the carbon samples collected in many of the studies found in the literature were separated and concentrated in order to measure their physical and chemical properties with many established analytical methods. The separation process can alter the size and physical properties of the carbon particles. Some of the techniques only separate the larger carbon particles with no regard to the smaller carbon and soot particles.

Despite these caveats, findings from the research programs referenced herein provide valuable background information to help guide this study and interpret the analytical results.

Sources of Unburned Carbon from Low-NO_x Combustion Technologies – An Overview

The combustion process in coal-fired boilers is extremely complex and cannot be covered in detail in this paper. It is, however, important to understand some basic aspects of why some carbon in the fuel does not burn completely and thus ends up in the flue gas entering the PCD.

In utility boilers firing coal, very small amounts of the fuel carbon pass through the boiler without being combusted, irrespective of the process conditions or the coal type. To put this in perspective, if a 10% ash coal (dry basis) burns to 99% completion, the unburned carbon content of the fly ash will be 9.1%. If burnout is 99.5%, the unburned carbon is 4.8% of the fly ash.

Evidence suggests⁸ that the unburned carbon does not bypass the combustion zone unreacted, but instead undergoes considerable changes depending on the design and operation of the boiler and the combustion system. In particular, conditions that limit NO_x formation also tend to increase the carbon content of the ash.

As soon as the coal enters the hot furnace environment it begins to decompose, and volatile gases evolve and burn leaving behind a porous solid residue called char. Coal minerals (ash) mostly remain with the char during devolatilization and initial combustion. From 50 to 80% of the coal becomes volatile at typical furnace temperatures. Figure 2 shows several forms of carbon that have been identified in fly ash samples using microscopy techniques.

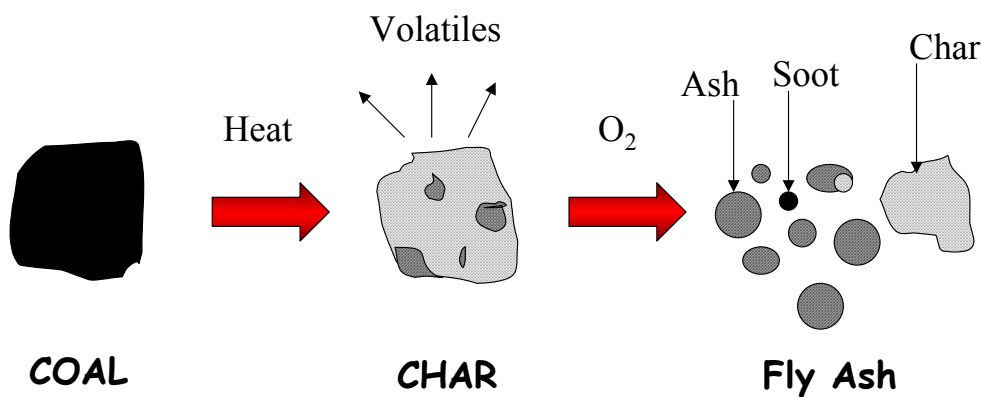


Figure 2. Coal Combustion.

After the volatiles start to burn, the char ignites too. The rate of char combustion is initially controlled by the amount of oxygen in contact with the coal. Oxygen is usually limited in low-NO_x flames (a major factor in unburned carbon formation), so most of the char burnout happens in the post-flame region of the furnace.

Char particles can be either amorphous or graphitic. Amorphous particles are porous and spongy, and often have irregular shapes. They are fairly similar in structure to the raw coal except that there is no volatile matter left in them. Graphitic char is usually smooth and rounded with a higher degree of structural ordering. Chemical reactions that take place during devolatilization leave these particles with less porosity and reactivity. A small fraction of carbon in the ash is attached to the inorganic fly ash particles. This carbon can be either amorphous or graphitic. Conversely, some larger char particles can have inclusions of inorganic material. Surprisingly, there is little evidence of char particles encapsulated by glassy inorganic particles. Under conventional combustion, soot particles should be relatively rare, but tests have shown that a significant amount of soot and soot aggregates can occur during low-NO_x combustion⁹. They are much smaller (some soot particles are in the nanometer size range) than residual carbon particles, and are formed when coal volatiles do not have sufficient air to burn.

Properties of Particulate Carbons

Unburned carbons (chars) typically have low surface areas and are only moderately porous. The pores tend to be larger than the size associated with good adsorbents. Activated carbon sorbents on the other hand have very large surface areas and variable pore sizes giving them high capacities to adsorb a wide range of compounds. Carbon particles emitted to the atmosphere could have mercury or other compounds adsorbed onto their surfaces. A summary of the properties is presented in Table 1.

