H-COAL® PROCESS MODELING AND SCALEUP

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ABSTRACT

H-Coal® Process commercial viability has been confirmed by a highly successful, 2-1/2 year demonstration program at the Catlettsburg, Kentucky, Pilot Plant. A major accomplishment was the confirmation of scale-up of the H-Coal® Process chemistry in the well-mixed ebullated-bed reactor system. This paper describes the H-Coal® Process Model developed from bench scale and PDU testing and its relationship to the Pilot Plant operations. Kinetics, hydrodynamics, and overall process simulation are described.

PROCESS BACKGROUND

The H-Coal® Process is a direct catalytic coal liquefaction process developed by Hydrocarbon Research, Inc. for conversion of coal into high-quality, clean liquid fuels[1]. The process can be modified to produce a range of products from a heavy fuel oil to an all-distillate (C_4-650°F boiling range) syncrude. The novelty of the H-Coal Process resides in the use of the commercially proven ebullated-bed reactor in combination with other process steps to achieve C_4-975°F liquid product yields in the range of 40 to 55 weight percent on dry coal. Over twenty coal types have been evaluated in more than 70,000 hours of operation. The history development, and advances of the H-Coal® Process have been extensively documented. Some details of the process and HRI's ebullated bed reactor are appended to this paper.

H-COAL PROCESS SCALE UP

The development path for commercialization of the H-Coal Process is similar to that used by HRI for scale up of the commercial H-Oil™ residuum and heavy crude hydroconversion process. The H-Oil reactor system was successfully scaled up from the bench, through the PDU, followed by a pilot plant demonstration unit and finally to the commercial scale plant. The diameters of experimental reactors employed for collecting process, engineering and scale-up data are listed below. All reactors have operated at commercial design conditions.

	H-Oil Reactor Diameter	H-Coal Reactor Diameter
Bench Unit	3/4 in.	3/4 in.
Development Unit	8-1/2 in.	6 & 8-1/2 in.
Pilot Plant	4 ft. 6 in.	5 ft.
Commercial Plant	13 ft.	10-13 ft.

In addition, HRI has constructed two cold-flow reactor models for fluid dynamics studies and the development of improved reactor internals: a 6 in. x 12 ft glass column and a 5 ft x 24 ft. carbon steel vessel. These cold flow studies, combined with data collected from the various experimental units, have provided the information to scale the effects of equipment size, pressure and temperature on reactor fluid dynamics and vessel internals designs.

The performance of the H-Oil unit at the Kuwait National Petroleum Company's (KNPC) Shuaiba Refinery is evidence of this successful scale up technique. The KNPC H-Oil unit has two reactor trains, each operating on vacuum residuum at about 25,000 B/D. Both reactors have been operating since 1968.

The H-Coal reactor has been thoroughly tested in bench- and PDU-size systems and has recently completed a 2 1/2 year demonstration program at the Catlettsburg, Kentucky H-Coal Pilot Plant. The H-Coal Pilot Plant operation has achieved many significant results including:

- Confirmation of H-Coal process chemistry and equipment scale-up.
- Establishment of process mechanical operability and reliability in commercial-scale equipment
- Collection of engineering design and operating data on critical equipment
  - Slurry Preheater
  - Letdown valves
  - Slurry pumps
  - Reactor and internals
- Accumulation of about 7000 hours of on-coal operations with over 56,000 tons of coal processed.

H-COAL REACTOR IS WELL MIXED AND THERMALLY STABLE

To determine the degree of catalyst backmixing in an ebullated bed, a radioactively-tagged catalyst sample was charged to the PDU reactor in separate operations with Kentucky and Wyoming coals. The amount of tagged catalyst in the daily withdrawals and subsequent reactor catalyst bed dump was determined. The residence time distribution of catalyst in the ebullated-bed was then analyzed. A detailed discussion of the experimental procedures, and some of the test results were reported in the literature. The estimated reactor Peclet number reported by Bickel and Thomas[3] for the Kentucky coal operation was 0.451. Using a modified dispersion model and taking into consideration the fluid dynamics during the catalyst addition and withdrawal operations, the Peclet number for the Wyoming coal operation was estimated to be 0.248 (detailed analysis to be published). These low Peclet numbers indicate that the catalyst mixing characteristics in the PDU ebullated-bed reactor approximated that of the CSTR.

