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Project Title: Cooperative Research on the Combustion Characteristics of Cofired Desulfurized Illinois Coal and Char with Natural Gas

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ABSTRACT

The objective of this project is to determine the contributions of coal type, sulfur type, temperature, and residence time on the coal combustion behavior (especially the effect on ash constituents) during cofiring with natural gas. The Drop Tube Furnace Facility (DTFF) at the University of Illinois at Urbana-Champaign (UIUC) together with appropriate modifications is the chief apparatus used in this investigation, with additional diagnostic tools including the scanning electron microscope and chemical analyses.

A review of literature related to the project was made, and a strategy outlined for carrying out the research. Different kinds of coal are injected into the drop tube furnace under various controlled conditions, and samples taken at various residence times are analyzed to show the transformation of the species. Effects of the controlling parameters are studied.

The DTFF is extended to larger sample collecting capability and higher temperatures, resulting in the establishment of the Ash Characterization Facility and the High Temperature Drop Tube Furnace. The Ash Characterization Facility enables continuous coal injection and sampling under controlled conditions. Several hundred milligrams of char or ash can be collected in one-half hour. The High Temperature Drop Tube Furnace uses a plasma torch to preheat the gas to over 2000 K and inject it into a ceramic tube which enters a furnace designed for 1700°C (1973 K) operation, so that temperatures and heating rates encountered by pulverized coal particles in the flames of large boilers or in the advanced slagging cyclone combustors can be simulated. An aerodynamic coal feeder works well in supplying coal continuously to the drop tube. A watercooled, Helium-quench sampling probe collects the solid samples. A scanning electron microscope is used to study the morphology of ash and char particles. A sulfur determinator, a gas chromatograph provide analytical means in the laboratory, and the Illinois State Geological Survey performs other necessary analyses of the samples.

Tests on cofiring coal with 1 to 4% methane show that sulfur retention in ash was related to temperature and residence time. The addition of methane caused changes in gas temperature profile in the tube and also changes in chemical composition of the gases. The overall effect on sulfur retention is seen to be a result of several complex interacting factors. Further detailed studies are necessary to clarify the contribution of each factor and to provide clues to the mechanism of the process.

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EXECUTIVE SUMMARY

The reduction of sulfur emissions from the combustion of Illinois coals is an important and urgent task. Research on the fundamental level can provide a better understanding of the process involved in clean coal technologies, and offer basic data and new approaches for improving the present technology or developing new technologies. This project considers the effects of cofiring natural gas with Illinois high-sulfur coals to determine the sulfur retention potential in ash as a function of sulfur content, sulfur type and natural gas concentration. Previous experimental results from UIUC have shown that cofiring natural gas and coal has the potential for reducing sulfur emissions by increasing sulfur retention in the ash. Additionally, other researchers have suggested that cofiring of natural gas and coal reduces ash deposition problems.

To study sulfur retention in ash and deposition problems, the mechanism and rate of sulfur and mineral transformation must be understood. A survey of the literature shows that present work on sulfur transformation are mainly limited to pyrite, and the details of transformation in coal under boiler conditions have not been well studied. Ash formation and behavior under boiler conditions have just begun to be studied in a fundamental way, and the range in conditions and the degree of quantification are still quite limited. The effects of high temperature on ash behavior such as those encountered in large boilers (~2000 K) or slagging cyclone furnaces (>2000 K) have not been studied due to lack of suitable facilities. Also, the effects of natural gas cofiring have not been completely investigated. Therefore, through the present project, we seek to obtain results contributing to the understanding of these subjects with the goal to find more effective ways to limit sulfur emission and to reduce boiler deposition.

In order to better study the above problems, the Drop Tube Furnace Facility (DTFF) at UIUC was modified in this project year to emphasize the study of ash and effects of high temperatures. This results in the establishment of the Ash Characterization Facility and the High Temperature Drop Tube Furnace. The Ash Characterization Facility is a larger diameter drop tube furnace with continuous coal feed and capable of collecting a few hundred milligrams of ash or char samples in a period of one-half hour. The heating rate is from $5 \times 10^3 \text{ K/s}$ to $5 \times 10^4 \text{ K/s}$ and the gas temperatures are from 1000 to 1700 K. Residence times from milliseconds to order of seconds are possible. A water-cooled, Helium-quench sampling probe collects the solid ash or char samples. A scanning electron microscope at the UIUC Electron Microscopy Center is used to study the morphology of particles at different stages of the combustion process. A sulfur determinator, a gas chromatograph and cooperative arrangement with the Illinois State Geological Survey provide chemical analysis for the samples gathered from these facilities.

In the High Temperature Drop Tube Furnace, the gas can be preheated to over 2000 K by the use of the plasma torch gas heater, developed during this period. The plasma torch works on nitrogen, consumes about 3kW electric power, and gives trouble-free operation for hundreds of hours. The gas is injected into a SiC tube heated in a furnace designed to work at a maximum temperature of 1700°C (1973 K). The gas temperature in the drop tube reached 1900 K, which is higher than the values reported in the drop tube furnace literature. Residence times up to 300 ms can be obtained in this tube.

Experiments on the coal combustion in the Ash Characterization Facility to date include combustion of coal at different temperatures and residence times with and without methane cofiring. Samples are collected and analyzed for sulfur, carbon and silicon content to determine the sulfur retention as a function of coal type, temperature, residence time and natural gas cofiring. The results show that methane cofiring changed the

temperature and composition distribution in the drop tube, and has a significant effect on the burning rate and sulfur content in the remaining solids. The overall effect of methane cofiring is seen to be a complex function of the physical (mainly temperature) and chemical factors involved. More detailed studies will be needed to clarify the mechanisms of sulfur transformation and retention in the process of combustion with methane cofiring. The details of the combustion studies are included in the FINAL TECHNICAL REPORT for the project entitled "Combustion of Illinois Coals and Chars with Natural Gas".

