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THE DEVELOPMENT OF COAL-BASED TECHNOLOGIES FOR DEPARTMENT OF DEFENSE FACILITIES

Phase III Final Report

By

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Work Performed Under Cooperative Agreement No. DE-FC22-92PC92162

For
U.S. Department of Energy
National Energy Technology Laboratory
P.O. Box 10940
Pittsburgh, Pennsylvania 15236

By
The Consortium for Coal-Water Slurry Fuel Technology
The Pennsylvania State University
C211 Coal Utilization Laboratory
University Park, Pennsylvania 16802

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DISCLAIMER

EXECUTIVE SUMMARY

The third phase of a three-phase project investigating the development of coal-based technologies for U.S. Department of Defense (DOD) facilities was completed. The objectives of the project were to: decrease DOD's dependence on foreign oil and increase its use of coal; promote public and private sector deployment of technologies for utilizing coal-based fuels in oil-designed combustion equipment; and provide a continuing environment for research and development of coal-based fuel technologies for small-scale applications at a time when market conditions in the U.S. are not favorable for the introduction of coal-fired equipment in the commercial and industrial capacity ranges.

The Phase III activities were focused on evaluating deeply-cleaned coals as fuels for industrial boilers and investigating emissions control strategies for providing ultra-low emissions when firing coal-based fuels. This was addressed by performing coal beneficiation and preparation studies, and bench- to demonstration-scale emissions reduction studies. In addition, economic studies were conducted focused on determining cost and market penetration, selection of incentives, and regional economic impacts of coal-based technologies.

Coal Preparation/Utilization

Research conducted under Phases I and II of this project revealed a number of specific areas where continued and/or more focused effort was required in order to develop more effective, more reliable coal processing systems. The specific objectives of the Phase III research on coal preparation were centered around focused investigations into specific coal-cleaning options and their associated ancillary operations and the integration of processing/cleaning operations for overall system optimization. As in the earlier phases of the project, emphasis was on fine-coal processing for the production of a high quality, micronized coal for dry coal and coal-water mixture (MCWM) applications.

Simulations were performed for open and closed grinding circuit configurations using grinding kinetics theory to determine an optimum grinding circuit for producing MCWMs. Data obtained from investigations from the first two phases of the project, and supplemented by additional experiments carried out in Phase III, were used to perform simulations for both conventional ball milling and stirred-media milling. These simulations clearly illustrated that

single-stage grinding in a conventional ball mill does not produce size distributions with the form required for stable coal-water mixtures. A general two-mill grinding circuit simulator, previously developed at Penn State, was used to evaluate the performance of a two-mill grinding circuit under various operating conditions to obtain design criteria for a full-scale system. Data from the simulator were used to develop a conceptual integrated grinding/cleaning circuit for preparing MCWMs.

Particle size separations were carried out using a high-speed, solid-bowl centrifuge under various operating conditions to classify -100 mesh Upper Freeport seam coal. Centrifuge performance was evaluated in terms of the size selectivity curves and corresponding performance parameters: cut size, sharpness index, and apparent bypass. The results indicated that the centrifuge was able to achieve cut sizes less than 10 μm under nearly all conditions, with sharpness indices generally ranging from 0.45 to 0.60. In most cases, the apparent bypass was less than 0.3.

The solid-bowl centrifuge was also used for density separations using a magnetite-and-water dense medium under various operating conditions for treating -100 mesh coal. Centrifuge performance was evaluated in terms of the clean coal yield and ash content while, for selected tests, partition curves were generated. In nearly all cases, clean coal could be produced at an ash content of less than 7%. The clean coal yields varied over a wide range, with values over 80% possible in some cases.

Density separations were also conducted using a magnetic fluid as the dense medium. The separations were carried out using several laboratory separation cells designed to operate within a Franz electromagnet. The results demonstrated that separations could be made at different densities using both batch and continuous separators. Magnet pole design was found to be critical in achieving a constant medium density.

