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

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Authors:

Wolfe, R.A.
Wright, R.E.
Im, C.J.
Henkelman, M.R.
O'Neal, G.W.
Wolfe, R.A.

Contractor:

Coal Technology Corporation
103 Thomas Road
Bristol City Industrial Park
Bristol, Virginia 24201

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DISCLAIMER

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CONTRACT INFORMATION

Contract Number	DE-AC21-87MC24116
Contractor	Coal Technology Corporation 103 Thomas Road Bristol City Industrial Park Bristol, Virginia 24201 703-669-6515
Contract Project Manager	Robert E. Wright
Principal Investigators	Chang J. Im Michael R. Henkelman Glenn W. O'Neal Richard A. Wolfe
METC Project Manager	Dr. Justin L. Beeson
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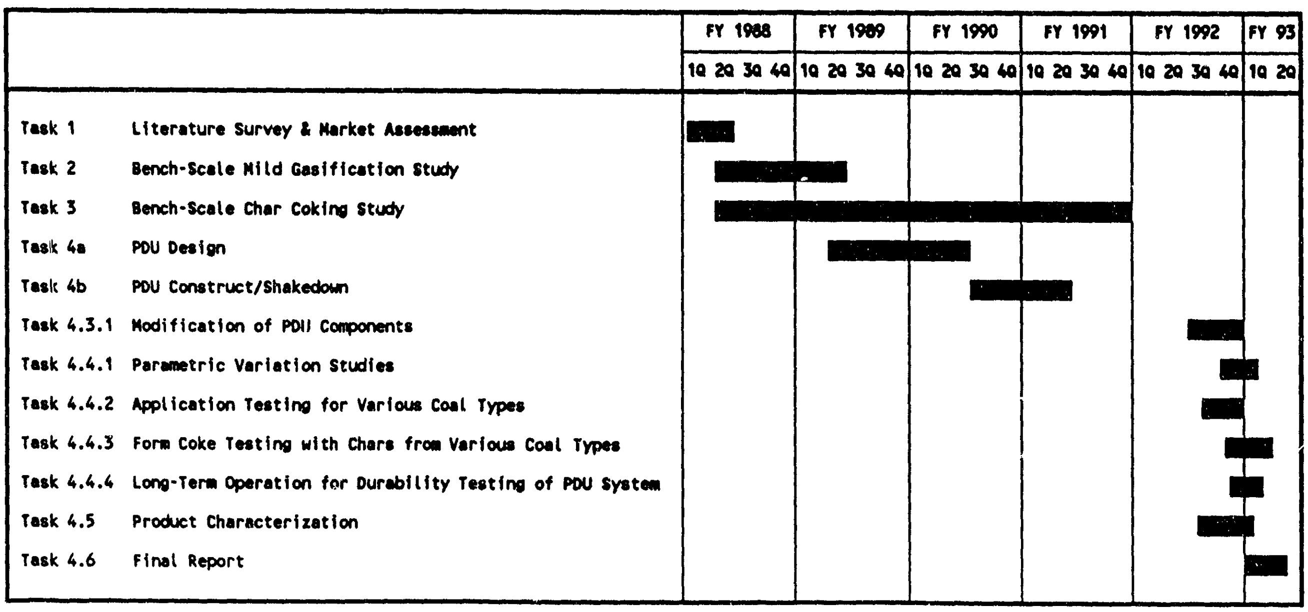

OBJECTIVE

The objective of this project is to develop a continuous mild gasification process to convert highly caking coals to coal liquids , char and coke for near term commercial application.

Coal liquids after fractionation can be blended with petroleum and used interchangeably with conventional fuels without modifications in gasoline and diesel engines.

Char can be used as a carbon source in the production of ferro-alloys and in mini-mills.

Coke can be produced by upgrading char through briquetting and calcining and for use in the steel industry foundries and blast furnaces.

In a step beyond the scope of the project, the plan is to finance, design and construct, in a partnership with others, a plant to produce coal liquids, char and coke in the initial range of 250,000 tons/year.