OBJECTIVES

This research seeks to characterize the effects of treatment processes and natural gas cofiring on the ash characteristics for Illinois coal, with the intent of finding better methods of reducing the sulfur emission and ash deposition problems. In order to achieve this objective, it is necessary to make detailed studies of sulfur and mineral transformation during combustion and behavior of ash under various conditions for parent and treated coals.

The specific objectives for the current project are to obtain ash transformation data under well controlled conditions in the Drop Tube Furnace Facility (DTFF) for Illinois coals, with and without natural gas cofiring. An understanding of the mechanisms and potential for reducing the sulfur emission and ash deposition problems can be obtained.

The tasks addressed to meet the objectives are the following:

1. Formulation of the complete strategy, methodology and outline for this research.
2. Modification of the experimental apparatus to suit the purpose of this particular project.
3. Performing preliminary experiments and refining the apparatus.
4. Obtaining reliable data in the improved apparatus, using available diagnostic tools.
5. Analysis of the data to arrive at an assessment of the effects of the various factors and an understanding of the processes involved.
6. Summary of the work performed and recommendations for future work.

INTRODUCTION AND BACKGROUND

Introduction

There are many significant research issues impacting the use of our coal resources including the mechanisms of pollutant emission and methods of pollution control, methods to achieve stable and efficient combustion, mineral matter transformation which contributes to slagging and fouling in boilers, database on fuels and combustion characteristics, diagnostic methods, combustion modeling, etc. At present, the environmental requirements as exemplified by the Clean Air Act Amendment of 1990 pose an important and urgent problem for the coal producers and users, especially to the state of Illinois. Due to the high sulfur content of the Illinois coals, many users may switch to low-sulfur coals in order to comply with the standards. There will be significant losses of markets for this fuel. Therefore it is of utmost importance and urgency to find better solutions to the sulfur pollution problems in the combustion of Illinois coal.

Illinois is actively involved in many of the clean coal research and demonstration projects. For instance, the Gas Reburning and Sorbent Injection Project has started at Hennepin and Springfield. The Low NOX/SOX Burner for Utility Cyclone Boilers

project is being carried out at Marion. The demonstration project of Mild Gasification of Illinois Coal has started at the Illinois Coal Development Park. Research on a fundamental level is also being supported. This fundamental research provides a better understanding of the processes involved in the clean coal technologies and offers basic data and new ideas to the pilot or industrial scale studies with the purpose of improving the present technology or developing new technologies.

Background

In recent years, the problem of fireside deposition in boilers has attracted a great deal of attention among research groups. This is because slagging and fouling on the heat transfer surfaces and in flow passages in boilers are the major cause of unplanned shutdown and loss of plant availability. Large economic losses are involved in these problems. With coal switching and coal blending occurring more frequently in practice, the deposition behavior of ash becomes more unpredictable, as the simple concept of "ash fusion temperature" will not give reliable predictions. Therefore scientists are trying to study ash formation and behavior in a fundamental way and hope to arrive at reliable predictive methods for deposition problems in boilers with different coals. A logical approach to this problem is to first study the processes of mineral transformation under conditions representative of those in the actual boiler furnaces, then quantify the mechanisms and form physical models, and subsequently combine these models into one comprehensive model which can be used to predict ash deposition behavior. Some fundamental processes of ash formation have been studied through analytical methods and some preliminary submodels have been presented. First attempts at establishing a framework for a comprehensive models are just being initiated.

Research efforts have identified two kinds of ash particles formed, a micron size formed by residual ash in char burnout, and a submicron fume ($\sim 0.05 \mu\text{m}$) formed by condensation of vaporized mineral components [1]. The evolution of fly ash during pulverized coal combustion including the fragmentation and/or agglomeration of coal and ash particles were studied [2]. Recently, an attempt was made [3] to predict relative fouling tendencies of a coal and a coal blend, based on computer controlled scanning electron microscopy (CCSEM) of the coal minerals. Results were compared with those of experimental deposition studies obtained in the pilot scale Combustion Research Facility, and they are encouraging.

The modeling of mineral matter transformation in coal combustion has also received some attention [4]. Submodels are obtained from bench scale tests done in many laboratories which include descriptions of 1) the coal minerals, 2) transformation of key mineral species, 3) particle transport and dispersion, 4) particle reaction and fragmentation, 5) deposition mechanisms, 6) particle sticking and 7) deposit characterization. The submodels are integrated into a comprehensive computer code for the simulation of mineral matter behavior during pulverized coal combustion. Results show promise. Additional studies of the mechanisms of coal ash formation under closely controlled combustion conditions have been performed [5]. Minerals in the original coal and in char and ash particles obtained in a drop tube furnace were analyzed by advanced methods such as CCSEM and SEM microprobe methods. Transformations in early and late stages of combustion were described.

In the study of mineral transformation during pulverized coal combustion [6], different coals showed different behavior in ash formation. In some coals, coalescence among illite, kaolinite and quartz minerals was dominant, while in others, pyrite fragmentation was observed. Transformation of pyrite and illite was studied separately [7].

Advanced analytical methods in the study of mineral transformation in coal combustion have also been performed [8]. CCSEM methods of coal mineral analysis were developed, and they used methods such as Mössbauer spectroscopy to supplement the analytical work.

Recently the Combustion Research Facility of Sandia National Laboratories engaged in the study of mineral matter transformation and char fragmentation [9, 10]. The long term goal is also the prediction of ash formation and deposition behavior of coals in combustion equipment.

