COAL LIQUEFACTION

QUARTERLY REPORT

JULY – SEPTEMBER 1976

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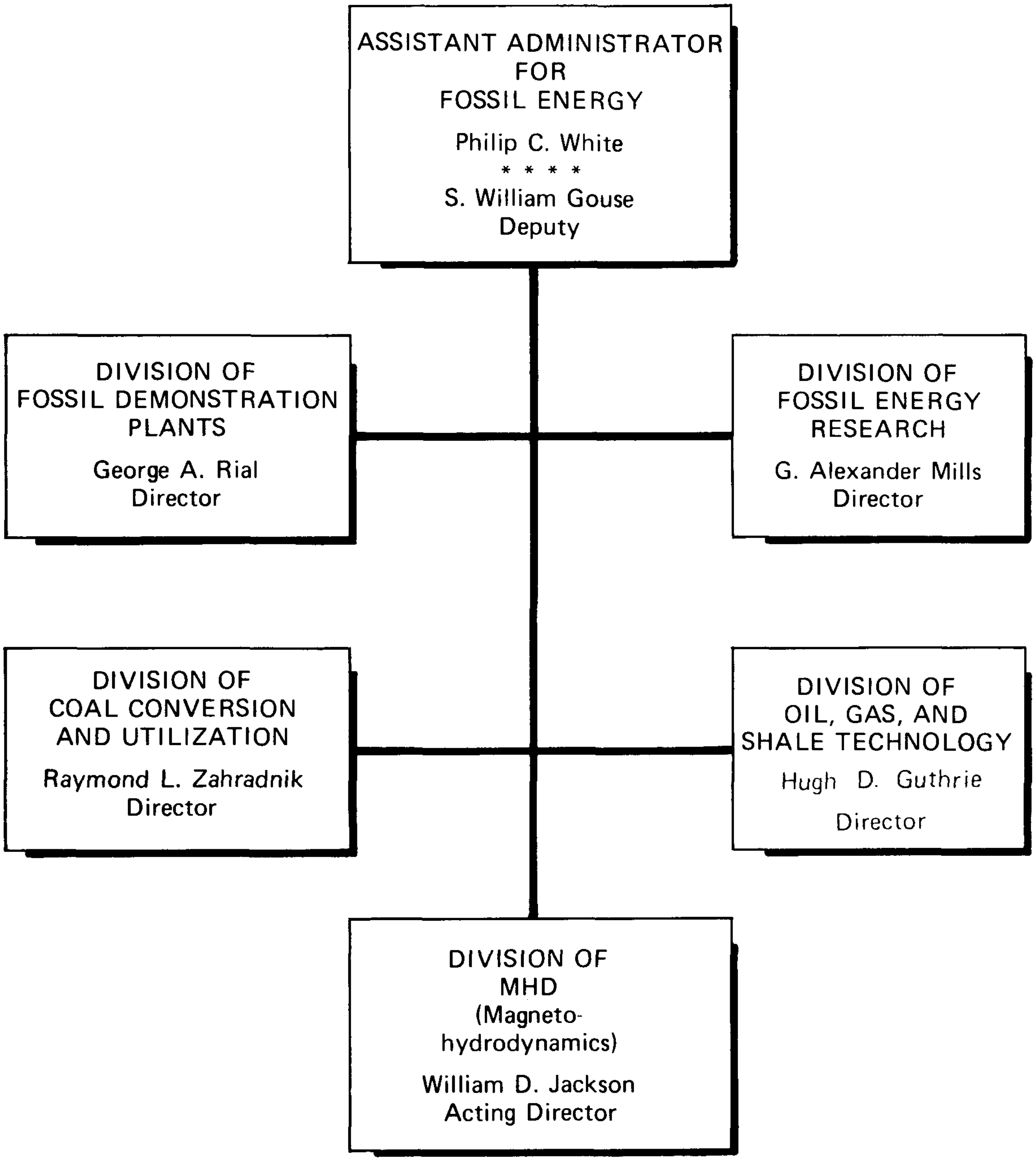
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EXECUTIVE SUMMARY

The United States has more energy available in coal than in petroleum, natural gas, oil shale, and tar sands combined. Nationwide energy shortages, together with the availability of abundant coal reserves, make commercial production of synthetic fuels from coal vital to the Nation's total supply of clean energy. In response to this need, the Office of Fossil Energy of the Energy Research and Development Administration (ERDA) is conducting a research and development program to provide technology that will permit rapid commercialization of processes for converting coal to synthetic fuels and for improved direct combustion of coal. These fuels must be suitable for power generation, transportation, storage, and residential and industrial uses.

The technologies selected for development—gasification, liquefaction, and combustion—satisfy an urgent need for a particular type of fuel, are potentially feasible both technically and economically (in terms of the costs of research and development and the final product) and will not exceed air, water, and solid pollution standards established by the Environmental Protection Agency (EPA). The emphasis given each technology varies, depending on such considerations as technical complexity, development stage (laboratory research including bench-scale tests and experiments with process development units, and pilot plant design, construction, and operation), variety of uses for the fuel produced, and urgency of the need that the technology is designed to satisfy.

ERDA's program for the conversion of coal to liquid fuels was begun by two of ERDA's predecessor agencies: Office of Coal Research (OCR) in 1962, and Bureau of Mines, U.S. Department of the Interior, in the 1930's. Techniques for converting coal to synthetic liquid fuels, originally developed in Germany in the early 1930's, are being improved to increase the supply of easily transportable and usable liquid fuel. Current emphasis is on the development of fuels suitable for firing industrial and electric utility boilers and gas turbines.

Recent improvements have provided better catalysts, reactor designs, and construction materials, and have led to more attractive processing economics and lower capital investment. Coal liquefaction can now be achieved under more moderate processing conditions and more rapidly than was the case in the 1930's. The advantage of coal liquefaction is that the entire range of liquid products, including fuel oil, gasoline, jet fuel, and diesel oil can be produced from coal by varying the type of catalysts and operating conditions used in the process. Furthermore, coal-derived liquid fuels also have the potential for use as chemical feedstocks.

To develop the most efficient means of utilizing coal resources, ERDA is sponsoring the development of several conversion processes, which are currently in different stages of development. Two of these processes are in the pilot plant stage. Responsibility for the design, construction, and operation of the pilot plants is assigned to Pittsburg & Midway Coal Mining Company for the Solvent-Refined Coal plant at Ft. Lewis, Washington and Ashland Synthetic Fuels, Inc. and Hydrocarbon Research, Inc. for the H-Coal pilot plant at Catlettsburg, Kentucky. Fluor Engineers and Constructors, Inc. is responsible for reactivating an ERDA-owned equipment test and evaluation facility in Cresap, West Virginia.

A process development unit is being designed by Foster Wheeler Energy Corporation to test the Synthoil process, which was originally developed by the U.S. Bureau of Mines. Associated laboratory research is being conducted by the Pittsburgh Energy Research Center (PERC), Sandia Laboratories, Argonne National Laboratory, Battelle Memorial Institute (Columbus Laboratories), Exxon Research and Engineering Company, and Morgantown Energy Research Center (MERC). The Clean Coke Process is being developed by USS Engineers and Consultants, Inc., a subsidiary of United States Steel Corporation. The results of laboratory tests and data obtained in the Clean Coke process development unit will be used for intended scale-up. The Rocketdyne Division of Rockwell International is conducting program to develop a technology which would permit the liquefaction of coal with short reaction times by direct hydrogenation in an entrained-flow reactor.

Research to improve solid-liquid separation using filtration techniques is being conducted by Johns-Manville Sales Corporation. The Dow Chemical Company is conducting a research and development program dealing with the use of coal-derived liquids as chemical feedstocks. R&D engineering services, dealing with the development and operation of a fluidized-bed hydrocarbonization reactor and the development of a process to carbonize residua, are being provided by Oak Ridge National Laboratory.

Continental Oil Company, with the participation of Shell Development Company, is developing the zinc halide hydrocracking process, a technique designed to produce clean liquid and gaseous fuels from coal extract and subbituminous coal. Studies are being conducted by the Occidental Research Corporation to evaluate the commercial potential of the company's flash pyrolysis coal liquefaction process. Together with the Electric Power Research Institute, ERDA has contracted with Southern Services Company to operate the Wilsonville, Alabama SRC plant under a variety of conditions and correlate the results with those from the Ft. Lewis, Washington SRC facility.

Using engineering and design technology similar to that practiced in the petroleum industry, the donor solvent liquefaction process is being developed by Exxon Research and Engineering Company. The same firm is also studying the potential of fluid coking to recover distillate liquids from coal liquefaction residues. To facilitate the possible future upgrading of primary coal liquids using technology now available in the petroleum industry, UOP, Inc. is characterizing a wide range of coal liquids. Cities Service Research and Development is determining the operating conditions and modifications necessary to utilize an existing ebullated-bed hydroprocessing unit to upgrade SRC extract.

The ***Solvent-Refined Coal (SRC) Process*** pilot plant operated for 62 days during the quarter. Approximately 1,500 tons of coal were processed, yielding about 750 tons of SRC product. Feed rates ranged from 21 to 32 tons per day with onstream factors of 63 to 71 percent. Plant modifications for the SRC II project are on schedule and ten experimental runs were completed on the SRC II process at the Merriam Laboratory.

Construction continued during the quarter on the ***Synthoil Process*** ten-ton-per-day process development unit. Mechanical completion of the facility is now scheduled for September 1977. Work at the Pittsburgh Energy Research Center included a successful run with a small-volume preheater and the processing of data from a previous 20-day catalyst life test. Efforts at Argonne National Laboratory dealt with additives to facilitate separation of solids from the Synthoil product. Studies at Sandia Laboratories involved catalyst improvement, ash effects, and kinetic studies.

Design activities for the ***H-Coal Process*** pilot plant continued throughout the quarter. Piping progress was over 50 percent complete and work in the coal hydrogenation and offsite areas was nearly finished by the end of the quarter. Tests were also conducted in the process development unit and the catalyst addition and withdrawal system was designed and installed. A 42-hour run was made in the anti-solvent precipitation unit and some PDU experiments were conducted to determine pressure drops across the recycle cup and cup risers.

Renovation of the ***Cresap Test Facility*** continued throughout the quarter. Engineering and design was 98 percent complete and procurement reached 99 percent completion. Work on the test program, as well as the development of test procedures, also continued on-schedule throughout the period. Personnel were hired in Operations, Technical Department, Maintenance and Engineering, and the Administrative group. An extensive training program is currently underway for all operations employees.

Five carbonization test runs were conducted on the ***Clean Coke Process*** during the quarter. Oil from the process development unit was processed in the flash vaporizer and binder preparation PDU to produce clean wash oil, light oils, and tar residues. The hydrogenation PDU and the catalytic rehydrogenator needed for start-up was also operated during the period. The properties and compositions of two synthetic binders were compared with a binder made from stripper bottoms from the hydrogenation PDU.

Twelve tests were conducted in the 500-pound-per-hour reactor for the partial ***Liquefaction of Coal by Direct Hydrogenation***. For the most part, operational difficulties were solved and relatively high conversions were obtained. Construction of the one-ton-per-day reactor system is proceeding rapidly toward a completion date of early November 1976. A draft annual report describing progress during the first year of the program was also prepared and submitted to ERDA for technical review.

Filtration Process and Equipment Studies conducted during the quarter involved 48 filtration runs in the rotary leaf pressure precoat filter. Six of these were made on a sample of Synthoil product and the remainder used Tacoma Filter Feed. Parameters studied during the tests included filter aid pore size, location of the rate-limiting resistance to flow in the filtering system, the effect of body feed, temperature, differential pressure, blade advance, and effect of feedstock agitation. Studies was 40 percent submergence at 5, 15, and 20 second immersion time, shell pressure, and degree of agitation of feedstock. The tests led to the conclusion that filtration rate is favored by higher temperatures and increased pressure differentials.

Studies dealing with the production of ***Chemicals from Coal*** involved the hydrotreating, hydrocracking of various fractions and reforming of hydrotreated naphtha from Synthoil, H-Coal, and SRC products. The results of the earlier tests indicated that some sort of contamination problem still existed even when using the gaseous presulfiding technique. The use of hydrogen sulfide was implemented in an effort to eliminate the problems, and the more recent results indicate that the technique was successful.

R & D Engineering Services dealing with ***Hydrocarbonization*** involved the modification of the bench-scale system to permit recirculating fluidized-bed operation. The reactor was tested on Wyodak subbituminous coal and only minor difficulties were encountered. The solids handling facility underwent shakedown operations during the quarter and approximately 100 pounds of coal was produced for future tests.

Data analysis of previous test results indicated that at 300 psig and 910°–1075° F there was neither significant consumption nor significant production of hydrogen.

R & D Engineering Services dealing with the ***Carbonization of Residue*** included four carbonization runs conducted during the quarter. All of the runs were terminated prematurely when plugs developed in the 1/2-inch line leading from the reactor to the cyclone. Several design changes were subsequently made to alleviate these problems. Two 1110° F agglomeration tests were also conducted with mixtures of Wyodak char and residue in ratios of 2:1 and 9:1. Agglomeration in both tests was slight.

Effort on the ***Zinc Halide Hydrocracking Process*** dealt with the reactivation of an existing continuous bench-scale hydrocracking unit and a continuous fluidized-bed combustion unit for the regeneration of spent halide melt. Initial break-in operation of the continuous bench-scale hydrocracker was done with coal extract.

Various subsystem tests were performed on the ***Flash Pyrolysis Coal Liquefaction Process*** PDU. The tests demonstrated that the plant was operable at normal capacity and process conditions. Work also continued on the modification of the unit to improve char circulation in the pyrolysis system. Characterization of the tar from the initial runs began and a sulfiding procedure for the hydrotreating catalysts was selected.

Operation of the Solvent Refined Pilot Plant at Wilsonville, Alabama was discontinued from July 10 to August 4 for maintenance and modifications. Tests began in early August on Illinois No. 6 coal. Attempts were made during the quarter to improve filtering operation.

Tests continued throughout the quarter on the ***Donor Solvent Liquefaction Process*** 100-pound-per-day recycle coal liquefaction unit. Comparative tests were also conducted on the 50-pound-per-day unit on North Dakota lignite and Illinois No. 6 coal.

Studies dealing with the ***Fluid Coking of Coal Liquefaction Residues*** involved the analytical characterization and bench-scale screening of the coke, liquid, and gas yields from H-Coal solvent precipitation underflow.

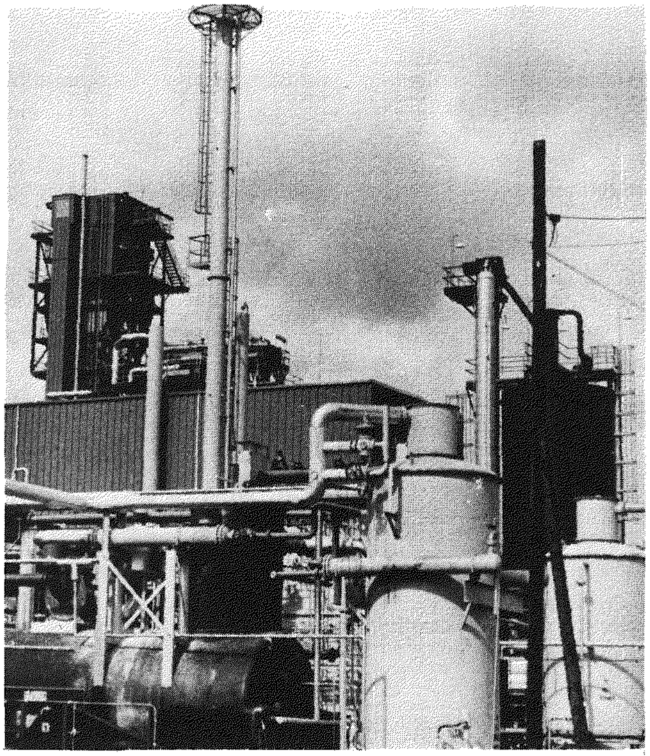
Efforts aimed at the ***Characterization of Coal Liquids*** included the filtering and hydrotreating of six primary coal liquids using a fixed bed of commercial catalyst. In all cases, catalytic hydrotreating substantially eliminated sulfur and benzene-insoluble matter.

Pilot plant studies of the ***Commercial Scale Ebullated-Bed Hydroprocessing of SRC Extract*** included the completion of analytical work on feed components. Preparations have also been made to ready the LC-Fining unit for operation.

I. SOVENT-REFINED COAL (SRC) PROCESS

THE PITTSBURG & MIDWAY COAL MINING COMPANY
MERRIAM, KANSAS

Plant Site: Tacoma, Washington
Contract No.: E(49-18)-496
Total Funding: $82,098,707
(*100% ERDA*)



INTRODUCTION

The SRC process is being developed by Pittsburg & Midway Coal Mining Company (P&M), a subsidiary of Gulf Oil Corporation, under the sponsorship of ERDA. The specific objectives of the contract are to (1) continue obtaining technical and economic data from the pilot plant to validate scale-up to commercial production and (2) provide large samples of solvent-refined coal and by-products from the process for market development studies. Figure I-1 provides a schedule for the development of the process.

PROCESS DESCRIPTION

The SRC process converts high-sulfur, high-ash coals to low-sulfur, low-ash liquid fuel. Figure I-2 shows a schematic of the process. The coal is first pulverized and mixed with a coal-derived solvent in a slurry mix tank. The slurry is combined with hydrogen and is then pumped through a fired preheater and passed into a dissolver. In this unit, the coal is hydrogenated and thereby depolymerized, leading to an overall decrease in product molecular weight and dissolution of the coal. The solvent is also hydrocracked in the dissolver unit, yielding lower molecular weight hydrocarbons ranging from light oil to methane. Yet another reaction occurring in the dissolver is the hydrogenation of the organic sulfur in the coal, thus producing hydrogen sulfide.

From the dissolver, the mixture passes to a separator where the gases are separated from the slurry of undissolved solids and coal solution. The raw gas is sent to a hydrogen recovery and gas desulfurization unit. The recovered hydrogen is then recycled and combined with the slurry coming from the slurry mix tank. Hydrocarbon gases are released and recovered and the hydrogen sulfide is converted to elemental sulfur.

The slurry of undissolved solids and the coal solution is then separated in a filtration unit. In a commercial-scale process, the solids would be sent to a gasifier-converter where they would react with supplemental coal, steam, and oxygen to produce hydrogen for use in the process. The coal solution passes to the solvent

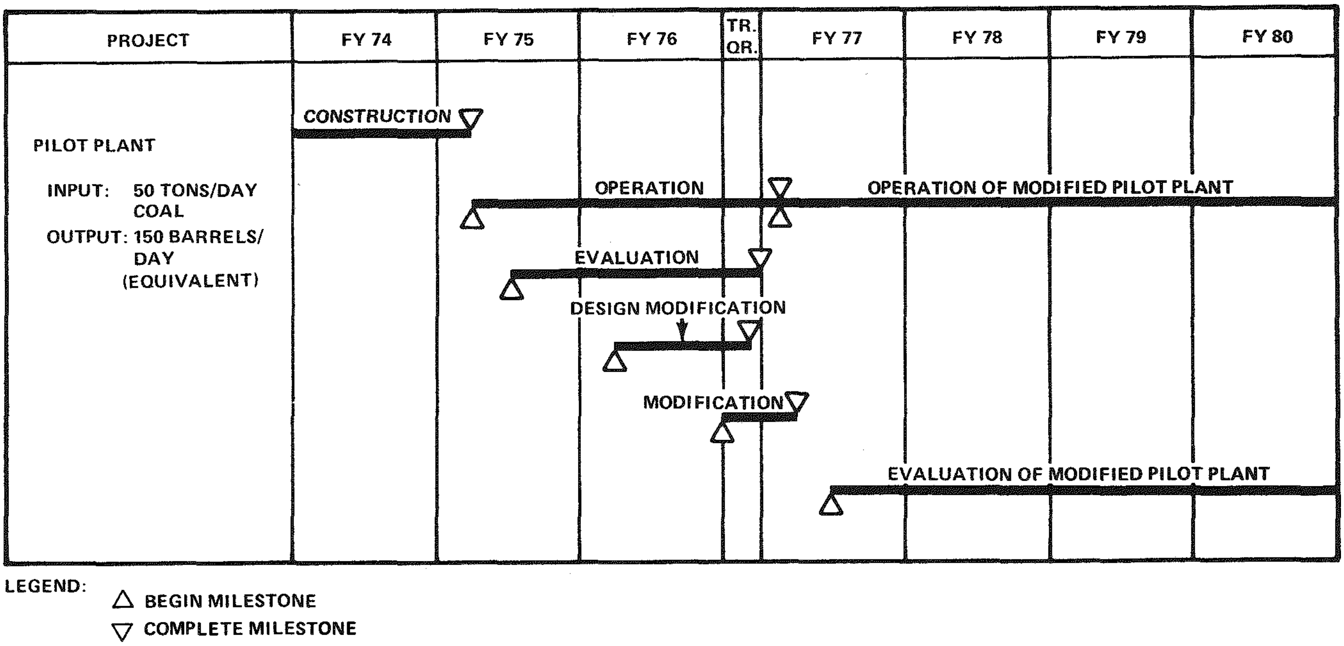


Figure I-1. SOLVENT-REFINED COAL PROGRAM SCHEDULE

recovery unit and the final liquid product, solvent-refined coal, is produced. This material has a solidification point of 350° F to 400° F and a heating value of approximately 16,000 Btu per pound.

HISTORY OF THE PROJECT

The SRC project was begun in 1962 when Spencer Chemical Company was awarded a research contract by the Office of Coal Research (OCR, now part of ERDA) to study the technical feasibility of a coal de-ashing process (now called the SRC process). In 1965, the process was successfully demonstrated in a 50-pound-per-hour continuous-flow unit, and the work on the contract was therefore completed. During the term of the contract, Gulf Oil Corporation acquired Spencer Chemical Company. After reorganization, the contract was assigned to the research department of P&M.

To develop the SRC process further, a contract was awarded to P&M to design, construct, and operate a pilot plant that would be capable of processing 50 tons of coal per day. In 1969, Stearns-Roger Corporation completed the design for the pilot plant but funds to begin construction were not available until late 1971. In June 1972, OCR extended its contract with P&M to construct and operate the pilot plant. Rust Engineering Company began constructing the pilot plant in July 1972 at Ft. Lewis, Washington, near Tacoma. As units were completed, preliminary tests of process equipment and instrumentation were performed. The pilot plant became fully operational in October 1974.

Eleven material balance runs were made on the SRC pilot plant in the distillate solvent mode during 1975. Five successful runs were also made during 1975 in the slurry recycle mode. Overall, the 50-ton-per-day plant operated smoothly throughout 1975.

The laboratory unit was also run in both the distillate solvent and slurry recycle mode during 1975. The variables investigated included filtration rate, solvent recycling, slurry residence time, concentration of coal in the slurry, and temperature.