BACKGROUND INFORMATION

The process for converting coal to char and hydrocarbon liquids is relatively simple and was commercially practiced in the United States (4, 5, 6, 7) in the 1920's and 1930's to make smokeless fuel, a premium product in its day. Coalite (1, 2, 3), a coal derived smokeless fuel, is still being produced in the United Kingdom today in a batch mild gasification process. The Hayes process (5, 6) was self-sustaining in that approximately half of the non-condensible pyrolysis gases were used to supply the heat needed to operate the retort.

In 1984, Coal Technology Corporation (then called UCC Research Corporation) began work under an earlier DOE contract on coal mild gasification via a batch process. This work provided valuable knowledge, but it became evident that a continuous process would be much better.

In the Coal Technology Corporation (CTC/CLC) process, coal is continuously moved by interfolded twin screws through a novel heated retort in the absence of air. The residence time of the coal in the Continuous Mild Gasification Unit (CMGU) is in the range of 30-50 minutes. The coal is heated to controlled temperatures between 800 and 1400 degrees F and is converted into char, condensible hydrocarbon liquids, small quantities of water, and non-condensible fuel gases. The coal derived fuel gases could supply all the required process heat, but for convenience, natural gas is used in the experimental unit. The process concept is particularly suitable for highly caking coals which cannot be processed in fluidized bed or moving bed furnaces.

The present project to develop a continuous process began in September 1987 and consists of four main tasks. Task 1, Literature Survey and Market Assessment, and Task 2, Bench-Scale Mild Gasification Study have been completed. Task 3, Bench-Scale Char Upgrading Study, has been underway since September 1989. In char upgrading studies, "green" uncured char briquettes have been prepared and calcined in 20-pound batches to evaluate the effects of char, binders, and heating conditions on final coke properties. A total of 150 formulations have been tested thus far in this work.

Work on Task 4, PDU Mild Gasification Study, has been in progress since February 1991, with the completion of a CMGU with a design rate of 1000 lb./hr. Since start-up of the CMGU, there have been 72 runs with a variety of operating conditions and coal types.

A paper presented at the Eleventh Annual Gasification and Clean-up Systems Contractor's Review Meeting (9) describes the CTC continuous mild gasification process, the key process items, and the initial operations. Since this information has already been published, it will not be repeated here.

SUMMARY OF WORK IN THE PAST YEAR

At the time of the last contractor's meeting, the CMGU was out of the shakedown period and 12 experimental tests had been made. These tests produced useful data. Basically, the CMGU function is to reduce the volatile content of coal below 10% at which point we consider that coal becomes char with a fixed carbon content in the range from 80% to 92%. It is possible to vary the coal feed rate, the heat input to the reactor, and the residence time in the reactor to obtain the desired char volatile content -- all dependent on the type of coal being processed. This combination of variables also affects the amount and type of coal liquids.

Concurrently, with the CMGU experiments, work was active on coke briquetting experiments. Here, the type and characteristics of the char's parent coal and the char volatile content affects the coke quality, along with the amount and type of coal binder used, and the amount and type of tar and pitch binder. In general, it has been found that a char volatile content of about 8% is desired for the briquetting operation. The CMGU generally has been operated with different coals to obtain char with around 8% volatile content.

Enough work had been done at this time a year ago to be confident that the twin screw process to produce char had the characteristics required for a good commercial process. It was already evident that we could make continuous coke that fully met industry standards. What needed to be done, and which has been done, was to firm up our understanding of the process, to eliminate problems as they became evident, and to improve the process where possible.

We now know very clearly that the CMGU works with any type of eastern bituminous caking coal that we have tested to date. We believe it will work with western coals but have not yet had an opportunity to test western coals. We know now that free swelling index, normally a very important quality for coking coals, is not of any concern in the CLC process. We know that we can operate the system to produce the desired volatile content of the char. We know there are clearly defined markets for the co-products.

ELIMINATING PROBLEMS

(A) The Internal Shaft Heaters - The CMGU as originally designed had an electric heater inside the screw shafts. The concept was also to heat the coal by external radiant gas heat through the screw housing.

