DOE/PC/91334--T24

## **TECHNICAL REPORT**

DDE/ PC/91334 -- T?"

DE92 007868

September 1, 1991 through November 30, 1991

**Project Title:** 

Combustion of Illinois Coals and Chars with Natural Gas DE-FG 22-91PC 91334

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# ABSTRACT

There are applications where the combined combustion of coal and natural gas offers potential advantages over the use of either coal or natural gas alone. For example, low volatile coals or low volatile chars derived from treatment or gasification processes can be of limited use due to their poor flammability characteristics. However, the use of natural gas in conjunction with the solid fuel can provide the necessary "volatiles" to enhance the combustion. In addition, natural gas provides a clean fuel source of fuel which, in cofiring situations, can extend the usefulness of coals with high sulfur content. The addition of natural gas may reduce SO<sub>x</sub> emission through increased sulfur retention in the ash and reduce NO<sub>x</sub> emissions by varying local stoichiometry and temperature levels. In this research program, studies of combined coal and natural gas combustion will provide particle ignition, burnout rates and ash characterization, that will help clarify the effect of coal and natural gas and identify the controlling parameters and mechanisms.

The Drop Tube Furnace Facility (DTFF) allows unique detailed measurements of coal particle combustion under well-controlled conditions which can exist locally in an actual combustor. Operating characteristics include the use of particles in the 50 to 200 micron size range, particle heating rates greater than  $10^4$  K/sec, gas temperatures from 1200 to 1700 K, variable gas composition and particle residence times from 5 msec to 1 sec. The combustion characteristics of single coal particles are determined through a novel set of diagnostic techniques including in situ simultaneous measurements of particle morphology, temperature and velocity. Imaging of particle shape enables the determination of particle surface area and, therefore, a more accurate quantification of particle burning rate. In addition collection and analysis of solids samples will provide data on sulfur retention, carbon burnout and ash properties relevant to slagging and corrosion in boilers.

The emphasis of the effort in the first quarter of this project was on the understanding of the ignition enhancement and burning rate processes during cofiring. Initial investigations into the ash characterization have also begun. Ignition measurements and predictions for low and high volatile coals indicate the thermal dominance of the ignition processes. Temperature and burning rate data were obtained and analyzed as a function of methane addition, oxygen concentration, coal type and residence time. For similar oxygen concentrations, methane addition does not significantly alter the carbon burning rate. Solid sampling experiments have been initiated for various methane concentrations, oxygen concentrations, coal types and residence times.

This project is funded by the U. S. Department of Energy (PETC) and by the Illinois Department of Energy and Natural Resources as part of their costshared program.

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### EXECUTIVE SUMMARY

During the last three years a unique combination of experimental facilities and optical diagnostics have been developed to permit *in situ* simultaneous measurement of individual coal particle size and shape, temperature, and velocity. In our Drop Tube Furnace Facility (DTFF), dilute coal particle suspensions are injected into a high temperature environment which duplicates the time-temperature histories of coal fired boiler systems. Using advanced optical diagnostics and conventional sampling techniques, experiments are conducted to measure the details of coal particle size, particle temperature, ash properties and the effect of the oxidizing species and temperature environment. The novel aspect of this facility is the simultaneous measurement of particle temperature, morphology, and residence time by the non-intrusive techniques

Diagnostic equipment used for experimental measurements within this DTFF include (1) an optical system for ignition delay determination, (2) a two-color pyrometer for single particle temperature measurements, (3) an imaging system for determining particle size, shape and structure, (4) an optical system for particle velocity measurements and (5) a sampling probe for particle collection. The capabilities of the DTFF include the combustion of individual coal particles (of diameters between 50 and 200  $\mu$ m) in high temperature environments (1200 to 1700 K) with practical heating rates (~10<sup>4</sup> K/sec) and residence times (5 msec to 2 sec). We also have continuous gas sampling equipment for NO/NO<sub>x</sub>, CO, CO<sub>2</sub>, O<sub>2</sub>, HC and SO<sub>2</sub> gas phase analyses and will soon have a gas chromatograph in operation for more detailed gas phase species analyses.

Several pre-combustion techniques to reduce coal sulfur content are currently under investigation including flotation cleaning, electrostatic separation, microbial desulfurization, and chemical treating, among others. Although these techniques are capable of removing sulfur and ash, they often also alter other coal characteristics such as volatile content, heating value and ash properties. Such changes can significantly influence the burning characteristics of the coal in terms of ignitability, heat release rates and char burnout, for example. The potential for co-firing treated coals and chars to enhance the combustion characteristics and to leverage sulfur retention by the ash is a very promising avenue for the future usage of Illinois coals. Such studies should provide information which will allow combustion engineers to assess the prospects of co-firing treated coals in large scale industrial systems and to project the performance of these coals in terms of key boiler design features such as ash handling, heat release rates, turn down ratios, char burnout, etc.

