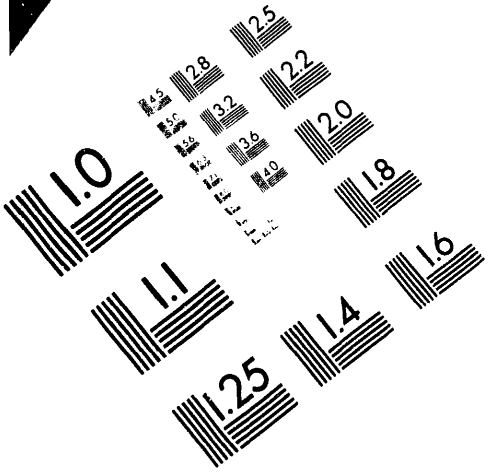

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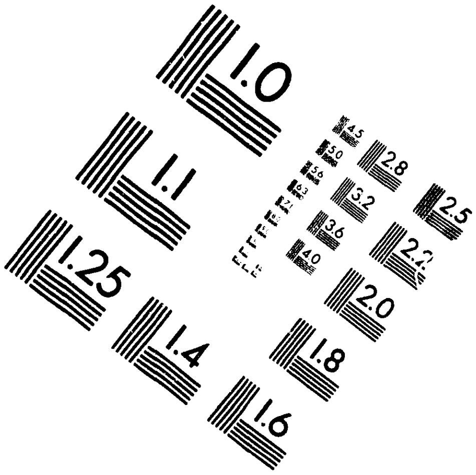

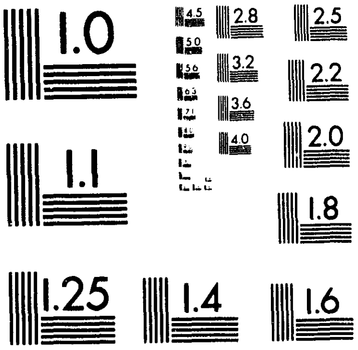

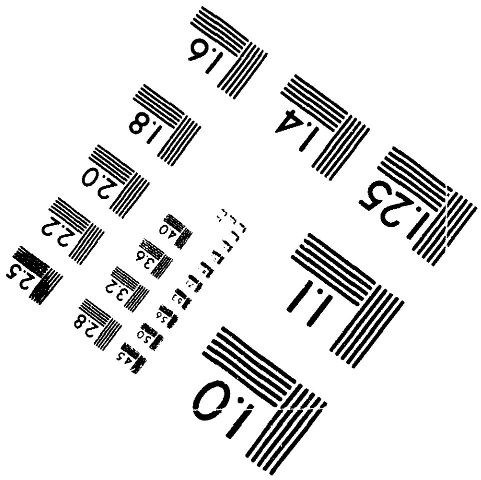

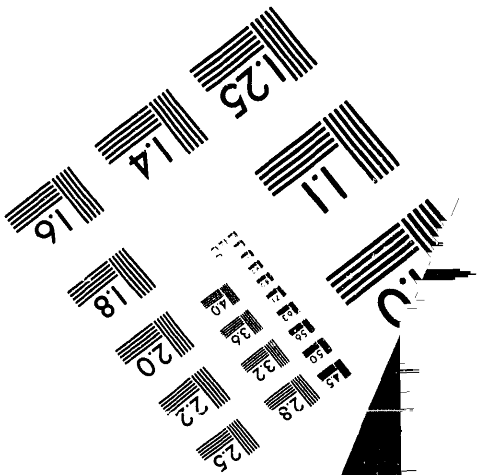

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TECHNICAL REPORT
March 1 through May 31, 1993

Project Title: **COMBUSTION AND EMISSIONS CHARACTERIZATION OF PELLETIZED COAL FUELS**

DE-FC22-92PC92521

Principal Investigator: Dr. S. Rajan
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Project Manager: Dr. Frank Honea
Illinois Clean Coal Institute

ABSTRACT

Pelletization of coal offers a means of utilizing coal fines which otherwise would be difficult to use. Other advantages of coal pelletization include: (a) utilization of low grade fuels such as preparation plant waste, (b) impregnation of pellets with calcium carbonate or calcium hydroxide sorbent for efficient sulfur removal, and (c) utilization of coal fines of low quality in combination with different types of binders.

The objective of this project is to investigate the carbon conversion efficiency and SO_2 and NO_x emissions from combusting pelletized coal fuels made from preparation plant waste streams using both limestone and calcium hydroxide as sorbent and cornstarch and gasification tar as binders. The combustion performance of these pelletized fuels is compared with equivalent data from a reference run-of-mine coal.

During this quarter, six different samples of coal pellets have been secured from ISGS researchers. The pellets are made from the spirals and flotation concentrate fraction of the Kerr-McGee preparation plant. Four samples contain limestone as sorbent in various Ca/S ratios, and cornstarch as binder. Two pellet samples contain calcium hydroxide as sorbent and are hardened by carbonation under atmospheric conditions.