Because of the large heat transfer surface provided by the screw flights, the internal shaft heating has almost twice the heating surface of the external heat. Unfortunately, in the early shakedown, the electric heaters failed. The failure was due to: (1) abrasion of the stationary calrod units against the rotating screw shaft because of very close clearances; (2) the difficulty of providing the electric heating unit with insulation adequate to withstand the 1200°F temperature that is required. It appeared that these problems could be overcome by a re-design. However, since recycled process gas heating would clearly be required for economic reasons in the commercial plant design, it was decided to seek an internal gas heater solution. This proved to be a very difficult and time consuming task because of the dimensions, the temperatures required, and the necessity to provide a heater that would not cause hot spots on the screw shafts.

After great effort, the heater that was selected is a gas pulse jet combustor with a combustion efficiency of 95% with a maximum heat input of 300,000 Btu/hr for each burner. This is similar to the pulse jet on the German V-2 rockets except that the V-2 produced thrust while ours will produce a jet of hot gases which can be controlled to produce a given temperature. These gas burners are located at the discharge end of the screws so the heat is supplied counter-currently with respect to the coal/char flow.

At the time of this report, the burners have been installed, they have been given preliminary tests and it seems certain they will perform as planned. These burners will permit higher coal feed rates and better temperature control of the reactor.

(B) Screw Drive Problems - The reactor, as originally designed, has a hydraulic drive capable of producing a forward and reverse motion of the screws. This was done with two thoughts: (1) to permit the forward/reverse ratio to be adjusted by a programmable computer to give a calculated retention time for the coal and char in the reactor; and (2) to obtain a degree of agitation for the coal/char to improve heat transfer.

Runs were made with many different combinations of timing for forward, pause and reverse. A formula was developed for calculating the resultant residence time in the reactor. As this experimentation advanced, it became apparent that this thinking, even though carefully developed, was incorrect. The residence time formula would apply perfectly for a freely flowing material such as sand, gravel, grain or coal without heat. However, in the case of heating coal to 800°F, the formula does not apply. The progress of the coal through the reactor is impeded as the coal passes through the plastic zone at 700°F to 800°F. The coal/char mix in the plastic zone takes on a consistency perhaps somewhat like peanut butter where it becomes difficult for the screws to handle. The coal/char mix dwells in the screws, gradually devolatilizing, with little forward motion. The total residence time in the screws is in the range of 30 to 50 minutes in all experiments without the shaft heater. It is expected that when tests start with the new shaft heater, the residence time will be greatly reduced, the heat transfer rate will be increased and it will be possible to increase the coal feedrate.

Repeated observations show that it takes about 1.5 hours after coal feed is started for a full scale plastic zone to develop. During this time, the power required to drive the screws gradually increases and approaches 40 horsepower. Simultaneously, as the required horsepower increases, the screw speed gradually decreases from 17 RPM to about 12 RPM. At around 1.5 hours after start of coal feed, the power to drive the screws rapidly drops and the screw speed increases. One can visualize and theorize that the dwell in the plastic zone accomplishes devolatilization of the coal which then becomes char which the screw can handle readily with less power required. The theory is that this forming, break-up and reforming of a plastic zone occurs at 1 to 2 hour intervals. This pattern may be altered greatly by the internal shaft heaters now installed.

At this stage of our experimentation, we need to evaluate more completely the forward/pause/reverse action of the screws. Some of the recent tests with a uniform forward drive appeared to work very well. If further work proves this to be the case, a much simpler and cheaper drive can be used on future reactors.

(C) Problems with the Screws - In the early operation stage while the operating procedures were still being learned, one particular incident occurred when the coal feed rate exceeded the power available in the screw drive resulting in the screws stalling. This occurred when the furnace was at the normal temperature of 1400°F. It is believed that stalling at this temperature resulted in bending the screws about 1 1/2 inches out of line measured at the mid-length of the screws. This was corrected by removing the screws and straightening them at a shop equipped with cold bending equipment.