The overall goals of our research program are to use the DTFF to (1) measure the ignition delays and burning rates of treated coals and chars as a function of char properties and natural gas concentration, and to (2) investigate the effects of co-firing of natural gas with Illinois high sulfur coals to determine the sulfur retention potential as a function of sulfur content, types of sulfur and natural gas concentration. Our work to date on the DTFF has shown that co-firing natural gas and coal has the potential for reducing sulfur emissions and enhancing the ignition characteristics of low volatile coals. First, long ignition delays and low burning rates associated with low volatile chars produced as part of a

desulfurization process are often unacceptable for conventional boilers. Co-firing of natural gas with these chars may provide adequate ignition and burning rate enhancement so that these chars can be viable fuels. Secondly, we have demonstrated that natural gas addition has the potential for reducing sulfur emissions by increasing sulfur retention in the ash. The effects of co-firing natural gas with Illinois coals and treated coals to determine sulfur retention potential as a function of sulfur content, types of sulfur and natural gas concentration has significant potential benefits.

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# **OBJECTIVES**

Several techniques are currently under investigation to remove or reduce the sulfur content of Illinois coals prior to combustion. These treatment processes can also alter other properties of the coal which can significantly change the combustion characteristics of the coal. Detailed measurements are needed to fully understand the effects of treatment and cofiring processes on the coal combustion ignition and burning rate characteristics. Sulfur retention in the ash has been shown to increase with methane addition supporting the suggestion of reduced sulfur emissions with natural gas addition not only due to replacement of high sulfur coal but also due to increased sulfur capture in the ash.

Thus, the objectives of this research program are: (1) to determine the ignition enhancement for Illinois coals, treated coals and chars as a function of coal and char properties and natural gas concentration, (2) to quantify the burning rate for the same parameters, and (3) to investigate the effects of the combustion of natural gas with Illinois sulfur coals and chars to determine sulfur retention potential as a function of sulfur content, type of sulfur, alkali-to-sulfur ratios and natural gas concentration. The Drop Tube Furnace Facility (DTFF) will be used to perform detailed measurements on burning coal particles. Coal particles are injected into the DTFF which has controlled and uniform temperatures (750 to 1600 °C, to be extended to 1750 °C), variable species concentrations (control of oxygen mole fraction and natural gas), variable residence times (5 msec to a few sec) and access for both physical and optical sampling. Measurements are made with a number of diagnostic techniques including particle velocity, particle morphology and devolatilization behavior by an imaging system which features a two-dimensional photodiode array mounted on the camera attachment of a photomicroscope, and particle temperature with a specially designed two-color pyrometer. A physical sampling probe is used to capture particles for average size, surface area and chemical analysis when needed for ash property determination.

# INTRODUCTION AND BACKGROUND

Research in our laboratory has shown that pre-treatment processes to reduce the sulfur in Illinois coals alters the combustion characteristics of the coals. Specifically, charring processes that result in a reduction of the sulfur in coal also reduce the volatile content of the coals. We have experimentally demonstrated that such treatment processes reduce the flame speed as compared to untreated coals. Our experiments on co-firing low volatile coals and natural gas show that small amounts of natural gas provide the necessary "volatiles" to ignite low volatile coals as though they are untreated high volatile coals. Additionally, our research has shown that natural gas addition has the potential for reducing sulfur emissions by increasing sulfur retention in the ash.

The combustion processes for coal particles are a complex interaction of both homogeneous and heterogeneous reactions. The overall combustion process, however, consists of two major steps: (1) thermal decomposition which involves the evolution of volatile matter as a result of particle heating and (2) char combustion. During thermal decomposition, the particle undergoes drastic changes in physical and chemical structure. Changes in particle size and pore structure that occur during devolatilization have a strong effect on subsequent char combustion. Thermal decomposition occurs much more rapidly than char combustion. The time required for thermal decomposition is on the order of 0.1 sec while the time required for char combustion is on the order of 1 sec. Thus, it is the combustion of char that determines the ultimate size of the combustor which is required to achieve complete combustion of the coal particle. Thermal decomposition and char combustion may occur separately or simultaneously, depending on the physical and chemical properties of the coal particle and combustor characteristics such as heating rate and oxygen concentration.

