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EDS COAL LIQUEFACTION PROCESS DEVELOPMENT: PHASE IV

Summary of EDS Predevelopment (1966-1975)

Interim Report

July 1978
Date Submitted

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Exxon Research and Engineering Company
Florham Park, New Jersey

U. S. DEPARTMENT OF ENERGY

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Interim Report

W. R. Epperly - Project Director

Exxon Research and Engineering Company
P. O. Box 101
Florham Park, New Jersey 07932

Submitted - July 1978

PREPARED FOR
UNITED STATES DEPARTMENT OF ENERGY
UNDER COOPERATIVE AGREEMENT NO. EF-77-A-01-2893

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United States Department of Energy,
The Carter Oil Company,
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Atlantic Richfield Company

EXXON DONOR SOLVENT PROCESS DEVELOPMENT
PHASES I AND II (1966-1975)
SUMMARY REPORT

Abstract

The Exxon Donor Solvent (EDS) coal liquefaction process is currently being developed to a state of commercial readiness by Exxon Research & Engineering Company under a jointly funded research program. Prior to this jointly funded program, Exxon carried out privately funded research over the 10 year period 1966 through 1975 which led to the selection of the EDS Process as the preferred route to produce liquid fuels from coal. This report summarizes the research program which led to the development of the EDS Process. The significant research findings and alternative processing options which were evaluated are discussed.

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1. INTRODUCTION

Exxon Research and Engineering Company (ER&E) is developing the Exxon Donor Solvent (EDS) Coal Liquefaction Process to a state of commercial readiness. This work is currently being carried out in a 240 million dollar research program sponsored by the U.S. Department of Energy, The Carter Oil Company (an Exxon affiliate), the Electric Power Research Institute, the Japan Coal Liquefaction Development Company, Phillips Petroleum Company and Atlantic Richfield Company. The jointly funded program which is scheduled to extend through mid-1982, features an integrated laboratory and engineering R&D program and construction and operation of a 250 T/D coal liquefaction pilot plant.

Prior to the jointly funded program, ER&E carried out studies which led to the selection of the EDS Process over several other options as the preferred route to produce coal liquids. Over 30 million dollars were expended by Exxon on coal liquefaction research over the ten-year period, 1966 through 1975. The technical feasibility of the EDS Process was established in small continuous pilot plants. A 0.5 T/D integrated liquefaction pilot plant was operated prior to 1970 and a 1 T/D pilot plant was started up in 1975 and is currently operating as part of the continuing R&D program. Throughout the period engineering economic studies were prepared and in 1975 the design of a 250 T/D coal liquefaction pilot plant was completed. The purpose of this report is to summarize the research during the Exxon funded period and to show how the EDS process evolved as the best alternative among the options studied.

Phase I - Predevelopment

Phase I, the Predevelopment Phase, spanned the period 1966 through 1973 and concentrated on identifying the preferred process flow scheme for coal liquefaction. This research was very broad in scope and was conducted in experimental equipment ranging in complexity from 30 cc batch units to the continuous 0.5 T/D coal liquefaction pilot plant (CLPP-1).

Studies were conducted to understand more fully the liquefaction process, seek major process improvements and assess the quality of coal liquefaction products. Examples of the laboratory studies carried out include:

- Liquefaction - bituminous and subbituminous coals and lignites, different reactor types (stirred tank, ebullating bed, tubular plug flow), broad range of process conditions, etc.
- Solvent Hydrogenation - catalyst screening, process variable studies, catalyst deactivation studies.
- Solids Separation - filtration, centrifugation, hydroclones, distillation
- Coking - yields, operability and product qualities versus process conditions, both delayed coking and FLUID COKING.*

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*"Service Mark"

- Hydrotreating - product quality and catalyst screening studies.
- Upgrading - hydroconversion, catalytic cracking and catalytic reforming studies
- Exploratory Studies - mechanism of coal liquefaction, major process improvements.
- Hydrogen manufacture - gasification of coal and liquefaction residues with steam

A wide range of temperatures, pressures and residence times was evaluated in both the liquefaction and solvent hydrotreating reactors. Liquefaction conditions generally ranged from 750-800°F at 350 psia pressure to 800-900°F at 2800 psia. Figure 1 summarizes the range of operating pressures and temperatures which was investigated in CLPP-1 and the bench scale liquefaction pilot units, and compares this range with the earlier German liquefaction work. The figure also shows the preferred range of operating conditions which has been selected for more detailed study in the current EDS Process Development Program.

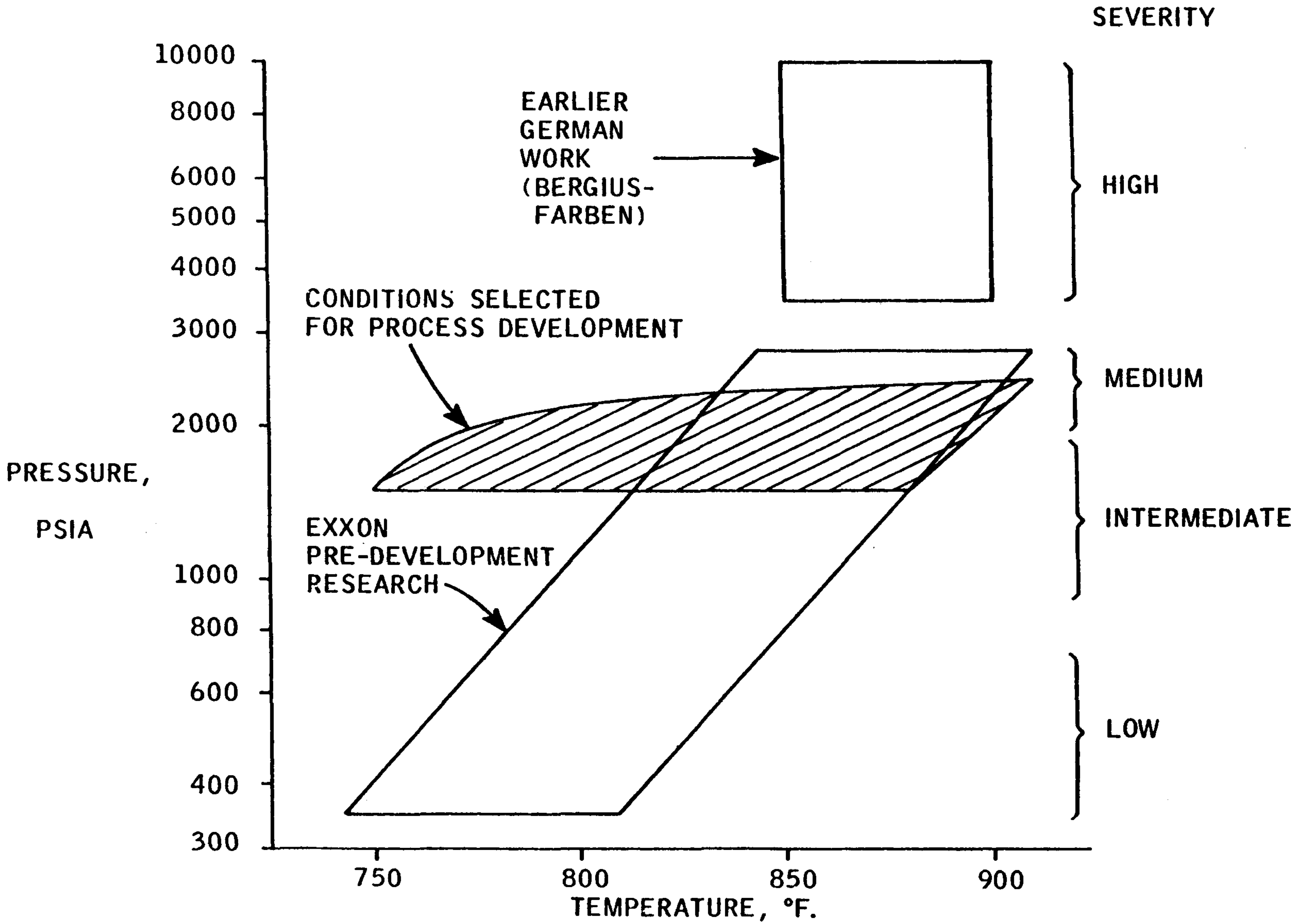
In support of the laboratory program a significant engineering effort was carried out in two major areas: engineering design and cost evaluations, and development of basic engineering technology. The engineering design studies consisted of numerous screening cases comparing various process alternatives, such as different liquefaction reactor pressure levels, alternative solids/liquid separation methods, etc., together with a major study design of a conceptual commercial plant which was completed in 1969. Alternative processes and feeds for producing hydrogen required for the process were also evaluated. The purpose of these studies was to identify the most attractive process arrangement and to define the data needed for further evaluations and eventually for the design of commercial plants. The need was established for a large pilot plant to obtain the required data for scaleup to a commercial size EDS plant at reasonable risk. Engineering and cost evaluations on competitive processes to produce liquids from coal were also carried out during this period to determine if development of the EDS process should proceed or be abandoned in favor of a process being developed by others.

The engineering technology program covered the development of basic engineering data outside the area of process operating conditions, yields, and qualities. During Phase I, studies were carried out to identify the areas requiring technology development. Extensive studies were carried out in the areas of solids separation, slurry heating and slurry rheology and support was provided for the ongoing laboratory and engineering R&D.

Based on these initial coal liquefaction R&D efforts, the basic features of the EDS process were established. The hydrogen donor process was shown to be preferred over non-donor and catalytic liquefaction. The intermediate severity operating conditions selected for the EDS process as shown in Figure 1 were found to be more economic as well as more operable.

FIGURE 1

PREFERRED LIQUEFACTION CONDITIONS SELECTED FROM BROAD DATA BASE



Solids separation devices such as filters and centrifuges were found to be costly and unreliable in coal liquefaction processing, leading to selection of distillation to recover recycle solvent and distillate liquid products. A separate catalytic solvent hydrogenation reactor was selected. This allows maintenance of high catalyst activity (due to removal of ash and heavy 1000°F+ coal liquids prior to hydrogenation) as well as more selective hydrogen addition to the coal products in liquefaction to minimize hydrogen consumption. The EDS process sequence offered the most promise for successful development and commercialization when compared to the other alternatives evaluated.

Phase II - Large Pilot Plant Planning and Design

Phase II, the Large Pilot Plant Planning and Design Phase, spanned the period 1974-75. A major effort during this phase was directed toward the basic design of the 250 T/D Exxon Coal Liquefaction Pilot Plant (ECLP). The large pilot plant efforts included:

- planning studies - evaluated alternative plant sizes, locations, flow plans, yields, operating conditions, required offsite facilities and developed a design basis.
- ECLP design - prepared complete Design Specifications with sufficient process and mechanical details so that a major engineering contractor could prepare a detailed design and construct the large pilot plant.
- ECLP investment - prepared detailed cost estimate based on Design Specifications to establish investment and enable cost control monitoring during detailed engineering and construction.

Concurrent with the ECLP planning studies, laboratory and bench scale work was underway to better define the preferred operating conditions within the narrower intermediate severity conditions of the EDS Process. The 50 and 100 lb/day Recycle Coal Liquefaction Units (RCLU) were used extensively during this period. Studies were carried out to obtain drying rate data for the slurry drying step. Bench scale studies were carried out on coking liquefaction bottoms as a way to increase overall liquid yields. A longer range research program was conducted to define liquefaction reaction mechanism and develop potential process improvements.
A major effort was construction and initial operation of the larger 1 T/D Coal Liquefaction Pilot Plant (CLPP-2). Product yields and qualities from this unit showed excellent agreement with the 50 lb/day RCLU unit demonstrating successful scaleup of the process by a factor of 40.

Engineering and cost studies were conducted to evaluate process improvements and provide process optimization. An in-depth study design of a commercial plant was started during this period and completed in 1976 during the jointly funded Phase III A Program. Alternative bottoms processes were evaluated and the FLEXICOKING* process was selected as the preferred method. FLEXICOKING provided additional liquid yields while providing a low BTU gas

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for plant fuel. Competitive technology was also reviewed and the EDS Process was shown to be competitive with the alternatives evaluated and showed the most promise for successful development. Various engineering technology programs were begun in conjunction with CLPP-2 and ECLP. These included slurry preheater and vacuum tower operating studies on CLPP-2, a cold reactor model simulation of the ECLP liquefaction reactors, and a program to obtain basic thermodynamic data (such as vapor/liquid equilibrium, viscosity, etc.) on EDS coal liquids.

Description of the EDS Process

The main features of the EDS Process as conceived at the end of the Phase I/II research program are described below and in Figure 2.

