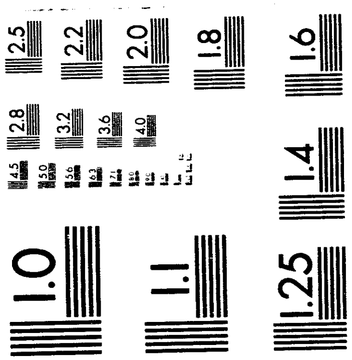
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1 of 1

DOE/PC/92521--T72

FINAL TECHNICAL REPORT
September 1, 1992 through August 31, 1993

Project Title: **COMBUSTION AND EMISSIONS CHARACTERIZATION OF PELLETIZED COAL FUELS** DE-FC22-92PC92521

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ABSTRACT

The feasibility of converting waste preparation plant coal streams into marketable fuels with good combustion and emissions properties has been investigated in this project. Coal pellets containing both hydrated lime and limestone as sorbents were made from a flotation column waste feedstock under a related Illinois Clean Coal Institute project. These pellets, which contain sorbent with Ca/S ratio varying from 0.8 to 2.4 were successfully burnt in a 4-inch internal diameter circulating fluidized bed combustor. Emissions levels of sulfur dioxide, oxides of nitrogen and hydrogen chloride were measured as a function of bed temperature. Mineral matter analyses of the combustion generated ash was made, and combustion efficiencies were studied. The pellet coals combustion performance was compared to that of an Illinois No. 6 run-of-mine coal from the same preparation plant.

Results show that the coal pellets, produced from the flotation column waste stream, are excellent fluidized bed combustor fuels. Calcium hydroxide impregnated pellets yielded lower sulfur dioxide emissions than limestone sorbent pellets for the same Ca/S ratio. Increase in Ca/S ratio with the hydrated lime sorbent produced a more rapid decline in sulfur dioxide emissions than with the limestone sorbent. Oxides of nitrogen emissions were generally on the order of 0.3 to 0.4 lbs/10^6 Btu. Hydrogen chloride emissions were found to be influenced more by Ca/S ratios than by bed temperature, and varied in the range of 0.0075 to 0.055 lbs/10^6 Btu in the present tests. Combustion efficiencies of the pellet fuels were about 98% without secondary cyclones recycle. EDX analysis of the mineral matter in the raw coal and the combustion-generated ash samples sheds light on the mineral transformations during combustion.

DISCLAIMER

EXECUTIVE SUMMARY

Extensive combustion experiments were performed in a 4-inch internal diameter circulating fluidized bed combustor (CFBC) on six coal-sorbent pellet fuels produced from a flotation column waste stream of the Kerr-McGee Galatia preparation plant. These pellet fuels were produced under subcontract by the Illinois State Geological Survey. These fuels are:

* Two calcium hydroxide impregnated pellets, hardened by air carbonation, with Ca/S ratios 0.9:1 and 1.85:1, respectively

* A coal-sorbent pellet containing a coarse limestone (75%<30 mesh) with Ca/S ratio 1.6:1 and cornstarch as binder

* Three coal-sorbent pellets containing a fine limestone (80%<200 mesh) with Ca/S ratios of 0.8:1, 1.6:1 and 2.4:1, respectively, and cornstarch as binder

The experiments were designed to study (1) the comparative effectiveness of hydrated lime and limestone as sorbents, (2) the influence of Ca/S ratio and (3) the effect of limestone particle size on sorbent effectiveness.

An Illinois No. 6 run-of-mine coal, also used as feedstock to the above preparation plant, was burnt in the CFBC for comparison purposes. Fluidization velocity was kept constant during the tests. Bed temperature and Ca/S ratios were variables. The carbon monoxide, carbon dioxide, oxygen, sulfur dioxide, oxides of nitrogen, and hydrogen chloride emissions were measured. Combustion efficiencies and carbon balances were calculated. Mineral matter transformations were studied using energy dispersive x-ray analysis. The test results are summarized below.

Sulfur Dioxide Emissions

Experimental results showed that at a Ca/S ratio of 0.85, the hydrated lime and the limestone yield roughly the same sulfur dioxide emissions with the hydrated lime holding a slight advantage. However, at a higher Ca/S ratio of 1.6, the sulfur capture effectiveness of the hydrated lime was superior to that of the limestone. Data from the tests indicate that, with this type of pellet fuel, a Ca/S ratio of 1.2-1.4 would result in SO_2 emissions of 1.0 lbs/10^6 Btu with the hydrated lime. The corresponding Ca/S ratio with the limestone sorbent was about 1.8. Sulfur dioxide levels from the mined coal selected for the present tests were much higher than those from the pellet fuels.

Oxides of Nitrogen Emissions

Measured oxides of nitrogen levels from the pellets ranged from 0.3 to 0.4 lb/10^6 Btu. These emissions were not strongly influenced by bed temperature. Increasing the Ca/S ratio resulted in slightly lower NO_x emissions. Limestone size in the pellets also influenced the NO_x emissions.

Hydrogen Chloride Emissions

Hydrogen chloride (HCl) concentrations in the combustion gases ranged from 0.0075 to 0.55 lbs/10^6 Btu. Generally, hydrogen chloride levels were found to decrease slightly at the higher bed temperatures of about 1600°F. Increasing the Ca/S ratio produced changes in HCl emissions that were different for the two types of sorbents investigated. Limestone particle size also influenced the HCl emissions.

Combustion Efficiency

High combustion efficiencies of 98% or more were realized with both limestone and hydrated lime sorbents in the present tests. Because the mined coal used for comparison had a high ash content, its combustion efficiency was 2-3% lower. Combustion efficiency was found to be insensitive to sorbent type or Ca/S ratio. Larger limestone particle sizes seemed to produce lower values of combustion efficiency.

Mineral Matter Analyses

The EDX mineral matter analyses showed reductions in the alkali metals in the combustion generated ash compared to that in the raw coal. Chlorine and sulfur concentrations were also reduced.

Summary

The pellet fuels, whose combustion and emission properties were studied in the present project are shown to be excellent fuels for fluidized bed combustors. They represent a means of making marketable products from waste coal streams and offer definite economic and environmental advantages.

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 secure pellets made by ISGS using froth flotation fines by mixing these fines with cornstarch as binder and containing limestone fines as sorbent.
3. to burn these pellets in a laboratory scale 4-inch diameter circulating fluidized bed combustor (CFBC).
4. 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.
5. 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.
6. 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 that 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-inch 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 that 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.

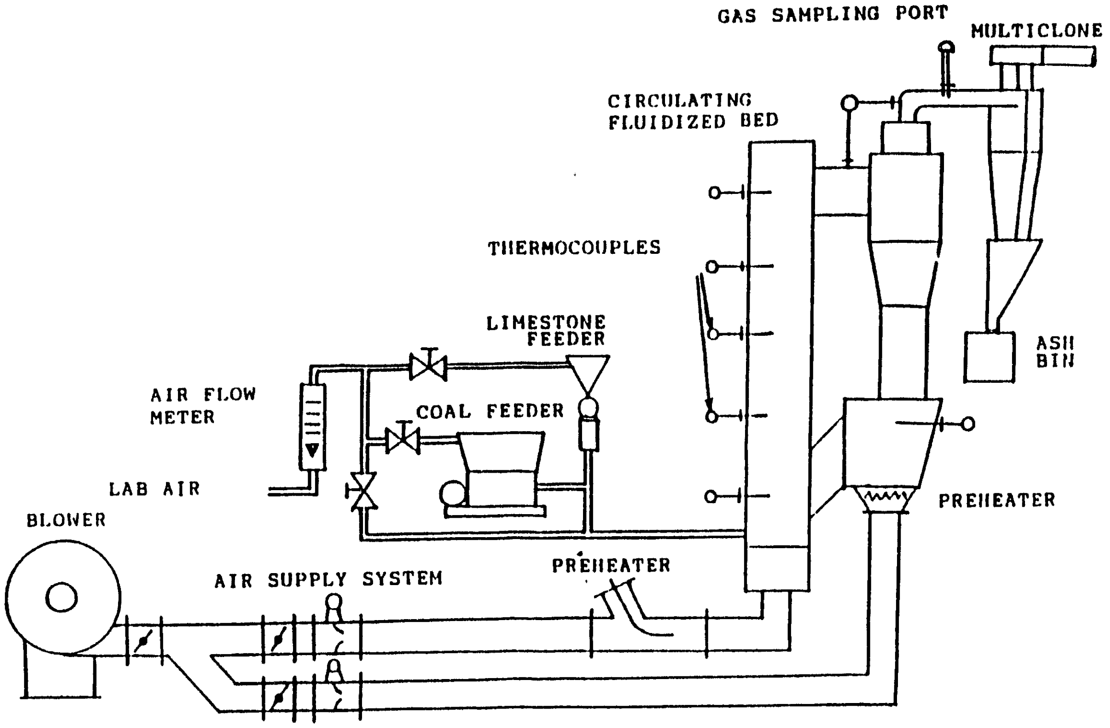


Figure 1. Schematic of 4-Inch Internal Diameter Circulating Fluidized Bed Combustor

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 re-injected into the bed. The multiclone solids are later analyzed for heat content, using an adiabatic calorimeter. Combustion gases are drawn off 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 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 $\approx$ 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 is injected during these tests.
- Six to ten test runs are made. Where pellet fuel quantities permitted, the test was run twice. 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, HCl 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 that contains a stream of moving particles suspended in a liquid. Light rays that strike particles are scattered through angles that 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.

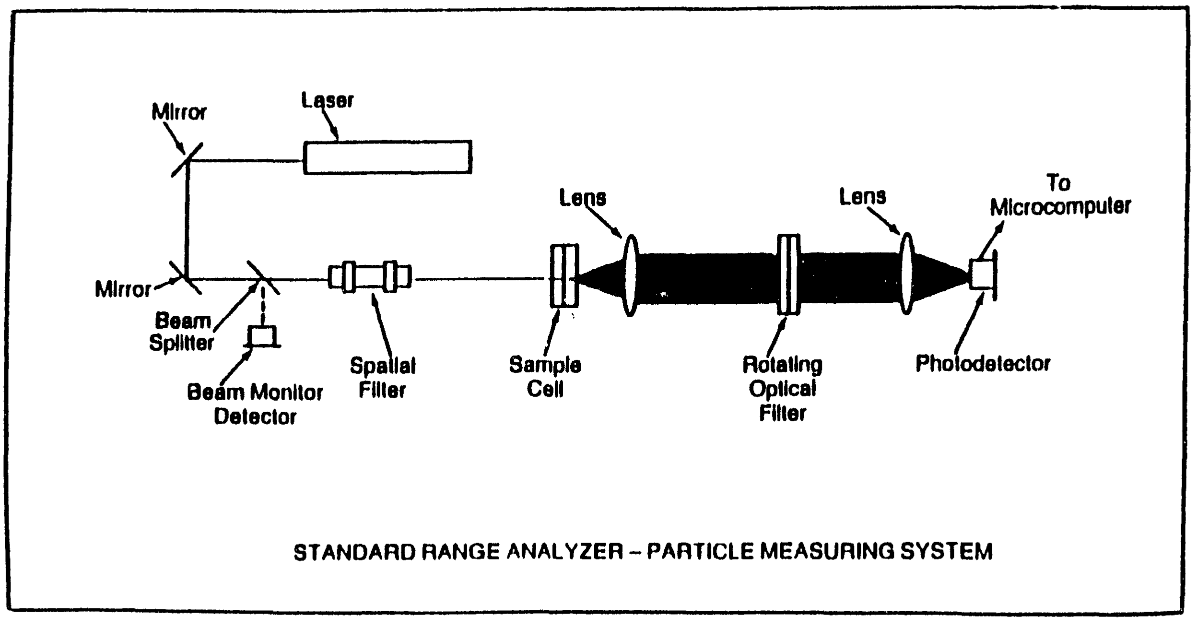


Figure 2. Schematic of Microtrak Particle Size Measurement System

(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 100 kV 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

Chemical and Particle Size Analysis of Fuels

Table 1 shows the chemical analysis of the flotation column fines used as the pellet feedstock and the Illinois No. 6 run-of-mine coal. The run-of-mine coal is that used as the feed for the preparation plant. The flotation column fines is the mixture of the spirals fractions and the centrifuge product. The moisture content of the column fines as used in the pellet is 5.40%, while that of the run-of-mine coal is 6.4%. The run-of-mine coal had a high ash content of 38.54% and a correspondingly low heating value of 9489 Btu/lb. It also had slightly lower sulfur and hydrogen contents compared to the flotation fuel.