The axial temperature variation in an ebullated-bed reactor is significantly smaller than that in a plug-flow (fixed-bed or moving-bed) reactor at the same conversion level. The axial temperature gradient (ΔT) in an ebullated-bed reactor can be estimated using the following expression:

$$\Delta T = \frac{-\Delta Hr}{\rho C_p} \frac{1}{1+R} (C_{A_f} - C_{A_1}) \quad (1)$$

where,

C_{A_f}, C_{A_1} = concentration of reactant in the feed and in the reactor outlet, respectively.

ΔHr = heat of reaction

R = dimensionless constant related to recycle liquid/feed ratio and internal liquid backmixing. This constant ranges from 5-15 in ebullated-bed operations.

ρ, C_p = average fluid density and heat capacity, respectively.

The calculated temperature gradients from equation (1) are in agreement with PDU and Catlettsburg Pilot Plant data as shown below:

MEASURED VS. PREDICTED H-COAL REACTOR TEMPERATURE GRADIENTS

(Illinois No. 6 Coal, Burning Star Mine)

	PDU-5	Catlettsburg
Reactor Internal Diameter	0.71 ft.	5 ft.
Reactor Temperature	850°F	845-851°F
Reactor Pressure	2,700 psig	3,000 psig
Reactor Temperature Gradient		
Measured	16-20°F	11-18°F
Predicted	16°F	16°F

For a commercial-size H-Coal reactor operating at conditions similar to that of the Catlettsburg Pilot Plant, a temperature gradient of about 7°F was predicted. This lower reactor temperature gradient was due to increased backmixing in large size reactor. The same calculation procedure predicted temperature variations of 2-5°F in commercial H-Oil® reactors, which is consistent with commercial experience. The higher temperature gradients observed in H-Coal® reactors were caused by higher heat of reaction and lower ebullating recycle flow in the H-Coal® Process.

In plug-flow reactors, such as fixed beds and moving beds, the temperature gradient across the reactor for an exothermic hydrocracking reaction can be calculated using the following equation:

$$T_1 - T_f = \left(\frac{-\Delta Hr}{\rho C_p}\right)\left(C_{A_f} - C_{A_1}\right) \quad (2)$$

where subscripts f and 1 denote reactor feed and outlet conditions, respectively. Comparing equations (1) and (2), one can deduce that the axial temperature gradient in a plug-flow reactor is about one order of magnitude higher than that in an ebullated-bed reactor when both are operating at the same conversion level.

The well-mixed ebullating-bed reactor system enables commercial reactor systems to utilize the exothermal heat of reaction to preheat the H-Coal reactor feed. This enables the coal slurry preheater to operate at a low temperature (~700°F at the Pilot Plant). No coke was found in the slurry preheater after 2-1/2 years of Pilot Plant operation because of the mild preheater severities. The H-Coal reactor system is energy-efficient, cost-effective, and has the potential for highest plant on-stream factors of any direct coal liquefaction process.

H-COAL PROCESS MODELLING

The H-Coal® Process has been successfully demonstrated on the bench scale with over 20 coal types including bituminous, sub-bituminous, lignite and brown coal.

The H-Coal® Process offers considerable product slate flexibility, ranging from an all-distillate syncrude to a maximum heavy fuel oil production. The H-Coal® Process meets these requirements through manipulation of key process variables including:

- Coal space velocity
- Catalyst type and replacement rate
- Hydrogen partial pressure
- Reactor temperature
- Recycle slurry composition

HRI has developed a tool to predict the effect of these key process variables on H-Coal yields and product qualities for Illinois No. 6 coal. The H-Coal® Process Simulator is a comprehensive computer model of the H-Coal reactor system at steady-state conditions. The Simulator includes recently updated correlations for H-Coal product qualities, reactor vapor/liquid equilibrium "K" values, and an H-Coal kinetic model. All reactor feed and effluent streams, reactor composition, and product yields and qualities are depicted. Procedures are contained in the Simulator for elementally balancing yields and calculating reactor slurry composition as a function of recycle slurry and recycle gas feed stream rates and compositions. The reactor slurry composition is an important parameter in the H-Coal kinetic model for predicting product yields.