Virtually all these fundamental studies of coal ash formation utilized some form of laminar flow reactor, most often a drop tube furnace. Reference 6 used a precombustor on top of the drop tube. References 9 and 10 employ an upward-flowing laminar-flow reactor with a flat flame for heating the gas and igniting the coal. Most facilities have gas temperatures lower than 1750 K because of the heating method and material limitations. The high temperature drop tube in references 1 through 3 used plasma heating with Argon gas up to 2100 K. Most facilities inject the coal on the centerline, but a few employed dispersed coal injection.

The problem of mineral transformation and ash formation and deposition is very complicated. The coal and its mineral composition, the method of firing, and the conditions of combustion all contribute to the ash behavior. A simple concept of ash fusion temperature cannot give reliable prediction on boiler deposition. A great deal of progress has been made in recent years to solve this problem through fundamental investigations, and important advances have been made. However, up to now, the studies have only begun to clear up some of the physical phenomena involved in mineral transformation and ash formation, and most understanding is generally qualitative. There are still many significant questions in all phases of the ash formation and deposition problem that must be answered. Quantitative results which can be used as database in comprehensive models for reliable prediction of boiler deposition problems are required.

UIUC has been performing basic research in coal combustion for many years. One of the most recent projects has been the study of combustion characteristics of desulfurized Illinois coal and char. An Entrained Dust Flow Facility (EDFF) and a Drop Tube Furnace Facility (DTFF) have been used to study the ignition, flame speed and emission characteristics of parent and treated Illinois coals. The DTFF provides the capability to study single particle combustion with simultaneous measurements of particle temperature, morphology and velocity of the coal. The effects of natural gas cofiring have been studied, and it was found that cofiring decreases ignition delay time for low volatile coals or chars. It was also found that addition of natural gas increased sulfur retention in the ash, so the reduction in sulfur emission could be more than the simple displacement by natural gas.

The availability of the DTFF as a research tool for accurately controlling the environment of coal particle combustion and the possibility of increased sulfur retention in ash make further study into the details of sulfur transformation an important next step. Since ash deposition is a major problem with boiler operation, ash behavior under conditions simulating the actual boiler is also a very important consideration. Therefore the DTFF is used to further study sulfur and mineral transformation as affected by coal type, treatment methods and natural gas cofiring.

Summary

This research project seeks to provide a better understanding on how sulfur and minerals are transformed in combustion and how the coal type, treatment methods and natural gas cofiring affect these transformations. Thus better methods of reducing sulfur emission and ash deposition problems may be found.

The original DTFF at UIUC was designed to obtain accurate in situ diagnostic data on individual coal particles during transit through the drop tube. It was not convenient to use the original furnace for gathering large amounts of ash in a short period of time. Also the temperature and heating rates of the original DTFF did not simulate the conditions in the hottest flames of large boilers or in the advanced slagging cyclone combustors. Therefore, the DTFF was modified to become an apparatus more convenient for ash studies and collection, with high temperature capabilities encountered in large modern boilers.

EXPERIMENTAL PROCEDURES

The emphasis of this project is on the transformation of sulfur under conditions representing those in the actual furnace, the mechanism of ash formation at high temperatures, and the effects of natural gas cofiring on these processes. To study sulfur retention in ash, the mechanism and rate of sulfur transformation must be better understood. The selected experimental technique is to heat coal particles in a drop tube furnace at boiler heating rates (10^4 ~ 10^5 K/s), temperatures (1250~2000 K) and residence times (tens of milliseconds to the order of seconds). The samples are quenched and collected for observation and analysis.

Ash characterization studies require sufficient quantities of char and ash samples for reliable analysis. Since the original DTFF was designed to work with single particle behavior of coal, collection of a sizable sample in this facility would take a very long time. So a continuous coal feed with higher flowrate in a larger diameter tube is required. This leads to the construction of the Ash Characterization Facility discussed below. To reach the high temperatures sought in this program a High Temperature Drop Tube Facility is used.

Ash Characterization Facility

The Ash Characterization Facility is diagramed in Figure 1. The central part of the ACF is a SiC furnace tube (35mm i.d., 44.5mm o.d., 0.75m long) mounted inside a furnace formed by B&W K-30 insulating firebricks rated at 1600°C maximum temperature. Eight Kanthal Super 33-3/6mm x 315mm long heating elements made of molybdenum disilicide material are arranged in two rows. These are heated by electricity from two variac transformers with step down transformers. The furnace can be run at 1450°C (1723K). Four Pt/Pt-Rh thermocouples monitor the temperature along the length of the tube.

A long brass tube with flow straightening vanes is mounted on top of the SiC tube. It provides a flow straightening and steadying section for the furnace tube. It greatly reduces the fluctuation in gas flow and gas temperature in the upper portion of the heated tube. A water-cooled coal injection tube can be inserted to any depth in this section, or even be lowered into the heated tube itself. By varying the position of coal injection, the heating rate for the coal particles can be effectively changed.

This tube permits a larger flowrate of coal with longer residence times. Coal particles are injected with a continuous feeder through the watercooled injector. Several hundred milligrams of ash or char can now be obtained in a period of 1/2 hour.

Figure 1. Ash Characterization Facility

High Temperature Drop Tube Furnace

The basic philosophy in laboratory study of pulverized coal combustion is to create controlled environments which are representative of the conditions in real combustion equipment and to study the behavior of the coal particles in these environments. The devolatilization, ignition, volatile combustion, char combustion, mineral transformation and ash behavior are studied in these controlled environments. This is the idea behind all the drop tube furnaces widely used for pulverized coal combustion study.