The run-of-mine coal was ground and sieved to a mesh size of (-14+18) mesh. The particle size analysis of the pellet feedstock is similar to that used in the previous year's research and is shown in Figure 3.

Chemical and Particle Size Analysis Sorbents

The sorbents used were obtained from the Mississippi Lime Company, Alton, Illinois. The chemical and particle size analysis of the limestone as provided by the company is given in Table 2 while that of the hydrated lime produced from the limestone is given in Table 3.

Processed Fuels Investigated

Pellet fuels processed from the above feedstocks and supplied to the present project under a subcontractual arrangement by Illinois State Geological Survey researcher David Rapp included the following:

TABLE 1

CHEMICAL ANALYSIS OF COAL FEEDSTOCKS
DRY BASIS %

	PELLET FEEDSTOCK	RUN-OF-MINE COAL
Ash	10.8	38.54
Sulfur	2.62	1.98
Hydrogen	4.56	2.70
Carbon	73.52	48.79
Nitrogen	1.42	1.27
Oxygen	7.10	7.48
Btu/lb	12,967	9,489

```
RELATIVE VOL. GRAPH            CUMMULATIVE GRAPH
 106  *3                       125  **********100
  75  ****7                     88  *********96
  53  *******12                 62  ********89
  38  **********16              44  *******76
  27  ********14                31  *****59
  19  ********13                22  ****44
  13  ******10                  16  ***31
 9.4  ****8                     11  **20
 6.6  **4                      7.8  *12
 4.7  **3                      5.5  7
 3.3  *2                       3.9  4
 2.4  0                        2.8  1
 1.6  0                        1.9  0
 1.2  0                        1.4  0
 0.8  0                        0.9  0
```

FIGURE 3. Particle Size Distribution and Cumulative Volume Graph for Pellet Coal Feedstock

TABLE 2

CHEMICAL AND PARTICLE SIZE ANALYSES OF LIMESTONE

Chemical Analysis

$CaCO_3$	97.80%	to	98.90%
$CaSO_4$	0.06	to	0.12
SiO_2	0.35	to	0.90
Al_2O_3	0.11	to	0.34
Fe_2O_3	0.04	to	0.10
$MgCO_3$	0.40	to	0.95
H_2O	0.02	to	0.08
P_2O_5	0.006	to	0.009
MnO	0.0010	to	0.0024

Physical Analysis

Passing 16 Mesh (1,180 microns)		100.0%	
Passing 20 Mesh (850 microns)	91.5%	to	96.0%
Passing 30 Mesh (500 microns)	70.0	to	81.0
Passing 60 Mesh (250 microns)	23.0	to	30.0
Passing 80 Mesh (180 microns)	13.5	to	20.0
Passing 100 Mesh (150 microns)	8.5	to	13.5
Passing 200 Mesh (75 microns)	3.0	to	8.5
Passing 325 Mesh (45 microns)	2.0	to	7.0
Particle Size Range (microns)	10	to	1,100

Source: Mississippi Lime Company, Alton, Illinois

TABLE 3

CHEMICAL AND PARTICLE SIZE ANALYSIS OF HYDRATED LIME

Chemical Analysis

$Ca(OH)_2$	96.0%	to	97.2%
CaO - Equivalent	72.6	to	73.6
CaO Total	73.6	to	74.3
$CaCO_3$	0.65	to	1.75
$CaSO_4$	0.05	to	0.10
S - Equivalent	0.012	to	0.024
SiO_2	0.38	to	0.65
Al_2O_3	0.20	to	0.30
Fe_2O_3	0.07	to	0.10
MgO	0.40	to	0.55
Free H_2O	0.04	to	1.0
P_2O_5	0.008	to	0.012
MnO	0.0015	to	0.0025

Physical Analysis

Minus 100 Mesh	100.0%
Minus 200 Mesh	98.5%
Minus 325 Mesh	92.0%

Source: Mississippi Lime Company, Alton, Illinois

- Two calcium hydroxide impregnated pellets, hardened by air carbonation, with Ca/S ratios 0.9:1 and 1.85:1, respectively

- A coal-sorbent pellet containing a coarse limestone (75%<30 mesh) with Ca/S ratio 1.6:1 and cornstarch as binder

- Three coal-sorbent pellets containing a fine limestone (80%<200 mesh) with Ca/S ratios of 0.8:1, 1.6:1 and 2.4:1, respectively, and cornstarch as binder

- A reference run-of-mine coal from Kerr-McGee Galatia mine

The reference run-of-mine coal was obtained directly from Kerr-McGee Corporation. The pellets were 0.125 inches in diameter and 0.375 inches long, as used in the present combustion tests. This size was selected as being suitable for efficient combustion in the 4-inch diameter CFBC unit. However, the size could be varied to match other equipment needs. As received, the pellets were quite moist and fragmented easily. They were air dried until the moisture content was less than 5%. Where cornstarch was used as binder in the above pellets, they contained 5% by weight of cornstarch.

Combustion Test Results

Test procedures and operating conditions for the combustion tests performed on the pellets and run-of-mine coal have been described above. In this section, the results of these tests are discussed under three headings:

1. SO_2, NO_x and HCl Emissions
2. Combustion Efficiency, and
3. Ash Mineral Analysis

1. SO_2, NO_x and HCl Emissions

1(a) Calcium Hydroxide Sorbent Pellets: Effect of Ca/S Ratio

Two pellets with Ca/S ratios of 0.90 and 1.85, respectively, were burnt in the CFBC unit. Fluidization velocity and pellet feed rates were kept constant. Bed temperature control was obtained by an in-bed heat exchanger. Figure 4 shows the sulfur dioxide emission levels using the calcium hydroxide sorbent in the pellets. For a Ca/S ratio of 1.85, very good SO_2 control is obtained for a feedstock sulfur content of 1.84%, the actual SO_2 emissions levels being

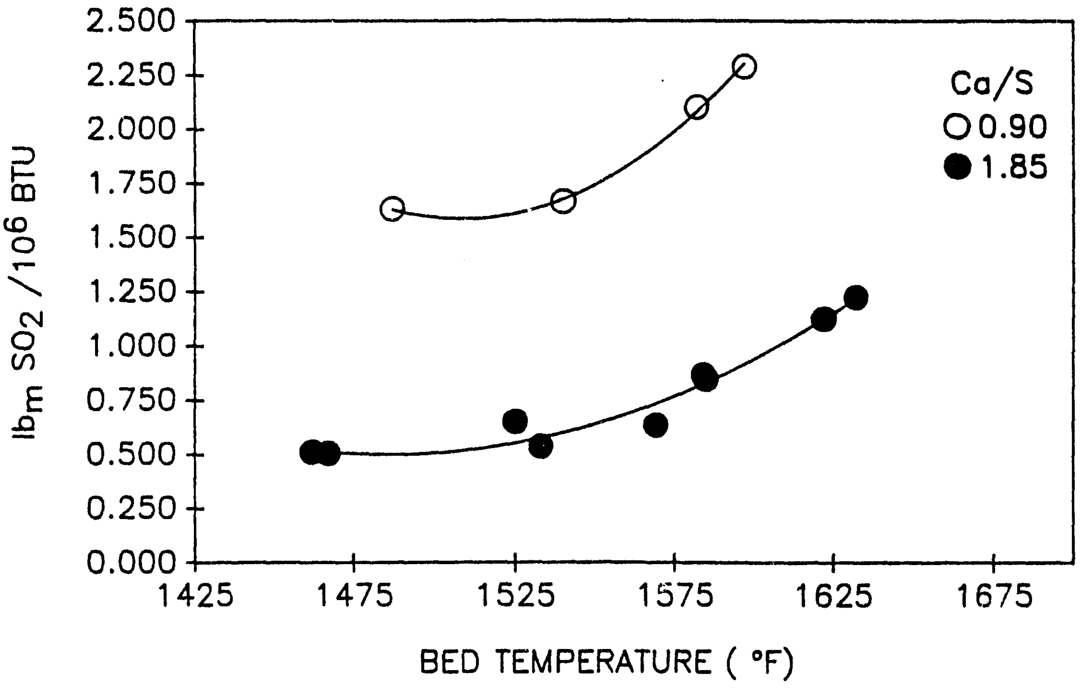


FIGURE 4. Effect of Bed Temperature and Ca/S Ratio on SO_2 Emissions with Calcium Hydroxide Sorbent

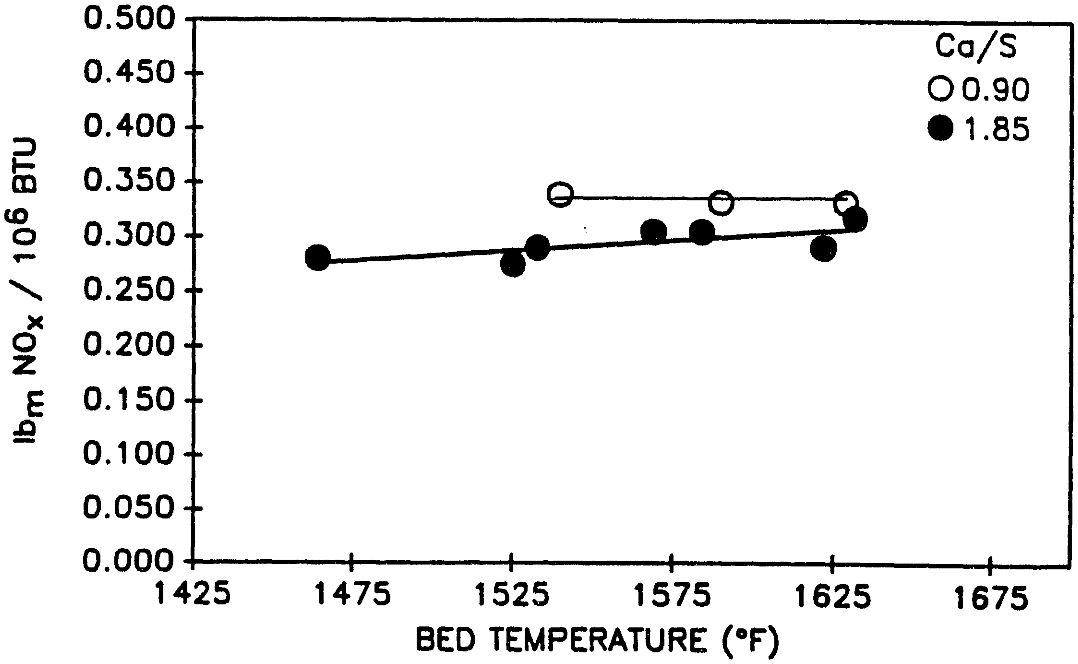


FIGURE 5. Effect of Temperature and Ca/S Ratio on NO_x Emissions, Calcium Hydroxide Sorbent

about 0.51 lbs/10^6 Btu at a bed temperature of 1510°F. The data also shows that roughly doubling the Ca/S ratio from 0.9 to 1.85 reduces the SO_2 emissions by more than 50%. Thus hydrated lime is a very effective sorbent when impregnated into the pellet matrix.

The effect of bed temperature and Ca/S ratio on the NO_x emissions from the calcium hydroxide sorbent pellets is shown in Figure 5. Increasing the Ca/S ratio resulted in a slight reduction in NO_x emissions. This is most likely the consequence of the endothermic calcination reactions occurring in the pellet. Air carbonation converts some of the calcium hydroxide to the carbonate. As Ca/S ratios are increased, endothermic calcination reactions yield lower pellet temperatures during the fluidized bed combustion process, which lead to lower levels of NO_x emissions. Generally, NO_x emissions increased slightly with bed temperature in the range of 1475-1625°F. However, bed temperature increases do not significantly increase NO_x emissions as seen in Figure 5. NO_x emissions with a feedstock nitrogen content of 1.74% were measured to be on the order of 3.0 to 3.5 lbs/10^6 Btu under the present test conditions.