After about 14 months of operation of the CMGU, one of the shafts developed a crack. This may or may not have been due to residual stresses caused by the cold bending described above. The crack may have been due to poor welding practices by the manufacturer of the screws. Because of the low tensile strength at 1400°F, there is a possibility that it could have been a torsion failure. There does not appear to be enough operating hours - only about 650 hours - for it to have been a fatigue failure. In any event, the first crack was repaired. Then, at intervals, four other cracks occurred and each failed on the same screw near the initial crack. Meanwhile, the other screw has had no problems. In the study of possible causes, it has been learned that although the manufacturer had previously made many other screws, the screw with five crack failures was the first of the two made for this project. It is therefore possible that the welding procedures were improved before the second screw was welded.

It became apparent that new screws would have to be made if the project was to continue. New screws were ordered with the shaft thickness almost doubled and with 310 stainless steel instead of 304 stainless steel. Every possible welding procedure was investigated in order to select the best method. The new screws will be installed by August 30 and can then operate with the new shaft heaters.

(D) Condenser Problems - After shakedown, the experimental tests were limited by plugging of the vapor lines from the reactor to the condensers. Usually within five or ten hours, the 4" diameter vapor lines became plugged with carryover of fine coal/char particles impregnated with coal tar. This required the periodic removal of the vapor lines to manually clear the pluggage. The first effort to solve this problem used a periodic purge of nitrogen which helped somewhat. The second step was to insulate the vapor lines which also helped. The third step was to electrically trace the vapor lines plus insulation to keep the lines at about 800°F. This third step solved the plugging problem although the nitrogen purge at half hour intervals has been continued as a precaution.

Another problem related to the condensers was the condensation of water vapor along with the hydrocarbon vapors. The collected coal liquid and water mixture could not be separated in the lab with normal distillation procedures. This problem was solved by insulating and heating the condensers so they operate above 212°F. Under this procedure, the coal liquids condense with less than 1% water content. The water vapors go along with the non-condensible vapors through a second condenser to collect the water and finally to the flare. This procedure completely solved the water separation problem.

Still another problem with the condensers had been the tar collection that occurred and the difficulties of removing the tar from the coal liquids. The shape of the bottom of the condensers has been altered and the bottom section of the condensers will be kept hot with strip heaters. The tar if kept hot separates by gravity from the coal liquids and can be withdrawn from the condenser bottom at intervals. By this method, tar and coal liquids separation can be accomplished. It has been found that this tar is an excellent binder for coke briquettes and will be available for the briquetting in about the amount required for the matching coke operation.

As a result of these condenser operating techniques, the coal liquids may only require very simple refining for use as motor fuels, particularly the diesel fraction.

(E) <u>Coal Bin Feed Difficulties</u> - The CMGU coal bin as designed is cylindrical with steeply sloping cone bottom. This should normally prevent "bridging" and rat-holing." However, all of the coal tested has been from coal preparation plants with a resulting 8-10% moisture content and a considerable fines content. This type of coal requires drying to a less than 5% moisture content to feed well with the CMGU coal bin. To meet this requirement, CTC designed and built an inclined gas heated screw conveyor that tests show is capable of drying typical coal from 11% moisture to 1% moisture at a rate greater than 1000 lb./hr. This then provides a free flowing coal for the CMGU bin, but it also reduces the heat load on the CMGU reactor which will give an increased coal feed rate capability with less water in the condensed liquids.

The problems that have been identified and discussed in this section of the report have all been solved. The CMGU is ready for tests with other coals, at higher feed rates, and with runs of longer duration. The solutions to the problems will result in a better and more economical commercial plant.

<u>GENERAL FINDINGS FROM THE EXPERIMENTAL TEST RUNS ON CMGU REACTOR</u>

The research work on the CTC/CLC process in the past year concentrated on finding the operating conditions to maximize the quality and quantity of the co-products of char, coal liquids and coke.

The CTC/CLC process successfully devolatilized all coals tested. As would be expected, the volatile content of the feed coal affects the yield of co-products, especially coal liquids.