There have been many studies of the kinetics of coal particle thermal decomposition in inert and oxidizing environments [1-8]. Such studies show that devolatilization rates and yields are a strong function of temperature and heating rate. Experiments in oxidizing atmospheres have shown devolatilization rate to increase with increasing temperature and increasing oxygen concentration. More recent studies [9, 10] involving direct observation of coal particles during devolatilization in oxidizing atmospheres at high temperatures have increased the understanding of fundamental processes occurring during thermal decomposition. These results suggest that for high heating rates, combustion is initiated by ignition of the volatile matter given off during thermal decomposition.

A detailed review of the combustion rates and mechanisms of coal chars is given by Smith [11]. For particle sizes typical of pulverized coal, both particle size and density decrease during burning. Particle size affects the reactivities and the rate controlling regime that governs particle burning. The effects of parameters such as temperature and oxygen concentration which also determine burning rate can be hidden by the effect of particle size if it is not a known quantity. Thus, in order to accurately characterize the burning process, particle size and temperature must be measured simultaneously. Experiments on char oxidation generally determine burning rates by direct sampling of gaseous products or solid products [6, 8, 12 - 14] where the measured particle burning rate is averaged over the particle size distribution. In order to gain a detailed understanding of the particle burning process, burning rates need to be determined on a single particle basis to account for the interdependence of size and temperature. The only practical means of making in-situ measurements of particle size and temperature is with optical techniques.

The need to measure particle size and temperature simultaneously was recognized by researchers at Sandia National Laboratories [15 - 18]. They have developed an apparatus capable of measuring individual particle length, temperature, and velocity simultaneously. Particle size information is obtained from information received through two slits rather than an entire particle image. They have also performed combustion experiments with pulverized coal particles with the measurement technique described above to determine particle burning rates.

At the University of Illinois, we have developed an experimental facility [19, 20] to study single particle combustion to provide *in situ* simultaneous particle temperature, morphology and velocity of the coal particles. Ignition experiments in the area of co-firing indicate that co-firing natural gas with low volatile coals can reduce the ignition delay time to levels which are more typical of high volatile coals. Imaging experiments showed the complex structure of the particles during co-firing combustion. Entire particle morphology information, rather than one length, can be obtained with our imaging system. The addition of natural gas may reduce SO<sub>x</sub> emissions through increased sulfur retention in the ash and reduce NO<sub>x</sub> emissions by varying local stoichiometry and temperature levels [21]. Our results for sulfur retention in the ash as a function of initial methane concentration suggest an increased sulfur retention in the ash with methane addition.

# EXPERIMENTAL PROCEDURES

The Drop Tube Furnace Facility (DTFF) design shown schematically in Figs. 1 and 2 has been discussed in detail in the original proposal. The DTFF allows detailed measurements of coal particle combustion under well-controlled conditions which can exist locally in an actual pulverized coal combustor. In the DTFF dilute concentrations of coal particles of the pulverized coal size range are subjected to a high heating rate and various residence times at high temperatures. Particles can be reacted in gaseous environments of widely varying composition and all phases of particle combustion, from devolatilization and ignition to carbon burnout of the char, can be observed. The general characteristics of the DTFF and measured quantities are summarized in Table 1.

**Operating Characteristics** Particle Size 50 - 200 microns Heating Rate 10<sup>4</sup> K/sec typical 1200 - 1700 K Gas Temperatures Residence Times 5 msec to 2 sec 0 to 30%Oxygen concentration **Measured Quantities** Ignition Delay Particle Size Particle Temperature Particle Velocity Particle Morphology Ash Content

Table 1. Summary of Drop Tube Furnace Facility (DTFF) capabilities.

The heated section of the reactor consists of a one meter long cylinder which houses high temperature electrical heating elements and ceramic fiber insulation. A fluidized bed particle feeder is used to provide a dilute concentration of particles. The feeder design allows the introduction of a stream of single particles into the reactor with each pulse. The particles and gaseous reactants enter a water-cooled jacket which prevents reactions prior to the heated test section. Two optical particle detectors are mounted at the end of the injector to detect when a particle exits the injector and to determine the particle injection velocity. The particle then enters the test section where particle heating rates are estimated to be on the order of  $10^4$  K/s. Particle residence times are varied over a wide range by changing gas velocity and the length of the heated process tube.

For ignition delay measurements, light exiting the reactor process tube from igniting particles is imaged onto a phototransistor. The elapsed time from the point at which a



Figure 1. Drop Tube Furnace Facility (DTFF)



Figure 2. Diagnostics for the DTFF

particle enters the reactor to the point at which it ignites is recorded as the ignition delay time. For solids sampling experiments, a water-cooled sampling probe is inserted through the lower section of the reactor and particles are collected at the plane of the optical test section.