Hydrogen chloride (HCl) concentrations were measured in the exhaust gases using a gas filter correlation HCl analyzer manufactured by Thermoelectron Corporation. Generally, these concentrations were on the order of 10-40 ppm. The lbs/10^6 Btu emissions of HCl measured during the combustion of the calcium hydroxide containing pellets are shown in Figure 6. During these tests with the hydrated lime containing pellets, it was found that the HCl emissions increased with Ca/S ratio. Nevertheless, the emissions were quite small, being on the order of 0.01 to 0.04 lbs/10^6 Btu.

1(b) Limestone Sorbent Pellets: Effect of Ca/S Ratio

The effect of Ca/S ratio on the SO_2 and HCl emissions was studied with pellets containing a fine limestone (80%<200 mesh). Results are presented in Figures 7 and 8. In general, SO_2 emissions increased with bed temperatures above 1525°F. As with the combustion of ordinary coal, increasing the Ca/S ratio of limestone sorbent decreases the SO_2 emissions from pelletized coal combustion. At 1525°F, increasing the Ca/S ratio from 0.8 to 2.4 resulted in a decrease of SO_2 emissions from 1.6 to roughly 1.0 lbs/10^6 Btu, a reduction of 37.5% compared to the Ca/S = 0.8 value.

The effect of Ca/S ratio on the NO_x emissions was similar to that shown in Figure 5 for the calcium hydroxide containing

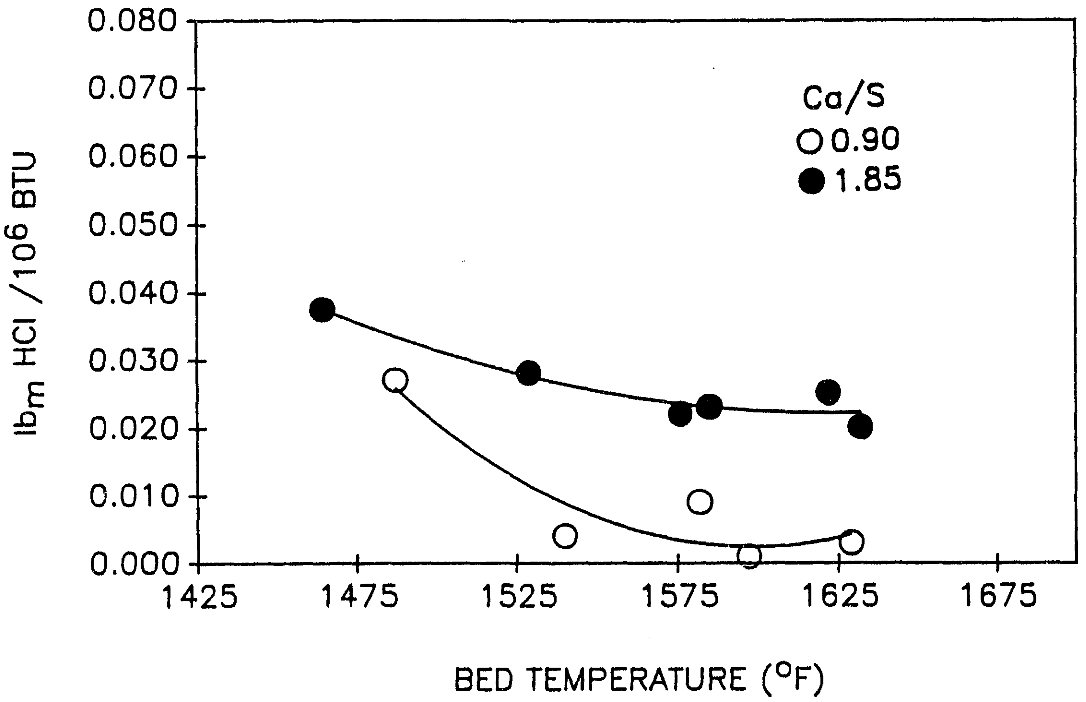


FIGURE 6. Effect of Temperature and Ca/S Ratio on HCl Emissions; Calcium Hydroxide Sorbent

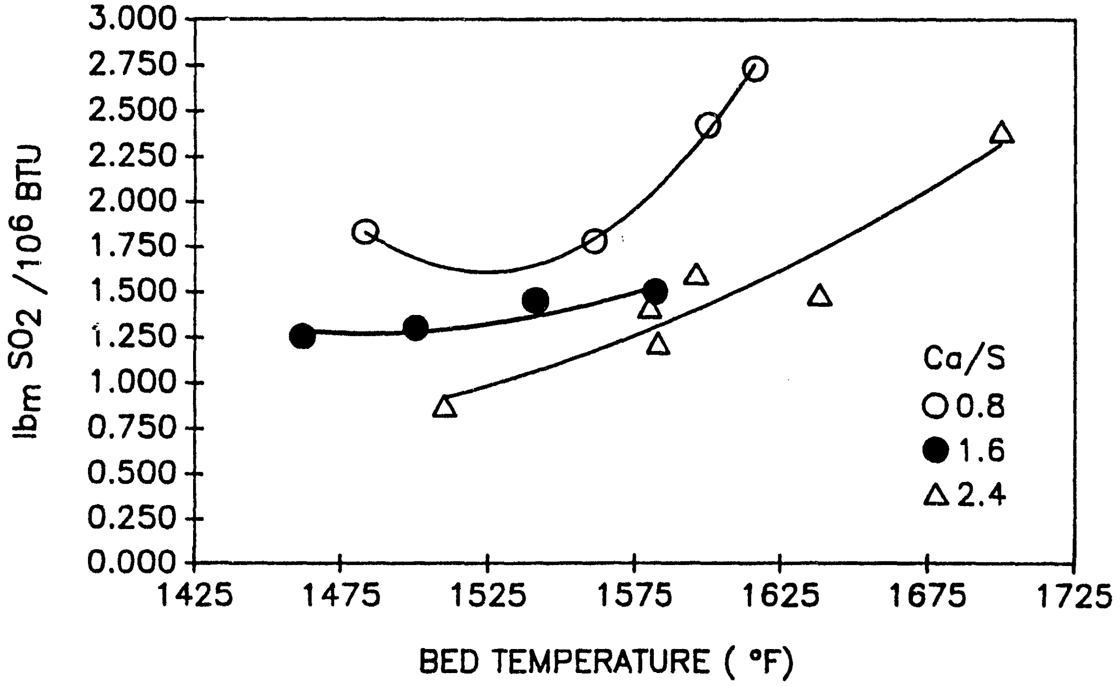


FIGURE 7. Effect of Temperature and Ca/S Ratio on SO_2 Emissions; Fine Calcium Carbonate Sorbent

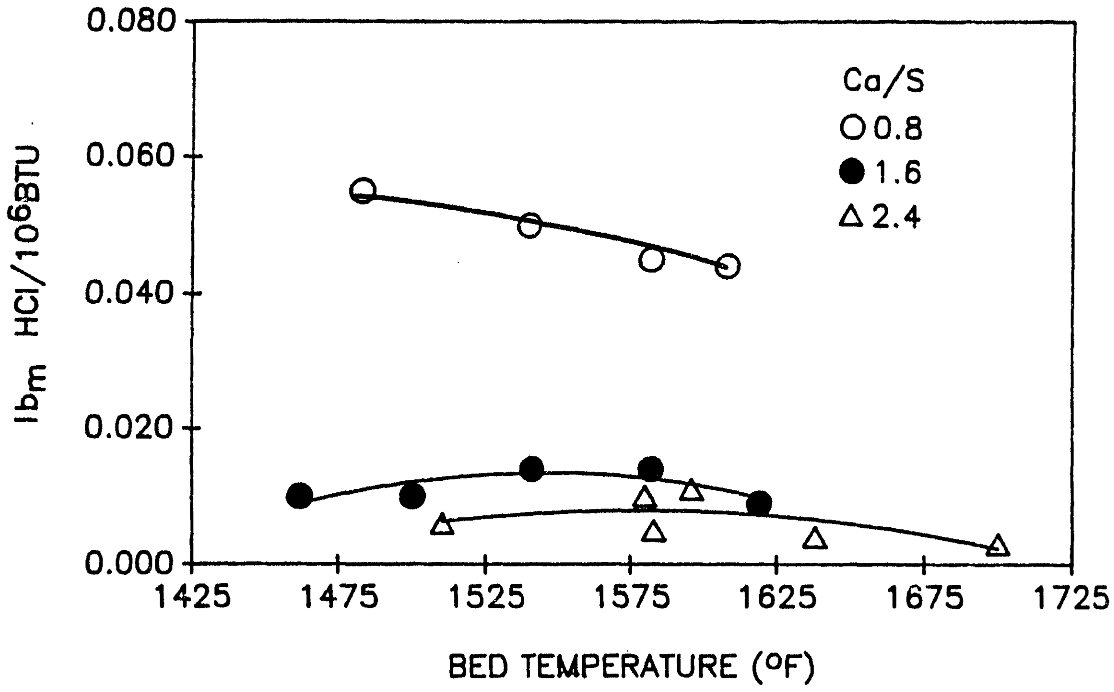


FIGURE 8. Effect of Temperature and Ca/S Ratio on HCl Emissions; Fine Calcium Carbonate Sorbent

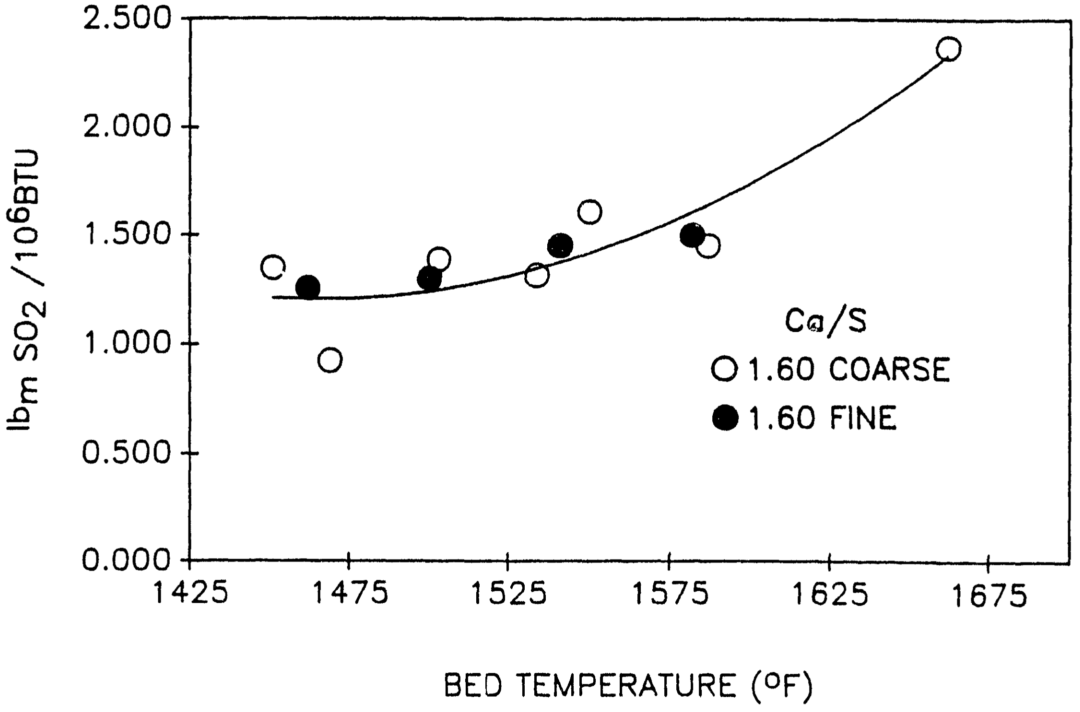


FIGURE 9. Influence of Limestone Sorbent Size on SO_2 Emissions

pellets. The actual NO_x values were slightly higher, being on the order of 0.33 to 0.4 lbs/10^6 Btu.

Figure 8 indicates that with the limestone sorbent, HCl emissions levels decreased as Ca/S ratio was increased, the rate of decrease being higher at the lower Ca/S ratios. During the present tests, HCl emissions of 0.0075 to 0.0575 lbs/10^6 Btu were measured as the Ca/S ratio was decreased from 2.4:1 to 0.8:1. At bed temperatures beyond 1550°F, there was also a slight decrease in HCl emissions with further increase in bed temperature.

