

DOE/MC/23289--T5

DE92 017759

DEVELOPMENT OF
MILD GASIFICATION PROCESS

Quarterly Report
for the Period: April - June, 1988

By:

Charles I.C. Chu
Thomas M. Derting

July, 1988

Work Performed Under Contract No.:
DE-AC21-87MC23289

For:

U.S. Department of Energy
Office of Fossil Energy
Morgantown Energy Technology Center
P.O. Box 880 / 3610 Collins Ferry Road
Morgantown, West Virginia 26505

By:

UCC Research Corporation
P.O. Box 1280 / 103 Thomas Road
Bristol, Virginia 24203

MASTER

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

EXECUTIVE SUMMARY

This report represents the work performed by UCC Research Corporation (UCCRC) from April 1, 1988 to June 30, 1988, under DOE Contract No. DE-AC21-87MC23289, "Development of Mild Gasification Process".

Four shakedown tests were conducted on the mild gasification unit (MGU) during this quarterly period - three utilizing Wellmore #8 bituminous coal and one utilizing lignite as feedstocks. No operational problems were experienced with the non-swelling lignite coal.

The common objective of the three tests conducted using Wellmore #8 bituminous coal was to alleviate the char plugging and discharge problems experienced in earlier MGU bituminous coal tests. From these tests it was found that: (1) filling the bottom 12 inches of the reactor tubes with coarse gravel would prevent plugging of the hot sweep gas cross-over pipes; and (2) the char can be discharged relatively easily if it is discharged hot.

A number of modifications have been made to the MGU during this quarterly period. These include: (1) An additional 6-inch flue stack was installed near the bottom of the furnace to reduce the temperature difference between the top and bottom regions within the furnace and thus provide for a more uniform temperature gradient within the reactor tubes; (2) the gas recycle compressor was dismantled and removed from the MGU and replaced with the original 3/4 horsepower vacuum pump utilized in the early MGU testing. The frequency and severity of the maintenance required by the compressor were the primary reasons for its removal; (3) a hydraulic by-pass device has been installed in the hydraulic lines to the two reactor gates located at the bottom of the reactor tubes in response to high hydraulic pressures breaking the reactor gate supports during the discharge of the char; (4) a slip-stream condensing system has been installed on the off-gas line from the reactor tubes on the MGU. The condenser will allow a representative coal liquid sample to be obtained for each specific test run; and (5) three gas flow meters were also installed on the MGU in a move toward obtaining a material balance. One flow meter was installed in the flare gas exit line, the second in the noncondensable sweep gas recycle line, and the third in the noncondensable gas line on the slip stream condenser. The three flow meters, in conjunction with noncondensable gas and liquid analysis of the slip stream liquids, will provide a good basis for constructing a material balance.

TABLE OF CONTENTS

| <u>Section</u> | <u>Page</u> |
|---|-------------|
| INTRODUCTION | 1 |
| Task 1. Test Plan for Optimization of the Mild Gasification Process | 2 |
| Objective | 2 |
| Discussion | 2 |
| Task 2. Optimization of the Mild Gasification Process . . . | 2 |
| Objective | 2 |
| Discussion | 2 |
| Task 3. Evaluation of Char and Char/Coal Blends as an Industrial Boiler/Blast Furnace Fuel | 5 |
| Objective | 5 |
| Discussion | 5 |
| Task 4. Analysis of Test Data and Preparation of Final Report | 6 |
| Objective | 6 |
| Discussion | 6 |
| REFERENCES | 7 |

INTRODUCTION

Oil currently accounts for over 42% of the total U.S. energy consumption and over 40% of the nation's oil is imported from foreign countries. The remaining oil reserve available in this country constitutes less than 6% of the proven total U.S. recoverable fossil energy reserves (1)*. Total U.S. coal resources are estimated at more than 3.9×10^{12} tons (2). The demonstrated coal reserve alone (the coal reserve that is proven and can be economically mined using present technologies and mining techniques) amounts to 488×10^9 tons. At the current annual U.S. coal production rate of approximately 900×10^6 tons, the demonstrated coal reserve alone will last more than 500 years. In light of this contrast in available coal and oil resources, it is very desirable to make good use of our abundant coal resource in our ever more difficult pursuit of energy independence.

