H-COAL™ PROCESS DEMONSTRATIONS, DEVELOPMENTS AND RESEARCH ACTIVITIES

A. G. Comolli, J. B. MacArthur, H. H. Stotler

Hydrocarbon Research, Inc., A Subsidiary of Dynalectron Corporation
P. O. Box 6047, Lawrenceville, New Jersey 08648

ABSTRACT

The H-Coal™ Process is a direct catalytic hydroliquefaction process for converting coal into high-quality, clean hydrocarbon liquids. The process has been developed and tested in laboratory equipment converting up to 3.5 tons of coal per day. The process has been scaled to 600 tons of coal per day in the recently-constructed Pilot Plant at Catlettsburg, Kentucky. A continuous coal feed, 45-day run processing Illinois No. 6 coal was successfully concluded in April of 1981, followed by a 131-day test run on the same coal completed in December of 1981.

Development work on the H-Coal Process and variants of the process to meet specific market requirements is underway at the HRI R&D Center. Research activities directed towards better catalysts and catalyst regeneration and a fundamental study of how various coals behave in the H-Coal Process are progressing.

Distillate yields and equipment performance were very encouraging. Further operations with Wyodak and Kentucky coals are planned.

HISTORY

H-Coal™ is a direct catalytic hydroliquefaction process based on the use of an ebullated-bed reactor for conversion of coal to desirable clean liquids. The process can be altered to produce a broad spectrum of hydrocarbon liquids ranging from an all-distillate synfuel to a low-sulfur fuel oil.

The H-Coal Process, a modification of the H-Oil™ Process, was invented by HRI and has been under development since 1963. About twenty coal types have been tested in over 60,000 hours of operation. Development and demonstration have been carried out in bench-scale units processing about 25 pounds of coal per day, in a PDU of 3.5 tons' coal capacity and in a Pilot Plant designed to process up to 600 tons of coal per day. The bench-scale units serve as the primary research and development tool for kinetics and process improvement studies, catalyst and coal evaluations, and staged processing. The PDU has been used to confirm the design bases, operating conditions, and modes of operation for the Pilot Plant, to produce large quantities of products for evaluation, and to develop and test specialized equipment. Large-scale demonstration of the ability to process different coals, the testing of commercial-size equipment, and production of large quantities of product for full-scale evaluation are accomplished at the Pilot Plant in Kentucky.

Illinois No. 6 coal is currently being processed at the Pilot Plant. Following the evaluations of Illinois and Kentucky coals in 1981, an extended operation is planned on a subbituminous Wyodak coal. Data developed on equipment performance and process observations will be used for the design of full-scale commercial H-Coal plants. A commercial H-Coal plant slated for erection in Breckinridge, Kentucky is now being

designed by Ashland Synthetic Fuels, Bechtel, and HRI under the sponsorship of the Department of Energy. Other commercial projects are in the planning stage.

Research and development is continuing at HRI's Research and Development Center in New Jersey and at H-Coal participant laboratories. Current studies are directed towards the development of new catalysts, catalyst regeneration, and multiple-stage processing of high-oxygen-content coals. Modeling of the ebullated bed is directed towards improving its design and performance. Microautoclave studies are underway to improve our understanding of the thermal and catalytic behavior of various coals in the H-Coal Process. Engineering studies, using linear programming to optimize process configuration, operating conditions, and product options for a complete coal liquefaction plant, are being conducted at HRI's Process Engineering Facility in Lawrenceville, New Jersey. Results are being used to guide experimental work and to aid in the planning of commercial plants.

The current sponsors, along with Dynalectron Corporation, HRI's parent company, are the U. S. Department of Energy, Ashland Oil, Inc., The Electric Power Research Institute, Standard Oil Company of Indiana, Conoco Coal Development Company, Mobil Oil Corporation, The Commonwealth of Kentucky, and Ruhrkohle, AG of West Germany.

H-COAL™ PROCESS

The H-Coal Process is based on the ebullated-bed reactor, a liquid-gas fluidized bed of catalyst. The reactor operates at relatively high temperatures and pressures with small amounts of catalyst added and removed daily, or as required, to maintain a desired state of activity. An aluminum-oxide-based catalyst extrudate with cobalt or nickel and molybdenum promoters is currently being used for coal liquefaction -

Support	- Aluminum Oxide
Promotors	- CoMo and NiMo
Diameter	- 1.4 to 1.7 MM
Length	- 1.8 to 10 MM
Density	- 0.55 to 0.75
Pore Distribution	- Bimodal
Pore Size, Average	- 50 and 130 Å

This catalyst, with a high surface area, is considerably more active than any naturally-occurring, disposable catalyst.[1] (Figure 1). The ebullated-bed reactor is completely backmixed and operates at near isothermal conditions.

