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Development of an Advanced, Continuous Mild Gasification Process for the Production of Co-Products

Quarterly Report
October - December 1993

Glenn W. O'Neal

February 1994

Work Performed Under Contract No.: DE-AC21-87MC24116

For
U.S. Department of Energy
Office of Fossil Energy
Morgantown Energy Technology Center
Morgantown, West Virginia

By
Coal Technology Corporation
Bristol, Virginia

MASTER

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EXECUTIVE SUMMARY

Formed coke experiments to select suitable binder coals were completed. Of five different candidates, a metallurgical coal blend from Koppers and a Sewell Seam coal were found to be good binder coals. Petrographic analysis of the binder coals were used in making the selections. Design of a 1000 pounds per hour continuous formed coke pilot plant is under way. Used equipment will be used wherever possible to lower the cost. Six pyrolyzer test runs were completed. Plugging of the shell and tube light oils condenser occurred for long test runs. A new spray contact condenser is being installed on the pyrolyzer using the original tar trap. Circulating coal liquids should dissolve tars and prevent plugging.

INTRODUCTION

Petroleum currently accounts for over 42% of the total energy consumption in the United States; over 40% of the petroleum consumed in the United States is imported from foreign countries. The remaining oil reserve available in the United States is less than 6% of proven recoverable fossil energy reserves while over 90% of the proven recoverable reserves are coal (1)*. Total coal resources in the United States are estimated to be more than 3.9 trillion tons (2). Just the demonstrated reserves, that is, the deposits that are proven and can be economically mined using today's technologies and mining techniques amount to 488 billion tons. At an annual production rate of 900 million tons per year, the demonstrated reserves alone will last more than 500 years. In view of the very abundant coal reserves and limited petroleum reserves, it would seem prudent to make good use of coal in our evermore difficult pursuit of energy independence.

Devising a continuous reactor system that can deliver a good quality co–products which require only minimal upgrading before being marketed is a major challenge. At present, mild gasification reactor configurations tend to fall into two broad categories: circulating or fluidized bed types characterized by high heating rates (up to 10,000 °C per second, or fixed or moving bed types characterized by slow (on the order of 0.2 to 0.5°C per second) heating rates. Circulating or fluidized–bed types produce high liquid yields at the expense of quality. Fixed or moving–bed types produce better quality liquids but in lesser quantities. An optimum reactor is envisioned as one which avoids the secondary reactions associated with slow heating rates and the quality problems associated with high heating rates. Importantly, an optimum reactor would be capable of processing highly caking coals. The reactor concept under investigation in this effort is an advanced derivative of a reactor once used in prior commercial practice which approaches the characteristics of an optimum reactor.

It is important that a mild gasification reactor interface easily with the subsequent product upgrading steps in which the market value of the products is enhanced. Upgrading and marketing of the char are critical to the overall economics of a mild gasification plant because char is the major product (65 to 75% of the coal feedstock). In the past, the char product was sold as a "smokeless" fuel, but in today's competitive markets the best price for char as a fuel for steam generation would be that of the parent coal. Substantially higher prices could be obtained for char upgraded into products such as metallurgical coke, graphite, carbon electrode feedstock or a slurry fuel

*Numbers in parentheses indicate the reference listed at the end of this report.

replacement for No. 6 fuel oil. In this effort, upgrading techniques are being developed to address these premium markets. Liquid products can similarly be upgraded to high market value products such as high-density fuel, chemicals, binders for form coke, and also gasoline and diesel blending stocks. About half of the non-condensible fuel gases produced by the gasification process will be required to operate the process; the unused portion could be upgraded into value-added products or used as fuel either internally or in "across the fence" sales.

The primary objective of this project is to develop an advanced continuous mild gasification process and product upgrading processes which will be capable of eventual commercialization. The program consists of four tasks. Task 1 is a literature survey of mild gasification processes and product upgrading methods and also a market assessment of markets for mild gasification products. Based on the literature survey, a mild gasification process and char upgrading method will be identified for further development. Task 2 is a bench-scale investigation of mild gasification to generate design data for a larger scale reactor. Task 3 is a bench-scale study of char upgrading to value added products. Task 4 is being implemented by building and operating a 1000-pound per hour demonstration facility. Task 4 also includes a technical and economic evaluation based on the performance of the mild gasification demonstration facility.