Most of the high-severity coal conversion processes that have been developed or are being developed, are too complicated, too expensive, or both, largely due to their reliance on very severe operating conditions and heavy uses of expensive hydrogen.

While conventional coal devolatilization (or "mild gasification") processes are among the oldest methods for obtaining liquid fuels from coal, they are also technically among the least complex. Mild gasification also has the advantages of higher thermal efficiencies than those of other routes to liquid synfuels from coal. Efficiencies of 85-90% can be expected from mild gasification processes, in contrast to only 50-70% for high-severity, indirect and direct liquefaction processes (3). Recent papers reporting various coal liquid qualities and hydrotreatment requirements also indicate that mild gasification liquids are generally superior in quality to those produced from high-severity coal liquefaction processes and require a substantially lesser degree of hydrotreating (3-8).

However, in the existing mild gasification processes, the relative quantities and properties of the co-products are not optimized to make the technology economically and environmentally viable. Many times, either the liquid yield is too low or the liquid quality is poor; and the main product, char (representing 65-75 wt.% of the coal feedstock), often cannot find its proper marketplace.

Under a previous contract with the Morgantown Energy Technology Center (METC), Department of Energy (DOE) Contract No. DE-AC21-84MC21108, UCC Research Corporation (UCCRC) built and tested a 1500 lb/day Mild Gasification Process Development Unit (MGU). Testing completed under the previous contract showed that good quality hydrocarbon liquids and char can be produced in the MGU. However, the MGU is not optimized. The primary objectives of the current project are to optimize the MGU and determine the suitability of char for several commercial applications. The program consists of four tasks: Task 1 - Test Plan; Task 2 - Optimization of Mild Gasification Process; Task 3 - Evaluation of Char and Char/Coal Blends as a Boiler/Blast Furnace Fuel; and Task 4 - Analysis of Data and Preparation of Final Report. Task 1 has been completed while work continues on Task 2.

* Numbers in parentheses designate references at the end of this report.

Task 1. Test Plan

Objective

The objective of this task is to develop a test plan for optimizing the mild gasification process.

Discussion

The test plan was completed and submitted to the Department of Energy in March, 1987.

Task 2. Optimization of the Mild Gasification Process

Objective

The objectives of this task are to: (A) modify the MGU to optimize the unit operation; (B) conduct parametric tests to determine the effect of process parameters on product (gas, condensible, and char) quantity and quality; and, (C) produce sufficient quantities of char and hydrocarbons in order to evaluate these products in various commercial applications.

Discussion

Four shakedown tests were conducted on the Mild Gasification Unit (MGU) during this quarterly period. The first of these tests was conducted using lignite coal and the remaining three were conducted using Wellmore #8 bituminous coal. No operational problems were observed during the first of these tests using the non-swelling lignite coal. Although the condensible liquid yield was low (approx 4% by weight), all of the MGU components appeared to be functioning satisfactorily.

The primary objective of the second test was to alleviate the char sticking/discharge problems experienced in the earlier tests using Wellmore #8 bituminous coal. It was believed that this condition could be improved by reducing the rate and degree of swelling in the coal bed. This was to be accomplished by: (1) reducing the flowrate of the hot recycle/sweep gas through the coal bed and; (2) shutting off the hot recycle/sweep gas when the coal was in its plastic stage. However, during the second test, the hot recycle/sweep gas was inadvertently allowed to run too long and both the inner and outer regions of the bed had entered the plastic stage before the sweep gas was shut off. The test was stopped and the MGU was allowed to cool. The test was resumed the following day and this time the hot sweep gas was shut off during the time that the coal was in its plastic stage. However, at the completion of the test, difficulties were still experienced with discharging the char from the reactor tubes.

During the third test (Wellmore #8 bituminous coal), the temperature of the exit gas from the reactor tubes was observed to be unusually low (157°F).

This indicated that the crossover pipes between the sweep gas heater tubes and the reactor tubes had plugged. Upon completion of the test, the hydraulic rams were able to discharge the char from one of the reactor tubes - but not the other. The char in the second tube had to be removed manually. The crossover pipes were then examined and found to be laden with gummy char. The crossover pipes were removed, cleaned, and welded back into place.

In order to alleviate future crossover pipe plugging problems, it was decided that the bottom 12 inches of the reactor tubes (approximately 2 inches above the crossover pipe openings) would be filled with coarse gravel. This was designed to prevent the coal from migrating into the crossover tubes during the coal's plastic stage and help to more evenly disperse the hot sweep gas through the coal bed.