Combustion and emissions characterization of these pellets in the laboratory scale 4-inch diameter circulating fluidized bed have been performed on some of the pellet samples. The pellets burn readily, and provide good bed temperature control. Preliminary results show good carbon conversion efficiencies. Oxides of nitrogen emissions are quite low and sulfur dioxide emissions are as good as or lower than those from a representative run-of-mine coal.

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EXECUTIVE SUMMARY

In late April, the following six 5-gallon pellet samples were received from ISGS collaborators:

(a) three pellet samples containing limestone sorbent (Ca/S ratio 1-3), using cornstarch as binder

(b) one pellet sample containing a coarser limestone and cornstarch as binder, with a Ca/S ratio of 2:1

(c) two pellets containing calcium hydroxide as sorbent and hardened by carbonation under atmospheric conditions, (Ca/S ratios of 1.1 and 2.3).

The pellets are processed from the coal obtained from the spirals and flotation concentrate fraction of the Kerr-McGee preparation plant at Galatia, Illinois. They are approximately 0.125 inches in diameter and 0.25-0.375 inches long. The size of these pellets is not fixed, but was selected only as being suitable for combustion in the laboratory scale 4-inch CFBC.

During the combustion evaluation of these pellets, the 4-inch internal diameter CFBC was first preheated on propane gas. When the bed temperature has reached about 1000°F, coal feed was initiated and the system brought up to operating temperatures of 1550-1600°F. A run-of-mine coal was used for this purpose. Steady state temperatures were usually attained after 40-50 hours of operation. For comparison purposes, tests were conducted with a run-of-mine coal, also obtained from the Kerr-McGee Galatia plant, and the pellets under similar operating conditions. The coal was fed using a calibrated screw feeder, while the pellets were fed using a vibratory feeder, both fuels being injected into the combustor pneumatically using a metered air supply. Variables measured during a test included air and fuel mass flows and temperatures at various locations in the combustor. Samples of the exhaust gas were taken through heated lines and the concentrations of CO, CO_2, O_2, NO_x and SO_2 measured. Timed samples of ash collected in the secondary cyclones and not recycled into the combustor were also collected for evaluating carbon conversion efficiencies.

Experiments were conducted with a pellet sample containing 5% calcium hydroxide as sorbent, which for the coal used in the pellets, translated to a Ca/S ratio of 1.1:1. Data was taken at different bed temperatures and compared with equivalent data, taken previously, from a run-of-mine coal.

Data from the experiments show that at a Ca/S ratio of 1.1:1 and a bed temperature of 1500°F, the emission levels from the pellet are on the order of 1.6 lbs/million Btu, and the NO_x emissions are 0.47 lbs/million Btu. The SO_2 values are high because the Ca/S ratio is low. Higher Ca/S ratios are expected to yield lower levels of SO_2. Comparative results with the run-of-mine coal are discussed in the body of the report.

In mid-May, the gas filter correlation HCl analyzer was received. Since one of the objectives of the present experiments is also to measure and investigate the HCl emissions from the combustion experiments, and since the quantities of pellet samples are limited, efforts since mid-May have focused on installation and start-up of the HCl analyzer. This will permit HCl emissions measurement, and data on HCl emissions will also be available from the resumed combustion tests to be conducted in the next quarter.

RESEARCH OBJECTIVES

During this one-year project period, the specific goals to be achieved are as follows:

1. to secure appropriate quantities of coal pellets made under the ICCI project at ISGS by mixing calcium hydroxide with the coal fines and carbonating the pellets to harden them.

2. to make pellets from coal fines (e.g. gob pile wastes) by mixing the coal with gasification tar or pitch impregnated with limestone.

3. to make pellets from gob pile waste coal and froth flotation fines by mixing these fines with corn starch as binder and containing limestone fines as sorbent.

4. to burn these pellets in a laboratory scale 4 inch diameter circulating fluidized bed combustor (CFBC).

5. to compare the carbon conversion efficiency, carbon balances, SO_2, NO_x and HCl emission levels and combustion waste analyses of the pelletized fuel with equivalent values obtained from a standard coal.

6. to analyze the mineral matter and elemental distribution in the combustion residues from the above fuels when burned in a laboratory scale 4-inch internal diameter circulating fluidized bed combustor.

7. to analyze the data and make appropriate recommendations.