A schematic of the H-Coal Process is shown in Figure 2. Heavy distillate and high-boiling residual oils are recycled in the syncrude and boiler-fuel modes to optimize conversion and heteroatom removal. Hydroclones partially remove solids from the heavy recycle oils. Further oil recovery and solids concentration are achieved through vacuum distillation in the syncrude mode and by solvent precipitation or Critical Solvent Deashing in the fuel-oil mode. By definition and selection the syncrude mode is a balanced operation and enough bottoms are produced to supply the net hydrogen required for coal liquefaction. The H-Coal Process can also be operated at conditions yielding less bottoms and more distillate when hydrogen is supplied by partial oxidation of coal and steam reforming the gases. The process can also be operated in the COIL™ mode, utilizing heavy and low-cost petroleum fractions in combination with coal to further increase desirable distillate yields.(Figure 3).

RECENT ACCOMPLISHMENTS

Pilot Plant

The Pilot Plant has several major objectives which cannot be met on laboratory and PDU-scale equipment. These are:

- Demonstrate the mechanical operability and reliability of commercial-scale equipment.
- Provide products for commercial testing at rates of 100 to 300 tons per day.
- Verify yields in commercial-size equipment.
- Determine appropriate materials for construction.
- Establish maintenance requirements for key items of equipment.

More detailed process flow diagram of the Pilot Plant are shown in Figure 4 and 5.

The most important recent milestone met by the H-Coal Process was the successful completion, on April 3, 1981, of a 45-day, continuous coal run on Illinois No. 6 Burning Star coal at the Catlettsburg Pilot Plant operated by Ashland Synthetic Fuels.[2] (Table 1). 9,850 tons of coal were liquefied at an average coal feed rate of 184 tons/day, with a maximum rate of 222 tons/day, producing three barrels of distillate per ton dry coal. The run was voluntarily shut down when a key hydroclone feed control valve failed and restrictions formed in the vacuum tower.

Reactor operations were smooth and trouble-free during the entire 45 days. The temperature drop across the reactor averaged 20°F. Catalyst was added at a rate of one lb/ton of dry coal; catalyst losses through attrition were less than 0.09 lb/ton of dry coal fed.

Data collected from the slurry feed system showed a uniform mixing of the coal and recycled oils, and oil/solids ratios from 1.2 to 1.8. The slurry feed pumps and circulating pumps performed reasonably well with feed pump packing and seal lives from 21 to 45 days. Experience with the helical coil preheater was excellent. The pressure drop during the run averaged 50 psig, and the coil was free of coke and erosion.

High pressure letdown and block valve operation, a major problem in early runs, was greatly improved. One valve was on stream for 23 days and was removed only when a line choke failed. Hydroclone efficiencies reached 30-40%. No serious erosion was noted; however, there was some corrosion in the atmospheric fractionator and the cooling water system. (See Tables 2, 3, 5 and Figure 6).

Another major operation on Illinois No. 6 coal was completed December 11, 1981, and the difficulties experienced in a similar Pilot Plant operation (Run 6) with the control of recycle oil inventory have been solved. The test objectives set by the DOE and the H-Coal industrial participants were met in late 1982 upon the completion of a 131-day test run with Illinois No. 6 coal being processed at the design conditions of 220 tons/day, 72% of the time. During this run the plant converted 19,200 tons of dried coal to low sulfur hydrocarbon distillate at three barrels of oil per ton of dry coal. A major achievement during the run was obtaining durability of valves

operating at high temperature and pressure with erosive coal, ash and oil slurries. One valve used for let-down service from high to low pressure with heavy oil and unconverted coal and ash achieved 1,245 hours of continuous service.

Research and Development Center

An important run in support of the Pilot Plant, involving testing a new catalyst with Wyodak coal, has been successfully completed. This run, PDU 10, was conducted in the 3.5 tons/day PDU, and included 46 days of syncrude mode operation. Coal conversion above 90% and C_4-975°F distillate yields of 47-53% on an MAF basis were achieved. (Table 4). Catalyst activity and solvent quality reached equilibrium in twenty days. In comparison to previous operations, the new catalyst, Amocat 1A, gave improved yields and lower hydrogen consumptions with lower oil viscosities. A catalyst tracer study conducted during the run, with assistance from Sandia Labs, indicated complete mixing of the ebullated bed system. (Figure 7). Inspection of the unit following the run showed complete freedom from calcium-carbon type deposits reported by other processors of subbituminous coals.[3]

Another major achievement of the HRI R&D Center has been the complete regeneration and activity recovery of the H-Coal catalyst. Previous regeneration consisted of carbon burnoff to recover 60% of the initial catalyst activity. The new techniques show total recovery of initial activity (Figure 8). This finding will have a significant impact on catalyst costs and distillate yields. The procedure for regeneration involved a simple physical and chemical treatment at atmospheric conditions.

