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PROJECT LIGNITE

FINAL TECHNICAL REPORT

March 28, 1972 - June 15, 1977

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I. SUMMARY

This final report describes work performed and results achieved under Project Lignite for the contract period from March 28, 1972, to June 15, 1977. It describes the development of the concept of the two-stage liquefaction of lignite, the first stage being the production of solvent refined lignite (SRL) by noncatalytic solvent hydrogenation, and the second the upgrading of the SRL by catalytic hydrogenation to a synthetic crude oil. The report covers the investigation of lignite liquefaction in a batch autoclave system, in a 50 lb/hr Process Development Unit (PDU) and additionally covers a series of economic studies of the technology developed.

The PDU was operated for thirty-two runs covering 1870 hours actually processing lignite under various conditions of pressure, temperature and throughput. A description of the unit is presented together with a typical material balance and yield structure.

The PDU was run at preheater outlet temperatures from 692° to 774°F, pressures from 1500 to 2500 psig, lignite feed rates from 14 to 61 lbs/hr, solvent to lignite ratios of 1.33 to 3.00, liquid hourly space velocities of 0.66 to 2.8 cu. ft. of slurry per hour per cu. ft. reactor volume. Conversions of over 90 percent were achieved under most conditions and possible solvent recovery indicated sufficient recovery for operation without addition of make-up solvent. Satisfactory solvent recovery was obtained at 2500 psig operating pressure. The longest run was for seven days including approximately 150 hours on stream charging lignite. The effect of process variables is reviewed with some correlations presented in equation form. Operational problems were associated with solids build-up in the reactor which caused plugging downstream and within the reactor as well as mechanical failure of pumps and control equipment. The solids-liquid separation system produced deashed solvent refined lignite, but did not operate satisfactorily for extended periods. The major corrosion problem was halide-induced stress cracking in the high pressure-high temperature sections noted during shake-down operation. These 316 stainless steel sections were replaced with Incoloy 800 which gave satisfactory service during the rest of the operations.

Batch autoclave experiments were done before and during operation of the continuous unit to develop design criteria, test process variables on a small scale, and otherwise serve a supporting role for the PDU. Parametric studies included

evaluation of start-up solvents, simulated solvent recycle, lignite storage, catalytic effect of cations, atmospheric composition, source of lignite, particle size, effects of carbonization and drying, pressure, direct hydrogenation, reaction time and particle size of lignite. Under a subcontract, conversion of the solvent refined lignite to light liquids by catalytic hydrogenation was investigated on the bench scale by faculty of the Department of Chemistry of the University of North Dakota.

Economic evaluations were made for conceptual commercial size plants using the discounted cash-flow (DCF) methods to arrive at a base unit cost. Sensitivity analysis of the unit cost was made for variations in the DCF rate, lignite cost, and labor costs. Liquefaction is a capital intensive process with the unit cost of SRL being strongly influenced by the DCF rate, and less strongly by the costs of lignite or labor. In all cases, the required selling price of the product was more than present prices for oil. For example, the investment required (mid-1975 prices) for a commercial SRL plant was $569,000,000 with total annual operating costs of $127,800. Unit cost of the SRL was from $2.33 to $3.14 per million Btu for a DCF rate from 12 to 20 percent.

II. INTRODUCTION

Production of liquid and environmentally acceptable fuels from our extensive coal reserves has received renewed interest because of the current energy situation in which over 50 percent of our petroleum requirements are imported. Direct liquefaction is attractive because of relative simplicity over prior gasification followed by synthesis, and it gives better yields than obtaining coal liquids by carbonization. Significant sulfur reduction is obtained and the mineral matter can be removed from the liquid phase to a low concentration. Solvent Refined Coal (SRC), which is solid at ambient conditions, or synthetic fuel oil equivalent could be used in steam electric stations without extensive clean-up of the combustion gases. Addition of more hydrogen to the SRC through catalytic hydrogenation would produce a liquid fuel having a H/C ratio suitable for use in the present mobile transport systems.

Of the several hydrogenation processes suggested for coal conversion, the use of a carbon monoxide atmosphere at high pressure and elevated temperature in the presence of a hydrogen-donor organic solvent has interesting possibilities for use with low rank coals. A synthesis gas mixture (H_2 plus CO) is also effective, giving high conversions without the expense of producing either pure hydrogen or pure carbon monoxide. However, most research activities in solvent refining have been done using bituminous coals and pure hydrogen.

At the University of North Dakota, research has been carried out over many years on the characteristics of lignite and the problems involved in its utilization. Initially, investigation of the liquefaction of lignite on a bench scale was done under sponsorship of the Great Northern Railway Company (Burlington Northern). Research activities were expanded in 1972 under a five year contract with the Office of Coal Research which subsequently was reorganized into the Energy Research and Development Administration followed by absorption into the Department of Energy. An extension of the five year contract by an additional year was negotiated. This report summarizes the activities and accomplishments during the initial contract, and is in three parts: (1) operation of the Process Development Unit (PDU); (2) investigations in batch autoclaves; and (3) cost studies evaluating process economics. Details have been reported previously in several publications including monthly, quarterly, and interim reports under contract as well as in the technical literature (1-26)*.

*Numbers in parentheses refer to items listed in the references section at the end of the report.

III. PROJECT SCOPE

The objective of the initial contract was to develop the necessary data for design of a lignite refinery which would produce high-quality solid, liquid, and gaseous fuels from Northern Great Plains Province Lignite. A five step experimental program was initially established to investigate process steps for possible integration into the operational scheme. These were:

1. Production of solvent-refined coal (SRC) from lignite.
2. Hydrogenation of SRC to produce liquids, and distillation to characterize the liquids.
3. Carbonization to produce char, liquids, and gases.
4. Direct hydrogenation of lignite to produce liquids, gases, and solid residues.
5. Drying of lignite as a separate process step.

The products obtained from such a complex could include pipeline gas, synthetic crude oil for refinery processing, solvent-refined lignite for solid or liquid fuels applications, and low-ash coke. In addition, raw materials for chemicals manufacture might be produced from the various fractions.

Development of details for complete design of the complex from Process Development Units was beyond the scope of the project. Consequently, most factors were investigated in the laboratory. Although a direct hydrogenation unit was designed and partially constructed, this phase of the investigation was dropped, and catalytic hydrogenation of SRL was done only by the Chemistry Department on the bench scale subcontract. Primary effort was directed toward operation of a 0.6 ton/day process development unit (PDU) designed for continuous solvent extraction of lignite at pressures up to 2500 psig and at temperatures up to 950°F in an atmosphere of reducing gases, usually synthesis gas, and with a hydrogen-donor solvent. Economic evaluations were made to determine the relative cost for production of the SRL.

IV. OPERATION OF THE PROCESS DEVELOPMENT UNIT (PDU)

A. INTRODUCTION

In early 1975, the Process Development Unit (PDU) was completed with the exception of the solid-liquid separation system. Some ten shake-down runs were completed in which the unit was operated with recycle of only solvent. The gas clean-up and recycle system and the solids separation system were not operated. Operational procedures were developed for correction of process upsets including plugging, leaky valves and instrument malfunction without total shutdown. The unit was nominally scheduled to operate for five-day weeks, each alternating with a week of shutdown for repairs, improvements, cleaning the system, interpretation of results and planning further runs. Most of the initial gross mechanical problems were corrected. Corrosion failures of 316 stainless steel culminated in shutdown of the plant after three preheater coils failed in operation and the dissolver heads developed cracks. A six month delay in the operation of the PDU followed, during which time the solid-liquid separation unit was completed and tested using unseparated SRC from the P&M pilot plant at Fort Lewis, Washington.

The plant was rebuilt with a new high pressure section of Incoloy 800 and put back into service in February, 1976. A schedule of operation during alternate weeks was then generally maintained through June, 1977. Reliability of system operation was improved, and most runs were carried through to the end of a five-day week, although some repairs and plug removals had to be done during the run. Some of the run conditions that were planned originally now appear to be inoperable, though improvements in equipment and techniques have resulted in higher yields, smoother operation, and better solvent recovery.

Through June of 1977, thirty-two runs were carried out with the reconstructed PDU and approximately 1870 hours of operation were accumulated in actually processing lignite to SRL. The longest run, M-29, was for 150 hours. Mechanical and operating problems with the solids-liquid separations unit resulted in limited successful operation. This section includes a description of the PDU, a summary of the effects of process variables, and a brief review of operating problems. Detailed yield data, test conditions and operations were included in the various Quarterly Technical Progress Reports (16), as well as a series of publications (1,4,5,9).

B. DESCRIPTION OF THE PDU

The continuous unit has a nominal design capacity of 50 lb/hr of lignite feed to produce approximately 15 lb/hr of solvent refined lignite (SRL). The melting point before deashing was usually in the temperature range of 300° to 400°F. Deashed SRL had lower melting points, sometimes as low as 200°F. Additionally, smaller quantities of lighter liquids and gases are produced.

Lignite with full "as-received" moisture content is pulverized and slurried with solvent. The slurry is pressurized, preheated and reacted at selected process conditions of temperature and pressure in a reducing gas atmosphere. Liquefaction products are separated as gases, liquids and SRL from the unconverted lignite and mineral matter. A simplified flow diagram is presented in Figure 1 while a brief description of the process by function areas follows:

1. Lignite Crushing and Pulverization

The feed lignite is shipped from the mine by rail or truck. Following initial size reduction with a jaw crusher, tramp iron is magnetically removed, and representative lots are obtained using a rotating sample splitter. The lots are then placed in 65 cu. ft. capacity storage bins and sealed in a nitrogen atmosphere until needed.

Individual bins are moved into the preparation building, hoisted by an electric crane and discharged through an iris valve into a secondary crusher. A second iris valve in the chute is used to help control dust emissions. A 70 cu. ft. surge bin below the crusher is used to correct for the disparity in capacity of the crusher and pulverizer. From the surge bin, the crushed lignite is fed into the pulverizer and discharged into 55 gallon drums prior to slurry preparation. Pulverization reduces the lignite to 100 percent passing 60 mesh and 90 percent through 200 mesh Tyler screens. Moisture content is reduced to about 30 percent during pulverization, although it is attempted to keep moisture loss to a minimum.

2. Slurry Preparation and Pumping

Included in this area are a pulverized lignite bin, a lignite feeder, a slurry mix tank, a slurry circulation pump and the high pressure reciprocating pumps.