INTRODUCTION AND BACKGROUND

The economic climate for coal usage can be improved by utilizing fines and coal cleaning wastes to produce clean burning reconstituted fuels such as coal-sorbent pellets. Currently, ICCI is supporting one such project at the Illinois State Geological Survey (ISGS) wherein different methods of making coal-sorbent pellets are being investigated. The ISGS method employs a mixture of calcium hydroxide and fine coal which is then hardened by carbonating the pellets formed. This provides an intimate mixture of calcium with the coal for effective sulfur capture. Additionally the coal and limestone sorbent can be mixed and pelletized using various binders such as gasification tars or pitch. The sulfur capture effectiveness of the ISGS method in comparison to other pellet forming techniques using other binding agents has not been investigated. The market potential of these fuels will depend on the combustion and emissions characteristics of the coal/sorbent pellets. This can be established only by combustion testing under conditions simulating actual boiler firing. Because of the small quantities of the fuels being produced, laboratory-scale combustors are best suited to demonstrate their utility and emissions characteristics.

There is a need to evaluate the burning characteristics of the coal-sorbent pellets to determine whether they can be burned easily and with good carbon conversion efficiencies. In the case of the coal-sorbent pellets, it is necessary to quantitatively evaluate the sulfur capture efficiency of the pellets, in comparison with the case where the coal and sorbent are fed separately, to establish the merits of the coal-sorbent pelletization process. Additionally, it is necessary to investigate how the pelletization process, namely the mixing of the sorbent with the coal matrix influences the porosity and carbon burnout histories of the coal. The sorbent Ca/S ratios needed to meet EPA requirements with the pellets and their carbon conversion efficiencies need to be evaluated in comparison with standard Illinois coals to demonstrate the usefulness of the pelletization process.

The research currently being performed in this project addresses the above needs to improve the usefulness of fuels produced from high sulfur coals.

EXPERIMENTAL PROCEDURES

I. Equipment and Instrumentation

The experiments are being conducted in the 4" internal diameter circulating fluidized bed combustor shown schematically in Figure 1. The combustor is lined with a castable refractory to reduce heat losses. As shown in Figure 1, a blower supplies fluidizing air which is split into two streams. The main stream enters the fast fluidized bed section of the combustor through a distributor plate specially designed to provide even fluidization. This section of the air duct also houses a propane-fired preheat system, which is utilized to bring the bed solids up to temperatures required to ignite the main fuel. Unburnt fuel, limestone and ash entrained by the gases in the main bed column pass through a refractory-lined hot cyclone, which traps the larger particles and deposits them into an auxiliary bubbling bed attached to the bottom end of the hot cyclone. The second smaller air stream enters this bubbling bed into which the carry-over solids from the fast fluidized bed trapped by the hot cyclone are deposited. A non-mechanical seal ensures that this unburnt fuel and bed solids flow from the bubbling bed into the fast fluidized bed and not vice-versa. Both air streams are metered with ASME nozzles and incorporate control valves for adjusting the flow velocities in the fast fluidizing and bubbling bed sections of the combustor.

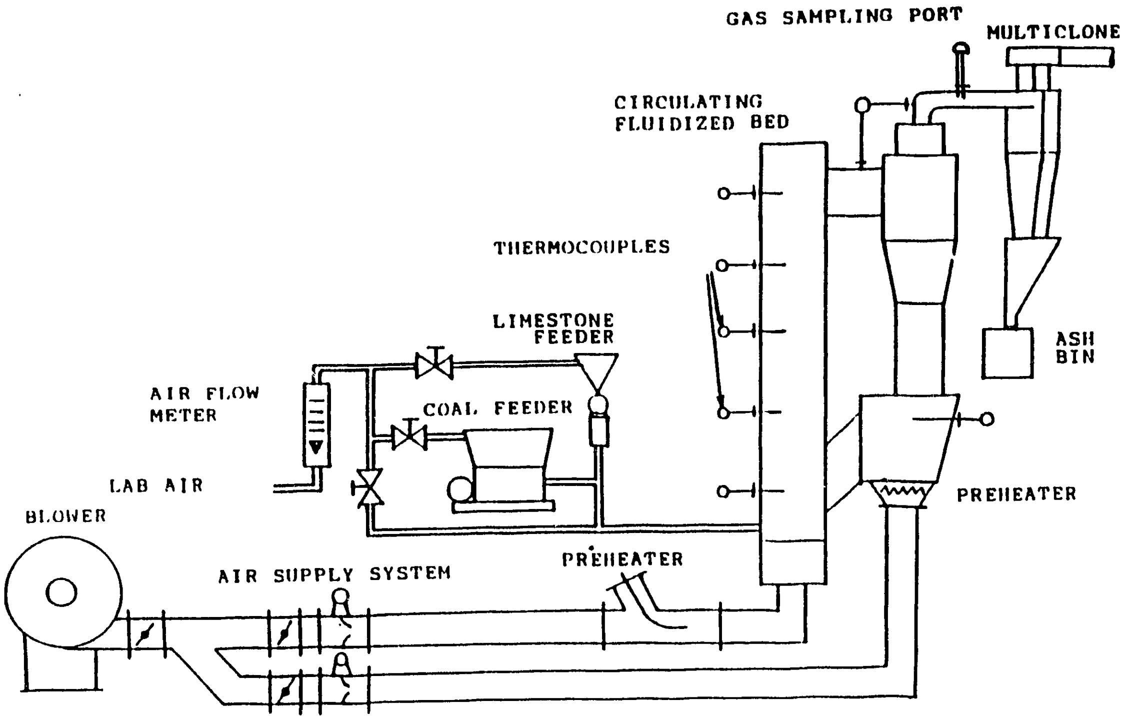

Figure 1. Schematic of 4-Inch Internal Diameter Circulating Fluidized Bed Combustor