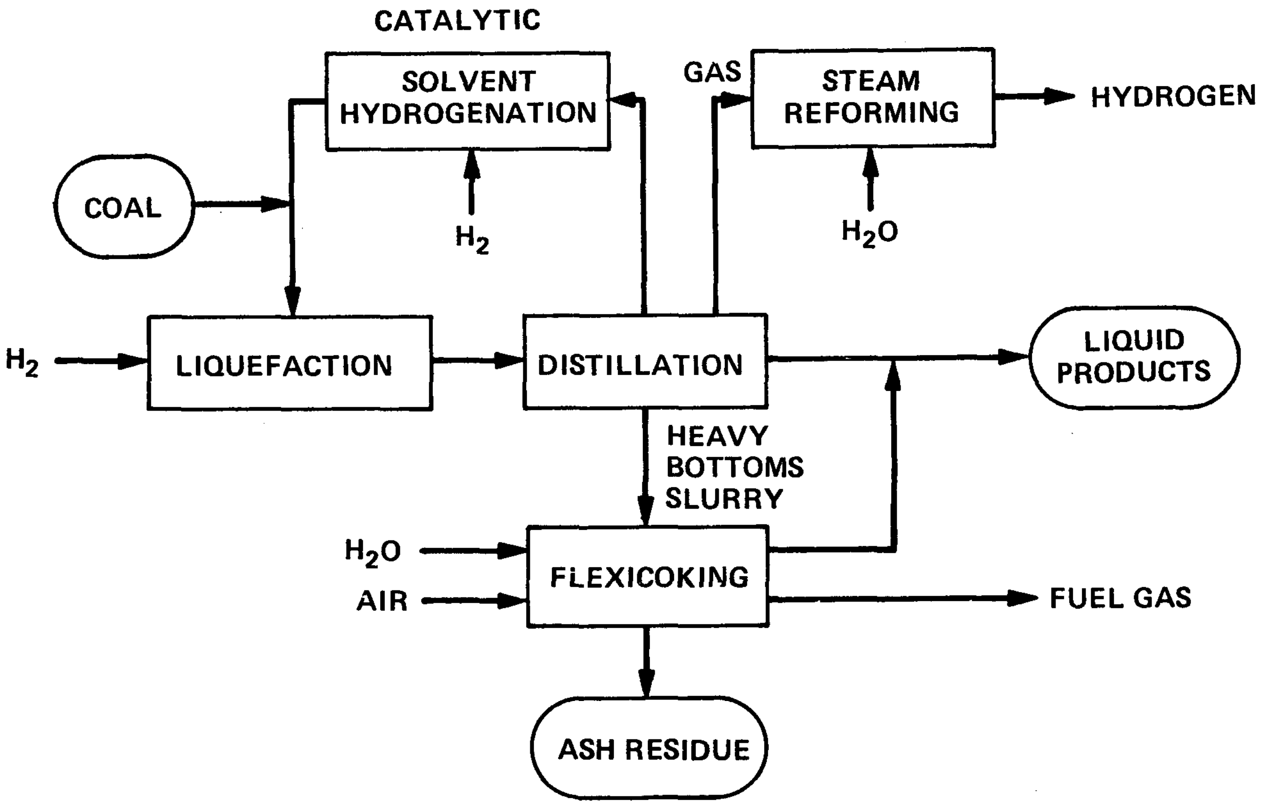
- Crushed coal is liquefied in a non-catalytic tubular reactor in the presence of molecular hydrogen and a hydrogen-donor solvent. The liquefaction reactor operates at 800-880°F and 1500-2000 psi.
- The hydrogen-donor solvent is a 400/850°F boiling range material. The solvent is a catalytically hydrogenated recycle stream fractionated from the middle boiling range of the liquid product. After hydrogenation, the solvent is mixed with fresh coal and fed to the liquefaction reactor.
- Slurry leaving the liquefaction reactor is separated by distillation into gas, naphtha, distillates and a vacuum bottoms slurry.
- The vacuum bottoms slurry is fed to a FLEXICOKING unit to produce additional liquids and provide low BTU gas for process fuel.
- Process hydrogen is provided by steam reforming of the light gases (C_1-C_2) produced in the process.

The EDS process is simple and the critical processing steps are adaptations of petroleum refining technology. The plant is "balanced" in that it is self-sufficient in both process fuel and hydrogen requirements. The process produces high yields of low sulfur liquids from bituminous or subbituminous coals or lignites. On Illinois bituminous coal, the liquid yield is about 2.6 barrels of C_4^+ distillate liquid per ton of dry coal. The liquid products include a coal naphtha which can be upgraded for use as a gasoline blendstock and a low sulfur fuel oil product. A distillate cut could also be recovered from the fuel oil product for upgrading to turbine fuel. Ammonia and elemental sulfur are the only byproducts of significance. By varying liquefaction conditions or adjusting solvent properties, product distribution may be varied over a wide range.

The following sections of this report describe the aforementioned studies carried out during the Exxon funded research program.

FIGURE 2

EXXON DONOR SOLVENT PROCESS



2. LIQUEFACTION

In 1966, ER&E initiated a program to find the preferred route to process liquid fuels from coal. The processes commercialized in Germany in the 1930's were reviewed, as were alternatives being proposed at that time by others. A summary of the competitive coal liquefaction technology evaluated during the Phase I and II period is presented in Section 9. ER&E's laboratory efforts explored several alternative methods of liquefying coal. By far the major effort was on a donor solvent liquefaction process having some similarity to the Pott-Broche process developed in Germany. In the initial process configuration, coal was liquefied at low pressure (~350 psig) with a donor solvent with no molecular hydrogen present. Other approaches were evaluated in the laboratory program, including catalytic, non-donor, other donor configurations, and the use of synthesis gas and water replacing hydrogen. From these efforts, the present EDS process configuration evolved as the best among the alternatives. The EDS process liquefies the coal at intermediate severity conditions (1500 psig, 840°F) with a hydrogen donor solvent as well as molecular hydrogen. The solvent is derived from the coal liquids product and separately hydrotreated and recycled to liquefaction. The laboratory studies carried out during the Phase I and II period to evaluate various coal liquefaction processing alternatives, operating conditions, and coal feedstocks are described below.

The liquefaction step of the EDS process was developed during Phase I of the EDS development program in a combined bench scale-process development unit effort. The approach employed deviated from the traditional research and development procedures which sequentially develop concepts originating from small bench experiments in progressively larger process development steps. With the exception of the initial batch screening studies, the majority of the concepts and innovations in the liquefaction development program originated in the 0.5 ton-per-day Coal Liquefaction Pilot Plant (CLPP-1). The bench scale efforts then followed to either verify or expand the results obtained from the larger process development unit efforts. This reversed sequence of development was due in part to problems associated with handling slurries. These problems were more easily solved in the larger scale CLPP-1 facility which employed larger tubing, control valves and pumps. During the latter parts of Phase I and Phase II, the slurry handling procedures were successfully applied to an intermediate sized processing facility capable of processing 50 pounds-per-day of coal feed. This liquefaction unit was later integrated with solids separation and solvent hydrotreating/fractionation steps to form the first of two Recycle Coal Liquefaction Units (RCLU). These highly integrated units were the major source of process variable data used for the design of the 250 ton-per-day Exxon Coal Liqufaction Plant (ECLP).

Bench Scale Studies

Bench scale studies were carried out on equipment ranging in size from the 30 cc batch tubing bombs to continuous once-through units feeding up to 50 lbs/day of coal feed. In the batch tubing bomb tests, coal feed and solvent were added, the bomb was pressurized with hydrogen and heated to the desired temperature in a sandbath. Once the desired residence time is achieved the bombs were quenched in water. Gas yield and composition were measured along with cyclohexane soluble and insoluble yields and water make. Multiple tests were conducted to improve test accuracy. The tubing bomb tests were supplemented by the larger (4.5 liter) batch autoclave apparatus and once-through continuous units to increase the quantity of coal liquefaction products for more extensive product quality tests. These bench scale studies were used extensively throughout the Phase I and II period and continue to provide useful data to the ongoing EDS R&D program. The more significant study results are summarized below:

- Early in Phase I, six different coal feeds were tested ranging from Western lignites to Eastern bituminous coal. Illinois No. 6 bituminous coal gave favorable results and was selected as the base feedstock for future studies.

- Increasing liquefaction pressure above 350 psig increased liquid yields.

- Increasing liquefaction temperature increased total conversion and conversion to benzene and cyclohexane soluble liquids up to a point. At high temperature (900°F) the liquid conversions decreased.

- The addition of hydrogen gas increased total conversion and conversion to liquids and favored the yield of C_4/700°F liquids.

- As residence time was increased at constant temperature, total conversion, gas yields, and light liquid (C_4/400°F) yields increased. Total liquid (C_4/1000°F) yield reached a maximum and then decreased at longer residence times.

- Total conversion and conversion to liquids improved with increased donor hydrogen level of the solvent.

- Synthesis gas (H_2/CO) and water were as effective as pure hydrogen as a liquefaction treat gas when a donor solvent was used. The percentage of hydrogen donated by the solvent was higher in synthesis gas liquefaction.

Early bench scale studies indicated significantly higher liquid yields for catalytic liquefaction over hydrogen donor liquefaction. Based on these results, catalytic liquefaction was tried out in CLPP-1 but was discontinued due to operability limitations. Subsequently, a 50 lb/day Liquid Fluidized Catalytic Unit (LFCU) was operated to evaluate catalytic liquefaction. After solving some initial operating problems, a 350 hour run was successfully completed. Although liquid yields were initially higher with catalytic liquefaction, the yields decreased rapidly due to catalyst deactivation. After processing about 800 lb coal/lb catalyst, C_4/1000°F liquid yield was equivalent to the donor solvent liquid yields. Based on these results and published data, further engineering evaluations and economics were developed. It was concluded that coal liquids from catalytic liquefaction would cost about the same as those from donor liquefaction while the catalytic system was more complex and required greater development.

Liquefaction reaction mechanism studies were also carried out with donor and non-donor solvents and with model coal liquid compounds. These studies gave considerable insight into the relative roles of the solvent donor hydrogen and of molecular hydrogen and of the importance of solvent quality (see Section 3). Several process improvements were found as part of these studies.

Continuous Integrated Pilot Unit Studies

Larger scale process development studies were carried out during Phases I and II in continuous integrated pilot plants ranging in size from 50 pounds per day to one ton per day coal feed. "Continuous" describes the mode of coal addition and "integrated" refers to the inclusion of several unit operations including coal slurrying, liquefaction, solvent hydrogenation, fractionation, solids separation, and gas cleaning. Typically both solvent and treat gas were recovered and recycled to the process. The initial development work was begun in 1967 in the 0.5 T/D Coal Liquefaction Pilot Plant (CLPP-1). This unit was large enough to produce sufficient quantities of coal-derived products (~1 B/D liquid product) for detailed bench-scale studies on the intermediate processing steps and for downstream product quality studies. On the other hand, it was small enough to enable rapid changes in design and flow sequence.

Coal Liquefaction Pilot Plant (CLPP-1)

During the two and one half year period in which CLPP-1 operated, 125 yield periods were conducted over a wide range of hydrogen donor operating conditions (and some nondonor operations) and processing configurations. The pressure of the liquefaction step was arbitrarily used to divide the CLPP-1 operations into low (below 700 psi), intermediate (700-2000 psi), and medium (2000-3000 psi) severity groups. More than ten donor process configurations were tested in sufficient depth to evaluate its advantages and limitations. Subsequent configurations were designed to improve prior limitations and to retain, where possible, the advantages. This evolutionary process eventually produced the Exxon Donor Solvent processing scheme. Some of the more significant results of the CLPP-1 operations are summarized below:

- Solids separation evolved from filters to centrifuges, to coking, to vacuum fractionation as described in Section 3. Vacuum fractionation was clearly the preferred method for the integrated operation.

- Extinction recycle of heavy coal liquids (700°F+) to Solvent Hydrogenation was feasible and did not hurt solvent quality or liquefaction yield. It did, however, reduce Solvent Hydrogenation flexibility to control solvent quality (and would probably severely limit catalyst life).

- Use of a heavy recycle solvent (400°F+ instead of the base 400/850°F solvent) at low severity donor liquefaction conditions with centrifugation for solids separation simplified the solvent recovery scheme and improved solvent balance. There was, however, a continuous buildup of heavy, refractory material in the system which led to excessive catalyst deactivation and lower liquefaction conversions.

- At low severity (350 psig) donor liquefaction conditions, increasing treat gas rate, solvent to coal ratio, and a 50% change in slurry velocity had no significant effect on conversion and yields. Increasing the solvent hydrogen level above the base value did not increase C_4/1000°F liquid yield but shifted the product slate to lighter C_4/700°F liquids. Decreasing the hydrogen level substantially lowered total conversion and C_4/1000°F liquid yield. Typical overall yields including liquefaction, solvent hydrogenation and coking are given in Table 1.

- At medium severity (2400 psig) <u>non</u>-donor solvent liquefaction conditions, liquid yields were lower compared to the low severity donor operation. Hydrogen consumption was lower and liquid product was less stable and of poorer quality. Increasing temperature above 875°F increased total conversion but selectivity to liquids decreased. Increasing hydrogen treat rate or solvent to coal ratio did not improve yields or operability. Typical yields are given in Table 1.

- At intermediate severity (1500 psig) donor solvent liquefaction conditions, it was found that high solvent tetralin levels provided higher conversion and liquid yield. Addition of molecular hydrogen increased liquid yields. Typical yields for intermediate severity donor liquefaction are given in Table 1. Unit operability was more trouble free than with the previous low severity donor and medium severity non-donor operations.

TABLE 1

TYPICAL COAL LIQUEFACTION YIELDS - ILLINOIS COAL

	Low Severity Donor(1)	Medium Severity Non-Donor(2)	Intermediate Severity Donor(2)
Temperature, °F	775	860	840
Pressure, psig	350	2400	1500
H_2	-4.1	-2.8	-3.1
Chemical Gases	15.3	13.8	13.5
C_1-C_3	11.3	12.8	6.5
C_4/1000°F	26.6	20.4	34.0
1000°F+	50.9	55.8	49.1
	100.0	100.0	100.0

(1) Includes delayed coking for solids separation.
(2) Includes vacuum fractionation for solids separation.

Based on these results and those from the bench scale studies, as well as the engineering evaluations carried out on the various flow schemes and processing economics, the intermediate donor liquefaction conditions using vacuum flash for solids separation were selected as the preferred process for producing liquid fuels from coal. With the basic flow scheme for the Exxon Donor Solvent process selected, further studies were carried out to define process variables and look for process improvements.

Recycle Coal Liquefaction Units (RCLU)

The CLPP-1 facilities were operated prior to 1970. In 1972, a smaller 50 lb/day Recycle Coal Liquefaction Unit (RCLU-1) was started up and in 1974 a 100 lb/day RCLU-2 began operations. These integrated pilot units provided reproducible yields consistent with the CLPP facilities yet on a smaller scale. Numerous studies were carried out at the selected intermediate severity donor liquefaction conditions. There were 291 yield periods on the RCLU units during Phase I and II. Some of the more significant results are described below:

- Investigation of various reactor types including stirred tank, external pumparound, and plug flow reactors led to the selection of the plug flow configuration. The plug flow reactor provided the highest liquid yields while being the simplest and least costly to construct and operate for a commercial plant.

- Over 68 yield periods were conducted to study process variables with Illinois coal. In Figure 3, typical RCLU yields of C_1-C_3 gas, C_4/1000°F liquid, and 1000°F+ bottoms, as well as hydrogen consumption are presented as a function of liquefaction residence time at 840°F (expressed in terms of reference values).

  Note that in the overall EDS processing scheme liquefaction liquid yield is supplemented by liquids obtained by coking of the 1000°F+ bottoms.