The objectives of the fourth test were to determine (1) if the addition of gravel would indeed prevent the coal from entering the crossover pipes, and (2) if charging the coal while the furnace was hot would reduce the char discharging problems. The furnace was preheated to 1200°F before the coal was loaded into the reactor tubes. The rate of hot recycle/sweep gas was also maintained at a much lower rate than that which was utilized in previous test runs. At the conclusion of the test, the furnace was allowed to cool over night. The following day, the hydraulic rams were not able to discharge the char from the cold reactor tubes. However, after the furnace was reheated to approximately 800°F, the char was easily discharged. It was therefore concluded that discharging the char while it is hot in all future MGU tests should greatly reduce the sticking problems experienced in the past. A liquid yield of approximately 6% by weight was obtained in the test run - approximately 2% greater than that obtained in previous test runs. The reactor off gas temperature also was found to be substantially higher than that of the past shakedown test runs, this time reaching 650°F. Filling the bottom of the reactor tubes with coarse gravel seems to have accomplished its intended purpose, as no plugging was found in the sweep gas cross-over pipes at the completion of the test run.

This fourth test (shakedown test #7) was the first test run in which the noncondensable gas stream was sampled and analyzed by the gas chromatograph. The results of the gas analysis are shown in Table 1. It can be seen from Table 1 that hydrogen and methane constitute the bulk of the noncondensable gas stream. Overall, this test run was much improved over earlier shakedown runs.

Table 1.
Gas Analysis* - MGU Shakedown Test #7

| <u>SAMPLE#</u> | <u>H₂</u> | <u>CO+N₂</u> | <u>CO₂</u> | <u>H₂S</u> | <u>CH₄</u> | <u>C₂H₆</u> | <u>C₂H₄</u> | <u>C₂H₂</u> | <u>C₃H₈</u> | <u>TOTAL</u> |
|----------------|----------------------|-------------------------|-----------------------|-----------------------|-----------------------|-----------------------------------|-----------------------------------|-----------------------------------|-----------------------------------|--------------|
| 1 | 30.7 | 8.4 | 2.0 | 0.4 | 35.5 | 4.8 | 13.2 | 3.5 | 0.1 | 98.6 |
| 2 | 35.5 | 6.4 | 2.2 | 0.5 | 37.3 | 4.4 | 11.6 | 1.4 | 0.05 | 99.3 |
| 3 | 30.1 | 15.9 | 4.3 | 0.5 | 34.2 | 4.7 | 8.0 | 1.4 | 0.2 | 99.3 |
| 4 | 29.7 | 14.4 | 4.9 | 0.4 | 35.2 | 5.0 | 8.5 | 1.3 | 0.1 | 99.5 |
| 5 | 27.0 | 13.5 | 5.3 | 0.5 | 34.5 | 8.3 | 10.1 | 2.9 | 0.7 | 102.8 |

* All values are volume percentages.

| <u>SAMPLE #</u> | <u>REACTION TIME</u> | <u>OUTER REACTOR TEMP.</u> | <u>INNER REACTOR TEMP.</u> |
|-----------------|----------------------|----------------------------|----------------------------|
| 1 | 30 MIN | 1302 °F | 426 °F |
| 2 | 69 MIN | 1306 °F | 650 °F |
| 3 | 107 MIN | 1299 °F | 840 °F |
| 4 | 132 MIN | 1308 °F | 952 °F |
| 5 | 157 MIN | 1075 °F | 995 °F |

A number of modifications were performed on the MGU during this reporting period. The features of many of these MGU modifications were presented during the Eighth Annual Gasification and Gas Stream Cleanup Systems Contractor Review Meeting, May 10-12, 1988. An additional 6-inch flue stack was installed near the bottom of the furnace to reduce the temperature difference between the top and bottom regions within the furnace. This bottom flue stack joins the original top flue stack outside the furnace to form one single combined stack. The two flue stack dampers control the distribution of the hot flue gases between the top and bottom of the furnace and thus provide for a more uniform temperature gradient within the reactor tubes.