Crushed and sieved coal is fed from a pressurized hopper via a screw feeder pneumatically into the dense portion of the fast fluidized bed, using metered high pressure air. Sized limestone, stored in a separate hopper, is fed simultaneously into the air stream, conveying the coal into the bed. Both coal and limestone feed systems have been calibrated individually.

Two quartz glass-lined observation ports, one located in the dense bed at the bottom, and the other located near the top in the dilute phase or transport section of the bed, serve for visual monitoring of the combustion process. The circulating fluidized bed combustor is instrumented with chromel-alumel thermocouples at various positions for measuring temperature. The thermocouples are connected to a selector switch and, thence, to a digital readout meter.

Solids too small to be captured by the hot cyclone are trapped in a multiclone, mounted at the hot cyclone exit. In the present system, these multiclone solids are not reinjected into the bed. The multiclone solids are later analyzed for heat content, using an adiabatic calorimeter. Combustion gases are drawn off from a point at the exit of the multiclone, filtered through 2-5 micron particulate filters, and conveyed via heated lines to an instrument panel for determining gas composition. Carbon monoxide and carbon dioxide are measured with Beckman NDIR analyzers, oxygen with a Beckman

755 paramagnetic analyzer, oxides of nitrogen, NO_x, with a Thermoelectron 10 AR chemiluminescent analyzer and sulfur dioxide with a Beckman IR analyzer.

Pellet Feed System

The coal-sorbent pellets are injected pneumatically into the dense bed using a vibrating feeder. Feed rates are controlled by changing the frequency of vibration. Feed rates are measured by feeding a known weight of pellets and measuring the time in which this known weight of pellets is utilized by the burner.

II. Test Procedures

CFBC Combustion and Emissions Tests

The combustion testing of the pellet fuels involves the following steps:

* The CO, CO_2, O_2, NO_x and SO_2 analyzers are calibrated at the beginning and at several times during a test burn.

* The CFBC combustor is filled with the proper amount of bed material (sand or limestone).

* The propane preheat system is fired the bed material and unit is brought up to about 1100-1200°F. This step takes several hours.

* Coal and limestone hoppers are filled with prepared standard coal and limestone sorbent, respectively.

* The coal feed is initiated and the CFBC unit is brought up to operating temperatures of around 1500°F on the standard coal. The operation of all sampling and control systems are checked.

* For tests with standard coal and pellet fuels, typical values of operating variables are as follows:

fluidization velocity 9 ft/sec
Ca/S ratio 1-4
Bed temperature ≈ 1450-1650°F

These parameters are kept constant with all the fuels, so that comparison of the combustion and emissions parameters can be made under identical conditions of operation.

* During the coal-sorbent pellet tests, the pellets are injected pneumatically into the bed. No additional limestone sorbent will be injected during initial tests. If SO_2 emissions are higher than EPA limits, further tests will be conducted with limestone injection.

* Six to ten test runs are planned to be made. Each test run is made after the combustor has reached steady state conditions. Combustor steady state conditions are usually achieved after 30-48 hours of operation. Where test fuel supplies are limited, the procedure adopted is to first bring the combustor to steady state operation on the standard coal or another Illinois coal, and then change the fuel feed to the test coal, or pellet fuel only for the duration of the steady state data acquisition period.

* The variables measured during a test include:

  - fuel and air mass flows
  - air superficial velocity
  - bed temperature
  - other temperatures at various combustor locations
  - combustion gas analysis comprised of CO, CO_2, O_2, NO_x and SO_2 emissions
  - test duration time
  - quantity of ash collected in cyclones during test period

Combustion generated ash and spent limestone from the pellets and standard coal tested are analyzed. The heat content of the elutriated unburnt carbon is determined from calorimetry tests. Spent limestone and ash are prepared on metal stubs and subjected to energy dispersive x-ray (EDX) analysis to determine the elements present in the samples.

Sample Analysis

(a) Proximate and Ultimate Analyses

Proximate and ultimate analyses of the raw coals used in making the pellets and the size graded reference Illinois No. 6 coal are obtained using standard ASTM procedures at the Coal Technology Laboratory at Carterville, Illinois.