To obtain a low sulfur char or coke, it is necessary to have a low sulfur feed coal since the CTC/CLC process does not remove sulfur.

Ash does not go into the coal liquids so the ash content of the char or coke will be slightly greater than the ash content of the feed coal on a weight basis. Thus, a low ash feed coal is desirable if a low ash char or coke is required.

After shakedown, there has now been a total of 70 test runs. Over this span of operation, a period of 13 months, there is no evidence that the twin screw retort is affected in any adverse way by the 17 different coals that have been tested so far.

The coals involved in test work in the past year are shown in Table 1 below. These are all eastern caking bituminous coals. These coals all performed satisfactorily in the reactor. No western coal has been tested yet. The sulfur and ash contents of these coals are consistent with the requirements for coke and char for the steel and ferro-alloy industries.

Work has been done to produce char for special metallurgical applications. There are indications that the CTC/CLC process yields char with good characteristics for a variety of new markets of a potential size from 200,000 - 500,000 tons per year.

CORRELATIONS FROM THE EXPERIMENTAL CMGU DATA

The CMGU data was carefully examined on Splash Dam, Sewell and Pocahontas (PLC) coals; the coals with the greatest number of test runs. These evaluations were made in an attempt to understand the CMGU process more clearly and obtain some correlations. First order kinetics were assumed for char formation, liquids formations and coal usage. Plots of natural logarithm of the concentration of these species versus time were used to obtain the rate constant.

TABLE 1 COALS USED FROM 8/30/91 TO 8/30/92

		Volatile	Ash	Fixed Carbon	Moisture
Sewell	Sewell	19.61	3.11	73.47	3.82
Beckley	Beckley	15.86	5.41	74.82	3.91
East Ky Metallurgical	-------	28.90	9.25	59.25	2.59
Pocahontas 3	Pocahontas	15.43	6.15	74.60	7.42

One thing that surprises experienced coke plant operators is that the CTC/CLC process accepts coal with any free swelling index. Moisture content is a factor only as related to coal flow through the feed bin and the star feeder. Specifically, a moisture content of 5% or less is desirable to avoid bridging and "rat-holing" in the coal bin feeding the reactor and avoidance of plugging in the star feeder. However, with the new coal dryer, recently installed, the coal moisture is no longer a problem.

For example, (Ln (coal) = kt + C; slope of Ln coal versus t is equal to k.

Since the values for the rate constants for both char and liquids formation are generally far apart, a different mechanism for each reaction is occurring. The coal is reacting through a complicated process.

There is a "lag time" between coal usage and char formation. Rate of char formation is significantly faster than the rate of liquids formation. Again, this implies a complicated process.

Approximately two hours of running time elapsed in each test before "steady state" or equilibrium was reached. Rates of reaction were calculated after two hours of running time. The power required to drive the screws in almost all tests increased to a peak within the first two hours, but varied all during the test. Less power is required for higher fluidity coal.

The correlations listed below were made individually for each coal (Splash Dam, PLC, and Sewell). The results are shown in the accompanying graphs for Sewell coal but are similar for the other coals.

(1) The lower the feed rate, the lower the char volatile content (see Figure 1).

(2) Char volatile content is inversely proportional to the char fixed carbon (see Figure 2).

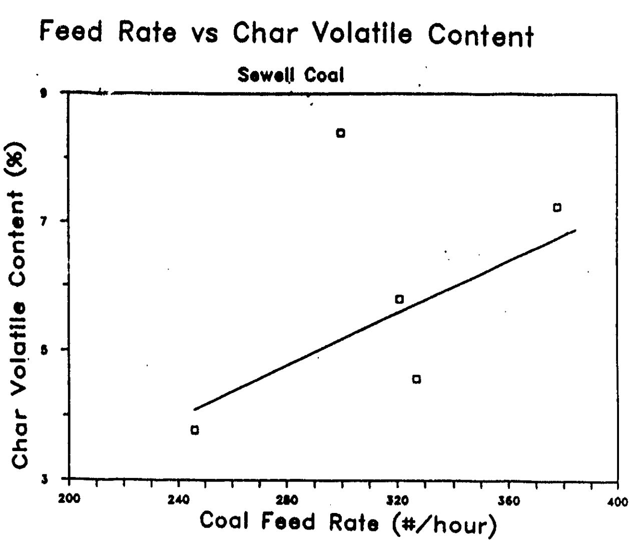

Figure 1

(3) The higher the coal feed rate, the lower the percent fixed carbon in the char (see Figure 3).