The pulverized lignite is charged from the 55 gallon drums into the feed bin of an Acrison volumetric feeder which discharges into a Marion mixer at a constant flow rate. Weigh cells are used to measure and control the weight rate of feeding. Flow of solvent to the mixer is controlled through a flow-recorder-controller to give the desired lignite-solvent ratio. Discharge from the mixer is controlled by the

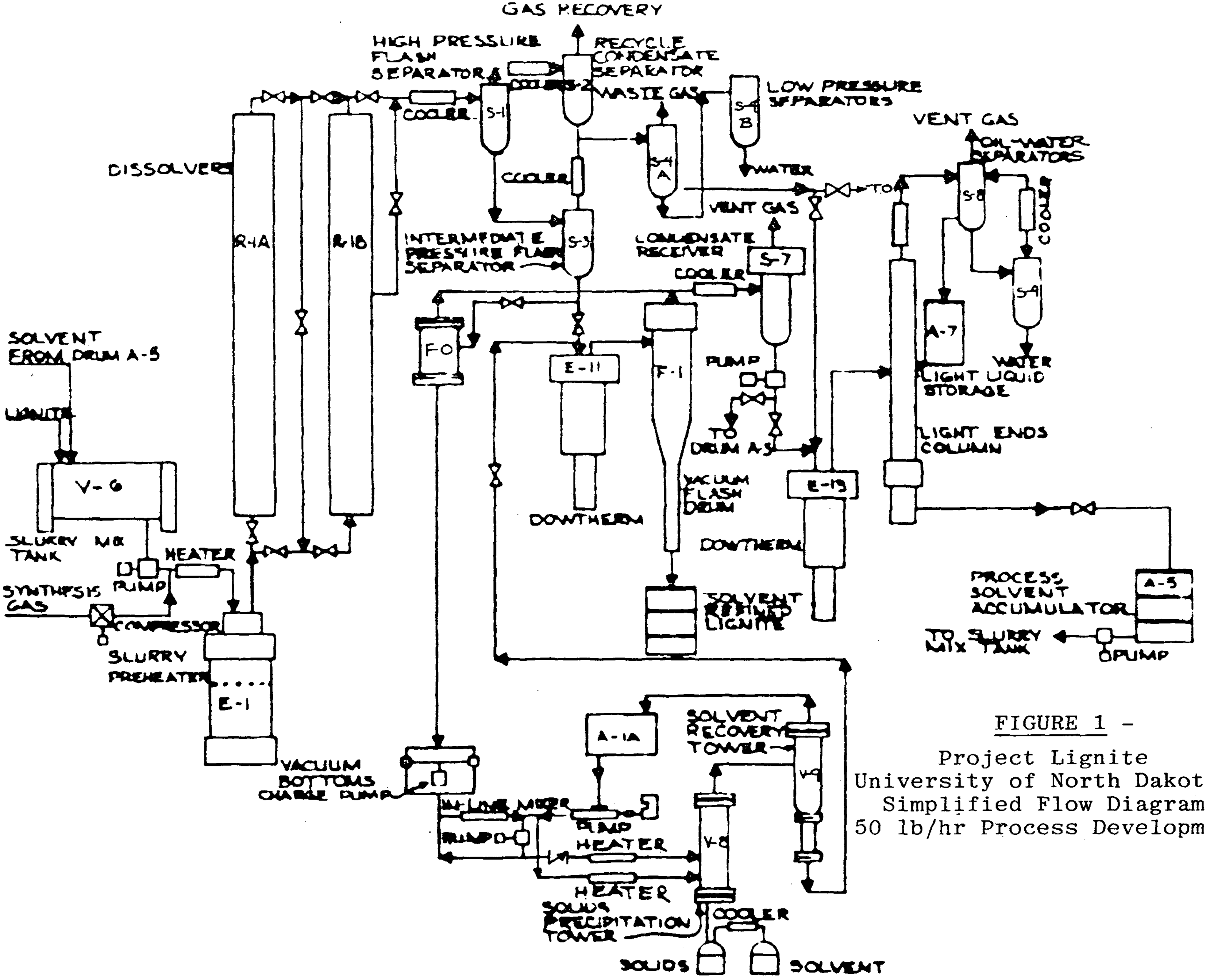


FIGURE 1 -

Project Lignite
University of North Dakota
Simplified Flow Diagram
50 lb/hr Process Development Unit

slurry level in the mixer tank. A jacket around the mixer allows water or steam to be circulated to control slurry temperature.

A lobe pump is used to circulate slurry around a mixer loop from which the slurry is fed to the suction side of the high pressure pumps at a pressure of about 50 psi. Sample connections in the circulating line allow sampling of slurry for density and viscosity measurements when needed.

3. Slurry Preheating, Dissolving and Gas Separation

A fluidized-solids slurry preheater, the dissolvers (reactors), and the high pressure vessels for separation of slurry, liquid and gaseous products are in this area. All process equipment and vessels were designed for operation at pressures to 2500 psig, and temperatures as high as 950°F. The preheater coil and dissolvers are constructed from Incoloy 800 and the high pressure separators are of cast 316 stainless steel.

The pressurized lignite-solvent slurry is combined with a gas nominally composed of cleaned recycle gaseous reactants combined with make-up hydrogen and carbon monoxide. Temperature of the slurry-gas mixture is raised to 300°F in a steam-heated exchanger and the stream then enters the coil in the fluidized-sand bath preheater from which it exits at about 750°. The sand in the bath is maintained approximately 25°F higher than the desired slurry-gas temperature or about 775°F. The mixture of lignite-solvent and gas flows through the 9/16-inch Incoloy 800 tube in the sand bath into one or both of the dissolvers-reactors, R-1A and R-1B. The dissolvers are constructed from 18-foot lengths of 4 7/8-inch OD by 3 7/8-inch ID Incoloy 800 tubing. Adiabatic conditions are approached in the reactors by using controllers and heating elements to maintain a zero temperature gradient between thermocouples in the insulation and at the walls of the reactors. Inlet to the dissolvers is at the bottom and the outlet at the top with an additional outlet near the center. This arrangement allows any multiples of 10-foot length to 40 feet to be used giving a four-fold variation in residence time of slurry without change in feed rate.

An air cooler is used to reduce the temperature of product from the reactor to about 500°F before the high pressure separator. The vapor from the separator is cooled to about 100°F in a water cooler and then enters the recycle condensate separator, S-2. The gas from this separator contains unreacted carbon monoxide and hydrogen, carbon dioxide, hydrogen sulfide, and the lighter hydrocarbons. After a pressure control valve, the gas enters the gas recovery and recompression area where it is cleaned and recycled. The pressure control valve regulates the pressure of the preheater-dissolver-high pressure separator system.

The liquid products from the recycle condensate separator consists chiefly of water, process solvent, and light hydrocarbon products. This liquid passes to the low pressure flash separator S-4A.

From the high pressure separator, the slurry products pass through a pressure release system to the intermediate pressure flash separator, S-3, which is at the same temperature as the high pressure separator. Light hydrocarbons and some solvent are vaporized and pass through a pressure regulator after which they are cooled to less than 100°F using water coolers. The cooled stream is combined with the liquids from the recycle condensate separator and both flow to the low pressure flash separator S-4A.

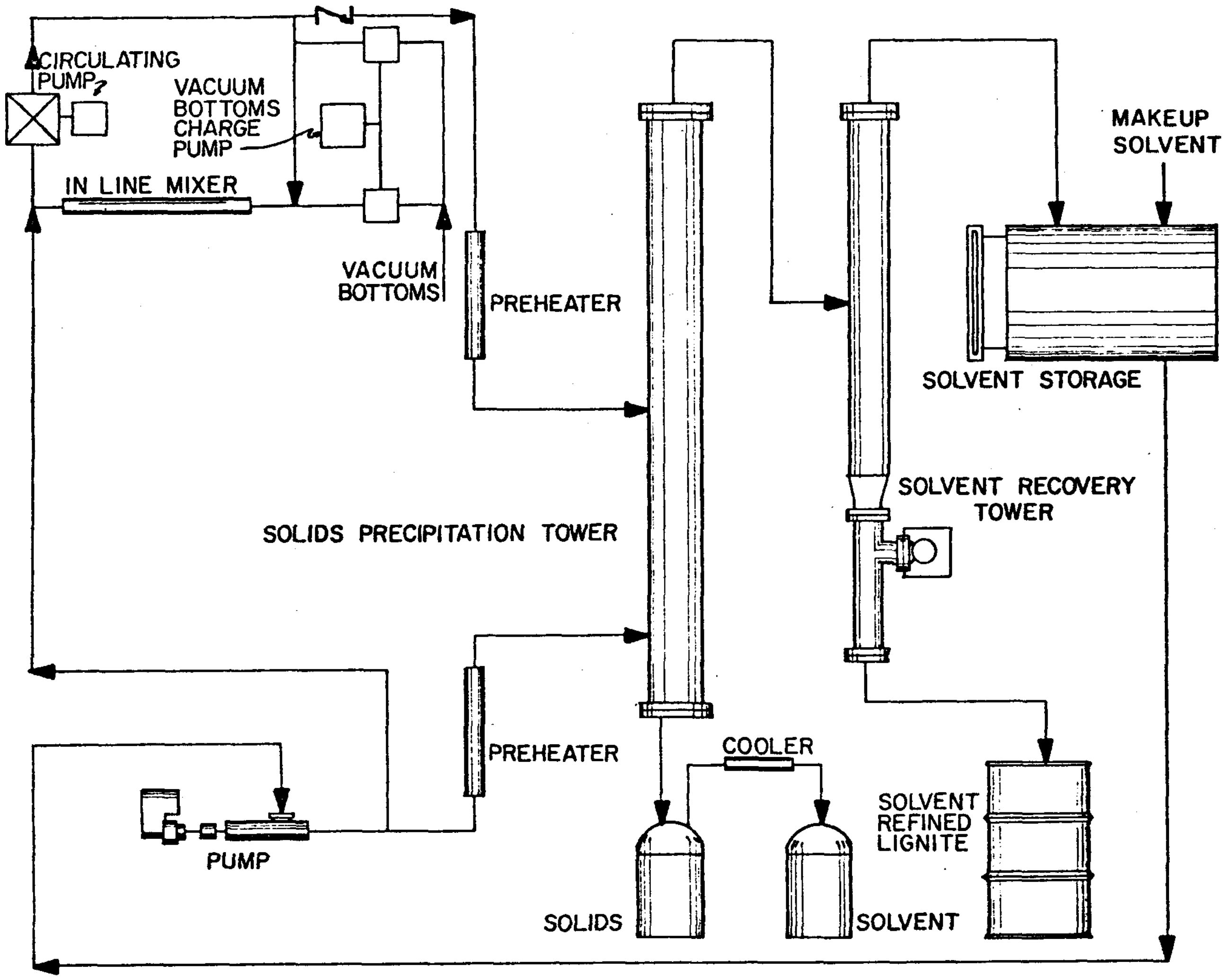
An aqueous phase is separated from the organic phase in the low pressure flash separators S-4A and S-4B with the organic phase transferred to the light-ends-column feed heater.

