Sulfur Retention in North Dakota Lignite Coal Ash

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Introduction

Emissions from power plants such as particulate and sulfur are regulated by state health agencies and the U.S. Environmental Protection Agency (EPA). Much of the sulfur in the flue gas of coal-fired power plants is in the form of SO_2, with 1%–3% in the form of SO_3. SO_2 can be effectively controlled using commercial flue gas desulfurization (FGD) systems such as wet scrubbers. Sizing of these FGD systems affects the capital cost as well as the operational cost of the units. When the FGD system is sized, a coal analysis provides the amount of sulfur present in the coal, all of which is assumed to be SO_2 at the entrance to the FGD unit. The FGD system is then sized according to this concentration of sulfur. However, during combustion and depositional processes in the combustion system, some of the sulfur is retained in the ash because of reaction with basic oxides in the coal inorganics. The alkali and alkaline-earth elements are primarily responsible for this capture phenomena, with calcium and sodium being the most important. By accounting for the sulfur capture before the FGD, a more accurate determination of the FGD size and efficiency can be made. This report presents data from combustion tests performed at the University of North Dakota (UND) Energy & Environmental Research Center (EERC) on lignite coal, with an emphasis on evaluating the sulfur retention capability of the ash derived from the combustion. Statistical analysis and modeling of the sulfur retention and evaluation of a previous sulfur retention model developed at the EERC by Gronhovd et al. (1) is also presented.

Experimental

Coal Characterization The coal used in this test was obtained from core drilling of a mine area. Samples from the core drilling were contained in plastic bags and ranged from 0.13 to 29.5 lb per sample. Samples were put into three categories according to their sulfur levels. The categories were devised with low-sulfur samples having 0%–0.67% sulfur, medium-sulfur with a sulfur range of 0.68%–1.15% sulfur, and high sulfur having a sulfur content in the coal of greater than 1.16%, all on an as-received, weight percent basis of coal. The individual sample bags were placed into barrels of the three categories. The combined samples were then air-dried and thoroughly mixed. Samples of the three coal divisions were then analyzed using computer-controlled scanning electron microscopy (CCSEM), chemical fractionation, and x-ray fluorescence (XRF) of the ash for bulk oxide content. Table 1 shows the results of the average ultimate analysis for the three sample sulfur levels on an as-received basis. The bulk oxide composition of the ash generated from the coal sample composites as determined by XRF is shown in Table 2.

Combustion Tests The coal composite blends were tested in the conversion and environmental process simulator (CEPS), an intermediate-scale downfired combustor that fires 0.5–2.0 kg (1–5 lb)/hr of fuel, simulates conditions of a full-scale utility boiler, and can generate realistic combustion test results for a variety of fuels and combustion conditions. Control of gas temperatures and composition throughout the CEPS is possible, independent of the heat capacity of the fuel, because of its external electric heating. Flue gas temperatures can reach a maximum of 1500°C (2732°F) in the radiant section and can be maintained at 500°–1200°C (932°–2200°F) in the convective pass section and 120°–250°C (248°–482°F) in the baghouse. There is ample access for sampling, observation, and optical diagnostics through access ports located throughout the CEPS. A personal computer displays and records temperature, gas flow, feed rate, and flue gas (O_2, CO_2, CO, SO_2, and NO_x) composition, which is sampled from ports in the radiant section and after the collection device. Figure 1 is a schematic of the CEPS.

The CEPS furnace test conditions were standard operating conditions to simulate pulverized coal (PC)-fired combustion of coal. Coal was fed at a rate of 5 lb per hour, and the first six heated sections were maintained at temperatures of 1000°, 1500°, 1500° 1400°, 1250°, and 1100°C starting at the top of the combustion zone. Air was introduced into the system at the rate of 30 standard liters per minute (SLPM) as primary air and 137 SLPM as secondary air. The ash generated in the system was collected at the baghouse as well as at the horizontal section just after the combustion chamber. This ash was analyzed for sulfur content and the amount of sulfur exiting the system obtained by difference when compared to the total sulfur contained in the coal. These data were used to evaluate the sulfur retention model developed at the EERC in a previous study.

Table 1. Ultimate Analysis for the Three Composite Samples (as-received basis)

	Low Sulfur	Medium Sulfur	High Sulfur
Hydrogen	6.49	7.12	6.66
Carbon	30.46	33.41	34.25
Nitrogen	0.50	0.56	0.54
Sulfur	0.51	0.95	1.61
Oxygen	47.25	48.24	42.47
Ash	14.78	9.74	14.49
Btu	5830.00	6008.50	5961.50

Table 2. XRF Bulk Oxide Composition of Ash of Coal Sample Composites (weight percent, mineral basis)

	Low Sulfur	Medium Sulfur	High Sulfur
Silica	39.13	23.16	24.18
Aluminum	13.14	8.95	7.66
Iron	1.94	5.65	14.87
Titanium	0.93	0.55	0.43
Phosphorus	0.29	0.19	0.11
Calcium	17.36	18.39	13.14
Magnesium	6.44	6.82	4.77
Sodium	5.31	6.61	4.53
Potassium	0.52	0.40	0.40
Sulfur	14.36	28.92	29.44
Barium	0.58	0.36	0.46

Results and Discussion

Regression Analysis Statistical analyses of nine different samples of recently obtained lignite and 40 other lignite samples from a previous study by Gronhovd et al. (1) have been used to determine the efficacy of a model developed at the EERC in a previous study to predict sulfur emissions. Because calcium seemed either to be heavily affected by other factors or else insignificant, it was not included as a factor in the new model. Instead, silicon and aluminum contents were