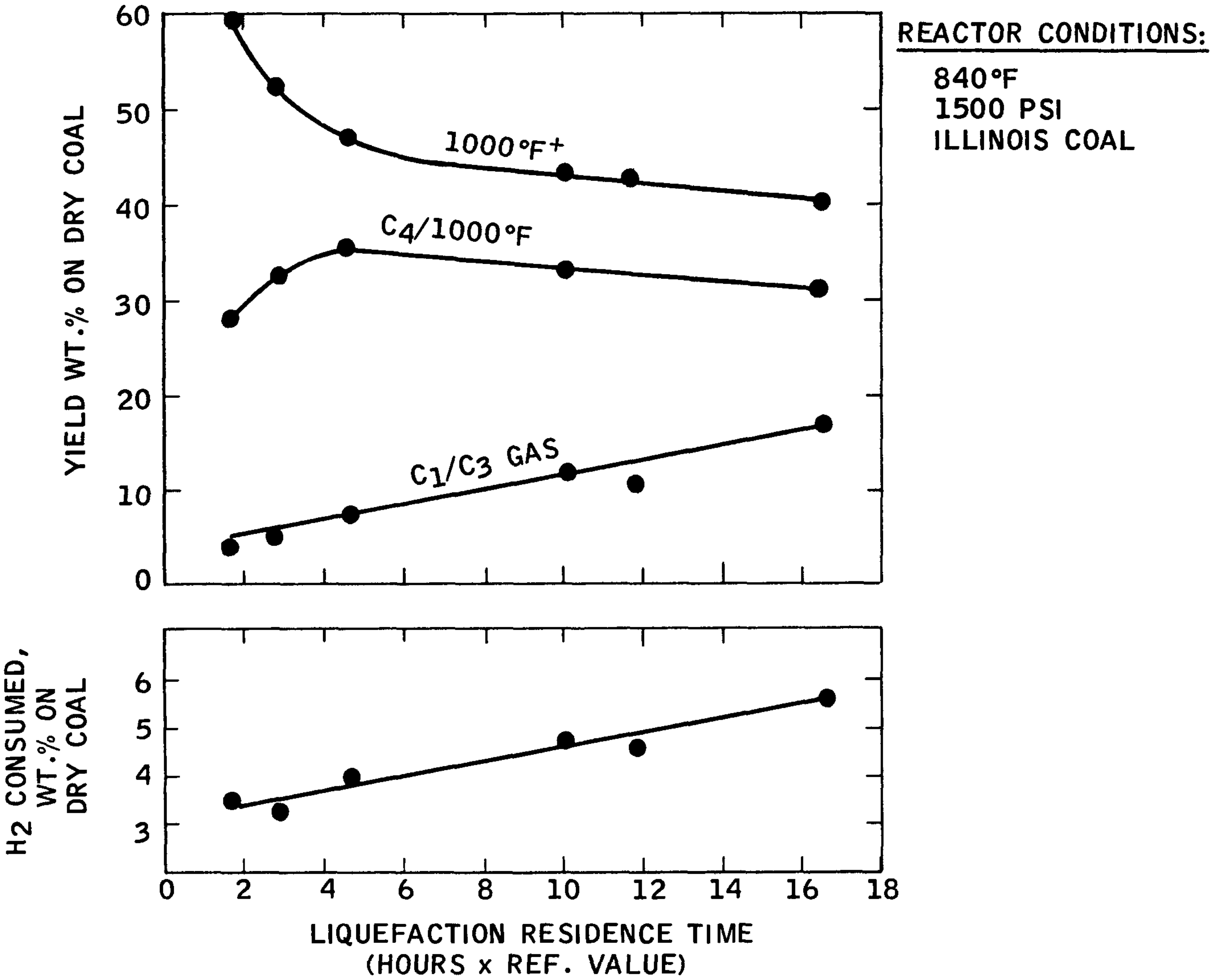
- Operations with a Wyoming subbituminous coal indicated lower liquid yields with the Wyoming coal as well as some operating difficulties. These difficulties were due to the high viscosity of the liquefaction bottoms and to formation of calcium carbonate deposits in the liquefaction reactor which led to reactor plugging. Favorable results were obtained in solving the operating problems and improving yields during the jointly funded Phase IIIA research program.

Coal Liquefaction Pilot Plant (CLPP-2)

The design and construction of the 1.0 T/D Coal Liquefaction Pilot Plant (CLPP-2) was initiated in 1974 with startup of the plant in 1975. This unit was similar to the final CLPP-1 processing configuration incorporating plug flow liquefaction reactors, vacuum flash solids separation, and a separate solvent hydrogenation reactor. Many improvements over the

FIGURE 3

EFFECT OF RESIDENCE TIME ON LIQUEFACTION YIELDS



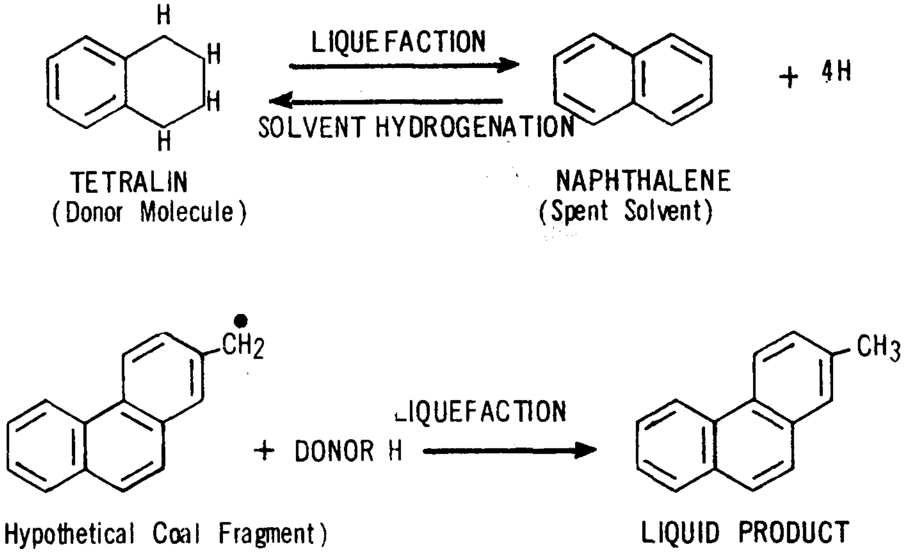
earlier CLPP-1 unit were incorporated. On-line process analyzers and sophisticated computer monitoring facilities were provided. Operations during Phase II were limited to startup, and solving initial operating and material balance closure problems. After these initial difficulties, the data obtained showed excellent agreement with that obtained on the RCLU units indicating successful scaleup of the EDS process by a factor of 40. Studies of process variables and equipment design, as well as production of large amounts of product for quality/upgrading tests began in earnest during Phase IIIA and are reported separately.

3. SOLVENT HYDROGENATION

As described previously, the recycle solvent quality is a critical variable which impacts on coal liquefaction yields and product quality. In the EDS process, the recycle solvent provides hydrogen for liquefaction; hence, it is given the name "donor solvent." The EDS solvent is derived from the coal liquid product and typically has a boiling range of 400 to 850°F. The coal liquids contain a large amount of aromatics and hydroaromatic compounds. By catalytically hydrotreating the solvent boiling range liquids in a separate fixed bed reactor, much of the aromatics are converted to hydroaromatics such as tetralin as shown below. The hydroaromatic then donates its hydrogen to the coal molecules during liquefaction and the reaction below is reversed.

FIGURE 4

DONOR SOLVENT REACTIONS



Without a high quality donor solvent present, a portion of the coal liquids formed during liquefaction will repolymerize. The donor solvent dramatically reduces repolymerization and provides high yields of distillate liquid products. Besides donating hydrogen, the solvent is the vehicle for transporting the coal into the liquefaction reactor and for promoting the dissolution of the coal particles. Numerous studies were carried out during the EDS Phase I and II development program to evaluate the effect of solvent quality on liquefaction performance and to establish the preferred operating conditions in solvent hydrogenation to produce the desired solvent quality. Some of these studies are described in this section.

There is extensive petroleum refining experience with the design and operation of fixed bed catalytic hydroprocessing for heavy oil hydrodesulfurization and hydroconversion. The EDS process draws upon Exxon's experience with fixed bed catalytic hydroprocessing and extends this technology to hydrogenation of the coal liquid donor solvent. In the EDS program, fixed bed catalytic solvent hydrotreaters were included in all CLPP-1 operations using recycle donor solvent. In these integrated operations, the hydrogenation reactor was coupled directly to the liquefaction reactor via the recycle solvent loop. The process variable studies for solvent hydrogenation were carried out in independent once-through bench scale units, which enabled more extensive evaluation of process variables. Over 1700 material balanced yield periods were carried out during the 1966 to 1975 period. Important variables such as different feedstocks, catalyst types, and reactor operating conditions (temperature, pressure, space velocity, treat gas rate, etc.) were evaluated to determine their effect on solvent quality.

Feedstocks

The feedstocks for the hydrotreating studies were typically obtained from the CLPP pilot plant operations at that time. Initially, a total centrifuge overhead product (centrate) obtained from low severity liquefaction was utilized. This feed contained a significant amount of 850°F+ boiling material as well as a small amount of solids. For this operation, the hydrotreater was operated to convert simultaneously this heavy material to the solvent boiling range (400/850°F) and provide high quality donor solvent. These operations were successful in meeting the solvent quality requirements; however, the hydrotreating catalyst deactivated rapidly. The rapid deactivation was attributed to operation at high temperature (to promote hydroconversion) and to the presence of the high boiling material and solids in the hydrotreater feed.

Several other solvent hydrogenation feedstocks were evaluated. These included coker liquid, spent solvent produced at intermediate and medium severity liquefaction conditions, and a simulated spent solvent produced by mildly hydrotreating a commercially available creosote oil (coal derived). The latter was used when spent solvent produced from the integrated pilot units was unavailable. Results from these studies supported the early solvent hydrogenation study results that the lighter (400/850°F) solvent was preferred to provide long catalyst cycle lengths with minimum catalyst requirements. Removal of solids prior to solvent hydrogenation was essential to prevent catalyst bed fouling.

Catalyst Type

Several commercially available as well as Exxon proprietary hydrotreating catalysts were evaluated during this period. These included cobalt/molybdenum, nickel/molybdenum, and nickel/tungsten catalysts of different sizes and physical properties (i.e., surface area, pore volume). These catalysts were evaluated from the standpoint of activity for the solvent hydrogenation reactions as well as activity and selectivity maintenance. Other important catalyst performance criteria such as sulfur, nitrogen, and oxygen removal were also closely monitored. Based on the Phase I and II data, a nickel/molybdenum catalyst was found to be preferred over the alternatives.

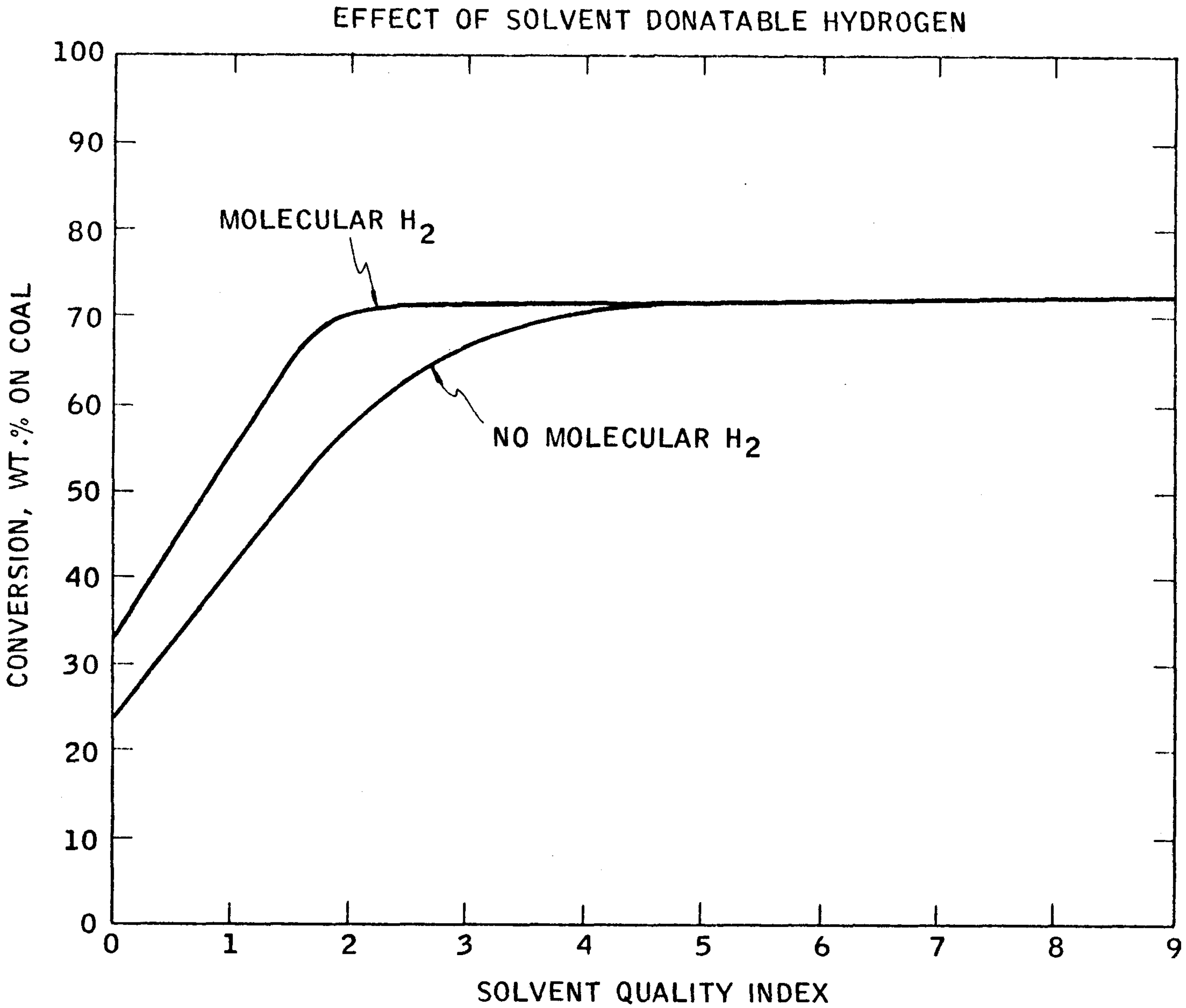
Reactor Operating Variables

Hydrotreating studies covered a wide range of operating conditions from 600 psig to 3000 psig. Treat gas rates were varied by a factor of 10, space velocities were varied by a factor of 20, and temperature covered a 400°F range. Treat gas compositions including pure hydrogen and synthesis gas (H_2/CO) and water mixtures were evaluated. Over this wide range of solvent hydrogenation operating conditions, catalyst activity, activity maintenance, and product quality were monitored. Base operating conditions were selected which would provide catalyst life of a year or greater while providing the required solvent donatable hydrogen content.