A second captive volume pressure letdown system discharges the slurry from the intermediate pressure flash separator S-3 to the partial flash separator F-0 with the discharge controlled by a displacement level controller located inside the vessel. Overhead from F-0 is combined with the overhead from vacuum flash drum F-1 and passed through a condenser to the vacuum condensate accumulator S-7. The bottoms from F-0 are sent to the solid-liquids separation system.

4. Solid-Liquid Separation

The separation system is designed for the high temperature extraction of solvent refined lignite plus solvent from mineral matter and unconverted lignite using toluene as a diluent. The flow diagram for this area is shown in Figure 2.

The bottoms stream from partial vacuum flash drum F-0 is pumped through an in-line mixer into a recirculating slurry loop together with the toluene deashing solvent. A toluene-slurry lobe pump discharges the slurry at pressures to 450 psig. The slurry is fed through an inline, cylindrical, 5 KVA heater where the process temperature is raised to between 350° and 400°F. The heated mixture is fed into the carbon steel settling tower which is 18 inches in diameter and 12 feet high. The cross sectional area is sufficient so that the terminal velocity of the settling particles is greater than the upward velocity of the toluene and SRL. Particles of one micron or larger size settle through the toluene solution moving upward at a velocity of 0.005 feet per second or less.



<u>FIGURE 2</u> - Solid-liquid separation unit diagram, 50 lb/hr process development unit.

At the base of the settling tower, a second stream of toluene enters at the temperature and pressure of the tower, flows upward, and promotes dissolution of any SRL which may be adhering to the ash and unconverted lignite. Mineral matter, unconverted lignite, and some toluene are withdrawn through a modified lock hopper operated intermittently by a timer. The toluene is flashed, condensed and collected. The ash and unconverted lignite are collected.

The liquid overhead from the settling tower, consisting of dissolved SRL, liquefaction solvent, and toluene, is depressurized, cooled in steam cooler E-6, and fed to the toluene recovery tower, V-9, which is 6 inches in diameter by 10 feet high and operates at 300°F or above and 5 psig. The toluene flashes, leaving the SRL-solvent mixture behind. The bottom stream is fed to preheater E-11 for vacuum flash separation of SRL and liquefaction solvent.

The overhead stream of toluene is condensed, stored in accumulator A-1, and recycled using a triple diaphragm pump, P-6. The toluene stream, at a pressure up to 450 psig, is split into two streams. One provides dilution of the vacuum bottoms and the other feeds through a line preheater E-9 to the wash toluene inlet on settling tower V-8.

5. Liquid Separation and Solvent Recycle

Two major distillation sections in this area are the vacuum flash section for separating process solvent from the liquefied product and the distillation column for separating light oil from recycle liquefaction solvent. The Dowtherm preheaters, storage tanks for all products, and solvent recycle facilities are also included in this area.

The slurry from the intermediate pressure flash separator S-3 at about 550°C goes into flash drum F-0. The bottoms stream is sent to the solid-liquid separations system. The return stream from the solid-liquid separations system, i.e. the bottoms from the toluene flash vessel V-9, is fed through preheater E-11 and into the vacuum flash drum F-1. The bottoms from F-1 is the SRL product. The overhead from F-1 is combined with the F-0 overhead, condensed, and sent to drum A-5A as part of the recycle solvent. The light oil from S-4A is charged to light ends column F-2. This column is operated at a pressure of 10 to 15 psig. The overhead product is a light oil, essentially a stabilized naphtha, consisting of hydrocarbons ranging from about C_5 to perhaps as high as 350-450°F in boiling point. The bottom product is also recycled through drum A-5A to the slurry mix tank.

6. Gas Purification and Recycling

The vapor products from the recycle condensate separator S-2, at a temperature of about 100°F, flow through a pressure control valve, in which the pressure is reduced to about 100 psig, to an absorption tower in which carbon dioxide and hydrogen sulfide are removed using a 10 percent by weight solution of sodium hydroxide. The upper two-thirds of the tower is packed with 1-inch polypropylene Pall rings on which sodium hydroxide solution is sprayed; the gas percolates upward. The sodium hydroxide from the bottom of the tower flows back to the storage tank through a level control valve, operating on the level of sodium hydroxide solution in the tower. The sodium hydroxide is circulated until it is spent, and then a fresh solution is charged from the other storage tank.

The gas stream from the acid gas absorption tower goes to the ammonia absorption tower, which uses a boric acid solution as the absorbing medium. The gas from the ammonia tower consists of carbon monoxide and hydrogen, plus light hydrocarbons and inerts. Part of the gas flows to a compressor surge drum through a positive displacement meter while a bleed stream flows to the thermal oxidizer, through a flow control valve, to limit the build-up of hydrocarbon gases and inert gases.

Two identical diaphragm compressors are provided, either of which has sufficient capacity for the requirements of the unit.

The feed hydrogen and carbon monoxide are stored in high-pressure tube trailers. The ratio of hydrogen to carbon monoxide in the feed is controlled by a ratio recorder controller. The hydrogen-carbon monoxide gas mixture is combined with the recycle gas just ahead of the suction of the compressor, with the flow rate controlled to maintain the desired suction pressure. After compression the gas flows through a flow controller to the slurry just ahead of the fluidized solids preheater E-1.

C. RESULTS FROM OPERATION OF THE PDU

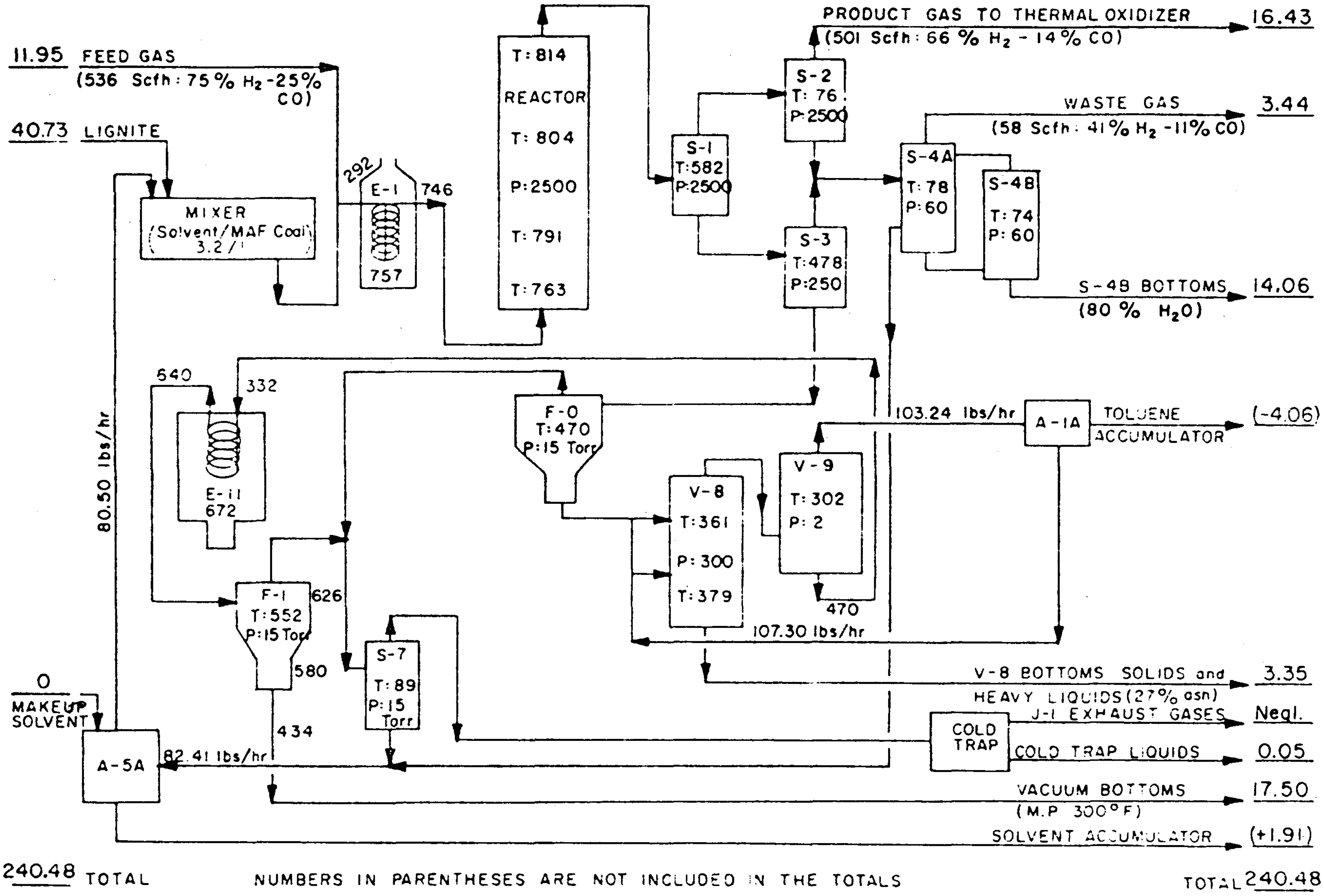
1. Typical Operations and Conditions

During the usual continuous five-day run, data were gathered for a series of twelve-hour yield periods for material balance purposes. In the flow diagram in Figure 3, mass flows and run conditions are given for a typical run, Run M-17AB, representing a composite for two successive yield periods. Pressure and temperatures are also given to indicate run conditions in important vessels. In this run, 41 lb/hr of 31.5 percent moisture lignite was processed using 536 SCFH of gas containing 75 percent H_2 and 25 percent CO, 81 lb/hr of recycle solvent, and 107 lb/hr of recycle toluene in the liquids-solids separation system.