The gas recycle compressor was dismantled and removed from the MGU and replaced with the original 3/4 horsepower vacuum pump utilized in the early MGU testing. The frequency and severity of the maintenance required by the compressor were the primary reasons for its removal. The 3/4 HP vacuum pump has a much lower capacity than the compressor, and therefore may not be capable of producing a flowrate great enough to operate the MGU in "recycle gas compression" mode. If this is indeed the case, nitrogen gas may be introduced to sweep the coal bed and all of the noncondensable gas from the vacuum pump will be vented to the flare, with the vacuum pump running in vacuum mode. The possibility of obtaining a larger capacity vacuum pump is also being investigated.

A hydraulic by-pass device has been installed in the hydraulic lines to the two reactor gates located at the bottom of the reactor tubes. In both of the last two tests conducted on the MGU, the reactor gate supports inside the char hopper have been broken by the hydraulic pressure exerted on them during the discharging of the char. The hydraulic plungers ("rams") generally require a hydraulic pressure of 800 to 1000 psi to discharge the char, which in these tests, created a force great enough to break the welds holding the reactor gate supports to the bottom of the furnace. The hydraulic by-pass will allow the rams to receive 800 to 1000 psi, while the hydraulic pressure to the reactor gates will not exceed approximately 300 psi.

A slip-stream condensing system has been installed on the off-gas line from the reactor tubes on the MGU. The condenser is basically a 2-stage system consisting of: (1) a 3-gallon canister which will be cooled with a dry ice bath, and; (2) a 1/2-inch diameter coiled copper pipe which will be cooled by an ice and/or dry ice bath. By diverting a fraction of the gas stream from the reactor bed to the slip stream condensing system, a representative coal liquid sample can be obtained from each test run.

Three gas flow meters were also installed on the MGU in a move toward obtaining a material balance. One flow meter was installed in the flare gas exit line, the second in the noncondensable sweep gas recycle line, and the third in the noncondensable gas line on the slip stream condenser. The three flow meters, in conjunction with noncondensable gas and liquid analysis of the slip stream liquids, will provide a good basis for constructing a material balance.

Task 3. Evaluation of Char and Char/Coal Blends as an Industrial Boiler/Blast Furnace Fuel

Objective

The objective of this task is to evaluate the MGU char product in three commercial applications. Tests will be conducted to determine the suitability of char in industrial/utility pulverized coal boilers, stoker coal boilers, and as a replacement for coke in foundry/blast furnaces.

Discussion

No work scheduled during this reporting period.

Task 4. Analyze Test Data and Prepare Final Report

Objective

The objective of this task is to analyze the test data generated during MGU testing and char evaluation. The performance of the individual process elements and overall process, including potential end uses for char, will be evaluated. Recommendations shall be made regarding further research and/or development of this mild gasification process.

Discussion

No work scheduled during this reporting period.

REFERENCES

1. T. R. Scollon, "An Assessment of Coal Resources, "Chemical Engineering Progress, June 1977, pp. 25-30.
2. J. M. Eggleston, "Bituminous Coal Marketing, "presented at the Third U. S. A. - Korea Joint Workshop on Coal Utilization Technology, Pittsburgh, October 5-7, 1986.
3. H. W. Parker, "Liquid Synfuels Via Pyrolysis of Coal in Association with Electric Power Generation," Energy Progress, Vol 2, No. 1, March 1982, pp. 4-8.
4. V. A. Nongbri, L. M. Lehman, and L. T. Wisdom, "Hydro-treatment of Cogas Pyrolysis Oil Via the H-Oil Process," F&P Division Reprints, AIChE, July 1980, pp. 690-715.
5. J. Caspers, R. Van Driesen, K. Hastings, and S. Morris, "Upgrading Solvent Refined Coal Hydrogeneration," F&P Division Reprints, AIChE, July 1980, pp. 768-775.
6. R. L. Graves, S. S. Trevitz, S. S. Lestz, and M. D. Gurney, "Screening Tests of Coal Pyrolysis as Diesel Fuel Extenders," presented at the SAE West Coast International Meeting, August 1984.
7. M. R. Khan and T. M. Kurata, "The Feasibility of Mild Gasification of Coal: Research Needs," DOE/METC-85/4019 (DE-85013625), July 1985.
8. R. L. Graves and E. C. Fox, "Diesel Fuels from minimally Processed Coal Pyrolysis Liquids: Exploratory Investigations," Proceedings, Intersociety Energy Conversion Engineering Conference, San Francisco, August 1984.

END

**DATE
FILMED
9/01/92**