1(c) Limestone Sorbent Pellets: Effect of Sorbent Size

The effect of sorbent size on emissions levels of SO_2, NO_x and HCl were studied by burning pellets containing two sizes of limestone both obtained from the Mississippi Lime Company. The fine limestone was in the size range of 80%<200 mesh on a mass basis and the coarse limestone was in the size range of 75%<30 mesh. Figures 9 and 10 show that, in the size range investigated, the limestone size had no discernible effect on either the sulfur dioxide or oxides of nitrogen emissions. The Ca/S ratio for the tests was 1.6:1, and the bed temperature varied in the range 1475 to 1675°F. It was found, however, that a finer limestone size was more conducive to lower HCl emissions as depicted in Figure 11. This trend is in keeping with that observed in the previous tests, described under section 1(b) above where a higher Ca/S ratio produced a reduction of HCl emissions. This data indicates that the more intimately the limestone is mixed with the coal pellet, and the larger the quantity of limestone, the lower the levels of HCl emissions, possibly due to chemical reactions occurring between the HCl gas and the calcined limestone.

1(d) Comparison of Calcium Hydroxide and Limestone as Pellets Sorbent

Figure 12(a) and (b) compare the sulfur dioxide emissions using the two sorbents. The Ca/S ratios in Figure 12(a) are 0.85 and 0.9 for the limestone and the hydrated lime, respectively. For Figure 12(b), the corresponding ratios are 1.6 and 1.85, respectively. In either case, the hydrated lime yields lower levels of sulfur dioxide emissions the difference being greater for the higher Ca/S ratios as expected.

The effect of sorbent type on NO_x emissions is not so pronounced as seen from Figure 13, although the calcium hydroxide sorbent yields slightly lower NO_x emissions.

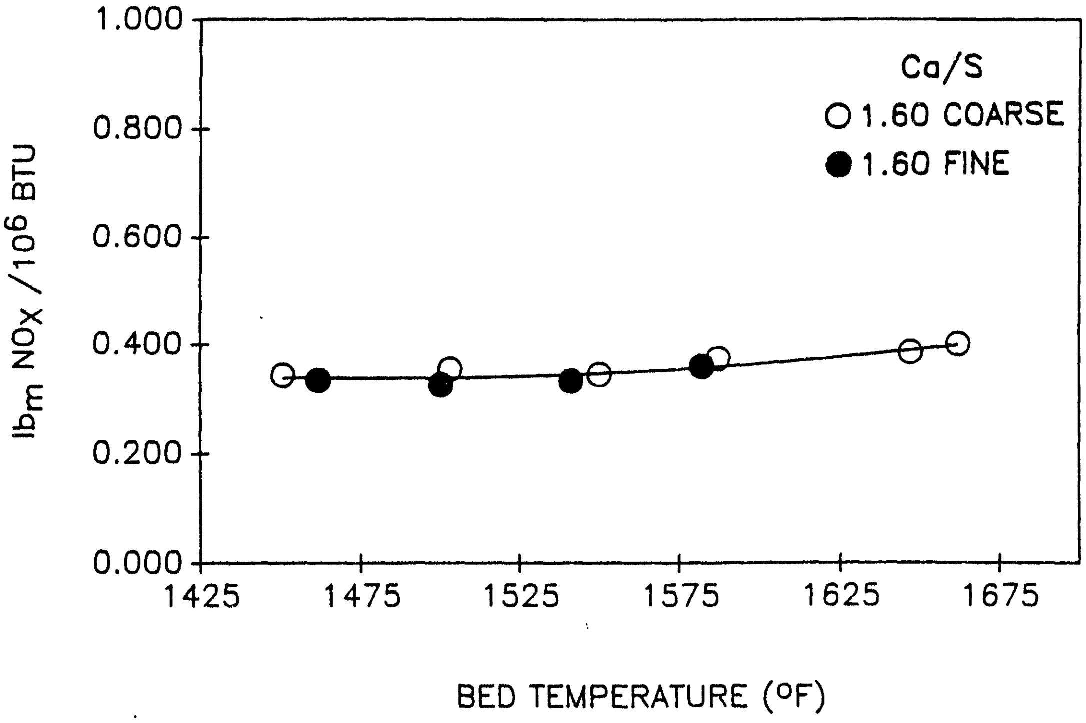


FIGURE 10. Influence of Limestone Sorbent Size on NO_x Emissions

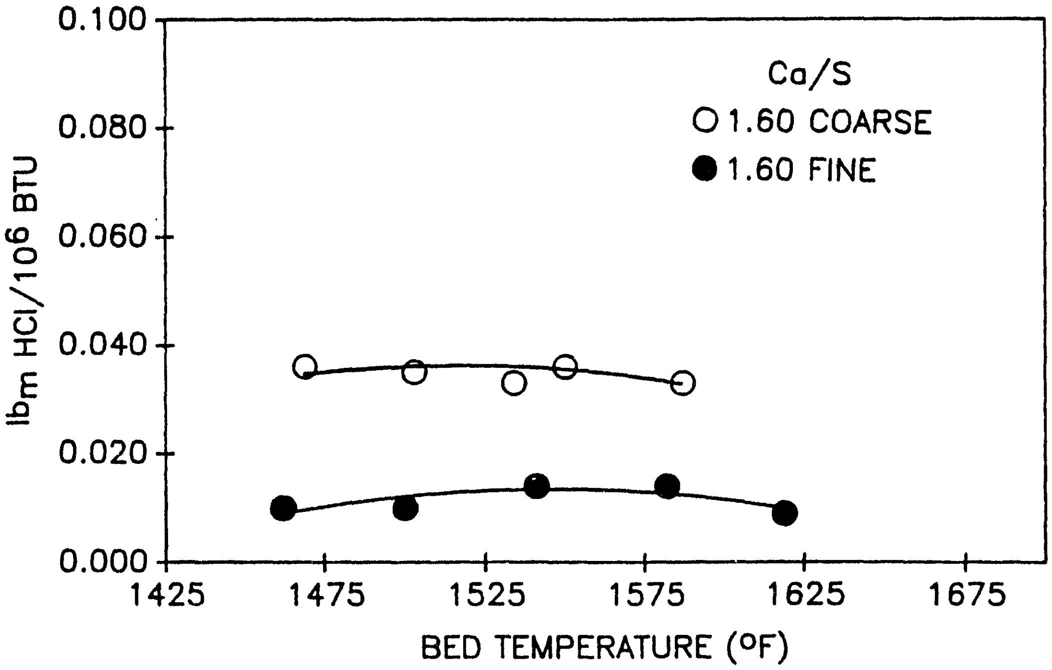


FIGURE 11. Influence of Limestone Sorbent Size on HCl Emissions

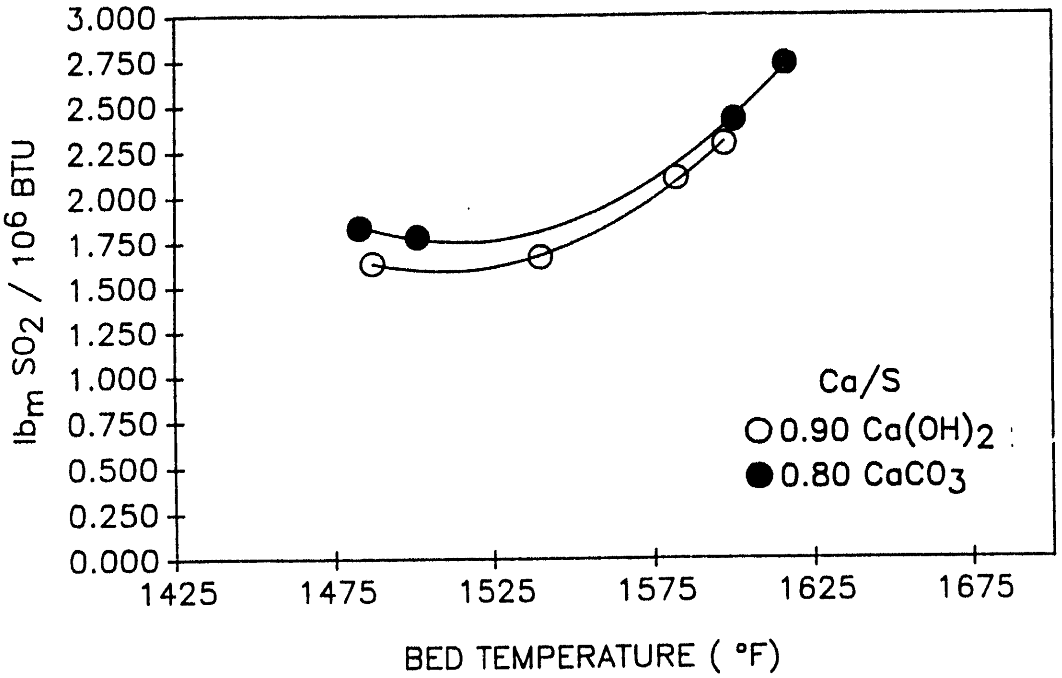


FIGURE 12(a). Comparative Performance of Calcium Hydroxide and Limestone as Sorbents; Ca/S Ratio ≈ 0.9

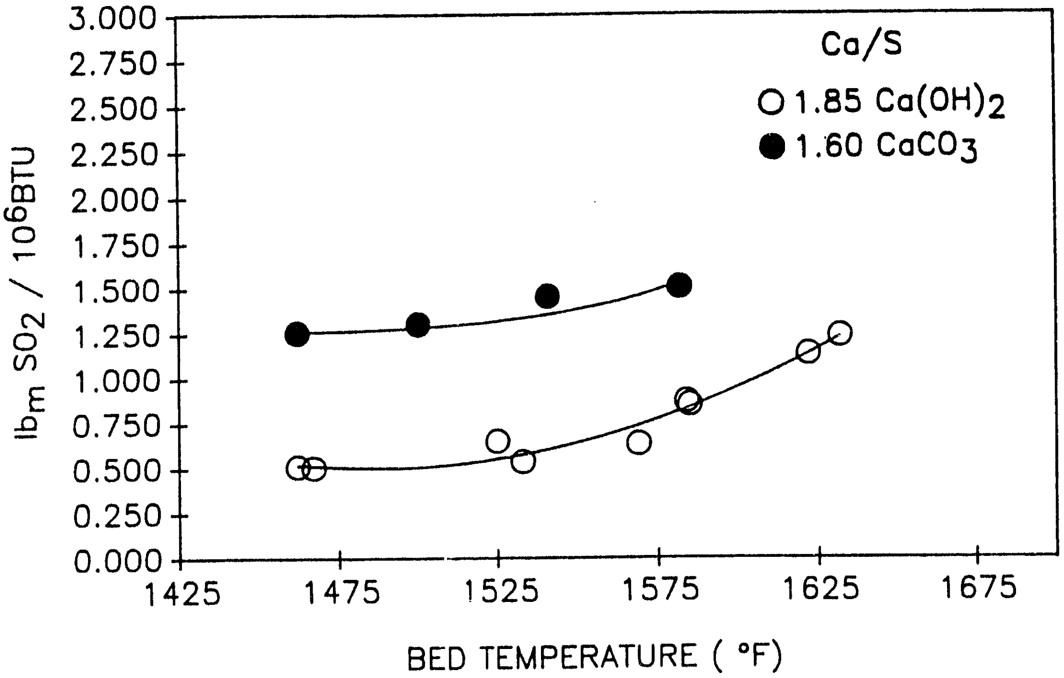


FIGURE 12(b). Comparative Performance of Calcium Hydroxide and Limestone as Sorbents at Higher Ca/S Ratios