TASK 1. LITERATURE SURVEYS AND MARKET ASSESSMENT

Objective

The objectives of this Task are: (1) to identify the most suitable continuous mild gasification reactor system for conducting bench-scale mild gasification studies; (2) to identify the most feasible chemical or physical methods to upgrade the char, condensibles and gas produced from mild gasification into high profit end products; and (3) to assess the potential markets for the upgraded products from this process.

Summary

This task was completed and the Topical Report was submitted and approved by the DOE in January 1988 (3).

TASK 2. BENCH-SCALE MILD GASIFICATION STUDY

Objective

The objective of Task 2 is to study mild gasification in bench-scale reactor(s) to obtain the necessary data for proper design of the one ton/hour mild gasification screw reactor in Task 4.

Summary

After much consideration, it was concluded that it would not be necessary or desirable to build a bench-scale reactor. Instead, data and experience from Dr. David Camp's single screw reactor at Lawrence Livermore National Laboratory provided much useful information for the design of the reactor for this project. In addition, the information available from the literature on the eight years of operation of the Hayes process at Moundsville, West Virginia and the earlier Lauck's screw reactor supplied valuable process design data.

TASK 3: BENCH SCALE CHAR UPGRADING STUDY

A petrographic binder coal study was completed this quarter to help better qualify binder coals. Using the best possible binder coal is crucial to the economics of the continuous coke process as it will allow using a minimum of pitch binder which is very expensive. Five coals were used as binder in coke tests at CTC. These same five coals were sent to Coal Petrographic Associates, Inc. in Pittsburgh for petrographic study and analysis. These coals consisted of Consol-Rend Lake Mine, Maple Creek-Pittsburgh Seam, Koppers, Lady H-Sewell Seam and Pinnacle-Pocahontas No. 3 Seam. The recipe used for the coke from these binder coals was 25% binder coal, 65% char, and 10% tar/pitch. This recipe was chosen because it would magnify the binder coal contribution to the final coke product. The coke was made varying only the binder coal. All other variables were held as close to constant as possible. The tests done on each binder coal were proximate, Gieseler plastometer, Arnu dilatometer, free–swelling index and petrographic analysis.

The work done by Coal Petrographic Associate for CTC is displayed as follows. The proximate and sulfur analyses are shown in Table 1. The results of the Gieseler plastometer, Arnu dilatometer and free–swelling index tests are shown in Table 2. The petrographic analyses are shown in Table 3. The positions of the five coals, relative to isostability curves, are shown in Figure 1. The coke reactivity (CRI) and strength after reaction (CSR) tests done by CTC on coke made from these binder coals are shown in Table 4.

Findings of the petrographic work done are presented below.

Consol–Rend Lake Mine

The Rend Lake sample had very poor rheologic properties (0 ddpm, no dilation and 3.5 FSI). This coal is marginally coking, high volatile (0.70% mean–maximum reflectance) and contains an abundance of reactive coal macerals (89.6%). The Rend–Lake coal produces highly reactive isotropic carbon forms in coked briquettes. The Rend Lake is a poor candidate as binder coal.

Maple Creek–Pittsburgh Seam

The sample of Maple Creek coal has the best rheological properties of the bituminous coals used in this study (29,984 ddpm and 7.5 FSI). The petrographic analysis indicates the Maple Creek sample is a fair coking, high volatile coal in rank (0.86% reflectance), with a moderately low total inert content (22.5%). Generally, coal of this rank produces granular or fine circular anisotropic carbon forms in coke. This type of carbon is less reactive than the carbon forms from the Rend Lake coal but more reactive than the carbon forms from the higher rank coals used in this study.

Koppers Coal

The Gieseler plastometer and Arnu dilatometer tests results (4,824 ddpm and 113% max. dilation) for the Koppers coal are lower than expected for this rank of coal. The sample had a good free–swelling index (7.5). The petrographic analysis of the Koppers coal indicates it should be fairly good coking, high volatile coal (0.94% reflectance), with a moderate total inert content (24.3).

