TECHNICAL REPORT

December 1, 1991 through February 29, 1992

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 effects on ash constituents) during cofiring with natural gas. The Drop Tube Furnace Facility (DTFF) at the University of Illinois at Urbana-Champaign (UIUC) is the chief apparatus to be used in this investigation, with additional diagnostic tools including scanning electron microscopy and other chemical analyses.

During the first quarter, a complete review of literature related to the project was performed, and a detailed strategy outlined for carrying out the research. It was determined that the DTFF would be modified to extend the operating range to larger sample collection capabilities and higher temperatures (up to 2000 K). A plasma gas heater was been designed and constructed so that temperatures and heating rates encountered by pulverized coal particles in the flames of large boilers or in the advanced slagging combustors are better simulated. Also, a LECO Sulfur Determinator for total sulfur analysis was refurbished, and arrangements were made with the Illinois State Geological Survey for the analysis of char and ash at UIUC.

In the current quarter, the modification of the DTFF has been completed. Now the facility includes a 1.5 m silicon carbide tube which can be used up to a temperature of 2100 K (as compared to 1750 K with electrical heating alone). A continuous coal feeder with a steady feedrate has been developed for the modified furnace. The plasma torch has been tested and calibrated, and a steady operation has been established with parameter variations of less than 2%. At a power input of 2.6 kW, the temperature of the nitrogen effluent can reach approximately 4000 K. Lower gas temperatures are obtained by reduced power and mixing with cold gases.

The specific ash characterization experiments are reported in the quarterly TECHNICAL REPORT for the project entitled "Combustion of Illinois Coals and Chars with Natural Gas." Note that preliminary runs with IBC106 coal with and without methane cofiring in the furnace have produced consistent results with the sulfur analyzed by the LECO Sulfur Determinator.

A series of tests will be performed in the next quarter to determine how the overall operating conditions affect sulfur transformation. The furnace will then be tested with the plasma heater to reach the designed gas temperature of 2000 K.

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EXECUTIVE SUMMARY

The reduction of sulfur emissions from the combustion of Illinois coal is very important and urgent. Research on the fundamental level can provide a better understanding of the processes 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 co-firing natural gas with Illinois high-sulfur coals to determine the sulfur retention potential as a function of sulfur content, sulfur type and natural gas concentration. Previous experimental results from UIUC have shown that co-firing natural gas and coal has the potential for reducing sulfur emissions by increasing sulfur retention in the ash. Additionally, other researchers have suggested that co-firing 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 literature shows that present works 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 for lack of a suitable apparatus. Also, the effect of natural gas co-firing has not been well studied. 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.

The Drop Tube Furnace Facility has been modified to provide an environment with a heating rate comparable to that in the boiler furnace (10^4 ~ 10^5 K/s) up to a temperature representative of boiler flame temperatures (1500~2000 K). Various residence times from tens of milliseconds up to seconds are possible. The samples are quenched and collected for observation and analysis. Both chemical analysis and scanning electron microscopy (SEM) have been and will be used. The high gas temperature will be obtained through using a plasma torch gas heater which will heat nitrogen to over 2000 K. Oxygen will be mixed in downstream to provide gas flow of the desired temperature and composition. A tungsten/rhenium thermocouple will be used to measure the high temperature in the inert gas. The coal is injected along the axis directly into the high temperature gas to achieve the high heating rate. A sampling probe positioned at various positions along the axis produces varying residence times. A LECO Sulfur Determinator loaned from another laboratory in the department can provide total sulfur values, and cooperative arrangement with the Illinois State Geological Survey (ISGS) will provide chemical analysis and SEM-EDX (energy-dispersive-X-ray analysis) methods for the char and ash samples.

The work in the first quarter consisted of the literature survey, exploring and defining the relationship between requirements of Clean Coal Technology, especially those directly affecting the use of Illinois coal, and the works of the present project, and setting up of the strategy of the present project. With the desired range of parameters defined, the apparatus modification was initiated. A larger diameter silicon carbide tube was ordered to replace the alumina tube so that high temperature operation is possible. A plasma torch gas heater was also designed and constructed. Initial ash characterization experiments were performed.

In the current quarter, the modification of the DTFF was completed. The apparatus is now relocated in the Mechanical Engineering Laboratory. The furnace has a 35 mm (i.d.), 1.5 m long, silicon carbide tube which can be used up to a temperature of 2100 K (as compared to 1750 K with electrical heating alone). The LECO Sulfur Determinator has

been fully calibrated and tested. It can give accurate total sulfur readings for coal, ash or char down to very small sample sizes (a few milligrams). Preliminary runs with IBC106 coal with and without methane addition have produced consistent results to those previously obtained. The plasma torch has been tested and calibrated. A set of conditions for steady running with variation in parameters less than 2% has been established. At a power input of 2.6 kW, the temperature of the nitrogen effluent is 4000 K, and lower temperatures are obtained by reducing power and mixing in cold gases. An adaptor has been constructed to connect the torch to the DTFF.