FIGURE 3: MASS RATES AND RUN CONDITIONS

	START	END
Date of Run	9/21/76	9/22/76
Time of Run	1530	1530
Length of Run	24 hrs	

Run No: M-17AB	
Units: lbs/hr, °F, psig	
LHSV: 1.30	GHSV: 375
Revised 5/9/77	



Feed and product compositions and characteristics are shown in Table 1, and the yield structure is shown as a bar graph in Figure 4. With a basis of 100 pounds of MAF lignite fed, 65.8 pounds of SRL and light organic liquids were produced, and 15.6 pounds of CO, 1.0 pounds of H_2 and 5.4 pounds of water were consumed. Conversion was a little more than 90 percent of the MAF coal.

The PDU has been operated at pressures from 1500 psi to 2500 psi and with preheater exit temperatures of 692°F to 774°F, resulting in dissolver exit temperatures from 683°F to 916°F. The lignite feed rate has been varied from 14 lb/hr up to 61 lb/hr and the weight ratio of process solvent to lignite from 1.33 to 3.00. The space velocity of the slurry and gas can be varied independently of feed rate because the two dissolvers can be used in series, one dissolver can be used with the other by-passed, or half a dissolver can be used. Thus the liquid hourly space velocity has been varied from 0.66 to 2.8 cubic feet of slurry per hour per cubic foot of reactor volume, and the specific coal rate from 14 to 70 lb/hr per cubic foot of reactor volume. However, complete yield periods were not realized at some values of the variables.

Operability of the unit was achieved, but the range of conditions under which sustained operation can be maintained is somewhat limited. "Operability" is used in the sense of being able to keep the plant going, even though solvent recovery may not be adequate.

The unit was operated successfully in runs at pressures of 1500, 2000 and 2500 psi. Temperatures over 900°F in the dissolver result in excessive coking and operational problems. It is necessary to operate at a preheater outlet temperature such that the dissolver temperatures will be less than 900°F, and other factors make even lower temperatures seem desirable. Under most conditions a 750°F preheater outlet temperature is satisfactory, but in some cases it was necessary to reduce it to as low as 690°F. Solvent to coal ratios of 1.4 to 3.00 were successfully used. A solvent to coal ratio of 1.33 resulted in continuing mixing and pumping troubles and in the production of high melting point vacuum bottoms that forced a shutdown. The original vertical slurry mix tank and centrifugal slurry pump would not operate satisfactorily below about a 1.5 ratio, but the Marion mixer and lobe slurry pump have worked well down to a 1.4 ratio.

The melting point of the vacuum bottoms was a good indicator of impending process difficulty during most of the runs. When the melting point (i.e. gradient bar softening temperature) reached 335°F for undeashed vacuum bottoms eventual shutdowns occurred because of inability of the Moyno vacuum bottoms pump to remove the material. However,

TABLE 1: FEED AND PRODUCTS - RUN M-17AB

FEED MATERIALS

Lignite
North American Coal Co., Zap, ND
Screen size 90%-200 mesh, 100%-60 mesh
Moisture: 31.5%

Feed Gas (mixed from tank trailers)

	Vol. %	Wt. %
CO	24.7	81.9
H_2	75.2	17.8

Startup liquefaction solvent
Gulf Oil Co. FS-120 Carbon Black Feed Stock
Boiling range (1.6 Torr)

	FS-120 Startup	Recycle Solvent
IBP-100°C	4.2	34.9
100-230°C	81.0	55.9
230-255°C	14.8	9.6
Specific Gravity	1.046	0.980

MATERIAL BALANCE FOR GAS COMPONENTS

	Vol % In	Vol % Out	lb/hr In	lb/hr Out
H_2	75.2	66.2	2.13	1.87
CO	24.7	14.4	9.78	5.81
CO_2	-	13.2	-	9.91
H_2S	-	0.2	-	0.18
CH_4	-	4.5	-	1.11
C_2H_6	-	1.1	-	0.58
C_3H_8	-	0.3	-	0.33
NH_3	-	0.1	-	0.04

ULTIMATE ANALYSIS OF MATERIALS

	Lignite Charged	Starting Solvent	Recycle Solvent	Vacuum Bottoms	Deashed SRL
C	45.22	89.03	83.09	80.20	87.42
H	6.43	8.11	9.14	5.20	5.67
N	0.64	0.12	0.20	0.98	1.07
S	0.45	2.23	1.09	0.90	0.98
O*	41.45	0.51	5.97	4.46	4.86
Ash	5.81	0	0	8.26	0

*By difference

PROPERTIES OF PRODUCT SRL

	F-1 Vacuum Bottoms (measured)	SRL (calculated)
Gradient Bar Melting Point °F	327°	-
Pyridine Solubles, Wt % ash-free	83.5	100
Specific Gravity	1.28	1.25
Heat of Combustion, Btu/lb	14,330	15,990

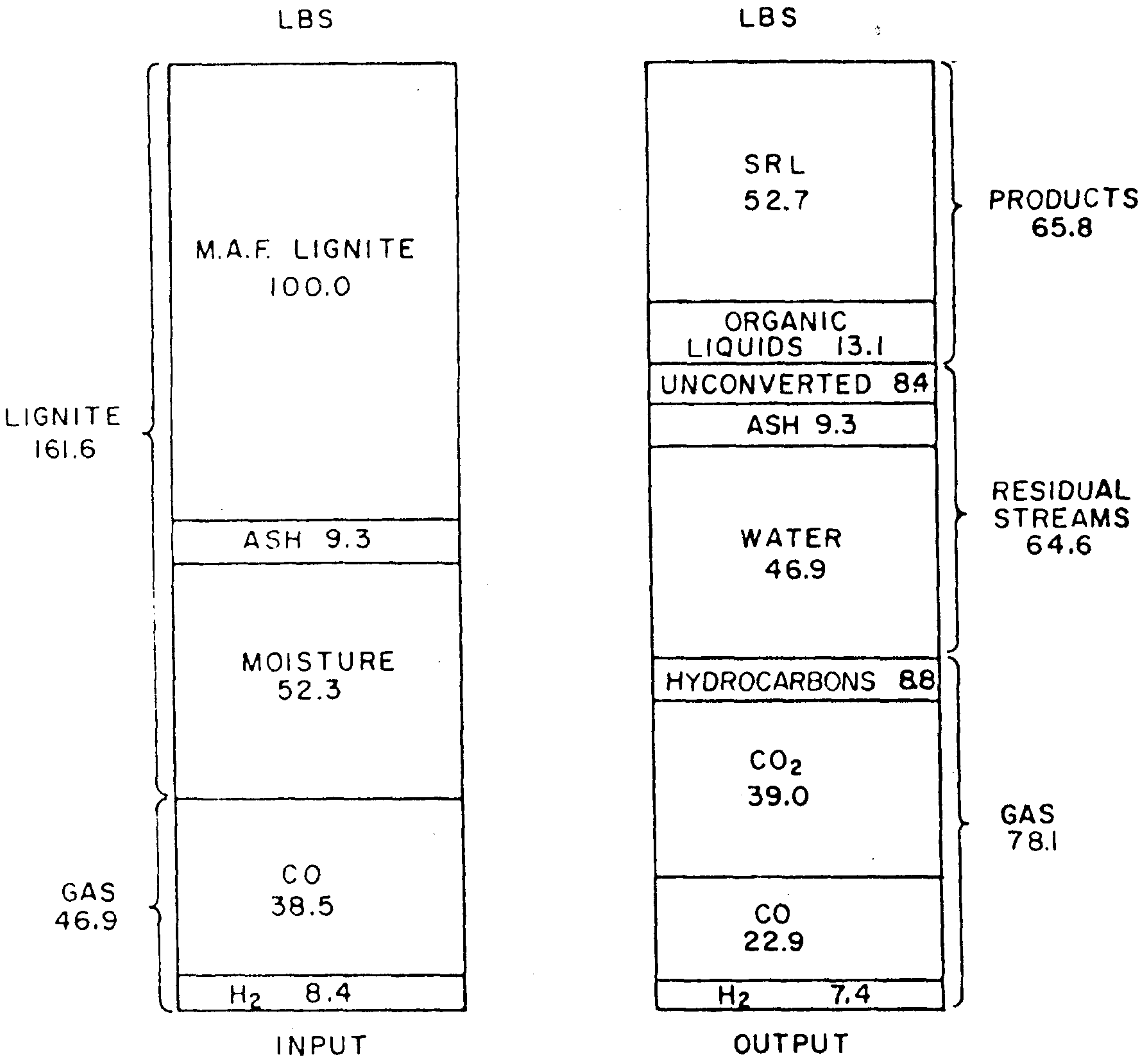


FIGURE 4: NET MATERIAL BALANCE, RUN M-17AB

BASIS: 100 LB. M.A.F. LIGNITE

replacement of the Moyno pump with a lock hopper system through which vacuum bottoms could be drawn off periodically under pressure improved this operation.

The vacuum flash has been operated successfully at temperatures in the range of 500° to 600°F. Above 600°F, because of the removal of too much distillate, the melting point of the bottoms may become too high for successful draw-off and coke may form in the drum. Too low a temperature in the lower section of the draw-off leg results in excessively high viscosity of the bottoms and difficulty in bottoms draw-off.

Liquid hourly space velocities of 0.66 to 1.6 hr^{-1} were used successfully, but an attempt to raise the LHSV to 2.8 hr^{-1} resulted in plugging because of low conversion and consequent high solids downstream. The gas rates have not been varied appreciably. Major space velocity variations are caused by varying the dissolver volume. The highest GHSV accompanied the 2.8 hr^{-1} LHSV, which was an inoperable condition.

Operation was successful at coal rates up to 38 pounds lignite per hour per cubic foot of reactor volume, but was unsuccessful at 70 lb/hr-cubic foot.

2. Solvent Balance

A critical question concerning the success of the SRL process is whether it will generate enough process solvent to maintain itself. On the PDU scale, enough solvent must be recovered to continue recycle for extended periods to provide process data that truly reflects performance of solvent generated from the lignite. The first runs showed about 85 percent solvent recovered for recycle, and consequently the runs originally programmed were altered to seek conditions under which solvent balance would be achieved. In later operation the solvent balance was gradually improved and in some runs better than 100 percent solvent recovery was actually realized when organic distillates that could be condensed or vacuum distilled were recovered. Thus solvent included all material boiling below 850°F.