Lady H–Sewell Seam

The Lady H sample is low in ash and sulfur contents (3.85 and 0.85%, respectively) and contains 28.59% dry, ash–free volatile matter. The sample is fairly low in fluidity (1,174 ddpm) and dilatation (±125% max. dilation) and has a free–swelling index (8.5 FSI). The rheologic properties of this sample were low for coal of this rank and were significantly lower than results obtained by our laboratory on previous samples of this coal. Petrographically, the Lady H sample is borderline high/medium volatile coal in rank with a 1.12% mean–maximum reflectance. In addition, the sample is relatively low in total inert content (19.9%).

Pinnacle–Pocahontas No. 3 Seam

The Gieseler plastometer and Arnu dilatometer test results (8 ddpm and 13% max. dilation) are on the low side for this product. The sample has a good free–swelling index (8.0 FSI). Petrographically, the Pinnacle sample is a medium range, low volatile coal in rank (1.61% reflectance), with a moderately high total inert content (27.0%).

CTC went into this study with an open mind to learn from petrographic study of binder coals. The approach by Coal Petrographics was to characterize a wide variety of coal ranks and compositions and relate their properties to their performance as binders in the CTC coke process. Much was learned to characterize binder coal from a petrographic approach. Coal Petrographic Associates recommends that in the future, mid–range coals with medium volatiles (about 1.25% reflectance) having a moderate inert content be obtained for testing.

In the past quarter CTC has used the binder coal data along with the DOE supported Penn State Coal Sample Bank and Database to locate potential binder coal sources. This database is a powerful source in indentifying coals and probably is unequaled anywhere in the world. From data supplied to Penn State over 1,130 coals can be analyzed to meet the required characteristics and bring the potential sources down to a manageable number.

With current equipment, continuous coke has been made which meets and exceeds conventional coke laboratory specifications. To continue to move toward market acceptance, quantities of coke are needed which would allow for furnace testing of CTC coke as well as other DOE funded coke projects.

Preliminary work has begun on an extension of this project which would construct a facility to continuously produce coke. The unique feature of this facility is it would be built with used equipment. A new rotary hearth furnace has been quoted at $1.3 million. If the furnace is half the total equipment, the total equipment cost would then be $2.6 million. Also, if the equipment cost is half the total construction cost, that would put the construction cost of a facility with new equipment at $5.2 million. By constructing the facility with used equipment, the total equipment cost would be approximately $300,000. With modifications to equipment, installation, and shakedown, the total project can be built for approximately $850,000 which is a major savings of government and DOE funds.

Coal Technology engineers are currently researching the used equipment market for the best possible equipment for this project. The primary focus of this search is the calciner. With the help of various used equipment brokers, the entire United States, Canada, and the United Kingdom are being searched. Since no calciner was built for this application, many characteristics have to be considered such as:

- Operating temperature and heat profile capabilities
- Capacity
- Cost of modifications
- Cost of relocating
- Coke product degradation

In addition to the calciner, a used briquetter has been located and price negotiated which will make large briquettes to be used for foundry coke or can be broken to form the irregular shapes of the proper size for blast furnace coke.

Excess government–owned equipment is also being explored for use on this project. Three pieces have been found: a motor control center, a weigh feeder, and a crusher which would be valuable to this project and are in the process of being transferred to it. This excess government–owned equipment inventory is continuing to be investigated to determine what other equipment may be available which would keep the cost of this project as low as possible.

Table 1

Proximate Analysis and Sulfur Content of the Indicated Coal Samples[1)]

Sample I.D.	Volatile Matter daf[2)]	Proximate Analysis, Wt. % (dry) Volatile Matter	Fixed Carbon	Ash	Total Sulfur, Wt. % (dry)
Consol Rend Lake CPA# 18237	36.27	34.20	60.10	5.70	0.80
Maple Creek Pittsburgh Seam CPA# 18240	37.72	35.19	58.10	6.71	1.50
Koppers CPA# 18238	33.59	31.68	62.62	5.70	0.70
Lady H Mine Sewell CPA# 18239	28.59	27.49	68.66	3.85	0.85
Pinnacle Pocahontas #3 Seam CPA# 18241	18.01	17.05	77.62	5.33	0.65

1) As determined by Coal Technology Corp.