The main parameters characterizing the environment of coal combustion are the gas composition, temperature and residence time. The particle heating rate, which is very important to the devolatilization and ignition processes, is determined by the temperature distribution and heating conditions. The gas composition is controlled by the feed gas, the temperature by the heating of the tube and preheating of the gases, the residence time

controlled by the length of the tube and/or the flow velocity. A variation of the position of coal injection or sampling effectively changes the tube length.

The gas temperature in the drop tube is usually limited by the material of the tube and method of heating. In the literature, most of the data with coal combustion have been obtained at gas temperatures below 1750 K (1477°C) [11-19]. One study of high temperature devolatilization [17] uses argon gas in a graphite tube heated by graphite elements and reached gas temperature of 2100 K. One recent study of study of high temperature combustion of pulverized coal [19] used a furnace without an inner tube and reached gas temperature of 1600°C (1873 K). Most of the drop tube furnaces run at temperatures around 1500 K or lower.

In the actual furnace of large boilers, flame temperatures up to 1650°C (1923 K) are commonly encountered. In advanced cyclone furnaces with liquid slag taps, flame temperatures are above 2000 K. To simulate these temperatures in the drop tube is no easy task. However, temperature is a very important parameter controlling the combustion of coal, behavior of ash, and the formation of pollutants, and it would be desirable to raise the temperature of the gas environment closer to the actual value encountered in the real situation.

To heat gases to temperatures above 2000 K, plasma heaters can be used. In fact, both arc [17, 18] and induction [16] plasma heaters have been used in drop tube furnaces. But due to difficulties in plasma heater design, the past work all used argon as the main working gas. Furthermore, since the flows were at low velocity and low Reynolds numbers, the gas quickly cooled down to the wall temperature if the wall was colder than the gas. Thus the highest temperatures for the reported data were limited to the tube temperature. Thus, it is necessary to have a high temperature heated tube even if the gas is preheated by a plasma torch to temperatures above 2000 K.

Present day electric heating technology can reach 1700°C (19731K) in furnaces using established materials. With new improved materials, 1800°C (20731K) furnaces can be built.

The electric heating elements used for 1700°C furnaces are the Kanthal Super 33 molybdenum disilicide elements, which are rated at maximum element temperature of 1800°C. Proper design of the element size and power dissipation for the desired furnace temperature is necessary, and the Kanthal Handbook should be carefully consulted. At 1700°C furnace temperature and recommended maximum power dissipation of the heating elements, the power dissipation is 6W/cm². For the present size furnace, Kanthal Super 33-6/12mm elements are required.

For even higher temperatures, the new product Kanthal Super 1900 heating elements can be used, but the cost is much higher. Special hot surface refractories are also required.

The thermal insulation for the 1700°C furnace is of the low-thermal mass type to allow rapid heating up. The Zircar Company supplies a series of fiber ceramic insulation materials. The hot surface is SALI material (an alumina fiber ceramic material) rated at 1700°C, or the ECO-25 material (a looser material, also rated at 1700°C). Outside of this is the blanket material alumina LD Mat, rated at 1650°C. The outside shell is RS-100 board, rated to 1300°C and quite strong.

The furnace tube material needs to withstand the high temperature and the oxidizing environment. It must not crack under the thermal stress caused by uneven or rapid heating/cooling. Alumina can stand high temperature and oxidizing environment. but

due to its high thermal expansion and low thermal conductivity, it may not stand the rapid heating too well. Therefore, a silicon carbide tube, which has much higher thermal conductivity and lower thermal expansion than alumina, was selected. It did not crack under the sudden application of plasma heated gas injection. However, it did crack when a large water-cooled tube was inserted repeatedly into the hot SiC tube in close proximity to the wall. So any vigorous cooling of part of the heated tube is to be avoided. SiC oxidizes at 1700°C . Therefore it should not be used at higher temperature than this value.

The present high temperature furnace design is shown in Fig. 2. Initial tests were run with Kanthal Super 33-3/6mm heating elements. When plasma torch was used, the furnace temperature exceeded 1700°C , and the element temperature was too high. Great care should be exercised to monitor the element temperature by watching the resistivity of the element, and to reduce the power input to the element if the temperature (indicated by resistivity) rises up to the limit of operation. In the future, Super 33-6/12 element and a suitable step-down transformer should be used for the high temperature furnace.

Figure 2. High Temperature Furnace

The temperature profile in the high temperature furnace tube is shown in Fig. 3. The furnace temperature with no gas flow was 1650°C (1923 K). With higher gas flow (unpreheated), the gas temperatures were lower because the tube wall was cooled by the

Figure 3. Temperature Profile in the High Temperature Drop Tube Furnace

incoming gas and could not be kept at the initial high temperature. When the incoming gas was heated by the plasma torch, there was less heat loss from the furnace, and the temperature became higher. Also, the temperature distribution became flatter, with the gas coming in at higher temperatures. The gas temperature reached 1625°C (1900 K), which is higher than the reported data obtained in other drop tube furnaces.

Plasma Torch Gas Heater

In scientific research and industry, there are needs for continuous gas streams at temperatures of a few thousand Kelvin, which are not easily reached by ordinary heating methods. Plasma generators can provide such gases quite conveniently, and can be used in many cases.

In the drop tube furnace for coal combustion studies, the gas needs to be preheated before it enters the tube so that the coal particles will enter a hot environment as soon as they leave the injector. In this way, high heating rates simulating those in the real boiler furnace may be achieved. Furnace type preheaters have often been used (e.g., reference [20]). Plasma heaters, either using the arc [21, 22] or high frequency induction [23], have been used to preheat argon gas as an inert in drop tube furnaces. Although the plasma preheaters can heat gases to high temperatures, the gas temperature in the test section soon reaches that of the surrounding tube due to heat loss to the wall. Thus the reported test data were all limited to the tube temperature. In reference [21], although a gas temperature of 2000K was reached, the reported data were only given up to 1750 K. In reference 22, a graphite tube heated by graphite elements was used, and arc heated argon gas was injected (no oxygen). The temperature reached was 2100 K for devolatilization studies at high temperatures.

