FINAL TECHNICAL REPORT
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Project Title: **ILLINOIS COAL GASIFICATION/REFORMING USING LOW-TEMPERATURE PLASMA**

ICCI Project Number: 05-1/4.1B-4
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ABSTRACT

A GTI led team is developing a novel technology, low-temperature Plasma Assisted Reforming (PAR), for production of hydrogen, syngas for Fisher-Tropsch synthesis, and fuel-gas for gas turbines from coal. In the PAR process, micronized coal is converted to product gas at temperatures of 400°C to 700°C. The product gas is fed into a warm gas cleanup unit for removal of sulfur, mercury and other contaminants and then into a purification unit for gas separation and CO_2 capture.

The project objectives include: Task 1, confirming the reactivity of Illinois coal in GTI's laboratory scale plasma reactor; Task 2, performing pilot-scale prototype tests of the plasma-assisted gasification reactor; and Task 3, based on the results of testing, performing a system analysis and market evaluation for three prospective applications of the technology for Illinois coal: production of hydrogen, fuel gas for gas turbines, and syngas for chemical synthesis. The first two efforts include studies of coal and processed chars characterization conducted in collaboration with Dr. John Crelling of SIUC. The plasma characterization study was conducted with participation of Dr. Saveliev of UIC. The Team evaluated the plasma technology applicable for Illinois coal and defined the presence of electric charges on coal particles after plasma treatment.

Coal reactivity was characterized in a fixed bed configuration in a laboratory scale semi-batch 200-W Dielectric Barrier Discharge (DBD) reactor. Results from the tests indicate that application of plasma and coal micronization increase gas production. In scale up studies, the operational feasibility of steam jet based micronization of Illinois coal was confirmed experimentally without adverse operational issues, such as plugging due to the caking propensity of the feed material. Major reaction products in the PAR tests were hydrogen (60%-80%), carbon dioxide (14%-29%), and carbon monoxide (4%-10%), while concentrations of hydrocarbons were below 3%. Test results showed an increase in gasification rates due to the use of plasma. Modeling results showed a 25% reduction in production costs and more than a 10% efficiency increase of the PAR technology compared to traditional gasification.

Page(s) 1-30 contain proprietary information.

EXECUTIVE SUMMARY

A GTI led team is developing a novel technology, low-temperature Plasma Assisted Reforming (PAR), for production of hydrogen, syngas for Fisher-Tropsch synthesis, and fuel-gas for gas turbines from coal. In the PAR process, micronized coal is converted to product gas at temperatures of 400°C to 700°C. The product gas is subsequently fed into a warm gas cleanup unit for removal of sulfur, mercury and other contaminants and then into a purification unit for gas separation and CO_2 capture.

The overall project objectives include: Task 1, confirming the reactivity of Illinois coal in GTI's laboratory scale plasma reactor; Task 2, performing pilot-scale prototype tests of the plasma-assisted gasification reactor; and in Task 3, based on the results of laboratory and pilot scale testing, performing a system analysis and market evaluation for three prospective applications of the technology for Illinois coal: production of hydrogen, fuel gas for gas turbines, and syngas for chemical synthesis. The first two efforts include studies of coal and processed chars characterization conducted in collaboration with Dr. John Crelling of SIUC. The plasma characterization study was conducted with participation of Dr. Saveliev of UIC. The Team studied the plasma technology applicable for Illinois coal and defined the presence of electric charges on coal particles after plasma treatment.

In Task 1: Laboratory Scale Testing, Illinois #6 coal reactivity was studied in a laboratory scale plasma reactor. The laboratory scale tests were conducted using three average coal particle sizes: 1 mm, 70 micrometers, and 10 micrometers. The reduction in particle size by passing coal through a steam jet mill micronization apparatus increases the particles surface area available for reaction, and potentially can alleviate operational issues associated with caking/agglomeration. There has been no caking of the IL coal observed for tests with an average particle size of 10 micrometers. After testing, the material had a morphology of powder similar to the original sample. Tests with larger particle sizes have shown significant caking. Particles agglomerated together on the tray and a "crust"-like surface was produced. This result is significant, because it shows that coal micronization can prevent caking and potential plugging of the continuous unit. Higher reaction rates were observed in tests with 10-micron particles, compared to tests with larger particles. The smaller particles also generated three times more dry product gas volume than the larger particles. Higher concentrations of hydrogen, carbon monoxide and methane were produced in tests with plasma compared to the tests without plasma. Similar concentrations of carbon dioxide in product gas were observed in tests with and without plasma. On the average there was a 10% to 20% increase in dry gas production rate observed due to plasma operation. Laboratory scale tests were conducted at 600°C and 700°C. Methane and hydrocarbons are produced at higher concentrations at lower process temperature, while hydrogen, carbon monoxide and carbon dioxide are the major products at higher temperature. Production rates of dry gas increase significantly at higher process temperature, as expected. No significant effect of process pressure on reaction rates and product gas composition was observed.