(b) Particle Size Analysis

Particle size analysis in the range below 125 microns is measured utilizing a Leeds and Northrup Microtrak Model 7995-10 particle size analyzer. A schematic of the instrument is shown in Figure 2. In this version of the instrument, a laser beam is projected through a transparent cell which contains a stream of moving particles suspended in a liquid. Light rays which strike particles are scattered through angles which are inversely proportional to their sizes. The rotating optical filter transmits light at a number of predetermined angles and directs it to a photodetector. Electrical signals proportional to the transmitted light flux values are processed by a microcomputer system to form a multi-channel histogram of the particle size distribution.

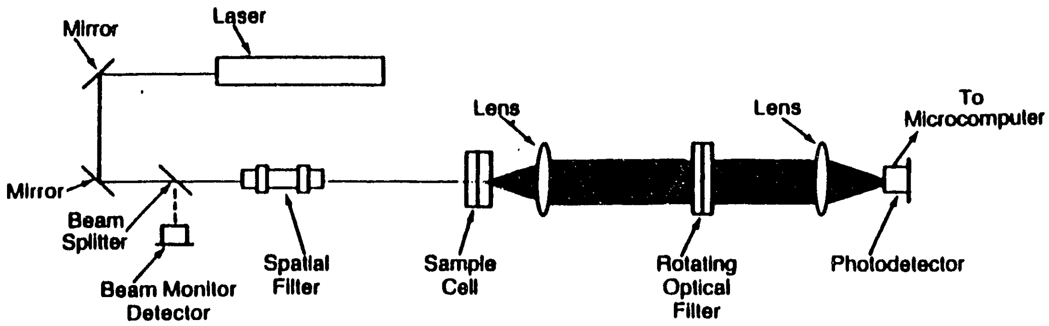

Figure 4. STANDARD RANGE ANALYZER – PARTICLE MEASURING SYSTEM

Figure 2. Schematic of Microtrak Particle Size Measurement System

(c) Mineral Matter Analysis

The mineral matter analysis of the coal in the pellet fuels and the reference Illinois No. 6 coal is conducted with a Hitachi H-600 analytical electron microscope operating both in the transmission and the scanning-transmission electron microscopy (STEM) modes. With STEM, a Tracor-Northern energy dispersive x-ray (EDX) Model 5500 analysis system was employed. The specimen samples were mounted on adhesive copper grids and examined at 100kV in the electron microscope. The samples were uncoated.

Data Analysis

From the measured data the following parameters will be computed:

- excess-air ratios
- Ca/S mole ratios
- carbon conversion efficiency
- sulfur capture efficiency %
- SO_2 emissions levels in lb/10^6 Btu
- carbon balances

RESULTS AND DISCUSSION

(a) Combustion Evaluation of Pellets

Pellet samples were received from ISGS personnel in late April, and combustion tests were begun forthwith. Table 1 gives the ultimate analyses of the coal used in the pellets (as furnished by ISGS) and that of the reference run-of-mine coal used in the present experiments. Both coals were obtained from the Kerr-McGee Galatia plant. The preparation plant waste coal used in the pellets contained 3.09% total sulfur on a DAF basis, while the run-of-mine coal had 1.92%. The mesh size of the run-of-mine coal is -14+20 Tyler mesh.

TABLE 1

Ultimate Analyses of Coal Used in Pellets
and the Reference Run-of-Mine Coal

	Pellet Coal	Reference Coal
Ultimate Analysis (wt.% DAF)		
Carbon	81.92	83.69
Hydrogen	3.98	6.07
Nitrogen	1.56	1.89
Sulfur	3.03	1.92
Oxygen	8.17	6.43
Heating Value, Btu/lb (Dry)	12,925	11,529

The combustion experiments were conducted as detailed in the section on Experimental Methods. Figure 3 shows the variation of measured SO_2 emissions with temperature. The pellet contained 5% calcium hydroxide which was equivalent to a 1.1:1 Ca/S ratio. As seen from the figure, the SO_2 emissions did increase with bed temperature beyond 1500°F. This variation is similar to that commonly observed with limestone as sorbent, and indicates that the sulfur capture reactions and mechanisms for both calcium hydroxide and calcium carbonate follow similar trends with temperature. These results are still considered preliminary, and further experimentation is planned to verify them.

As measured, the SO_2 emissions are quite high, and this is to be expected for the low Ca/S ratio of 1.1:1. Further experiments with these pellets and pellets containing 10% calcium hydroxide will be conducted in the next quarter.