In reference 24, the authors studied high particle temperature combustion of pulverized coal in high oxygen concentration gases. The furnace used did not have a heated tube, and the gas was preheated and entered the furnace with direct radiation heating by the walls and the LaCrO_3 heating elements. The gas temperature reached 1600°C (1873 K) in the tests.

The aim of the present work is to study sulfur and mineral transformations in coal under controlled conditions simulating real boiler furnaces. In large boilers and cyclone furnaces flame temperature can reach 1700°C or even higher. Thus it is desirable to be able to test coal combustion at these high gas temperatures. However, due to limitations in heating and in insulating materials, most furnaces are limited to temperatures below 1500°C. A 100°C increase at such temperature levels is not easily achieved. At present, 1700°C furnace technologies are available (e.g. see reference 25), and plasma gas heaters can readily produce high temperature gases. So it is reasonable to expect that high-temperature drop tube furnaces operating at 1700°C (1973 K) are possible.

Assuming that nitrogen gas at 2000 K is flowing down the tube with a velocity of 1 to 2 m/s, the flow rates required for two different sized tubes are as follows (specific volume of nitrogen at 2000 K and 0.1 MPa is 5.937 m³/kg):

<u>Tube diameter</u>	<u>Velocity</u>		
	<u>1 m/s</u>	<u>1.5 m/s</u>	<u>2 m/s</u>
35 mm	0.162 g/s	0.243 g/s	0.324 g/s
50 mm	0.331 g/s	0.497 g/s	0.662 g/s

A gas preheater capable of providing such gas flows has been constructed. A d.c. arc plasma torch has been designed to give steady and repeatable performance, to be free from operational and maintenance problems, and to operate easily. Test results show that these requirements have been well met

Details of the construction, calibration and operation are in Appendix A. The torch contains a thermionic cathode, a gas swirl chamber and a stepped anode. The cathode is a small thoriated tungsten rod embedded in a water-cooled copper tip. This enables the tungsten rod to be well-cooled on the sides and limits the arc root to the center of the rod. The erosion of the cathode in non-oxidizing atmosphere is kept to very low values, and the life under the present operating conditions ($I < 50A$, 1 atm, N_2) can be hundreds of hours.

The anode is made of copper, water cooled. The arc is drawn through the throat by the action of the swirling gas, and attaches to the enlarged passage downstream. The point of attachment of the arc depends on the gas flow rate, the arc current, and the amount of swirl. However, the attachment point will not go beyond the exit plane of the anode, thus limiting the arc length and the arc voltage. The gas swirl tends to rotate the arc root around the periphery of the anode passage, but due to the small current with the accompanying small diameter of the arc column, the aerodynamic force is not large enough to rotate the arc root evenly all the way around the periphery but only covers part of the circular path. At the present conditions of operation, there is no evident sign of serious erosion, and the anode should last for several hundred hours of operation. The gas injection ring has one tangential hole to produce a swirling gas necessary for stabilizing the arc on the axis. Sealing of the gas injection ring in the torch body is achieved by using paper gaskets at both ends of the ring. The number of holes (one) was arrived at through experimentation for best effect on arc stabilization.

The arc is started by a high-frequency, high-voltage discharge provided by the power supply. Once the gas breakdown occurs in the gas between the electrodes, arc current flows through the gap and the arc is formed. It is immediately swept into the final configuration by the aerodynamic action of the blowing gas. A mechanical contact starter consisting of a spring-loaded tungsten rod and actuated by a solenoid-lever system was also successfully employed to start the arc. It can be used if the power supply does not have the high frequency, high-voltage starting circuitry.

Nitrogen is used as the working gas of the plasma torch and a mixing section is provided for mixing in oxygen or other gases downstream of the torch exit. This is done because it is necessary to have the composition of the gas in the drop tube furnace closely simulating the furnace gas environment in the actual boiler. It is more difficult to run the plasma torch on nitrogen than on argon because of the differences in thermodynamic and transport properties of the two gases. In fact, all the other investigators who used plasma heating for coal studies used argon as the main working gas. With suitable design of the torch and a suitable power supply, steady and continuous running on pure nitrogen has been achieved in the present plasma torch.

The power supply used for this plasma torch is a plasma cutting power unit of the inverter design (L-TEC PCM82i). This type of power supply gives high output voltage (open circuit voltage 280V, rated output voltage 120V) at low current (10-80A, 50A continuous), which is suitable for this torch running on pure nitrogen. The inverter design is a new development in plasma cutting power supplies. It features a rectifier, an inverter which produces a high frequency a.c. current with feedback control, and a final rectifier which provides the output d.c. current. Since the inverter works at high

frequency with a sensitive feedback circuit, the power supply is very compact (only 285 mmW x 485 mmH x 515 mmD, weighing 44 kg), and the current control is fast responding and accurate.

Gas from cylinders is metered through rotameters. Circulating cooling water is provided by a pump through proper filters. Water pressure of about 0.3 MPa (~45 psig) is used.

A set of performance data for the plasma torch with the mixing section attached (but no gas mixed in) is shown in Fig. 4. These data can be used in selecting the setting of the plasma torch. When additional gases are mixed in the temperature is lowered, as shown in Fig. 5. The arc voltage values are for a given position of the cathode relative to the anode. If this position is adjusted, higher or lower arc voltage will result.