Solvent Characterization

The molecular composition of the recycle solvent is an important controllable variable in the EDS process. Extensive research was carried out to characterize the quality and define the molecular composition of the preferred solvent. Model coal compounds such as tetralin and naphthalene were studied as liquefaction solvents. Several solvent parameters were evaluated to determine the most important solvent characteristics needed to provide high coal liquefaction liquid yields. A single parameter was defined, Solvent Quality Index (SQI), which varies with the degree of solvent hydrogenation. Details of the Solvent Quality Index are proprietary; however, the significant effects of this process variable on liquefaction yields are illustrated in Figure 5. As seen in this figure, at intermediate severity conditions of the EDS process without hydrogen present, coal conversion as measured by cyclohexane solubility increases from 25 wt% on coal to over 70 wt% as the SQI increases from 0 to 4. With the addition of molecular hydrogen to the liquefaction reactor, the solvent quality requirements are significantly reduced. With molecular hydrogen present, the maximum coal conversion is reached at a SQI of about 2. Note that even with molecular hydrogen present, liquefaction conversions are significantly lower without adequate solvent quality.

FIGURE 5


EFFECT OF SOLVENT DONATABLE HYDROGEN
MOLECULAR H_2
NO MOLECULAR H_2
CONVERSION, WT.% ON COAL
SOLVENT QUALITY INDEX
0
10
20
30
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4. HYDROGEN MANUFACTURE

Hydrogen was identified in Phase I as an essential ingredient in the production of liquids from coal. Hydrogen supply must be considered for any commercial coal liquefaction plant and several alternative methods for manufacturing hydrogen were evaluated as part of the EDS process development. The various methods and the laboratory and engineering studies conducted are described in this section.

A logic diagram, as shown in Figure 6, was developed to identify the potential methods of hydrogen manufacture. As can be seen in the figure, the primary hydrogen source can be from light hydrocarbons, water or heavy hydrocarbons. Water was indicated to be the major source of hydrogen in the future since light hydrocarbons ranging from methane to naphtha were projected to be in short supply and of high cost while heavy hydrocarbons such as coal contain low percentages of hydrogen.

The second series of alternatives concerns methods for recovery of hydrogen directly from water. One method, direct decomposition, yields both hydrogen and oxygen. The most practical way presently available for accomplishing this is by electrolysis. However, screening studies have shown that, even when taking a substantial oxygen credit, the cost of power must be below the generally projected levels for this process to be competitive. As a result, our efforts were concentrated on the reduction of water to hydrogen, which is the only other possible recovery method.

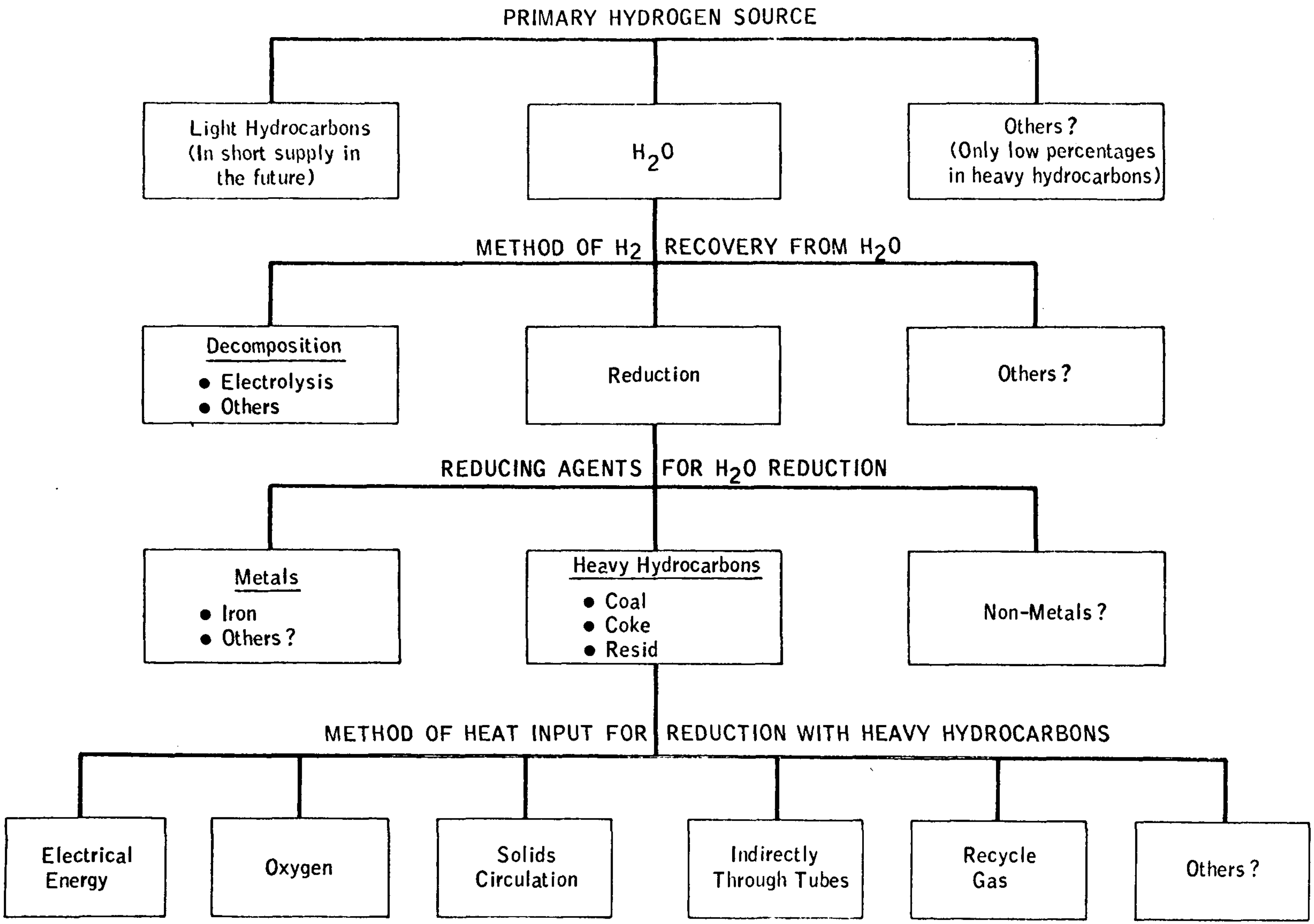
One class of reducing agents which can be used for the reduction of water are heavy hydrocarbons such as coal and coke. These reducing agents tie up the oxygen in the water as CO_2, which is rejected to the atmosphere. A second group of reducing agents is metals. In this case, reduction of water is accompanied by the formation of the metal oxide. However, the oxide must in turn be reduced, for recycle back to the process. Thus, reduction with a carbonaceous material is still required. Screening studies were made for a system using iron or tin as the metal reducing agent (steam-iron process). These studies indicated that although the use of an iron or tin intermediate does provide some benefits, on balance direct reduction with carbon will be less complex and less expensive. No interesting non-metal systems other than hydrocarbons for water reduction were uncovered. Such a system would offer the same potential disadvantage as a metal system; namely, that the oxygen must eventually leave the system as CO_2 since carbon is the only material for which disposal to the surroundings is not a potential pollution hazard or prohibitive in cost.

Thus, discovery of a radical new approach to hydrogen manufacture did not seem probable and as indicated by the final set of alternatives, we concentrated our search for the most economic future hydrogen process on consideration of methods of heat input and reactor configurations for the well-known reaction of H_2O with coke or coal. Heat input by electrical energy, oxygen injection, circulating solids, indirectly thru tubes, and by recycle gas were evaluated. Alternative reactor systems such as entrained, dense fluid bed and moving bed were also evaluated.

FIGURE 6

POTENTIAL METHODS OF HYDROGEN MANUFACTURE

LOGIC DIAGRAM



Hydrogen via Steam Gasification

A laboratory research program was carried out to develop a process to produce hydrogen by steam gasification of coal, coke or coal liquefaction bottoms. The general process scheme envisioned consisted of gasifying coal, coke, or coal liquefaction bottoms with steam at high temperatures (1400-2000°F) in a fluid bed reactor to produce a synthesis gas containing mostly carbon monoxide and hydrogen. The synthesis gas would then be further upgraded to the desired hydrogen purity by water gas shift, carbon dioxide and sulfur removal, and methanation.

Laboratory studies on steam gasification were carried out in units ranging from small bench scale units to the 0.5 T/D Integrated Gasifier-Burner Unit (IGBU). A wide range of process variables for fluid bed gasification were evaluated including:

- temperature
- total pressure
- steam partial pressure
- steam rate
- residence time
- char particle size
- particle surface area
- hydrogen and carbon monoxide inhibition
- coal type (bituminous, subbituminous)
- gasification catalysts

Typically, the gasifier effluent hydrogen content ranged between 50 and 65%. Operating problems were experienced while feeding caking Illinois coal to the gasifier. Agglomeration and gasifier feed system plugging were abetted through close control of the char/coal feed ratio.

The concept of the Transfer Line Burner was evaluated to provide heat to the gasifier. With this concept, char from the gasifier is partially combusted with air in a separate transfer line burner. The char is heated to temperatures several hundred degrees above the gasifier bed temperature by the carbon-oxygen reaction and returned to the gasifier where excess heat is rapidly absorbed by the endothermic steam-carbon gasification reaction. This concept provides a heat balanced gasification process without the need for costly oxygen feed and with an appreciably lower CO_2 content in the product gas (due to separation of the transfer line burner flue gas from the product gas).

In 1971, the steam gasification program objective shifted from hydrogen production to SNG. Studies continued in a proprietary Exxon funded coal gasification research program. The key elements of steam gasification for SNG and hydrogen were similar and therefore the technology developed for SNG would also be applicable for production of hydrogen.

Hydrogen via Steam Reduction with Metals

Exploratory research studies were carried out during Phase I to evaluate steam-metal processes for producing hydrogen. The general process scheme envisioned consisted of reacting a metal with steam at delivery pressure for the hydrogen consumer and at the lowest temperature which was kinetically feasible.

$$M + H_2O \longrightarrow MO + H_2 \text{ (general reaction)}$$

The possibility of generating hydrogen directly at high pressure is attractive because the compression facilities account for a large part of the capital costs and utility requirements for hydrogen manufacture. The lower temperatures favor the equilibrium limited hydrogen-steam ratio in the product. The oxidized metal would be removed to a reduction zone at low pressure, react with a reducing gas such as CO to reduce the oxide to the metal or to a lower oxide. A molten metal which could be pumped into the high pressure hydrogen generator was thought to be preferable.

Experimental work was carried out with iron, zinc and tin as the metal for steam reduction. Studies evaluated both the hydrogen generation step and the subsequent oxide reduction. The separate steps were repeated to determine whether activity declines during cyclic operation. Reaction kinetics were evaluated to determine the extent of metal utilization at the conditions of interest.

A variation of the steam-iron process concept was evaluated in which coal or coke was used to reduce the iron oxide instead of a reducing gas such as CO. Laboratory data showed that relatively pure hydrogen (94%) could be produced through this technique. Experimental work with the metal-steam processes was discontinued when economic evaluations indicated that the steam gasification processes could produce hydrogen at lower cost.

Economic Evaluations

Economic evaluations were carried out on a wide variety of process options for producing hydrogen for coal liquefaction. Screening studies identified the more favorable process options from the standpoint of both economics and likely commercial development. The preferred processes were then studied in more detail. The economic studies evaluated a wide variety of feedstocks for hydrogen manufacture including natural gas, light hydrocarbon gases, liquid hydrocarbons, coal, coke and coal liquefaction bottoms. The hydrogen manufacturing processes evaluated included conventional Steam Reforming, Partial Oxidation, Fluid Bed Gasification, CO_2 Acceptor, Steam-Iron, Electrolysis of Water, Moving Bed Gasification, Entrained Bed Gasification and Oxygen FLEXICOKING.

Initial studies identified Fluid Bed Gasification as the preferred process for producing hydrogen from coal, coke, or liquefaction bottoms. This conceptualized process was evaluated with the endothermic heat of reaction for steam gasification provided by electrical heating, oxygen addition, or through circulating hot char. The latter transfer line burner concept for heat input was incorporated into the Fluid Bed Gasification process as the preferred heat input method. Process economics carried out through 1974 favored this process for hydrogen manufacture over the alternative coal/coke/bottoms gasification processes.

As the Exxon Donor Solvent process evolved, liquid yields, bottoms production, and fuel balances also were changing. During Phase II, screening studies indicated significant cost savings could be achieved through FLUID COKING* of coal liquefaction bottoms to produce additional coal liquid products with subsequent gasification of the coke using Fluid Bed Gasification to produce the hydrogen required in the process. Further studies were then carried out on various coke gasification alternatives. Coke gasification with air was considered for providing plant fuel while using conventional steam reforming (of the light hydrocarbons produced in the liquefaction process) for producing hydrogen. The FLEXICOKING process, which integrates FLUID COKING with air gasification of the coke, in combination with steam reforming for hydrogen production was found to be the most economic process. FLEXICOKING and steam reforming were selected for the 1975/76 EDS Commercial Plant Study Design.

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5. SOLIDS SEPARATION

The separation of solids (unconverted coal and mineral matter) from the liquefaction reactor effluent is a critical processing step in producing coal liquids. Of equal importance is the interaction between the solids separation technique employed and the subsequent processing (if any) or end use of the solids concentrated stream. The interaction of these steps must result in an operable and efficient processing configuration which maximizes the utilization or recovery of valuable constituents originally present in the coal feed.

During Phase I of the EDS development (1966-73) several approaches to solids separation were evaluated. Since these studies typically required large quantities of representative feedstock, the 0.5 T/D coal liquefaction pilot plant (CLPP-1) was a key factor in conducting the solids separation program. Also, solids separation/liquids recovery was absolutely essential for achieving integrated operations and solvent balance with the CLPP-1 facility.