Table 1. Comparison of Physical Properties – Unburned Carbon vs. Activated Carbon

Property	Unburned Carbon (Char)	Powdered Activated Carbon
Particle Size – MMD	Unknown and variable	15-18 microns
Large Fraction	180-350 microns	120 microns
Smallest Fraction	Submicron soot	2-5 microns
Particle Shape	Amorphous Char – Irregular and porous Graphitic Char – Crystalline and structured Soot – Unknown	Irregular, porous
Surface Area	10-60 m ² /g	500-1100 m ² /g (600m ² /g avg)
Bulk Density	0.26 g/cm ³	0.5 g/cm ³
Skeletal Density	2-3 g/cm ³	2-3 g/cm ³

Analytical Techniques to Characterize Carbon Particles

From the literature^{10,11,12} researchers have analyzed unburned carbon for a variety of physical and chemical properties. Table 2 summarizes the major properties and some of the most common analytical techniques used to characterize them.

Table 2. Physical and Chemical Properties of Particulate Carbons and Analytical Methods

Property	Analytical Method(s)
Particle size	Cascade Impactor, Series Cyclone Microtrac, Elzone 5380 Particle Analyzer, SEM
Particle morphology/petrography	Reflected light microscopy, Scanning Electron Microscopy (SEM), High Resolution Transmission Electron Microscopy (TEM)
Surface Area	BET Nitrogen adsorption
Bulk Density	ASTM D2854-96
Skeletal Density	Helium Pycnometry
Porosity	Mercury porosimetry, Carbon tetrachloride activity
Adsorption Capacity	Iodine number
Oxidation Reactivity	Thermo Gravimetric Analysis
Crystal Structure	Raman microprobe
Carbon Content	LECO Infrared spectrometry, Loss on Ignition (LOI)
Elemental Analysis Trace Elements	X-Ray Photoelectron Spectroscopy (XPS) Time-of-flight secondary ion mass spectroscopy (TOF-SIMS) Ash digestion, Metal analysis
Resistivity	IEEE Standard 548
Cohesivity	Electrostatic Tensiometer (modified)

Sampling Techniques

Collecting samples of unburned carbon particles that enter and leave the PCD can be challenging and expensive. Sampling for particulate at the inlet of the PCD will be much more straightforward than collecting outlet samples. EPA Methods 5 and 17 mass trains, cyclones and impactors can be run in short test periods. Care is needed to obtain large inlet samples without plugging/blinding thimbles and filters. Dilution techniques designed to capture small amounts of PM for SEM and TEM analysis will need to be extremely short in duration. Particulate emissions (PCD outlet) from most power plants equipped with PCDs are very low and present the opposite challenges for sampling. Long sample times will be necessary to collect quantities sufficient for lab tests. In general, plants equipped with ESPs will have higher particulate emissions than plants with fabric filters, but in most cases even ESPs collect over 98% of the incoming particles. Collecting samples large enough to conduct a battery of analytical tests can be time consuming and expensive. It is anticipated that at least 2 to 5 grams of sample will be required. The samples will be a mixture of mineral fly ash and carbon. The carbon fraction may be low in some cases, which might require a separation and concentration process to run some analytical procedures.

The literature search included an investigation of what type of automated or continuous analyzers^{13,14,15} were available to measure and/or collect particulates and carbon from flue gas. While several of these technologies (beta-gauge particulate monitors, nephelometers, black carbon monitors) showed promise, none were considered suitable for the specific needs of the proposed test program without special modifications which are as yet unproven.

Gaps in the Current Knowledge Base

While much is known about the properties of unburned carbons from PCD hopper samples, the data may not be representative of the particulate carbon as it enters a PCD. In order to predict the performance of the PCD, it is important to determine the size distribution, and shape and density of the in-flight particles.

Particle Size Distribution – This is among the most important parameters that determine the performance of both ESPs and FFs. There is insufficient data on the size distribution of unburned carbon in post low NO_x combustion retrofits. While hopper samples confirm the presence of large carbon particles little is known about carbon in the smaller fractions. Isokinetic sampling measurements using impactors or cyclones are necessary to determine the representative in-flight PSD. Further analysis of these samples will be necessary to differentiate between carbon and mineral fly ash for the various size classifications and can be accomplished by light or electron microscopy.

Particle Density – As with a particle's size, its density will influence its behavior in PCDs, especially ESPs. For example, large, low-density char particles have been found in ESP outlet hopper samples. The density of carbon is much lower than that of mineral fly ash. There may also be variation of densities among the different char and soot types. Skeletal density measurements have been conducted on some of the larger char particles, but not on the carbon in the smaller size fractions. Particle density is an input for some ESP models.

Particle Resistivity – There is a wealth of information on fly ash resistivity from most coal types. Measurements have been made both in-situ and in the laboratory. When the carbon content of the fly ash is moderate to high (>10%) these measurements become difficult. Mineral fly ash typically has resistivities ranging from 10¹⁰ ohm-cm to 10¹³ ohm-cm. Carbon is very conductive in comparison, with resistivities below 10⁷ ohm-cm. In-situ and laboratory resistivity measurements are difficult to perform on heterogeneous dusts like fly ash with high carbon content. These dusts have both high and low resistivity particles and either one could skew the resistivity measurement results. Likewise, their electrical behavior in an ESP is difficult to predict. The current ESP models do not accommodate this phenomenon.