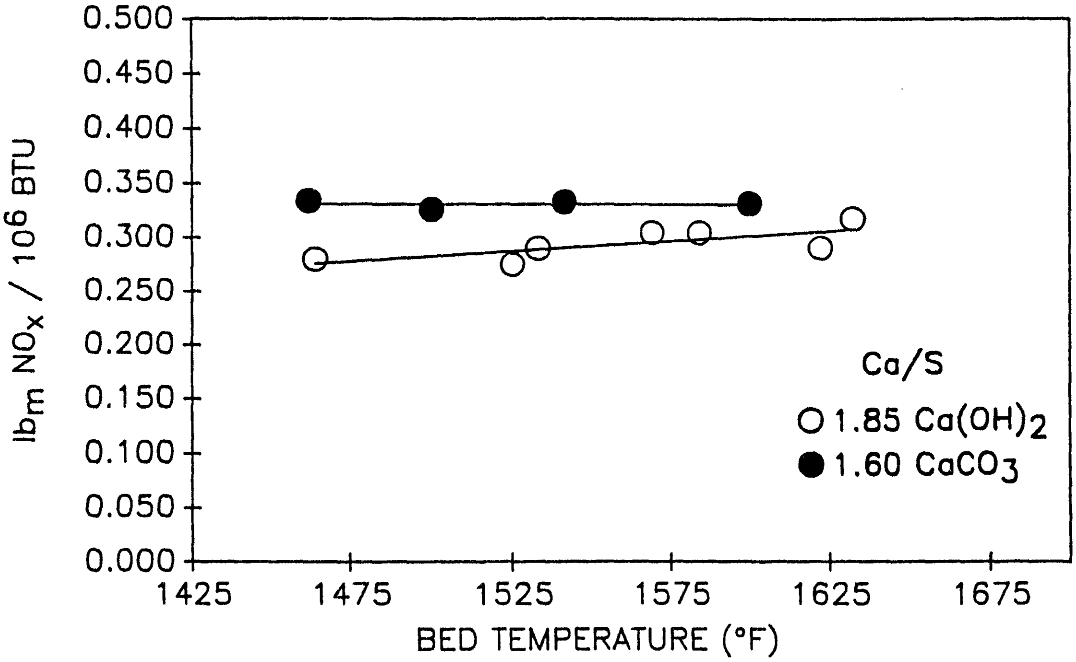


FIGURE 13. Influence of Sorbent Type on NO_x Emissions

The influence of sorbent type on HCl emissions is shown in Figures 14(a) and (b) for the Ca/S ratios mentioned above. In this study, it was found that at low Ca/S ratios, pellets containing limestone sorbent emitted higher HCl levels, ranging from 0.0575 to 0.042 lbs/10^6 Btu, compared to the corresponding values of 0.025 to 0.005 lbs/10^6 Btu for the hydrated lime in the bed temperature range of 1480 to 1625°F. However, with higher Ca/S ratios of 1.6 for the limestone and 1.85 for the hydroxide, the use of limestone as sorbent produced lower HCl emissions, Figure 14(b). Thus, the trends in HCl release and reaction within the combustor seem to be opposite to each other for the two sorbents evaluated. Further work is necessary to confirm these trends and to investigate the mechanisms involved.

1(e) Comparison of Pellet and Run-of-Mine Coal Emissions

During the combustion of the run-of-mine coal in the CFBC unit, bed ash material had to be drained off at regular intervals before each test data point to maintain a uniform bed height. This was because of the high (38.4%) ash content of the coal. Although the coal contained 1.98% sulfur on a dry basis, its low heating value of 9489 Btu/lb influenced its SO_2 emissions levels on a lb/10^6 Btu basis. The run-of-mine coal tested was the feedstock supplied to the preparation plant from which the spirals and flotation concentrate fractions were taken to make the pellets.

Figure 15 compares the SO_2 emissions from the run-of-mine coal and the fine size limestone impregnated pellets with a Ca/S ratio of 1.6. For this high ash content run-of-mine coal, bed temperature did not strongly influence the SO_2 emissions, but compared to the limestone-containing pellets, SO_2 levels were quite high. This is attributed partly to the high ash content of the coal and the somewhat poor combustion characteristics of this particular sample of coal. The effect of Ca/S ratio, for a bed temperature of 1550°F, is shown in Figure 16 for the three fuels tested.

Figures 17 and 18 compare the NO_x and HCl emissions of the fine limestone impregnated pellets and the run-of-mine coal. The mined coal NO_x emissions were somewhat higher and its HCl emissions lower than corresponding pellet values. The mine coal contained 0.25% chlorine while the pellet feedstock had 0.311% on a dry basis.

2. Combustion Efficiency Results

Combustion efficiency of the test fuels was measured on a heating value basis. The timed ash samples collected from

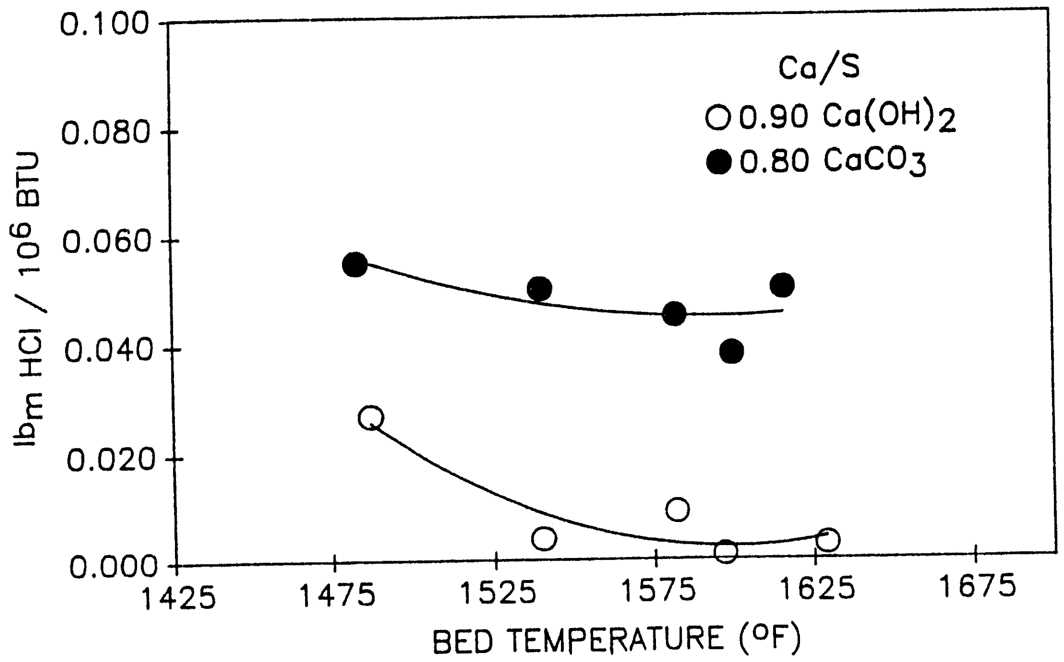


FIGURE 14(a). Influence of Sorbent Type on HCl Emissions; Ca/S Ratio ≈ 0.9.

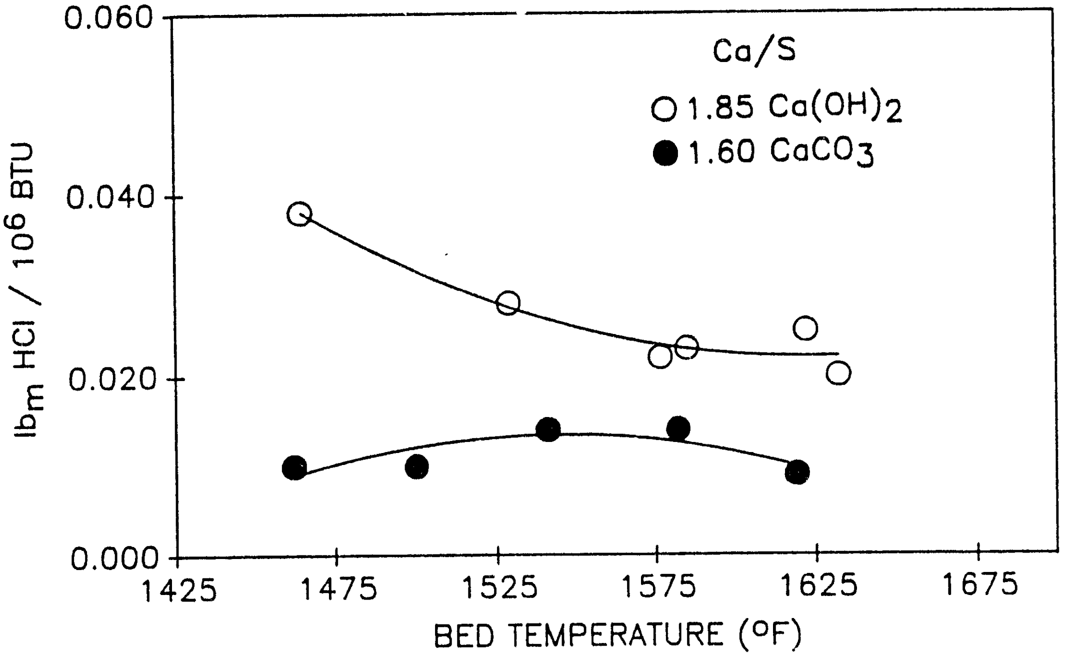


FIGURE 14(b). Influence of Sorbent Type on HCl Emissions at Higher Ca/S Ratios

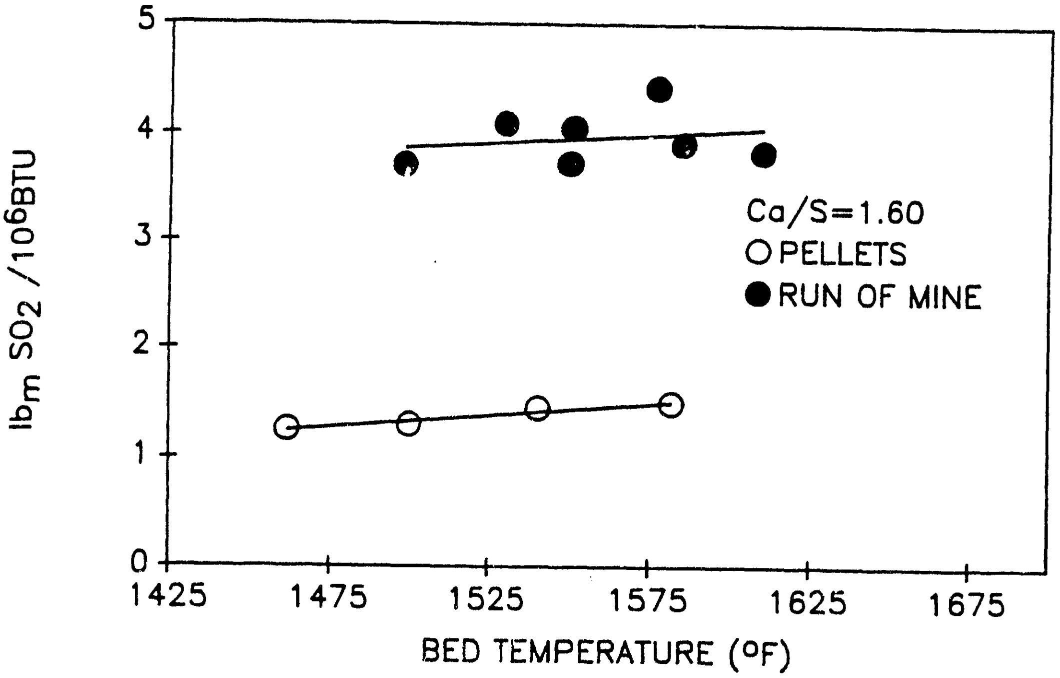


FIGURE 15. Comparative SO_2 Emissions from Run-of-Mine Coal and Fine Limestone-Containing Pellets

