Task 3.16 - Low-Cost Coal-Water Fuel or Entrained-Flow Gasification

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**By
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SUBTASK 3.16 - LOW-COST COAL-WATER FUEL FOR ENTRAINED-FLOW GASIFICATION

1.0 INTRODUCTION

Continued interest in gasification technologies has led to the need for more technological advances in the area of fuel cleanup and fuel feed systems, which invariably affect the other components comprised by gasification systems. Some entrained-flow gasifiers require the fuel to be a slurry form or a coal-water fuel (CWF). Recent technological advances at the Energy & Environmental Research Center (EERC) have led to potential means for improving efficiency and air toxics control for gasifiers that utilize CWF. Highly reactive low-rank coals present an attractive CWF gasification feedstock. Hydrothermally treating low-rank coals allows a CWF to be formulated that has an elevated solids content, which reduces the amount of water fed to the gasifier, thereby decreasing the amount of oxygen needed to gasify the coal. Preliminary measurements show that the process would increase the solids content from 53 to 63 wt%, giving a 20% improvement in energy density.

2.0 OBJECTIVES

The specific objective of this research project is to assess the potential process efficiency and pollution control benefits that may result from applying the hydrothermal, or hot-water-drying (HWD), process to low-rank coals as related to entrained-flow gasification systems. Project emphasis is on identifying more efficient coal dewatering and CWF formulation methods prior to gasification.

A favorable estimate of incremental cost for integrated hydrothermal drying depends in part on increasing the particle size of the feed coal from minus 100 mesh to minus 28 mesh for the purpose of simplifying the slurry concentration process. Two options will be reviewed for dewatering or concentrating the processed slurry: 1) depressurization and then concentration with sieve bends or 2) partial dewatering at system pressure with hydroclones. Both have their own merits; sieve bends are a low-cost alternative, while hydroclone application would not require additional pumping sections prior to gasification. Various CWF samples with different particle-size distributions and solids concentrations will be sent to equipment vendors for application review. Also, in-house EERC models will be used to verify the suitability of the fuel for gasification systems.

3.0 HYDROTHERMAL SYSTEMS

The EERC, under the sponsorship of the U.S. Department of Energy (DOE), has developed an efficient nonevaporative hydrothermal process for converting high-moisture low-rank coal into high-energy-density clean CWF containing on the order of 60% dry coal solids finely dispersed in 40% water to form a stable, pourable liquid fuel having a heating value of about 7000 Btu/lb or 12,000 Btu/lb dry. The system that was developed (Figure 1), is capable of continuously hydrothermally treating a couple of tons per day of fine coal. Briefly, the system consists of the

Figure 1. EERC 7.5-tpd HWD pilot plant.

following major unit operations: slurry preparation, pumping, preheat, reaction, pressure letdown, production recovery, and HWD coal dewatering.

The coal to be processed is first pulverized with a hammer mill to less than 20 mesh and then slurried to the desired coal-to-water ratio. A high-pressure pump, capable of pumping highly viscous feed slurries up to 2500 mPa-s is used to deliver the slurry to the preheat section at the desired system operating pressure. Slurry flow rate is controlled using a variable-speed motor.

A series of four heat exchangers is used to preheat the slurry to the desired processing temperature. A double-pipe steam heat exchanger first heats the slurry up to 80°C, whereafter the slurry is heated in a series of three condensing Dowtherm vapor-liquid heat exchangers. The nominal ratings of the electric immersion heaters are 22, 22, and 30 kW, respectively.

The slurry, after exiting the fourth preheater at the desired processing temperature, is then directed to a series of two downflow reactors. Typical slurry outlet temperature is 300°C to 330°C. The process piping is configured to allow using a single reactor to attain a residence time of about 10 minutes at conditions. The reactor is equipped with four 2-kW externally mounted heaters to achieve isothermal temperature control.

After processing at the desired temperature and residence time, the coal slurry is throttled through pressure-reducing valves with a resultant flashing of steam and process gas. The gas-water vapor stream is cyclonically separated from the concentrated HWD coal slurry. It is then drawn through a multipass water-cooled condenser, whereafter the noncondensable process gas phase is sent to a natural gas-fired incinerator fired at 800°C. Condensate is collected for possible recycle.