An integrated centrifugal/froth-flotation system was evaluated, with the aim of extending the size range of fine coal cleaning further. It was found that for a given clean coal yield, the integrated process was able to produce a clean coal with a lower ash content than either dense-medium cycloning or flotation alone.

Surface-based processes such as froth flotation are particularly attractive for deep cleaning of fine coal for MCWM applications because of their relative insensitivity to particle size. Laboratory-scale, batch testing of fine coal flotation, conducted in Phases I and II of the project was extended to continuous, pilot-scale studies. A two-stage, rougher/cleaner circuit with a capacity of 8 gpm was used in these tests. The results indicated that performance in the continuous system roughly paralleled that in the batch tests, but that flotation rates and recoveries were consistently lower. The differences were attributed primarily to low mobility and stability of the froth. Continuous flotation tests were also conducted in a pilot-scale, fully instrumented flotation column. It was found that fine coal with an ash content of 9.4% could be cleaned to about 5% at 75 – 80% yield in a single stage operation under appropriate conditions of air flow rate, solids feed rate and concentration and froth-phase gas hold-up.

Continuous dry processing of fine coal using triboelectrostatic separation was also investigated as an extension of the batch testing carried out under Phases I and II of the project. A specially designed test unit was shown to separate fine coal selectively, but at lower efficiency than in the equivalent batch systems. Design modifications to improve separation efficiency were investigated. Preliminary evaluations of an integrated grinding/dry separation system provided encouraging results.

Emissions Reduction

The studies of emissions from coal-fired industrial boilers, that were initiated in the first two phases of the project, were extended into Phase III and involved fundamental, pilot-scale, and full-scale demonstrations and included: 1) evaluations of NO_x reduction strategies such as low-temperature selective catalytic reduction (SCR) and reburning by injecting a biomass-based product and a MCWM; 2) a pilot-scale investigation of simultaneous SO_2 removal/NO_x reduction technology injecting a biomass-based pyrolysis product into the combustion chamber; 3) a fundamental study of formation/destruction mechanisms of polycyclic aromatic hydrocarbons (PAHs) follwed by demonstration-scale testing; 4) comprehensive studies of trace elements and mercury emissions investigating the effect of coal cleaning and particulate removal devices; 5) the development of a unified methodology for simultaneously measuring trace element emissions and mercury speciation; 6) an investigation of fine particulate matter removal using ceramic filters; and 7) the development of a molecular basket to capture CO_2 emissions from the flue gas of coal-fired boilers.

A low-temperature SCR study was initiated in Phase II with the objectives of identifying and/or devleoping a NO_x reduction catalyst that is compatible with the typical operating conditions and the economic constraints of industrial boilers, establishing the limitations of candidate NO_x reduction catalysts so that their implementation in pilot- and demonstration-scale tests will be straightforward, and identifying maximum allowable transients that the catalyst can be exposed to before losing effectiveness. Bench-scale testing was successfully completed in Phase II and the results were scaled up for pilot-scale testing in Phase III. Pilot-scale tests demonstrated that a low-temperature SCR catalyst can achieve greater than 20% reduction at typical utility of industrial baghouse temperatures. Further work is necessary before this concept would be commerically accepted.

A reburing study was performed to understand the mechanism by which NO_x emissions are reduced when cofiring coal with MCWM. The results confirmed the potential of MCWM as a reburn fuel for significant NO_x reduction as MCWM exhibits reductions similar to that of natural gas. Reburn effectiveness depends strongly upon the reburn zone stoichiometery, which is controlled by the reburn fuel heat input and by the air staging configuration. The results indicated that the gas phase reactions did not contribute significantly to the reduction of gas phase NO_x that was produced in the initial stages of combustion but rather it was reactions with the carbon/char that resulted in the lower NO_x emissions.

A model was developed for NO reduction through homogenous gas phase reactions when a biomass-derived product, made from biomass pyrolysis products that have been reacted with air and a lime/slurry, was used as a reburn fuel. The model was validated through thermal gravimetric analysis and pilot-scale combustion testing. The study showed that the percentage of NO_x reduction using the biomass-derived product could be predicted using the gas phase reaction model. Levels of NO_x reduction were correlated with pyrolysis gas yield.