In longer continuous runs in which all recoverable organic liquids were recycled, lignite conversion decreased as the runs progressed, apparently because the increasing amount of lighter materials in the solvent reduced the solvent effectiveness. Hence in later runs the organic liquids boiling below about 400°F were removed by distillation from the recycle solvent, and the solvent retained its effectiveness. Satisfactory solvent recoveries have only been obtained at 2500 psig dissolver pressure.

3. Results of Variable Study

The following is a discussion of the effects of the principal operating variables on process results. Summary data for the runs upon which this discussion is based are presented in Table 2. The runs were selected, with certain exceptions, based upon the following criteria:

1. Runs made subsequent to Run M-11, when operating techniques and product workup procedures had been developed to satisfactory levels.
2. Runs in which the solid-liquid separation system was not in use, because solids hold-up in the precipitation tower made it difficult to measure product distribution. Two exceptions were made for runs illustrating certain conditions which were not covered by runs without the SLS system.
3. Runs having overall material balances (product recovered) between 98 and 101 percent. Here again a few exceptions were made to illustrate certain conditions.
4. All runs meeting the above criteria conducted at 2500 psig with approximately 1/1 CO/H_2 ratio in the charge gas.
5. Certain other selected runs at lower pressures (1500 or 2000 psig) and selected runs with 3/1 or 1/3 CO/H_2 ratio in the charge gas.

a. Temperature

The yields of the major products (totaling 100 percent of the MAF coal) are plotted against temperature in Figure 5 for the runs at 2500 psig and 1/1 H_2/CO. Unconverted coal is defined as the pyridine-insoluble ash-free material remaining after liquefaction. The net product (oil and solvent refined lignite or SRL) is all of the distillable and/or pyridine soluble material minus the solvent charged. The gas yield is the weight of dry gas out minus the weight of gas charged. The water yield, which is generally negative since water is usually consumed, is the water recovered minus the moisture charged with the lignite. Any change in ash, which is usually small and may be either positive or negative, is included with the water so that the four major product yields will total 100 percent of the MAF coal. The temperature plotted is the maximum measured dissolver temperature, usually that recorded 15 feet up from the bottom of the 18-foot dissolver, but sometimes the dissolver outlet. The strong reaction exotherm in the dissolver complicates the interpretation of temperature as a variable. Initially the preheater outlet temperature was held as closely as possible

TABLE 2: SUMMARY DATA OF PDU OPERATIONS

Yield Period	M-16A(1)	M-17B(2)	M-21D(2)	M-23A	M-27D	M-27E	M-27F	M-28D(1)
Symbol								
Hrs on Coal at end of YP								
Since Reactor Cleaning	19	41	76	27	48	60	72	34
During Run	19	41	76	27	52	64	76	69
Cumulative	914	962	1123	1199	1409	1421	1433	1502
Reactor Used	B	A	B	B	A	A	A	B
Coal Charged, lbs/hr	48	37	48	49	53	51	47	48
Gas Charged, SCF/Hr	435	537	612	554	475	485	483	489
Solvent/Coal Ratio	1.77	2.17	1.87	1.83	1.83	1.82	2.12	2.09
LHSV, Hr^{-1}	1.43	1.27	1.40	1.41	1.53	1.52	1.50	1.52
GHSV, Hr^{-1}	305	376	429	387	332	339	338	342
Coal Ratio, lbs/Hr/CF	33.9	25.7	33.5	34.1	37.1	35.9	32.9	33.4
Gas Rate, SCF/lb Coal	9.0	14.5	12.8	11.4	8.9	9.4	10.3	10.2
Mol % H_2 in Gas Charged	24.6	74.2	71.6	71.5	48.8	51.2	51.0	57.6
Gas Consumed, SCF/lb Coal	3.5	2.6	3.7	1.6	5.0	3.4	4.3	5.2
Mol % H_2 in Gas Consumed	-11.8	37.8	60.7	37.1	31.7	31.5	33.7	40.5
Yields, Wt % of MAF Coal								
Light Oil (540°F-)	15.3	24.7	8.3	10.0	9.9	7.5	14.5	22.1
Solvent (540°F+)	-12.9	-11.4	-11.3	-33.1	-6.9	-7.6	-5.2	-22.7
Distillable Oil	2.4	13.3	-3.0	-23.1	3.0	-0.1	9.3	-0.6
SRL	53.1	50.4	60.4	57.9	50.3	59.0	52.1	60.6
Total Product	55.5	63.7	57.4	34.8	53.3	58.9	61.4	60.0
H_2O and Ash	-10.2	-5.6	-1.3	-0.2	-7.5	-6.4	-2.7	-10.6
Unconverted	15.5	6.2	13.2	37.9	15.9	17.0	9.2	20.9
H_2O and CO	-44.8	-20.1	-18.6	-12.2	-39.1	-28.1	-34.5	-38.4
C_1-C_3 HC's	15.7	10.6	6.7	8.6	16.2	7.6	9.3	10.0
CO_2	67.8	43.8	41.8	30.7	60.4	51.4	56.6	57.0
Net Gas	39.2	35.7	30.7	27.5	38.3	30.5	32.1	29.7
Solvent Recycle								
Actual	98.8	100.6	67.2	86.7	79.2	84.1	87.0	93.4
Potential-Total	100.9	103.8	99.0	92.0	101.1	99.9	102.8	99.8
Potential-540°F+	94.6	94.6	95.4	86.4	97.1	93.7	98.3	92.9
Material Balance, %	96.7	97.9	101.0	99.6	100.1	98.9	99.9	96.2
Conditions								
Temperature, °F								
Preheater Outlet	744	745	746	764	707	699	717	763
Reactor Inlet	-	-	746	741	706	698	717	734
Reactor - 3 ft	784	774	-	791	742	714	727	771
Reactor - 15 ft	858	826	-	832	843	817	824	809
Reactor Outlet	849	-	828	831	-	-	-	-
Pressures, psig								
Reactor	2500	2500	2000	1500	2500	2500	2500	2500
H_2 Equiv Gas Consumed, Wt % of MAF Coal	2.85	2.21	3.04	1.33	3.96	2.83	3.58	4.40

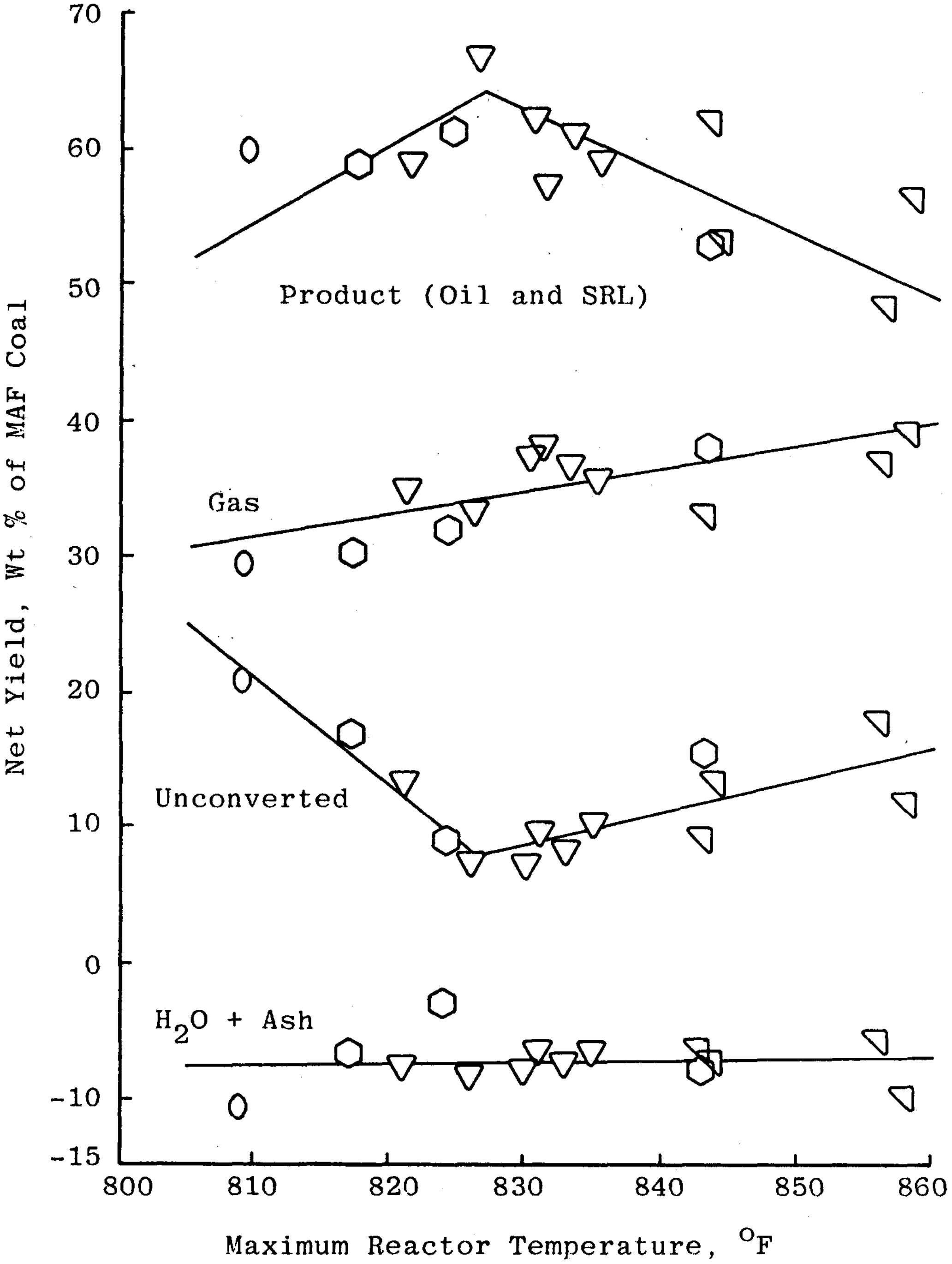
(1) Used in spite of slightly low material balance to illustrate effect of low hydrogen in charge gas (M-16A) and effect of lower temperature (M-28D).

(2) Solid-Liquid Separation Unit in operation during run.