Figure 4 shows a typical comparison of the SO_2 emissions from the CaOH-impregnated pellet and the reference coal. In the experiments with the run-of-mine coal, the limestone was fed

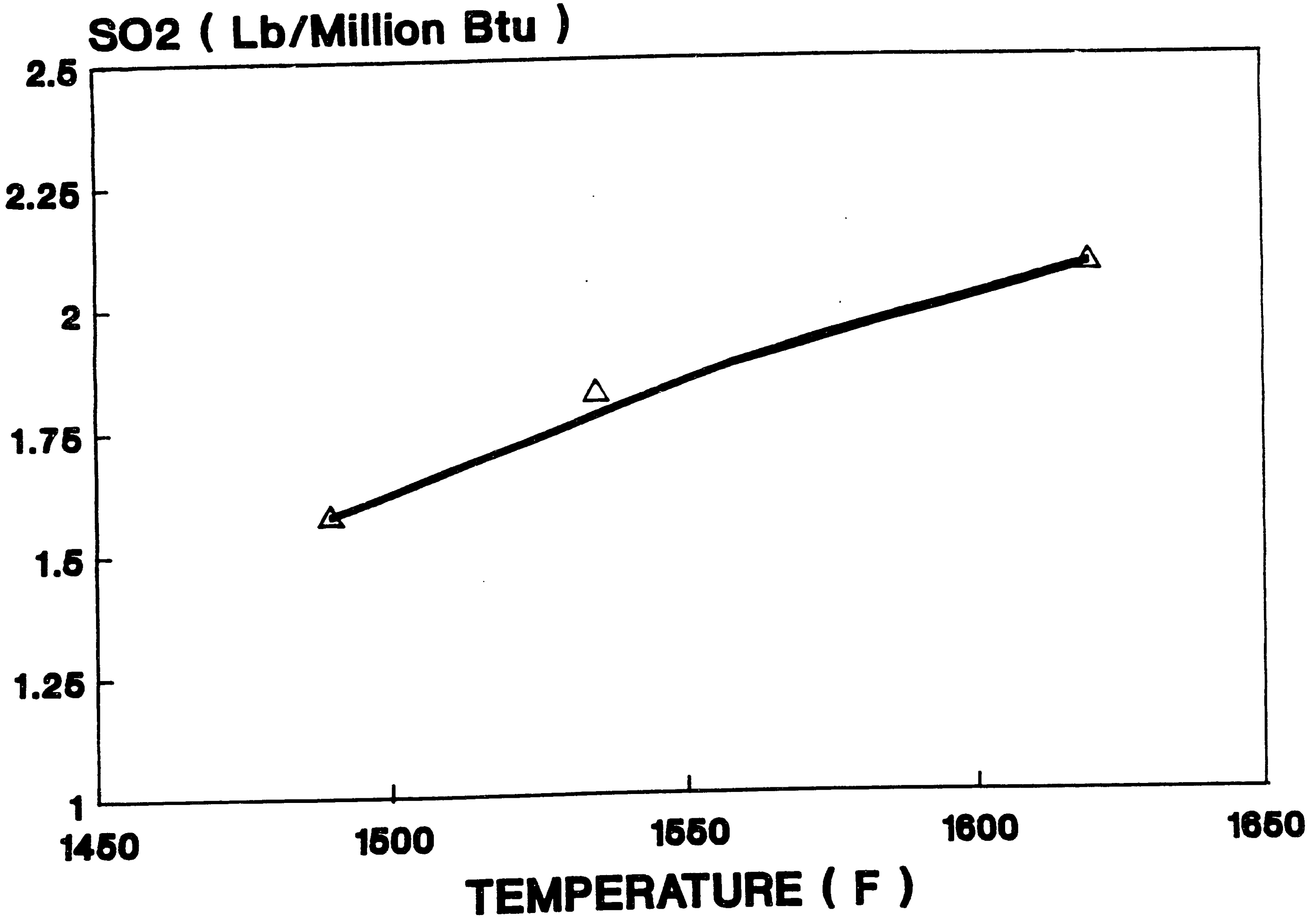

Figure 3. Influence of Bed Temperature on SO_2 Emissions, Measured with CaOH-Impregnated Pellets; Ca/S = 1.1:1