PROGRESS DURING JULY-SEPTEMBER 1976

Summary

The Ft. Lewis SRC pilot plant operated for 62 days during the quarter, producing 747 tons of SRC. During a 21-day operating period in July, coal feed rate varied from 2,400 to 3,500 pounds per hour (6,400 to 8,700

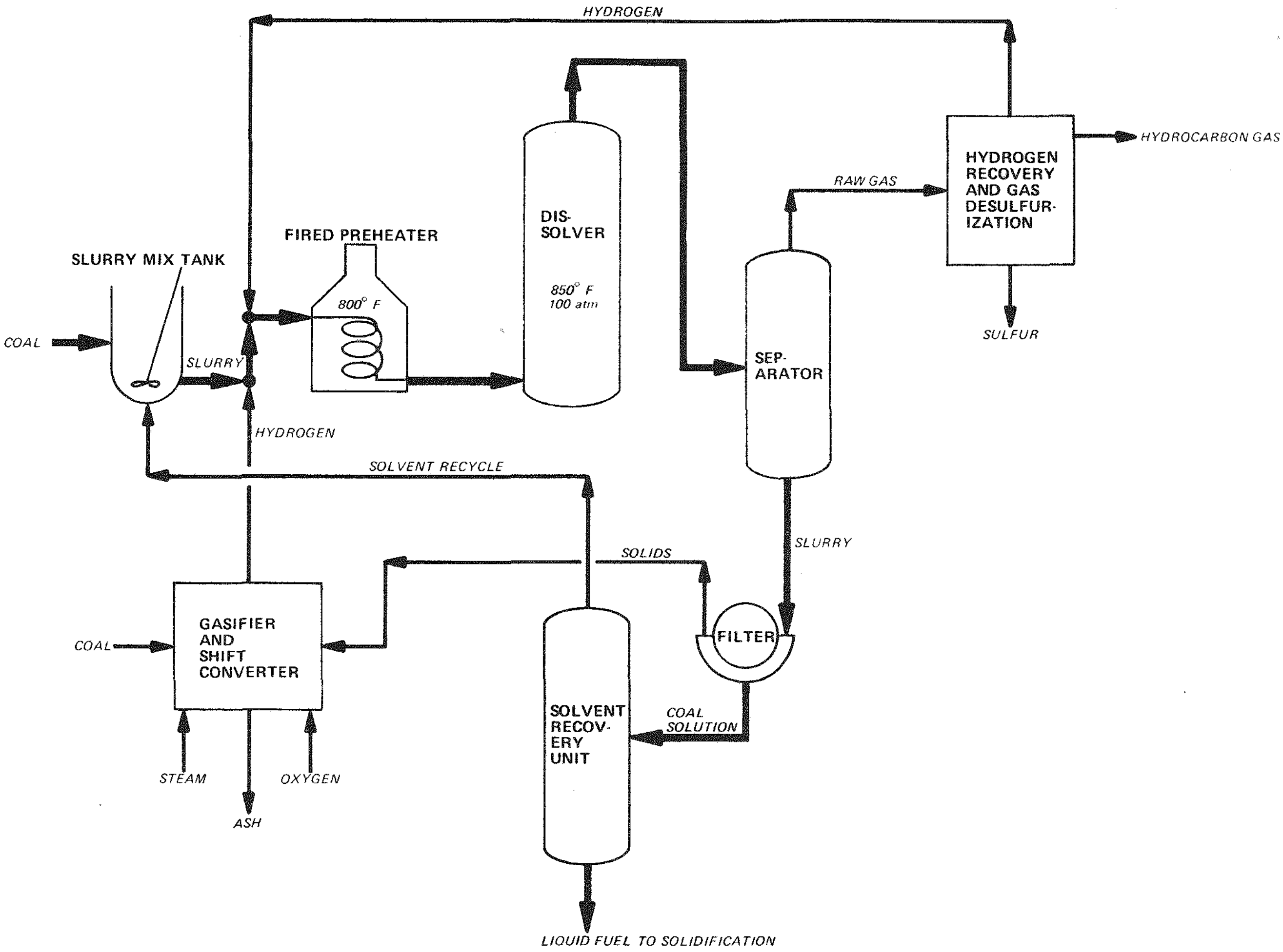


Figure I-2. SRC PROCESS SCHEMATIC

pounds per hour of slurry) at a solvent-to-coal ratio of 1.5 and dissolver operating conditions of 1,500 psig and 850° F. The mineral separation and solvent recovery areas were the major source of plant problems during the month. The plant processed 502 tons of Kentucky bituminous coal during August operations at an average rate of 22.8 tons per day. An on-stream factor of 71 percent was achieved during that period. A total of 612 tons of coal were processed in September at an average feed rate of 32.2 tons per day with an on-stream factor of 63.3 percent.

A series of plant debottlenecking studies were initiated during the quarter. The areas investigated thus far include the vacuum flash system and the light ends column reboiler. Pilot plant modifications for the SRC II project are on schedule, with the engineering, concrete and sewer, and structural steel work being 98, 90, and 75 percent completed, respectively. At the Merriam Laboratory, ten experimental runs were completed in the series designed to develop the SRC II process (slurry recycle).

Pilot Plant Operations

During July, the plant processed coal for 21 days, averaging 21.4 tons of coal feed per stream day. A total of 449 tons of coal were processed at feed rates of 2,400 to 3,500 pounds per hour (6,400 to 8,700 pounds per hour of slurry), a solvent-to-coal ratio of 1.5, and dissolver operating conditions of 1,500 psig and 850° F. The operation yielded 233 tons of specification SRC product.

During the July operations, the mineral separation and solvent recovery areas were the major source of plant problems. Filter "A" produced 2.73 million pounds of filtrate at an average rate of 89 pounds per square foot per hour. Filter "B" processed 0.97 million pounds of filtrate at a rate of 170 pounds per square foot per hour. Filter "A" was opened four times during the month; twice for rescreening, once for screen repairs, and once to clear a cake discharge problem. The Johnson screen on Filter "B" failed and was replaced with a conventional Dutch Weave Screen.

The mineral residue dryer was decoked twice during July and a leak developed in the filter feed surge vessel heat exchanger. Solvent recovery operations were interrupted twice during the month by corrosion-caused leaks in the wash solvent column.

The typical analysis of on-specification SRC product during the July production run was:

Carbon	87.24 wt.%
Hydrogen	5.76 wt.%
Nitrogen	2.13 wt.%
Sulfur	0.73 wt.%
Oxygen (by diff.)	3.93 wt.%
Ash	0.21 wt.%
Heating Value	15,986 Btu/lb. (Dulong)
Fusion Point	335° F (Gradient Bar)

The plant was on stream for 22 days in August, producing over 214 tons of SRC product from 502 tons of Kentucky bituminous coal. The on-stream factor was 71 percent at an average feed rate of 22.8 tons per day. Filter "A" required minor repairs during the month and was rescreened once. Filtrate rate after rescreening averaged 126 pounds per square foot per hour. During the entire operating period, the filter processed 1.8 million pounds for an average rate of 101 pounds per square foot per hour. Filter "B" functioned well during the August runs with no downtime for screen maintenance or repair. Total filtrate processed by Filter "B" was 1.5 million pounds for an average rate of 133 pounds per square foot per hour.

A typical analysis of on-specification SRC during August was:

Carbon	87.06 wt.%
Hydrogen	5.63 wt.%
Nitrogen	2.14 wt.%
Sulfur	0.80 wt.%
Oxygen (by diff.)	4.21 wt.%
Ash	0.16 wt.%
Heating Value	15,860 Btu/lb. (Dulong)
Fusion Point	346° F (Gradient Bar)

During 19 on-stream days during September, the pilot plant processed 612 tons of coal, yielding 300 tons of SRC product. This represented an average feed rate of 32.2 tons per day for an on-stream factor of 63.3 percent. Following the run, an extensive inspection of the vessels and exchangers revealed cracking of the alloy liners in the dissolver, the intermediate pressure flash drum, and recycle condensate separator. None of the cracks, however, were considered serious enough to require replacement at that time. Extensive corrosion was also observed in the wash solvent column and overhead vapor line as well as in the reboilers of both the light ends column and wash solvent column. Replacement may be required in the near future.

The filters were opened five times for repairs during the September operating period. Filter "A" cycles for the month totalled 14, with total filtrate produced at 3.085 million pounds for an average filter rate of 140 pounds per square foot per hour. Eight filtration cycles were completed on Filter "B", producing 0.924 million pounds of filtrate for an average rate of 140 pounds per square foot per hour.

The typical analysis of on-specification SRC product during September was:

Carbon	87.67 wt.%
Hydrogen	5.70 wt.%
Nitrogen	2.22 wt.%
Sulfur	0.75 wt.%
Oxygen (by diff.)	3.52 wt.%
Ash	0.14 wt.%
Heating Value	16,045 Btu/lb. (Dulong)
Fusion Point	330° F

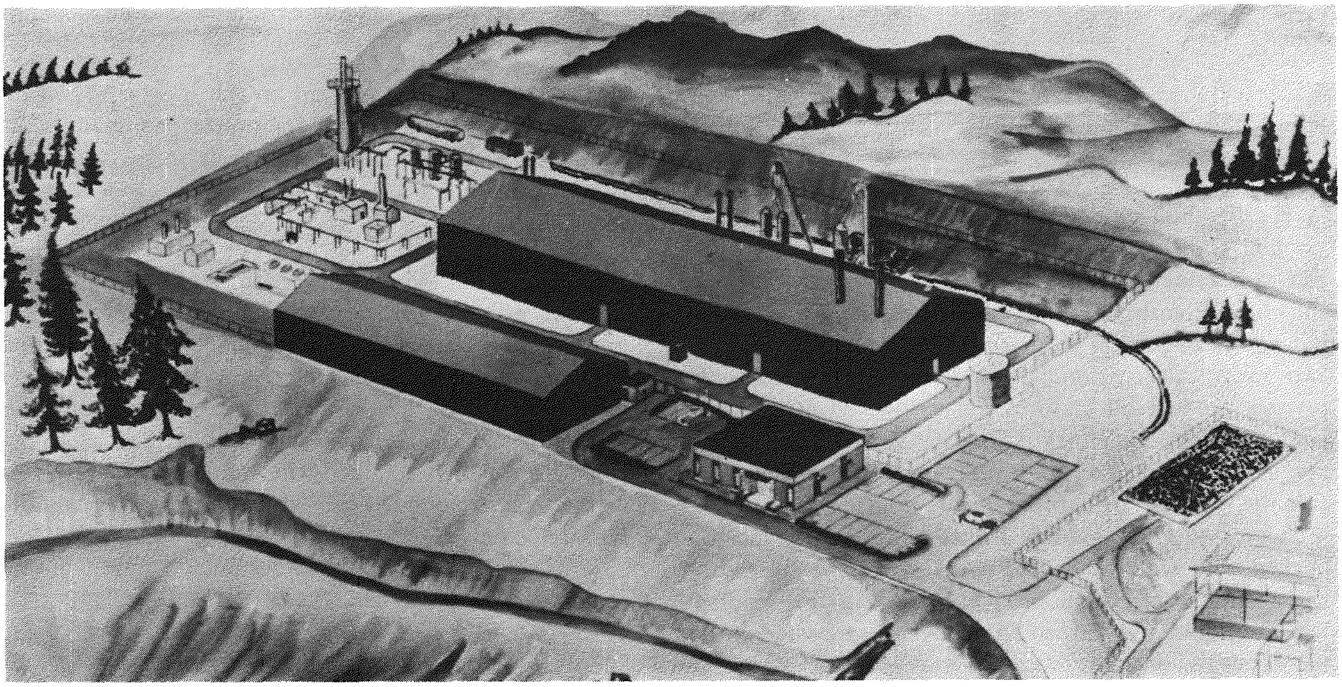
A series of plant debottlenecking studies were initiated in August to identify areas of the process which limit plant throughput. The first test was made around the Solvent Recovery Area light ends column reboiler. It was determined that in order to increase the capacity of this column, either more heat duty must be obtained from the present reboiler, or the column feed must be preheated. Upon completion of this test, a study of the performance of the vacuum flash system was begun. The results of this study showed that the occurrence of "tar" spots on the Sandvik belt were the result of poor distribution on the belt rather than a non-equilibrium condition in the vacuum flash drum.

Pilot plant modifications for the SRC II project were on schedule throughout the quarter. As of the end of September, the engineering, concrete and sewer, and structural steel work was 98, 90, and 75 percent complete, respectively. Procurement of equipment and materials was 77 percent completed.

Laboratory Studies

Ten SRC II runs were completed during the quarter. These tests were a continuation of a series to develop the SRC II process (slurry recycle). The runs provided information on the effect of residence time on product yields. The preliminary results showed that increasing residence time from 1.0 to 1.5 hour resulted in an increased gas yield and decreased distillation residue yield.

II. SYNTHOIL PROCESS



FOSTER WHEELER ENERGY CORPORATION
LIVINGSTON, NEW JERSEY

Total Funding: $24,425,776

SANDIA LABORATORIES
ALBUQUERQUE, NEW MEXICO

Total Funding: $1,636,000

ARGONNE NATIONAL LABORATORY
ARGONNE, ILLINOIS

Total Funding: $1,574,000

BATTELLE MEMORIAL INSTITUTE, COLUMBUS LABORATORIES
COLUMBUS, OHIO

Total Funding: $156,000

EXXON RESEARCH AND ENGINEERING COMPANY
BAYTOWN, TEXAS

Total Funding: $100,000
Plant Site: Bruceton, Pennsylvania

INTRODUCTION

Development of the Synthoil process, initiated by the U.S. Bureau of Mines, is currently being managed by ERDA through the Pittsburgh Energy Research Center (PERC) in Pennsylvania. The objective of this project is to determine the technical and economic feasibility of the process for scale-up to commercial use. Foster Wheeler Energy Corporation is responsible for designing and managing the construction of a ten-ton-per-day process development unit to test the Synthoil process. PERC is conducting support research for the design of the unit. In addition, PERC is monitoring laboratory research on various aspects of the Synthoil process being conducted by Sandia Laboratories, and Argonne National Laboratory. Research being conducted at Exxon Research and Engineering and Battelle Memorial Institute is being monitored by the Morgantown Energy Research Center (MERC) in West Virginia. The schedule for the Synthoil project is shown in Figure II-1.

PROCESS DESCRIPTION

The Synthoil process is a hydrodesulfurization process that converts high-sulfur coal to a low-sulfur, low-ash synthetic fuel oil. A schematic of the process is shown in Figure II-2. The coal is first crushed to 70 percent through 200-mesh and dried to approximately two weight percent moisture. It is then mixed with process product oil to form a slurry of 35-40 percent coal. The resulting slurry is then combined with hydrogen and fed into a fired preheater, which contains ceramic pellets to improve heat transfer. From the preheater, the slurry enters a fixed-bed catalytic reactor packed with catalyst pellets of cobalt molybdate on a silica-alumina base. The product mixture is then cooled and the liquid and unreacted solids are separated from the gases.

Hydrogen propels the slurry so violently through the fixed-bed catalytic reactor that plugging of the bed by the mineral matter in the coal is prevented as the coal becomes liquefied. The turbulence of the slurry promotes mass and heat transfer in the slurry, which in turn promotes hydrodesulfurization and liquefaction.

The liquids and unreacted solids leave the bottom of the separator and pass into a centrifuge. The solids are separated and fed into a pyrolyzer. Part of the liquid leaving the centrifuge is recycled to the mixer and the remainder is drawn off as fuel oil product. The solids are further pyrolyzed to yield additional quantities of product oil and an ash residue. This residue, which contains some carbonaceous material, is sent to a gasifier and shift converter to produce additional hydrogen.

The separator off gases are sent through a gas purification system where they are separated into five product streams: hydrogen sulfide, water, ammonia, hydrocarbon gases, and hydrogen. The hydrogen sulfide is sent to a sulfur recovery system yielding elemental sulfur as the product. The hydrocarbon gases are fed to the gasifier and shift converter. The hydrogen stream which is mixed with the hydrogen produced in the gasifier and fed into the slurry stream entering the preheater.

HISTORY OF THE PROJECT

Work on this process began in 1969 at PERC. The initial work on the Synthoil process used a reactor

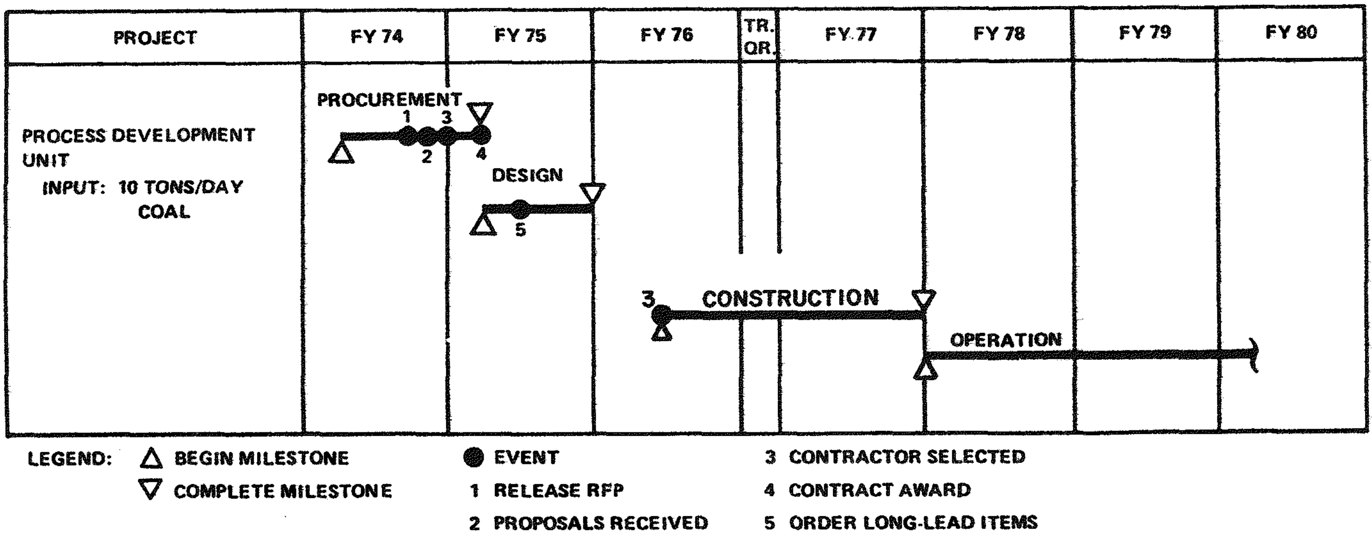


Figure II-1. SYNTHOIL PROGRAM SCHEDULE

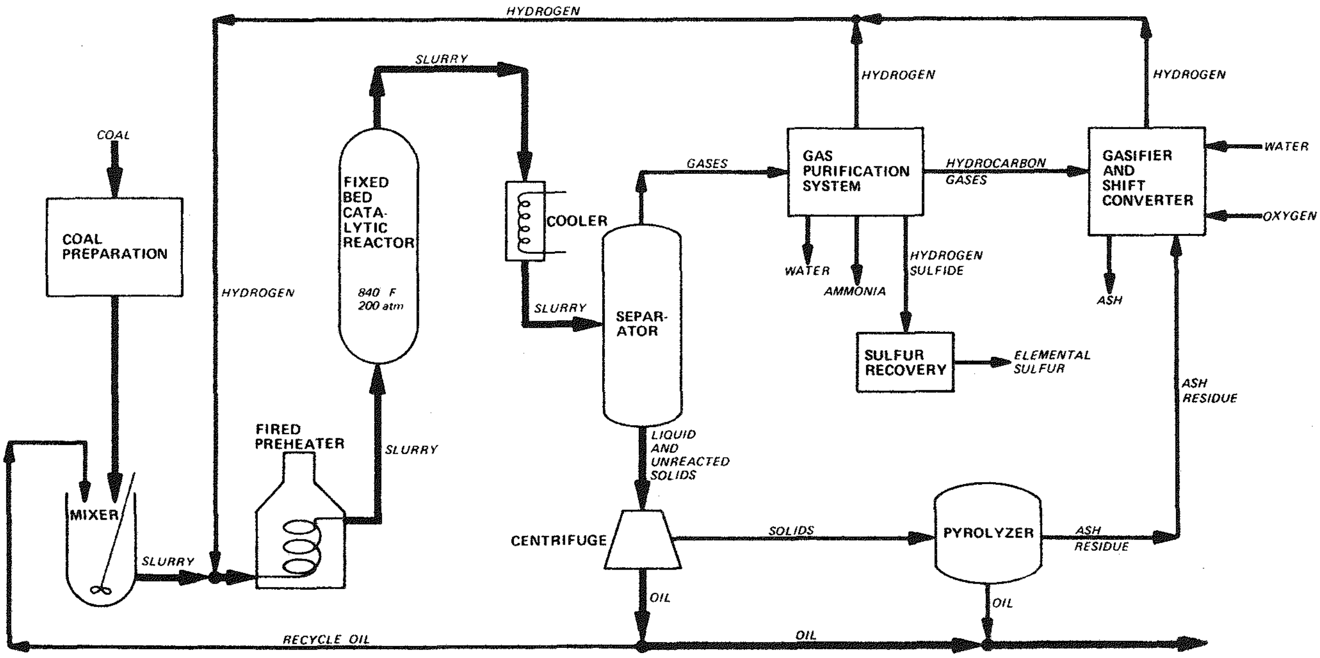


Figure II-2. SYNTHOIL PROCESS SCHEMATIC

with an internal diameter of $\frac{5}{16}$-inch in a bench-scale plant that processed five pounds of slurry per hour. Experimental work was conducted on various coals, such as Pittsburgh seam, Indiana No. 5, Middle Kittenning Ohio No. 6, and Kentucky strip coal. All of these types of coal were satisfactorily converted to low-sulfur fuel oil with no significant attrition of the catalyst. Experiments were conducted to study the effects of hydrogen flow rate, the percentage of coal in the feed slurry, the recycle rate of the product oil, and the effect of hydrogen sulfide in the recycle gas.

To demonstrate the broad applicability of the process, a ½-ton-per-day slurry unit was constructed that used a reactor of 1.1-inch ID made of two interconnected stainless steel pipes that were each 14.5 feet long. The operations were carried out on several types of coal at reactor pressures of 140 atm and 280 atm and 840° F. High yields of low-sulfur and low-ash fuel oil were obtained. At the lower reactor pressure the calorific value, product oil yield, and hydrogen consumption was also lower. However, the sulfur and ash content of the product oil was higher than that of the oil produced at 280 atm. Operation at lower pressure would be favorable if the higher sulfur and ash content were environmentally acceptable. Economic analyses will decide the appropriate pressure for this process, considering the capital and operating costs for high-pressure and low-pressure operations, and the environmentally acceptable sulfur and ash content of the product oil.

The overall design package for the process development unit was completed during 1975. The construction bid packages for both site preparation and plant construction were also completed during 1975. In addition, long lead equipment was placed on order. Actual field construction of the facility began during the fourth quarter of 1975.

PROGRESS DURING JULY-SEPTEMBER 1976

Summary

Construction continued on the ten-ton-per-day process development unit (PDU) during the quarter. Construction is essentially complete on the Administration and Maintenance/Warehouse/Laboratory Buildings and structural steel for the Process Building was erected and siding installed. Mechanical completion of the facility is now scheduled for September 1977.

Process development activities at the Pittsburgh Energy Research Center (PERC) included a successful run with a smaller volume 1.1-inch ID x 42-foot-long preheater. Devolatilization of Synthoil solids on a Sandvik steel belt drying system was also investigated. During this period, a decision was made to terminate filtration studies and concentrate solids separation studies on centrifugation. Other PERC activities included the processing of data from a 20-day run with the miniature catalyst life-testing unit. The results indicated that the catalyst was highly effective during the 472 hours of on-stream time.

Efforts by Argonne National Laboratory dealt with additives to facilitate separation of solids from Synthoil gross product and solvents to remove solids from the coal liquefaction product. Studies by Sandia Laboratories involved catalyst improvement, ash effects, and kinetic studies.

Discussion of Activities

The ten-ton-per-day PDU is being constructed in order to obtain scale-up data for the Synthoil process. Construction is essentially complete on the Administration and Maintenance/Warehouse/Laboratory Buildings. Construction continued on schedule for the Process Building and the Underground Piping and Electrical, with expected completion in late November 1976. Structural steel for the Process Building was erected and siding was installed. Concrete floors were poured and the concrete foundations were completed for the coal preparation and reactor areas. Mechanical completion of the facility is now scheduled for September 1977.

During process development studies at PERC, a successful run was conducted with a smaller volume 1.1-inch ID x 42-foot-long preheater. The preheater was maintained at 450° C and only slurry feed and make-up gas were heated in the unit. The recycle gas was heated separately to 450° F and introduced directly into the reactor, thereby increasing the residence time of slurry in the pretreater. The smaller preheater allowed more product viscosity increase with run time than the large preheater.

Devolatilization of Synthoil solids on a Sandvik steel belt drying system was also investigated during this reporting period. The results indicated that the solids could be devolatilized to a non-sticky, manageable solid by this technique. Installation of such a devolatilization system in the ten-ton-per-day PDU was recommended to convert the solids generated in the PDU to a non-sticky, odor-free substance prior to disposal.

As part of the cold-model studies, a new method was developed to measure liquid residence time of two-phase gas-liquid concurrent upflow in a packed bed. The new method is quicker and more reproducible compared to measuring the difference in weight of a partially saturated and unsaturated packed bed.

Processing of data was completed for catalyst life-test CLT-8, a 20-day run conducted last quarter. Although steady state conditions were not achieved during the run, the results indicated that the Harshaw cobalt-molybdate catalyst was highly effective. A comparison of these test results with those obtained in the ½-ton-per-day unit under comparable conditions with the same coal indicated that steady state results from the life-testing unit will correlate with those from the larger unit. No operations were made in the life-testing unit this quarter, as extensive maintenance and revision work was being performed.

Work also continued on the Synthoil-II ½-ton-per-ton PDU. The preheater was modified to provide greater flexibility in terms of residence time and temperature profiles and the slurry feed system was partially redesigned to allow handling of the coal slurry at higher temperatures with lower pressure drops. A number of components were received during the quarter and specifications were prepared for a variety of others.

Construction of the heat transfer test facility continued throughout the quarter. The model reactor was constructed and the fabrication of other equipment is approximately 95 percent complete. Approximately 80 percent of the instrumentation was received and the installation of the test facility was approximately 75 percent complete by the end of September.

Studies continued at Sandia Laboratories in the area of catalyst improvement. Elemental mapping and stepped point analysis of spent catalyst samples by the electron microprobe were completed. During the study of ash effects, several autoclave runs were conducted with West Virginia coal to establish experimental run conditions, followed by the initiation of tests to determine the reactivities of several different coals. Studies are also underway to find a technique to selectively demineralize coal without affecting the maceral distribution. Construction also continued on the facility for the kinetic studies.

Studies conducted by Argonne National Laboratory dealt with the separation of solids from Synthoil pro uct. Part 1 of the investigation concerned additives to facilitate the separation of solids. A variety of surfactants and agglomerating agents were added to the Synthoil product to determine the effectiveness of these agents in decreasing the viscosity of the product or inducing agglomeration of suspended solids. More than 24 additives were tested, however, only two caused a reduction in the viscosity of the product.