(4) The lower coal feed rates result in higher fuel ratios in the char (see Figure 4).

(5) The lower feed rates result in lower char yields (see Figure 5).

It is expected that the data will be better as the operating techniques improve, thus, reducing the scatter shown in the graphs.

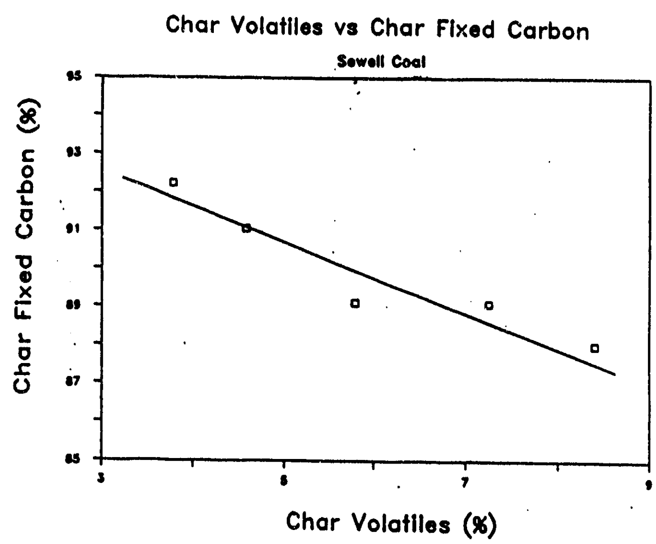

Figure 2

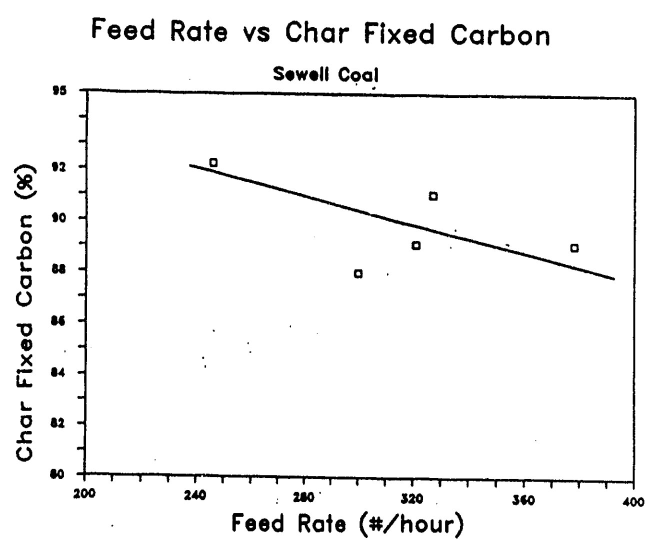

Figure 3

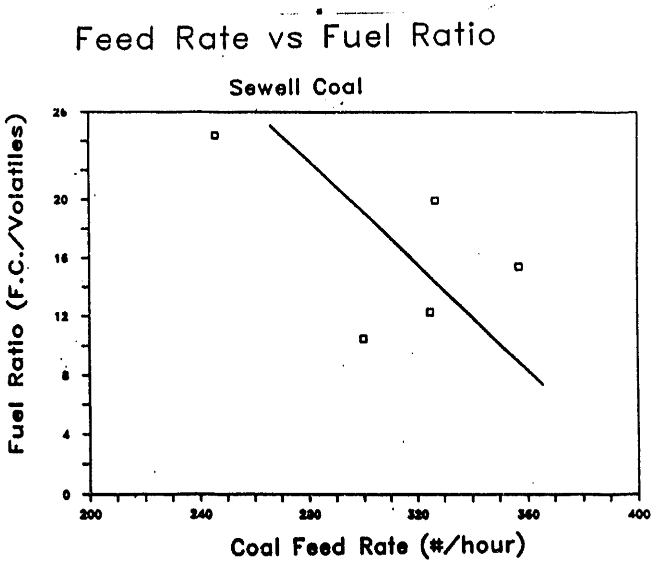

Figure 4

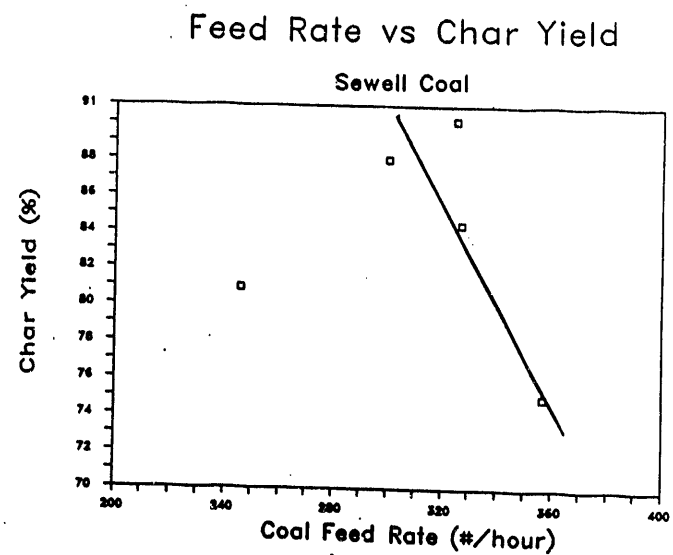

Figure 5

GENERAL FINDINGS FROM EXPERIMENTAL TESTS ON COKING

The research work in the past year on coke briquetting consisted of 27 briquette tests. An important step was the design and installation of the new evaluator for determining the Coke Reactivity Index (CRI).

Coke Test No. 145 on March 24, 1992, produced the best coke briquettes of the entire series of 149 tests. The results for Test No. 145 are shown in Table 2.

Work in the past year shows that there are permutations of char/coal/binder, heating cycle and other variables that require testing to obtain the lowest cost formulation with the best coke specifications.

The coal tar produced as a part of the coal liquids on the CTC/CLC process is an excellent binder for briquetting. The quantity of coal tar from the CTC/CLC process matches what is required. Equivalent purchased coal tar is marketed at $300 per ton so use of the CMGU coal tar enhances the profitability of the CTC/CLC process.

TABLE 2
COKE SPECIFICATIONS VS. BEST TEST RESULTS

	Target Specification	Test Results
Coke Strength Index (CRI)	<30	30.4
Coke Strength After Reaction (CSR)	>55	67.0

The test results are as reported by a third party, UEC-USX Engineers & Consultants, located in Pittsburgh, Pennsylvania.

This data proves that very good coke can be made by the CLC process. There is great confidence that it can be done on a commercial scale.

The first 106 briquette tests were formulated with char made on the batch mild gasification unit (MGU). After the char from the continuous mild gasification unit (CMGU) became available in May 1991, the tests were formulated from that time until now with CMGU char. Up until Test No. 147, there was doubt that the quality of briquettes made with CMGU char was as good as that made with MGU char. This question was resolved with Test No. 147 which proved to make the best coke of all the tests that have been made. A number of hot briquetting tests were made with the conclusion that hot briquetting does not look attractive at this time. The best hot briquettes had a crushing strength about half that of cold briquettes. This factor, and other disadvantages, eliminates the hot briquetting route from further consideration in this current program. Hot briquetting should be evaluated more completely in the future.

Ten of the coke tests were made with a cheaper poorly coking coal with a free swelling index of 2-3. The best coke in this test series had a crushing strength of about two-thirds of the best coke made with coking coal. It is expected that there will be interest in coke made from a non-coking coal at a somewhat reduced price; therefore this testing could be expanded.