The EERC currently uses either a solid bowl centrifuge or a recessed filter press to dewater the product slurry, producing damp filter cake and filtrate. For system development for gasification, more advance separation equipment or modifications to coal particle size must occur.

4.0 VENDOR SOLICITATION

Numerous filtration/separation equipment vendors were consulted to determine the more appropriate equipment to use to dewater the hydrothermally treated slurry at pressure and temperature. The vendor response was that more than likely for fine coal dewatering, the line pressure would have to be released, the slurry dewatered, and finally pumped into the slurry gasification systems. After initial dewatering, the product solids may also have to be reconcentrated to the pumpable slurry. The level of dewatering will of course vary dramatically with the type of separation equipment that is selected.

Initial efforts were focused on hydrothermal processing of particle size 28 mesh $\times$ 0 and partial dewatering with sieve bends or hydrocyclones. Both methods represented an economically attractive option, adding only estimated \$0.2/mmBtu and \$0.4/mmBtu, respectively (1). However, technically, these options may not work for this application. Hydroclones, according to two different vendors, would be able to handle the pressure and the particle size, but would not be able to dewater to above 50 wt% solids. Based on previous experience with other slurries, both estimated the limit to be less than 40 wt% solids. The sieve bend application cannot handle the line pressure. Similar to other filtration options, the slurry would have to be depressurized and fed to a system by a centrifugal pump from surge tanks. In addition, a larger particle-size distribution is required for high efficiencies.

5.0 HYDROTHERMAL SYSTEM DEVELOPMENT

Based on the latest round of discussion, the EERC altered the existing hydrothermal system to process larger particle size and elevated temperature. Membrane separation technology has also been a dewatering technology which may work for pressure application. The larger particle size testing required a batch process mode since it is not feasible to continuously heat-treat lump coal in a slurry because of coal particle settling properties and slurry pump and valve limitations. Elevated temperature tests will review ability to reduce trace metals and sulfur fractions in the coal. In addition, the EERC has also issued a provisional patent which extends the EERC hydrothermal technology used to produce slurry fuels from low-grade feed materials. The EERC will also continue to track advancements in filtration/separation techniques which are currently available for use to dewater the slurries at pressure and temperature. A complete equipment description of process equipment will be compiled for the final report due March 31, 1997.

5.1 Larger Particle Size and Extended Temperatures

Both bench-scale and pilot-scale hydrothermal process systems were modified to process larger-particle-size material ($\frac{1}{4}$ -in. $\times$ 0) and higher temperature conditions. The bench-scale modifications included a multigram unit, a 40-cm³ reactor, which was built as an extraction vessel and charged with 10–20 g of coal during experimentation. Bench-scale extractions were performed on the multigram apparatus (Figure 2) that was assembled to increase the scale of the process from

Figure 2. Schematic of the multigram subcritical fluid extraction apparatus.

the milligram to the 10–20-g scale. A pneumatically operated pump capable of continuous pumping water at 10 to 300 cm³/min through the fixed bed of coal was used. Results of multigram testing showed that operating conditions of subcritical water at 350°–420°C and 2300 psig consistently reduced the sulfur content by over 50% when applied to Illinois bituminous coal with yields ranging from 50 to 90 wt%, depending upon temperature.

Scaleup from the multigram unit to the pilot scale was accomplished by reproducing the batch process of the multigram unit in the pilot unit, resulting in an approximately 1000-fold increase in coal capacity. A schematic of the hydrothermal setup in the batch processing mode is shown in Figure 3. A superheater was installed for heating water to final temperature after leaving the Dowtherm heaters and just prior to entering the reactor. Top and bottom frits prevented the coal from leaving the reactor and allowed the water to carry away the extracted sulfur and hazardous air pollutants (HAPs). A water-cooled condenser was selected for collecting the effluent water. Although a less desirable process from the commercial perspective, it had some advantages during the initial stages. Some of those advantages included the following:

- The batch unit provided a much broader range of temperatures (ambient to 500°C) and pressures (ambient to 6000 psig) with which to experiment.
- The batch unit generated smaller amounts of process water containing sulfur and HAPs.
- The batch unit allowed for treatment of larger coal particle sizes.

EERC RT12656.CDR

Figure 3. Schematic of pilot-scale high-pressure and -temperature unit for subcritical water batch mode extraction coal experiments.