With the apparatus preparation close to completion, experiments can now begin on the study of the effects of operating conditions on transformation of sulfur in coal during combustion. The first series of test will be on the effect of methane co-firing on sulfur retention in ash. The variables will be coal type, furnace temperature, oxygen content, residence time, and methane content. The sulfur, silicon (as a tracer material), and carbon content of the ash sample will be determined. From these data, some understanding of the sulfur transformation during combustion can be obtained and further study into the mechanisms can be outlined. Later, the furnace will be tested with the plasma torch heater to gas temperatures of 2000 K. A longer sampling probe will be built to enable samples to be taken at various residence times.

Additional ash characterization experiments are reported in the quarterly TECHNICAL REPORT for the project entitled "Combustion of Illinois Coals and Chars with Natural Gas."

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OBJECTIVES

The overall objective of the project is to determine the effects of treatment processes and natural gas co-firing on the ash characteristics for Illinois coal, with a view towards 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 year are to obtain sulfur and mineral transformation data under well controlled conditions in the Drop Tube Furnace Facility (DTFF) for two Illinois coals (high pyritic and high organic sulfur) and their treated products, with and without natural gas co-firing, thereby gaining an understanding of the mechanism and potential in reducing the sulfur emission and ash deposition problems. Each set of data will consist of a number of data points taken at various residence times under identical conditions of gas temperature, composition and flow velocity.

The tasks to be completed in order to meet the objectives for the current year are the following:

1. Formulation of the complete strategy, methodology and outline for this research combined with initial ash characterization experiments. (1st quarter)
2. Modification of the experimental apparatus and methods of analysis. (1st - 2nd quarter)
3. Refine the apparatus and perform further experiments. (2nd quarter)
4. Running of the experiments to obtain a series of reliable data. (2nd - 4th quarter)
5. Analysis of the data to arrive at an assessment of the effects of the various factors and an understanding of the processes involved. (4th quarter)
6. Project completion and recommendations for future work. (4th quarter)

At the end of the second quarter, the tasks 1 through 3 above have been completed as planned. The remaining tasks will be performed in the next two quarters. Also the next period will include the initial testing at high temperatures as provided by the plasma torch.

INTRODUCTION AND BACKGROUND

There are many significant research issues impacting the use of our coal resources including the mechanism of pollutant emission and methods of pollution control, methods to achieve stable and efficient combustion, mineral matter transformation which mainly affects slagging and fouling in boilers, combustion modeling, database on fuels and combustion characteristics, diagnostic methods, etc. At present, the environmental requirements as exemplified by the Clean Air Act Amendment of 1990 is posing an important and urgent task to 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 and jobs. 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.

UIUC has been doing 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 co-firing have been studied, and it was found that co-firing 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 per cent substitution 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 worth studying. Therefore it was decided to use the DTFF in a further study of sulfur and mineral transformation as affected by coal type, treatment methods and natural gas co-firing.

The results of this research project will provide a better understanding on how sulfur and minerals are transformed in combustion and how the coal type, treatment methods and natural gas co-firing 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 periods 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 is being 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, ash formation and behavior at high temperature, and the effects of natural gas co-firing 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 the coal particles in a drop tube furnace at a heating rate comparable to that in boiler furnaces (10^4 – 10^5 K/s) up to a temperature representative of boiler flame temperatures (1250–2000 K) and keep them at various residence times from tens of milliseconds to the order of seconds. The samples are quenched and collected for observation and analysis.

Initial ash characterization experiments have been performed with the existing Drop Tube Furnace Facility. The specific ash characterization experiments are reported in the quarterly TECHNICAL REPORT for the project entitled "Combustion of Illinois Coals and Chars with Natural Gas." These experiments will continue along with facility modifications to obtain broader operating conditions as noted above.

The DTFF has been modified to provide the capability of operating at gas temperatures up to 2000 K with continuous coal injection along the axis. Modifications have also been performed to produce larger quantities of char or ash samples in shorter periods. A larger diameter tube (35 mm i.d.) silicon carbide tube, 1.5 m in length, was installed in the furnace. This tube permits a larger flow rate of coal with longer residence times to be considered. Coal particles are injected with a continuous screw-type feeder into the high temperature gas to achieve high heating rates. Thus, a sizable ash sample can now be obtained in a matter of minutes.

The project includes the following major steps:

1. Preparation of the Apparatus

The high-temperature DTFF has been moved to the newly remodeled Mechanical Engineering Laboratory. The plasma torch, shown in Fig. 1, has been tested and calibrated. The coal feed system has been developed. Continuous samples can be obtained with a water-cooled sampling probe.

2. Initial Testing of the Method of Studying Sulfur Transformation

For one sample with a specified gas temperature and oxygen concentration, samples will be quenched at residence times ranging from 10 ms to 1 s. After determining the extent of devolatilization and burnout, mass balances and total sulfur determinations will be performed. Once consistent trends are obtained in these environments, a set of samples will be sent to ISGS for elemental and sulfur type analysis. This set of tests will not only make sure that the experimental method is reliable, but also provide data on sulfur transformation under one particular set of conditions.