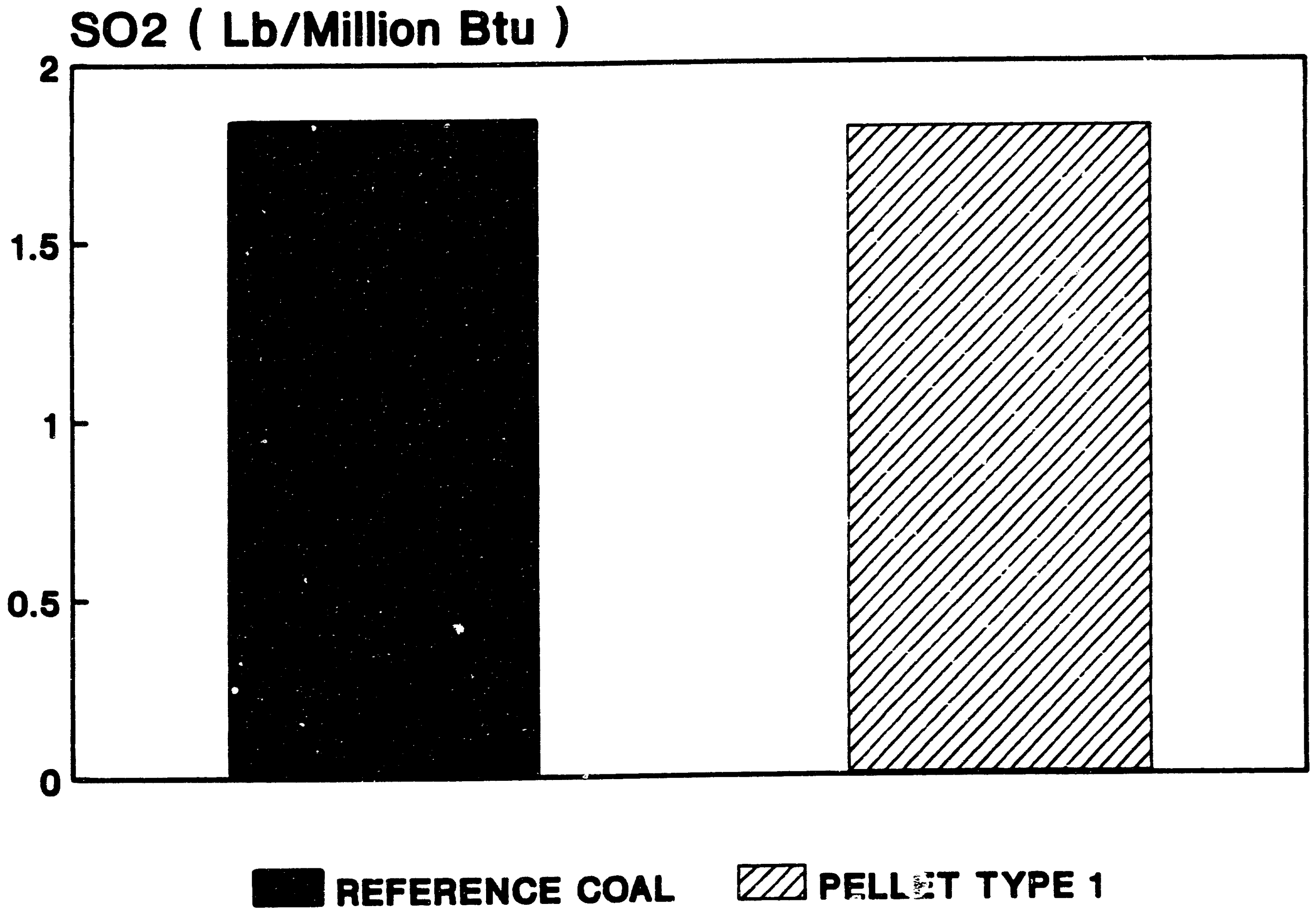

Figure 4. Bar Chart Illustrating Comparative SO_2 Emissions Levels from CaOH Containing Pellets and Run-of-Mine Coal