PLANS

R & D Center

HRI plans to continue the development and demonstration of catalyst regeneration and to continue the development of new and more specific H-Coal catalysts.[4,5] Other foreign and domestic coals will be studied and processed.

Further R&D Center activity, involving multi-stage processing and the use of cleaned coals and new catalysts, is included in a contract with DOE extending through 1982.

Pilot Plant

At the Pilot Plant in Catlettsburg, operation on Wyodak and another bituminous coal will continue through 1982, followed by a turnaround and the processing of other domestic and foreign coals.

Commercial Plant

Work has recently been completed on the process design, cost estimation, economic analysis and environmental assessment for a commercial-scale H-Coal liquefaction plant. This plant is to be located in Breckinridge County, Kentucky and is designed to feed approximately 22,500 tons/day of run-of-mine Illinois No. 6 coal to produce a nominal 50,000 barrels per day of hydrocarbon distillate products. (Table 5). The plant would be owned and operated by a partnership of companies headed by Ashland Oil Company. Construction is scheduled to begin in 1983 with initial production of H-Coal synfuels in 1988.

References

(1) "Chemical Studies on the Synthoil Process-Mineral Matter Effects" Final Report: September 1, 1975 to September 30, 1977. B. Granoff, et al.

(2) "H-Coal™ Pilot Plant Operations", James B. MacArthur, Harold H. Stotler, Alfred G. Comolli. 1981 Fuels and Lubricants Meeting, Houston, Texas. November 5-6, 1981.

(3) "Recent Developments In The Processing of Western Coal With the H-Coal™ Process", M. Merdinger and A. G. Comolli, Hydrocarbon Research, Inc. Sixth Annual EPRI Contractors' Conference on Coal Liquefaction, Palo Alto, California. May 13, 1980.

(4) "Recent Advances in the H-Coal™ Process", C. D. Hoertz, Ashland Oil, H. H. Stotler and A. G. Comolli, Hydrocarbon Research, Inc. Presented at the Southeast Regional American Chemical Society Meeting in Lexington, Kentucky, November 11, 1981.

(5) "Catalyst Development for Coal Liquefaction", J. A. Mahoney, Amoco Oil Company. H-Coal™ Research TAC Meeting, Catlettsburg, Kentucky. September 15, 1981.

TABLE 1. ANALYSIS OF ILLINOIS No. 6 COAL

ULTIMATE ANALYSIS	
Carbon	69.66
Hydrogen	5.10
Nitrogen	1.22
Sulfur	3.60
Ash	10.48
Oxygen (difference)	9.88
MINERAL ANALYSIS	
Silica	46.62
Alumina	18.95
Ferric Oxide	19.36
Lime	5.97
Sulfur Trioxide	4.26
Others	7.23

TABLE 2. H-COAL PILOT PLANT - OPERATING CONDITIONS FOR TYPICAL DAY - 3/27/81 - 3/28/81

	Actual	Target
Hydrogen Pressure, psi	3000	3000
Reactor Temperature, °F	845	850
Coal Feed, Tons/Day	222	219
Oil-to-Solid Ratio	1.65	1.75
Slurry Feed Rate, Lb/Hr	55,400	-
Space Velocity, Lbs Dry Coal/Hr/Ft^3	31.6	31.2
Hydroclone recycle flow, % of slurry oil	47	66

TABLE 3. H-COAL PILOT PLANT - PRELIMINARY YIELDS* FOR TYPICAL DAY - 3/27/81 - 3/28/81

	ACTUAL		TARGET	
	Wt. PERCENT	BBL/TON	Wt. PERCENT	BBL/TON
C_1-C_3	10.96	-	10.68	-
Naphtha (C_4-400°F)	22.71	1.66	18.74	1.40
Distillate (400-975°F)	23.89	1.46	28.33	1.63
Residuum Oil (975°F$^+$)	21.62	0.94	19.00	0.86
Unconverted Coal	3.47	-	5.78	-
Ash	11.22	-	11.51	-
TOTAL		4.06		3.89

*Dry Coal Basis

TABLE 4. H-COAL™ PILOT PLANT RUN NO. 8

OBJECTIVES

1. Demonstration of solvent balance during periods of coal feed.
2. Demonstration that satisfactory hydroclone efficiencies (30 percent) can be achieved.
3. Operation of the Lean Oil Absorption System.
4. Confirmation of the yields obtained in the Process Development Unit Run No. 5.
5. Accumulation of engineering data.