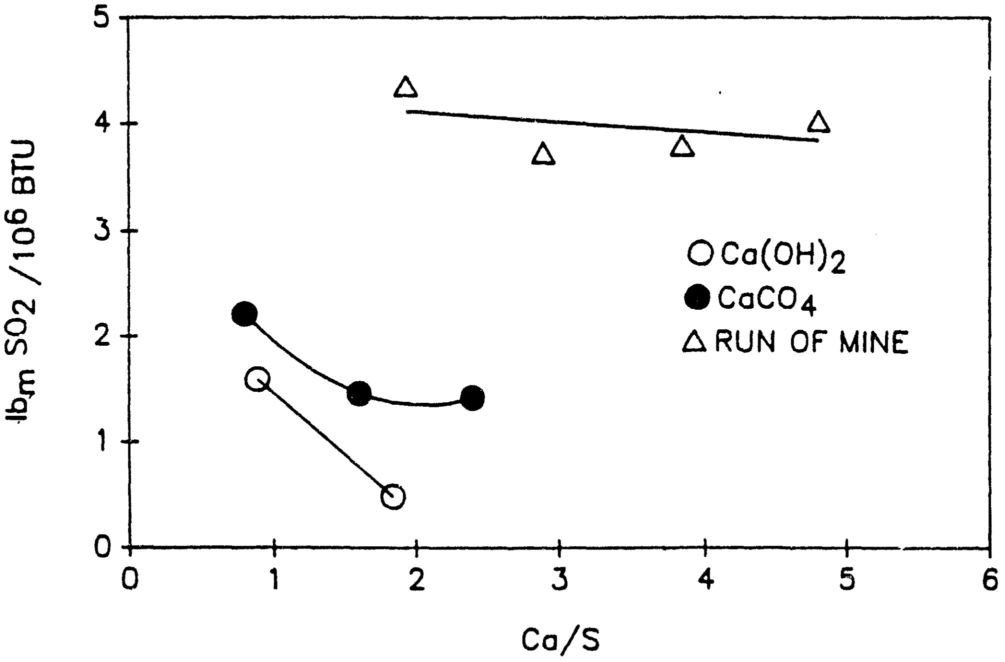


FIGURE 16. Effect of Ca/S Ratio on SO_2 Emissions

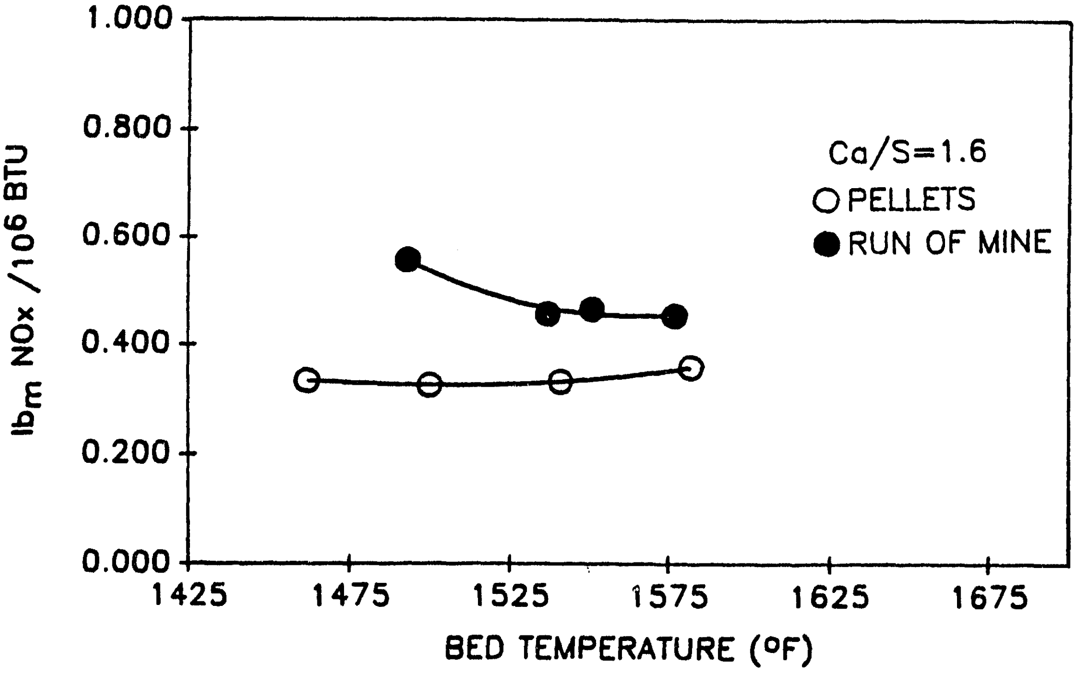


FIGURE 17. Comparative NO_x Emissions from Run-of-Mine Coal and Pellets

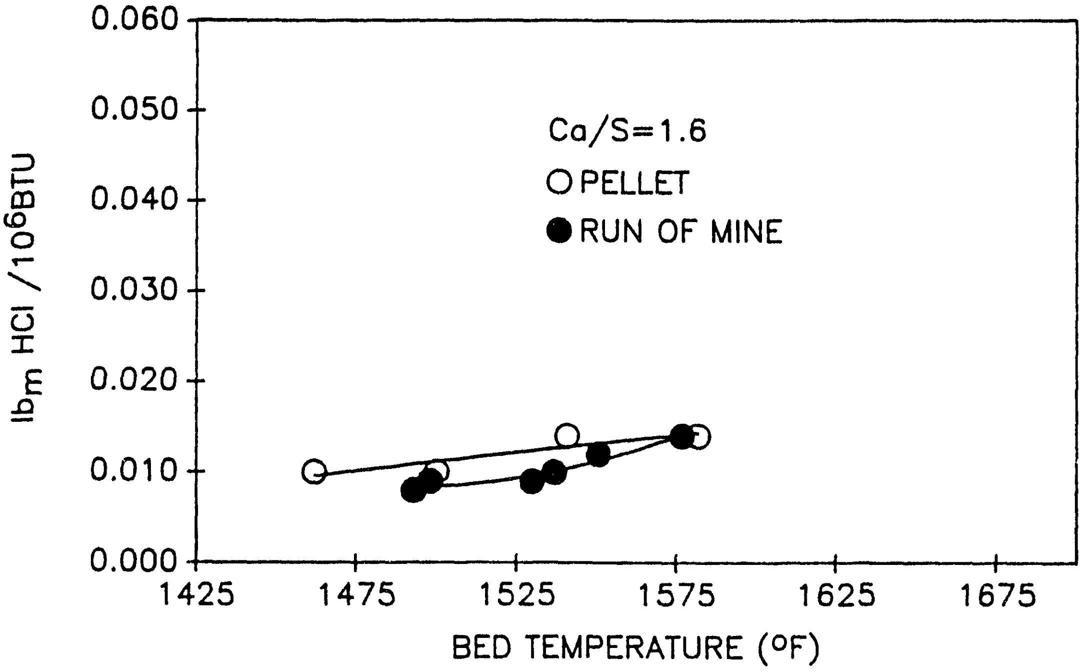


FIGURE 18. HCl Emissions Levels from Pellets and Run-of-Mine Coal

the secondary cyclones in Figure 1 were weighed and its heating value determined. Combustion efficiency was then calculated from the equation:

$$\frac{\text{Combustion}}{\text{Efficiency}} = \frac{\text{Mass fuel fed} \times \text{Heating Value} - \text{Mass ash rejected} \times \text{Its Heating Value}}{\text{Mass fuel fed} \times \text{Heating Value of Fuel}}$$

As seen from Figures 19 and 20, very high combustion efficiencies, on the order of 98%, were measured for the pellet fuels. Also, there was no effect of Ca/S ratio on combustion efficiency with both the hydrated lime or limestone sorbents. With fine limestone at a Ca/S ratio of 1.60, about 2% higher efficiencies were measured than with coarse limestone in the pellets, Figure 21. The high ash run-of-mine coal had an efficiency of about 96%, Figure 22.

3. Ash Mineral Matter Analysis

The raw feedstocks and the ash samples collected from the secondary cyclones were low temperature ashed to remove organic matter and were mounted on stubs and subjected to energy dispersive x-ray analysis to determine mineral matter content. The combustion ash samples selected for the comparisons were all taken at a bed temperature of about 1550°F and at a Ca/S ratio of 1.6. Figures 23-26 compare the percent of potassium, sodium, sulfur and chlorine in the raw feedstocks and the combustion generated ash. For the hydrated lime pellets, both potassium and sodium levels in the combustion generated ash, Figures 23, 24, were much lower than the limestone containing pellets, but the corresponding chlorine levels, Figure 25, were somewhat higher. This could mean that, with hydrated lime, the alkali metals combine more easily with chlorine to form solid products at the temperatures. Potassium and chlorine levels for the limestone containing pellets did not diminish as much in the ash, Figures 23, 25. Ash samples from the mined coal also showed a decrease of alkali metals. These results indicate that alkali vapors do result from both mined coal and pellets, which can be the cause of tube corrosion. Figure 26 shows a reduction of sulfur content in the ash indicating the release of sulfur from the coal and its subsequent reaction with the sorbent.

CONCLUSIONS

An in-depth investigation has been performed on the combustion and emissions properties of pelletized coal fuels in a 4-inch diameter circulating fluidized bed combustor. The pellets were made from a waste stream of the Kerr-McGee

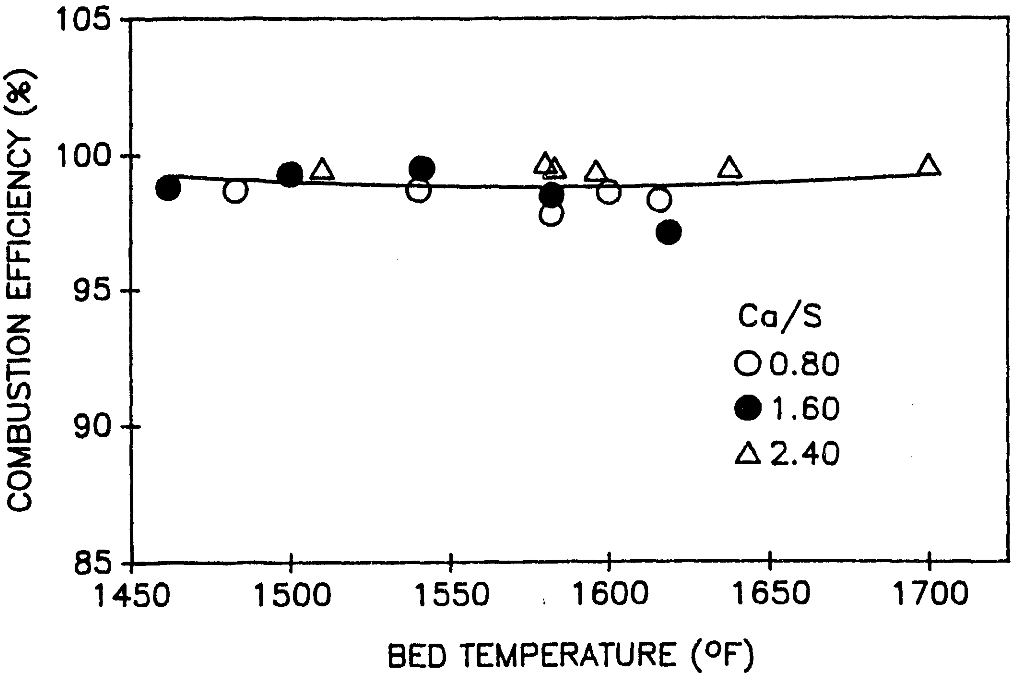


FIGURE 19. Effect of Ca/S Ratio and Bed Temperature on Combustion Efficiency; Fine Limestone Sorbent

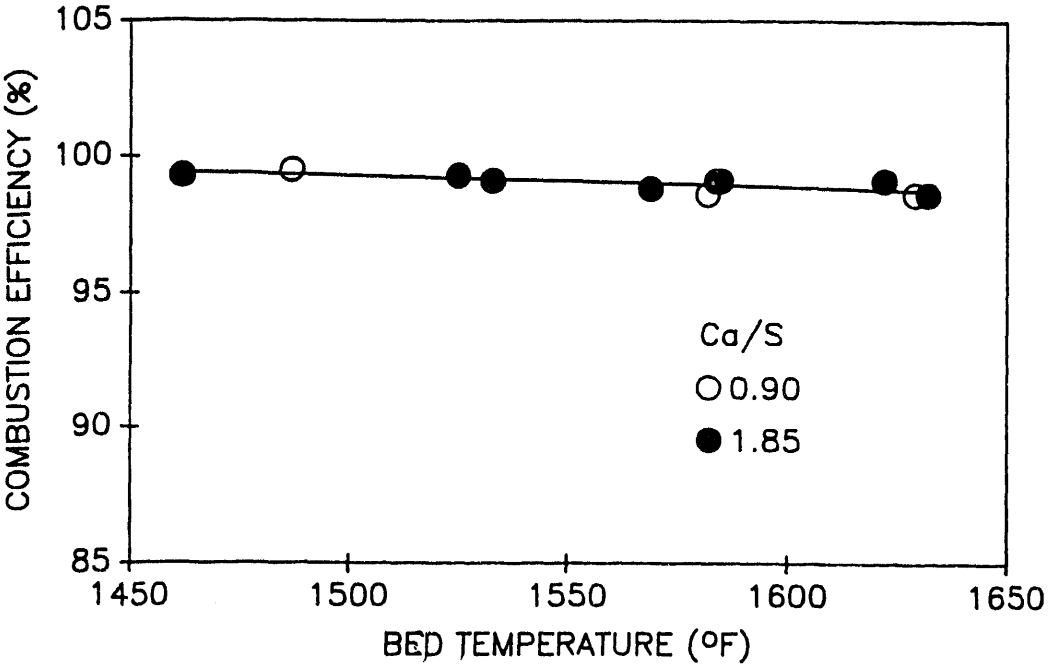


FIGURE 20. Effect of Ca/S Ratio and Bed Temperature on Combustion Efficiency; Hydrated Lime Sorbent