HRI has developed a semi-empirical kinetic model based on a back-mixed reactor system. The conversion reactions are assumed to be irreversible. The reactions are first order except for preasphaltene and asphaltene conversion. The reaction mechanism assumed is shown in Figure 1. Coal is made up of inert components (ash and inert organic material) and reactive components. The primary reactions involve the conversion of reactive coal to preasphaltenes and gases (including hydrocarbon and heteroatoms). Secondary reactions involve the conversion of preasphaltenes to asphaltenes, distillates (C_4-400°F, 400-650°F, and 650-975°F) and gases. Finally, in tertiary reactions, asphaltenes convert to distillates and gases, and distillates convert to lighter boiling components.

The kinetic model is incorporated into the H-Coal® Process Simulator. In the Simulator, empirical correlations are used to separate the distillate components into narrow (50°F) boiling range fractions. These components are utilized to obtain better estimates of reactor composition (through vapor/liquid equilibrium flash calculations), physical/chemical properties of the product streams, and for elemental balancing of the H-Coal® Process yields.

Table 1 summarizes the data base used to develop the H-Coal Kinetic Model for Illinois No. 6 Coal (Burning Star Mine). All process variables cover a wide range with the exception of temperature. The temperature effect has been modeled based on prior data obtained on a different Illinois coal (Monterey Mine).

TABLE 1. H-COAL KINETIC MODEL - RANGE OF DATA USED

COAL TYPE:	Illinois No. 6 (Burning Star Mine)
CATALYST:	HDS-1442A
NUMBER OF YIELD PERIODS:	140
PROCESS UNITS:	BENCH, PDU
PROCESS VARIABLE RANGES	
Coal Space Velocity	25 - 80 lb/hr/ft^3
Pressure	1450-2700 psia
Hydrogen Partial Pressure	1100-1900 psia
Temperature	840 - 852°F
Catalyst Age	0-600 hours
	0-3000 lb coal/lb catalyst
Operating Mode	Bottoms Recycle
Reactor Stages	Single

The H-Coal Kinetic Model components, correlating parameters, range of yield data, and standard deviations of the correlations are summarized in Table 2. Coal conversion is correlated as a function of coal space velocity, reactor temperature, and the concentration of reactive coal in the reactor. The Illinois coal is very reactive and coal conversion data covered a narrow range of 87-96 weight percent of MAF coal. Hydrogen partial pressure effects were not statistically significant. Although catalyst age and activity influence solvent quality, no catalyst impact on coal conversion could be correlated from available data. The standard deviation for the total bench and POU data base was 1.2%.

TABLE 2. REVISED H-COAL KINETIC MODEL

COMPONENT	FORM OF CORRELATION	RANGE OF YIELDS % MAF COAL	STANDARD DEVIATION
Coal Conversion	F_1 (SV,T,C)	87-96	1.2
Preasphaltenes	F_2 (SV,T,AGE,H_2PP,C)	1.3-20	-
Toluene Conversion	F_1-F_2		
Asphaltenes	F_3 (SV,T,AGE,H_2PP,C)	11-30	2.5
975°F$^-$ Conversion	F_1-F_2-F_3	45-80	2.0
650-975°F Liquid	F_4 (F_1,F_2,F_3,SV,C)	6-14	2.3
400-650°F Liquid	F_5 (F_1,F_2,F_3,F_4,SV,C)	5-30	2.1
C_4-400°F Liquid	F_6 (F_1,F_2,F_3,F_4,F_5)	10-40	3.1
C_1-C_3 Gas	F_7 (F_1,F_1,F_2,F_4,F_5)	5-15	1.7

Preasphaltene and asphaltene conversions were correlated as a function of coal space velocity, catalyst age, hydrogen partial pressure, reactor temperature, and reactor concentration. Statistical performance was significantly improved by correlating the toluene conversion (coal conversion minus preasphaltene yield) and 975°F$^+$ conversion (coal conversion minus preasphaltene and asphaltene yield). This eliminated data scatter associated with small differences between large numbers. Standard deviations of 1.8% and 2.0% were achieved for the toluene conversion and 975°F$^+$ conversion, respectively. Yields of distillates (C_4-400°F, 400-650°F, 650-975°F) were similarly correlated. The data, however, were more limited and data scatter more significant. Further data are required to confirm these reactions rate constants.