Part 2 of the Argonne studies dealt with the addition of solvents to the product to remove solids by promoting their precipitation. The tests showed the promoter-solvent de-ashing scheme to be a potentially attractive solids-liquid separation method, and kerosene was found to be an effective promoter solvent for the Synthoil gross product.

III. H-COAL PROCESS

HYDROCARBON RESEARCH, INC.
TRENTON, NEW JERSEY

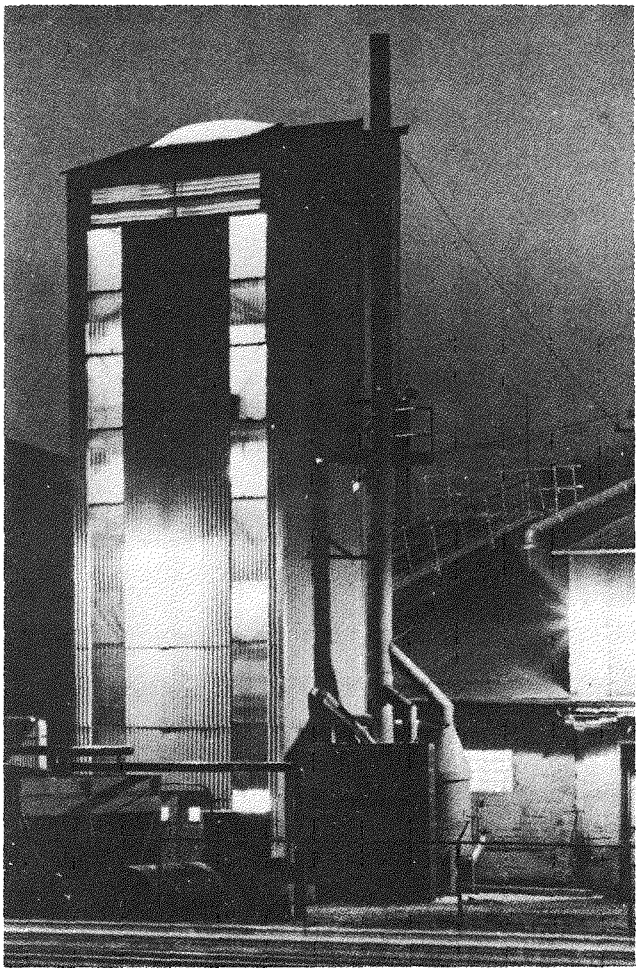
Plant Site:	Catlettsburg, Kentucky
Contract No.:	E(49-18)-1544
Total Funding:	$44,351,586 *ERDA: $32,692,102* *Industry: $11,659,484*

INTRODUCTION

The H-Coal process is being developed by Hydrocarbon Research, Inc. (HRI) under the joint sponsorship of (1) ERDA, (2) a private industry consortium composed of Electric Power Research Institute, Ashland Oil, Inc., Conoco Coal Development Company, Mobil Oil Company, and Standard Oil Company (Indiana), and (3) the Commonwealth of Kentucky. The overall objectives of this project are to further develop the H-Coal process and to demonstrate its technical and economic feasibility on larger scales. The specific objectives are to:

- Perform laboratory research on all phases of the H-Coal process, using the existing bench-scale unit and process development unit to establish design criteria.
- Design a pilot plant capable of converting 600 tons of coal per day to 2,000 barrels per day of low-sulfur boiler fuel.
- Procure equipment and materials for the pilot plant.

Ultimately, the pilot plant will be constructed and operated to provide data for further evaluation of the process and design of a full-size commercial plant. The overall program plan is shown in Figure III-1 .



PROCESS DESCRIPTION

The H-Coal process is a catalytic hydroliquefaction process that converts high-sulfur coal to boiler fuels and synthetic crude. A schematic of the process is provided in Figure III-2. Coal is crushed to minus 60 mesh, dried, slurried with recycled oil, and then pumped to a pressure of about 200 atm. Compressed hydrogen is added to the slurry and the mixture is preheated and charged continuously to the bottom of the ebullated-bed catalytic reactor. The upward passage of internally recycled reaction mixture maintains the cata-

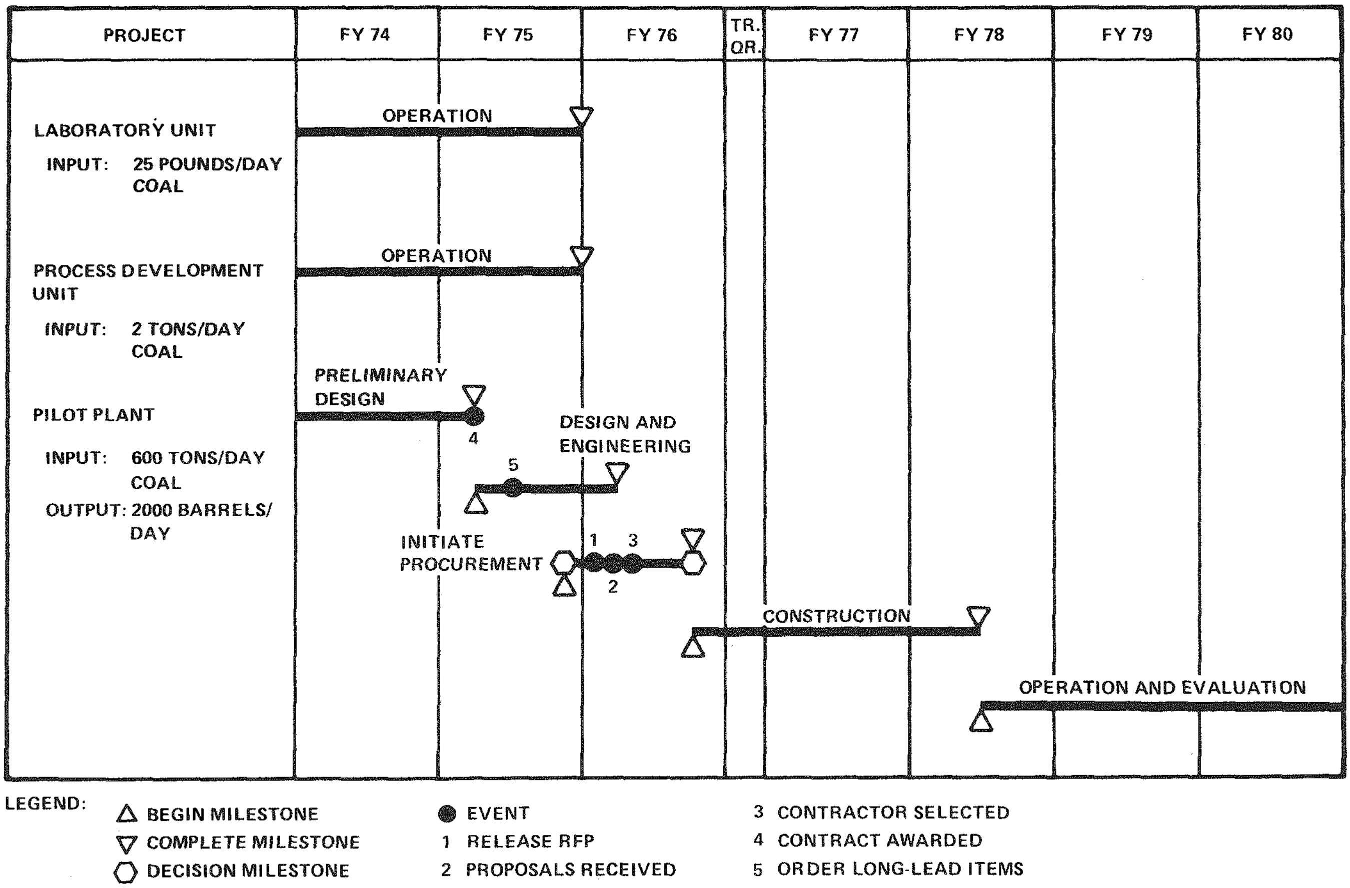


Figure III-1. H-COAL PROGRAM SCHEDULE

lyst in a fluidized state. Catalyst activity is maintained by the periodic addition of fresh catalyst and the withdrawal of spent catalyst. The temperature of the ebullated-bed catalytic reactor is controlled by adjusting the preheater outlet temperature. Typically, the temperature of the mixture entering the reactor is 650° F to 700° F.

Vapor product leaving the top of the reactor is cooled to separate the heavier components as a liquid. Light hydrocarbons, ammonia, and hydrogen sulfide are absorbed from the gas stream and sent to a separator and a sulfur recovery unit. The remaining hydrogen-rich gas is recompressed and combined with the input slurry. The liquid from the condenser is fed to an atmospheric distillation unit. The liquid-solid product from the reactor, containing unconverted coal, ash, and oil, is fed into a flash separator. The material that is flashed off is passed to the atmospheric distillation unit which yields light and heavy distillate products. The bottoms product from the flash separator (solids and heavy oil) is further separated with a hydroclone, a liquid-solid separator, and by vacuum distillation.

The gas and liquid products, composed of hydrocarbon gas, hydrogen sulfide, ammonia, light distillate, heavy distillate, and residual fuel, may be further refined as necessary. A portion of the heavy distillate is recycled as the slurry medium. The stream containing the unreacted carbon and some liquid will eventually be processed in a commercial installation to produce additional hydrogen needed for the process.

The specific operating conditions of the H-Coal process affect the type of fuel produced. For example, to produce synthetic crude, more hydrogen is required and there is a lower yield of residual fuel oil. In this mode, the separation of the solids (unconverted carbon and ash) from the liquid can be accomplished by vacuum distillation and a special liquid-solid separation unit is not required. To produce as major products clean fuel gas and low-sulfur residual oil, the tempera-

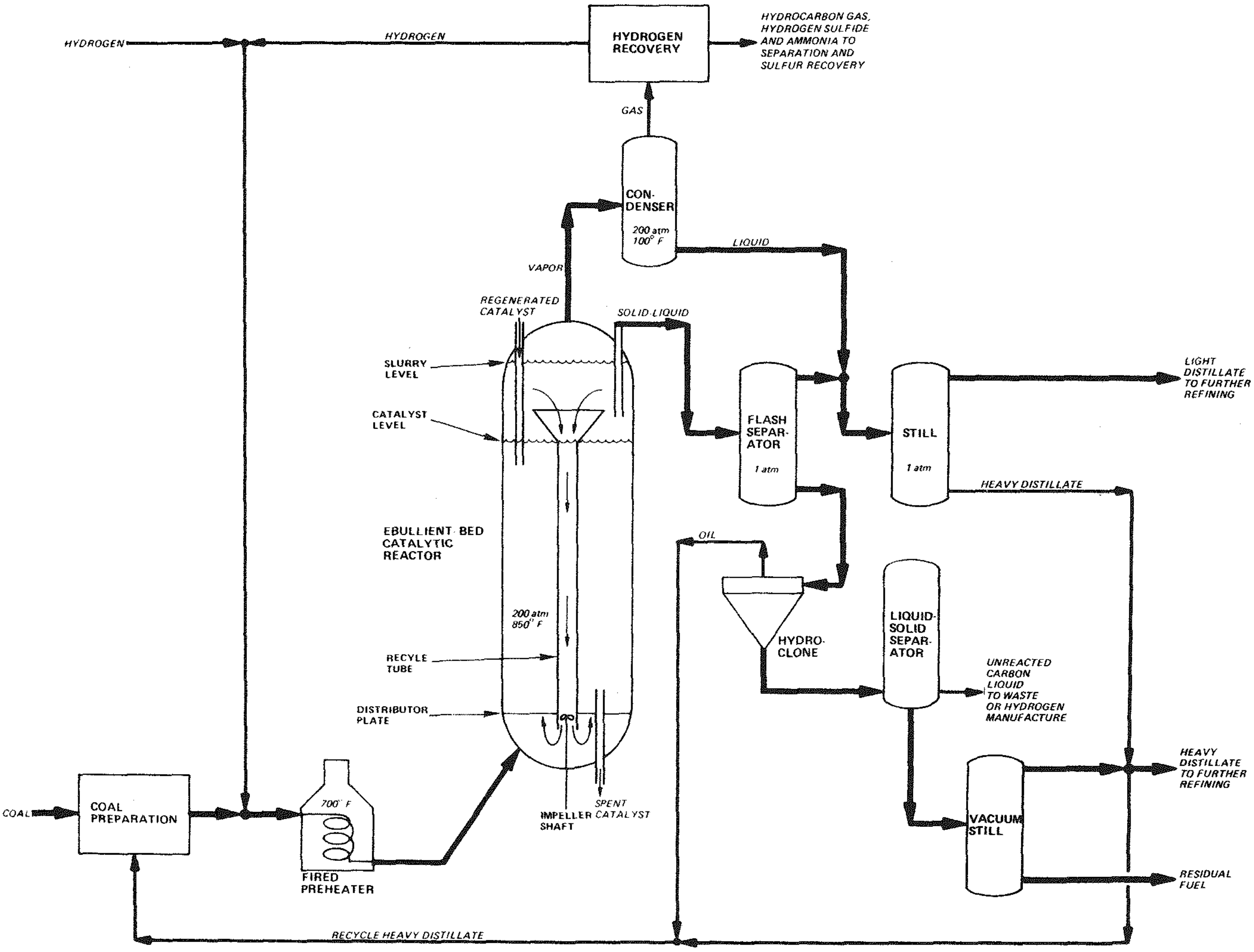


Figure III-2. H-COAL PROCESS SCHEMATIC

ture and pressure in the ebullated-bed reactor are lowered, and less hydrogen is required.

Before the H-Coal process can become commercially and economically competitive, an adequate supply of hydrogen must be generated from the process itself. The H-Coal process requires between 14,000 and 20,000 standard cubic feet of hydrogen for each ton of coal processed, depending on the type of oil produced. In the pilot plant some of the hydrogen required has been obtained from the gas produced by the ebullated-bed reactor. Additional hydrogen has been purchased to meet processing demands. In a commercial operation, hydrogen demand will be met by manufacture on site. Because the H-Coal process converts about 90 percent of the carbon contained in the coal to a liquid, the feed to the hydrogen plant could be liquid rather than solid. This suggests that commercial hydrogen manufacturing processes could be adapted. The solid cake char could also be used to produce hydrogen.

Another of the principal unsolved problems in the H-Coal process (and in other liquefaction processes) is the effective separation of the solids from the fuel products. In the H-Coal process, hydroclones have shown to be capable of removing about two-thirds of the solids and are useful in partially separating the solids so that the liquids can be recycled for slurrying the coal. HRI has also been investigating other methods of separating solids from liquids, such as magnetic separation, filtration, centrifugation, and solvent precipitation. Magnetic separation has shown only limited effectiveness. Separation of the solids by filtration has not been conclusive, although filtration rates of 150 pounds per hour per square foot and relatively dry filter cakes have been achieved by using continuous drum pressure filters.

HISTORY OF THE PROJECT

The H-Coal process was developed by HRI as a further application of the ebullated-bed processing technology originally used to convert heavy oil residues from petroleum into lighter fractions (H-Oil process). Early development of the H-Coal process, beginning in 1964, involved research with a bench-scale unit and a process development unit and preparation of a conceptual process design. An independent evaluation in 1968 confirmed the technical and economic feasibility of the H-Coal process.

The H-Coal process proved to be highly flexible in bench-scale and process development unit tests. The bench-scale unit can process 25 pounds of coal per day, the process development unit can process three tons of coal per day. Many types of coal have been tested satisfactorily on the bench-scale level, including eastern, midwestern, and western bituminous coals, western subbituminous coals, lignite from Texas and North Dakota, and Australian brown coal. The bench-scale unit has also provided information on the effect of process variables (temperature, pressure, coal feed rate, slurry oil composition, and catalyst age) on coal conversion and product yield. The process development unit tests demonstrated sustained H-Coal process operation with satisfactory control of the catalyst bed expansion.

Based on the data obtained from the bench-scale and process development units, design and engineering of the pilot plant began under the current contract in December 1973. During 1974, design of the coal preparation section of the pilot plant was completed to the extent that engineering and plant layout could begin. HRI concentrated, however, on the design and engineering for the ebullated-bed reactor, since the equipment needed for this section requires long lead times. Engineering for other sections of the pilot plant was started in late 1974. Also during 1974, three possible sites for construction of a pilot plant were evaluated. Based on detailed environmental assessments, it was decided during the second quarter of 1975 to locate the plant adjacent to the Ashland Oil, Inc. refinery in Catlettsburg, Kentucky.

The laboratory research in 1974 concentrated on bench-scale operation, liquid-solid separation, and process development unit modifications. A number of bench-scale runs were conducted to test regenerated, high-density, and aged catalysts. Tests were also conducted to investigate the mechanism of catalyst deactivation and the effects of low hydrogen partial pressure and high gas velocities between the bench-scale and process development unit. The process development unit was modified to ensure effective and safe operation, but the unit was not operated until 1975.

Pilot plant design and engineering, as well as laboratory research, was conducted during 1975. During that time the decision was made to construct the pilot facility adjacent to an Ashland Oil, Inc. refinery in Catlettsburg, Kentucky. Except for the steam supply, this arrangement would allow the utility system of the pilot plant to be shared with that of the refinery. All process descriptions, equipment specifications, and planning drawings were completed for the coal preparation section. Work in the hydrogenation area involved the modification and rearrangement of major equipment to conform to the physical characteristics of the new plant site. Preliminary process designs were also completed for the solid-liquid separation techniques of filtration, centrifugation, and solvent precipitation.

Laboratory research during 1975 concentrated on catalyst performance, hydroclone performance, and the production of low-sulfur fuel oil. Laboratory research was also conducted on the three solid-liquid separation techniques mentioned above.

PROGRESS DURING JULY-SEPTEMBER 1976

Summary

The Phase II/III agreements, originally scheduled for completion last quarter, have not yet been signed. Design activities continued throughout the quarter on the pilot plant. In the coal preparation area, effort centered on civil, structural, and electrical drawings. Piping progress is over 50 percent complete and the overall effort in the planning system and vessel groups is nearly finished. Work also neared completion in the areas of coal hydrogenation and offsites.

One test was made in the process development unit. During this test, the problem of poor separator performance reappeared. Steps were taken to alleviate a reoccurrence of this problem. Design of the catalyst addition and withdrawal system was completed and installation began. Several other reactor modifications were made prior to PDU Run 5.

A 42-hour run was made in the continuous anti-solvent precipitation unit and some preliminary PDU experiments were conducted to determine pressure drops across the recycle cup and cup risers. Also during the quarter, the PDU cold flow model was revised slightly and a correlation for fluid hydrodynamics of an ebullated bed, based on existing data, was completed.

Pilot Plant Design and Engineering

The Phase II and III agreements, originally scheduled to be signed last quarter, have not yet been signed. Due to the delay in this area, it will not be possible to have formal "Certified for Construction" grade design drawings available for a November 1, 1976 construction start.

Efforts continued throughout the quarter on plant construction. Progress in the coal preparation civil and structural areas is over 50 percent complete and all electrical drawing work is nearing completion. Overall effort in the planning system and vessel groups is nearly finished. Work in all areas of the coal hydrogenation section is over 85 percent completed. In order to retain the viability of each of the alternative reactor internal design approaches, Kobe Steel was authorized to purchase the necessary materials for both ebullating pump suction cone designs. One design will be for the full diameter cone with risers and the other is for a cone equivalent in diameter to 75 percent of the free area of the H-Coal reactor with a two-foot-high straight side.

In the solids-separation area, a 42-hour run was made in the continuous anti-solvent precipitation unit. Feed was the hydroclone underflow from the PDU and anti-solvent was H-Coal naphtha. At an anti-solvent to slurry ratio of approximately 0.8 to 0.9, poor separation was achieved for the first 16 hours. The anti-solvent to slurry ratio was increased to about 1.1 and satisfactory solids separation was achieved: 0.1 percent ash in the overflow and 20-35 percent ash in the underflow. This operation continued for approximately 12 hours and then modifications were made to analyze for the effect of slurry aging.

Process Development Unit Operations

PDU Run 4 was conducted during the quarter. During the last few days of the run, the earlier symptoms of poor separator performance reappeared. This poor performance was thought to be caused by the resid concentration being higher than that planned for the pilot plant. In addition, the relatively long residence time in the separator and resulting low flow rates may be a partial cause of the problem. A preliminary recommendation to solve the problem was to increase the liquid velocity in the lower half of the settler to suppress settling of solid materials.

Upon inspection of the PDU following the run, eroded areas were discovered on the wall of the low-pressure reactor liquid flash drum opposite the tangential inlets. The eroded areas were filled with stellite and will be observed during the next few runs to determine if reduced gas velocities, caused by efficient external separator operation, will eliminate the erosion problem.

Also during the quarter, some preliminary experiments on the PDU model were conducted to determine pressure drops across the recycle cup and cup risers. Total pressure drops ranged from 0.5 to 0.9 psig and pressure drops across the riser were between 0.2 and 0.5 psig. A correlation for fluid hydrodynamics of an ebullated bed, based on existing data, was completed during the quarter. This correlation establishes a relationship indicating the effect of gas rate on bed expansion utilizing a heptane-nitrogen system and a method of predicting bed expansion based on liquid and particle terminal velocity.

IV. CRESAP TEST FACILITY

FLUOR ENGINEERS AND CONSTRUCTORS, INC.
LOS ANGELES, CALIFORNIA

Project Site:	Cresap, West Virginia
Contract No.:	E(49-18)-1517
Total Funding:	$45,699,780 *ERDA: $43,042,140* *Industry: $2,657,640*

INTRODUCTION

Under contract to ERDA, Fluor Engineers and Constructors, Inc., is renovating and will operate an ERDA-owned facility in Cresap, West Virginia. Industrial financing is being provided by American Electric Power Corporation ($1 million), Allegheny Power Corporation ($1 million), and Fluor ($657,640). This facility, inactive since 1970, contains much of the equipment required for coal liquefaction. Fluor is to (1) renovate the Cresap facility to test the various critical mechanical components required for coal liquefaction and (2) operate the renovated facility to demonstrate an integrated process for the production of low-sulfur liquid fuel. Renovation of the Cresap facility, rather than construction of a new facility, will result in earlier operation of the pilot plant and initiation of the test program. The project schedule is provided in Figure IV-1.

PROCESS DESCRIPTION

In this process, shown schematically in Figure IV-2, coal is converted to low-sulfur liquid fuel. The coal is first crushed to 100-mesh and then combined with [illegible]omatic solvent in a mixer. The coal-solvent slurry is then pressurized and preheated. The slurry passes to an extractor where coal dissolution occurs. The product stream is then treated in a liquid-solid separation unit where the coal extract and solvent are removed, leaving a thickened slurry. This stream is sent to a carbonizer where the material is converted to char, oil, and gas. The oil is recycled to the liquid-solid separation unit and the char and gas are recovered.

One of the more persistent problems encountered in coal liquefaction processes is the separation of solids from the liquid product streams. At the Cresap Test Facility, several methods of separation will be tested. One of the most reliable methods for concentrating the solids is a system of hydroclones which was successfully used during prior pilot plant operations. Other processes to be evaluated will be filtration and chemical precipitation.