In Task 2: Scale up study of the PAR process with Illinois coal, the goals were to evaluate the performance of a micronizer operated with Illinois coal, determine coal conversion into product gas and product gas composition in an entrained flow reactor, and characterize DBD plasma in a mixture of Illinois coal and steam. GTI's Pilot scale DBD reactor facility was used to study the PAR process with Illinois coal. The PAR process combines three major technologies – micronization, plasma, and coal/steam reforming. First, coal is micronized to a particle size below 20 micrometers in a steam jet micronizer mill, and then plasma is applied to the mixture of steam and micronized coal. The role of micronization is to increase the active reaction surface of coal particles, and the role of plasma consists in creating active reaction sites on coal surface and in promoting chemical reactions of gasification. Additionally, plasma destroys oils and tars produced during coal devolatilization. One of the main concerns in application of the PAR process to Illinois coal is the coal's high caking propensity, which can create conditions amenable for plugging the process equipment during continuous operation. Prior to starting PAR tests, the feasibility and performance of the micronization process using Illinois coal was studied and compared to the available micronization data obtained with PRB coal, and petcoke. Illinois coal was micronized at three distinct coal feed rates without plugging the equipment due to the caking propensity of the coal. The tests confirm the feasibility of Illinois coal micronization within the PAR technology. Micronization performance with Illinois coal was similar to that of the PRB coal. A total of eight PAR tests were conducted in the entrained flow reactor with Illinois coal at two process temperatures of 400°C and 500°C and a residence time of 3 seconds. Significant efforts were expended to achieve higher temperatures, but they were unsuccessful due to caking on the surfaces of electrical heaters at heater surface temperatures above 600°C and plugging of the heater passages. One of the alternative approaches to achieve 600°C and 700°C temperatures could be to preheat the coal/steam mixture to 500°C and add small amounts of oxygen to increase temperature. Major reaction products in the PAR tests were hydrogen (60%-80%), carbon dioxide (14%-29%), and carbon monoxide (4%-10%), while concentrations of hydrocarbons were below 3%. Carbon conversion into product gas was between 8% and 27% without plasma and between 14% and 54% with plasma. All conversion results are obtained at the process residence time < 3 seconds. Petrographic analysis of processed char shows the presence of cenospheres, the presence of particles with reaction rims, and an increase in high reflectance particles. Additional effort is needed to close carbon and ash balances in the entrained flow reactor tests. Also, additional tests at higher process temperatures and longer residence times are recommended to increase carbon conversion.

Task 3: Performance analysis of the PAR system based on experimental data has shown advantages of this system in thermal efficiency and production costs compared to conventional gasification. For hydrogen production, thermal efficiency is increased by 13% using coal-fired steam production and by 6% using gas-fired steam production. Decreases in production costs of hydrogen are \$1.63/MBtu for the coal-fired method and \$1.32/MBtu for the gas-fired approach. The other benefits include lower capital costs due to elimination of ASU and shift reactor. Estimated capital costs for ASU and shift reactor are at the level of \$1.7 - \$2.3 \$/MBtu H_2. The other capital and operating costs are of the same order for both technologies and can be estimated [4] at the level of \$13/MBtu. Total costs of hydrogen produced via conventional gasification would be about $19/MBtu, while a PAR-based process would yield $14.5/MBtu that is competitive with cost of hydrogen produced via natural gas steam reforming ($14.8/MBtu). By using gas-fired modification of the PAR process, hydrogen can be produced for $14.8/MBtu, and another valuable product - char that can be used as low-NO_x non-caking fuel [7]. Similar economical benefits are projected for syngas production. Results of the system studies indicate that running a PAR process at coal conversions of 60% to 70% at low temperature and combusting unconverted char to produce steam for the PAR reaction could be most efficient scheme for hydrogen production.