The H-Coal® Process Simulator includes empirical correlations of hydrogen consumption, and heteroatom (COx, H_2S, NH_3, H_2O) yields. Liquid product qualities are calculated by kinetic (for S,N) and empirical correlations.

The H-Coal® Process Simulator has been used extensively to predict H-Coal® Process performance over a wide range of operating conditions and to evaluate process improvements. Significant improvements have been made in increasing the recycle solvent boiling range (400°F$^+$ to 600°F$^+$) and extincting the heavy vacuum gas oil stream. This improves process economics when a transportation product slate is desirable.[4] Also, recent studies have shown that the hydroclones used for preparation of recycle slurry oil may be eliminated in most cases without sacrificing liquid yields. The Simulator has been used to verify Pilot Plant performance.

H-COAL PILOT PLANT YIELDS PREDICTIONS CONFIRMED

The H-Coal Pilot Plant had a successful 131 day run with Illinois No. 6 and HDS-1442A catalyst (Run 8) during the second half of 1981. The run was conducted with daily catalyst addition and withdrawal to maintain a target catalytic activity. During this run, 19,200 tons of dried coal were processed with the reactor processing coal or oil slurry 100% of the time and coal fed for 72% of the time. The data obtained at the Pilot Plant are felt to be representative of what would be achieved in long term commercial operations.

Yield data were obtained in both September and a four-day material-balance period (October 29-November 2). The yield data obtained during the material-balance period were considered most representative due to lined-out H-Coal operations at steady-state conditions during this period.

Table 3 presents a comparison of yields obtained at the PDU scale with those obtained during the four-day material-balance period at the Pilot Plant for the Illinois No. 6 bituminous coal. Table 4 presents similar data for Wyodak subbituminous coal. These data show equivalent yields at the two scales and confirm the scaleup of H-Coal® Process chemistry. The H-Coal Kinetic Model and Process Simulator closely predicted the demonstrated Pilot Plant performance. These results provide confidence in HRI's ability to project commercial scale plant performance based on results obtained in bench and PDU units.

TABLE 3. H-COAL PROCESS CHEMISTRY SCALE-UP ILLINOIS NO. 6 COAL

UNIT	PDU	PILOT PLANT
Nominal Size, T/D	3.5	200
YIELDS	PERCENT	PERCENT
C_1/C_3	10.68	11.77
C_4/400°F	18.74	22.41
400/975°F	28.33	25.27
975°F$^+$ Residuum	19.00	21.26
Unconverted Coal	5.78	3.46
Ash	11.51	11.31
Total Distillate Oil	47.07	47.68
Residuum Plus Coal	24.78	24.72

TABLE 4. H-COAL PROCESS CHEMISTRY SCALE-UP WYODAK COAL

UNIT	PDU		PILOT PLANT	
Nominal Size, T/D	3.5		200	
YIELDS	PERCENT	B/T	PERCENT	BTU
C_1/C_3	99.8	-	9.29	-
C_4/400°F	22.12	1.62	25.95	1.93
400/975°F	24.06	1.40	23.93	1.41
975°F$^+$ Residuum	11.27	0.51	10.65	0.48
Unconverted Coal	10.72	-	9.12	-
Ash	8.84	-	9.13	-
Total Distillate Oil	46.18		49.88	
Residuum Plus Coal	21.99		19.77	

SUMMARY

The H-Coal® Process follows the same successful development and scale up path used for the H-Oil Process. Results of reactor testing and modeling are in agreement with plant data, confirming that ebullated beds are well-mixed and highly stable. A kinetic model and overall process simulator have been developed and shown to accurately predict results from bench, PDU, and Pilot Plant scales. Pilot Plant results confirm scaleup of the H-Coal® Process chemistry. Data obtained in future bench and PDU units may be used with confidence for design of commercial scale plants.