The temperature in the calciner will be very important as shown in Table 3.

commercial application. As a result, commercial considerations were included with all technical and operating decisions as the project moved forward.

Information is now available on the probable sales, sales price, plant operating cost, plant construction cost and total capital. This information has enough accuracy to permit a good ten year income analysis. The data shows a very profitable commecial plant can be operated.

FUTURE WORK ON THE CTC/CLC PROCESS

(1) Now that the new pulse jet shaft heaters are installed, it will be necessary to conduct experiments to determine the best combination of heat input from the shaft heaters and radiant heaters.

(2) The next objective after (1) above will be to determine the maximum coal feed rate, hopefully, to meet or exceed the original 1000 lb/hr design capacity.

(3) The effect of operating temperature of the reactor on quality and quantity of the co-products has not yet been investigated. Up until recently, the gas controls on the radiant burners did not permit operation of the reactor temperature much below 1400°F, so all runs to

TABLE 3
EFFECT OF CALCINER TEMPERATURES ON
COKE STRENGTH AND VOLATILE CONTENT

	Coke Crush Strength	Volatile Content
2.5 Hr. Coke @ 1832°F	1500 lbs.	4.76
VERSUS		
1 Hr. Cure @ 850°F and 1.5 Hr. Coke @ 1832°F	2772 lbs.	0.72

COMMERCIAL PROJECTIONS

From the start of this work five years ago, the objective has been to develop the technology of the twin screw mild gasification process for date have been made in the range of 1350°F to 1400°F. Revisions to the gas control system on the radiant heaters and the installation of the new shaft heaters will now permit a wide latitude in the operating temperature of the reactor. The plan is to begin exploratory runs at lower temperatures immediately. A thorough study of the affect of temperature will be conducted if justified by the exploratory runs.

(4) Up to the time of this report, work has been exclusively done using eastern bituminous coking coals. Work in the future is planned to test eastern non-caking coals and western coals.

(5) The present condensers on the CMGU appear to work well in collecting water free coal liquids. The coal liquids comprise light boiling hydrocarbons that are suitable for use in motor fuels and a higher boiling fraction suitable for use as binder in coke briquetting. Methods need to be developed to separate, heat and/or refine the coal liquids to obtain the most profitable co-products.

(6) More test runs need to be made, especially now that the shaft heaters are available, to determine if the forward/reverse action of the twin screws is necessary. If work shows no advantage to the present forward/reverse action, then the hydraulic drive should be replaced with a quieter, simpler and more powerful variable speed electric drive.

(7) With the completion of all or most of the above six items, work will be scheduled for 24 hour operations. The purpose would be to demonstrate stable operation of the system over longer periods. These extended runs will employ the evolutionary operation technique of making small changes to single variables during the run to find the best operating conditions.

FUTURE WORK ON COKE BRIQUETTING

(1) Now that we have the excellent new equipment for determining Coke Reactivity Index (CRI), we will replicate some of the best coke formulations to confirm their CRI. Fine tuning adjustments to the formulations can also be made so that the commercial plant will have the best CRI values at the lowest possible production cost.

(2) Several coals with promising petrographic characteristics will be tested to determine their effect on coke qualities in the CTC/CLC process, both for use to make char and for use as binder to make briquettes.

(3) The present 20 lb batch calcining oven has served well as a screening tool. However, it now should be replaced with a 20-50 lb batch oven that permits rabelling action (turning over the coke) which would more nearly simulate the continuous large scale calciner that would be used in a commercial plant.

(4) The ultimate need is to design and build a continuous 2000 lb/hr coke plant that matches the capacity of the continuous mild gasification unit. This would take care of two needs:

1. The perfection of the present coking process using char as a feedstock; and,

2. To have a continuous 2000 lb/hr unit to evaluate coals. Char formulations using char produced from a number of different DOE sponsored processes. This type of facility would allow the production of sufficient quantities of coke for testing in various industry foundry and blast furnace applications.

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