In situ imaging of the particles is desired because of the complex shape and structure of coal particles and the changes that occur in that structure due to combustion. Images of the nonspherical burning particles are obtained using a 640 x 480 photodiode array digital camera. A strobe light source is used to back-light and effectively "freeze" the particle. A microscope lens is used to magnify the particle image by as much as 35 times, allowing details of the particle shape, structure and size to be observed. The test section also provides optical access for the two-color pyrometer. The coal particles are imaged with a 1:1 magnification mirror system onto two Indium Antimonide (InSb) Infrared detectors. The ratio of these signals is used to calculate temperature. Calibration and testing of the two-color pyrometer indicates that individual particle temperatures are measured to within 40 K.

As discussed earlier the particle velocity at injection is determined by using two detectors separated by a know distance and recording the transit time for the particle from the first to the second detector. The velocity of the particle at the optical probe is determined by measuring the distance the particle travels between the time the particle is at the center of the two-color pyrometer probe volume and the time at which the strobe is triggered for the imaging system.

Solids sampling is by physically sampling, quenching and extracting samples. Particles are collected as they enter the optical test section with a probe that consists of a water-cooled jacket surrounding a stream of helium. The helium mixes with the sample at the probe inlet to quench the reaction. A one micron filter is positioned in the probe to collect particles.

Thus, the experimental methodology is to use the Drop Tube Furnace Facility to perform detailed measurements under controlled conditions. Specific variables controlled are the coal type, environment temperature, oxygen concentration, methane concentration, and residence time. The *in situ* measurements include particle temperature, particle residence time and particle morphology; physical ash sampling measurements include size, surface area and chemical analysis. The experimental research program goals are (1) to measure the ignition enhancement for Illinois coals, treated coals and chars as a function of coal and char properties and natural gas concentration, (2) to quantify the burning rate for the same parameters, and (3) to investigate the effects of the combustion of natural gas with Illinois sulfur coals and chars to determine sulfur retention potential as a function of sulfur content, type of sulfur, alkali-to-sulfur ratios and natural gas concentration.

# **RESULTS AND DISCUSSION**

As indicated in the section above, the three elements of this project are ignition enhancements studies, burning rate determination, and the ash characterization experiments. Various coals have been considered. The following table presents the general characteristics of the coals obtained to date. 
 Table 2: Coal Types Investigated

Designation	Volatiles/Sulfur	Ash	
PSOC-1519	36.54 % Volatiles 1.94 % Sulfur	10.52 % Ash 0.15 % Alkali Metals Organic/Sulfatic/Pyritic=1.82/0.1/0.02 %	
PSOC-1508	15.70 % Volatiles 0.64 % Sulfur	7.44% Ash 0.37 % Alkali Metals Organic/Sulfatic/Pyritic=0.50/0.00/0.14 %	
PSOC-1524	32.78 % Volatiles 1.85 % Sulfur	11.04 % Ash 0.19 % Alkali Metals Organic/Sulfatic/Pyritic=0.44/0.02/1.39 %	
PSOC-1551	32.11 % Volatiles 1.80 % Sulfur	10.37 % Ash 0.25 % Alkali Metals Organic/Sulfatic/Pyritic=0.85/0.01/0.94 %	
IBC-104	37.9 % Volatiles 3.90 % Sulfur	14.5 % Ash Organic/Sulfatic/Pyritic=2.54/0.10/1.26 %	
IBC-106	39.9 % Volatiles 3.67 % Sulfur	11.3 % Ash Organic/Sulfatic/Pyritic=1.97/0.04/1.62 %	

### Ignition Enhancement Investigation

The analysis of the ignition enhancement data is continuing through the first portion of this project. Ignition delay data for tabled and floatation treated IBC-104 and IBC-106 coals without methane addition, and the high volatile PSOC-1519 and low volatile PSOC-1508 coals as a function of methane addition, have been obtained. The results are repeated in Figures 3 and 4. Most of the effort during this quarter has been on the interpretation and analysis of this data.

The model for ignition includes the effects of methane combustion, particle geometry, particle swelling, variable properties, and devolatilization. With inputs of initial conditions of the gas and particle at the reactor inlet and the axial wall temperature profile of the reactor, the axial mean temperature and velocity profiles of the gaseous species using the measured wall temperature profile are calculated. The gas temperature is determined by considering convective heat transfer with the wall and the temperature rise due to gaseous combustion. The temperature and velocity history of the coal particle is determined from these known environmental conditions and a particle energy balance. Numerical integration of the energy equation yields particle temperature as a function of position. Particle temperature as a function of time can then be determined from the particle velocity history. The deviation of raw coal particles from spherical as well as particle swelling was determined by measuring the average radius of the two-dimensional images.