2) Dry, ash-free volatile matter

Table 2

Gieseler Plastometer, Arnu Dilatometer and Free-Swelling Index Test Results of the Indicated Coal Samples

Coal CPA#	Consol Rend Lake 18237	Maple Creek Pgh Seam 18240	Koppers 18238	Lady H Sewell Seam 18239	Pinnacle Pocahontas #3 Seam 18241
Gieseler Plastometer					
Maximum Fluidity, ddpm	0	29,984	4,824	1,174	8
Maximum Fluidity Temp., deg C	NA*	441	445	452	485
Softening Temp., deg C	NA	388	402	398	455
Solidification Temp., deg C	NA	484	488	496	507
Plastic Temp. Range, deg C	NA	96	86	98	52
Arnu Dilatometer					
Percent Dilation	-30	+197	+113	+125	+13
Percent Contraction	-30	-26	-26	-29	-27
Initial Softening, deg C	375	369	372	381	429
Initial Dilation, deg C	429	411	426	432	471
Maximum Dilation, deg C	NA	453	456	468	495
Free-Swelling Index	3.5	7.5	7.5	8.5	8.0

*NA = Not Applicable

Table 3

Petrographic Maceral Ccmposition of the Indicated Coal Samples

Coal CPA#		Consol Rend Lake 18237	Maple Creek Pgh Seam 18240	Koppers 18238	Lady H Sewell Seam 18239	Pinnacle Pocahontas #3 Seam 18241
Reactives						
Vitrinoid Type	5	3.9				
	6	39.1				
	7	28.9	10.2	1.3		
	8	6.3	43.7	16.3		
	9		13.7	31.9	0.7	
	10		0.7	15.6	31.4	
	11				32.1	
	12				8.8	
	13					0.7
	14					3.5
	15					25.6
	16					31.8
	17					7.6
Exinoids		3.9	5.7	5.8	3.7	0.5
Resinoids		6.5	0.9	1.5	0.7	0.4
Semifusinoids		1.0	2.6	3.3	2.7	2.9
	Total	89.6	77.5	75.7	80.1	73.0
Inerts						
Semifusinoids		2.1	5.3	6.6	5.5	11.3
Micrinoids		2.5	10.6	12.0	8.5	6.9
Fusinoids		2.5	2.5	2.4	3.6	5.8
Mineral Matter		3.3	4.1	3.3	2.3	3.0
	Total	10.4	22.5	24.3	19.9	27.0
Composition Balance Index		0.37	0.81	0.83	0.67	3.74
Rank Index		2.63	3.21	3.67	4.31	7.09
Mean-Max. Ro in Oil, %		0.70	0.86	0.94	1.12	1.61
Calculated Stability Factor		< 10	38	50	59	58

Table 4

COAL TECHNOLOGY CORPORATION

Japanese Method Coke Reactivity (CRI) and Strength After Reaction (CSR) Testing

Coal	CRI	CSR
Koppers	37.7	34.5
Sewell	38.7	34.4
Pinnacle	38.2	23.1
Maple Creek	45.6	13.3
Rend Lake	52.4	0.0

COAL PETROGRAPHIC ASSOCIATES, INC.