It can be seen from these data that high temperature gas flows at the desired temperatures and flow rates can be readily obtained from the plasma torch. Operation of the torch is steady, can be continuous for many hours (or days, if necessary), and the power consumption is in the range of 1~3 kW. The equipment are of a manageable size. Other than the present application of preheating gas for the drop tube furnace, this plasma torch can be used in many applications where there are needs for high temperature gas streams.

Solids Sampling

The sampling probe consists of four concentric thin-walled stainless steel tubes. The outer three form the cooling water jacket and the inner one supplies a stream of helium quench gas. The helium is injected inward near the tip of the probe to quench the reaction of the particles entering the probe opening. A vacuum pump draws the gas through the probe, and ash samples are collected in the stainless steel gauze filter near the probe tip and also on the filter paper at the bottom.

The ash samples are analyzed for total sulfur in the laboratory with the LECO Sulfur Determinator. This instrument uses a high temperature furnace with oxygen gas to burn out all the sulfur in the sample. Infrared absorption in the instrument test cell provides the quantity of resulting sulfur dioxide. A computer shows the results of sulfur content. This instrument is very sensitive and accurate even with a few milligrams of sample. However, the high temperature ceramic tube in the instrument is prone to failure if the furnace is cooled down too rapidly and too often. The other elements are analyzed by the Illinois State Geological Survey (ISGS). Silicon is used as a tracer to calculate the material balance, assuming that it is kept in the coal ash without evaporation throughout the combustion process.

The Scanning Electron Microscope in the Center of Electron Microscopy of UIUC is used to observe the morphology of the coal ash and char.

The variables in these experiments are coal type, furnace temperature, methane concentration and residence time. Residence time is controlled by the flow velocity and tube length. The details of the experimental methods for these ash collecting tests are given in the FINAL TECHNICAL REPORT for the project entitled "Combustion of Illinois Coals and Chars with Natural Gas." When reliable data are obtained as a function of these variables, the sulfur transformation mechanisms can be assessed and comparisons with model predictions are possible.

Figure 4 (a). Performance of the Plasma Torch (with the mixing chamber)

Figure 4 (c). Performance of the Plasma Torch (with the mixing chamber)

Figure 4 (c). Performance of the Plasma Torch

Figure 5. Effect of Mixing Cold Gas with Plasma Flow on the Exit Gas Temperature

RESULTS AND DISCUSSION

The results of the work performed are of two kinds: the new experimental capabilities obtained by the modification and construction of the testing apparatus, and the results of coal combustion with and without methane addition.

The new test capability consists of the original Drop Tube Furnace Facility for detailed study of single particle coal devolatilization, ignition, combustion and ash transformation, the Ash Characterization Facility for studies emphasizing the transformation taking place in the char and ash, and the High Temperature Furnace stresses the effects of high temperature and heating rate on combustion and ash behavior. The diagnostic capabilities consist of the LECO Sulfur Determinator for total sulfur analysis of the solid samples, the Hewlett-Packard Gas Chromatograph for the gas sample analysis, the Scanning Electron Microscope in the UIUC Electron Microscopy Center, and the cooperative arrangement with the Illinois State Geological Survey on analysis of char and ash samples.

The capabilities of the Ash Characterization Facility is summarized in the following table:

<u>Operating Characteristics</u>	
Particle Size	< 350 microns
Heating Rate	1 to 5×10^4 K/sec typical
Gas Temperatures	1700 K
Residence Times	0 to 1.5 sec
<u>Measured Quantities</u>	
Particle Size	
Gas Analysis	
Particle Morphology	
Ash Content	

The capabilities of the High Temperature Drop Tube Furnace are summarized in the following table:

<u>Operating Characteristics</u>	
Plasma Torch Gas Preheater	On or Off
Gas Temperatures	Up to 1900 K
Heating Rates	1 to 4×10^5 K/sec
Residence Times	0 to 300 msec
<u>Measured Quantities</u>	
Particle Size	
Gas Analysis	
Particle Morphology	
Ash Content	

The gas temperature in the High Temperature Furnace is higher than the reported data in other drop tube furnaces. This is made possible by the use of high temperature furnace design and the plasma torch preheating of gases. At present, this seems to be near the limit of the gas temperature in drop tube furnaces for oxidizing atmosphere because of the materials limitations. Other heating methods may be employed for higher heating rates and/or particle temperatures, e.g. pulsed laser heating. For testing under higher gas temperatures, methods avoiding constraints of wall conditions may be considered, e.g. coal particle combustion testing in plasma heated jets, or tests using the shock tubes.

The test results of coal combustion in the Ash Characterization Facility are given in detail in the FINAL TECHNICAL REPORT for the project entitled "Combustion of Illinois Coals and Chars with Natural Gas".

These results were obtained after many preliminary tests in which many anomalies were discovered and resolved by improvements in the test apparatus and diagnostic method. The tests were made with char and ash sampling under different temperatures and residence times with 0 to 4% methane cofiring. The results showed that the combustion rate increased with methane cofiring. Sulfur retention in the ash decreased with increasing temperature and residence time. The effect of methane cofiring on sulfur retention in the ash is seen to be a complex one, with thermal and/or chemical factors playing important parts. Thus it is necessary to clarify the mechanism of the combustion process with or without methane cofiring in order to well define its effects.