TABLE 2 (CONT) SUMMARY DATA OF PDU OPERATIONS

Yield Period	M-29B	M-29C	M-29D	M-29F	M-29G	M-29H	M-30A	M-30B	M-30C	M-30D
Symbol										
Hrs on Coal at end of YP										
Since Reactor Cleaning	53	65	77	26	38	50	36	48	64	76
During Run	53	65	77	119	131	143	36	48	64	76
Cumulative	1555	1567	1579	1621	1633	1645	1688	1700	1716	1728
Reactor Used	B	B	B	A	A	A	B	B	B	B
Coal Charged, lbs/hr	46	48	46	47	45	45	50	49	58	56
Gas Charged, SCF/hr	386	388	390	384	383	393	389	387	384	388
Solvent/Coal Ratio	1.95	1.93	2.00	1.95	2.03	2.03	1.86	1.86	1.47	1.53
LHSV, Hr^{-1}	1.39	1.43	1.40	1.42	1.40	1.40	1.45	1.42	1.46	1.45
GHSV, Hr^{-1}	270	271	273	269	268	275	272	270	268	271
Coal Rate, lbs/hr/CF	32.3	33.4	32.0	32.8	31.4	31.6	34.7	34.1	40.7	39.4
Gas Rate, SCF/lb Coal	8.4	8.1	8.5	8.2	8.5	8.7	7.8	7.9	6.6	6.9
Mol % H_2 in Gas Charged	49.0	49.3	48.8	50.0	49.6	50.0	58.2	54.3	54.6	55.2
Gas Consumed, SCF/lb Coal	3.2	2.9	2.2	1.8	2.9	2.9	3.1	3.2	2.8	2.6
Mol % H_2 in Gas Consumed	25.9	24.4	9.4	0	18.6	23.5	41.4	31.5	31.2	32.1
Yields Wt % of MAF Coal										
Light Oil (540°F-)	12.9	21.0	1.6	2.6	24.9	23.7	27.5	29.1	21.1	14.6
Solvent (540°F+)	3.5	0.1	14.9	8.2	-14.3	-7.0	-16.7	-11.3	-11.6	1.6
Distillable Oil	16.4	21.1	16.5	10.8	10.6	16.7	10.8	17.8	9.5	16.2
SRL	44.9	46.0	45.9	48.3	46.9	42.7	43.0	39.2	39.5	46.3
Total Product	61.3	67.1	62.4	59.1	57.5	59.4	53.8	57.0	49.0	62.5
H_2O and Ash	-7.0	-8.2	-7.6	-7.6	-6.1	-6.1	-6.7	-9.2	-5.1	-5.8
Unconverted	8.7	7.5	7.5	13.4	10.0	10.7	13.8	12.3	18.5	9.8
H_2 and CO	-27.7	-25.7	-23.7	-21.4	-28.2	-25.9	-22.5	26.7	-23.8	-21.3
C_1-C_3 HC's	10.6	8.4	6.6	6.6	10.3	8.3	13.7	15.1	12.0	8.2
CO_2	53.4	49.4	52.8	48.6	54.2	52.1	46.8	50.4	48.6	45.7
Net Gas	37.0	33.6	37.7	35.1	38.6	36.0	39.1	39.9	37.6	33.5
Solvent Recycle										
Actual	93.3	97.9	100.6	96.0	94.5	95.3	92.4	89.3	92.3	94.3
Potential-Total	105.4	107.1	105.1	103.5	103.3	105.2	103.7	106.1	104.0	106.8
Potential-540°F+	101.3	100.0	105.4	103.2	95.0	97.4	93.8	95.7	94.4	100.8
Material Balance, %	99.1	100.8	98.4	98.6	100.6	100.8	101.0	98.4	99.1	99.5
Conditions										
Tempertures, °F										
Preheater Outlet	714	707	713	719	714	710	744	740	719	712
Reactor Inlet	704	699	706	704	707	705	725	723	703	697
Reactor - 3 ft	770	751	758	773	776	772	783	785	772	759
Reactor - 15 ft	833	826	830	821	831	835	844	858	856	843
Reactor Outlet	823	807	810	787	814	804	821	831	832	816
Pressures, psig										
Reactor	2420	2400	2420	2450	2450	2450	2400	2400	2400	2400
H_2 Equiv Gas Consumed, Wt % of MAF Coal	2.60	2.38	1.86	1.53	2.44	2.37	2.61	2.69	2.39	2.17

FIGURE 5 - Yields of Major Products vs. Maximum Reactor Temperature



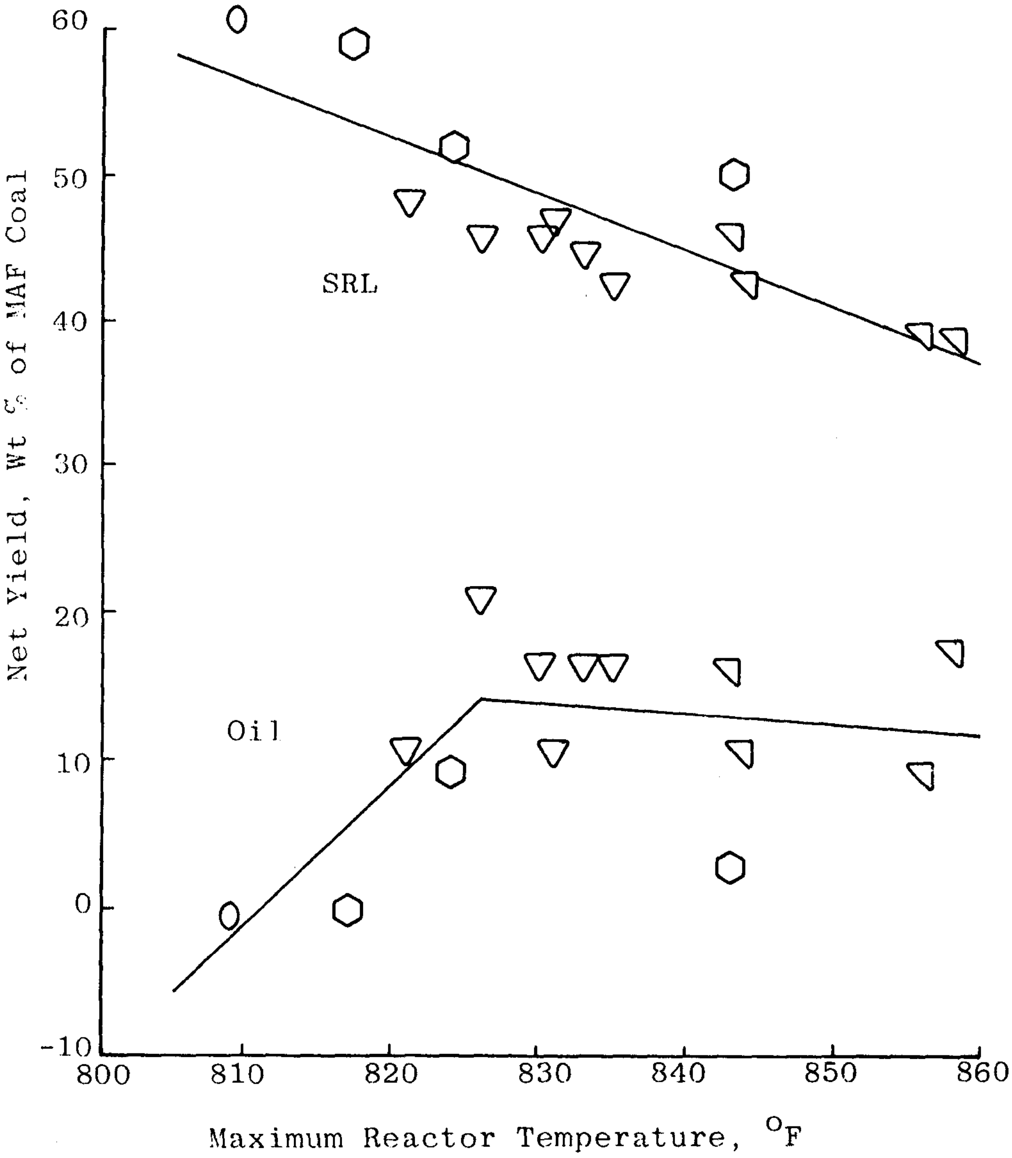
to 750°F. However, the large temperature rise through the dissolver led in some cases to maximum temperatures of about 900°F, so the preheater outlet temperature was lowered to give lower dissolver temperatures. The array of reactor temperatures shown in the plots reflects about a 65°F spread in preheater outlet temperatures. The temperature effects are further confounded with the effects of charge rates of coal, solvent, and gas and at gas composition, but these variables changed only slightly and thus Figure 5 reflects predominantly the effect of temperature.

The water consumption seems to decrease very slightly with increasing temperature, while the gas yield seems to increase regularly. The lines shown for these two products in Figure 5 were determined by linear regression of the net yield versus temperature. The correlation coefficient for the gas yield (r) is 0.767 as compared with the critical value at the 5% level of 0.532. Since the water consumption is essentially constant, the correlation coefficient has little meaning.

The oil and SRL yield appears to go through a maximum while the unconverted coal appears to go through a minimum between 826 and 830°F. Using the data for unconverted coal in runs at 826°F and below for one linear regression and the data at 830°F and above for another, two regression lines of opposite slope were established. The correlation coefficients (r) for these two regression lines were 0.977 and 0.730 for the lower and higher temperature ranges respectively, as compared with the critical values at the 5% level of 0.878 and 0.666. These two lines intersect at 826.3°F and 8.08 weight percent unconverted coal. Since the regressions for gas and water were done on different sets of data than the regressions for the net products, a consistent relation for net products yield versus temperature could not be determined by regression, but was determined by difference between the sum of the other three yields and 100 percent. The significance of the apparent maximum in conversion (minimum in unconverted coal or pyridine insoluble organic matter) is that conversion increases with increasing temperature, while solvent polymerization or condensation with the coal matter or coking also increase with temperature until the rate of formation of pyridine insoluble material exceeds the rate of conversion of the coal.

In Figure 6, the yield of products is divided into the two main products, oil and SRL, again plotted against maximum reactor temperature. The SRL yield seems to follow a general pattern of decreasing with increasing temperature, and the line determined by regression had a correlation coefficient of 0.831 as compared with the critical value at the 5% level of 0.532. Here again the line for the oil yield was determined by difference between the SRL line and the product line from Figure 5.