Figure 3 shows time to ignition for tabled and flotation treated coals. There is only a slight difference in the ignition delay for these coals since, as previously noted, the flotation process physically separates ash particles from the sample. Ignition studies on these coals



Figure 3. Measured ignition delay for IBC-104 and IBC-106 tabled and treated coals.



Figure 4. Ignition delay results as a function of methane concentration for PUOC-1508 (LV) and PSOC-1519 (HV) coals.

including the contributions of methane remain to be conducted, yet the analysis of both the high and low volatile coals (PSOC-1519 and PSOC-1509) verses methane addition has been considered. The aim of this ignition effort is to understand and model the ignition process for coals resembling treated and parent Illinois coals.

Measured and predicted ignition delays for the high and low volatile coals are shown in Figure 4. As the methane concentration is increased, the time to ignition of both coals is significantly decreased. This reduction is attributed to the increased particle heating created by the methane reaction. As expected, the low volatile coal requires a longer ignition time than the high volatile coal. However, the addition of 1% methane by volume to the low volatile coal results in ignition times similar to that of the high volatile coal with no methane addition. The dashed curves represent the predicted ignition times using the model and an assumed ignition temperature of 1080 K. The difference in coal volatility on ignition time was accounted for using a devolatilization model, assuming the released volatiles burn as methane and that five percent of the resultant heat released is absorbed by the particle.

The measured and predicted time to ignition for variation in bulk gas species has also been investigated. Differences in ignition times that result from different species is attributed to the different gas properties of the mixtures, and the model predictions agree with the experimental results when gas properties are accounted for in the calculations.

It is important to note that practical systems are likely to use natural gas rather than methane. Although natural gas is largely methane, it also contains ethane which is known to enhance ignition. Experiments using natural gas show no appreciable difference in particle ignition between the two fuels. Our predictions for ignition delay for both fuels confirms this behavior.

### Burning Rate Determination Results

The general effects of methane concentration on particle burning rate are addressed for two coals: the high volatile coal (PSOC-1519) and a low volatile coal (PSOC-1508). Particles in the size range of 125 to 150  $\mu$ m were used in the experiments. The emphasis during this quarter has been placed on data analysis and interpretation.

Instantaneous particle burning rates were determined using measured particle temperatures and information from particle images. A particle energy balance yields an absolute particle burning rate in micrograms of carbon per second. Thus, the value of the absolute particle burning rate was directly affected by the method used to determine particle external surface area. This is illustrated by Figure 5 which shows calculated particle burning rates for measurements at several oxygen concentrations using two different methods to determine particle surface area: the revolution method and the equivalent sphere method. The revolution method has been developed by our research group, and it provides more accurate measures of the particle surface area, volume and surface area-to-volume ratio than the equivalent sphere concepts. For typical predictions of real surface morphology, the revolution method can be expected to predict the values to within about 20% for surface area and the surface area-to-volume ratio, and less than 10% for volume. (Equivalent sphere methods always have larger errors, typically double those of the revolution method.) For the equivalent sphere method, an average particle diameter was first determined using the projected area of the two-dimensional particle image  $(2(A_y/\pi)^{0.5})$ . The surface area of a sphere with this diameter was then used as an estimate of the particle surface area. Each point in Figure 5 represents the burning rate of one particle. As the figure shows, the equivalent sphere method consistently underpredicted particle burning



Figure 5. Calculated particle burning rates for different methods of particle surface area determination.

rates because it did not take into account the irregular shape of the particle. Irregularities in particle shape result in higher surface areas. Thus, the revolution method was used in all subsequent particle burning rate calculations.

Figure 6 shows the effect of methane concentration on particle temperature for the high volatile coal ~100 msec after ignition. For this set of experiments, the oxygen concentration prior to methane combustion was 20% for all cases. At each methane concentration, a range of particle temperatures was observed. For the lower methane concentrations (0 and 1%), particles in the low temperature region were near burnout, similar to the 20% oxygen case. At the higher methane concentrations (2 to 4%), particle imaging showed the presence of low temperature soot-like particles. For methane concentrations of 0, 1, 2, 3 and 4%, the oxygen concentrations after complete combustion of the methane were 20, 18, 16, 14, and 12%, respectively. Figure 6 also shows that as the methane concentration was increased, the maximum particle temperatures with 2 and 4% methane were lower than those without methane by ~100 and 200 K, respectively. The solid line represents the predicted diffusion-limited temperatures using the bulk gas oxygen concentrations after methane combustion. The diffusion-limited model adequately predicts the reduction in particle temperature with methane concentration.