CONCLUSIONS AND RECOMMENDATIONS

- A. The test capabilities of the Drop Tube Furnace Facility have been extended by establishing the Ash Characterization Facility and the High Temperature Furnace. These facilities enable larger quantities of ash and char to be collected in a relatively short time period under conditions covering a wide range of temperature, gas composition and residence time. The High Temperature Furnace has a plasma torch gas preheater working on nitrogen gas and the drop tube can reach gas temperature of 1900K, which is considerably higher than the reported values of drop tube tests in the literature. The aerodynamic coal feeder and the water-cooled sampling probe developed during this year worked well on these drop tube furnaces.

The scanning electron microscope, the LECO Sulfur Determinator, the Hewlett-Packard gas chromatograph all work well and can provide analysis data on the solid and gas samples. Together with analyses performed by the Illinois State Geological Survey on the carbon, silicon and sulfur species in the solid samples, they constitute the diagnostic capabilities on coal ash and char collected in the drop tube furnaces.

It is recommended to use these facilities fully to investigate the devolatilization, combustion, sulfur transformation, and ash behavior of various types of coal in the next year.

- B. The experiments made with coal samples with and without CH₄ cofiring showed that CH₄ cofiring changes the temperature distribution and gas composition in the drop tube, resulting in significant changes in the combustion behavior of the coal. The sulfur retention in the ash is seen to be strongly affected by the temperature and residence time. The process is a complex one, and detailed studies are needed to

clarify the mechanisms involved. It is recommended that tests be made at various temperatures and residence times to study the transformation of sulfur and other components as a function of the combustion process, and to study the effect of high temperatures on burning rate and ash behavior.

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APPENDIX A: THE PLASMA TORCH GAS HEATER

Drawing of the Plasma Torch

Figure A.1. Schematic of Plasma Torch

Calibration and Performance of the Plasma Torch

Since the gas jet has a temperature of a few thousand Kelvin, thermocouples cannot be used for temperature measurement. The average temperature of the gas jet is obtained by energy balance and using equilibrium thermodynamic properties. The torch and mixing section were tested as one unit, so the temperatures and efficiencies given are those at the exit of the mixing section. The arrangement for calibration was either of the two shown below:

1. Calibration Arrangement 1.

From the energy equation,

$$\dot{m}_g (h_2 - h_1) = U I - \dot{m}_{H_2O} c_p \Delta T$$

where

h_2 = enthalpy of exit gas (hot),

h_1 = enthalpy of inlet gas (cold),

U, I = arc voltage and current, respectively,

$\dot{m}_{H_2O}$ = flow rate of cooling water,

$\dot{m}_g$ = flow rate of working gas,

ΔT = temperature rise of cooling water, and

c_p = specific heat of water.

Thus, h_2 can be calculated from the measured values of $U, I, \dot{m}_{H_2O}, \dot{m}_g, \Delta T$, and known thermodynamic data. T_2 is obtained from the h - T data of the gas.

Thermal efficiency of the system is defined as

$$\eta = [(h_2 - h_1) \dot{m}_g] / (UI) = 1 - (\dot{m}_{H_2O} c_p \Delta T) / (UI)$$

It can be seen that if the efficiency is low (i.e. heat loss is a large part of UI), the percentage error in temperature and efficiency will be larger.

2. Calibration Arrangement 2.

In the second arrangement, a calorimeter is connected to the exit of the mixing section and cools down the gas to a low exit temperature T_e . This temperature can be measured by a thermocouple. The energy equation for the calorimeter is:

$$\dot{m}_g h_2 - \dot{m}_{H_2O} c_p \Delta T = \dot{m}_g h_e$$

or

$$h_2 = h_e + (\dot{m}_{H_2O} c_p \Delta T) / \dot{m}_g$$

and T_2 is obtained from h_2 .

This arrangement gives better accuracy especially at low efficiencies. The water flow through the calorimeter should be steady and adjusted to give a good reading of ΔT .

In the present work, both arrangements have been tested, and comparable accuracies have been obtained. The data presented here were obtained with arrangement 1.

Operating Procedures

1. Safety Precautions

The power supply has an open circuit voltage of 280V and the operating voltage with the arc may be 120V. Contact with these voltages may be lethal. Make sure that the cathode circuit is well insulated at all points.

The high temperature N_2 will form harmful nitric oxides when in contact with air. The exhaust gases should be vented when testing in the open jet mode.

The noise from the open jet can be harmful to the ear, especially at higher power levels. Protective ear plugs should be worn.

The bright light from the arc contains ultraviolet rays which are harmful to the eye. Arc welding goggles must be worn when looking directly at the arc.

2. Before Starting Torch

Make sure that all connections for gas, water and electricity are correctly made and secure. Make sure there is a constant supply of cooling water and working gas (dry nitrogen).

3. Starting the Plasma Torch

- a. Turn on the circulating water pump.
- b. Turn on the power supply main switch, set current control to 25A.
- c. Turn on gas supply, adjust N₂ flow through the torch to 120 on rotameter.
- d. After making sure the gas and water are flowing, turn on the arc starting switch. The torch should start after a 2-second delay. The switch should be left on.

4. Running the Plasma Torch

- a. Adjust the N₂ flow to desired value (not below 80 on rotameter).
- b. Adjust the arc current to desired value (not over 50 A).
- c. Record arc voltage, current, gas flow, check from time to time.

5. Terminating the Plasma Torch

- a. Turn off the starting switch. The arc should stop.
- b. Turn off gas flow and power supply main switch. Keep the cooling water on until the furnace is cooled down sufficiently.

6. Check Points During Operation

- a. Cooling water circulating normally both in the torch body and in the cathode. No water leak at any point.
- b. Gas flow steady at set rate.
- c. Arc voltage steady at calibrated value. When new electrodes are installed, adjustment in the axial position of the cathode may be necessary to reproduce the arc voltage value at given arc current and gas flow.
- d. If any abnormality occurs, stop the plasma torch and examine the torch, the gas, water and electrical circuits.