REFERENCES

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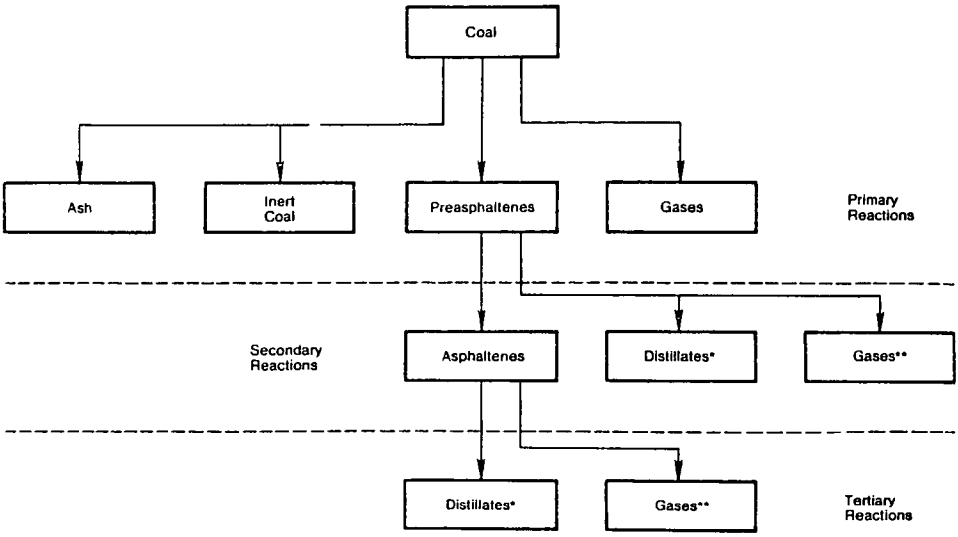
3. Bickel, T. C., and Thomas, M. G., IEC Process Design and Development, Vol. 21, No. 3, July, 1982.

4. MacArthur, J. B., J. E. Duddy, A. S. Ambegaonkar, and A. V. Moomjy, "H-Coal Liquids-Upgrading Upstream or Downstream?" AIChE 1983 Spring National Meeting, Houston, TX, March, 1983.

FIGURE 1

H-Coal® Kinetic Model Reaction Mechanism



*Distillates are Separated into 3 Components Including C_4/400°F Naphtha, 400/650°F Middle Distillate, and 650/975°F Heavy Distillate.

**Gases Include Hydrocarbons and Heteroatom Compounds.

APPENDIX

H-COAL PROCESS DESCRIPTION

Figure 1 presents a simplified flow diagram of the H-Coal Process. Coal is slurried with a process-derived oil, pumped to reactor pressure, mixed with hydrogen, preheated and fed to the reactor. There, coal, recycle oil, and hydrogen react in the presence of a catalyst. Depending on the process severity selected, the net product yield can be all-distillate material or, at low severities, a distillate and a heavy fuel oil. The reactor typically operates at temperatures of about 800-850°F and pressures of 2500-3000 psig. A portion of the reactor effluent is treated to reduce its solids content. Low-solid-content oil is recycled as a slurry oil for the coal. The balance of the liquid is fractionated to produce an all-distillate product. The vacuum residuum, containing nondistillate oils, unconverted coal, and ash, can be fed to a partial oxidation unit to produce the hydrogen for the process.

EBULLATED-BED REACTOR DESCRIPTION

Figure 2 is a sketch of the ebullated bed reactor which is the heart of the H-Coal reactor design. The reactor feed and recycle streams from the ebullating pump enter the bottom of the reactor. The liquid flow causes the catalyst bed to expand and fluidize. The catalyst remains in the bed. The reactor products, including the unconverted coal and ash solids, leave the bed and are separated for further processing. Because the catalyst is constantly in motion, a portion can be withdrawn and replaced to maintain high catalyst activity. On a daily basis, about one or two percent of the catalyst inventory is removed for this purpose. The ebullated-bed reactor system has over 30 unit-years of commercial operations in HRI's H-Oil® petroleum residuum hydroconversion process. The H-Coal® catalyst now being used has also been demonstrated commercially in H-Oil operations.

The ebullated-bed reactor allows intimate contact between catalyst particles, hydrogen, and the coal-oil slurry to achieve essentially isothermal reaction conditions and provide low constant reactor differential pressure. Other major advantages of the H-Coal reactor system are that:

- High liquid yields and qualities are achieved in the presence of a synthetic catalyst and are not dependent on the catalytic effect of coal ash.
- Continuous catalyst replacement controls deactivation, provides constant product quality, will permit continuous catalyst regeneration, and provides for high on-stream factors.
- Operating conditions can be varied to meet flexible product slate requirements.