The CLPP-1 studies during Phase I investigated filtration, centrifugation, delayed coking, and vacuum tower applications. In addition, separate laboratory scale-up studies were conducted involving centrifugation, FLUID COKING, solvent de-ashing, and hydroclones. The results of the Phase I solids separation studies led to the selection of the vacuum distillation/ FLEXICOKING processing configuration for the EDS process during Phase II (1974-75). The major laboratory studies and results which led to this selection are summarized below.

Filtration

Filtration was the primary mode of solids separation employed in the initial operations of CLPP-1. Both batch and semicontinuous filters were used in the initial process development work. The batch filters were conventional horizontal plate units and the semicontinuous filter was an experimental pressure rotary filter. The horizontal plate units, because of their inherent batch design, were primarily used as backups for the rotary filter and centrifuge systems. Even in this limited capacity, the frequent disassembly, cleaning, and precoat procedures associated with the batch units proved troublesome and very labor intensive. In addition, seal problems resulted in bypassing feed slurry into clean filtrate streams. When the seal problems were not limiting, the batch filters were capable of providing filtrates containing from 0 to 0.8 weight percent solids. Filtering cycles were limited, however, to about 45 minutes. Because of the batch filter experiences, the bulk of our filtration data were obtained from the semicontinuous rotary filter.

A rotary filter is essentially a rotating drum submerged in the solids containing solution with a filter cake or precoat layer deposited on its outer surface. The undesirable solids are separated from the liquid as it passes through the precoat either by external pressure or internal vacuum. Filtered solids and part of the precoat layer are continuously removed from the rotating drum by an advancing knife edge to maintain filtration rates. Filtration rates depend on type of precoat, drum rotation, knife advance, conditions of precoat after cutting and washing, drum submergence and pressure drop across the filter media. Typically, filtration rates of 10 gph/sq. ft. are considered to be acceptable in commercial applications.

The rotary filter used in our CLPP-1 laboratory program was designed for pressures up to 500 psig and temperatures up to 500°F. Total drum filter area was approximately 5 sq. ft. Operations of the rotary filter were not trouble free. Erratic feed control, fluctuating drum submergence level, and filter pressure drop variations caused repeated loss of or compression of precoat. Nevertheless, much meaningful data were generated from the resulting operations. At typical operating conditions of 300°F and 30 psig differential pressure, filtration rates of about 2 gph/sq. ft. of filter area were achieved. During these operations, the feed from liquefaction to filtration contained about 7 wt.% solids. Filtrate from the rotary drum unit contained about 0.4 wt.% solids. The longest period of operation at these conditions was 24 hours.

The technical feasibility of employing rotary pressure filters to separate unconverted coal and mineral matter from liquefaction slurry effluent was demonstrated during these studies. However, the low filtration rates experienced, coupled with high precoat consumptions and overall operating difficulties did not favor filtration as a commercial means of separating solids from liquefaction product.

Centrifugation

Two types of centrifuges were evaluated in the 1966-1973 Phase I development program. These included both the solid bowl-scroll discharge units and a nozzle-disc unit. The first is designed for concentrating a solids stream while the second has greater capacity for clarifying overhead liquids (centrate). The solid bowl units provided the basic solids separation function for the 0.5 T/D CLPP-1. Since the solid bowl centrifuges employed in CLPP-1 were operated at only a fraction of their design capacity, they also provided the liquid product clarity required for integrated CLPP-1 operations. During the Phase I development program the solid bowl units were also evaluated at or near design capacities in a parallel solids separation scale-up program. The nozzle-disc centrifuge was evaluated only in the latter scale-up program.

Two solid bowl-scroll discharge centrifuges were used in the CLPP-1 operations. In the solid bowl centrifuge, slurry feed enters at one end of a rotating bowl, solids are forced to the outside wall where a scroll conveyor removes them from the bowl. The solids discharge end of the bowl is usually conical to allow drainage of liquid. Clarified liquid flows inward and is skimmed off with an overflow dam located at the opposite end of the bowl. Mechanical factors such as size, speed, and operating conditions are limiting in the application of solid bowl centrifuges. For example, one centrifuge was originally designed for about 15 psig pressure at a maximum of 250°F. The pressure limitation was based on the case seal design and the temperature limitation was based on satisfactory lubrication of the bearings located between the centrifuge scroll and bowl. Working in conjunction with the manufacturer′s design engineers and Exxon′s lubrication experts, the operating limits of the solid bowl centrifuge were increased to about 100 psig at 450°F. However, larger versions of this centrifuge could still be temperature limited because of the tremendous stresses exerted at the periphery of the bowl. At the elevated operating conditions, 5000 g′s force, and 10 gph slurry feed rates, this unit was very efficient in both clarifying liquid and concentrating solids. Typically, it was capable of producing clarified liquid containing less than about 0.5 wt.% ash and a corresponding underflow containing about 45 wt.% solids.

The nozzle-disc centrifuge which was evaluated was too large to be incorporated in the CLPP-1 operations program. Therefore, a special test "Pad" was built to handle up to 200 gph of 500°F feed slurry. The nozzle-disc centrifuge, unlike the solid bowl scroll centrifuge, was designed to operate at temperatures to 600°F and pressures of 150 psig. Briefly, feed enters at the top of a suspended bowl containing a stack of closely-spaced conical discs. The feed distributes outward at the center of the rotating bowl leaving the liquid to then work inward between the discs. The concentrated solids slurry is discharged through nozzles located at the periphery of the rotating bowl. Part of the solids slurry from the nozzles is automatically recycled back to the bottom of the centrifuge bowl. The remaining concentrated solids slurry is removed as centrifuge underflow. The clarification capabilities are improved by increasing rotational speed, increasing the diameter of the discs and decreasing the spacing between the discs.

The nozzle-disc centrifuge capabilities were evaluated on the test "Pad" with various coal liquefaction slurries accumulated from CLPP-1 operations. Typically, with about 6 wt% feed solids concentration at temperatures of 420°F, the solids concentration of the clarified liquid was about 0.8 wt%. Process improvements were evaluated which later decreased this solids concentration to about 0.2 wt.%. The solids concentration in the underflow was typically 15-18 wt%. This is about one-third the solids concentration obtained with the solid bowl centrifuge.

Based on the results obtained during this program, the performance of both solid bowl and nozzle-disc centrifuges could be predicted. In general, the performance of the nozzle-disc centrifuge at design capacities provided product clarities comparable to or superior to the solid bowl centrifuges utilized in CLPP-1. Since the nozzle-disc centrifuges are designed for higher pressure and temperature operations, their applications to coal liquefaction processing should be easier than the solid bowl units. Much liquid product, however, is lost in the underflow and would need to be recovered by subsequent downstream processing.

Coking

Use of centrifuges did not provide solvent balanced operations in the early CLPP-1 operations. Solvent boiling range material was lost to the centrifuge underflow which typically contained about 55 wt% liquids. An effort was therefore undertaken to recover additional liquids from the centrifuge underflow. Initial success in bench scale studies led to the incorporation of a delayed coker into the CLPP-1 flow scheme. The combined solids separation scheme of centrifugation followed by delayed coking of centrifuge underflow provided recoveries of 80 to 90% of the liquids and solvent balanced operation. This was a major breakthrough in the EDS development program and led to more in-depth studies of the coking step.

Delayed coking is a high temperature (~900°F), long residence time pyrolysis process. Gas and liquid products are continuously removed while coke accumulates in a soaking drum which is periodically removed from service for coke removal. Delayed coking was used in several CLPP-1 yield periods at low severity donor liquefaction conditions. Generally, the liquid yields obtained at low severity conditions were about 20 wt% on dry coal feed and the quality of these liquids, particularly their distillability, was poor. Consequently, the low severity processing scheme relied on coking and hydroconversion to supplement liquefaction liquid yields and upgrade liquid product quality.

The initial success with delayed coking led to a more extensive evaluation of FLUID COKING. FLUID COKING is a continuous, non-catalytic pyrolysis process commercialized by Exxon Research and Engineering. It produces hydrocarbon gases, liquids, and coke from heavy, high boiling liquid hydrocarbons such as petroleum vacuum residuum. Studies were carried out on coking several liquefaction streams in a Continuous Stirred Coking Unit. The streams coked included the centrifuge underflow and overflow, recycle solvent, and total liquefaction effluent from different liquefaction operating conditions. An "all coking" solids separation scheme was found to be preferred over the centrifugation/coking method due to its simplicity and lower cost. FLUID COKING of total liquefaction effluent was selected for the low severity donor solvent liquefaction base case and this was used as the 1969 Commercial Plant Study Design basis.

A significant effort was then undertaken to evaluate FLUID COKING of total liquefaction reactor effluent. These studies investigated process variables (reactor temperature, residence time, feed type, steam dilution, etc.), ash and coke particle strength and fines make, product quality, and unit operability limits (bogging). As the EDS development program progressed, significant yield and operability improvements in the liquefaction section were achieved by increasing reactor pressure to the intermediate and medium severity liquefaction conditions. High liquid yields and good operability results were obtained using vacuum distillation instead of centrifugation or coking. Coking was therefore deemphasized in the Phase I program.

During the Phase II period (1974-75), FLUID COKING was again evaluated to provide additional coal liquid yields. At the intermediate severity donor solvent liquefaction conditions, 1000°F+ bottoms were produced in excess of that required for hydrogen production. The excess bottoms which contained coal ash and was high in sulfur was of questionable value as a process byproduct. Studies were therefore carried out to recover additional liquid products from liquefaction bottoms. Bench scale coking studies showed that coking could increase total liquid yields to 40 to 45 wt% on dry coal, depending on the operating conditions chosen for liquefaction. Larger scale coking tests were carried out during the jointly funded phase of the EDS program and are reported separately. As a result of these coking tests, however, the combination of vacuum distillation followed by coking of the vacuum bottoms became an integral part of the EDS process. This process configuration was incorporated into the 1975/76 EDS Commercial Plant Study Design (see Figure 7).

Hydroclones

Hydroclones (liquid cyclones) are a potentially attractive alternate for separating solids from liquids because they have no moving parts, and they can produce higher concentrations of solids than nozzle-disc centrifuges. However, they are sensitive to unstable flow conditions and highly subject to abrasion and/or plugging problems. Hydroclones do not normally provide as good clarification as that of nozzle-disc centrifuges. When used to separate fine particles, up to 300 small diameter (10 mm) hydroclones have been used in parallel. Such installations are currently used in refineries to remove fine catalyst particles from heavy hydrocarbon streams.

The basic driving force for the separations in hydroclones is the pressure drop across the hydroclone. Its capability to remove small particles from a liquid stream is also a function of unit diameter. Therefore, smaller diameter units operating at high pressure drops should remove finer particles. However, the effectiveness of hydroclones is very sensitive to geometry, flow conditions and other system and process variables. Consequently, as in the case of other separations techniques, pilot unit testing is essential to evaluate hydroclone performance.

To evaluate the capabilities of hydroclone separation in the liquefaction program, a single 10 mm diameter hydroclone, constructed from stainless steel, was tested. The test program was carried out over a wide range of variables:

- Feed concentration: 1 to 6 wt.% solids
- Feed rates: 0.9 to 1.6 gpm
- Pressure drop: 50 to 200 psig
- Overflow/underflow product ratio: 0.8 to 10
- Underflow concentration: 2 to 48 wt% solids

Efficiency data were obtained using both aqueous slurries of talc particles and coal products produced from the CLPP-1 operations.

Based on the results of the test program, we concluded that hydroclones do not clarify liquid as well as nozzle-disc centrifuges nor concentrate solids as well as solid bowl centrifuges. However, hydroclones can achieve higher concentrations of solids than nozzle-disc centrifuges. Therefore, depending on the particle sizes to be removed and the concentration of solids desired, hydroclones could be used either to replace or complement centrifuges.

Solvent De-Ashing

A brief study was carried out in batch equipment to evaluate the use of light hydrocarbons to reject the high-ash fraction in the liquefaction reactor effluent. The study was carried out in a de-asphalting treater normally used for petroleum feedstocks. The solvents investigated were n-butane, n-heptane, and blends of these solvents with methyl-ethyl-ketone. The liquefaction product tested was generated during the CLPP-1 low severity donor solvent operations with Illinois coal. The resulting data showed that none of the solvents was selective enough to reject mineral matter and unconverted coal without rejecting significant quantities of coal liquids. Also, the bottoms phase was extremely difficult to remove from the laboratory equipment. Because of the non-selectivity observed in these tests, work on solvent de-ashing with petroleum derived solvents was suspended.

Vacuum Distillation

A vacuum flash tower was used extensively in CLPP-1 operations. Initially vacuum distillation was used to separate solvent boiling range material from heavier coal liquids. Later, in conjunction with the medium severity liquefaction studies, emphasis turned to using vacuum distillation to separate unconverted coal and mineral matter from liquefaction effluent. In the later operation, liquefaction reactor product slurry was fed to the vacuum tower flash zone at 675-700°F and 25 mm Hg absolute pressure. The 1000°F+ vacuum bottoms containing the solids was removed from the bottom of a barometric leg. During this period 36 yield periods and over 1000 hours of operation were logged with the vacuum flash tower in the integrated CLPP-1 pilot plant.

Much valuable operating experience was gained with the CLPP-1 vacuum tower. Initially, operations were limited to temperatures of 650-675°F in the flash zone. This was due to a significant amount of thermal cracking and de-hydrogenation experienced with the liquefaction slurry feed at higher temperatures and the long residence time (~4 hours) of the bottoms slurry in the vacuum tower barometric leg during non-donor liquefaction operations. Operations and liquid recoveries significantly improved when CLPP-1 was changed from the early non-donor liquefaction scheme to the hydrogen donor liquefaction scheme.

Vacuum distillation proved superior to all solids separation equipment previously used in CLPP-1 from the standpoint of continuous operability; minimal maintenance or operator attention required; consistency of results; and the ability to provide solvent balanced operation. Also, engineering studies showed vacuum distillation to be more economically attractive then filtration, centrifugation, hydroclones, or coking of the liquefaction effluent.

Experience gained with the CLPP-1 vacuum tower was incorporated into the CLPP-2 facilities during the 1974-75 period. The barometric leg was replaced with a shorter bottoms section and a bottoms withdrawal pump. This modification reduced the residence time of the vacuum bottoms in the tower from 4 hours to 30 minutes. Several different bottoms pumps were evaluated for this service. In view of the success achieved with distilling liquefaction products from operations with hydrogenated solvents, a preheat furnace was provided capable of increasing the vacuum tower flash zone temperature to 750°F. As part of the engineering technology program, experiments were designed to obtain design and scale-up information related to entrainment of solids, transfer line performance, pump performance, and operability limits for the CLPP-2 vacuum tower. These experiments were and are being carried out as part of the current jointly sponsored EDS Program.