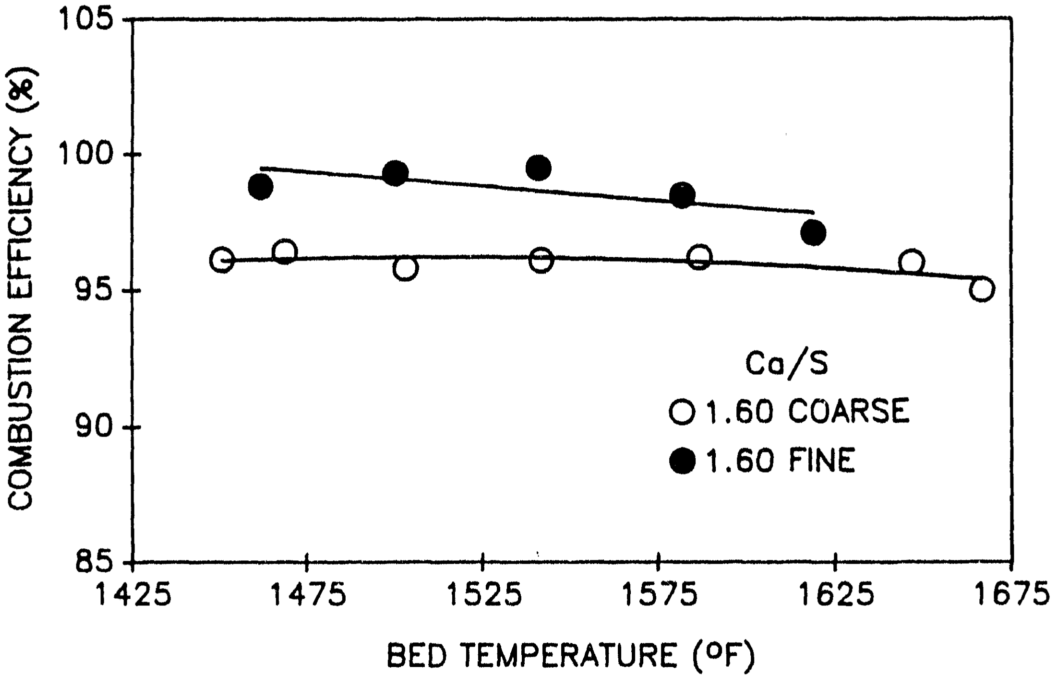


FIGURE 21. Influence of Limestone Size on Combustion

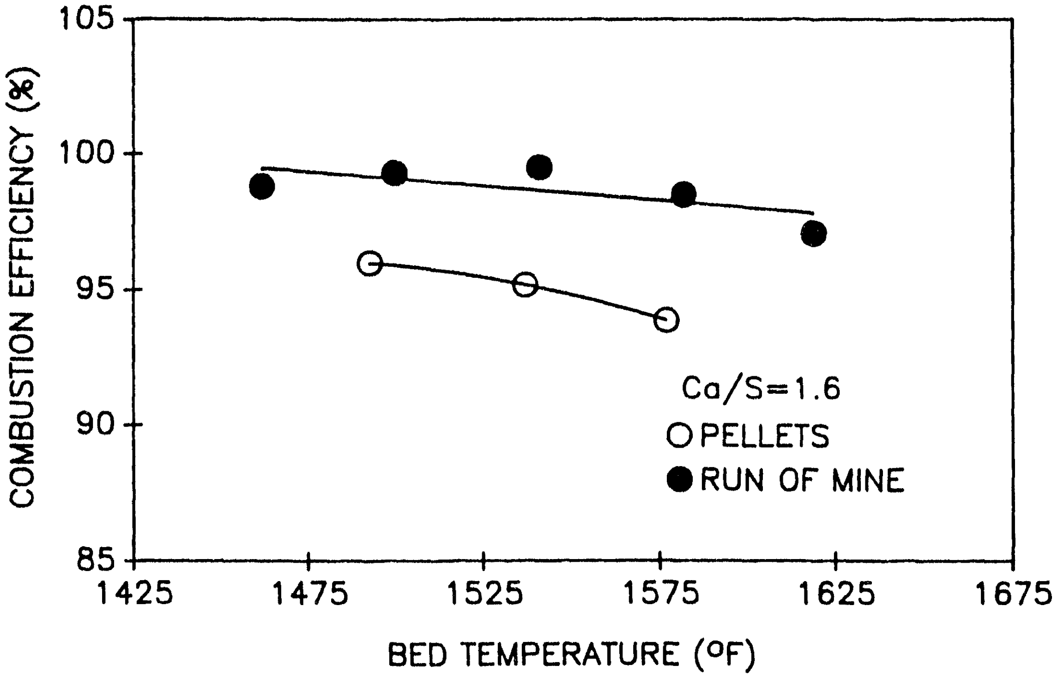


FIGURE 22. Combustion Efficiencies of Pellets and Run-of-Mine Coal

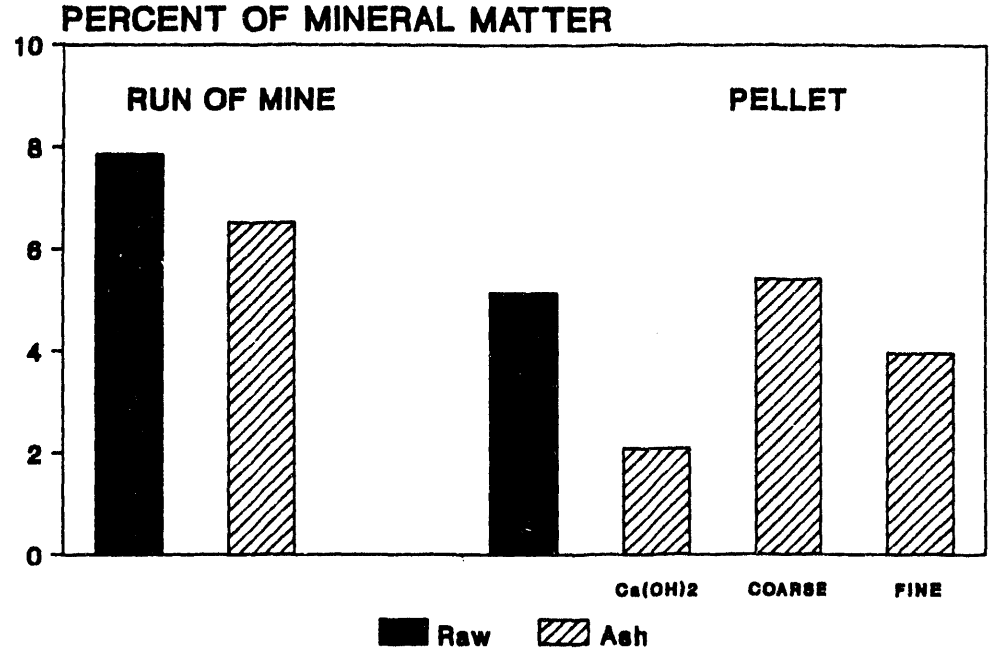


FIGURE 23. Potassium Transformations During CFBC Tests

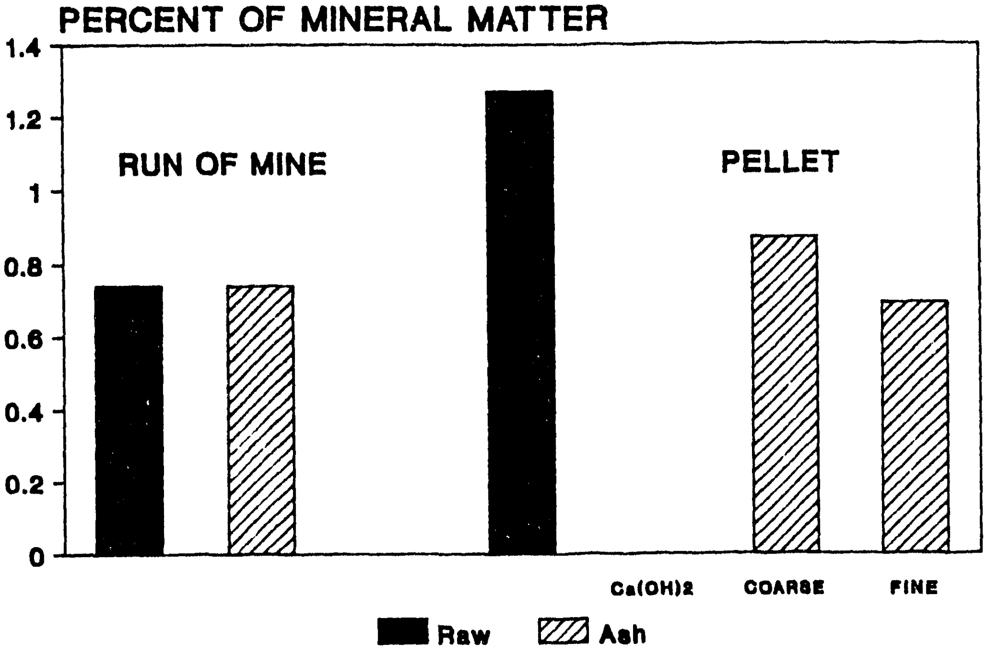


FIGURE 24. Sodium Transformations During CFBC Tests

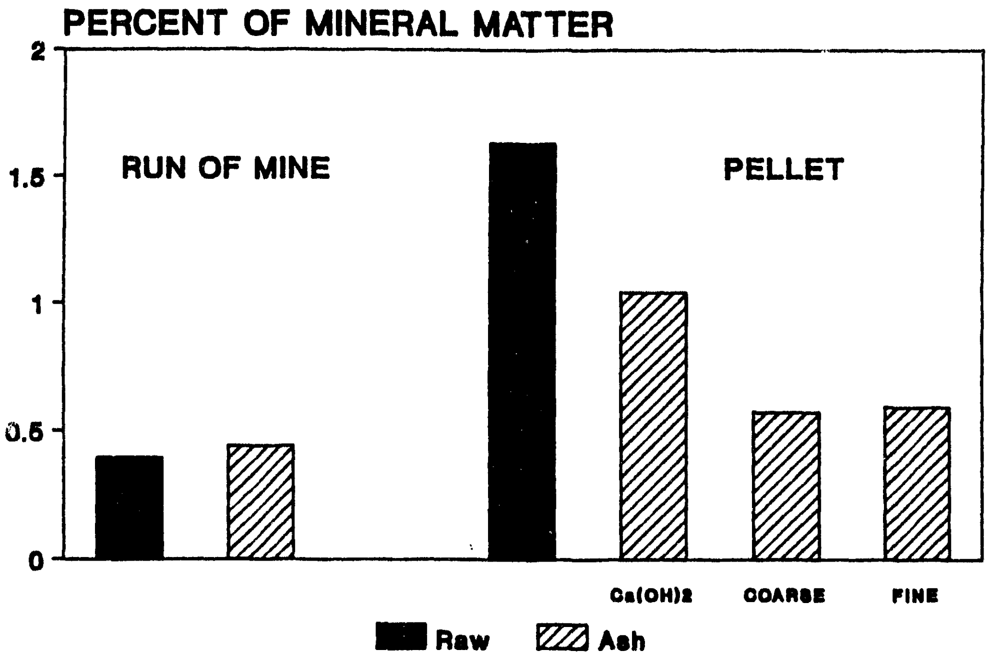


FIGURE 25. Chlorine Transformations During CFBC Tests

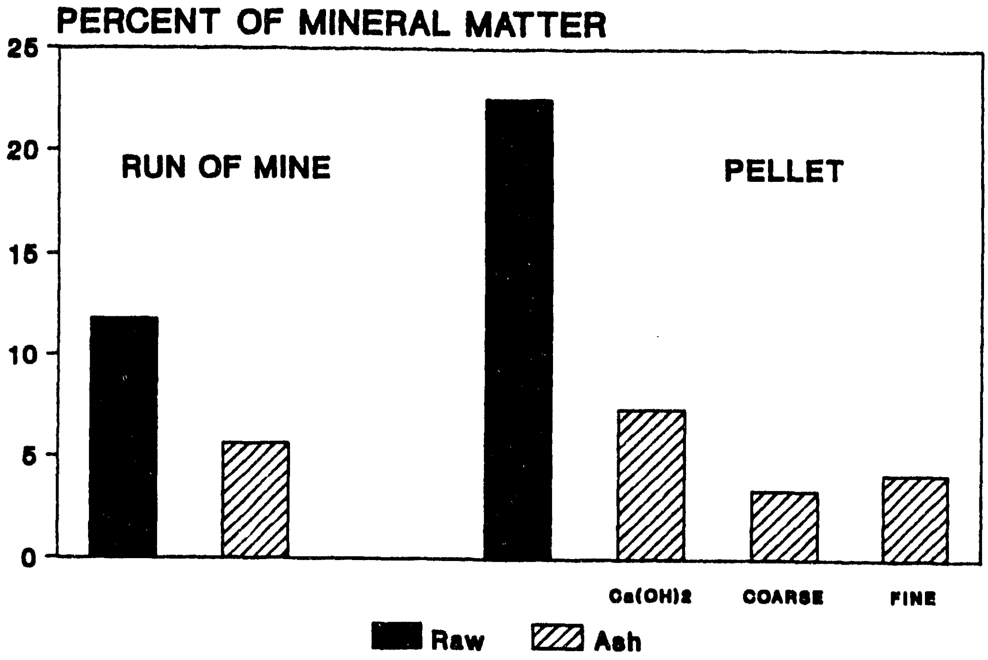


FIGURE 26. Sulfur Transformations During CFBC Tests

Galatia preparation plant. Both hydrated lime and limestone sorbents were evaluated as sulfur retaining materials. The effects of Ca/S ratios and limestone sorbent size were studied. Comparison of pellet fuels performance with that of the mined feedstock coal to the preparation plant has been made.

The CFBC combustion results show that the pellet fuels are excellent candidates for fluidized bed combustors. Not surprisingly, calcium hydroxide is a much better sorbent than limestone for pellet fuels as has been demonstrated for ordinary coal. With the hydroxide, Ca/S ratios of around 1.2-1.4 result in SO_2 emissions of 1 $lb/10^6$ Btu or less. With the limestone sorbent Ca/S ratios of about 1.8 are desirable for similar SO_2 emission levels. Oxides of nitrogen emissions of between 0.3 to 0.4 $lbs/10^6$ Btu were measured with the pellets, the feedstock coal of which had 1.42% nitrogen.

Hydrogen chloride concentrations in the combustion gases were also measured in the present tests, and ranged from 0.0075 to 0.055 $lbs/10^6$ Btu. With calcium hydroxide sorbent, increases in Ca/S ratio produced increased HCl emissions, while with limestone sorbent, Ca/S ratio increases resulted in less HCl emissions.

Combustion efficiencies with the pellets were very good being on the order of 98%. Because of the high ash content of the Illinois No. 6 run-of-mine coal selected for the present tests, its combustion efficiency was lower, and its SO_2 and NO_x emissions higher than corresponding data from the pellet fuels.