Figure 7 shows the corresponding calculated particle burning rates for the measurements in Figure 6. Particle burning rates for 0 and 1% methane ranged from ~0 to 10  $\mu$ g/s while burning rates for 2 to 4% methane were slightly lower and in the range of ~0 to 8  $\mu$ g/s. This reduction in burning rate was due to the reduction in oxygen with increasing methane addition. Also, the figure shows that the 1% methane case had more particles with higher burning rates than did the 0% methane case. At this residence time the 20% oxygen case (0% methane) had many particles near burnout. The addition of 1% methane reduced the particle burning rate during the combustion process such that at the same residence time more particles were still burning.

Particle temperature and image measurements were also obtained for the high and low volatile coals at different residence times with and without methane addition. Figure 8 shows the effect of methane addition on calculated particle burning rates for high volatile coal. Burning rates for particles in air at 0 msec are not shown since the particle images were obscured by volatile and/or soot clouds present at ignition. However, the figure shows that particles at 0 msec with 2% methane had ignited and were burning. At both 50 and 100 msec, the range of particle burning rates for the air and 2% methane cases were similar but at 100 msec the 2% methane case had more particles with higher burning rates than did the air case. This was a result of the reduced oxygen concentration with methane addition lowering burning rates during particle combustion which increased the time required to reach burnout. At 200 msec, particle burning rates with and without methane addition were essentially zero, indicating burnout.

Figure 9 shows the effect of methane addition on the calculated particle burning rates for low volatile coal. At 0 msec, the rate of carbon loss for low volatile coal particles in air was essentially zero while most of the particles in 2% methane were burning. Similar results are shown for 50 msec. At 100 and 200 msec, particles for both cases had similar ranges of burning rates, but the 2% methane case had a few more particles in the lower burning rate region. Note that at 200 msec, the low volatile coal particles for both cases were still burning while the high volatile coal at 200 msec had a burning rate of essentially zero. Part of this was a result of the low volatile coal igniting later than the high volatile



Figure 6. Effect of methane concentration on particle temperature.  $[20\% O_2 \text{ prior to CH}_4 \text{ combustion, high volatile coal } \sim 100 \text{ msec after ignition}]$ 



Figure 7. Effect of methane concentration on rate of carbon loss. [20% O<sub>2</sub> prior to CH<sub>4</sub> combustion, high volatile coal ~100 msec after ignition]



Figure 8. Effect of methane on rate of carbon loss at several residence times.  $[20\% O_2 \text{ prior to CH}_4 \text{ combustion, high volatile coal}]$ 



Figure 9. Effect of methane on rate of carbon loss at several residence times. [20% O<sub>2</sub> prior to CH<sub>4</sub> combustion, low volatile coal]

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coal, but the low volatile coal also had a higher amount of fixed carbon present and thus should take longer to reach burnout.

### Ash Characterization Studies

Our previous work has indicated that the sulphur retention in coal ash may increase with natural gas coal firing. At present, the mechanisms behind this process are not fully understood. In order to quantify this phenomenon, a series of experiments were proposed and have been initiated. These experiments consist of collecting solid and gas phase reaction products for a variety of experimental parameters. These reaction products will then be analyzed for their sulfur forms and quantity. Many of the experiments have been performed yet, due to conflicting results received from the chemical analysis laboratory, many of these experiments are currently being rerun.

The variables in these experiments were the coal type, oxygen concentration and methane concentration. Several coals were used in the experiments (see Table 1). These coals were chosen to have variable pyritic to organic sulphur content, variable alkali content and different volatile content. A summary of the some of the conditions for samples obtained to date are in Table 3. Two sizes of furnace tubes were used. A small 0.008 m dia. tube and a larger 0.0125 m dia. tube. The small bore tube was used so that previous experimental data could be repeated as a check for consistency. The large bore tube was chosen for the purpose of decreasing the experimental run time for each experiment. A total of 42 samples were initially taken and sent for analysis.

The solid samples were to be analyzed for carbon, silicon, total sulphur and sulphur forms (organic, pyritic and sulfatic) as per ASTM-2492, yet there was insufficient sample to perform all the determinations. Many of these tests are being rerun. Also, note that in the quarterly report for our project entitled "Cooperative Research on the Combustion Characteristics of Cofired Desulfurized Illinois Coal and Char with Natural Gas" the parameter range of the Drop Tube Flow Facility is being extended to provide more comprehensive range for subsequent data.