6. PROCESS ENGINEERING AND ECONOMIC STUDIES FOR COMMERCIAL SCALE COAL LIQUEFACTION PLANTS

Throughout the 1966-75 period, process engineering and economic studies paralleling the laboratory program were undertaken. The engineering studies concentrated on

- evaluation of laboratory data
- development of conceptual commercial plant designs based on the then current laboratory operations
- studies of process variables and processing alternatives to define incentives and give direction to the laboratory effort
- development of cost parameters for coal liquefaction process comparisons

The results of these studies led to the choice of process sequence and processing conditions which constitute the EDS process and are to be used in the 250 T/D pilot plant, ECLP.

Early predevelopment work concentrated on completion of study designs which evaluated catalytic coal liquefaction, donor liquefaction, and non-catalytic liquefaction (solvent refined coal). Studies were carried out for bituminous, sub-bituminous, and lignite coal feeds for a commercial plant producing 100 kB/D of gasoline. Filtration, centrifuging, coking, and distillation were evaluated for solids separation. Early studies were based on low severity donor liquefaction (350 psia, 750°F). Both hydrocracking and catalytic cracking were evaluated for conversion of the heavy coal liquids to gasoline.

In 1969, a major study design was completed to develop a set of investments and economics which would provide a sound basis for assessing the outlook for producing gasoline from coal. Approximately 10 man years of engineering effort was expended to develop this conceptual plant design. The design was based on the low severity (350 psig, 775°F) donor liquefaction process. In the study design flow scheme, hydrogen from the donor solvent only was reacted with the coal. The spent solvent was then regenerated in a separate Solvent Hydrogenation reactor. Additional conversion and solids separation was provided by FLUID COKING of the liquefaction reactor effluent with the fluid coke product sold as boiler fuel. Hydrogen for the process was provided by steam reforming natural gas. Heavy coal liquids were converted by catalytic cracking to gasoline blend stocks and naphtha was upgraded via hydrofining and catalytic reforming to gasoline.

Investments and economics were developed for a plant startup in 1972. The investment which included 15% project contingency was 200 M$ for a 30 kB/CD coal conversion plant which included product upgrading to gasoline. With 100% equity financing and appropriate product values (such as 11.2¢/gal for gasoline), a DCF return of only 1.4% was realized. These results led to cost reduction and process improvement studies. Cyclic POWERFORMING* of the full

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range (160/400°F) naphtha was used to improve gasoline octane and recover more hydrogen. The hydrogenation reactors were modified to use liquid water for feed quench (cooling). Hydrogen was recovered from purge gas streams by cryogenic separation, and $C_3^=$ and $C_4^=$ olefins were alkylated to produce additional gasoline blend stocks. These and other process improvements improved the economics and increased the DCF return to 4.2% for the "improved base case".

Recognizing that in the time frame that coal liquefaction is practiced commercially natural gas may be unavailable or very expensive, a series of "no natural gas" alternatives were developed. In these cases, various combinations of using C_3^- gas produced in the process, coke (char) produced in FLUID COKING and/or coal were used as the hydrogen plant feed and for process fuel. Combinations of steam reforming, fluid bed gasification, partial oxidation and coal fired process furnaces were evaluated (see Section 4). Study results showed that an increase in natural gas cost by only 10¢/MBtu (+20%) would make the partial oxidation (of coal/char) for hydrogen production cases a standoff with steam reforming of natural gas. Thus, steam reforming of purchased natural gas was dropped as the hydrogen generation method in future studies. Fluid Bed Gasification of the byproduct char was shown to be the preferred method for production of hydrogen. An "Updated Study Design" was prepared to provide a detailed up-to-date economic assessment of low severity liquefaction which could serve as a base for screening alternatives. This study served in later years as a base point to evaluate other process configurations.

Subsequent to the 1969 Study Design, economic evaluations were carried out to find an improved coal liquefaction process. Various process configurations were screened in conjunction with the laboratory studies described in Section 2. These processing options included:

- low severity donor liquefaction (350 psig, 775°F)
- non-donor liquefaction at medium severity (2500 psig, 850°F)
- donor liquefaction with synthesis gas ($H_2/CO/H_2O$) at medium severity
- intermediate severity donor liquefaction (1800 Psig, 825°F)

The plant size for each case was set at 30,000 B/D of C_4/1000°F coal liquids produced from Illinois coal. The results of this study are summarized below.

	Low Severity Donor	Non-Donor	Syn Gas	Intermediate Severity Donor
Investment	Base	+ 13%	Base	Base
Coal Liquids Cost	Base	+ 9%	+7%	-7%

Intermediate severity donor coal liquefaction was shown to produce coal liquids at about 7% lower cost than the low severity donor liquefaction base case. In addition, as described in the sections on Liquefaction and

Solids Separation, operations of the 0.5 T/D pilot plant (CLPP-1) were the most trouble-free with these intermediate severity liquefaction operating conditions with vacuum fractionation for solids separation. The thrust of the EDS research program was then directed toward the development of this process configuration which became known as the Exxon Donor Solvent Coal Liquefaction Process.

Numerous process improvement studies were carried out on the basic EDS process configuration. Process variables such as reactor temperature, residence time, pressure, and solvent quality were studied to determine their impact on process economics. Alternative coals were evaluated as were several bottoms processing/hydrogen production alternatives (described in Section 4). In 1975, work began on a comprehensive EDS Commercial Plant Study Design using the block flow plan shown in Figure 2. The coal conversion plant was designed to feed 24 kT/SD of cleaned Illinois No. 6 coal to produce 60 kB/SD of C_3+ coal liquids. The principal products from the plant are C_3 and C_4 LPG, naphtha and low sulfur fuel oil. The LPG and LSFO leave the plant as finished products while the naphtha requires further downstream processing prior to its use as gasoline blend stock or in aromatics production. The coordination flow plan for the EDS Study Design is shown in Figure 7. An overall thermal efficiency for the plant of 63.3% was calculated. Planning, process engineering and most of the equipment design for the EDS Study Design were completed during Phase II (1974/75) while the remaining equipment design, investments, and economics were developed in 1976 during the jointly sponsored research program, Phase IIIA. Complete results of this study are presented in DOE Report FE-2353-13, EDS Coal Liquefaction Commercial Plant Study Design.

In summary, the process engineering and economic studies provided guidance for laboratory studies as well as defining the preferred processing conditions for coal liquefaction. This led to the logical evolution of the EDS Process. As the coal liquefaction data base is expanded and additional process engineering and economic studies are carried out, further improvements are anticipated in the EDS Process and confidence in the process reliability and commercial attractiveness are expected to increase.

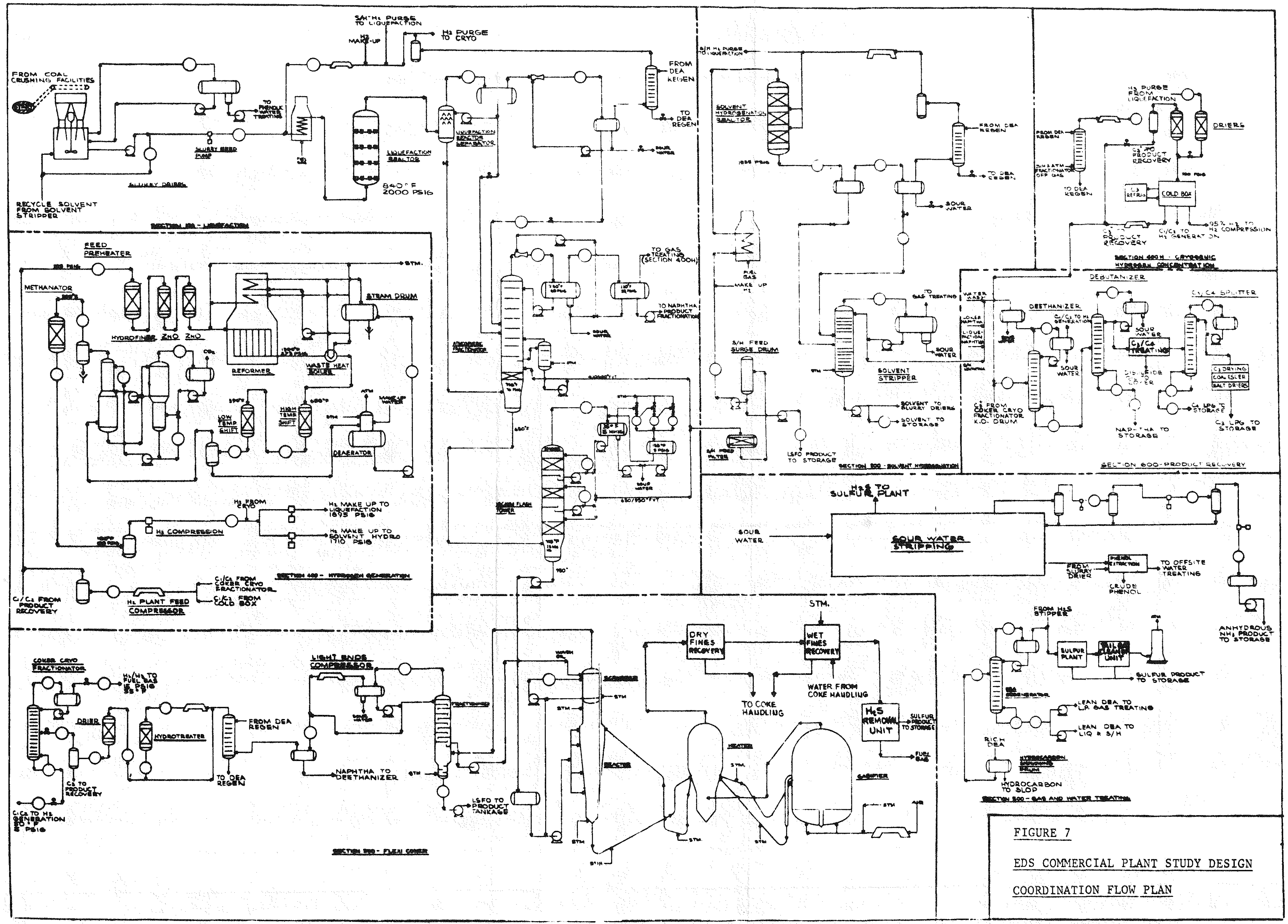

FIGURE 7

EDS COMMERCIAL PLANT STUDY DESIGN

COORDINATION FLOW PLAN

7. SUPPORTING RESEARCH

Numerous supporting research programs were conducted as part of the EDS integrated R&D program. These programs increased our overall understanding of coal processing, guided the development of a technically feasible processing concept and produced fundamental physical data for use in the design of commercial coal liquefaction plants.

Two types of supporting research programs were conducted during the Phase I and II period: on-going programs such as research in analytical and petrographic techniques, which contributed to the development program on a day-to-day basis; and specific engineering technology development programs such as coal slurry drying, slurry preheat furnace tube coking, chemical engineering unit operation and physical properties studies which were directed to longer range aspects of developing the EDS Process. This section summarizes some of these studies.

Slurry Drying

In most coal liquefaction processes it is advantageous to remove the moisture from coal prior to subjecting the coal slurry to liquefaction conditions. By doing so the problems associated with slurry preheating and the required size of the downstream liquefaction reactor are reduced substantially.

Conventional methods of drying coal usually employ hot oxygen containing gases. The low heat capacities of gases require the circulation of large quantities of gas in these applications. Consequently, not only are the energy requirements large but there are problems associated with removing fine solids from the gas so as not to lead to an air pollution problem. When drying a combustible material like coal, potential fire and explosion hazards are created if the oxygen content of the drying system is not properly controlled. In addition, oxidation of the coal may occur. Oxidation may lead to lower liquid yields in the liquefaction step.

To avoid the problems associated with gas drying systems, ER&E developed a process to simultaneously dry the coal while preparing the coal slurry feed for liquefaction. In this scheme, crushed coal and hot recycle solvent are fed separately to a well-stirred tank maintained at about 250-350°F and near atmospheric pressure. Moisture evolves from the coal particles and bubbles up through the hot solvent. No oxygen comes in contact with the hot coal. The evolving moisture generates a foam, so space is provided for this. The evolved moisture along with some stripped solvent vapor is withdrawn from the slurry drier and condensed. Condensed solvent is returned to the drier while the water is removed for subsequent water treating. Adequate residence time is provided to dry the coal slurry to less than 4 wt% on dry coal feed. Heat required to vaporize the water from the coal is provided by recycling a portion of the slurry through a heat exchanger.

Laboratory tests were carried out to evaluate the effects on drying rate of particle diameter, temperature, nature of the solvent, coal type, and degree of agitation. Quantitative foam volume data and analyses of the evolved water were collected. The coal drying system was modeled to enable adjustments to conditions and coals other than those tested. Engineering design studies evaluated alternative process conditions and equipment options to arrive at a preferred unit design. The preferred features have been incorporated into the design of the 250 T/D pilot plant (ECLP).

Slurry Heating

The preheating of coal/solvent slurry or coal/solvent/treat gas feeds to liquefaction were identified early as potentially a major technical problem in the development of the EDS process. The slurry furnace "tube side" operating conditions of about 840°F and 2000 psig with a high solids stream were beyond the range of available process data. The operating problems which might be encountered included:

- plugging of lines, valves, and pump check valves
- erosion of lines and equipment
- fouling and plugging of heat exchanger and furnace tubes

To avoid these problems, adequate pilot plant experience and specialized engineering data for scale-up are necessary. Studies were carried out in the 0.5 T/D pilot plant (CLPP-1) as well as in a number of parallel engineering technology facilities. The data obtained included the following:

- Furnace tube coking data - Evaluated coking rates in CLPP-1 as a function of slurry film temperature (up to 950°F), slurry velocity, and solvent/coal ratio.

- Slurry viscosity data - Obtained viscosities for both swelling and non-swelling coals as a function of temperature, solvent/coal ratio, coal size and solvent type in CLPP-1 and in a separate slurry pumparound loop. These data are essential to heat exchanger and furnace tube design as well as pumps and piping.