7. Maintenance

After about 50 hours of running time or when operation shows irregularity, examine the electrodes for erosion. If there is excessive erosion, the parts should be replaced by new ones.

APPENDIX B: THE HIGH TEMPERATURE FURNACE

Operation

1. Heating up the furnace:
 - a. Check over the rig, make sure all parts are in order.
 - b. Turn on the cooling water, make sure all lines are flowing properly.
 - c. Turn on the exhaust fan (switch in 115F MEL).
 - d. Turn on the electricity to the heaters. At high furnace temperatures, check the resistivity value of the heating elements by calculating from the voltage and current readings. Make sure the elements do not exceed 1750°C in temperature.
 - e. Monitor the furnace temperature with the Type B thermocouple. It should not exceed 1700°C.
2. Turning on the plasma torch:
 - a. Check the connections, gas, water and electricity supplies.
 - b. Operate plasma torch on heated furnace only.
 - c. Turn on the arc quickly after the gas is turned on, preheat some minutes at 25A.
 - d. Adjust gas flow and current to desired values (current not over 50A).
3. Turning off the plasma torch:
 - a. Reduce current to 25A.
 - b. Turn off the arc, then quickly turn off the gas.
 - c. Leave the cooling water on.
4. Cool off the furnace:
 - a. Turn off the power to heating elements, turn off exhaust fan.
 - b. Leave the cooling water on over night.
 - c. Shut off cooling water the next morning when the temperature is low.

Aerodynamic Feeding of Pulverized Coal

In the drop tube experiments, a controllable and steady coal feed is very important. The mechanical screw feeder often gave trouble with stoppage, so an aerodynamic feeder was developed. In the ash characterization experiments, a continuous feed rate of the order of 0.1 to 0.5 g/min is required. The aerodynamic feeder works well in this range.

The feeder works on the principle as shown in Fig. B.1. A certain amount of pulverized coal of given size range is contained in an ordinary plastic syringe for medical use. A carrier gas (N_2) is metered through the inlet tube of about 1.1 mm i.d. and blows downward toward the surface of coal particles. It fluidizes a small portion of the coal and forms a plume of fluidized cloud of coal particles. The outlet tube has its opening located near the top of this cloud, and thus the outflowing gas carries the coal particles with it. The coal in the bottom half of the syringe is pushed upwards by the motor/screw mechanism at a fixed rate. With proper adjustments, the outflowing coal just balances the upward movement of the coal surface, so that a steady state is reached.

The motor is a 1 rpm synchronous motor, the screw threads used are 20, 24, 28 and 32 threads per inch, and the diameter of the syringe are 19 and 25.4 mm. These combinations give coal feed rates from 0.15 to 0.43 g/min.

The important points for making the feeder work properly are: the opening of the outlet tube should be smooth and slightly enlarged, the inside diameter of the outlet tube should be at least 6 times the particle diameter to avoid bridging, the openings of the tubes should be beveled, the bevels should face each other, the distance between the openings should be adjusted to optimum value, the gas flow should be adjusted to optimum value.

The feeder works reliably for pulverized coal under 200 μm size. Future improvements may involve using a variable speed motor with speed reduction gears to obtain continuously variable feed rates.

Figure B.1 Aerodynamic Feeder

FINAL PROJECT MANAGEMENT REPORT

September 1, 1991 through August 31, 1992

Project Title: Cooperative Research on the Combustion Characteristics of Cofired Desulfurized Illinois Coal and Char with Natural Gas

Project Investigator: Richard O. Buckius, UIUC

Other Investigators: Cheng-Kang Wu,
Herman Krier, and
James E. Peters
UIUC

Project Monitor: Kenneth K. Ho, CRSC

COMMENTS

The attached budget material indicates that all budget activity has been completed. There were additional expences that were charged to other accounts.

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 cost-shared programs.

Projected and Estimated Expenditures by Quarter

Quarter*	Types of Cost	Direct Labor	Materials & Supplies	Travel	Major Equipment	Other Direct Costs	Indirect Cost	Total
Sept. 1, 1991 to Nov. 30, 1991	Projected	6,350	303	250		1025	743	8,671
	Estimated	5,370	70	768		2,328	854	9,930
Sept. 1, 1991 to Feb. 29, 1992	Projected	12,700	606	500		2,050	1,586	17,442
	Estimated	11,705	1,052	768		3,252	1,678	18,455
Sept. 1, 1991 to May 31, 1992	Projected	19,052	909	750		3,075	2,378	26,164
	Estimated	17,532	1,175	993		3,941	2,364	26,005
Sept. 1, 1991 to Aug. 31, 1992	Projected	25,402	1,213	1,000		4,100	3,282	36,097
	Estimated	24,962	1,175	993		5,686	3,281	36,097

Cumulative by quarter

COSTS BY QUARTER

(Place Your Project Title Here)

Months and Quarters

○ = Projected Expenditures

△ = Estimated Actual Expenditures

Total CRSC Award \$ 36,097

SCHEDULE OF PROJECT MILESTONES

Project Title: Cooperative Research on the Combustion Characteristics of Cofired Desulfurized Illinois Coal and Char with Natural Gas

Principal Investigator: Richard O. Buckius, UIUC

Other Investigators: Cheng-Kang Wu, Herman Krier, and James E. Peters, UTUC

Project Monitor: Dr. Kenneth K. Ho, CRSC

Milestones:

- A. DTFF aggregate flotation parent and treated coal parent ash data
- B. DTFF aggregate flotation and treated coal ash data
- C. Technical Reports
- D. Project Management Reports

DATE

FILMED

2 / 24 / 94

END

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