The coal extract and solvent leaving the liquid-solid separation unit are heated and fed into a flash still where the heavy components of the coal extracted are separated from the light liquids. The light liquids are

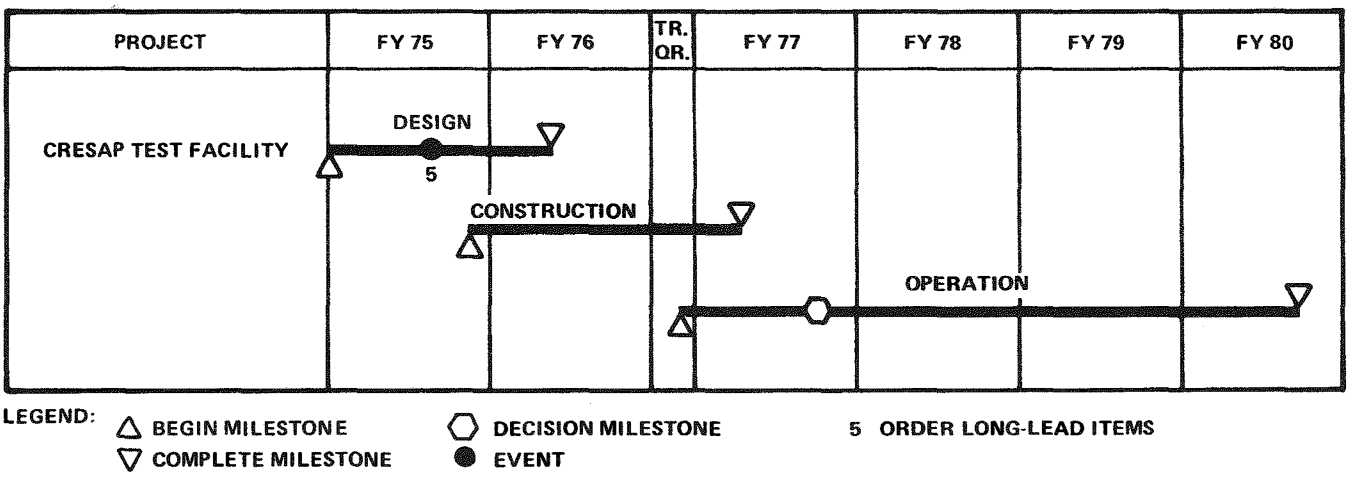


Figure IV-1. CRESAP TEST FACILITY PROGRAM SCHEDULE

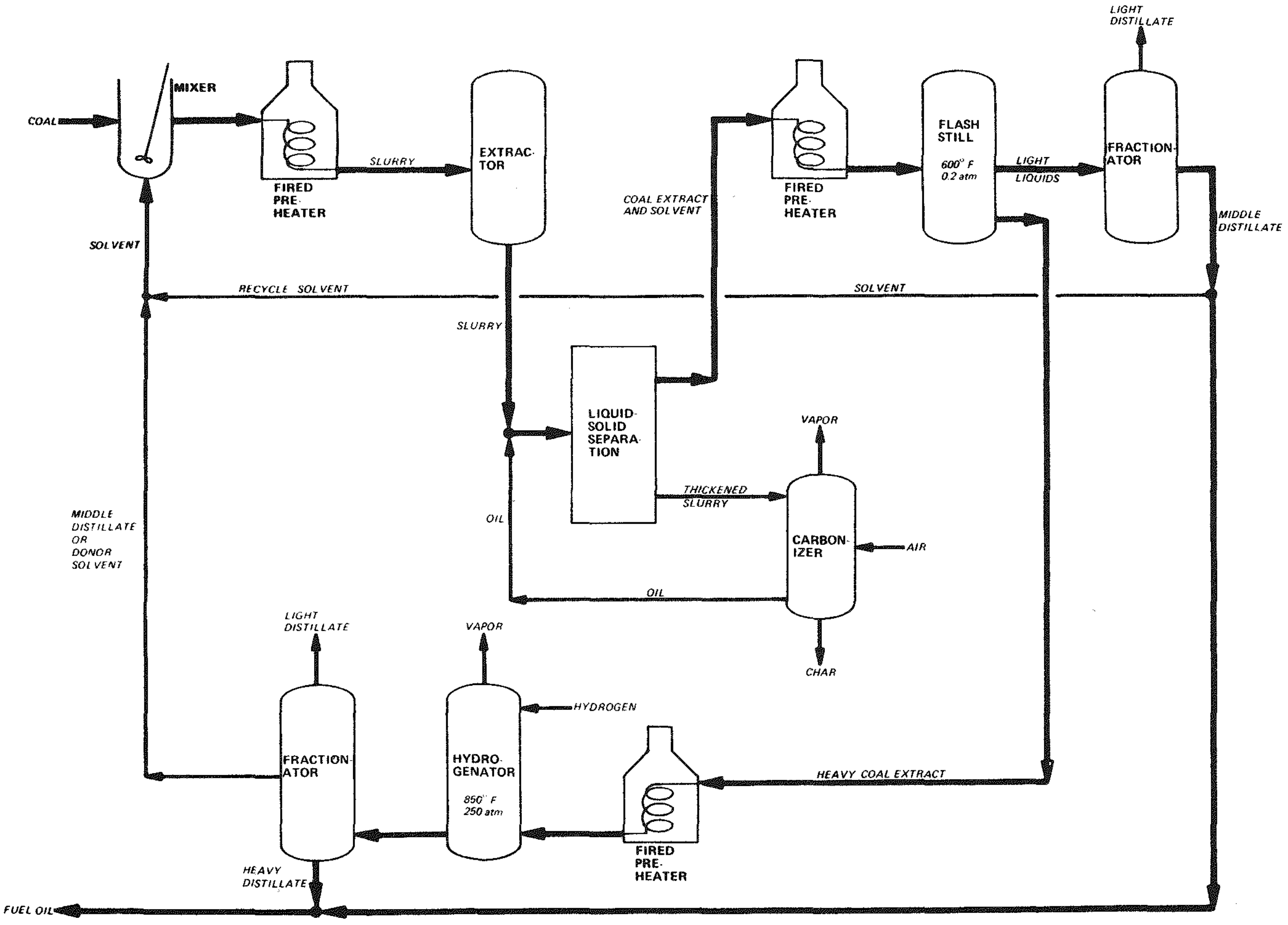


Figure IV-2. LIQUID FUEL FROM COAL PROCESS SCHEMATIC then fractionated into light and middle distillates. The middle distillate may either be recycled as a component of coal solvent or recovered as product fuel oil. The heavier components of the coal extract are heated in a fired preheater, hydrocracked, and fractionated into three distillate fractions. The middle distillate is used as coal solvent and the heavy distillate is recovered as fuel oil product.

HISTORY OF THE PROJECT

Reactivation of the Cresap facility to develop an integrated process test center for producing low-sulfur liquid fuel from coal was initiated in May 1974. Since then, the facility has been upgraded to meet environmental requirements and current design standards of piping, electrical, and mechanical equipment codes. A task force has been established to organize, coordinate, and direct the early testing program.

Renovation of the pilot plant involved the refurbishing of existing equipment and the installation of new equipment to replace outmoded or worn out equipment. Inquiry packages were developed for most of the new equipment, and flow diagrams and specifications were prepared for the refurbishing of some of the existing equipment. The Liquefied Coal Development Corporation, a wholly-owned subsidiary of Fluor, was formed during 1975 to operate the pilot plant.

PROGRESS DURING JULY-SEPTEMBER 1976

Work continued on the project to renovate the Cresap Test Facility. Engineering and design was 98 percent complete and procurement reached 99 percent completion. Much of the work on the test program planning and test procedures has been completed also. Engineering and design for the test effort is 62 percent complete and procurement completion has reached 72 percent.

Throughout the reporting period, the refurbishing work progressed at a steady pace on all of the scheduled aspects of coordinating the home office engineering, design, and procurement activities, and a high level of activity continued in the field. There were a few reported problems regarding material delivery delays and vendor quality control, but they were considered to be relatively minor and were resolved as they arose.

The work on the test program progressed according to schedule, and while a few delays and problems appeared from time to time, none were considered to be insurmountable. Preparation and development of the test procedure continued during this report period. The first draft was completed for many of the individual test elements.

Field construction activities continued throughout the quarter. The following units and subunits were mechanically complete by the end of the quarter.

- Coal feed preparation unit
- Fuel oil system
- Steam generation unit
- Steam and condensate distribution system
- Cooling water system
- Boiler feed water system
- Plant and instrument air
- Fire water system
- Tank farm
- Stretford unit
- Sanitary sewer system and treatment plant
- Waste water treatment
- Related lab facility
- Sulfur loading facility

Recruiting remained a major operational consideration as several people were hired in Operations, Technical Department, Maintenance and Engineering, and the Administrative Group. It was reported that the Affirmative Action goals were being met. An extensive training plan was underway for the employees.

V. CLEAN COKE PROCESS

USS ENGINEERS AND CONSULTANTS, INC.
UNITED STATES STEEL CORPORATION
PITTSBURGH, PENNSYLVANIA

Plant Site: Pittsburgh, Pennsylvania
Contract No.: E(49-18)-1220
Total Funding: $11,792,329
ERDA: $8,254,630
Industry: $3,537,699

INTRODUCTION

The Clean Coke process is being developed by USS Engineers and Consultants, Inc., a subsidiary of United States Steel Corporation, under the sponsorship of ERDA. The contract was initiated in 1972 under the auspices of the Office of Coal Research (OCR, now a part of ERDA). The objective of the project is to design a pilot plant that is capable of converting low-grade, high-sulfur coal to low-sulfur, low-ash metallurgical coke, chemical feedstocks, and liquid and gaseous fuels. The pilot plant design will be based on the results of laboratory tests and data obtained from process development units. The schedule for developing the process is shown in Figure V-1.

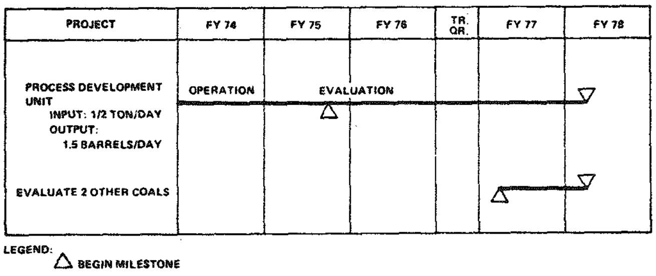
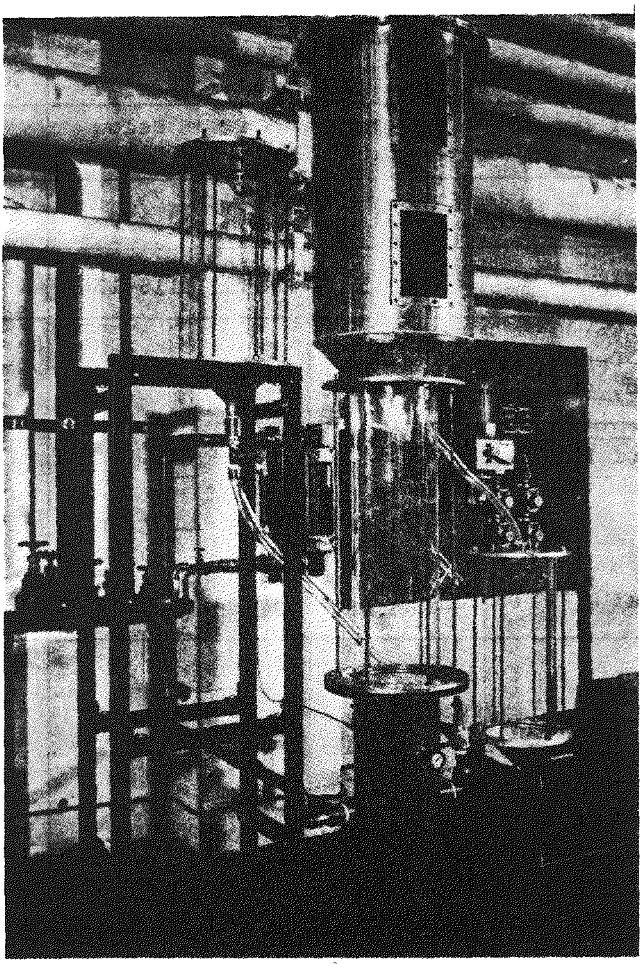


Figure V-1. CLEAN COKE PROCESS SCHEDULE



PROCESS DESCRIPTION

The Clean Coke process is shown schematically in Figure V-2. This process combines coal carbonization and hydrogenation in an environmentally acceptable manner to produce solid, liquid, and gaseous fuel products. No external hydrogen source is required and only minimal amounts of external energy will be consumed.

The feed coal is first pulverized and classified into two portions. One portion is sent to the carbonizer.

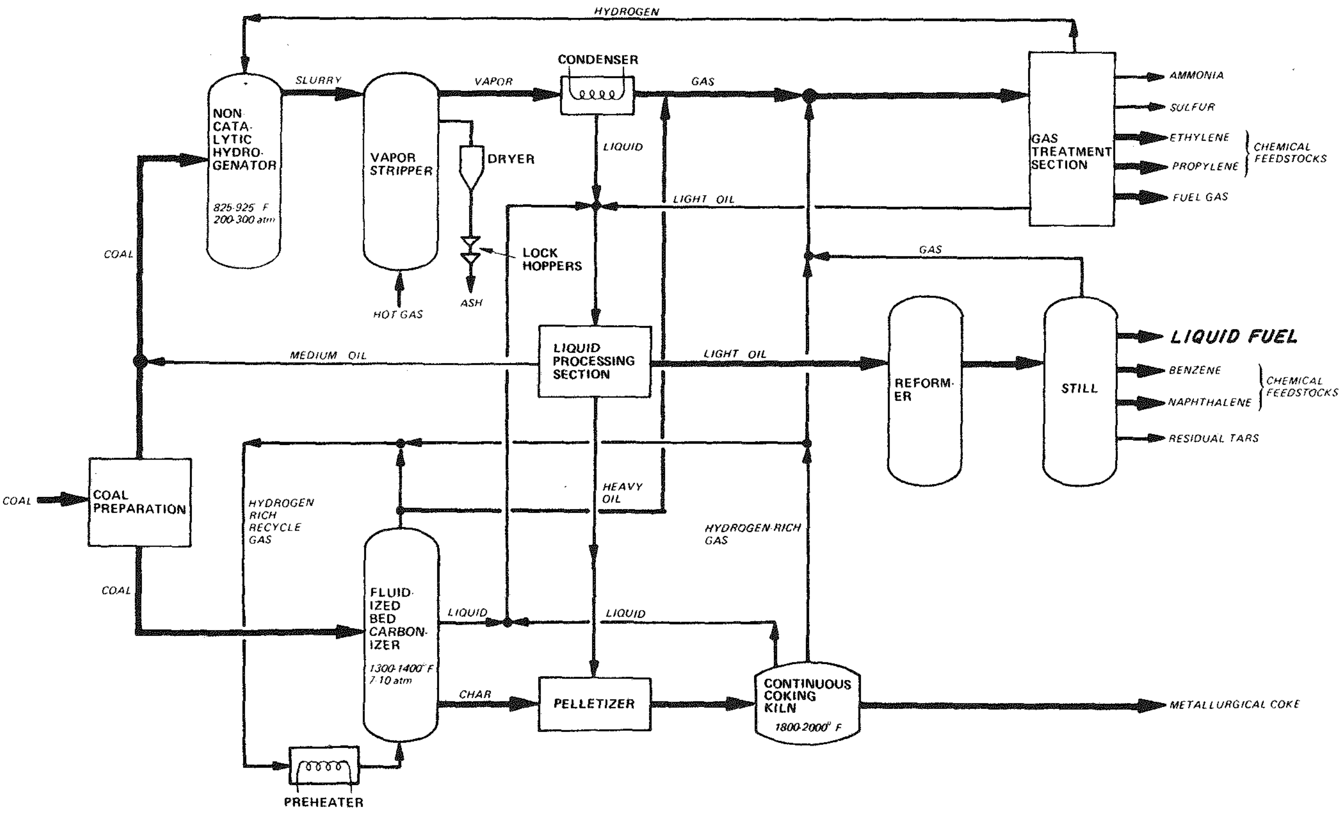


Figure V-2. CLEAN COKE PROCESS SCHEMATIC

The coal is pyrolyzed in a fluidized-bed yielding char, liquid, and a hydrogen-rich gas which is subsequently cleaned and recycled to fluidize the bed. The hydrogen-rich atmosphere of the bed provides a hydrodesulfurization environment that reduces the high-sulfur coal to low-sulfur char. This char is then pelletized using a process-derived heavy oil binder and the resulting pellets are cured and calcined in a kiln. The products are low-sulfur metallurgical coke and a hydrogen-rich gas.

The other portion of the coal is fed to the hydrogenation unit where it is slurried with a medium-weight oil recycled from the liquid processing section. This slurry is reacted with hydrogen recovered from the gas treatment section. The non-catalytic hydrogenation process is conducted at approximately 900° F and 270 atmospheres.

The hydrogenated slurry is sent to a vapor stripper where the lighter components in the slurry are vaporized by countercurrent contact with hot process gas, leaving an ash which is removed through a lock hopper. The vapor/hot gas stream from the stripper unit is fed to a condenser, the resulting liquid stream is further treated in the liquids processing section, and the uncondensed gas is sent to the gas treatment section.

Gases from all operations are treated in a common facility, which includes ammonia and acid gas removal, a sulfur production unit, an ethylene plant, and a cryogenic gas separation system.

The products from a liquids processing unit are light, medium, and heavy oils. The lighter oil is treated to produce gas, gasoline, benzene, naphthalene, and residual tars. The medium oil is recycled as slurry oil and the heavy oil is used as a coke binder.

Two major advantages of the Clean Coke process design are that no hydrogen is produced for the liquefaction portion and that metallurgical-grade coke is product from high-sulfur, high-ash coal.

HISTORY OF THE PROJECT

Laboratory and bench-scale development studies were initiated by U.S. Steel in 1969, using Illinois No. 6 seam coal to establish the technical feasibility of the Clean Coke process. Emphasis was placed on coal preparation, carbonization-desulfurization of coal in fluidized beds, and high-pressure hydrogenation of coal. The effects of temperature, pressure, reaction time, and type of feed on the carbonization reaction were investigated. The purpose of the bench-scale hydrogenation studies was to determine the effects of reaction time, temperature, and pressure on the conversion to liquid and gaseous products. Preliminary investigations of coke production were also conducted.

Under contract to ERDA since 1972, U.S. Steel is operating the process development units. The carbonizer is currently operating and the variables affecting carbonization are being tested. Initial testing has begun on the hydrogenation process development unit.

PROGRESS DURING JULY-SEPTEMBER 1976

Summary

Five carbonization process development unit (PDU) runs were completed during the quarter. Two of these were at low temperature (first stage) and three were at high temperature (second stage). Total PDU operating time is 3,334 hours, including 2,235 hours at 1,250°F and higher. Fluidization tests were also conducted during the quarter to determine the minimum fluidizing velocities for feeds with broader particle size distribution than that processed to date.

In the hydrogenation PDU, conversion of coal to oil was approximately 58 percent, probably because of the low hydrogen content (6.95 percent) of the slurry oil. Rehydrogenation of slurry oil has been modified to permit an increase in hydrogen content to about eight percent.

The flash vaporizer of the binder-preparation PDU processed more than 33,000 pounds of liquid blowdown from the carbonization PDU. Approximately 19,000 pounds of clean wash oil was recovered for recycle and 1,900 pounds of tar residue were produced for binder-preparation. The properties and compositions of two synthetic binders for cokemaking were compared with those of binder made from stripper bottoms obtained during a coal hydrogenation run.

Carbonization Process Development Unit

In addition to general maintenance work on the PDU, the unit was operated in two low-temperature (first stage) runs and three high-temperature (second stage) runs. In the first low-temperature run (820° F), 85 hours of steady-stage operation produced 3,650 pounds of semichar from 4,542 pounds of preoxidized Illinois No. 6 seam coal. The semichar produced was used as feed for second-stage carbonization tests. Average feed rate was 54 pounds per hour, for a residence time of 27 minutes. The volatile matter and sulfur contents of the semichar product were 21 and 1.7 weight percent, respectively.

A 1,325° F test was then made to study the effect of lower pressure (100 psia) on carbonization/desulfurization of the semichar feed. Compared with data from tests run at 165 psia, the char produced at the lower pressure had a higher volatile matter content (3.1 vs. 2.0 weight percent), a lower bulk density (13.8 vs. 21.7 pounds per cubic foot), and only a slightly higher sulfur content (0.73 vs. 0.70 weight percent). Char yield was not affected by pressure, but gas production was substantially greater at the lower pressure. Based on the test results, it was determined that the effect of pressure alone cannot be demonstrated in the continuous reactor because of the interdependence of the variables in the system. Because of this, future PDU runs will be made at 165 psia and the pilot plant will be designed for pressures of at least 165 psia.

The third carbonization run was a 1,400° F run consisting of two tests to study the effect of high hydrogen sulfide levels in the recycle gas on char sulfur content. The tests showed that increasing the level from 100 to 500 ppm increased the sulfur content from 0.21 to 0.71 percent, but a further increase in hydrogen sulfide content to 1,000 ppm had virtually no effect on sulfur content.

The last two PDU runs were made to obtain material balance data. These runs consisted of a five-day 820° F run and a ten-day 1,400° F run. Total PDU operating time is now 3,334 hours, including 1,099 hours of first-stage carbonization and 2,235 hours of second-stage carbonization. Approximately 16.5 tons of Illinois No. 6 seam coal have been processed to date.

The carbonization PDU has thus far been operated with coal feed size 20 x 65-mesh. However, the process design study includes testing of feed having a broader particle-size range to examine the effect of particle size on desulfurization and provide information useful in scaleup to larger fluidized-bed carbonizers. Fluidization tests were therefore run in the ten-inch mock-up of the carbonizer reactor with two coal fractions, 10 x 100-mesh and ⅛-inch x 100-mesh. The minimum fluidizing velocity for each of the size ranges was:

20 x 65-mesh	0.63 feet per second
10 x 100-mesh	0.99 feet per second
⅛-inch x 100-mesh	1.95 feet per second

The fluidizing velocity for the 10 x 100-mesh feed is reasonable, and thus this size range will be studied in future PDU runs. The 1.95 feet per second velocity required for the larger size range is too large for the existing system and thus will not be further evaluated.

Hydrogenation Process Development Unit

The hydrogenation PDU was successfully used for a coal hydrogeneration run during the quarter. Following the run, an analysis of residual solids indicated that conversion of coal to oil was low, approximately 58 percent, based on moisture- and ash-free coal. The hydrogen content of the slurry oil, 6.95 percent, was probably too low to effect good conversion of the coal.

Overhead product from the stripper, collected from the coal conversion run, was distilled to remove light and heavy ends, as well as recover middle oil. The 2,000 pounds of middle oil which was recovered was subsequently catalytically hydrogenated to produce a slurry oil containing nearly eight percent hydrogen. Approximately the same quantity of previously prepared slurry oil was hydrogenated again to produce a slurry oil containing 7.7 percent hydrogen. These hydrogenated middle oils will be used in future runs to determine the effect of higher hydrogen content in the slurry oil on coal conversion efficiency. A coal conversion run was attempted in September, but was unsuccessful due to significant plugging problems.

Binder Preparation Process Development Unit

The flash vaporizer of the binder preparation PDU processed the liquid blowdown from the carbonization PDU runs to separate and recover the water, recycle wash oil, and heavy tar residues in the blowdown mixtures. The was oil inventory was also increased by processing a conventional coke-oven tar fraction to produce 16 drums of fresh wash oil. In an effort to minimize plugging problems in the flash vaporizer, blowdown liquids from the gas/gas interchanger pot are now being passed through a 50-mesh screen before collection at the carbonization PDU.

The properties and compositions of two synthetic binders for cokemaking were compared with those of binder made from stripper bottoms obtained during a coal hydrogenation run. One of the synthetic binders was prepared from fluxed solvent-refined coal (SRC) product and the second was prepared by blending Clairton hard pitch and tar oil. It was noted that the properties of the two hydrogenation-derived binders were similar, thus indicating that the fluxed SRC product should be a satisfactory substitute for the Clean Coke-derived binder.

VI. LIQUEFACTION OF COAL BY DIRECT HYDROGENATION

ROCKETDYNE DIVISION
  ROCKWELL INTERNATIONAL CORPORATION
CANOGA PARK, CALIFORNIA

Plant Site: Canoga Park, California
Contract No.: E(49-18)-2044
Total Funding: $994,375
(*100% ERDA*)

INTRODUCTION

Under contract to ERDA, the Rocketdyne Division of Rockwell International is conducting a research program to develop a technology to liquefy coal via direct hydrogenation. This technique would involve rapid mixing, reaction, and quenching of a gaseous hydrogen and pulverized coal mixture to form hydrocarbon liquids. These products would be suitable for conversion to fuel oil or chemical feedstocks.

The program is divided into two phases. Phase I involves the design and fabrication of cold-flow equipment and the operation of the equipment in a series of tests to evaluate methods of transport and injection of pulverized coal into the reactor.

Upon completion of Phase I, the results of the cold-flow tests were submitted to ERDA for a decision on proceeding with Phase II. Upon Phase II authorization, small-scale reactor testing was initiated to evaluate the effects of reactor configuration and operating conditions. Based on these results, a larger reactor, with diverse injection methods and a coal flow rate of approximately one ton per hour, will be built and tested. Test results and analyses will be evaluated to determine the feasibility of this process. The schedule for the development of this process is shown in Figure VI-1.

In addition to the ERDA funded effort, Rockwell International is sponsoring a corollary effort to provide supporting facilities.

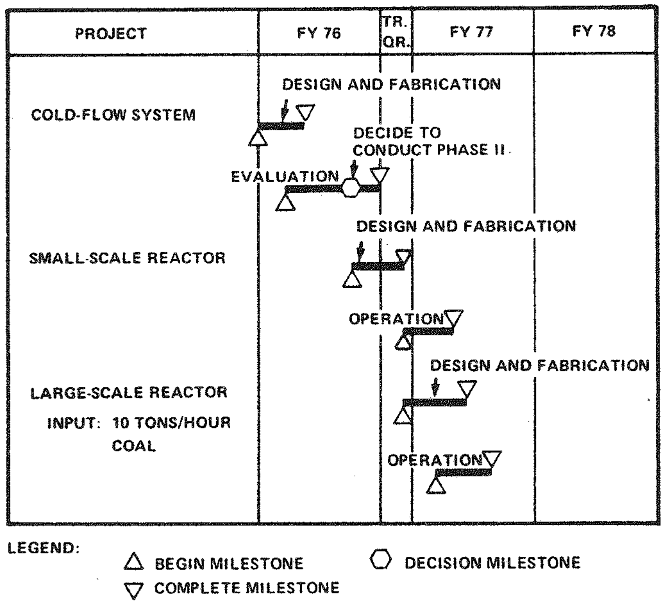


Figure VI-1. PROGRAM SCHEDULE FOR LIQUEFACTION OF COAL BY DIRECT HYDROGENATION

PROCESS DESCRIPTION

The basic concept of this process for the liquefaction of coal is that high liquid yields are favored by rapid mixing, reaction, and quenching of a mixture of pulverized coal and hot gaseous hydrogen. In this process, as shown in Figure VI-2, coal is fed into the coal feeder from a batch feeder, with the transport gas. The coal passes through the feeder, is partially fluidized with hydrogen at the conical exit, and then passes to the entrained-flow reactor where it is mixed rapidly with 1500° F hydrogen. The pulverized coal and hydrogen react for about 10 to 100 milliseconds. The reactor is expected to be operated over a pressure range of 35 to 100 atm and a temperature of approximately 1800° F.