Figure 1. Schematic of the plasma torch.

3. Study of Effects of Certain Factors on Sulfur Transformation

With the method of study established, several parameters will be varied to investigate their effects. Important contributions are the extent of natural gas co-firing, amount and kind of sulfur in coal, gas temperature and residence time. From these investigations, it is hoped that some insight on how to better control SO_x emission may be obtained.

4. Study of Ash Formation and Behavior

With the use of an optical microscope, scanning electron microscope (SEM), and SEM with energy-dispersed x-ray spectrum (EDX), the ash formation, melting and coalescence processes will be studied to better understand the effects of temperature, natural gas co-firing, and coal type on ash behavior. Additionally, the possibility of studying deposition tendencies in the drop tube furnace will be considered.

5. Study of Combustion Rates at High Temperatures

Burning rate data for various coals and chars will be obtained at gas temperatures up to 2000 K. Such high temperature data are needed for modelling of combustion in large boiler furnaces and cyclone slagging combustors but have not been obtained before due to apparatus constraints.

6. Study of Sorbent Addition to Coal

Sorbents can be injected along with sulfur species in the gas to study the governing mechanisms under known temperatures and gas compositions. These data can be used to understand the processes in sorbent injection.

The above steps may not be all completed within one year. According to the proposed project, work described in items 1 through 4 will be completed before August 31, 1992.

RESULTS AND DISCUSSION

Work Performed During the Second Quarter

In this current reporting period (December 1, 1991 to February 29, 1992), the following tasks have been performed:

1. The modified DTFF has been assembled in the newly remodelled Mechanical Engineering Laboratory. The facility consists of the furnace, the heating elements, the electrical supply circuits, the cooling water circuits, the coal feeding system, the ash sampling system, and the control panel. The drop tube inside the furnace is a 1.5 m long silicon carbide tube. The coal feeder can feed the pulverized coal at a controlled rate. The system has been tested to 1750 K with the electric heater alone. All the subsystems work well.
2. Preliminary tests using IBC106 coal with and without methane addition have been performed in the modified DTFF. Ample ash samples for analysis were collected in about 15 minutes. The samples were analyzed for sulfur in the LECO Sulfur Determinator. This instrument gave accurate results even with small samples of a few milligrams when care was taken to calibrate the instrument. The results showed trends of increased sulfur retention in the ash with addition of methane, yet the absolute sulfur content was dependent on the operating conditions such as temperature and residence time. More experiments will be performed before significant amount of data can be gathered and definite conclusions reached.
3. The plasma torch has been tested and calibrated. The configuration of the cathode and anode has been optimized for arc stability and low erosion. A set of conditions has been established to yield steady operation for hours. At an electric input power of 2.6 kW, a nitrogen flow with a temperature of 4000 K was obtained. This temperature was determined from a calorimeter attached to the torch exit and measuring the heat transfer to the water and the calorimeter exit gas temperature. The gas temperature from the torch was calculated through an energy balance and equilibrium thermal properties of nitrogen. Lower gas temperatures are obtained by using lower arc power (reducing arc current) and by mixing in low temperature gases downstream of the torch. An adapter for connecting the torch to the drop tube has been made.

Work Scheduled for the Third Quarter

A series of experiments will be performed in the modified facility, at first without the plasma gas heater and with temperatures below 1750 K. The key variables will be furnace temperature, methane concentration, residence time and coal type. Total sulfur will be analyzed with the LECO instrument, and silicon, carbon and type of sulfur will be analyzed by the Illinois State Geological Survey. A longer sampling probe will be constructed to

obtain samples at various heights along the drop tube. The plasma torch gas heater will be connected to the drop tube and high temperature ash data will be gathered.

CONCLUSIONS AND RECOMMENDATIONS

To study sulfur retention in ash and deposition problems, the mechanism and rate of sulfur and mineral transformation must be understood. Our literature survey shows that present works 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 studies under boiler conditions are also lacking. Therefore, 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.

The Drop Tube Furnace Facility has been modified during the current quarter to enhance ash production and collection, so that sulfur transformation in coal during combustion can be more easily and thoroughly studied. The high-temperature plasma torch gas heater has been tested and calibrated and will be installed on the DTFF in the next quarter. Preliminary combustion tests have been run in the modified DTFF using IBC106 coal and consistent results have been obtained, showing that the apparatus is ready for use in actual data taking.

A series of tests devoted to the study of effects of coal type, operating conditions and natural gas co-firing on sulfur transformation should be run in the next quarter. With the preparatory work on the apparatus and diagnostics largely completed, the emphasis should be on experiments and the analysis of the results, while the final improvements on the apparatus are continued.

PROJECT MANAGEMENT REPORT

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COMMENTS

The attached material indicates that this budget is very close to predictions. The slight overage is a result of a sooner-than-expected use of materials and supplies funds.

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