Two tests with 25 lb of raw $\frac{1}{4}$ -in. $\times$ 0 North Dakota lignite and Illinois bituminous coal was processed at 400°C using the modified pilot-scale batch system. For these initial tests, water flowed through the fixed bed of coal at a rate of approximately 60 lb/hr for 60 min. The heaters were then turned off, and water continued to flow through the coal bed for 4 additional hours. After the sample had cooled, the solids were recovered and analyzed. Despite reduced water to coal exchanges compared to the multigram system, the results from the pilot scale for both coal types were encouraging. The initial test results indicated more than 55% reduction in sulfur for the Illinois coal. The volatile content of the lignite was reduced from 43 to 15 wt%, and the fixed carbon was concentrated from 48 to 76 wt%. The lignite moisture content was reduced from 32.7 to less than 1 wt%. The lignite-drying test was conducted at too high of a temperature, indicated by the level of devolatilization. This lignite char product had better adsorption characteristics, like activated carbon.

The existing continuous slurry system was also modified to include higher temperatures for extended residence time. The continuous mode allows processing of the fine coal at a reduced residence time. The continuous configuration bypassed the superheater used in batch processing and included a second reactor to increase the residence time to 20 minutes at temperature. In this mode, the pulverized coal was slurried with water, and the final product was collected as a filter cake. Tests were conducted on the two different Illinois coals at temperatures as high as 340°C. Solids recoveries were approximately 85% for both coal types. Periodic samples of water, gas, and product slurry were taken at each condition to determine sulfur and material balance. The volatiles content of the product remained essentially unchanged, and the sulfur level was decreased by more than one-third. In addition, the mercury content was 87% less than in the feed, and the selenium content was 46% less than in the feed.

5.2 Membrane Separation Technology

Through funding made available by Illinois Clean Coal Institute (ICCI), Williams Technologies, Inc. (WTI), and Clark Rajchel Engineering (CRE) recently completed an investigation to produce high-quality coal-water slurries from preparation plant fine coal streams using high-shear cross-flow separation (2). This technology was proposed to replace or enhance conventional thickening processes by surpassing normally achievable solids loadings. Dilute ultrafine (minus 100 mesh) solids slurries can be concentrated to greater than 60 wt % and remixed, as required, with dewatered coarser fractions to produce pumpable, heavily loaded coal slurries. Since the technology seems to work best with preheated, finely ground slurries, the EERC requested information for the potential application to the hydrothermal system.

According to literature provided by CRE, this dewatering technology utilizes cross-flow membrane separation and heat and feed pulsation to effect a separation of water from the fine-particle slurries. The separation of water from coal-water slurry is effected by pumping the slurry across a fine pore membrane surface. The membrane is vibrated radially at high frequency (50–60 Hz), which prevents the pores from being blinded by the finest slurry particles. The product is a thickened slurry. In previous laboratory work, fine particle slurries approaching 60 wt% coal have been produced (2). Major advantages of the technology include the following:

- It eliminates flocculants required by both filtration and thickening processes.
- It achieves higher solids concentrations than those achievable in conventional thickeners.
- Slurry concentration equipment is compact in comparison to other fine-particle dewatering/thickening equipment. The footprint of the commercial machinery is approximately 8 by 8 ft, including pumps.
- Equipment is simple, requiring maintenance similar to that required by a pump.
- It provides a pumpable fuel for coal gasification and combustion technologies.
- It has a high potential for use as a NO_x-reducing reburn fuel.

According to CRE, the system can be modified to treat pressurized and heated slurries in the range of hydrothermal process. CRE has also had discussions with DESTEC on using this technology with their gasification systems. The EERC will continue discussions, and if appropriate, forward coal samples to CRE for pilot-scale testing.

5.3 EERC CWF Advancements

Through its extensive development activities, the EERC has been able to optimize the hydrothermal process to maximize desirable slurry characteristics. These efforts have resulted in valuable knowledge that the EERC considers its property. This information includes, but is not limited to, a provisional patent application that was filed on June 14, 1996, entitled "Methods to Enhance the Characteristics of Hydrothermally Prepared Slurry Fuels." The knowledge considered as proprietary information noted in this patent application includes the effect of shear force and time on the viscosity of hydrothermally treated solid fuel; the effect of temperature on the quality of

coal-water fuel produced by hydrothermal treatment; and the use of mixed feedstocks, including different particle sizes to produce enhanced slurries. The patents and technology identified above have been transferred by the Energy & Environmental Research Center Foundation to benefit the State of North Dakota, University of North Dakota (UND), the EERC, and the inventor and technical staff of the EERC involved in the development. Additionally, the EERC has proprietary knowledge on the scaleup relationship between bench autoclave to pilot- to full-scale systems, and the applicability of carbonized slurry fuels to advanced power systems.