OPERATING CONDITIONS

Dry Coal Feed Rate	219 T/D
Space Velocity	31 lb/hr/ft^3
Average Reactor Temperature	850°F
Oil/Solids Feed Ratio	1.75 lb/lb
Gas Flow to Reactor	550,000 SCFH
Reactor Pressure	3,000 psig
Inlet Hydrogen Partial Pressure	2,400 psig
Outlet Hydrogen Partial Pressure	1,900 psig
Catalyst Replacement Rate	1 lb/ton dry coal fed

TABLE 5. H-COAL PILOT PLANT RUN NO. 8

RESULTS

1. Solvent balance was demonstrated during periods of coal feed. During a four-day material balance, 335 barrels of solvent (fractionator bottoms) were accumulated. Approximately 1,000 barrels of heavy distillate and 5,000 barrels of light distillate were accumulated were accumulated during Run No. 8.

2. Satisfactory hydroclone efficiencies were demonstrated. Efficiencies as high as 30 percent were achieved.

3. The Lean Oil Absorption System was operated successfully during Run 8. On-stream time was approximately 90 percent.

4. An excellent material balance test was achieved over a four-day period from october 29 until November 2. Plant conditions were stable, and the yields were in agreement with Process Development Unit Run No. 5.

5. Engineering data was obtained for the following:

   a. Slurry Mix System
   b. Slurry Heater
   c. Vapor Liquid Equilibrium
   d. Lean Oil Absorber
   e. Fractionation
   f. Hydroclone
   g. Partition Coefficients of Phenol in Hydocarbon and Water
   h. Heat of Reaction
   i. Heat Transfer Coefficients for Critical Exchangers
   j. Chloride Distribution
   k. Bowl Mill
   l. Coal Weigh Feeder
   m. Letdown Valve Program
   n. Corrosion and Erosion Monitoring
   o. Volumetric Efficiency of Slurry Charge Pumps
   p. Reduced Exchanger Washwater Rates
   q. Slurry Viscosity
   r. Characterization of Stripped Process Water

TABLE 6. COMPARISON OF YIELDS - ILLINOIS NO. 6 COAL - RUN NO. 8

	Pilot Plant		PDU 5	
	lbs/100 lb Dry Coal	Bbl/ton	lbs/100 lb Dry Coal	Bbl/ton
C_1-C_3	11.77		10.68	
C_4-400°F	22.41	1.69	18.74	1.40
400-650°F	16.46	0.99	20.37	1.21
650-975°F	8.81	0.46	7.96	0.41
Residuum + Unconverted Coal	24.70		24.80	
TOTAL		3.14		3.02

TABLE 7. PDU 6 and 10, WYODAK COAL PRODUCT YIELDS

YIELDS, W % MAF COAL BASIS

	PDU 10	PDU 6
C_1-C_3	11.2	13.7
C_4-400°F	24.6	28.0
400-975°F	27.2	20.2
975°F$^+$ (Residual Oil)	11.9	12.7
Coal Conversion, W %	91.5	93
Hydrogen Consumption, W % Dry Coal	5.75	6.25