The reactor effluent is passed into a quench section which has a set of water spray nozzles. From there the effluent passes into a char receiver.

HISTORY OF THE PROJECT

Development of a process for the liquefaction of coal by direct hydrogenation was started in August 1975. Work to date has focused on a series of tests designed to determine the most effective means of transporting coal to the point of injection into the reactor. These tests were conducted in both a transparent feeder mechanism and a cold-flow unit. Design of a one-ton-per-hour reactor also began during 1976.

PROGRESS DURING JULY-SEPTEMBER 1976

Summary

During the quarter, 12 additional tests were conducted in the 500-pound-per-hour reactor. For the

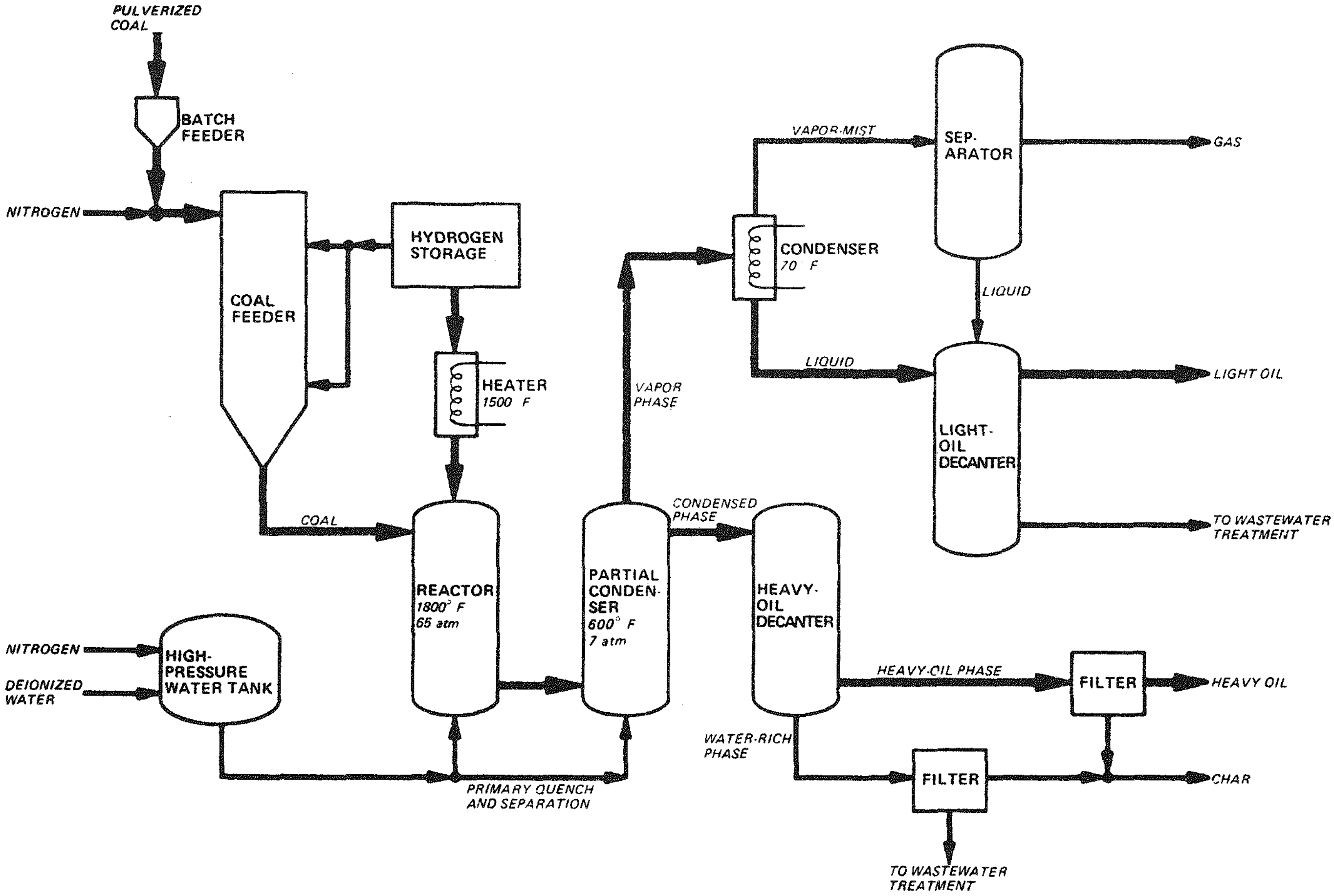


Figure VI-2. SCHEMATIC OF LIQUEFACTION OF COAL BY DIRECT HYDROGENATION most part, operational difficulties were solved and relatively high conversions were obtained. In addition, a critical review of the product analysis procedure revealed no errors in the methods used. The draft annual report describing progress during the first year of the program was also prepared and submitted to ERDA for technical review. Construction of the one-ton-per-hour reactor system is proceeding rapidly toward a completion date of early November. The program is currently three weeks behind schedule. Initial testing of the system is scheduled for early December.

Discussion of Activities

The twelve additional reactor tests conducted on the 500-pound-per-hour unit demonstrated that the char agglomeration and wall adhesion problems encountered during earlier tests did not occur at sufficiently high reactor temperatures. The reactor temperatures were obtained through partial combustion of preheated hydrogen with oxygen. Successful operation was obtained with oxygen addition in a preburner upstream of the reactor or in the reactor itself, although overheating problems were encountered with the concentric tube injector when oxygen was introduced in the preburner. The test results indicated that the location of oxygen addition had little effect on the overall carbon conversion but does have a significant effect on the conversion to gases. The data suggest that there is an increase in overall carbon conversion with both increased residence time and increased velocity in the reactor. The liquid yield also is increased with increasing residence time and velocity.

Tests were conducted with two injector configurations, a concentric tube and a 4-on-1, in a basic reactor configuration. Analysis of the test results also suggested that carbonaceous material was being carried out in the condensed phase with the product gas. Therefore, the reactor system is to be modified to add a bag filter to the product gas system to recover this finely divided material.

The product analysis methods were critically reviewed by several individuals during the quarter to assure that the evaluations were correct. The analysis methods were confirmed by this review.

A draft annual report describing progress during the first year of the program was prepared during the quarter and submitted to ERDA for technical review. The report covered the cold-flow testing conducted during Phase I and the reactor testing effort made through July.

Construction of the one-ton-per-hour reactor facility continued toward a scheduled completion date three weeks behind schedule. Based on this schedule, testing of the unit will be initiated early in December.

VII. FILTRATION PROCESS AND EQUIPMENT STUDIES

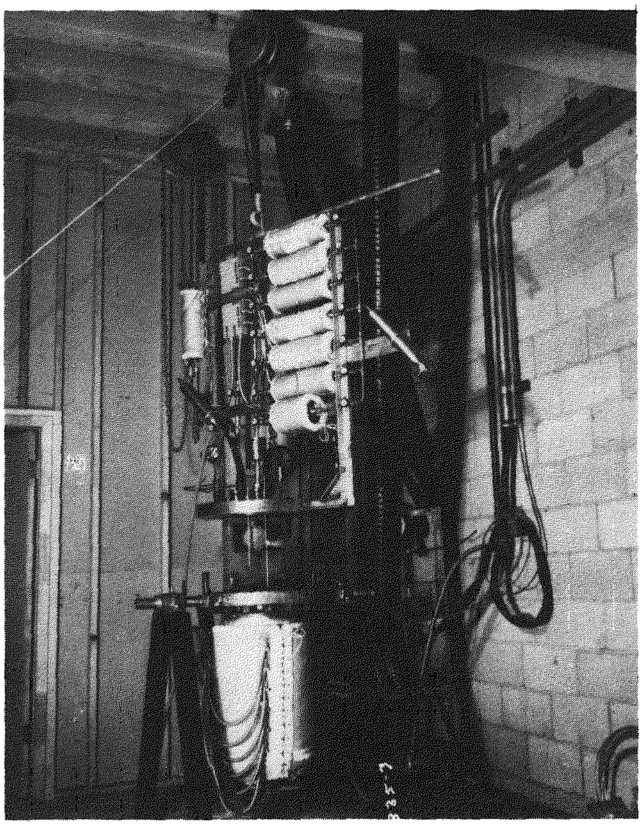
JOHNS-MANVILLE SALES CORPORATION
DENVER, COLORADO

Project Site: Denver, Colorado
Contract No.: E(49-18)-2007
Total Funding: $190,000
(100% ERDA)

INTRODUCTION

Under contract to ERDA, Johns-Manville Sales Corporation is conducting research to improve solid-liquid separation, a critical step in all coal liquefaction processes. The first phase of this program was initiated in May 1975. Johns-Manville is studying rotary drum pressurized precoat filtration. The objective of the research program is to achieve major improvements in the filtration process and in the filtration equipment used in the coal liquefaction process. The goals are to double the filtration rate, reduce filter aid usage, and demonstrate the results on a pilot-plant scale.

The filtration research program is divided into two phases. The objectives of Phase I are to (1) develop basic knowledge of the coal oil filtration process and its dependence on liquefaction process parameters and (2) apply the knowledge gained to improve the performance of filtration operations in existing liquefaction plants. Phase II involves the design and construction of a rotary drum pressurized precoat filter capable of processing the product flow from an existing liquefaction pilot plant. Phase I is being funded entirely by ERDA and Phase II is being supported by the Electric Power Research Institute (EPRI) in cooperation with the ERDA effort. One of the criteria established for a viable system is the capability of filtering in excess of 20 gallons of solid-liquid mixture per square foot [illegible] hour. Data obtained from operation of the pilot scale filtration unit will be used to design filters for commercial plants.



PROGRAM DESCRIPTION

Phase I of the project is being implemented through laboratory bench-scale research. The initial step involves the design and construction of a laboratory test facility that will simulate the operation of a rotary drum pressurized precoat filter capable of operating at 750° F and 27 atm. A series of tests will then be conducted to study pressure precoat filtration operating parameters at the temperatures and pressures expected in normal pilot plant operations. In these tests, two liquefaction process liquids are to be used: solvent-refined coal (SRC) from both the Wilsonville and Tacoma pilot plants and Synthoil boiler fuel supplied by the Pittsburgh Energy Research Center.

Upon completion of these tests, the apparatus may be moved to a Pittsburg and Midway (P&M) facility at DuPont, Washington. Here it would be used to establish the relationships between test results obtained on fresh filter feed and those on oil products that have been cooled and tested in Denver. Due to the chemical and physical changes which may take place during aging and irreversible alterations which may occur due to various thermal histories, it is vital that product behavior during pilot testing be accurately correlated to the anticipated behavior during commercial operations.

In cooperation with P&M, Johns-Manville will apply its filtration knowledge to improve the filtration rate and reduce the filter aid usage on existing Goslin-Birmingham filters. Similar filter process optimization studies will then be conducted at other ERDA-sponsored coal liquefaction plants. Johns-Manville is also responsible for assisting in the selection of a suitable precoat filter for the Cresap Test Facility in Cresap, West Virginia. This investigation will include (1) the feasibility of modifying the existing filtration equipment to accommodate the experimental work and (2) assistance in the start-up and establishment of operating conditions for the filter system selected.

PROGRESS DURING JULY-SEPTEMBER 1976

Summary

During this reporting period, 48 filtration runs were conducted in the rotary leaf pressure precoat filter. Six of these runs were made on a sample of Synthoil product and the remainder used Tacoma Filter Feed. The major emphasis of test runs conducted during July was to determine the dependence of filtration rate and efficiency on process parameters. Also investigated were the significance of filter and pore size, location of the rate-limiting resistance to flow in the filtering system, and the effect of body feed. Runs conducted during August studied the effects of temperature, differential pressure, blade advance, and immersion time as well as shell pressure, body feed, additional let-down of feedstock, and degree of agitation of the feedstock. The bulk of the filtration effort during September was devoted to the Tacoma Filter Feed (SRC product), however, initial work also began on the study of Synthoil product.

The results of the filtration tests enabled the investigators to draw some preliminary conclusions. It was determined that, in general, filtration rate is favored by increased blade advance, higher temperatures (up to 600° F), and by increased pressure differentials (up to 35 and 40 psid). It was also found that filtration rate may be increased by shorter immersion time and the introduction of body feeds, i.e., modification of the rate-limiting factor of coke formation on the surface of the product.

Filtration Tests

Fifteen filtration runs were made in July. In addition to a study of the dependence of filtration rate and efficiency on process parameters, also studied were filter aid pore size, location of the rate-limiting resistance to flow, and the effect of body feed. For all runs, pre-precoats of FIBRA-FLO 7C were used and precoats of antiflocculant-treated diatomite filter aids were deposited from slurries in 50/50 toluene-xylene (TX) mixed solvent. The feedstock used was Tacoma Filter Feed.

To study the effect of filter aid pore size, tests were made using both AF 7 precoat (with a median pore size of four micrometers) and AF 5 precoat (with a median pore size of 7.5 micrometers). No evidence was found of a significant rate decrease with decreasing pore size. This finding suggested that viscous drag in the precoat was not a rate-limiting factor. Thus, the use of finer grades of filter aid, which should limit penetration of particulates and, therefore, require a less rapid blade advance, is indicated.

Other July filtration runs led to the conclusion that under the conditions imposed, the rate-limiting resistance to flow resides in the filter cake formed by the SRC solids on or near the surface of the precoat. This observation would therefore indicate that filtration rate could be improved by using shorter immersion times and the introduction of body feeds. The use of body feeds fits in well with the expectation that a commercial liquefaction facility would desire to regenerate and reuse a major portion of the filter aid. Both economics and waste disposal indicate this is desirable. Since some degradation of permeability of the filter aid will occur during regeneration, it is expected that this regenerated material will be well suited for use as body feed.

Filtration tests during August were designed to study the parameters expected to affect the rate-limiting role of the filter cake itself. A total of 17 filtration runs were made on Tacoma Filter Feed, exporing the usual parameters (temperature, differential pressure, blade advance, and immersion time) as well as shell pressure, body feed, additional let-down of feedstock, and degree of agitation of the feedstock.

The results of the tests indicated that there was little or no effect of shell pressure on filtration rate. The trend, if any, was for increased rate with increased pressure. A 29 percent let-down of as-received Tacoma Filter Feed with Tacoma Process Solvent did result in improved filtration rates, but not sufficiently improved to accommodate the increased volume. It was also determined that high rates of filtration are favored by higher temperatures (up to 600° F) and by increased pressure differentials (at least up to 35 and 40 psid). In work with body feed, blade advances of two mils per cycle appeared adequate to maintain favorable rates, whereas with unaltered Tacoma Filter Feed, two-mil advances have proven insufficient and even five-mil advances resulted in rates considerably suppressed relative to those attained with ten-mil advances.

The tests also showed that thorough agitation of charges containing body feed is desirable to keep all solids in suspension throughout a run. There was no significant effect of agitation on filtration rate of unaltered feedstock.

Seven filtration tests were conducted in September using Tacoma Filter Feed and record flow rates of 388 pounds per square foot per hour were obtained for untreated feedstock at 604° F and 40 psid. These results demonstrated the advantage of using higher temperatures and pressure differentials.

The last six tests conducted during September were made on a sample of Synthoil product received from the Pittsburgh Energy Research Center. The tests were hampered by possible sample contamination and a malfunction of the blade advance mechanism. The test results did, however, indicate that reasonable filtration rates may be expected in future runs using this feedstock.

VIII. CHEMICALS FROM COAL

THE DOW CHEMICAL COMPANY
MIDLAND, MICHIGAN

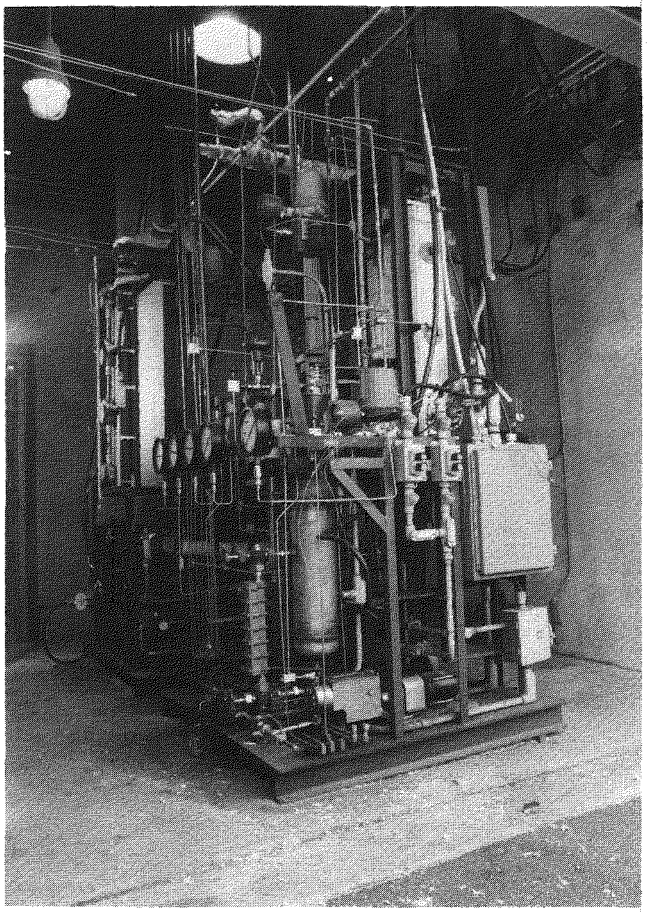
Project Site: Midland, Michigan
Contract No.: E(49-18)-1534
Total Funding: $342,896
(100% ERDA)

INTRODUCTION

Historically, coal was used as a chemical feedstock, but petroleum and natural gas were substituted as cheaper feedstocks when they became available. However, because of the recent rising costs and decreasing availability of petroleum and natural gas, The Dow Chemical Company, under sponsorship of ERDA, is studying the possibility of using coal-derived liquids as chemical feedstock. The current contract was initiated in 1974. Dow is testing liquids from four coal liquefaction processes (COED, Synthoil, H-Coal, and SRC) in standard petrochemical processing experiments and is evaluating these results to select one process for further study. The selection will be based on process design, quality of chemicals produced, and overall plant economics. For the process selected, further development will include material balances and utility requirements for each major process step, a preliminary process flowsheet, and estimates of capital and operating costs for a commercial plant.

PROGRAM DESCRIPTION

The liquids produced by the four processes are tested in standard petrochemical processing operations to determine the advantages and disadvantages of using coal-derived liquids for petrochemical feedstocks. The experimental work is being conducted in small units capable of operating unattended at pressures up to 204 atm., temperatures up to 1300° F, and feed rates from 25 to 400 milliliters per hour.



The processing sequence used for all of the samples is:

(1) Distillation into nominally three cuts—straight-run naptha (350° F or less), middle distillate (350° F to 650° F), and heavy gas oil (650° F to 950° F).
(2) Hydrocracking of the middle-distillate and heavy gas oil to reduce molecular weight.
(3) Hydrotreating of the straight-run naphtha and hydrocracked naphtha.
(4) Reforming of the hydrotreated naphthas to maximize aromatics.

Steam coil cracking of some of the fractions is also performed to evaluate potential ethylene yields.

Characterization studies on all of the samples in the hydroprocessing experiments are performed using the following procedures:

- Component analysis of the hydrocarbons.
- Atmospheric, vacuum, and American Society for Testing Materials (ASTM) distillations.
- Elemental analysis (carbon, hydrogen, oxygen, nitrogen, sulfur).
- Trace metal analysis.
- Liquid chromatography for class separations (aliphatics, monoaromatics, diaromatics).

PROGRESS DURING JULY-SEPTEMBER 1976

Synthoil

Hydrocracked naphthas were hydrotreated in experiments using a catalyst charge that had been used to hydrotreat H-coal material. Sulfur data indicated that after 100 hours on stream, the sulfur contamination experienced during presulfiding for the H-coal run was washed out and sulfur reduction to less than two ppm was achieved. This synthoil feedstock was re-hydrotreated over Cyanamid Aeroform HDS-9A catalyst during August and September. Results indicated that sulfur was reduced to less than 1.5 ppm and nitrogen reduced from 19.6 to 8.6 ppm.

The re-hydrotreated naphtha was reformed during September. The data patterns from the reformate were shown to be similar to those of the COED and H-Coal naphthas. However, with the Synthoil material, the aromatic yield in the C_6-C_9 range was slightly lower than in the other two.

H-Coal Syncrude

During July, results from naphtha hydrotreating tests conducted in June, in which the catalysts were presulfided with hydrogen sulfide gas, were evaluated. Results indicated that some sort of contamination problem still existed even when using the gaseous presulfiding technique. The use of hydrogen sulfide was implemented in an effort to eliminate contamination problems that had been experienced in using carbon disulfide in light naphtha for presulfiding. The naphtha was re-hydrotreated in August and September. Analytical results indicated that sulfur levels were reduced to 3.4 ppm and nitrogen was reduced to 6.4 ppm. Reforming tests were also performed during September. Components data for both the feedstock and product demonstrated much the same pattern as was observed in COED hydrotreating/reforming experiments. The H-Coal product had a much higher aromatic yield in C_6-C_9 reformate, however. Hydrogen consumption calculations are expected to be made following the completion of data reduction and elemental balance analysis.

SRC Product

First-pass hydrocracking experiments initiated last quarter were completed in July. The resulting product was distilled to yield 57 pounds of hydrocracked naphtha and 114 pounds of middle distillate. Second-pass hydrocracking experiments were completed in August. This composite product was distilled to yield 10.9 pounds of hydrocracked naphtha and 23.7 pounds of middle distillate. Hydrotreating experiments on the cracked naphthas were initiated in September. However, problems were encountered in the removal of nitrogen during normal hydrotreating conditions. Testing of the naphtha revealed that various forms of pyridine were present and attempts to remove the nitrogen by increasing hydrotreating reactor pressure to 2500 psig were made. No test results were available by the end of the quarter.

IX. R&D ENGINEERING SERVICES-HYDROCARBONIZATION

OAK RIDGE NATIONAL LABORATORY
OAK RIDGE, TENNESSEE

Contract No.: E(49-18)-1740
Total Funding: $2,135,000
(*100% ERDA*)

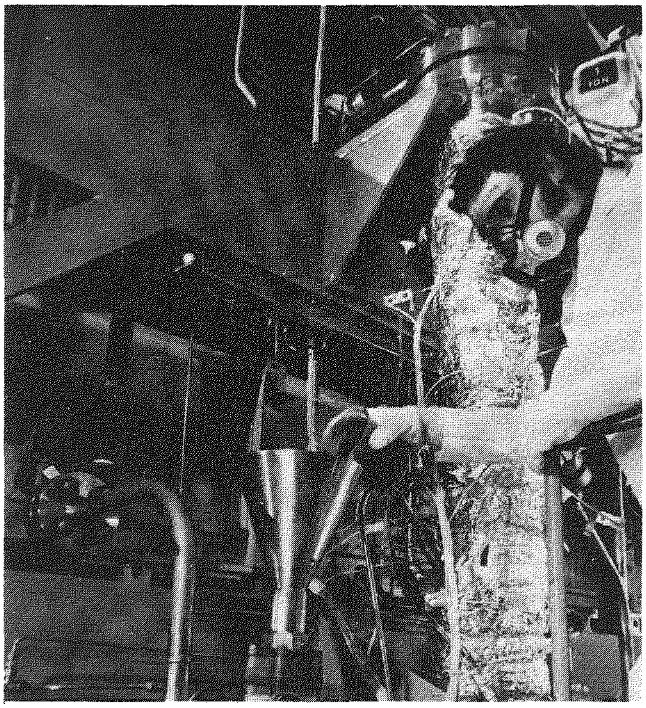
INTRODUCTION

The Oak Ridge National Laboratory (ORNL) is conducting several research and development projects in support of the increased utilization of coal as a source of clean energy. Under this agreement, begun in 1974, ORNL is to estimate the optimum operating conditions of a fluidized-bed hydrocarbonization reactor and design, construct, and operate a fluidized-bed reactor capable of continuously processing ten pounds of coal per hour. Operating conditions for this reactor will be at least 20 atm and up to 1,200° F.

PROCESS DESCRIPTION

Hydrocarbonization is an important type of coal liquefaction process which combines high-temperature, high-pressure, and fluidized-bed operation using hydrogen-rich gas for fluidization. The hydrocarbonization process can produce controlled yields of desulfurized char, liquid fuels, and substitute natural gas. The product char is suitable for use in boilers, without stack gas treatment units. The liquid products may be composed of naphtha, light gas oil, and heavy gas oil. A flow diagram of the bench-scale hydrocarbonization unit is shown in Figure IX-1.