- Heat transfer data - Measured heat transfer and system pressure drop data for a range of coal sizes, solvent types, solvent/coal ratios and temperatures for both the coal/solvent and coal/solvent/gas mixtures. New empirical correlations were developed for predicting coal slurry heat transfer coefficients for both turbulent and transition flow.

- Engineering design studies - Evaluated alternative fuels for coal liquefaction furnaces including coal and low BTU gas fired process furnaces.

The slurry heating studies included an analysis of scaleup requirements for a commercial plant slurry furnace as well as design of the slurry preheater furnace for the 250 T/D pilot plant (ECLP) as discussed in Section 10. The slurry preheater coking studies are presently continuing in conjunction with the operation of the 1 T/D pilot plant (CLPP-2).

Liquefaction Reactor

"Cold model" studies were carried out to simulate the three phase (coal/solvent/treat gas) coal liquefaction reactor. These tests are important to provide knowledge of the hydrodynamic characteristics of reactors to adequately scale up the results of the pilot plant program to a commercial plant size. In addition, they are needed to assess any operability constraints on the commercial plant.

The "cold model" tests were carried out in a 6-inch diameter by 10-feet high column at ambient temperature and pressure. Heptane and nitrogen were used to simulate the properties of the donor solvent and treat gas/vaporized solvent, respectively. Batch and continuous coal slurry feed operations were carried out with coal particle size up to minus 8 mesh and with coal concentrations up to 41 wt%. Bed expansion data were obtained over a range of gas and liquid velocities, coal particle size, and solvent/coal ratio. The presence of stagnant zones in the reactor were monitored, as was the particle size segregation. Large particles were found to concentrate near the inlet distributor indicating that it would be necessary to provide facilities for withdrawing larger particles from a commercial plant reactor to minimize potential reactor/distributor plugging problems.

A larger 24-inch diameter "cold model" was built to extend the liquefaction reactor studies to evaluate the inlet distributor design and solids withdrawal. These studies were carried out in the current jointly funded research program and are separately reported.

Other Engineering Technology Programs

Several other engineering technology programs were carried out during the Phase I and II period. Thermodynamic properties of coal liquids and coal slurries were obtained where possible from the on-going laboratory program. In certain areas, additional studies were required. A program was initiated with Brigham Young University to measure the vapor/liquid equilibrium of H_2-solvent mixtures at elevated temperature and pressure as well as density, viscosity and surface tension of coal liquids.

A materials test program evaluated erosion/corrosion of coal conversion streams (coal/solvent slurries and liquefaction reactor products). Where possible, data from petroleum processing were used and correlations were extended to the coal liquefaction conditions. Equipment such as pumps and instrumentation for coal slurry service were tested at proposed commercial plant operating conditions. These engineering technology programs are an essential part of the integrated R&D approach and are continuing through the jointly funded research program.

8. PRODUCT QUALITY AND COAL LIQUIDS UPGRADING

A significant effort was undertaken to determine coal liquefaction product quality as well as the upgrading required to make finished fuel products. Planning studies were also carried out to determine the value of coal liquids as a refinery feedstock. Some of these studies are described below.

Product Quality

A wide range of products can be produced via the EDS coal liquefaction process. These can include high BTU gas, low BTU gas, liquified petroleum gas (LPG), naphtha, low sulfur fuel oil (LSFO), and liquefaction bottoms (a solid product). The liquid products can be combined to form a synthetic crude product. In addition, chemicals such as phenols and aromatics may be extracted. As the EDS process was directed towards liquid fuels production, little effort was undertaken for evaluation of potential chemicals production. Also, the gas and solid fuels were considered byproducts of the liquid fuels operations. Analyses of the coal liquids typically produced in the EDS process are presented on Table 2. Included are the qualities of raw and hydrotreated naphtha and fuel oil as well as analysis of the raw pyrolysis (coking) liquid product. The product analyses were obtained at the intermediate severity donor solvent operating conditions at the end of the Phase I/II period.

Product Flexibility

The EDS process has the flexibility to vary the product slate among liquid, gas, and solid products for fuels or chemicals production. By varying the distillation cut points, products such as home heating oils and turbine fuels could be recovered separately from the heavy fuel oil (some product hydrotreating may be required). The process operating severity (i.e. temperature, residence time, solvent quality) can also be changed to shift the liquid product slate between light and heavy products. The data for Illinois coal indicate the flexibility to vary the naphtha ($C_5/400^oF$) to fuel oil (400^oF+) ratio over the range of 1:2 to 2:1.

Product Upgrading

The initial objective of the Exxon coal liquefaction program was to produce naphtha boiling range products that would be suitable for producing gasoline or gasoline blend stocks. Various methods were therefore evaluated for upgrading and converting 400^oF+ coal liquids to naphtha. During Phase II, the program objective was expanded to include middle distillates and low sulfur fuel oil products.

TABLE 2

Typical EDS Product Qualities

	Heavy Naphtha(1)(2)		400°F+ Fuel Oil(1)		Pyrolysis Liquid
	Raw Liquid	Hydrotreated Liquid	Raw Liquid	Hydrotreated Liquid	Raw Liquid
Nominal Boiling Range, °F	160/400	160/400	400/1000	400/1000	400/1200
Distillation, 15/5					
10 wt%	222	198	477	462	450
50 wt%	356	314	694	657	946
90 wt%	391	360	812	774	1100
Specific Gravity, 60/60°F	0.87	0.80	1.08	1.01	1.22
Elemental Analysis, wt%					
C	85.60	86.80	89.40	90.80	88.60
H	10.90	12.90	7.70	8.60	5.90
O	2.82	0.23	1.83	0.32	3.10
N	0.21	0.06	0.66	0.24	1.30
S	0.47	0.005	0.41	0.04	1.10
Higher Heating Value, BTU/lb.	18,300	19,300	17,100	18,100	16,100

Notes:

(1) Excludes pyrolysis liquid
(2) Excludes C_6/160°F naphtha cut

Laboratory hydrotreating studies were carried out with the following objectives:

- Naphtha hydrotreating to reduce O, N, and S to acceptable levels for catalytic reforming. Processing conditions more severe (~750 psig, 650°F) than petroleum naphtha hydrotreating were required to meet specifications.
- Middle distillate hydrotreating to prepare hydrocracking feedstocks.
- Gas oil (350/970°F) hydrotreating to prepare catalytic cracking feedstocks.

Coal liquids conversion studies were conducted in the areas of catalytic reforming, hydrocracking, and catalytic cracking. Results of these studies were that (1) coal liquids in the naphtha boiling range make excellent catalytic reforming feedstocks due to their high naphthene and aromatics concentrations and (2) coal liquids are much more difficult to convert to lighter products than petroleum feeds.

Coal Liquids Planning Studies

Evaluations were made throughout the Phase I and II period as to the cost and preferred route for converting coal liquids to lighter products. A study was also undertaken to evaluate coal liquids as a refinery feedstock in comparison to Arabian Light crude. The results of the latter study had a significant impact on setting the product slate for the EDS Commercial Plant Study Design (1975/76) and in giving direction to the research program. Using a proprietary linear programming model, two coal liquids feedstocks from the EDS process (a hydrotreated syncrude and a naphtha/LSFO product mix) were evaluated as feed to a 100,000 B/D grass roots refinery running mixtures of EDS coal liquids and Arabian Light petroleum crude. The overall conclusion of the study was that the most economical way to utilize heavy coal liquids is by using them directly as heating oil and fuel oil products and to increase conversion of heavy petroleum fractions to maintain gasoline and distillate production. The specific conclusions are as follows:

- Coal liquids are worth up to 2$/Bbl more than the Arabian Light crude (@ 12.50$/Bbl) they displace when they constitute 25% or less of total crude input.
- Upgrading the 450°F+ fraction by hydrotreating in the liquefaction plant did not increase the value of coal liquids.
- Catalytic reforming of the 350°F- naphtha provided high yields of hydrogen and high octane gasoline.
- Coal liquid fractions boiling above 350°F had high values as 0.7% sulfur fuel oil due to their low sulfur content and high volumetric heat content.

- There are substantial disincentives for converting 350°F+ coal liquids by either catalytic cracking or hydrocracking.
- The 350°F+ coal liquids should be utilized as low sulfur fuel oil or heating oil, displacing petroleum fractions to catalytic cracking feed.

9. COMPETITIVE TECHNOLOGY

Throughout the Phase I/II period, economic and engineering evaluations were prepared for a number of alternative routes to liquid fuels from coal. The purpose of these studies was to determine whether the EDS process had an economic advantage compared to the alternatives, as well as compare the key process features from a standpoint of operability and likely commercial development. This section summarizes the results of these studies. It is not the intent in this discussion to imply that other processes cannot or should not be developed.

In general, there are three levels of processing severity used to derive liquid fuels from coal. The least severe approach is coal pyrolysis. Heat is used to drive off or "distill" the volatile portions of the coal, but without adding hydrogen. Liquids, gas, and a large quantity of residual char are formed.

A second level of severity uses heat, pressure, and hydrogen. The coal is processed under high pressure at moderate temperatures using a liquid solvent to transport the coal and hydrogen to enhance liquid yields. Volatile matter is driven off and portions of the coal molecule are broken down. Hydrogen is added to chemically react with some of the carbon to form hydrocarbons with molecular weights lower than the original coal molecules. This general approach is usually described as coal liquefaction. The EDS process falls in this category.

Finally, the third level of severity is to convert the coal molecule into hydrogen and carbon oxides through very high temperature processing and then catalytically recombine the hydrogen and carbon oxides into hydrocarbon materials. This approach incorporates coal gasification with steam and oxygen followed by the Fischer-Tropsch synthesis reaction to produce the liquid hydrocarbons. Alternatively, the hydrogen and carbon oxides could be catalytically reacted to form methanol.

Pyrolysis

Several coal pyrolysis processes, as shown below, were evaluated. These processes differ in the method of heat addition to the pyrolysis reactor, the pyrolysis temperature, and the coal heat up rate (or residence time). Proposed commercial schemes differ widely as to downstream product upgrading and product slate. The table below summarizes the processes evaluated, the heat addition method, and the reported liquid yield as percent of Fischer Assay (a standardized laboratory pyrolysis test).

The calculated liquid cost showed no economic advantage over the EDS process. Pyrolysis liquid yield was lower than for EDS even without considering the additional liquid yields obtained from FLEXICOKING of the EDS liquefaction bottoms. The value and potential market for the pyrolysis char was uncertain. Thus, development of the EDS process was continued.

TABLE 3

COMPARISON OF PYROLYSIS PROCESSES

Process	Heat Addition Method	Liquid Yield(1)
Garrett	Circulating hot solids	200
Lurgi-Ruhrgas	Circulating hot solids	180
FLUID COKING/ FLEXICOKING	Circulating hot solids	100
TOSCOAL	Circulating ceramic balls	100
FMC-COED	O_2 injection into fluid bed	100
Combustion Retort	Direct air injection into moving bed	84

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(1) Percent of Fischer Assay

Coal Liquefaction

Liquefaction processes utilize increased processing severity to convert coal into liquids by the reaction of coal with hydrogen at high temperature and high pressure. The broad range of conditions at which liquefaction has been tested is from about 750°F and 300 psig up to 900°F and 10,000 psig. Processes under development during Phase I and II were generally under 5000 psig. The coal liquefaction processes differ in severity (temperature, pressure), in the nature of hydrogen addition (hydrogen donor solvent, molecular hydrogen, or both), the use of catalyst (none, liquefaction catalyst, or solvent hydrogenation catalyst), means of solids separation and method of hydrogen production. Product slates vary from a solid fuel product (Solvent Refined Coal-SRC) to high quality distillate liquid products and some include SNG co-products. The processes that were evaluated and their major features are described in Table 4.

An assessment was made of these processes from the standpoint of operability and ease of commercial development. The analysis reflected both experimental work carried out in the EDS Phase I/II program and literature references. In our judgement, the EDS process was found to have advantages over the alternative processes in the following areas:

- Solids Separation - Use of vacuum distillation is preferred over the alternatives based on the experimental work reported in Section 3.

- Liquefaction Reactor - Use of a simple, plug flow reactor is judged to be more operable, reliable, and easier to develop than those containing catalyst or internal/external recirculation.

- Separate Solvent Hydrogenation - This protects the catalyst from heavy coal liquids (1000°F+) and coal solids which cause rapid catalyst

TABLE 4

COMPARISON OF EDS PROCESS WITH OTHER LIQUEFACTION SYSTEMS

	CONSOL Donor	PAMCO SRC	H-Coal
Liquefaction Temperature, °F	725-800	800-900	800-900
Liquefaction Pressure, psig	250-500	1000-1500	3000
Liquefaction Reactor Type	Upward Plug Flow, No Catalyst, Internal Mixers (?)	Upward Plug Flow, No Catalyst	Catalytic Ebullating Bed
Solvent Hydrogenation	Separate Ebullation Bed Bed	None	In-Situ in Liquefaction Reactor
Catalyst Life, mos.	2-5 (?)	---	ca. 1
Solids Separation	Hydroclones	Filtration	Vacuum Distillation
Product	$C_4/1000^{\circ}F$ Liquids	Solvent Refined Coal	$C_4/1000$ Liquids

	Synthoil	EDS
Liquefaction Temperature, °F	800-900	800-900
Liquefaction Pressure, psig	4000	1500-2000
Liquefaction Reactor Type	Upward Plug Flow, Fixed	Upward Plug Flow, No Catalyst
Catalyst Bed		
Solvent Hydrogenation	In-Situ in Liquefaction	Separate Fixed Catalyst Bed
Reactor		$\sim$1 year
Catalyst Life, mos.	1	
Solids Separation	Centrifugation or Filtration	Vacuum Distillation
Product	C_4+ Liquids (Appreciable Portion of $1000^{\circ}F+$)	$C_4/1000^{\circ}F$ Liquids

deactivation. Process control in liquefaction is simplified since the high release of reaction heat occurs in fixed bed catalytic hydrogenation reactor (demonstrated petroleum technology) rather than in liquefaction.