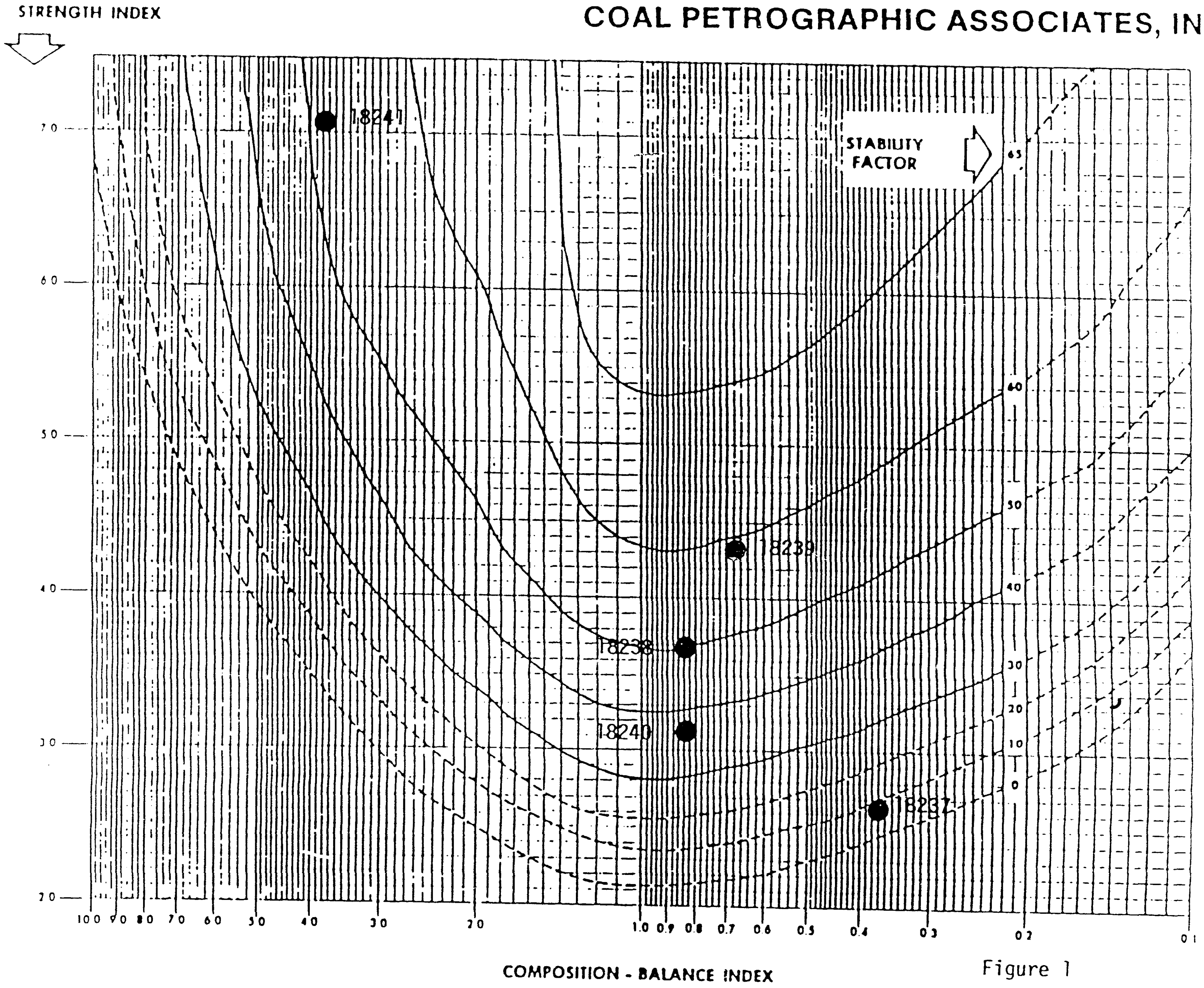

Figure 1

Shapiro and Gray, 1964

TASK 4. 1000 LB/HR CONTINUOUS MILD GASIFICATION UNIT (CMGU)

Test Runs 42–93 through 47–93 were completed during the Fourth Quarter of 1993. Design of a tar trap condenser for the pyrolyzer was a major effort during the Quarter. The original design included a tar trap directly connected to the pyrolyzer which discharged to a packed column for separation of heavy oils. A water cooled, shell and tube, condenser on top of the packed column removed light oils from the pyrolyzer off gases with the non–condensibles going to a flare. The basis for the condenser design was removal of tar before cooling the off gases to prevent plugging of the light oils condenser. The tar separation was not efficient enough to prevent plugging of the light oils condenser during long test runs. A new light oils spray contact condenser is being installed on the existing tar trap. The circulating light oils will dissolve any tars that exit the tar trap and should not plug during long test runs. the new condenser will be completed in January 1994.

Glenn W. O'Neal
Project Manager