FIGURE 6 - Yield of Oil and Solvent Refined Lignite vs. Maximum Reactor Temperature



In Figure 7, the oil yield is further divided into light oil, boiling below about 540°F, and the solvent fraction boiling between 540°F and SRL. The correlation coefficient (r) for the regression line for the light oil was 0.577 as compared with the critical value at the 5% level of 0.553. Again, the solvent yield was determined by difference, and there resulted a maximum of 1.6 weight percent solvent at 826.3°F, with a breakeven range for the solvent between 823°F and 830°F. However, there are solvent yields higher than these, so that other factors, either operating variables or variations in product work-ups and analyses, come into play in affecting the apparent solvent yield. However, breakeven solvent operation seems practical over only a fairly small temperature range.

Yields of carbon dioxide and of the lighter C1 to C3 hydrocarbons are shown in Figure 8. Carbon dioxide appears to decrease with increasing temperature while the C1 to C3 hydrocarbons increase with temperature. The lines determined by regression had correlation coefficients (r) of only 0.333 for carbon dioxide and of 0.644 for hydrocarbons as compared with the critical value of 0.532 at the 5% level.

b. Space Velocity and Solvent/Coal Ratio

The yield data for the major products for all of the runs used in the preceding temperature study were adjusted to a common temperature of 826.3°F by applying the slopes of the yield-temperature relations to each of the actual yields for each run. The adjusted yields are plotted against superficial liquid hourly space velocity (LHSV) in Figure 9. A multiple regression study of yields versus LHSV and solvent to coal ratio (S/C) resulted in the lines shown in the figure. Over the ranges of the variables shown, which are slightly greater than those actually experienced during the runs themselves, there are relatively small effects. The greatest differences are in the gas yields, where increasing the space velocity from 1.36 to 1.56 resulted in a 4.2 percent decrease in gas yield at constant solvent/coal ratio, while all other yields increased slightly. Increasing the solvent/coal ratio from 1.30 to 2.30 resulted in a 4.7 increase in gas yield at constant space velocity with slight decreases in other yields. This latter effect is thought to be the result of relatively greater gas production from solvent than from coal when more solvent is present. The changes in all yields are summarized in the following:

	Change in Yields, Wt% of MAF Coal	
	LHSV, 1.36→1.56	S/C, 1.30→2.30
Net Product	+0.8	-2.4
Net Gas	-4.2	+4.7
Net Water	+0.4	-0.7
Unconverted Coal	+3.0	-1.6

FIGURE 7 - Yields of Light Oil and Solvent vs. Maximum Reactor Temperature

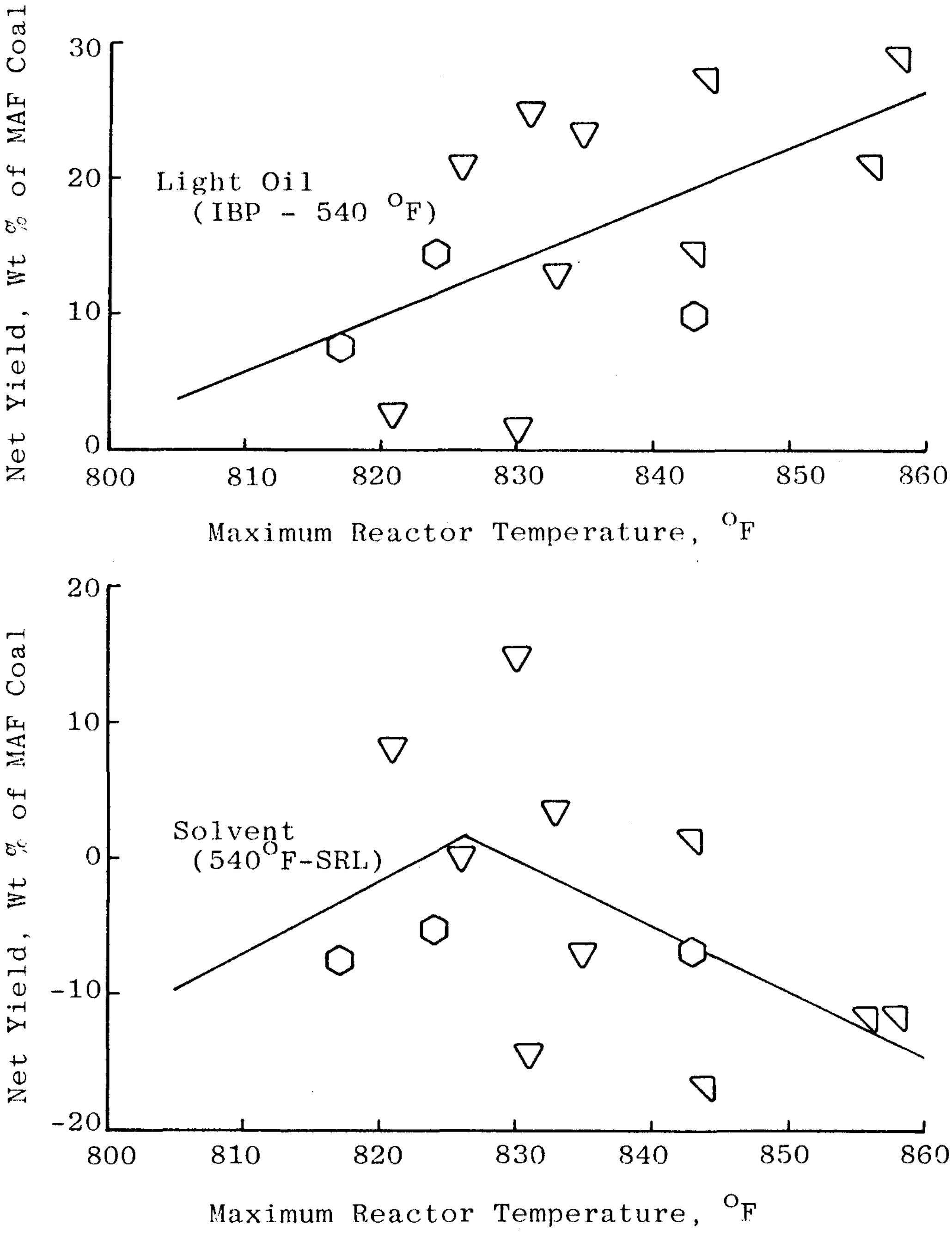


FIGURE 8 - Yields of Carbon Dioxide and Light Hydrocarbons vs. Maximum Reactor Temperature

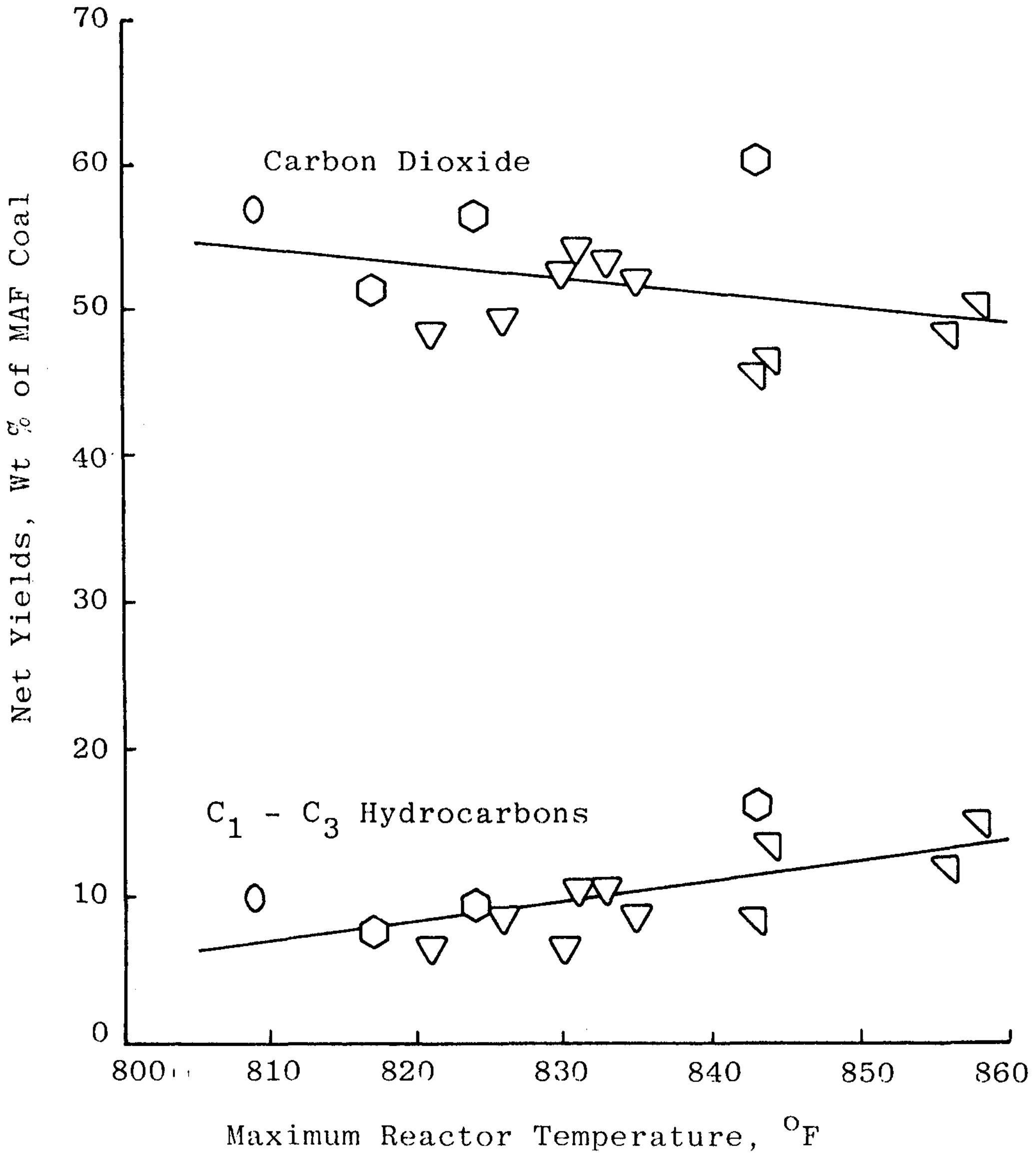
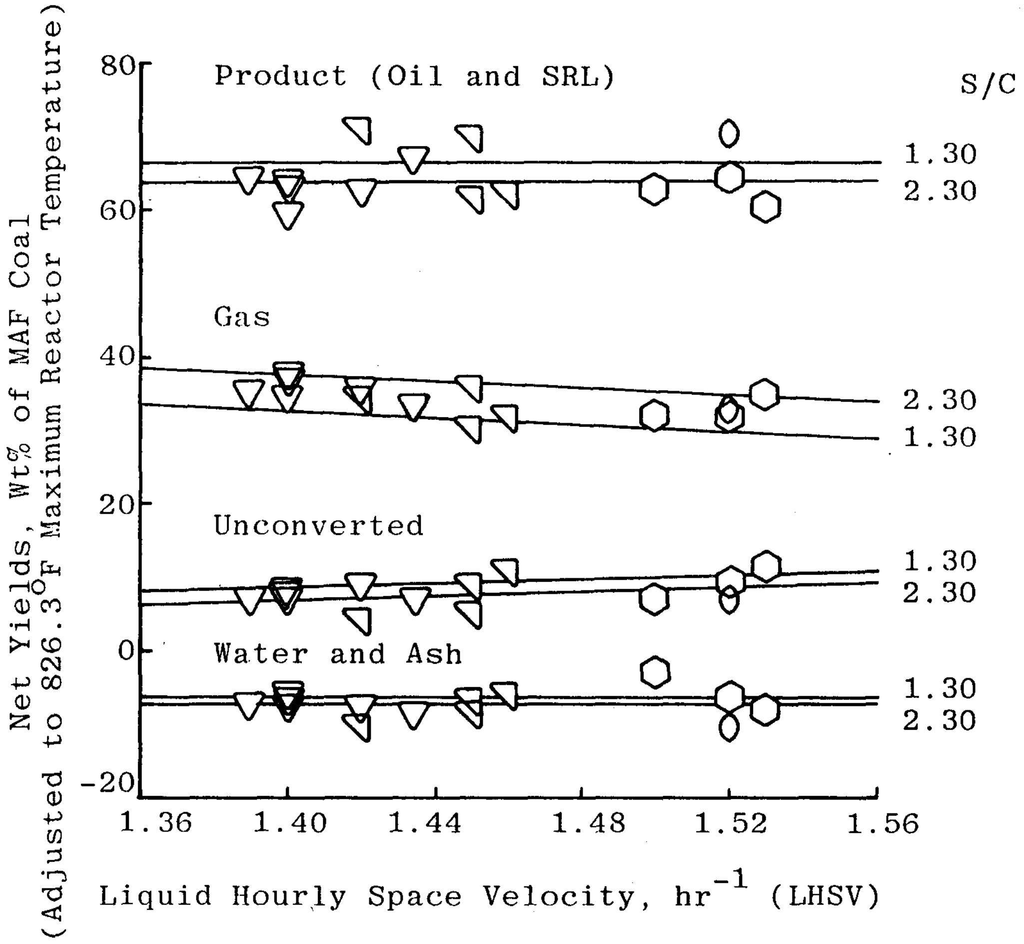