The work under this contract is divided into three tasks:



- Review and evaluation, involving the analysis of prior work to estimate operating conditions and guide reactor design.
- Experimental development, which includes the investigation of hydrocarbonizer components at atmospheric pressure and temperatures up to 1,200° F.
- Bench-scale studies involving the design, construction, and operation of continuous hydrocarbonization system capable of processing ten pounds of coal per hour at temperatures up to 1,200° F and pressures as great as 20 atm.

An intensive literature survey conducted during 1975 led to the preliminary conclusions that very little reaction takes place below 750° F and the yield of both oil and gas increases directly with temperature. A maximum oil yield was noted at 1,020° F to 1,110° F, however gas production increased proportional to

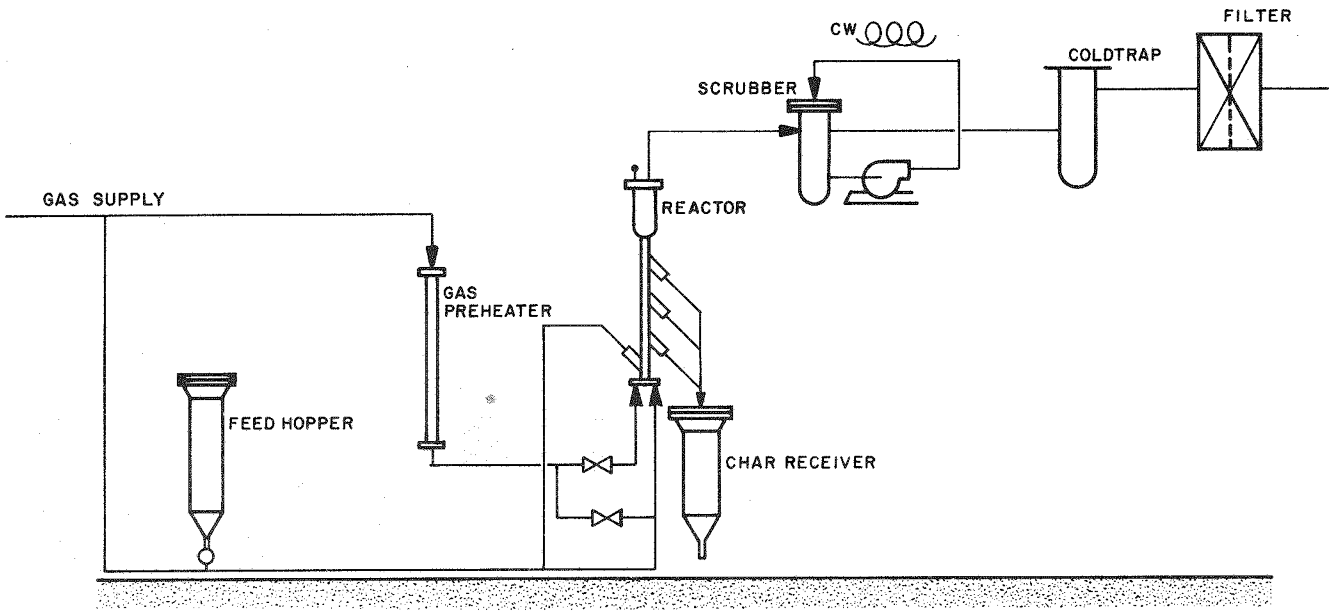


Figure IX-1. FLOW DIAGRAM OF BENCH-SCALE HYDROCARBONIZATION SYSTEM

temperature and therefore did not show an optimum over the temperature range studied. Two Lucite mock-ups were constructed to study the mechanics of hydrocarbonization processes and the ten-pound-per-hour bench-scale unit was built. The overall performance of this unit proved satisfactory.

PROGRESS DURING JULY-SEPTEMBER 1976

Modification of the bench-scale system to permit recirculating fluidized-bed operation was completed in early August. The reactor was successfully tested in Experiment HC-7, a run in which Wyodak subbituminous coal was fed at a rate of 14.8 pounds per hour for 6.9 hours, with the reactor operating at 820° F and 300 psig. During the run, minor difficulties were encountered with the electrical heaters and controllers, thus limiting operation to below design temperature.

By September, the solids handling facility was sufficiently completed to permit shakedown and coal preparation. Approximately 100 pounds of coal, dried to 10 percent moisture and sized at —50 + 140-mesh, was produced for HC-8 from 505 pounds of coal containing approximately 27 percent moisture. Equipment failures hindered two attempts at initiating Experiment HC-8, however they were corrected for resumption of experiments in October.

Data analysis also continued throughout the quarter on results from previous tests. It was found that at 300 psig and 910°-1075° F there was neither significant consumption nor significant production of hydrogen. Trace element analysis of HC-2 products were provided by the Analytical Chemistry Division.

X. R&D ENGINEERING SERVICES-CARBONIZATION OF RESIDUE

OAK RIDGE NATIONAL LABORATORY
OAK RIDGE, TENNESSEE

Contract No.: E(49-18)-1740
Total Funding: $325,000
(100% ERDA)

INTRODUCTION

The Oak Ridge National Laboratory (ORNL) is conducting several research and development projects in support of the increased utilization of coal as a source of clean energy. Under this contract, begun in November of 1975, ORNL is to:

- Review the experience with the low-temperature carbonizer at the Cresap Test Facility.
- Modify the existing atmospheric batch reactor to operate continuously on solids-laden residue from ERDA processes.
- Operate the above reactor with material from the H-Coal, Solvent Refined Coal (SRC), and Consol Synthetic Fuel (CSF) processes.

PROCESS DESCRIPTION

The process to be used by ORNL to evaluate the residual materials from the various liquefaction processes is shown schematically in Figure X-1. In the process, the residual materials will be converted to char by carbonization. The feedstock will be transported from the feed pot to the reactor by a stream of inert gas. Additional inert gas will be preheated and fed to the reactor for fluidization. The fluidized-bed reactor will be operated at temperatures up to 1,200° F. The char level in the reactor will be maintained by a char overflow pot connected to the reactor by two overflow tubes. Inert gas passing up through the central draft tube will transfer the product char from the reactor to a cyclone where the gas and solid phases are separated. The gaseous effluent will be subsequently scrubbed and filtered prior to being discharged.

A literature review has been conducted and the bench-scale batch reactor was modified to convert it from batch feed to continuous feed with respect to char. In addition, a second feed system was designed, involving the mixing of the residue with toluene to form a slurry which can be injected into the reactor as a spray.

PROGRESS DURING JULY-SEPTEMBER 1976

Four residue carbonization runs were conducted during the quarter. All runs were determined prematurely when plugs developed in the ½-inch line leading from the reactor to the cyclone. Some of the runs were termed partially successful in that no large agglomerates formed in the reactor. The temperature during these runs was 1200°-1230° F and the feed had a Wyodak char/residue ratio of 4:1.

During September, several design changes were made to the system to alleviate the solids carryover and plugging problems encountered in the previous tests. Following these modifications, a run was attempted but it did not achieve steady operation due to feeding

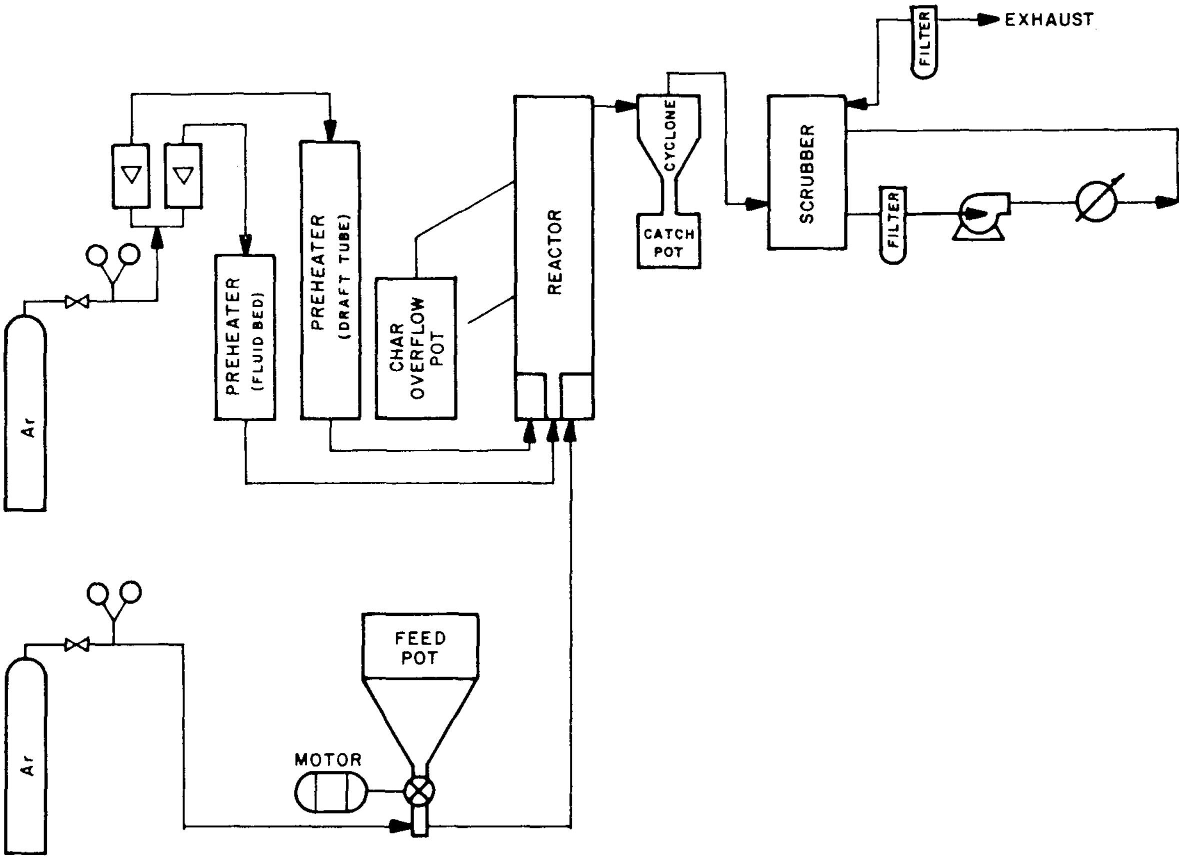


Figure X-1. FLOW DIAGRAM OF EVALUATION PROCESS

problems and a plug in the internal cyclone. These problems may have been caused by the 2:1 char/residue ratio, the lowest yet used in the reactor.

Two 1110° F agglomeration tests were also conducted during the quarter with mixtures of Wyodak char and residue in the ratios of 2:1 and 9:1. Agglomeration in both tests was slight and the agglomerates which did form were softer and more porous than those formed in previous tests with residue alone.

XI. ZINC HALIDE HYDROCRACKING PROCESS

CONOCO COAL DEVELOPMENT COMPANY
LIBRARY, PENNSYLVANIA

Project Site:	Library, Pennsylvania
Contract No.:	E(49-18)-1743
Total Funding:	$6,450,335 *ERDA: $5,650,335* *Industry: $800,000*

INTRODUCTION

Continental Oil Company, with the participation of Shell Development Company, a division of Shell Oil Company, is under contract to ERDA to develop the zinc halide hydrocracking process. The overall objective of the project is the production of clean liquid and gaseous fuels, of which approximately fifty percent will be distillate, equivalent to four barrels per ton of coal on a moisture and ash-free (MAF) basis.

Specifically, experimental studies will be performed involving both continuous bench-scale and process development unit work dealing with the investigation of zinc halide as a catalyst for the hydrogenation and hydrocracking of coal extract and subbituminous coal. Studies will also involve the development of an economical regeneration process for the efficient recovery of zinc halide from the spent melt. The schedule for the process is shown in Figure XI-1.

PROCESS DESCRIPTION

The zinc halide hydrocracking process is designed to maximize the conversion of subbituminous coal into light distillates by severe catalytic cracking. A schematic of the process is provided in Figure XI-2. The coal is dried and pulverized and then introduced into a feed tank where it is slurried with a process-derived recycle oil. The slurry feed is transferred to a hydrocracking reactor where it is mixed with hydrogen and the zinc chloride catalyst. In the reactor, the coal is cracked to distillates in the gasoline range, producing on distillation a clear 90 RON gasoline without further processing, and a small quantity of very low sulfur, low-nitrogen fuel oil. The gas is subsequently separated from the liquid in a product receiver. Spent catalyst is then fed to a regenerator for catalyst recovery.

PROGRESS DURING JULY-SEPTEMBER 1976

Summary

An existing continuous bench-scale hydrocracking unit (two-pounds-per-hour capacity) and a continuous fluidized-bed combustion unit for the regeneration of spent halide melt (five-pounds-per-hour capacity) were refurbished and reactivated. Initial break-in operation of the continuous bench-scale hydrocracker was done with coal extract.

Discussion of Activities

The bench-scale continuous regenerator was operated for extended periods to yield material balance

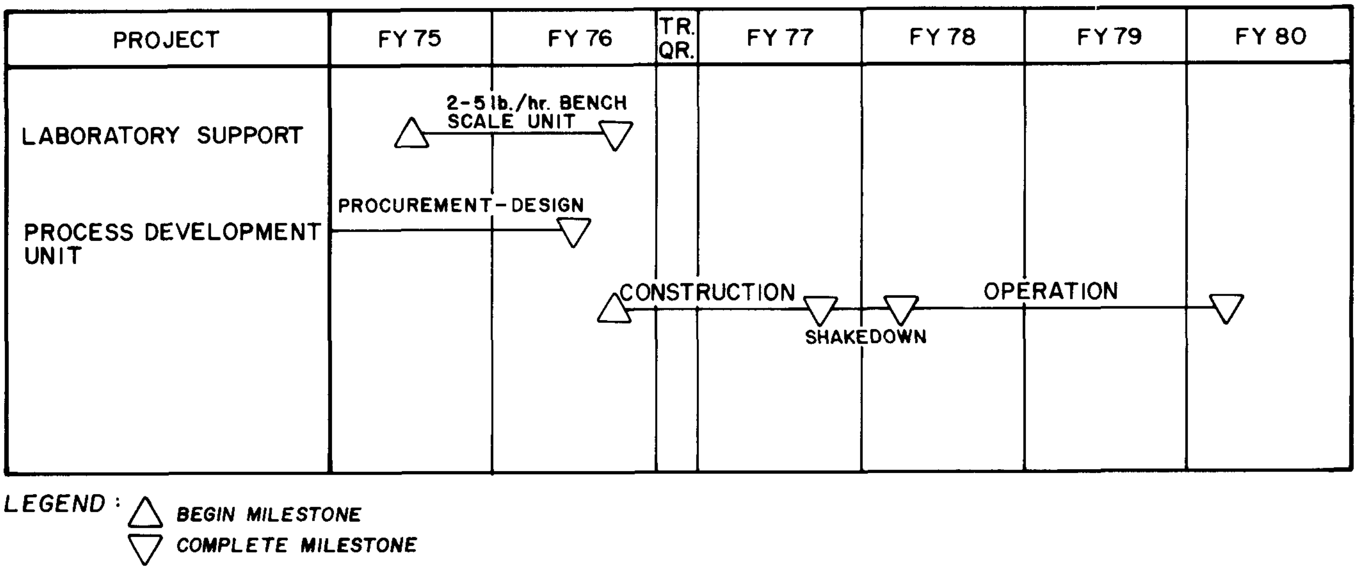


Figure XI-1. ZINC HALIDE HYDROPROCESSING PROJECT SCHEDULE

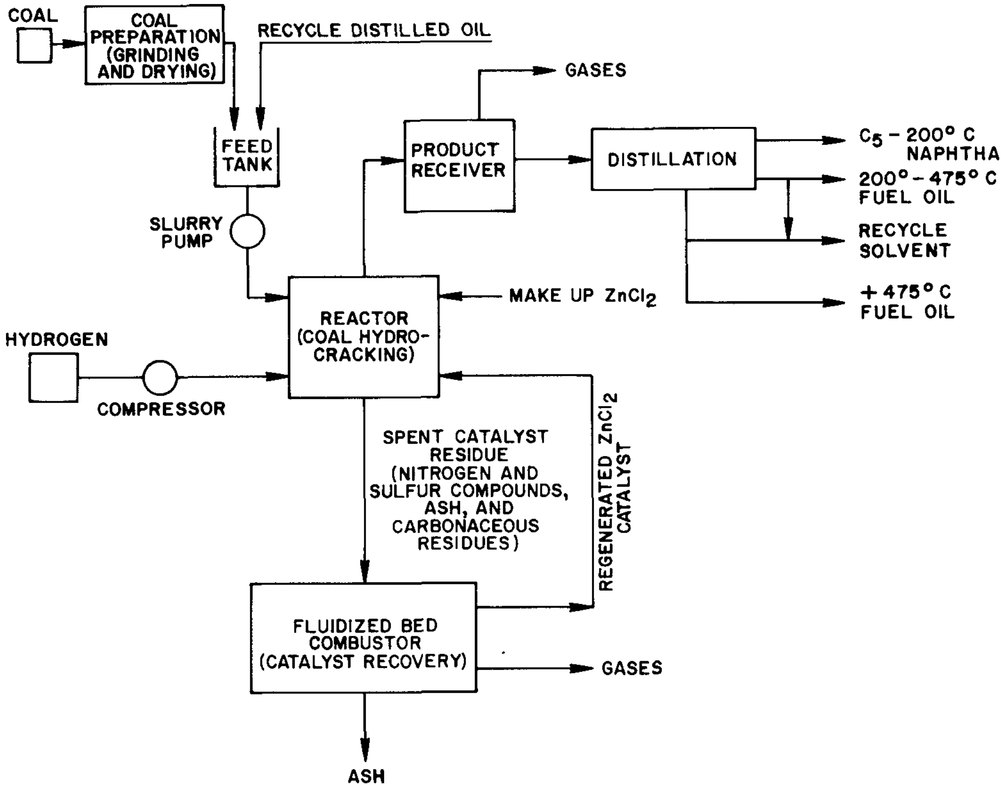


Figure X1-2. ZINC HALIDE HYDROCRACKING PROCESS FLOWSHEET data to determine zinc and chloride recoveries. Runs at various levels of excess air indicated that the unit was operable (no buildup of ash in the fluidized-bed) at 1700° and 1750° F. At 1800° F and 50 percent excess air, it was marginally operable, with ash accumulation similar to that at 1800° F and 15 percent excess air. The temperature at which no sintering occurs in the bed increases with hydrogen chloride concentration in the feed air. Bed sand was recovered and showed little reaction with coal ash. The unit will be modified and used for development of a continuous process to recover zinc chloride from coal ash collected in the regeneration cyclone.

The operating crew was transferred to the continuous bench-scale coal liquefaction unit, placing it in operating condition. The program to be conducted is a process variables study on hydrocracking subbituminous coal under various temperatures (725° to 800° F), hydrogen pressures (2,000 and 3,000 psig), and residence times (30 to 120 minutes).

Process Development Unit

A critical path diagram was designed to measure progress. Vessel design on the 100-pound-per hour process development unit (PDU) is complete and bids are being considered and orders placed. Notice of a delay in major vessel delivery was received, resulting in a two to three month delay in hydrocracker construction.

Laboratory Studies

Supporting laboratory studies were conducted on secondary recovery of zinc chloride from coal ash and on the kinetics of coal liquefaction using the zinc chloride system. A batch autoclave run designed to hydrocrack Colstrip coal showed that regenerated zinc chloride catalyst is as active as fresh catalyst.

XII. FLASH PYROLYSIS COAL LIQUEFACTION PROCESS

OCCIDENTAL RESEARCH CORPORATION
LA VERNE, CALIFORNIA

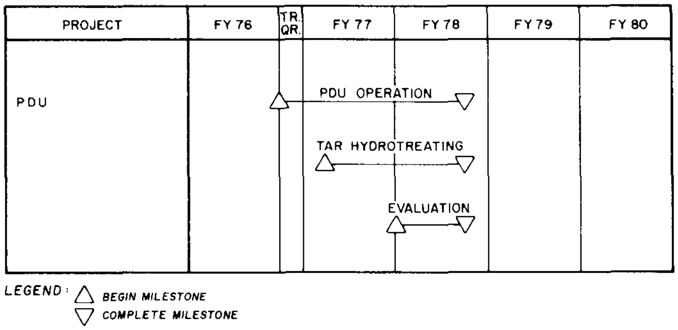
Plant Site: La Verne, California
Contract No.: E(49-18)-2244
Total Funding: $3,780,000
(100% ERDA)

INTRODUCTION

The Occidental Research Corporation (ORC), a wholly-owned subsidiary of Occidental Petroleum Corporation, will evaluate the commercial potential of its flash pyrolysis coal liquefaction process under the sponsorship of ERDA. Figure XII-1 provides the schedule for the project.

The main objectives of the program are to:

- Demonstrate that caking coals can be processed continuously in a specially designed single-stage pyrolysis reactor without oxidative pretreatment, and that this method will result in a significantly higher yield of liquids than other proposed pyrolysis processes.
- Conduct extended runs in the three-ton-per-day process development unit (PDU) in order to obtain steady state heat and material balances.
- Produce and recover large quantities of the primary tar, and to evaluate methods for upgrading this material to a clean fuel or synthetic crude oil.
- Continue development of specific areas of the pyrolysis and liquids collection systems to ensure a technologically sound basis for future scale-up.
- Obtain sufficient process and environmental data for detailed design of a larger plant and conduct an assessment of the potential commercial viability of the process.



gure XII-1. FLASH PYROLYSIS PROJECT SCHEDULE

PROCESS DESCRIPTION

The ORC process consists of rapidly pyrolyzing coal particles at a temperature of less than 1400° F in an entrained stream of hot coal char and a gas, substantially free of oxidizing constituents. The process features short residence time of coal particles thereby increasing the throughput per unit of cross sectional reactor area, and producing a relatively high yield of liquid products. Figure XII-2 provides a schematic flow diagram of the process.

After the coal is extensively milled and screened, it is transported to the pyrolysis reactor by heated

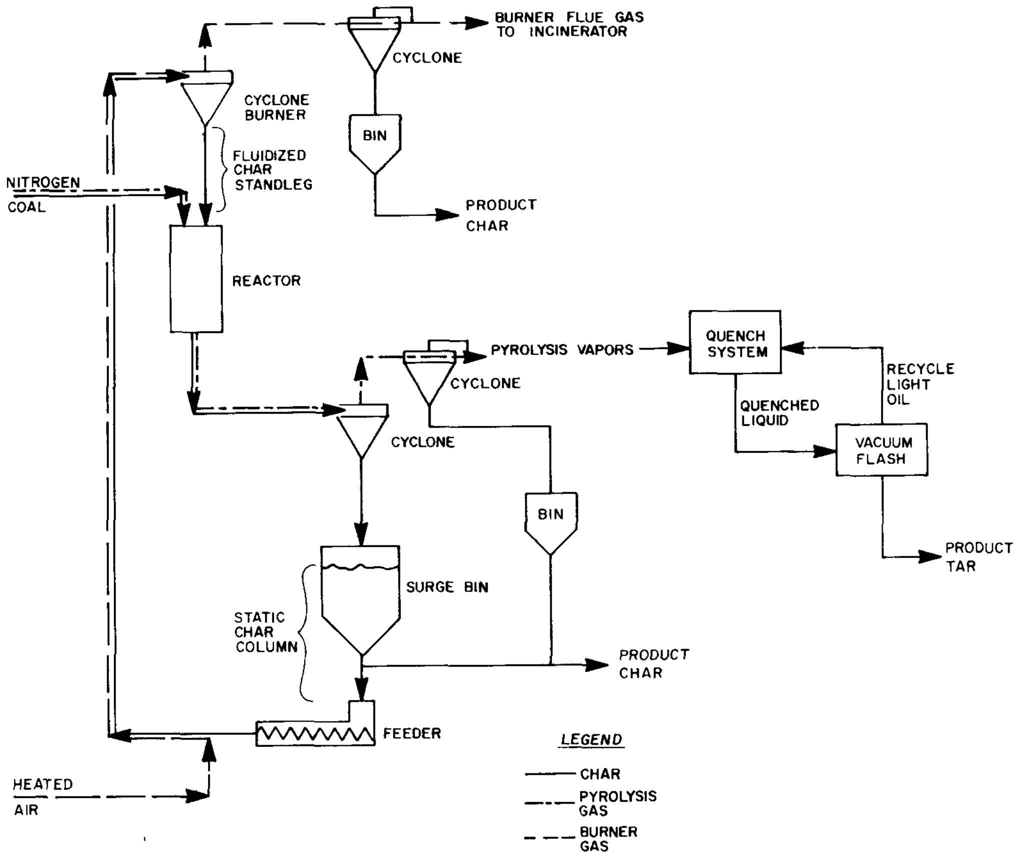


Figure XII-2. FLASH PYROLYSIS PROCESS FLOWSHEET

nitrogen at rates of up to 300 pounds per hour. In the reactor, the coal is mixed with recycled hot char which rapidly heats the coal to a pyrolysis temperature in the range of 950° to 1350° F. The char circulation rate ranges from five to ten times that of coal.

Char is separated from the pyrolysis vapors by a series of three cyclones. A portion of the char is cooled as product and the remainder is fed to the char heater where it is partially burned with air to raise its temperature to 1200° F to 1700° F for recycle to the reactor. The second and third stage cyclones remove fine char from the pyrolysis vapor prior to quenching in the tar collection system. This system consists of three subsections: two tar recovery stages and a vacuum flash unit.