FIELD TEST PROGRAM

A field test program has been proposed to fill the gaps in the data necessary to characterize the particulate carbon entering and leaving PCDs. Tests will be conducted at coal-fired plants representing the major coal/low NO_x burner/PCD combinations and at a plant with ACI.

The test program will consist of two components: Sample collection and laboratory analyses.

A wide variety of manual and automated sampling methods was considered for the project. Because of concerns that particles could agglomerate in bulk samples, inertial particle sizing was selected. Cascade impactors do not collect sufficient samples for analytical purposes, so a five-stage series cyclone will be used in an EPA Methods 5 or 17 sample train to collect adequate samples segregated by size. Cyclones will be run at the inlet and outlet of PCDs. The particles collected in each stage of the cyclone can then be tested for carbon content, density and mineral composition. In addition, bulk samples will be collected from PCD hoppers for comparison. On the plants with ESPs, a portable opacity monitor and an in-duct camera will be used to document transients and rapping spikes.

Once the cyclone samples and bulk samples have been collected they will be tested for a variety of properties. In some cases it may be necessary to separate the mineral fly ash from the carbon.

- Particle size;
- Particle shape;
- Particle density; and
- Carbon content.

Two additional properties, resistivity and cohesivity, are also of strong interest but the existing measurement techniques are not considered suitable for high carbon fly ashes.

The collected samples will be analyzed by the methods shown in Table 3.

Table 3. Analytical Tests to be Conducted on Collected Samples

Property	Method
Total Carbon	Infrared Spectrometry (LECO)
PSD (hopper samples)	Micro-trac, SEM
Skeletal Density	Helium pycnometry
Shape/Size	Reflective light microscopy, SEM

Sample Preparation – Carbon Concentration

The collected samples will be a mixture of carbon and mineral fly ash particles. While it is of interest to characterize the “as-collected” sample, the presence of mineral fly ash masks the properties of the actual carbon particles. For this reason, it is desirable to separate the carbon from the mineral fly ash. Several techniques have been used and can be broadly divided into subcategories of either wet or dry separation. Wet separation techniques include froth flotation and agglomeration and are not considered appropriate for this project because they could alter the properties of the carbon particles. Dry techniques are based on sieving, triboelectric separation and density gradient centrifugation. A stepwise combination of these techniques has been developed and used with success by the University of Kentucky CAER¹⁶. There maybe a limitation to the ability to separate the particles effectively in the smaller size fractions.

Laboratory Analytical Techniques

Particle size and shape – Aerodynamic particle size information will be provided by the Staged-Cyclone tests. Reflective light microscopy and/or SEM (if conventional microscopy is insufficient) analysis will provide additional information on the size and shape of the particulate collected in the cyclone samples. SEM analysis can be used to examine individual particles and distinguish between carbon and mineral particles. This would provide a crosscheck of the cyclone data since some carbon particles may contain mineral inclusions.

Particle Density – Skeletal density is the measure of particle density that excludes pore volume and is the density measurement that best represents the aerodynamic behavior of particles in flue gas streams and PCDs. Helium pycnometry is used to measure skeletal density and requires a bulk sample of at least 1ml in volume.

Carbon Content – While the ASTM LOI technique is a widely accepted measure of unburned carbon, there is a wealth of evidence that it may overstate the amount of carbon in fly ash samples. Dehydration of calcium compounds and evolution of volatile organic compounds (VOCs) can occur in the burnout phase of the LOI test and be incorrectly attributed as carbon. Infrared spectrometry is a more accurate measure of carbon content, especially on high calcium fly ashes and will be conducted on bulk and cyclone samples.

STATUS

The widespread application of low NO_x combustion technologies like LNBS and the potential retrofit of ACI to control mercury emissions changes the characteristics of the particulate matter that enters ESPs and fabric filters. Both the amount and properties of particulate carbon in the fly ash can impact the performance of the PCD. The existing computer models and empirical data used to size and design PCDs do not accurately represent the new PM characteristics. The objectives of this program were to gather what information is known about particulate carbon from low NO_x combustion technology and ACI, to identify the gaps in the knowledge base and to develop a test program to close those informational gaps. This will enable modelers and designers of PCDs to more accurately predict the impacts on PCD performance.

A great deal of research has been conducted to characterize unburned carbon from low NO_x combustion technologies. However, these efforts were for the purpose of optimizing combustion technologies and understanding the impacts on fly ash utilization. Little work has been done to characterize the properties of the in-flight carbon particles entering or leaving PCDs. There is some information on the amount of unburned carbon, but not its particle size distribution or true density while suspended in flue gas. A field sampling and analytical program have been proposed to address these and other unknowns.

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