FY02-XLII-108

LOW-TEMPERATURE NO_X REDUCTION USING HIGH-SODIUM LIGNITE-DERIVED CHARS

CONTRACTOR: Energy & Environmental Research Center

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PARTICIPANTS

Sponsor	Cost Share
Department of Energy (EERC/JSR/DOE)	$380,000
BNI Coal Ltd.(Cash)	$ 40,000
BNI Coal Ltd (In-Kind)	$240,000
Minnkota Power Cooperative (Cash)	$ 40,000
Luscar (Cash)	$ 40,000
Basin Electric(Cash)	$ 40,000
EPRI (Cash)	$ 40,000
ND Industrial Commission (Approved 6/19/02)	$ 40,000
ND Industrial Commission (Approved 10/01)	$160,000
Total	**$ 1,020,000**

Project Schedule – 24 Months
Contract Date – 3/14/02
Start Date – 5/22/02
Completion Date – 5/22/04
Time Extension – 6/30/04

Project Deliverables
Quarterly Reports – 6/30/02 √
9/30/02 √; 12/30/02√_; 3/30/03 _√
6/30/02 √_; 9/30/03√; 12/30/03_√
3/31/04 (√);
Final Report – 2/08/05 √

OBJECTIVE / STATEMENT OF WORK

The primary goal of this project is to reduce NO_x emissions from cyclone-fired boilers. A secondary goal is the oxidation of Hg^0 to Hg^{++}. The project includes the production of chars from high-sodium lignite, bench-scale testing, construction of a small demonstration-scale circulating fluid-bed reactor (CFBR), testing in the CFBR, and technical and economic evaluation. NO_x and Hg emissions are environmental issues subject to future regulation. Control of NO_x emissions from lignite-fired cyclone-fired boilers is a unique challenge that is not readily accomplished with current SCR or SNCR technology. Likewise, mercury emissions from lignite-fired systems are a unique challenge because mercury occurs as Hg^0. This project addresses two distinct and unique challenges facing the lignite industry.

STATUS
The CarbNO$_x$ project was initiated during this quarter with a kickoff meeting for project sponsors held on May 22. The agenda included presentations on the background for and details of the project followed by a discussion of materials to be tested for char production. Project activities initiated during the quarter focused on procurement of coal samples, developing protocols for bench-scale char production and analysis, laboratory-scale testing of prepared char, and drafting a design for an NO$_x$ reduction reactor. Discussion with the project sponsors resulted is selecting lignite samples currently being mined by BNI Coal from the Center Mine, Beulah–Zap from the Freedom Mine, and Luscar coal. Protocols have been developed for the synthesis of char from various coal samples. Analytical procedures for laboratory characterizations have been completed. The reactor has been designed, and the procurement of materials has begun.

April – June 2002 Quarterly Report. Preliminary data acquisition for the feasibility of this process will be carried out on the flue gas exiting a coal combustion unit – the conversion and environmental process simulator (CEPS). This process involves contacting hot ash-laden flue gas with carbonaceous materials. The initial design for this purpose is a spouting bed to avoid ash buildup and the concomitant plugging of the carbon bed. A granular bed filtration system will be designed and constructed for the management of the ash.

July 1 – September 30, 2002 Quarterly Report

Project activities focused on developing protocols for bench-scale char production and analysis, laboratory-scale testing of prepared char, and fabrication of a NOx reduction reactor for pilot-scale testing of the process. A new tube furnace capable of heating a 2.5-inch reactor tube was received and installed in the laboratory.

Carbons were prepared from Luscar char and Freedom mine coal. Granular carbons were analyzed for surface area NOx absorption studies. Activation studies examining the influence of CO2 versus steam for activation of Freedom coal was also studied, with steam providing a four-fold increase in surface area. Activated chars will be prepared and undergo bench-scale tests to compare NOx reduction effectiveness and to optimize char production.

Jan 1 – Mar 31, 2003
Large-scale batches of activated carbon were produced in the 2.5-inch tube furnace reactor. Three successful CEPS runs were completed and preparations began for CEPS tests during the next quarter. NOx reductions of up to 78% were observed.

Apr 1 – Jun 30, 2003
Bench scale test results indicate a maximum reduction of NOx of up to 98 %. In tests with SO2 addition, NOx levels were reduced by up to 90%. However, SO2 levels at the outlet were below the inlet concentration, suggesting that the SO2 was being captured by the char or were converted to other constituents such as SO3, COS, etc.

July – Sept, 2003
Using a Freedom mine derived lignite, char was prepared and activated at a temperature of 750 C. Bench scale testing indicated good reduction of NOx, but CO emissions remain a concern. Economic studies on the process will be initiated and additional studies will be conducted on reducing CO emissions.

Oct 1 – Dec 31, 2003
The effect of variations of reactor temperature was evaluated using a Freedom mine derived lignite char. At temperatures ranging from 475 to 512 C, only partial drops in NO levels were observed. At 550 C, the initial mass of char is sufficient to completely reduce the NO to N_2, but a loss in char coupled with insufficient kinetics leads to unreacted NO exiting the reactor bed. At 587 C to 625 C, nearly all of the NO gas was reduced. At low temperatures, the kinetic reduction of the char-NO reactions are slow; at higher reactor temperatures, a higher rate of NO reduction and a high selectivity was observed.

Jan 1 – April 30, 2004. An economic evaluation of the CarbNOx process as applied to commercial-scale facilities is being developed in conjunction with Ohio State University.

Final Report. The primary goal was to demonstrate the ability of lignite-derived char to effectively reduce NO_x levels from combustion gases produced from a cyclone-fired combustion system firing North Dakota lignite.
The pilot-scale tests showed that the process can reduce NO_x more than 98%. However, NO_x reduction by activated carbon depends on reaction temperature: the higher the reaction temperature, the greater the NO_x reduction. Tests confirmed that the process can reduce NO_x more than 95% even in the presence of higher levels of SO_2. However, the oxygen content needs to be less than 5% because oxygen competes directly with the NO_x reduction reaction by consuming the carbon.

The pilot-scale tests showed that the process produced CO, which is a concern for the commercial application of the process. The $CarbNO_x$ reactor was modified to improve temperature control of the reactor as high CO emissions are associated with higher operating temperature. Although the CO concentration at the outlet of the reactor decreased at lower operating temperature, the NO_x reduction rate decreased as well compared to the NO_x reduction at higher operating temperature. An attempt was made to convert the CO at the outlet of the reactor with the addition of an EERC-prepared Ni_2O_3 catalyst bed. However, no significant change in CO emissions was observed.

Conclusion: The $CarbNO_x$ technology could be added to coal-fired power plants to remove the NO_x generated in the combustion furnace from the flue gas stream before it leaves the stack. The process could use activated carbon prepared from North Dakota lignite coal. However, the results of this research show that the technology needs to overcome excess carbon monoxide production before it can be applied to a full-scale coal combustion power plant.