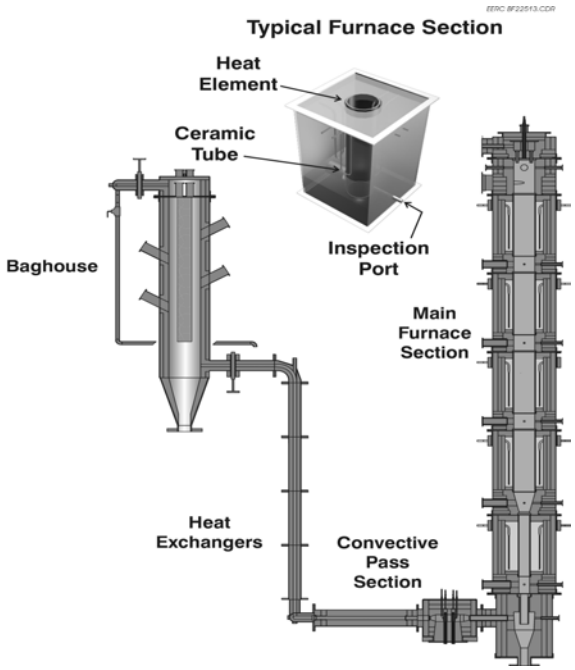

Figure 1. Schematic of the EERC CEPS.

included, which had seemed important to sulfur emissions in the previous tests. Aluminum was later disregarded as neither its effect nor its interactions proved important (the previous tests seemed to find a correlation between Al_2O_3 and sulfur emissions, but this may have been due to a correlation between aluminum and some other element). The final model developed in this study was as follows:

$$173.5!16.77(Na_2O)!2.207(SiO_2)!78.62(S)+0.2608(Na_2O)(SiO_2) +7.898(Na_2O)(S)+1.961(SiO_2)(S)$$

where the Na_2O and SiO_2 are given in weight percent oxide from the bulk ash XRF analysis and the sulfur is from the weighted average ultimate analysis from the three composite samples. The sulfur was computed on a moisture-free basis by weighting the measured sulfur level for each sample that went into the composite by the amount of sample in grams. These values are listed in Table 3 along with the measured sulfur in the collected combustion ash. The model r^2 term was 0.65, and its standard deviation was 10.1. This r^2 was slightly lower than the model developed previously at the EERC by Gronhovd. Statistically, the new model developed using the Grohovd data and the new lignite data has slightly better accuracy than did Grohovd's previous model. The results of the newly developed model are shown in Figure 2 and Table 4.

Table 3. Sulfur Levels in the Coal and the Collected Combustion Ash

	wt% in Coal (coal basis)	wt% in Combustion Ash (ash basis)
High Sulfur	3.29	2.1
Medium Sulfur	1.57	4.5
Low Sulfur	0.95	4.3

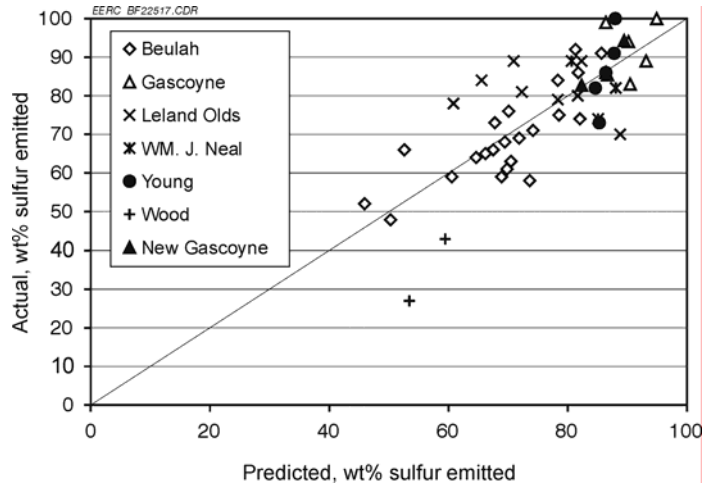

Figure 2. Predicted vs. actual percent sulfur emissions based on our current model.

Table 4. Predicted Versus Actual Sulfur Emitted from the Combustion Tests (percent of sulfur in coal)

	Predicted	Actual
High Sulfur	79	93
Medium Sulfur	70	83
Low Sulfur	87	87

Conclusions. The coal samples used in this study were divided into three composite samples. The coal came from core drilling of the lignite mine area. Core samples ranged in size from 0.13 to 29.5 lb per sample. The three composite samples were compiled according to sulfur levels of low, mid, and high, having values of 0%–0.67%, 0.68%–1.15%, and 1.16% and above, respectively. CCSEM analysis showed increasing amounts of pyrite and pyrrhotite and decreasing quartz and clays as the sulfur level increased. Chemical fractionation data indicate minor differences in the amount of material organically bound and carbonate materials in the three composite samples. The combustion tests of the three composite samples showed sulfur retention in the ash of roughly 7%–17% of the total sulfur in the coal. A new model developed from the data set produced in this study, as well as the data set from Gronhovd's previous study, indicates a slight increase in predictive capability over Gronhovd's previous model.

The regression model indicates that the original model developed by Gronhovd at the EERC in previous studies remains an adequate representation of the sulfur emissions expected from low-rank coals (1). While the newly developed model is statistically superior, the difference is minimal. Another point of the study is that the results from combustion studies performed in the EERC CEPS match the full-scale data from the previous study very well, validating the use of this pilot-scale equipment for modeling reactions in full-scale systems.

References

1. Some Studies on Stack Emissions from Lignite-Fired Powerplants: Technology and Use of Lignites Proceedings: Bureau of Mines, University of North Dakota Symposium 67, North Dakota, May 9–10, **1973**.