6.0 CONCLUSIONS

- Hydroclones and sieve bends may not be technically feasible for dewatering hydrothermal products.
- Bench- and pilot-scale modifications to an existing continuous hydrothermal system were completed. For the first time, lump coal was treated at temperatures up to 400°C, producing a lower-sulfur, low-moisture lump coal product.
- Pilot-scale subcritical water extraction of fine coal in the continuous mode at temperatures up to 340°C at approximately 2300 psig for extended residence time reduced the sulfur by as much as one-third. In addition, results also indicated removal of more than 87% of Hg and 46% of Se.
- Membrane separation technology was identified as a candidate for dewatering hydrothermal slurries at pressure and temperature.
- Provisional patent was developed by the EERC to advance hydrothermal treatment of low-grade slurry fuels.

7.0 FUTURE WORK

- Complete equipment descriptions.
- If appropriate, prepare small batches of hydrothermally treated slurries for dewatering vendors.
- Issue final report.

8.0 REFERENCES

1. Anderson, C.M. "Low-Cost Coal-Water fuel for Entrained-Flow Gasification," semiannual report for the period Jan. 1 through June 30, 1996, for U.S. DOE DE-FC21-93MC30097; EERC publication, 1996.

2. Ehrlinger, H.P., Harrett, D., Fonseca, A., Maurer, R. "Innovative Process for Concentration of Fine Particle Coal Slurries," final technical report for the period Sept. 1, 1995, through Aug. 31, 1996; DOE Cooperative Agreement DE-FC22-92PC92521 (Year 4), ICCI Project Number 95-1/5.2A-3P.

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1. Program/Project Identification No. DE-FC21-93MC30097		2. Program/Project Title TASK 3.0 ADVANCED POWER SYSTEMS				3. Reporting Period 10-1-96 through 12-31-96																																																																																																								
4. Name and Address Energy & Environmental Research Center University of North Dakota PO Box 9018, Grand Forks, ND 58202-9018 (701) 777-5000		5. Program Start Date 01-12-93																																																																																																												
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11. Major Milestone Status	Units Planned	
	Units Complete	
3.12 Small Power Systems Commercialization Plan	P	
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3.15 Impacts of Low-NOx Combustion Fly Ash and Slagging	P	
	C	
3.16 Low-Cost CWF for Entrained Flow Gasification	P	
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3.17 Hot-Gas Cleanup	P	
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12. Remarks

13. Signature of Recipient and Date

14. Signature of DOE Reviewing Representative and Date

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1. Program/Project Identification No. DE-FC21-93MC30097		2. Program/Project Title TASK 3.0 ADVANCED POWER SYSTEMS		3. Reporting Period 10/1/96 through 12/31/96	
4. Name and Address Energy & Environmental Research Center University of North Dakota PO Box 9018, Grand Forks, ND 58202-9018 (701) 777-5000		5. Program Start Date 01-12-93			
		6. Completion Date 12-31-97			
Milestone ID. No.	Description	Planned Completion Date	Actual Completion Date	Comments	
Subtask 3.12	Small Power Systems Commercialization Plan Develop commercialization plan	3-97			
Subtask 3.15	Impacts of Low-NOx Combustion on Fly Ash and Slagging				
a	Modification of CEPS for Low-NOx combustion and ash deposition	3-96	7-96	Completed	
b	Low-NOx fly ash generation and analysis	6-96	10-96	Completed	
c	Low-NOx slagging tests and analysis	8-96	12-96	Completed	
d	Submit draft topical report and article for publication	3-97		Revised Date	
Subtask 3.16	Low-Cost CWF for Entrained Flow Gasification				
a	Procure samples for vendors	3-97			
b	Technical evaluation for dewatering	3-97		Delayed	
Subtask 3.17	Hot-Gas Cleanup				
a	Screening of PFBC sorbents	6-96	9-96		
b	Optimization of selected PFBC sorbents	12-96	12-96		
c	Optimize conditions for tar-cracking catalysts	9-96			

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