PROJECT SUMMARY

Equipment Inventory Report:

No equipment was purchased during the duration of this contract.

List of Publications:

Rajan, S., Bruyns, H. and Ho, K., "CFBC Combustion and Emissions Properties of Processed Preparation Plant Slurry Fuels," Proceedings, 12th International Conference on Fluidized Bed Combustion, ASME, 1993.

Rajan, S. and Bruyns, H., "Pelletized Coal Fuels Combustion in a CFBC Burner," Paper presented at Coal, Energy and Environment, Joint International Conference, Ostrava, Czechoslovakia, October 12-16, 1992.

Hazardous Waste Report:

No hazardous wastes were generated during the course of the experiments.

PROJECT MANAGEMENT REPORT
June 1 through August 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

EXPENDITURES - EXHIBIT B

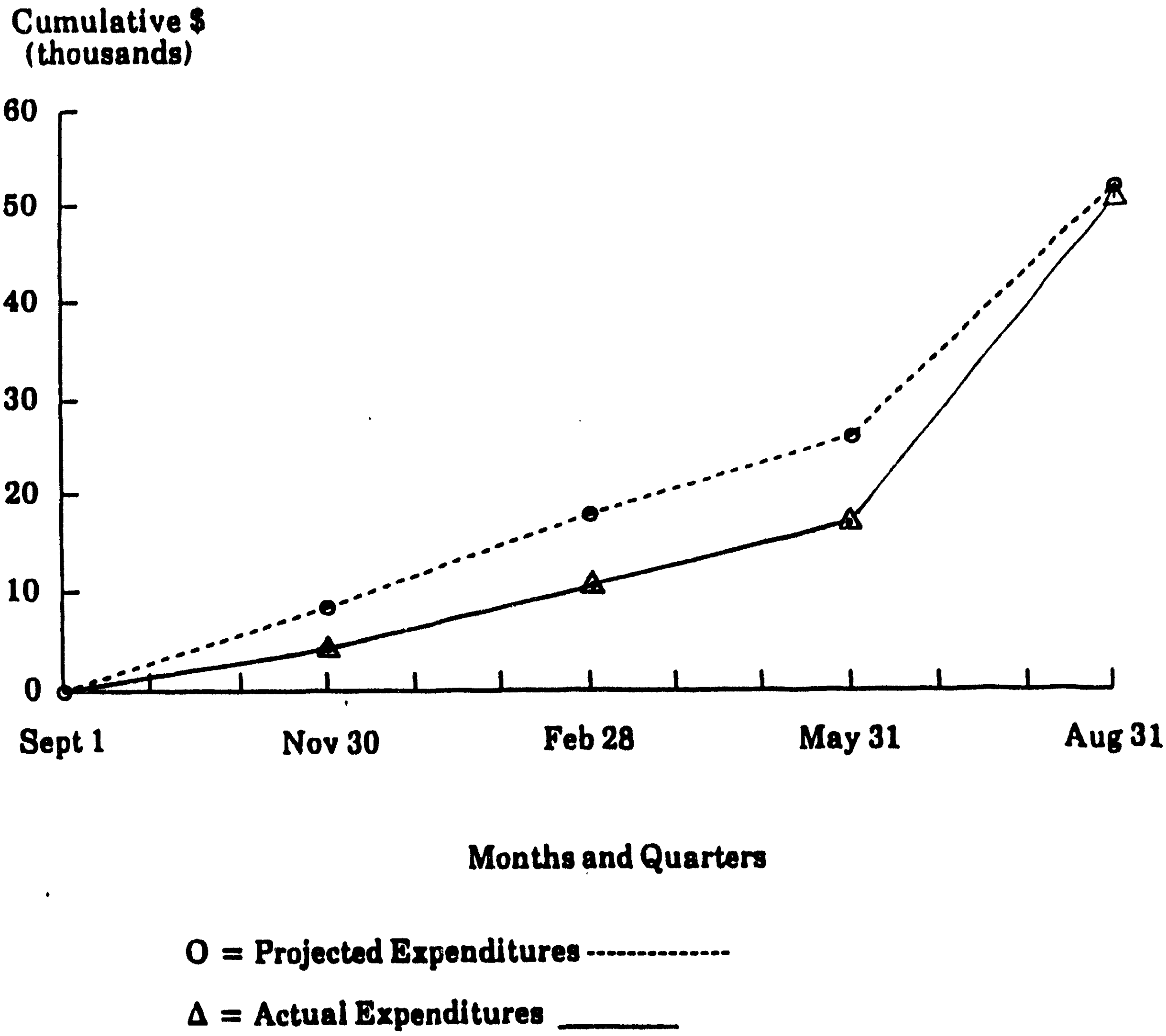
Cumulative Projected and Estimated Actual Expenditures by Quarter

Quarter*	Types of Cost	Direct Labor	Materials and Supplies	Travel	Major Equipment	Other Direct Costs	Indirect Costs	Total
Sept. 1, 1992	Projected	4,937	1,125	0	0	1,625	769	8,456
to Nov. 30, 1992	Estimated Actual	2,000	700	0	0	1,000	400	4,100
Sept. 1, 1992	Projected	9,874	2,250	0	0	3,250	3,250	16,981
to Feb. 28, 1993	Estimated Actual	6,000	1,500	0	0	2,000	1,000	10,500
Sept. 1, 1992	Projected	14,811	3,375	0	0	4,875	2,376	20,137
to May 31, 1993	Estimated Actual	8,000	3,200	0	0	3,500	2,300	17,000
Sept. 1, 1992	Projected	35,198	4,424	500	0	6,500	4,740	51,362
to Aug. 31, 1993	Estimated Actual	38,500	3,500	500	0	5,000	4,132	51,362

*Cumulative by quarter.

CUMULATIVE COSTS BY QUARTER - EXHIBIT C

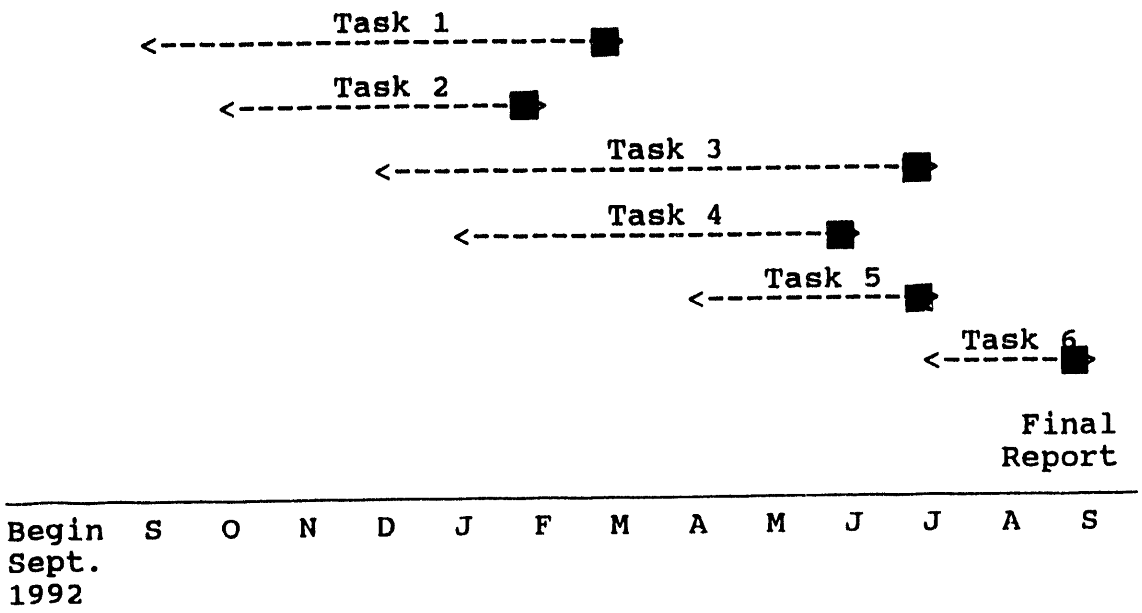
Combustion and Emissions Characterization of Pelletized Coal Fuels



Total CRSC Award $52,138

SCHEDULE

A schedule for the major tasks is given below.



Task 1.	Fuels Procurement and Manufacture
Task 2.	Fuels Testing and Analysis
Task 3.	Circulating Fluidized Bed Combustor Tests
Task 4.	Combustion Residues Analysis
Task 5.	Data Analysis
Task 6.	Final Report Preparation

DATE
FILMED
4/7/94

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