- Product Quality - A liquid product as produced in the EDS process has a wider range of possible uses, including turbine fuel, heating oil, gasoline, boiler fuel, and refinery feedstock. A solid fuel (SRC) would be probably limited to boiler fuel and present space problems for storage in certain metropolitan areas. The improved stability of the liquefaction reactor product in EDS and other processes employing hydrogenation catalysts improves operability in downstream processing and solids separation.
- Feed Preparation - EDS crushes coal to minus 8 mesh and then dries in a slurry drier. Processes employing direct catalytic liquefaction require a smaller size coal which is more expensive to prepare. Also, drying via fluid bed drying can oxidize coal and result in loss of liquid yield.

The EDS process was found to be competitive in cost with the alternative coal liquefaction processes for the same coals and products. The most important criteria in considering different processes was thought to be the probability for successful technical development, reliability, and flexibility. EDS was felt to have advantages in this area compared to the alternatives.

Gasification/Liquids Synthesis

The most severe processing approach for making liquid hydrocarbons from coal is the combination of coal gasification followed by Fischer-Tropsch synthesis. This process sequence has been demonstrated commercially in South Africa by SASOL. At SASOL, multiple Lurgi coal gasifiers convert the coal to a raw synthesis gas. This gas is fed to the Fischer-Tropsch reactors where the hydrogen and carbon monoxide in the synthesis gas are converted into liquid hydrocarbons. A large amount of hydrocarbon gas and chemical byproducts are also produced. Liquid yields and processing efficiency by this process were found to be relatively low compared to the direct coal liquefaction processes. Also, the economics of Fischer-Tropsch synthesis appeared unfavorable for fuel products. The new coal liquefaction processes and specifically the EDS process offered significant improvements over the commercialized Lurgi/Fischer-Tropsch technology.

Alternately, methanol may be produced from the synthesis gas produced from coal gasification. Synthesis of methanol at 50-100 atm. (such as via the ICI process) is widely practiced commercially on synthesis gas produced by steam reforming of light hydrocarbons. Also, both the Lurgi and Koppers-Totzek coal gasification processes represent commercially demonstrated technology for producing synthesis gas from coal.

Thus, both of the major steps required for production of methanol from coal have been practiced commercially.

Evaluations of various schemes for producing methanol indicated that methanol was higher in cost than coal liquids as a source of energy. Also, studies reported in the literature indicated that there could be problems associated with using methanol in gasoline, although it could find other applications where a clean burning fuel was required.

10. EXXON COAL LIQUEFACTION PILOT PLANT (ECLP) PLANNING AND DESIGN

To meet the goal of developing the EDS technology to commercial readiness, an integrated R&D approach is being used. This approach includes an active R&D program based on bench scale research, small pilot unit operation and engineering/design studies integrated with the operation of a 250 T/D pilot plant (ECLP). In some areas such as process equipment design for coal slurry service, adequate data for process scaleup cannot be obtained on the smaller scale (e.g. 1 T/D pilot plant) equipment. Thus the large pilot plant (250 T/D) operations provide a smooth transition from the laboratory to the design of a commercial plant. We believe the integrated R&D approach offers an effective strategy for development of complex technology while minimizing the cost.

Selection of The Development Strategy

Several commercialization strategies for the EDS process were considered before the definitive planning and design of the 250 T/D Pilot Plant for the EDS coal liquefaction process was started. The objective of the selected strategy was to bring the EDS technology to the stage that direct duplication of facilities in subsequent EDS plants could be carried out at reasonable risk. This would be done at a minimum total cost - initial investment plus operating/fix up costs. Options evaluated include:

- Construction of a commercial plant directly from the results of the 1 T/D pilot plant (CLPP).
- Construction of a 10,000 B/D demonstration plant (single commercial size train), followed by construction of a single commercial plant.
- Construction of an appropriately sized large pilot plant from which scale up to a single commercial plant would be made.

The third of these alternatives was chosen as the development strategy. It was concluded that the least expensive, easiest, and quickest way to achieve integrated operation is in a large pilot plant rather than in a commercial unit. The basic advantages are: (1) changes to make the plant operable can be made more rapidly and at lower cost because smaller equipment is used, (2) more alternatives can be tested because their total effect on cost and schedule is less, (3) the pilot facility can be designed to process and test more than one coal, and (4) the failure to recognize a potential environmental problem in the large pilot plant has a less severe impact on the surroundings because of the relatively small size of the facility.

Definitive Planning for ECLP

Definitive planning for the large EDS Coal Liquefaction Pilot Plant (ECLP) was started early in 1974, basic onsite design started in September 1974 and cost estimating work was started in 1975. This work was essentially completed by the end of 1975. The definitive planning work for ECLP consisted of setting the basis for the plant design in the following major areas:

- Process Basis Items
    - Basic process flowplan including alternative operations
    - Unit operating conditions
    - Unit yields
- Project Basis Items
    - Feed and product sources, dispositions, characteristics, etc.
    - Utility supply and other services sources
    - Expected plant operating life and service factors
    - Special research program requirements - materials test program, data logging, etc.

The results of these planning studies were a series of design basis specifications covering the onsite and offsite facilities from which the design specifications were prepared.

ECLP Flowplan

Figure 8 shows the schematic flowplan for the base case design for ECLP.

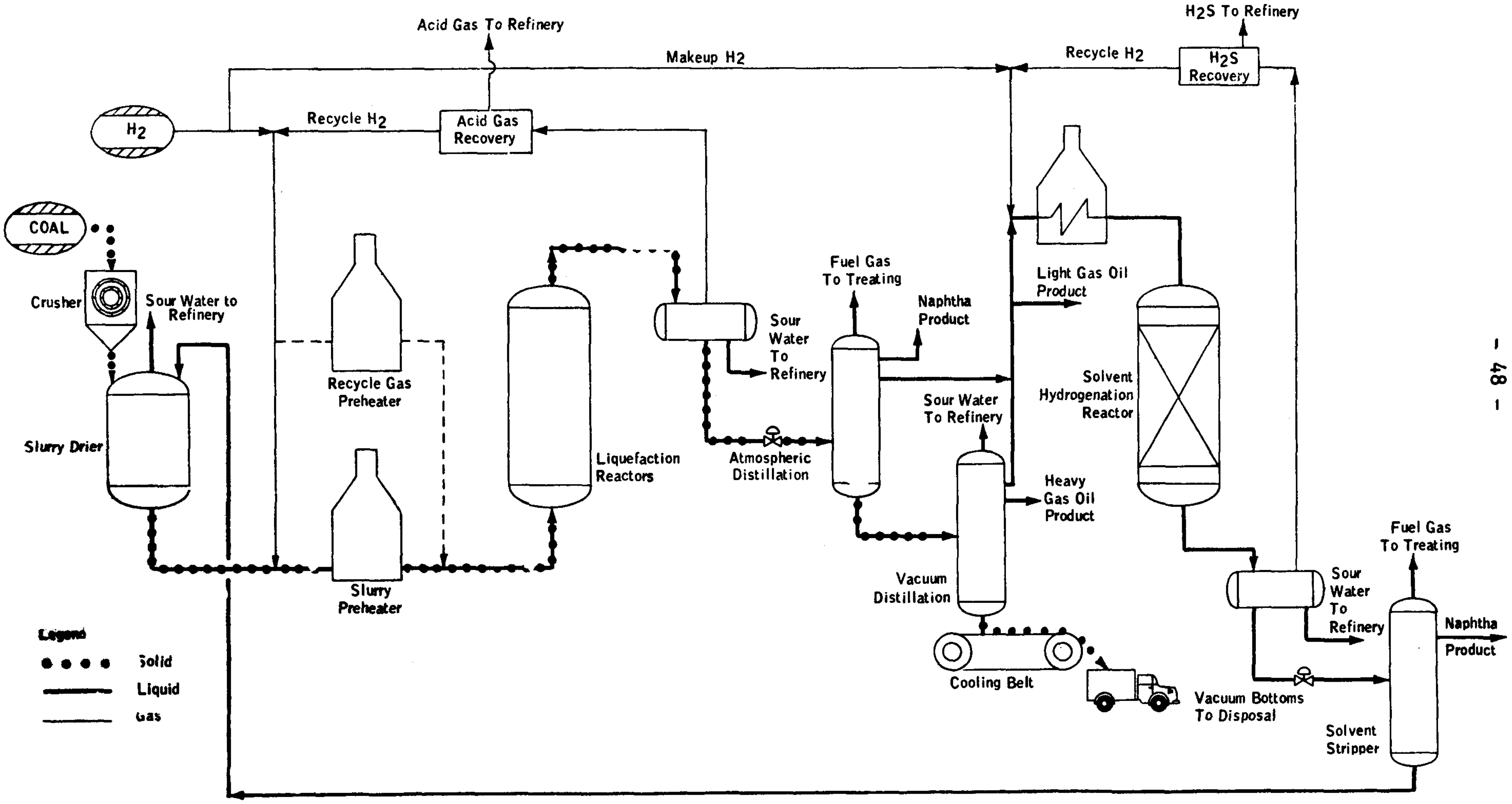
Coal from storage is crushed and then fed to the slurry drier. Here the coal is slurried with a 400/850°F hydrogen donor solvent and heated to drive off the water that enters with the feed coal. After drying, the coal slurry is pumped and heated, mixed with a preheated hydrogen rich recycle gas and fed to the liquefaction reactors which operate at about 2000 psig and 840°F. In the liquefaction reactors, hydrogen from the donor solvent and the gas phase is transferred to the coal to liquefy a substantial portion of the coal.

Products from the liquefaction reactors are separated into (1) a gas stream which is scrubbed for ammonia and hydrogen sulfide removal and recycled to liquefaction; and (2) a liquid/ solids stream which is processed by atmospheric and vacuum distillation into different boiling range liquid cuts (naphtha, solvent, gas oils, etc.) and a vacuum bottoms cut which contains the solids (unreacted coal and ash). The vacuum bottoms slurry is solidified for disposal. Selected liquid streams are fed to Solvent Hydrogenation or to product storage. The offgas from the Atmospheric Fractionator Tower and the Vacuum Stripper Tower is sent to Fuel Gas Treating.

In Solvent Hydrogenation, the spent solvent is rejuvenated by hydrogenation over a fixed catalyst bed. Naphtha and gas oil in the feed are also upgraded. Liquid feed is pumped to pressure, mixed with recycle treat gas and preheated to reactor conditions. Effluent from the Solvent Hydrogenation Reactors is cooled and separated into a hydrogen

FIGURE 8

SCHEMATIC FLOW PLAN
EXXON COAL LIQUEFACTION PILOT PLANT (ECLP)



rich recycle gas and a Solvent Fractionator Tower feed. The recycle gas is scrubbed for ammonia and hydrogen sulfide removal and recycled to Solvent Hydrogenation. The recycle solvent is separated from the naphtha in the Solvent Fractionator and recycled to the liquefaction reactor.

A gas Oil Hydroconversion Unit was originally included in the project to convert vacuum gas oil to additional solvent. Additional laboratory data obtained after the design was completed showed this unit to be unnecessary and it was deleted from the ECLP project. FLEXICOKING of coal liquefaction bottoms and steam reforming to produce the hydrogen required for a commercial EDS plant have not been included in ECLP. Both of these processes are commercially-proven technology for petroleum processing. The EDS integrated R&D program includes laboratory and engineering studies to extend the FLEXICOKING technology to coal liquefaction bottoms. Further study of bottoms processing technology, recently concluded, recommended that a large scale demonstration of FLEXICOKING on ECLP coal liquefaction bottoms be a part of the EDS cooperative development program. A 750 B/D FLEXICOKING prototype unit, adjacent to the ECLP site in Exxon's Baytown Refinery, is available for the demonstration.

Alternative Operations

In addition to the base case operation just described, provision was made in the design for a number of alternative operations. For example, the plant was designed to process a high moisture content Wyoming subbituminous coal as well as the base case Illinois No. 6 bituminous coal; minus 30 mesh dry coal as well as minus 8 mesh wet coal feed can be supplied to the slurry drier; and processing of alternative boiling range feeds in Solvent Hydrogenation is possible since both the atmospheric fractionator in liquefaction and the solvent fractionator can produce side stream products. In addition, provision has been made to operate at various levels of pressure, temperature, residence time and solvent to coal ratio in the liquefaction reactor; various flash zone temperatures in Product Distillation; and higher pressure in Solvent Hydrogenation. Two other alternatives which represent a major change in the process flowplan and involve integration of the Liquefaction and Solvent Hydrogenation treat gas circuits have also been provided. They have the potential for reducing plant investment for a commercial EDS plant, but may be more difficult to operate because of the closer integration of the Liquefaction and Solvent Hydrogenation sections.

Choice of ECLP Plant Size

A substantial effort was devoted to setting the size of ECLP. This effort involved both process engineers and technology specialists. Each processing step in the ECLP facility was analyzed for scaleup requirements. Based on this analysis, the coal liquefaction reactors and the coal slurry preheat furnace were identified as the key equipment items which set the basis for the minimum size for ECLP. Detailed consideration of both of these areas led to the conclusion that 250 T/D is the proper minimum size for ECLP.

The studies showed that the large pilot plant reactor should be designed to duplicate commercial performance in the following key aspects:

- Same distributor design and operating characteristics
- Same gas/liquid velocities
- Same solid suspension characteristics
- Same gas/liquid/solid contacting
- Same plug flow behavior
- Same solids withdrawal system for largest particles

Based on a detailed analysis of the above factors, a two-foot diameter reactor was chosen as the proper size to provide the required scaleup data. This reactor is essentially a vertical "core" of a 10-15 foot diameter commercial reactor. The two-foot diameter reactor and the gas velocity which engineering studies indicated would insure good particle suspension and fluidization set the ECLP coal feed rate at about 250 T/D.

The analysis of scaleup requirements for the slurry preheater showed that the following design parameters must be essentially the same in the large pilot plant as in a commercial slurry preheat furnace to provide adequate scaleup and design basis:

- Flow regime (turbulent)
- Film temperature
- Residence time
- Upper and lower flow velocity limits
- Burner and coil configuration

Based on analysis of the coal slurry preheater operating requirements, a reasonable minimum tube size was selected to provide the scaleup and operability data. This tube size also corresponds to a coal feed rate of about 250 T/D.

ECLP Cost and Schedule

Based on the design specifications prepared for ECLP, a detailed estimate was prepared for the costs of detailed design, procurement and construction. For an April 1, 1976 contract award and September 1, 1978 mechanical completion, the estimated investment was 100 M$. The project, however, was delayed in order to secure financial support. The revised schedule projected mechanical completion in late 1979 at an estimated cost of 110 M$.

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