# CONCLUSIONS AND RECOMMENDATIONS

The first quarter of this project has focused on the understanding of the ignition enhancement and burning rate processes during cofiring and on the initial measurements of ash characteristics. Ignition measurements and predictions for low and high volatile coals indicate the thermal dominance of the ignition processes. A comprehensive model has been developed to predict all observable trends. Temperature and burning rate data were obtained and analyzed as a function of methane addition, oxygen concentration, coal type and residence time. For similar oxygen concentrations, methane addition does not significantly alter the carbon burning rate. A diffusion limited particle burning rate model, which uses local oxygen concentrations, adequately predicts particle temperatures and, hence, particle burning rates.

Solid sampling experiments have been performed for a variety of experimental parameters. The variables in these experiments were the coal type, oxygen concentration and methane concentration. The samples were analyzed for their sulfur forms and quantity, yet inconsistencies in the chemical analysis provided by the external laboratory make further measurements necessary.

Coal	Tube (inches)	Carrier Gas (mole %)	Oxygen (mole %)	Methane (mole %)
Small Di 1519 1519 1519 1519 1519	ameter tube (5/10 8 8 8 8 8 8	5") 100 (N <sub>2</sub> ) 80 (N <sub>2</sub> ) 80 (N <sub>2</sub> ) 80 (N <sub>2</sub> )	0 20 20 20 20	0 0 2 4
1519 1519 1519 1519 1519 1519	12 12 12 12 12	100 (N <sub>2</sub> ) 100 (N <sub>2</sub> ) 80 (N <sub>2</sub> ) 80 (N <sub>2</sub> ) 80 (N <sub>2</sub> )	0 0 20 20 20 20	0 4 0 2 4
1519 1519 1519 1519 1519	24 24 24 24	100 (N <sub>2</sub> ) 80 (N <sub>2</sub> ) 80 (N <sub>2</sub> ) 80 (N <sub>2</sub> )	0 20 20 20 20	0 0 2 4
Large Di 1519 1519 1519 1519 1519 1519 1519 151	ameter tube (1/2 24 24 24 24 24 24 24 24 24 24	$\begin{array}{c} 100 & (N_2) \\ 96 & (N_2) \\ 80 & (N_2) \end{array}$	0 0 20 20 20 20 20 20	0 4 0 1 2 3 4
1519 1519 1519	24 24 24	90 (N <sub>2</sub> ) 90 (N <sub>2</sub> ) 90 (N <sub>2</sub> )	10 10 10	0 1 2
1524 1524 1524 1524 1524	24 24 24 24 24 24	100 (N <sub>2</sub> ) 96 (N <sub>2</sub> ) 80 (N <sub>2</sub> ) 80 (N <sub>2</sub> ) 80 (N <sub>2</sub> )	0 0 20 20 20	0 4 0 2 4
104 104 104 104 104	24 24 24 24 24 24	100 (N <sub>2</sub> ) 96 (N <sub>2</sub> ) 80 (N <sub>2</sub> ) 80 (N <sub>2</sub> ) 80 (N <sub>2</sub> )	0 0 20 20 20 20	0 4 0 2 4
1551 1551 1551	24 24 24	80 (N <sub>2</sub> ) 80 (N <sub>2</sub> ) 80 (N <sub>2</sub> )	20 20 20	0 2 4
1508 1508 1508 1508 1508	24 24 24 24 24 24	80 (N <sub>2</sub> ) 80 (N <sub>2</sub> ) 80 (N <sub>2</sub> ) 80 (N <sub>2</sub> ) 80 (N <sub>2</sub> )	0 0 20 20 20 20	0 4 0 2 4