The PAH work, initiated in Phase II, was continued into Phase III to study the effect of temperature and oxygen concentration on the formation and destruction of PAHs in a laminar-flow reactor and a full-scale industrial boiler. Trends in PAH formation and destruction were observed as a function of temperature and oxygen level in the fundamental study; although, availability of oxygen is the more dominant factor in PAH formation and destruction than temperature, which was observed in both the reactor and boiler. Only napthalene and phenanthrene were detected in the demonstration boiler flue gas whereas the laminar-flow reactor, with decreased air-to-fuel mixing, resulted in significantly higher PAH emissions.

Three mercury and trace element activities were performed, which involved evaluating ceramic filters as a control option for mercury removal, evaluating trace elements emissions from deeply-cleaned coals produced in DOE's Premium Clean Coal Program, and developing a sampling train and methodology capable of simultaneously sampling mercury species and all inorganic trace elements. As part of the ceramic filter evaluation, a long-term demonstration of the filters was performed, similar to the one conducted in Phase II of the project, to assess the technical feasibility of using ceramic filters in lieu of fabric filters.

The ceramic filter chamber was modified after the second phase of the project (first 1,000-hour ceramic filter demonstration) to enhance filter regeneration, a new set of filters was installed, and the system was operated for ≈930 hours. The filters were regenerable; however, the pressure drop continued to increase, which is believed to be primarily due to the plugging of the filter substrate pores following the loss of the membrane through erosion. This is a concern for commercialization in a boiler setting and needs to be addressed by the manufacturer. The ceramic member filter particulate collection efficiencies ranged from 99.80 to 99.86%, which were higher than baghouse collection efficiencies using high-performance bags. The stack particulate emissions were below DOE's LEBS target of 0.01 lb/million Btu; however, the design of the ceramic membrane filters evaluated was not able to meet DOE's HIPPS target of 0.003 lb/million Btu.

The ceramic membrane filters were more efficient in reducing trace elements in the stack gas, accounting for an additional 45 to 61% reduction (excluding mercury) over that removed by the fabric filters. The ceramic membrane filters effectively removed 79% more mercury from the gas phase than the fabric filters, even at elevated temperatures. The use of the ceramic membrane filter is a potential control option for both particulate and gas phase mercury emissions provided the high pressure drop issue can be resolved.

The effect of coal cleaning on trace elements emissions was investigated using cleaned coals from CQ, Inc. (cleaned by a combination of heavy media cyclones, water spirals, and froth flotation) and from AMAX Research & Development (cleaned either by advanced flotation or selective agglomeration) to investigate the basic assumption that there is a direct relationship between the depletion/enrichment of an element in the fuel to its depletion/enrichment in the flue gas. The data from the AMAX fuels indicate that there is not always a direct relationship between the reduction of the trace elements in a fuel and a corresponding reduction in emissions.

By contrast, the data obtained from the CQ Inc. fuels suggested that there was a direct relationship between depletion/enrichment of an element in the fuel and its concentration in the flue gas. These different results suggest that trace element emissions can not be predicted solely from their concentration in the fuel. The interaction of the inorganics within the combustion system often affects the partitioning and concentration of the trace elements in the gas and ash streams. Therefore, the effectiveness of extensive coal cleaning in the reduction of trace element emissions should be determined on an individual basis, taking into account the coal composition and system configuration.

A study was performed to develop a sample train and methodology capable of simultaneously sampling mercury species and all inorganic trace elements EPA has listed as inorganic hazardous air pollutants (IHAPs). The advantage of such a system is that it will enable IHAPs and mercury species to be sampled simultaneously, thereby reducing time and cost for stack gas sampling. The train and metholdolgy developed, referred to as the PSU Method, consist of components of both the Ontario Hydro Mercury Speciation Method and Method 29 (total metals) sampling trains. Of particular interest was whether the differences in the PSU Method train configuration compared to the Method 29 and Ontario Hydro trains compromised the measurement of either or both of the total trace elements or mercury species. This was found not to be the case as statistical analysis of the data verified the use of the PSU Method for measuring trace elements and mercury species in combustion flue gas.