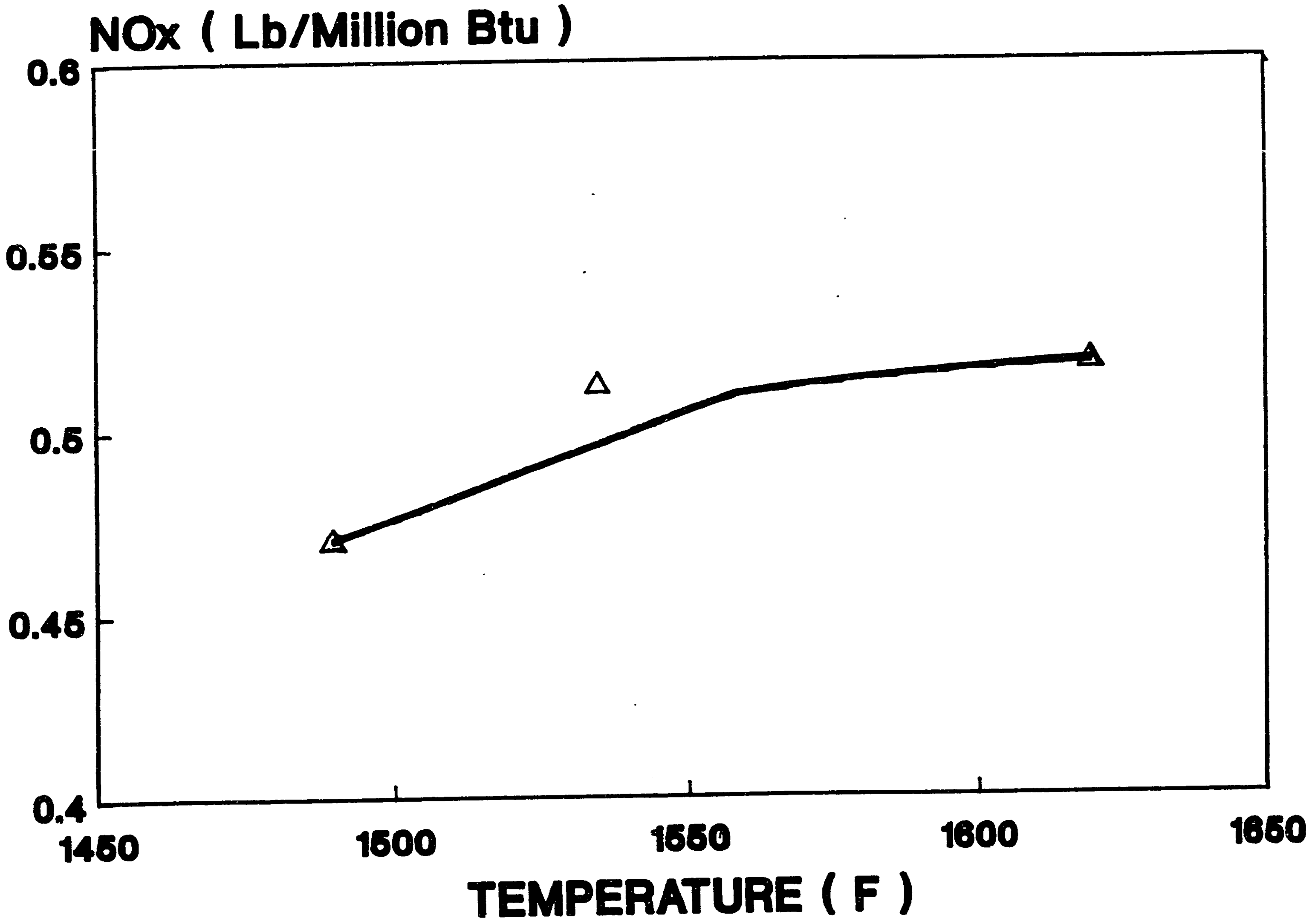

Figure 5. Influence of Bed Temperature on NO_x Emissions While Firing the 5% CaOH-Impregnated Pellets

separately. The Ca/S ratio used with the coal was 1.15:1. Figure 4 shows that for equivalent Ca/S ratios, the SO_2 emissions from the calcium hydroxide containing pellets and the run-of-mine coal using limestone as sorbent are quite similar. Again, additional exhaustive experiments are planned for the next quarter to verify this result.

Figure 5 shows the variation in NO_x emissions from the pellets as a function of bed temperature. NO_x levels from pellets are generally low compared to that from the coal, and varied from about 0.48 to 0.53 lbs/million Btu in these preliminary experiments. This is attributed to the lower temperatures surrounding the pellets.

(b) Hydrogen Chloride Analyzer Installation

The gas filter correlation Model 15 hydrogen chloride analyzer ordered from Thermo Electron Environmental Systems was received in mid-May. Upon receipt of the analyzer, further combustion testing was temporarily postponed. The rationale for this was that since one of the objectives of the present investigation was to measure the HCl emissions, and since the quantities of the pellet samples were limited, it would be necessary to bring the HCl analyzer on-line before the pellet samples were exhausted. This would enable HCl concentrations in the exhaust gases to be measured along with the emission levels of SO_2, NO_x, CO, CO_2, etc.

One of the requirements in the HCl analyzer installation is that it be located as close to the combustor sampling point as possible. For this reason, a separate platform is being attached to the combustor frame, such that the analyzer can be located within the 10 ft distance specified by the manufacturer. This installation, along with the heated sample line is proceeding on schedule and should be completed shortly. Combustion testing with the pellets will then be resumed.

(c) 12th International Conference on FBC Paper Presentation

Based on the work funded during the 1991-1992 fiscal year, a paper was presented at the 12th International Conference on Fluidized Bed Combustion co-sponsored by EPRI and the American Society of Mechanical Engineers (ASME) in San Diego. The conference dates were May 9-13, 1993. The title of the poster paper was "CFBC Combustion and Emissions Properties of Processed Preparation Plant Slurry Fuels," by S. Rajan, H. Bruyns and K. Ho. No travel funds from ICCI were requested for the conference. Travel was sponsored by the Department of Mechanical Engineering at Southern Illinois University.

CONCLUSIONS

Combustion characterization of calcium hydroxide and limestone impregnated pellets has been initiated, and preliminary results on SO_2 and NO_x levels from a pellet sample containing 5% calcium hydroxide has been presented. The gas filter correlation hydrogen chloride analyzer has been received in mid-May and is being installed. Combustion and emissions data, along with HCl emissions levels, will be available in the next quarter.

PROJECT MANAGEMENT REPORT
March 1 through May 31, 1993

Project Title:	COMBUSTION AND EMISSIONS CHARACTERIZATION OF PELLETIZED COAL FUELS
Principal Investigator:	Dr. S. Rajan Mechanical Engineering and Energy Processes Southern Illinois University Carbondale, IL 62901
Project Manager:	Dr. Frank Honea Illinois Clean Coal Institute

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