Intra-Particle Sulfur Capture During Battelle Treated Coal Combustion Under Simulated Turbine Conditions

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Introduction

For the last several years, Battelle Columbus Laboratories has been actively conducting research on the development of a practical, cost-effective process for improving both the fixed-bed gasification(1) and the boiler combustion(2) characteristics of high-sulfur, agglomerating, low-reactivity bituminous coal. The product of this research is a calcium-impregnated coal called Battelle Treated Coal (BTC), given the acronym BTC. The properties of BTC are much different from those of the raw coal from which it is processed(1,2). Specifically, these differences are:

- Increased ASTM volatile matter content.
- Higher char reactivity.
- Reduced tar formation tendency.
- Hotter ash fusion temperatures.
- Diminished coal particle agglomeration tendency.
- Enhanced sulfur retention by the ash.

As near-term markets for gasifier and boiler coal diminish or stabilize, respectively, Battelle has expanded the search for other old or new applications for bituminous coal that would also benefit from the alterations that could be given it by the Battelle calcium impregnation process. Recent planning studies by the United States Department of Energy (U.S. DOE) have forecast a need for coal-fired combustion turbines for the repowering of electric utilities when they become capacity-limited or deficient in the early-to-mid 1990s(3). The purpose of the program reported here is to demonstrate the promise of utilizing BTC as a fuel in advanced, coal-capable combustion turbines.

This paper is the second(4) in a series to present the results of a U.S. DOE-sponsored program designed to determine the intra-particle sulfur capture efficiency of BTC under simulated combustion turbine conditions. The BTC consisted of either of two high-sulfur (>4 percent by weight) Illinois No. 6 coals that had been impregnated with calcium-containing catalysts using a proprietary Battelle process. The simulated turbine combustion conditions consisted of super-atmospheric pressure, ultra-high excess air levels, average firing temperatures, and near-quantitative carbon conversion. Combustion test results reported here will focus on the parametric and comparative effects of raw coal organic sulfur content, calcium source, and calcium impregnation technique on sulfur capture efficiency. The effects of pressure and temperature on sulfur capture efficiency, as well as on ignition, burnout, and ash deposition, were the subjects of the earlier report(4).

Experimental

A coal-fired combustion turbine simulator was constructed at Battelle by converting a continuous high-pressure reactor from a gasifier to a combustor(4). A schematic of the combustion turbine simulator is shown in Figure 1. This device is capable of simulating(3,4) the following combustion turbine conditions:

- Super-atmospheric pressure: 4 atmospheres.
- Maximum coal flame temperature: 2600 F.
- Maximum firing temperature: 1900-2100 F.
- Maximum liner temperature: 1000-1500 F.
- Primary fuel/air ratio: 20 percent excess air.
- Overall residence time: 150-300 milliseconds.
- Carbon conversion efficiency: 99+ percent.

A premixed methane/air pilot flame was first used to ignite the down-flowing pulverized coal feed at atmospheric pressure. The coal firing rate was then adjusted to about 40,000 Btu/hour. Primary air amounted to 20-30 percent of the total air required for combustion at 20 percent excess air. Secondary air made up the remainder. Temperatures were continuously monitored down the vertical axis of the combustor at locations in the pre-to-post flame zones using Inconel-sheathed Chromel-Alumel thermocouples. Firing temperature was adjustable over a 200 F range by either varying the amount of tertiary dilution air introduced or by oxygen enriching the secondary air to various levels. After combustion was completed within the 12-foot long, 6-inch diameter combustion chamber, samples of gaseous and solid products were collected using an uncooled stainless steel probe or a cyclone mounted in the exhaust, respectively. Flue gases were analyzed for CO_2, CO, O_2, and SO_2 using conventional analytical instrumentation(4). Once the diagnosis of atmospheric-pressure combustion conditions was completed, the system was pressurized to 65 pounds per square inch absolute (psia) and the gas/solid sampling procedure repeated.

The combustion test plan consisted of experiments designed to determine the comparative in situ sulfur capture efficiency, under simulated combustion turbine conditions of: (a) raw pulverized bituminous coal, (b) pulverized BTC prepared from this raw coal, and (c) a physical mixture of the raw pulverized coal and pulverized lime with the same amount of calcium added as for the BTC.

The two raw bituminous coals used were both from the Illinois No. 6 seam. As Table 1 shows, both had total sulfur contents of greater than 4.50 weight percent. The difference between them was that one's total sulfur was predominately organic, whereas the other's consisted of a near-equal split between organic and inorganic sulfur.

The raw coals were first dried to about 2 percent moisture, pulverized to 85-90 percent minus 200 mesh (75 microns), and then fired.

To produce BTC, coarse raw coal, 100 percent minus 20 mesh (850 microns), was first treated via the proprietary Battelle calcium impregnation process (1) to increase its molar Ca/S ratio to the levels listed in Table 1. Either commercial grade or technical grade lime was used as the source of the calcium. The difference between these calcium sources is purity and cost with the commercial grade being less pure and less expensive. The BTC was then dried to 2-3 percent moisture, pulverized to 85-90 minus 200 mesh, and then fired.