FIGURE A-1

Simplified H-Coal® Process Flow Diagram

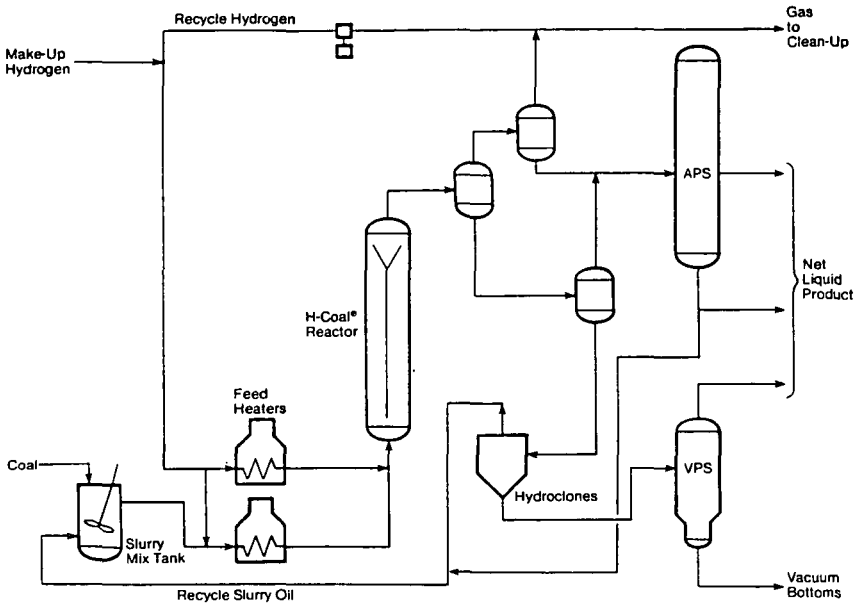
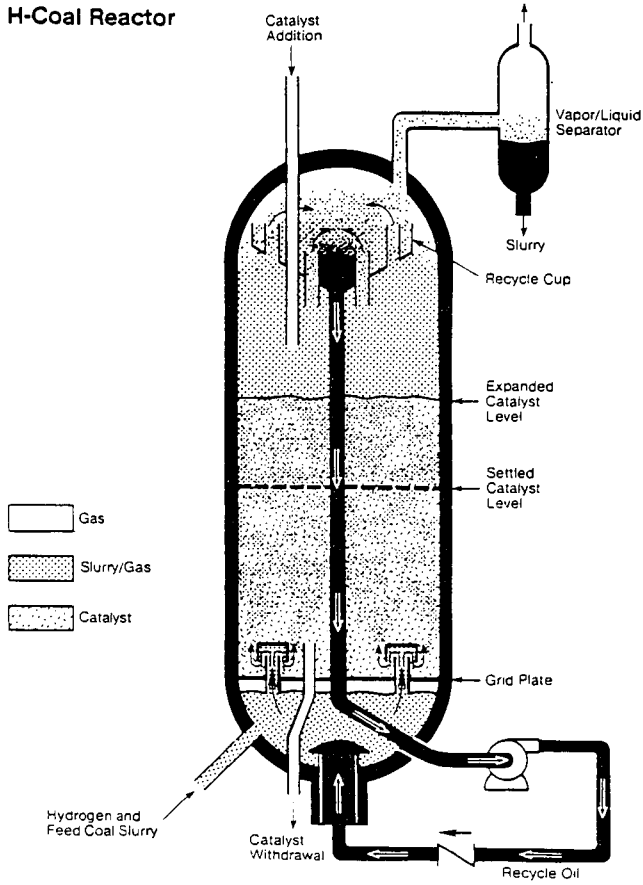


FIGURE A-2


H-Coal Reactor
Catalyst Addition
Vapor/Liquid Separator
Slurry
Recycle Cup
Expanded Catalyst Level
Settled Catalyst Level
Gas
Slurry/Gas
Catalyst
Grid Plate
Hydrogen and Feed Coal Slurry
Catalyst Withdrawal
Recycle Oil