Novel CO_2 "molecular basket" absorbents, based on polyethylenimine (PEI)-modifed mesoporous molecular sieve of MCM-41 type (MCM-41-PEI), were successfully developed for CO_2 removal from combustion flue gas. Carbon dioxide was selectively separated from boiler flue gases by using the adsorbent. The adsorbent adsorbed little N_2 and O_2. The selectivity of CO_2/NO_x was 2.5 for natural gas-fired flue gas and the separation selectivity for CO_2/SO_2 and CO_2/NO_x were 10.7 and 2.86, respectively, for coal-fired flue gas. The desporption of CO_2 was complete; however, very little NO_x and SO_2 desorbed after adsorption indicating the need for pre-removal of NO_x and SO_2 from the flue gas mixture before capture of CO_2 by the PEI based "molecular basket" adsorbent.

Economic Evaluation

The objectives of this activity were to determine cost and market penetration, selection of incentives, and regional economic impacts of coal-based fuel technologies. In addion, DOD's fuel mix was determined and a national energy portfolio constructed that minimizes energy price shock effects.

A market penetration model was formulated to find the equilibrium optimal mix of boiler retrofit technology adoption (to MCWM firing) among a sample population of watertube boilers located in the Pennsylvania counties of Cambria and Indiana (two main coal-producing counties) and, after model development, for an expanded study region of the entire state. It was estimated that only 40% of Pennsylvania's $\approx$7,000 watertube boilers can be considered for MCWM retrofiitting, of which 36 would benefit from adopting the retrofit technology.

A study was performed on the selection of incentives for commercializing the MCWM technology. The study focused on the microeconomic responses generated by the firms to the incentives. The results show that in the existence of a no-risk opportunity, a tax incentive is the best alternative to offer to the industry to induce an increase in their profits.

Community sensitivity to using coal in a power facility was studied using methods which integrated economic valuation with techniques used in psychology to characterize risk perceptions to value the welfare impacts due to the presence of energy production facilities. Results from the study show that an individual's willingness to pay for risk prevention or reduction is a function of their perceptions of the health, environmental, aesthetic, and economic impacts as well as their socio-economic characteristics. Individuals appear unable to distinguish between the probability and the severity of a risk in the manner suggested by the definition of risk. Perceived environmental, health, and aesthetic impacts play a larger role in determining option prices than potential economic impacts, explaining in part why residents may oppose a facility even when it will likely bring economic benefits to an area.

A review was performed to estimate the effects on the U.S. economy and its energy sectors of conservation strategies to reduce CO_2 emissions. The analysis was undertaken with a 20-sector computable general equilibrium model by simulating various responses to command and control, carbon tax, and carbon emission permit policies. The results indicate that the characterization of energy conservation as a no-regrets strategy is too strong. In all of the simulations, energy sectors stand to lose, though, in some cases, not anywhere near as much as would be expected. Each of the simulations of mandated conservation also lead to decline in output and employment for the U.S. economy. In contrast, some of the price-induced conservation repsonse strategies also simulated have a neutral impact on the overall economy.

An activity to examine U.S. military fuel consumption and describe alternative technologies that could be currently or potentially applied to the military in order to comply with U.S. government directives to lower emissions, lower dependence on imported oil, and to become more energy efficient was performed. It was shown that for both power/heat generation and transport applications, there is a variety of possible alternatives that could be applied for military use. Energy use in the military could also be significantly affected by new energy management systems and implementation of modified, more efficient, conventional technology.