For the third fuel, finely-pulverized (85-90 percent minus 200 mesh) raw coal was physically mixed with either the commercial or the technical grade lime, each having a particle size of 100 percent minus 325 mesh (45 microns), and then fired. The physical mixtures' molar Ca/S ratios were adjusted to be as equivalent as possible to that of the BTCs (see Table 1).

Results

The following are the significant new results regarding sulfur capture of these latest combustion tests, the results of which are documented in Table 1:

- Inherent sulfur capture by raw Illinois No. 6 seam coal appears to be independent of sulfur form, i.e., organic vs inorganic.
- Sulfur capture by Illinois No. 6 seam coal treated via the Battelle calcium impregnation process (BTC) apepars to be independent of sulfur form and grade of lime at both low and high Ca/S ratios.
- Sulfur capture by BTC made from Illinois No. 6 seam coal is at least 50 percent more efficient than that achievable upon physically mixing raw coal and either grade of lime.
- Sulfur capture by BTC at low levels of calcium impregnation exhibits a positive pressure dependence over the entire range of pressures studied.
- All other positive effects of Battelle calcium impregnation on raw coal combustion performance(4) -- higher ash fusion temperatures, lower free-swelling indices, smaller fly ash, negative pressure dependence for NO_x -- are observed for the organic sulfur enriched Illinois No. 6 seam coal.

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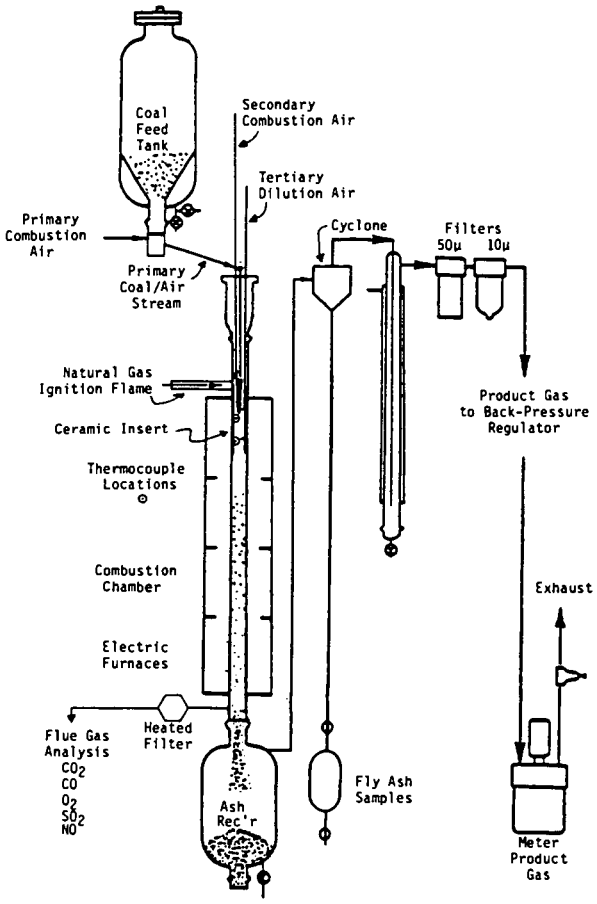

FIGURE 1. BATTELLE COMBUSTION TURBINE SIMULATOR

TABLE 1. SIGNIFICANT RESULTS OF TESTS TO STUDY THE EFFECT OF ORGANIC SULFUR CONTENT AND CALCIUM SOURCE ON SULFUR CAPTURE AS A FUNCTION OF PRESSURE, CALCIUM IMPREGNATION TECHNIQUE, AND Ca/S RATIO

Illinois No. 6 Seam Coal		Calcium			Sulfur Capture	
Total Sulfur (Weight %, DAF)	Organic Sulfur (%)	Impregnation Technique	Calcium Source	Ca/S Ratio	@15 PSIA (%)	@70 PSIA (%)
5.24	72	None	Nature	0.08	12±5	18±9
4.55	47	None	Nature	0.17	17±9	13±6
5.24	72	BTC	CGL	1.12	61±2	80±3
4.55	47	BTC	CGL	1.31	60±5	81±6
5.24	72	BTC	CGL	3.50	75±4	92±3
4.55	47	BTC	CGL	4.11	83±7	95±8
5.24	72	PM	CGL	0.98	35±10	57±18
5.24	72	PM	CGL	3.10	59±7	74±3
5.24	72	BTC	TGL	1.15	68±2	82±2
5.24	72	BTC	TGL	3.25	86±1	96±2

BTC = Battelle Treated Coal
PM = Physical Mix of Raw Coal and Lime
CGL = Commercial Grade Lime
TGL = Technical Grade Lime