In the first recovery stage, raw pyrolysis gas is quenched to about 210° F, most of the heavy tar vapors are condensed, and any entrained char is removed. The pyrolysis gas is cooled to approximately 80° F in the second recovery stage, where water and light oils are removed. The vacuum flash unit is used to separate the quench liquid and recovered tars into high and low boiling fractions, the high boiling fraction representing the main product tar.

Maintenance of a pressure balance between the char burner and coal pyrolysis sections is an important control requirement of the PDU pyrolysis system. The char, moving in a continuous cycle through both the burner and pyrolysis sections, must be isolated from the gases flowing through these sections. The char is used to form gas seals at two points between the process sections. As the seals are particularly sensitive to the pressure differential across them, a key control item is the balancing of the pressures of the gas streams to maintain these gas seals.

HISTORY OF THE PROJECT

ORC constructed their three-ton-per-day PDU fr development of other pyrolysis processes, and revis and improved the unit for use in this contract. ORC and the Commonwealth of Kentucky have a cost-sharing joint venture aimed at providing a detailed design for a 250-ton-per-day pilot plant using the process.

PROGRESS DURING JULY-SEPTEMBER 1976

As the process development unit (PDU) had been extensively modified since its operation prior to the inception of this contract, various subsystem tests were performed before operating the revised facility. The key tests involved circulation of hot char through the units and supplying heat by the combustion of some of the char. Shakedown and operation of the unit will be demonstrated with subbituminous non-caking coal prior to operation with caking coals.

The modified PDU was tested under normal operating conditions during its first run in August. Feed coal capacity of three-tons-per-day, design char circirculation rate of 1000-1600 pounds-per-hour, and heat provided by char combustion achieved the main run objective of demonstrating the plant to be operable at normal capacity and process conditions. Char circulation stability test demonstrated good control of the system pressure balance.

Work during the remainder of the quarter was devoted to modification of the PDU to improve char circulation in the pyrolysis system. Installation of a revised tar collection system began, and pre-startup check-out of the first and second recovery stages was started during September.

Characterization of the tar from the first bench-scale pyrolysis run has begun and a sulfiding procedure for the hydrotreating catalysts has been selected. The method of hydrotreating will be to contact the tar with high pressure hydrogen in the presence of an appropriate, commercially available, catalyst. A continuous fixed-bed unit will be used in the sulfiding of fresh catalysts, regeneration of used catalysts, and for activity tests. A batch autoclave will be used for screening of new catalysts, preliminary hydrotreating work, and the evaluation of used catalysts. Finally, the hydrotreating work will be conducted in a continuous fixed-bed unit. A high pressure test facility is now under construction and the hydrotreating units are to be moved into the facility when it is completed.

XIII. OPERATION OF SOLVENT-REFINED COAL (SRC) PILOT PLANT

SOUTHERN COMPANY SERVICES, INC.
BIRMINGHAM, ALABAMA

Plant Site:	Wilsonville, Alabama
Contract No.:	E(49-18)-2270
Total Funding:	$8,729,892 *ERDA: $5,741,892* *Industry: $2,988,000*

INTRODUCTION

The operation of the solvent refined coal (SRC) pilot plant at Wilsonville is jointly sponsored by ERDA and the Electric Power Research Institute (EPRI). The schedule for the project is shown in Figure XIII-1. The objectives of the project are to:

- Correlate the results of operation at the Wilsonville SRC pilot plant with those from the Ft. Lewis, Washington, SRC pilot plant.
- Test several coals of various ranks and locations to determine the effects of process variables on product yield and quality.
- Evaluate SRC process improvements.
- Evaluate improvements in equipment and operations.

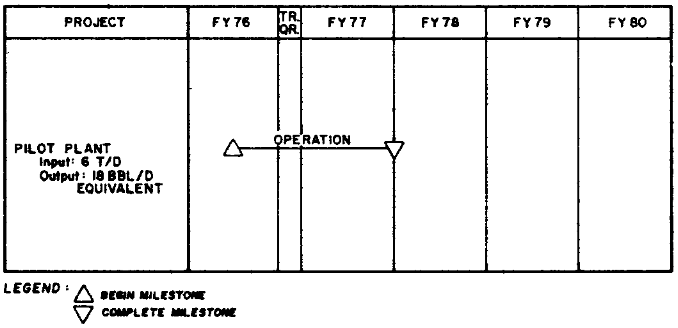


Figure XIII-1. SRC PILOT PLANT OPERATION SCHEDULE

PROCESS DESCRIPTION

The SRC process converts high-sulfur, high-ash coals to a clean distillate fuel and a clean high-Btu gas. A schematic of the process is shown in Figure XIII-2. The pulverized coal is mixed with two to three parts process-derived solvent. Hydrogen is then added to the coal-solvent slurry. The mixture is pumped to a preheater and then fed into a single-stage reactor or dissolver which operates at 800°-875° F, and a total pressure of approximately 1700 psig. Under these conditions, approximately 93 percent of the carbonaceous material in the coal feed is dissolved during the residence time of approximately 45 minutes. The product gases from the dissolver are separated and sent to a clean-up system to remove the hydrogen sulfide (about 60 percent of the organic sulfur in the coal is converted to hydrogen sulfide) and convert it to elemental sulfur. The liquid slurry is then subjected to a mineral separation step wherein the undissolved solids are removed. Process solvent is recovered from the coal solution by distillation and recycled to slurry the coal

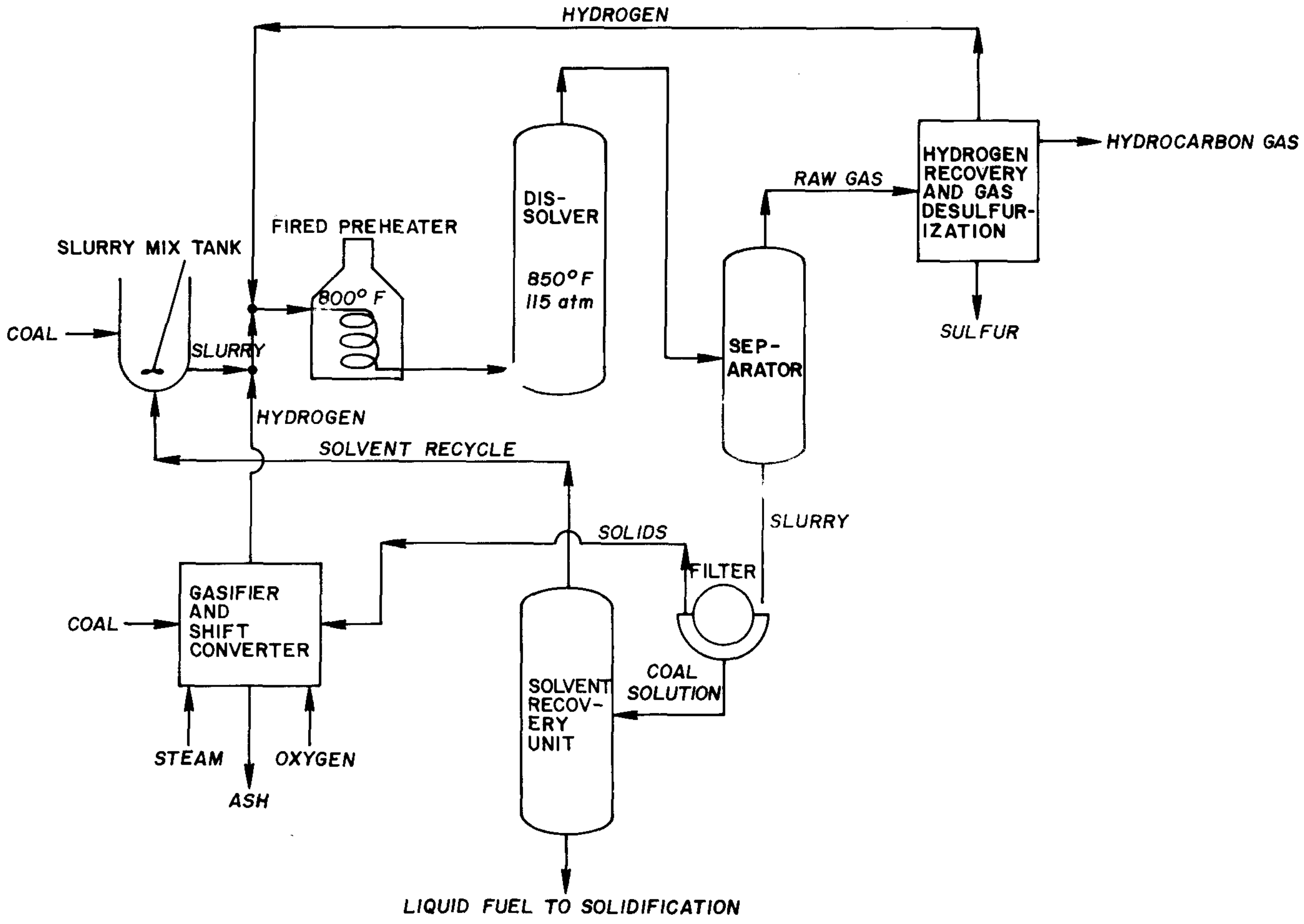


Figure XIII-2. SRC PROCESS FLOW DIAGRAM

feed. The residue which remains is the solvent refined coal product, which solidifies at about 350° F.

HISTORY OF THE PROJECT

In March 1972, the Edison Electric Institute and the Southern Electric System began a joint project to study the key steps in the SRC process. Consequently, Catalytic, Inc. designed, built, and is operating the six-ton-per-day pilot plant. The facility was completed in August 1973. The Electric Power Research Institute (EPRI) assumed the responsibilities of the Edison Electric Institute for utility industry sponsorship in April 1973.

The pilot plant began operation in January 1974, and by the end of 1975, had been in operation for more than 7,800 hours, including periods of 45 and 75 days of sustained operation. One subbituminous and four bituminous coals have been tested. SRC product meeting plant specifications of 0.16 percent maximum ash and 0.96 percent maximum sulfur has been produced from each coal. A fuel meeting these specifications and having a heating value of 16,000 Btu per pound could be used to achieve compliance with most emission limits and EPA's "new source performance standards."

In January 1976, ERDA joined EPRI as a co-sponsor of the pilot plant operation. To stimulate operation at the Ft. Lewis SRC pilot plant, a mixture of coals from the Kentucky No. 9 and 14 seams were used for plant operation. Coal feed rates as high as 75 pounds of coal per hour per cubic foot of dissolver volume, almost three times the design feed rate, we achieved. Material balance data for ten runs were obtained, allowing correlation of the results from both the Wilsonville and Ft. Lewis pilot plants. Empirical models to aid in scaling up to larger plants were developed for predicting conversion, sulfur removal, and filtration rates for operation with Kentucky No. 9 and 14 coals.

PROGRESS JULY-SEPTEMBER 1976

The plant was shut down from July 10 to August 4 for maintenance and modifications. In early August, tests began on Illinois No. 6 coal. In the separation area, attempts were made to improve filtering operation with new filter precoats and a new filter draining procedure.

XIV. DONOR SOLVENT LIQUEFACTION PROCESS

EXXON RESEARCH AND ENGINEERING
COMPANY
BAYTOWN, TEXAS

Contract No.: E(49-18)-2353
Total Funding: $12,740,360

INTRODUCTION

Under the terms of this contract, details of prior development work on the Exxon Donor Solvent (EDS) process conducted between 1966 and January 1976 are confidential. The program schedule for this project is shown in Figure XIV-1. The objectives of this contract are to:

- Expand the process data base from experimental units of up to one-ton-per-day to confirm the design for the construction of a 250-ton-per-day pilot plant.
- Provide data on the compatibility of coal-derived products with petroleum liquids, and on the storage behavior and end use potential of raw and hydrotreated coal liquids as well as blends of coal liquids and petroleum liquids.
- Provide data for the design of large scale coking and gasification units integrated with the EDS process.
- Provide economic estimates for conceptual commercial plants.

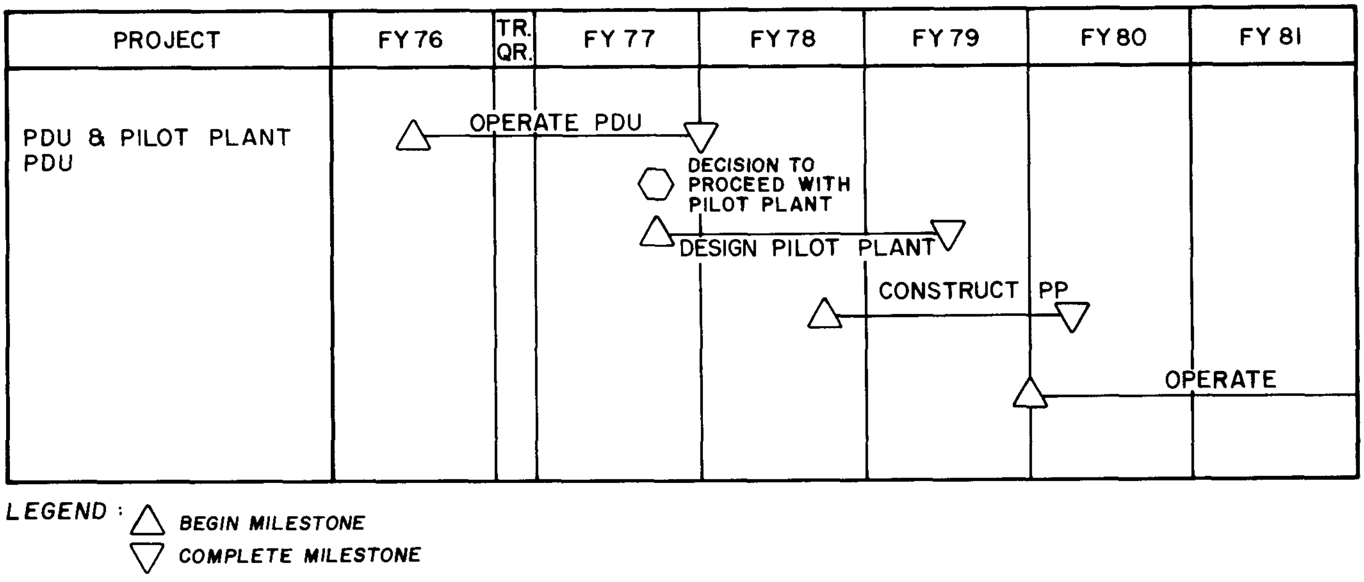


Figure XIV-1. DONOR SOLVENT PROJECT SCHEDULE

- Provide a data base to improve the design scale-up capability of critical components and plant sections.

PROCESS DESCRIPTION

Steps in the Donor Solvent Process involve engineering and design technology similar to that practiced in the petroleum industry. The process was designed to permit process control, allow for feeding different coals, and allow product distribution to vary based on market demand. In this process, the donor solvent provides a mechanism to allow intimate contact between hydrogen and dispersed coal fragments. The donor hydrogen content of the spent solvent is restored after liquefaction in the solvent hydrogenation reactor. This step also reduces the sulfur, nitrogen, and oxygen contents of the recycle solvent. The donor hydrogen content is the major parameter used to evaluate the effectiveness of various catalysts and processing conditions to produce good quality donor solvent. A schematic of the process is provided in Figure XIV-2.

The coal is dried and ground to 30-mesh and mixed with recycle solvent to form a slurry. Hydrogen is normally preheated separately and combined with preheated slurry at the reactor inlet. An alternate mode of operation is to combine the hydrogen with the slurry before preheating.

After liquefaction, the product goes to the first stage of separation. Water is removed, hydrogen is recycled, and heavy liquid products are sent to the vacuum separation section.

The primary vacuum flash tower removes the mineral matter and unreacted coal. The secondary vacuum tower is used to remove a heavy distillate which becomes the endpoint of the recycle solvent. This overhead from the secondary vacuum flash tower is then combined with the lighter liquid stream recovered from the liquefaction reactor separator system and fed to the solvent hydrotreating section, consisting of fixed-bed catalytic reactors. The solvent and naphtha from the solvent hydrotreating section are then separated by fractionation. Most of the solvent is recycled, as is the hydrogen, after undergoing cleanup in a monoethanolamine (MEA) scrubbing system.

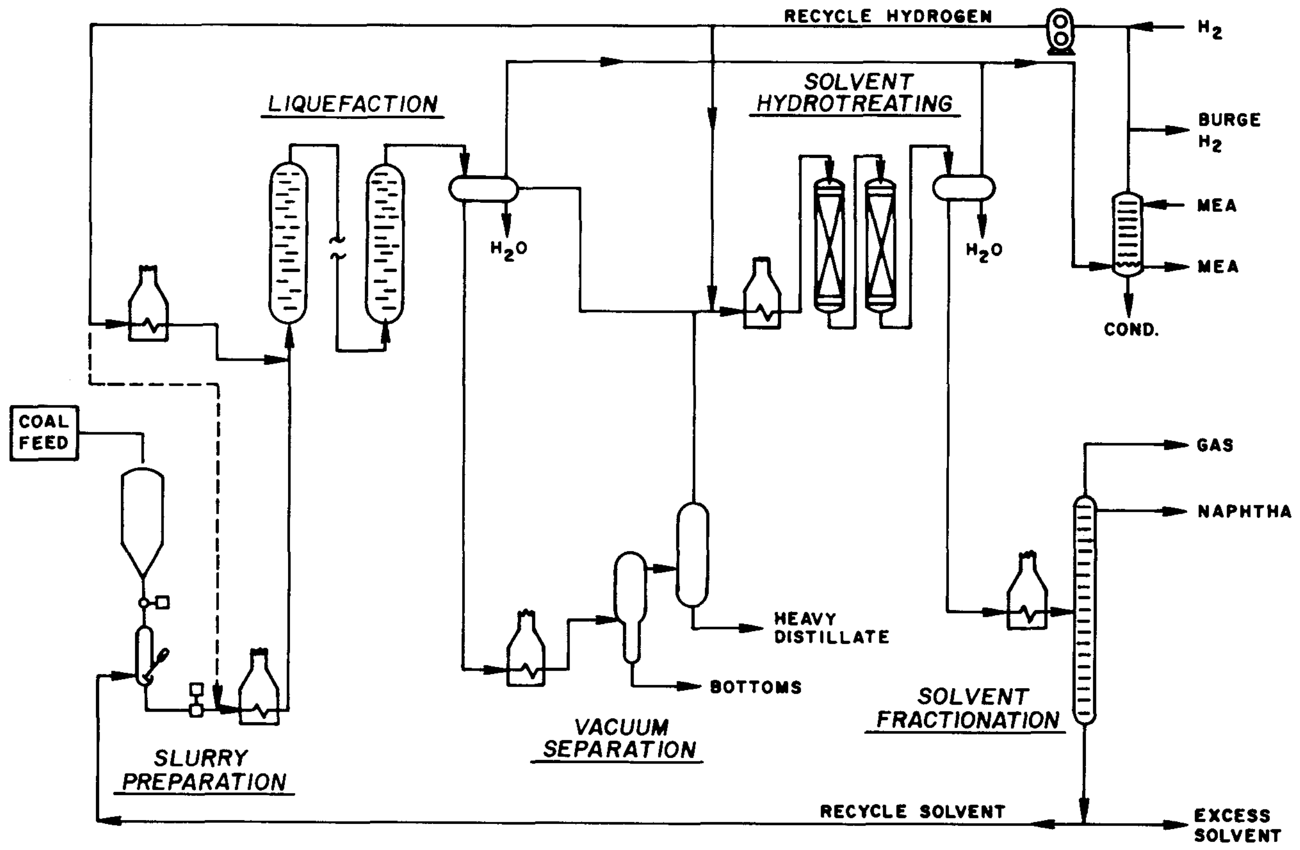


Figure XIV-2. DONOR SOLVENT LIQUEFACTION PROCESS

HISTORY OF THE PROJECT

Exxon's research on the EDS process began in 1966 and has resulted in a commercial study design. A semi-empirical kinetic model of liquefaction yields from Illinois coal has been expanded to include recent results from high-severity liquefaction experiments. Progress has been made in the development of a computer model of the entire integrated EDS process. The model will simulate, and link together in a single interfacing package, the six major processing sections: liquefaction, solvent hydrotreating, coking, cryogenic hydrogen recovery, steam/methane reforming, and final product separation.

Evaluation of several process alternatives have begun, including development of a computerized process alternative model to be used as a basic tool for the process engineering and economic studies which are planned. Engineering studies have been initiated to identify and develop equipment and engineering data needed for a safe, operable, and reliable EDS commercial plant.

PROGRESS DURING JULY-SEPTEMBER 1976

As two-stage liquefaction is a potential improvement to the basic EDS process, tests continued on the two-stage 100-pound-per-day recycle coal liquefaction unit. Comparative tests were made in the 50-pound-per-day recycle coal liquefaction unit on both North Dakota lignite and Illinois No. 6 coal. The EDS process alternatives computer model is nearing completion.

XV. FLUID COKING OF COAL LIQUEFACTION RESIDUES

EXXON RESEARCH AND ENGINEERING COMPANY

Contract No.: E(49-18)-2422
Total Funding: $11,312,470

INTRODUCTION

Exxon Research and Engineering Company, under the sponsorship of ERDA, will study the potential of fluid coking to recover distillate liquids. The primary objective of this contract is to evaluate the applicability of current commercial fluid coking technology to processing of residues from one selected coal liquefaction process. The program schedule for this project is shown in Figure XV-1.

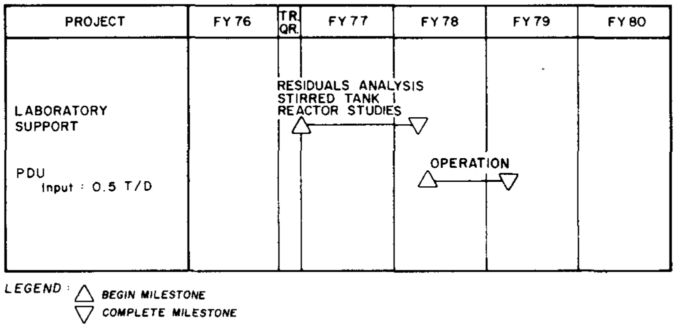


Figure XV-1. FLUID COKING PROJECT SCHEDULE

PROGRAM DESCRIPTION

The treatment of residual, solids-containing liquids produced by various coal liquefaction processes is important in determining the overall economic viability of the process. Solids are removed from primary coal liquids by methods such as hydrocyclones, vacuum distillation, and solvent de-ashing. The solids-containing residues from these operations may contain additional valuable liquid products which may be recovered by fluid coking.

Three samples of coal liquid residue produced in ERDA-sponsored coal liquefaction projects will be analyzed and processed using bench-scale equipment. One of these selected coal liquid residues will be processed in a nominal two-barrel-per-day fluidized-bed pilot unit.

PROGRESS DURING JULY-SEPTEMBER 1976

The first of three candidate coal liquefaction residues to be evaluated was H-Coal solvent precipitation underflow. Analytical characterizations and a bench-scale screening study of the coke, liquid, and gas yields were started on September 16. This represented a slight delay due to late shipment of the sample.

XVI. CHARACTERIZATION OF COAL LIQUIDS

UOP, INC.
DES PLAINES, ILLINOIS

Contract No.: E(49-18)-2010
Total Funding: $369,914

INTRODUCTION

UOP, Inc. is under contract to ERDA to demonstrate the nature and degree of possible upgrading of primary coal liquids using technology now available in the petroleum refining industry. The objectives of this contract are to:

- Evaluate the processability of the primary products from each of the current coal liquefaction processes.
- Estimate the optimum conditions for processing coal liquids in current hydrorefining units.
- Correlate bench-scale and analytical data in order to estimate processability on the basis of analytical data alone.
- Assess the economic viability of feeding raw coal liquids to existing refineries in order to produce a full slate of finished products.
- Identify a preferred method for processing coal liquids in an existing refinery.
- Identify areas where research and development appear to be essential in order to realize commercial utilization of coal liquids as refinery feedstocks.

PROJECT DESCRIPTION

Product liquids from coal liquefaction plants differ from petroleum crude in that they are deficient in hydrogen, have a higher polynuclear aromatic content, higher level of inorganic impurities, and are generally heavier stocks.

The response of existing full-scale refineries to almost any petroleum crude feedstock can be accurately predicted from chemical and physical characteristics of that stock. These characteristics, along with bench-scale refining unit test data, have been corroborated by full scale refinery experience. This approach, formally termed "characterization", will be used to estimate the result of processing coal liquids in existing oil refineries.

PROGRESS DURING JULY-SEPTEMBER 1976

Six primary coal liquids were filtered and hydrotreated under commercial conditions, using a fixed bed of commercial catalyst. These included Synthoil, H-Coal hydroclone underflow, and four SRC process streams. In all cases, catalytic hydrotreating substantially eliminated sulfur and benzene-insoluble matter.