Table 3. Solid Sampling Experiments

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### REFERENCES

- 1. Kobayashi, H., Howard, J.B., and Sarofim, A.F., "Coal Devolatilization at High Temperatures," <u>Sixteenth Symposium (International) on Combustion</u>, The Combustion Institute, 1976, p. 411.
- 2. Jamaluddin, A.S., Truelove, J.S., and Wall, T.F., "Devolatilization of Bituminous Coals at Medium to High Heating Rates," <u>Combustion and Flame</u>, Vol. 63, 1986, p. 329-337.
- 3. Solomon, P.R., Hamblen, D.G., Carangelo, R.M. and Krause, J.L., "Coal Thermal Decomposition in an Entrained Flow Reactor: Experiments and Theory," <u>Nineteenth Symposium (International) on Combustion</u>, The Combustion Institute, 1982, pp. 1139-1149.
- 4. Maloney, D.J. and Jenkins, R.G., "Coupled Heat and Mass Transport and Chemical Kinetic Rate Limitations During Coal Rapid Pyrolysis," <u>Twentieth Symposium</u> (International) on Combustion, The Combustion Institute, 1984, pp. 1435-1443.
- 5. Scaroni, A.W., Walker, P.L., and Essenhigh, R.H., "Kinetics of Lignite Pyrolysis in an Entrained Flow, Isothermal Furnace," <u>Fuel</u>, Vol. 60, 1981, pp. 71-76.
- 6. Jost, M., Leslie, I., and Kruger, C., "Flow-Tube Reactor Studies of Devolatilization of Pulverized Coal in an Oxidizing Environment," <u>Twentieth Symposium (International) on Combustion</u>, The Combustion Institute, 1984, pp. 1531-1537.
- 7. Timothy, L.D., Sarofim, A.F., and Beer, J.M., "Characteristics of Single Particle Coal Combustion," <u>Nineteenth Symposium (International) on Combustion</u>, The Combustion Institute, 1982, pp. 1123-1130.
- 8. Saito, M., Sadakata, M., and Sakai, T., "Measurements of Surface Combustion Rate of Single Coal Particles in Laminar Flow Furnace," <u>Combustion Science and Technology</u>, Vol. 51, 1987, pp. 109-128.
- 9. McLean, W.J., Hardesty, D.R., and Pohl, J.H., "Direct Observations of Devolatilizing Pulverized Coal Particles in a Combustion Environment," <u>Eighteenth Symposium</u> (International) on Combustion, The Combustion Institute, 1981, p.1239.
- 10. Samuelsen, G.S., Trolinger, J.D., Heap, M.P., and Seeker, W.R., "Observation of the Behavior of Coal Particles During Thermal Decomposition," <u>Combustion and Flame</u>, Vol. 40, 1981, p.7.
- 11. Smith, I.W., "The Combustion Rates of Coal Chars: A Review," <u>Nineteenth</u> <u>Symposium (International) on Combustion</u>, The Combustion Institute, 1982, p.1045.
- 12. Wells, W.F., Kramer, S.K., Smoot, L.D., "Reactivity and Combustion of Coal Chars," <u>Twentieth Symposium (International) On Combustion</u>, The Combustion Institute, 1984, pp. 1539-1546.

- 13. Young, B.C. and Smith, I.W., "The Kinetics of Combustion of Petroleum Coke Particles at 1000 and 1800 K: The Reaction Order," <u>Eighteenth Symposium (International) on</u> <u>Combustion</u>, The Combustion Institute, 1981, pp. 1249-1255.
- 14. Ayling, A.B. and Smith, I.W., "Measured Temperatures of Burning Pulverized-Fuel Particles, and the Nature of the Primary Reaction Product," <u>Combustion and Flame</u>, Vol. 18, 1972, pp. 173-184.
- 15. Mitchell, R.E. and Niksa, S., "Interpreting Spatially-Resolved Mass Conversions and Two-Color, Single-Particle Temperatures for Burning Polydisperse Pulverized Fuel Streams," Sandia National Laboratories, SAND83-8707 (1983).
- 16. Tichenor, D.A., Mitchell, R.E., Henchen, K.R., and Niksa, S., "Simultaneous In Situ Measurement of the Size, Temperature and Velocity of Particles in a Combustion Environment," Sandia National Laboratories, SAND84-8628 (1984).
- 17. Mitchell, R.E. and McLean, W.J., "On the Temperature and Reaction Rate of Burning Pulverized Fuels," <u>Nineteenth Symposium (International) on Combustion, The</u> Combustion Institute, 1982, p.1113.
- 18. Mitchell, R.E., Tichenor, D.A., and Hencken, K.R., "Overall Burning Rates of Coal Chars from Simultaneous Measurements of Particle Size, Temperature and Velocity in Burning Polydisperse Pulverized Coal Streams," <u>Heat Transfer in Fire and Combustion</u> Systems, ASME, 1985, p.37.
- Schroeder, A. R., D. A. Thompson, H. Krier, J. E. Peters, and R. O. Buckius, "Preliminary Study of the Effects of Natural Gas Co-Firing on Coal Particle Combustion," <u>Seventh Annual International Pittsburgh Coal Conference</u>, Pittsburgh, PA, 1990.
- Schroeder, A. R., D. C. Johnson, R. O. Buckius, H. Krier and J. E. Peters, "Effects of Natural Gas Co-Firing on Coal Particle Ignition," Central States Section, The Combustion Institute, 1991.
- 21. Nettleton, M. A., "Temperature Measurements on Burning Coal Particles in a Radiating Enclosure," <u>Comb. and Flame</u>, Vol. 9, 1965, pp. 311-316.



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