Portfolio theory was used to demonstrate how the energy mix consumed in the U.S. could be chosen if the goal is to reduce the risks to the domestic macroeconomy of unanticipated energy price shocks. The results indicate that the electric utility industry is operating very close to the minimum variance position with a risk aversion strategy. In contrast, overall energy consumption in the U.S. is far from an efficient mix. A shift towards coal consumption would reduce price volatility. With the inclusion of potential externality costs, the shift remains away from oil but is towards natural gas instead of coal. To achieve such shifts, policymakers could use regulation or tax incentives to industries to encourage the use of certain fuels. Of course, a minimum risk portfolio does not imply a minimum cost portfolio, and selecting a low-risk portfolio may lead to higher energy costs overall. The costs associated with an occasional energy price shock may be less than the cost associated with energy dependence or a dramatic shift towards coal, synfuels, and other alternative energy sources.

Evaluation of Deeply-Cleaned Coals as Boiler Fuels

Deeply-cleaned coals produced by Cyprus/Amax Research & Development Center were evaluated as industrial boiler fuels. The coals, produced in DOE's Premium Fuel Program using advanced column flotation and oil agglomeration cleaning techniques, were tested by Penn State to evaluate their combustion, emissions, and handling characteristics. The testing at Penn State complemented the Premium Fuel Program, which had the overall objectives of producing an alternative fuel that will emit less toxic emissions, moving advanced cleaning technologies from the laboratory to a fine coal cleaning palnt, and producing highly-loaded MCWMs that are competitive with fuel oil. The Penn State activities included modifiying a MCWM circuit to handle filter cake to produce MCWMs, determining the MCWMs atomization performance, stability, and rheological characteristics, performing bench-scale evaluations of filter cake handleability, and conducting pilot-scale and demonstration-scale combustion tests to evaluate combustion performance and emissions, specifically trace elements.

The objective of the filter cake handleability study was to evaluate the flow characteristics of fine, filter-cake coal using the Jenike shear-testing procedure and to investigate the use of a pilot-scale test bin for validation of the results. The Jenike shear tester was modified to eliminate inherent weaknesses of the system to increase the sensitivity and range of results. Tests were performed using fine, filter-cake material, the corresponding parent coal, and various blends of the two. The results from this work showed that the effective yield locus, a measure of the stresses required to maintain steady flow in a consolidated powder, appears to be relatively insensitive to mixture composition and moisture content. Increasing coarseness due to the addition of the coarser, parent coal caused a slight increase in the frictional resistance to flow, while moisture addition reduced friction, probably by a lubrication effect. The effects of mixture composition, i.e., size consist, on the (static) yield locus, a measure of the stresses needed to initiate flow in a consolidated powder, revealed that blends of the filter-cake material with the parent coal tended to be more cohesive than either of the separate components. This rather surprising result is attributed to enhanced packing in the blends and indicates potential materials-handling problems if coal fines are added to the normal feed coal in an existing system. Increased moisture content increases cohesion, probably through capillary forces, but reduces internal friction, presumably by lubrication. Because of their bimodal size distributions, the coarse/fine blends generally packed to higher bulk density than the separate components. The filter-cake material was slightly compressible – bulk density increases with applied stress – while the parent coal is essentially incompressible. Flow functions for these materials indicated that while the flowabilities of the filter cake and parent coal were relatively insensitive to consolidation, those for the blends tended to decrease as consolidation stresses were increased. It is postulated that consolidation stresses applied to the blends affected the fine component disproportionately, leading to enhanced cohesion despite very little change in overall bulk density. Measurements of wall friction against a steel reference material showed very little variation with mixture composition and lead consistently to angles of wall friction of about 27°.

The filter cakes were prepared into MCWMs that exhibited satisfactory handling and short-term storage properties (the MCWMs were prepared without stabilizers or particle size manipulation) for use as industrial boiler fuels. The combustion performance of the MCWMs was good, with the exception of one of the MCWMs (i.e., prepared from the Taggart seam coal cleaned by column flotation), and combustion efficiencies for a suite of tests ranged from ≈97 to 98%. The filter cake MCWMs performed as well or better than MCWMs prepared in the single- or double-stage grinding circuit. The level of natural gas cofire that was required to achieve these combustion efficiencies was 23 to 30%.

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