XVII. COMMERCIAL SCALE EBULLATED-BED HYDROPROCESSING OF SOLVENT-REFINED COAL EXTRACT

CITIES SERVICE RESEARCH AND
DEVELOPMENT COMPANY
CRANBURY, NEW JERSEY

Plant Site: Lake Charles, Louisiana
Contract No.: E(49-18)-2038
Total Funding: $591,122

INTRODUCTION

Cities Service, under ERDA sponsorship, is determining the operating conditions and modifications necessary to utilize an existing LC-Fining unit (ebullated-bed hydroprocessing unit) to upgrade a production quantity of Solvent Refined Coal (SRC) extract. The specific objectives of this contract are to:

- Determine the ebullated-bed conditions required to hydrotreat SRC extract at severity levels ranging from desulfurization to extensive hydrocracking.
- Upgrade a production quantity of SRC extract from the Ft. Lewis pilot plant using commercial-scale ebullated-bed hydroprocessing equipment.
- Demonstrate that the SRC process and ebullated-bed hydroprocessing can be combined to provide a valuable liquid fuel or refinery feedstock from coal and thereby accelerate commercial utilization of the SRC process.

PROCESS DESCRIPTION

Solvent Refined Coal (SRC) extract is a hydrocarbon mixture containing significant quantities of oxygen, nitrogen, sulfur, and ash. SRC extract composition and properties are similar to heavy sour petroleum crude and process-derived residua. These are problem stocks which must be upgraded by treatment with hydrogen before they can be further refined to finished products. Ebullated-bed hydrotreating can upgrade these stocks and maintain continuous operation for extended periods of time as well. A flow diagram of the process is shown in Figure XVII-1.

The SRC extract, together with hydrogen, is fed upward through the reactor as a liquid/gas mixture at a velocity such that the catalyst is in continuous motion. Part of the reactor effluent is recycled back through the reactors to maintain the necessary liquid velocity through the reactor system (the ebullated-bed). Because of the movement of catalyst particles in the liquid/gas medium, deposition of tar and coke is minimized and fine solids entrained in the feed will not lead to reactor plugging. Reactor effluent is cooled by exchange, and a vapor-liquid separation is made. After scrubbing, the hydrogen is recycled and liquid product is either stored or further fractionated.

Bench-scale pilot plant studies will be performed for the purpose of determining operational conditions for the commercial unit. Engineering studies will define the degree of effort and resources required to adapt an ebullated-bed unit to process a specified quantity of SRC extract.

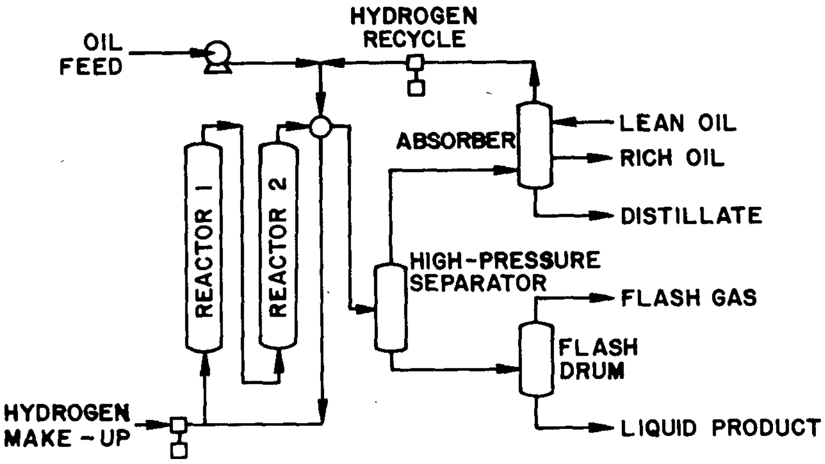


Figure XVII-1. EBULLATED-BED HYDROPROCESSING PROCESS SCHEMATIC

PROGRESS DURING JULY-SEPTEMBER 1976

Pilot plant studies included the completion of analytical work on feed components. Possible commercial feed blends are being studied to determine the highest SRC to solvent ratio having low solids formation and meeting viscosity requirements. Initial preparations to ready the LC-Fining unit for operation have begun.

GLOSSARY

absorption—an imprecise term suggesting the taking up of one substance by another by either a physical process or a chemical combination.

acceptor—calcined carbonate that absorbs carbon dioxide evolved during gasification, liberating heat.

acid gas removal—the process of selectively removing hydrogen sulfide and carbon dioxide from a gas stream.

activated carbon—carbon obtained by carbonization in the absence of air, preferably in a vacuum; has the property of absorbing large quantities of gases, solvent vapors; used also for clarifying liquids.

adiabatic—any process where heat is neither given off nor absorbed.

adsorption—the process by which the surface of a solid or liquid attracts and holds any atom, molecule, or ion from a solution or gas with which it is in contact.

agglomerate—assemblage of ash particles rigidly joined together, as by partial fusion (sintering).

anthracite coal—hard coal containing 86 to 98 percent fixed carbon and small percentages of volatile material and ash.

API—American Petroleum Institute.

API gravity—a scale adopted by the API for measuring the density of oils;

$$° \text{API} = \frac{141.5}{\text{Specific gravity, } 60°\text{F}/60°\text{F}} - 131.5$$

aromatic hydrocarbon—a cyclic hydrocarbon containing one or more six-carbon (benzene) rings.

ash—solid residue remaining after the combustion of coal.

ASTM—American Society for Testing Materials.

autoclave—a vessel, constructed of thick-walled steel, for carrying out chemical reactions under high pressures and temperatures.

bench-scale unit—a small-scale laboratory unit for testing process concepts and operating parameters as a first step in the evaluation of a process.

binder—carbon products, tars, etc., used to impart cohesion to the body to be formed; a coal-extract binder may be used to prepare formed-coke pellets from non-coking coals.

bituminous coal—a broad class of coals containing 46 to 86 percent fixed carbon and 20 to 40 percent volatile matter.

blow down—periodic or continuous removal of water from a boiler to prevent accumulation of solids.

bottoming cycle—the lower temperature thermodynamic power cycle of a combined-cycle system.

Btu—British thermal unit, the quantity of energy required to raise the temperature of one pound of water one degree Fahrenheit.

BTX—benzene, toluene, xylene; aromatic hydrocarbons.

caking—the softening and agglomeration of coal as a result of the application of heat.

ination—the process of heating a solid to a high temperature to cause the decomposition of hydrates and carbonates.

calorific value—the quantity of heat obtained by the complete combustion of a unit mass of a fuel under prescribed conditions.

carbon fiber—fine filaments of carbon about eight microns in diameter which are used in composite materials, being bound with resins.

carbonization—destructive heating of carbonaceous substances with the production of a solid, porous residue, or coke, and the evolution of a number of volatile products. For coal, there are two principal classes of carbonization, high-temperature coking (about 900° C) and low-temperature carbonization (about 700° C).

catalyst—a substance that accelerates the rate of a chemical reaction without itself undergoing a permanent chemical change.

centrifuge—an apparatus rotating at high speed which utilizes the centrifugal force generated to separate materials of different densities, e.g., undissolved residue from coal solution in the SRC process.

char—the solid residue remaining after the removal of moisture and volatile matter from coal.

Claus process—industrial method of obtaining elemental sulfur through the partial oxidation of gaseous hydrogen sulfide in air followed by catalytic conversion to molten sulfur.

coal—a readily combustible rock containing more than 50 weight percent and more than 70 volume percent of carbonaceous material including inherent moisture, formed from compaction and induration of variously altered plant remains similar to those in peat.

coalification—metamorphosis of vegetable debris into coal.

coke—strong porous residue consisting of carbon and mineral ash formed when bituminous coal is heated in a limited air supply or in the absence of air. Coke may also be formed by thermal decomposition of petroleum residues.

coke breeze—the fine screenings of crushed coke usually passing a ½-inch or ¾-inch screen opening.

combined cycle—two sequential thermodynamic power conversion systems operating at different temperatures.

combustion gas—gas formed by the combustion of coal, e.g., burning.

combustor—a vessel in which combustion takes place.

coupon—a polished metal strip used to measure the rate of corrosion of the metal in a specific gaseous or liquid environment.

cracking—the partial decomposition of high-molecular-weight organic compounds into lower-molecular-weight compounds, generally as a result of high temperatures.

crude gas—impure gas produced in a gasifier.

culm—the waste or slack from anthracite mines or preparation plants consisting of fine coal, coal dust, and dirt.

cyclone separator—essentially a settling chamber to separate solid particles from a gas, in which gravitational acceleration is replaced by centrifugal acceleration.

degasification—a process for removing naturally occurring methane from coal seams.

delayed coking—a process wherein coal is subjected to a long period of carbonization at moderate temperatures to form coke.

demineralization—removal of mineral matter (ash) from coal by solvent extraction, usually under hydrogen atmosphere.

depolymerization—the change of a large molecule into simpler molecules usually accompanied by the substitution of hydrogen for oxygen in the molecular structure.

destructive distillation—the distillation of coal accompanied by its thermal decomposition.

desulfurization—the removal of sulfur from hydrocarbonaceous substances by chemical reactions.

devolatilization—the removal of a portion of the volatile matter from medium- and high-volatile coals.

diatomaceous earth—a yellow, white, or light-gray, siliceous porous deposit made up of opaline shells of diatoms; used as a filter aid, paint filler, adsorbent, abrasive, and thermal insulator. Also known as kieselguhr.

diatomite—See Diatomaceous Earth.

dissolution—the taking up of a substance by a liquid with the formation of a homogeneous solution.

distillation—a process of vaporizing a liquid and condensing the vapor by cooling; used for separating liquids into various fractions according to their boiling points or boiling ranges.

dolomite—a carbonate of calcium and magnesium having the chemical formula $CaMg(CO_3)_2$.

Dowtherm—trademark for a series of eutectic mixtures of diphenyl oxide and diphenyl used as high-temperature heat-transfer fluids.

ebullated bed—gas containing a relatively small proportion of suspended solids, bubbles through a higher density fluidized phase with the result that the system takes on the appearance of a boiling liquid.

economizer—heat exchanging mechanism for recovering heat from flue gases.

effluent gas—gas given off from a process vessel.

elutriation—the preferential removal of the small constituents of a mixture of solid particles by a stream of high-velocity gas.

endothermic reaction—a process in which heat is absorbed.

enthalpy change—the increase or decrease in heat content of a substance or system which accompanies its change from one state to another under constant pressure.

entrained bed (flow)—a bed in which solid particles are suspended in a moving fluid and are continuously carried over in the effluent stream.

eutectic—that combination of two or more components which produces the lowest melting temperature.

exothermic reaction—a process in which heat is liberated.

extraction—a method of separation in which a solid or solution is contacted with a liquid solvent (the two being essentially mutually insoluble) to transfer components into the solvent.

extractive coking—similar to delayed coking process, with the emphasis on high tar yields to produce liquids.

filter aid—finely divided solids used to increase efficiency of filtering.

filter cake—the moist residue remaining from the filtration of a slurry to produce a clean filtrate.

filtrate—a liquid free of solid matter after having passed through a filter.

filtration—the separation of solids from liquids by passing the mixture through a suitable medium, e.g., cloth, paper, diatomaceous earth.

Fischer assay—method for determining the tar and light oil yields from coal or oil shale; conducted in a retort under an inert atmosphere with a prescribed increase in temperature to 500° C.

Fischer-Tropsch catalyst—catalysts developed for the catalytic synthesis of liquid fuels from coal-derived synthesis gas; catalysts contain principally iron, cobalt, nickel, or ruthenium.

Fischer-Tropsch process—method of hydrogenating mixtures of carbon monoxide and hydrogen produced from coal, lignite, or natural gas by means of steam, at 1–10 atmospheres and 360–410° F to yield liquid and gaseous fuels, and a wide spectrum of industrial chemicals.

fixed-bed—stationary solid particles in intimate contact with fluid passing through them.

fixed carbon—the solid residue, other than ash, obtained by destructive distillation; determined by definite prescribed methods.

flash carbonization—a carbonization process characterized by short residence times of coal in the reactor to optimize tar yields.

flue gas—gaseous combustion products.

fluidization (dense phase)—the turbulent motion of solid particles in a fluid stream; the particles are close enough as to interact and give the appearance of a boiling liquid.

fluidization (entrained)—gas-solid contacting process in which a bed of finely divided solid particles is lifted and agitated by a rising stream of gas.

fluidized-bed—assemblage of small solid particles maintained in balanced suspension against gravity by the upward motion of a gas.

fly ash—a fine ash from the pulverized coal burned in power station boilers, or entrained ash carried over from a gasifier.

fractionation—distillation process for the separation of the various components of liquid mixtures.

freeboard—the space in a fluidized-bed reactor between the top of the bed and the top of the reactor.

free swelling index—a standard test that indicates the caking characteristics of coal when burned as a fuel.

Friedel-Crafts reaction—a substitution reaction, catalyzed by aluminum chloride in which an alkyl (R-) or acyl (RCO-) group replaces a hydrogen atom of an aromatic nucleus to produce a hydrocarbon or a ketone.

fuel cell—a galvanic cell in which the chemical energy of a conventional fuel is utilized to produce electricity.

fuel gas—low heating value (150–350 Btu/scf) product generally utilized on site for power generation or industrial use.

gasification of coal—the conversion of solid coal into a gaseous form by various chemical reactions with steam.

gasifier—a vessel in which gasification occurs, usually utilizing fluidized-bed, fixed-bed, or entrained-bed units.

heat capacity—quantity of heat required to raise the temperature of one pound of a substance one degree Fahrenheit.

high-Btu gas—a gas having a heating value of 900 to 1,000 Btu per standard cubic foot, which approaches the value for natural gas.

higher-heating value (HHV)—the heat liberated during a combustion process in which the product water vapor is condensed to a liquid and the heat of condensation is recovered.

hydroclone—a small cyclone extractor for removal of suspend solids from a flowing liquid by means of the centrifug forces set up when the liquid is made to flow through a tight conical vortex.

hydrocoking—coking of tars, SRC, etc., under hydrogenating conditions to form liquid products.

hydrocracking—the combination of cracking and hydrogenation of organic compounds.

hydrogasification—gasification that involves the direct reaction of fuels with hydrogen to optimize formation of methane.

hydrogenation—chemical reactions involving the addition of gaseous hydrogen to a substance in the presence of a catalyst under high temperatures and pressures.

hydrogen donor solvent—solvent, such as anthracene oil, tetralin (tetrahydronaphthalene), decalin, etc., which transfers hydrogen to coal constituents causing depolymerization and consequent conversion to liquid products of lower boiling range which are then dissolved by the solvent.

hydrotreating—a process to catalytically stabilize petroleum or other liquid hydrocarbon products and/or remove objectionable elements from products or feedstocks by reacting them with hydrogen.

ideal gas—any gas whose equation of state is expressed by the ideal gas law, namely $PV = nRT$ where P is the pressure, V is the volume, R is the gas constant, T is the absolute temperature, and n = number of moles.

ignition temperature—the minimum temperature necessary to initiate self-sustained combustion of a substance.

industrial gas—See Fuel Gas.

inerts—constituents of a coal which decrease its efficiency in use, e.g., mineral matter (ash) and moisture in fuel for combustion.

in situ—in its original place, e.g., underground gasification of a coal seam.

intermediate-Btu gas—synthesis gas product with a higher heating value between 350 and 500 Btu per standard cubic foot.

lignite—brownish-black coal containing 65–72 percent carbon on a mineral-matter-free basis, with a rank between peat and subbituminous coal.

limestone—sedimentary rock containing 50 percent carbonate (CO_3) of lime or magnesia. Chemical formula (for culcite limestone) is $CaCO_3$.

liquefaction—conversion of a solid to a liquid; with coal, this appears to involve the thermal fracture of carbon-carbon and carbon-oxygen bonds, forming free radicals. These radicals abstract hydrogen atoms yielding low molecular weight gaseous and condensed aromatic liquids.

liquefied petroleum gas (LPG)—those hydrocarbons that have a vapor pressure (at 70° F) slightly above atmospheric (such as propane and butane); kept in liquid form under a pressure higher than 1 atm.

lock hopper—a mechanical device that permits the introduction of a solid into an environment of different pressure.

low-Btu gas—a gas having a heating value up to 350 Btu per standard cubic foot.

lower heating value—the heat liberated by a combustion process assuming that none of the water vapor resulting from the process is condensed, so that its latent heat is not available.

MAF—moisture and ash-free; a term that relates to the organic fraction in coal.

mesh—measure of fineness of a screen, e.g., a 400-mesh sieve has 400 openings per linear inch.

thanation—the production of methane (CH_4) from carbon monoxide or dioxide and hydrogen.

methane—CH_4, a colorless, odorless, and tasteless gas, lighter than air; the chief component of natural gas.

methanol—methyl alcohol, CH_3OH.

micron—a unit of length equal to one millionth of a meter; 10^{-6} meter.

moving bed—particlized solids in a process vessel that are circulated (moved) either mechanically or by gravity flow.

natural gas—naturally occurring gas extracted from sedimentary structures consisting mainly of methane and having a higher heating value of approximately 1,050 Btu per standard cubic foot.

noncoking coal—a coal that does not form coke under normal coking conditions.

olefinic hydrocarbon—a class of unsaturated hydrocarbons containing one or more double bonds and having the general chemical formula C_nH_{2n}.

open cycle—a thermodynamic power cycle in which the working fluid passes through the system only once and is then exhausted to the atmosphere.

peat—an unconsolidated, hydrophilic, yellowish-brown to brownish-black, carbonaceous sediment, formed by accumulation of partially fragmented and decomposed plant remains in swamps and marshes which retains more than 75 percent inherent moisture and less than 12 percent mineral matter in saturated natural deposits.

petrochemicals—those derived from crude oil or natural gas, or their coal-derived substitutes; they include light hydrocarbons such as butylene, ethylene and propylene, the raw materials for the production of plastics by polymerization.

phenols—a group of aromatic compounds having the hydroxyl (OH) group directly attached to the benzene ring.

pilot plant—a chemical process plant containing all the processes of a commercial unit, but on a smaller scale, for the purpose of studying the technical and economic feasibility of the process.

pipeline gas—a methane-rich gas that conforms to certain standards and has a higher heating value between 950 and 1,050 Btu per standard cubic foot.

plenum chamber—an enclosed space through which air is forced for slow distribution through ducts.

precoat—layer of suitable filtering medium, e.g., diatomaceous earth, laid down on a rotary filter cloth prior to operation.

prilling tower—a tower that produces small solid agglomerates by spraying a liquid solution in the top and blowing air up from the bottom.

process development unit—a system used to study the effects of process variables on performance; sized between a bench-scale unit and a pilot plant.

proximate analysis—analysis of coal based on the percentages of moisture, volatile matter, fixed carbon (by difference), and ash, using prescribed methods. Reported on different bases, such as as-received (or as-fired), dry, mineral-matter-free (mmf), and dry mineral-matter-free (dmmf).

purification—removal of a wide range of impurities present in gases from coal gasification.

pyrolysis—thermal decomposition of organic compounds in the absence of oxygen.

quenching—cooling by immersion in oil, water bath, or water spray.

Raney nickel catalyst—specially prepared nickel catalyst used in the hydrogenation of organic materials and the methanation of synthesis gas to methane.

raw gas—See Crude Gas.

reactivity—susceptibility to chemical change; for example, in coal liquefaction, the reactivity of the coal for conversion to liquid products is a function of the coal rank, among other things.

reactor—vessel in which coal-conversion reactions take place.

Rectisol process—a process for the purification of coal-gasification gas based on the capability of cold methanol to absorb all gas impurities in a single step; gas naphtha, unsaturated hydrocarbons, sulfur componds, hydrogen cyanide, and carbon dioxide are removed from the gas stream by the methanol at temperatures below 0° C.

reducing gas—a gas which, at high temperatures, lowers the state of oxidation of other chemicals.

reforming processes—a group of proprietary processes in which low-grade or low molecular weight hydrocarbons are catalytically converted to higher grade or higher molecular weight materials; also applies to the endothermic reforming of methane, for the production of hydrogen, by the reaction of methane and steam in the presence of nickel catalysts.

refractory—a material capable of withstanding extremely high temperatures and having a relatively low thermal conductivity.

residence time—time spent by a typical particle in a particular zone.

saturated hydrocarbon—a carbon-hydrogen compound with all carbon bonds filled; that is, there are no double or triple bonds as in olefins and acetylenes.

scrubber—apparatus in which a gas stream is freed of tar, ammonia, and hydrogen sulfide.

seam coal—coal which is intermediate in rank between bituminous coal and anthracite; contains 8 to 22 percent volatile matter and from 91 to 93 percent carbon.

semi-water gas—a mixture of carbon monoxide, carbon dioxide, hydrogen, and nitrogen, obtained by passing an air-steam mixture through a hot bed of coke, having a higher heating value of about 120 Btu per standard cubic foot.

sensible heat—that heat which results in only the elevation of the temperature of a substance with no phase changes.

shift conversion—process for the production of gas with a desired carbon monoxide content from crude gases derived from coal gasification; carbon monoxide-rich gas is saturated with steam and passed through a catalytic reactor where the carbon monoxide reacts with steam to produce hydrogen and carbon dioxide, the latter being subsequently removed in a scrubber employing a suitable sorbent.

sintering—the agglomeration of solids at temperatures below their melting point, usually as a consequence of heat and pressure.

slag—molten coal ash composed primarily of silica, alumina, iron oxides, and calcium and magnesium oxides.

slurry—a suspension of pulverized solid in a liquid.

solvation—the association or combination of molecules of solvent with solute ions or molecules.

solvent—that component of a solution which is present in excess; liquid used to dissolve a substance.

solvent extraction—selective solution of coal constituents from finely divided coal particles into a suitable solvent after intimate mixing, usually at high temperatures and pressures in the presence of hydrogen, with or without a catalyst, followed by phase separation.

solvent refined coal (SRC)—a coal extract derived by solvent extraction; a brittle, vitreous solid (m.p. 300° F to 400° F) containing about 0.1 percent ash and about 10 percent of th sulfur in the original coal feedstock; calorific value is about 16,000 Btu per pound; may be used as a clean fuel for power generation by combustion; utilized for the production of high-grade metallurgical coke, anode carbon, and activated carbon by coking, or hydrogenated to produce synthetic crude oil.

space velocity—volume of a gas (measured at standard temperature and pressure) or liquid passing through a given volume of catalyst in a unit time.

specific gravity—ratio of the weight of any volume of a substance to the weight of an equal volume of water at 4° C.

specific heat—heat capacity of a substance as compared with the heat capacity of an equal weight of water.

standard cubic foot (SCF)—the volume of a gas at standard conditions of temperature and pressure. The American Gas Association uses moisture-free gas at 60° F and 30 inches of mercury (1.0037 atm) as its standard conditions. The pressure standard is not universal in the gas industry: 14.7 psia (1.000 atm) and 14.4 psia (0.980 atm) are also used. The scientific community uses 32° F and 1 atm as standard conditions.

stoichiometry—the definite proportions in which molecules react chemically to form new molecules.

stripping—the removal of the more volatile components from a liquid mixture of compounds.

subbituminous coal—the rank of coal between bituminous and lignite, classified by ASTM as having a range of heating values between 8,300 and 11,000 Btu per pound on a moisture and mineral-matter-free basis.

substitute natural gas (SNG)—a gas produced from coal, oil sands, or oil shale conforming to natural gas standards.

superficial velocity—the linear velocity of a fluid flowing through a bed of solid particles calculated as though the particles were not present.

superheater—a heat exchanger which adds heat to the saturated steam leaving a boiler.

syncrude—synthetic crude oil; oil produced by the hydrogenation of coal, coal extracts, oil sands, or oil shale, which is similar to petroleum crude.

synthesis gas—a mixture of hydrogen and carbon monoxide which can be reacted to yield a hydrocarbon.

tail gas—a gas issuing from a gas-treatment unit which may be recycled to the process or exhausted.

tar (coal)—a dark brown or black, viscous, combustible liquid formed by the destructive distillation of coal.

therm—a unit of heat used as a basis for the sale of natural gas; equal to 100,000 Btu.

topping cycle—the higher temperature thermodynamic power cycle of a combined-cycle system.

turndown ratio—the minimum ratio of actual flowrate to design flowrate at which a process unit can be operated.

ultimate analysis—the determination by prescribed method of carbon and hydrogen in the material as found in the gaseous products of its complete combustion, the determination of sulfur, nitrogen, and ash in the material as a whole and the estimation of oxygen by difference; may be reported on different bases, such as as-received (or as-fired), dry, miner matter-free (mmf), and dry mineral-matter-free (dmmf).

Venturi scrubber—a gas cleaning device which involves the injection of water into a stream of dust-laden gas flowing at a high velocity through a contracted portion of a duct, thus transferring the dust particles to the water droplets which are subsequently removed.

volatile matter—those constituents of coal, exclusive of moisture, that are liberated from a sample when heated to 1750° F for seven minutes in the absence of oxygen.

water gas—gas produced by the reaction of carbon (in coal or coke) and steam to yield mixtures of carbon monoxide and hydrogen; similar to synthesis gas.

water gas shift—the reaction between water vapor and carbon monoxide to produce hydrogen and carbon dioxide or the reverse:

$$CO + H_2O \rightleftarrows H_2 + CO_2.$$

working fluid—a gas stream which directly does work, e.g., powering a gas turbine.