FIGURE 1. H-COAL CATALYST

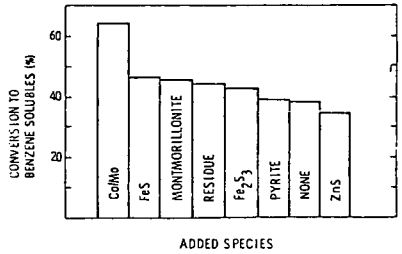

FIGURE 2. H-COAL PROCESS DEVELOPMENT UNIT

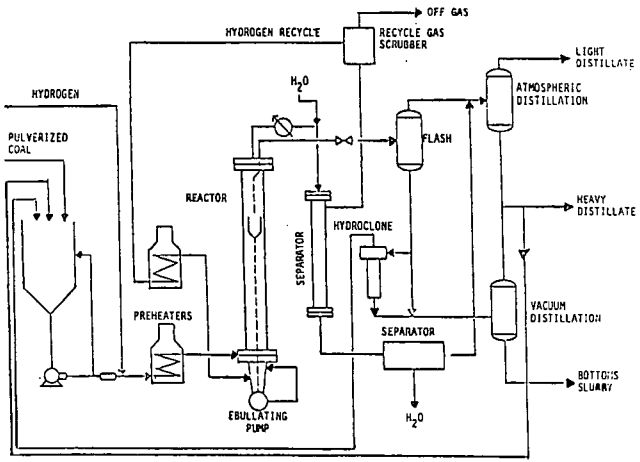

FIGURE 3. COIL™ CONFIGURATION

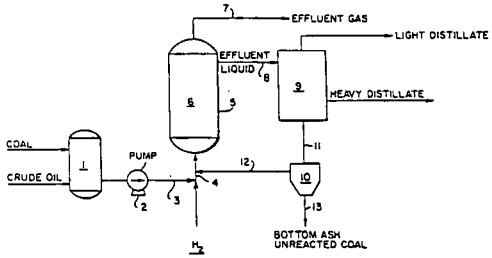

FIGURE 4. COAL LIQUEFACTION

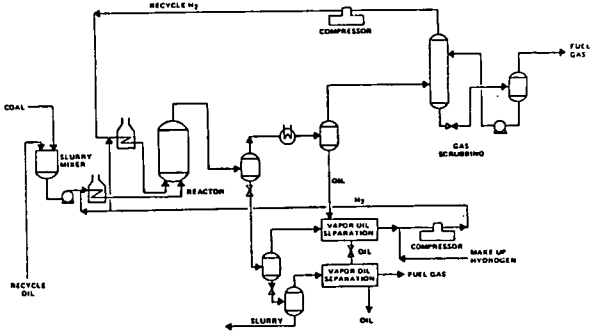

FIGURE 5. OIL RECOVERY

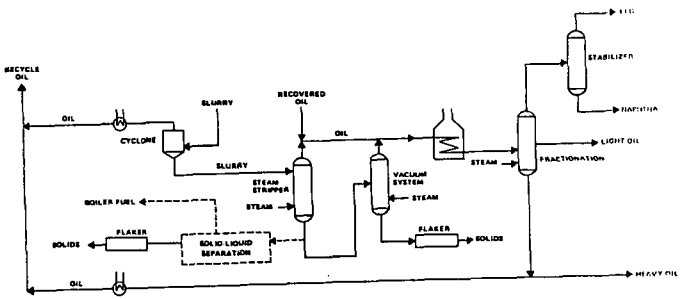

FIGURE 6. H-COAL PILOT PLANT OPERATING DATA - Run No. 6 - ILLINOIS COAL

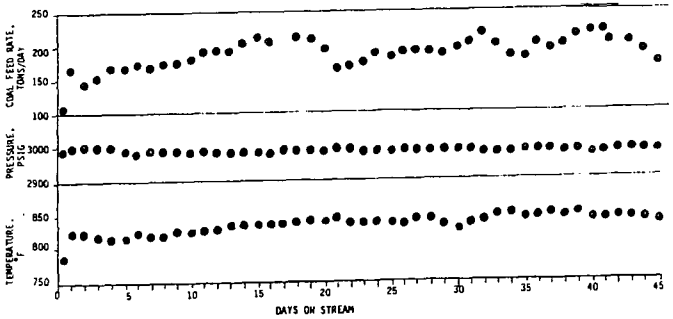

FIGURE 7. PDU 10 PRELIMINARY RADIOACTIVITY IN CATALYST WITHDRAWAL

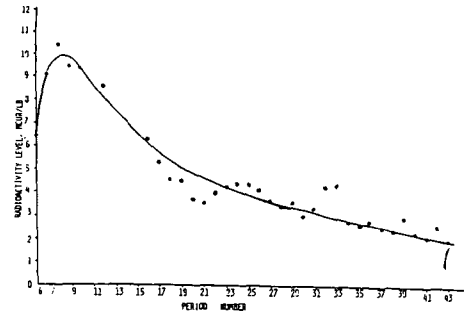
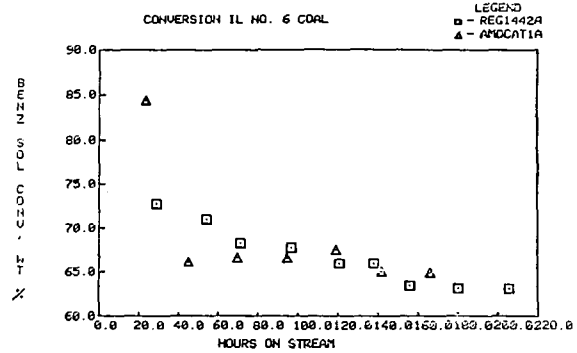
FIGURE 8. CONVERSION ILLINOIS NO. 6 COAL