FIGURE 9 - Yields of Major Products vs. Liquid Hourly Space Velocity with Solvent/Coal Weight Ratio as Parameter (Yields Adjusted to Constant Temperature)



The overall regression equations expressing the yields of major products as functions of temperature, space, velocity, and solvent/coal ratio at 2500 psig with 1/1 H_2/CO charge gas are as follows:

Net Gas = G = 177.768T - 20.795S + 4.719R - 173.120
Net Water = W = 20.706T + 2.239S - 0.701R - 35.604

Below 826.3°F

Unconverted = U = -799.669T + 14.741S - 1.628R + 1018.403
Net Product = P = 601.195T + 3.815S - 2.390R - 709.679

Above 826.3°F

Unconverted = U = 253.411T + 14.741S - 1.628R - 336.173
Net Product = P = -451.885T + 3.815S - 2.390R + 644.897

where

T = temperature variable, (°F + 460)/1000,
S = Superficial Liquid Hourly Space Velocity, LHSV, hr^{-1},
R = Solvent to Coal Ratio, S/C, Lbs. Solvent/Lb. Raw Coal.

The regression equations for the yields of all products as functions of temperature only, ignoring the effects of other variables, are as follows:

Net Gas = G = 177.768T - 194.334
Net Water = W = 20.706T - 33.685
Net SRL = SRL = -372.992T + 529.971
Net Lt. Oil= LO = 407.359T - 511.455
CO_2 = CO_2 = -97.708T + 178.343
C_1- C_3 HC's = HC = 138.598T - 169.079

Below 826.3°F

Unconverted = U = -799.669T + 1036.696
Net Product = P = 601.195T - 708.677

Above 826.3°F

Unconverted = U = 253.411T - 317.889
Net Product = P = -451.885T + 645.908

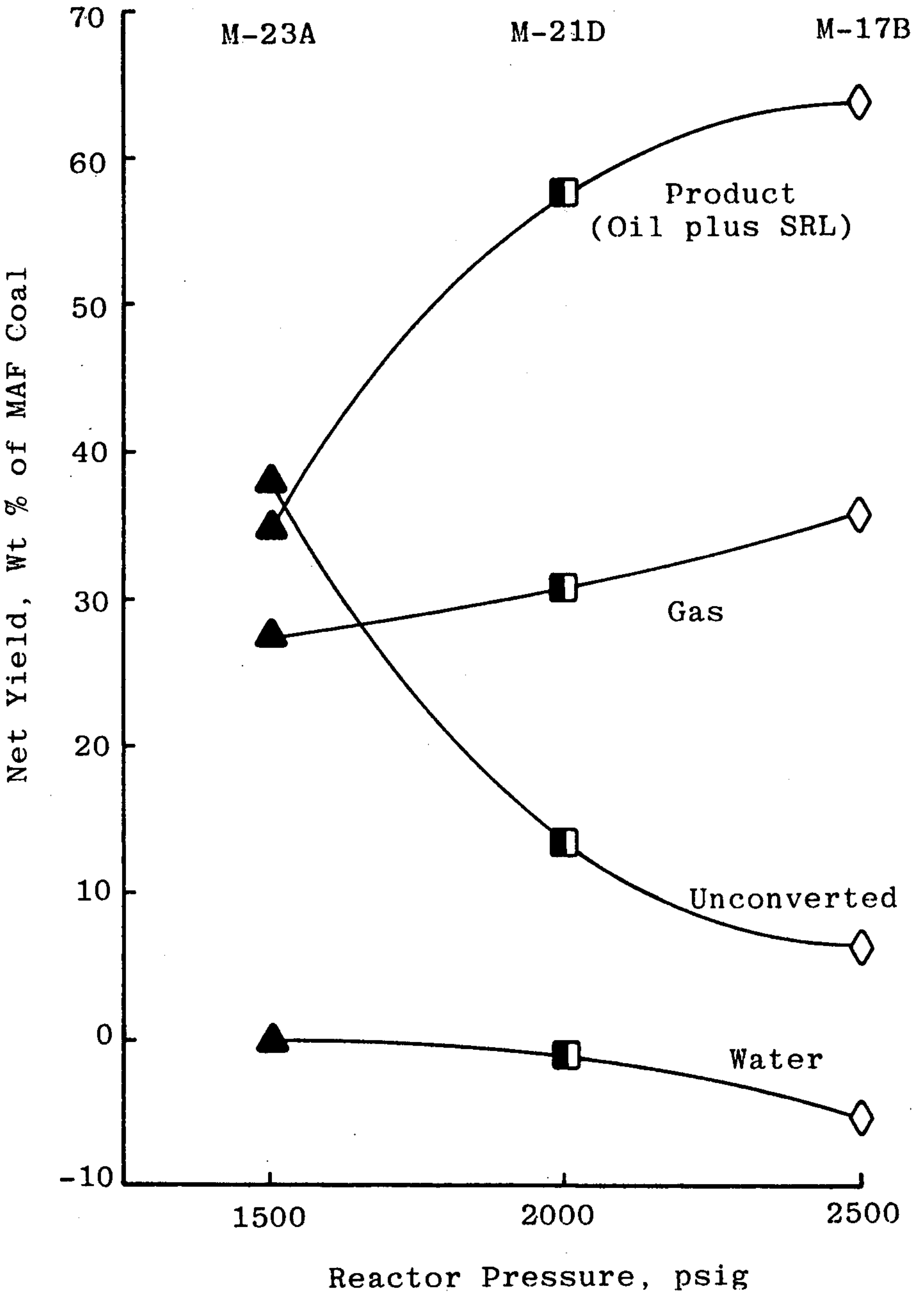
From these, the yields of other products may be derived:

Total Oil = Net Product - SRL
Solvent = Total Oil - Light Oil.

c. Pressure

The yields of major products from runs made at different pressures but at otherwise comparable conditions are shown in Figure 10. These runs were made with a H_2/CO ratio of roughly

FIGURE 10 - Yields of Major Products vs. Reactor Pressure (826-832°F, 1.3-1.4 LHSV, 1.8-2.2 S/C Ratio, 71-74% H_2 in Charge Gas, 11-13 SCF Gal/Lb Coal)



3/1 in the charge gas, so they are not directly comparable with the runs discussed previously. Conversion, gas yield, product yield, and water consumption all increase greatly with increasing pressure.

The distributions of the liquid and solid products are shown in Figure 11 as functions of pressure. The SRL yield decreases somewhat with increasing pressure, and the light oil yield increases. There is a large loss of solvent boiling range material at the lower pressure, which decreases as the pressure increases, while the light oil yield increases with pressure. The total oil yield shows net losses at 2000 psig and lower, though an appreciable net yield is experienced at 2500 psig. However, the solvent does not nearly approach a breakeven point.

The yields of carbon dioxide and of light hydrocarbons are plotted in Figure 12. The carbon dioxide yield increases with increasing pressure, while the light hydrocarbons yield changes little.

The results of the runs at the different pressures indicated that the PDU should be operated at the maximum permissible pressure, 2500 psig, to give any opportunity for operating in solvent balance.

d. Composition of Feed Gas

Though most runs in the PDU were made with a simulated synthesis gas having a hydrogen to carbon monoxide ratio of about 1/1, some runs were made with ratios of about 3/1 and 1/3. Only one run was attempted and only one yield period was completed successfully with the 1/3 H_2/CO ratio. The high CO content resulted in the production of large amounts of CO_2 in the off gas, which make it difficult to control the reactor pressure. The CO_2 expanding through the back pressure regulating valve resulted in temperatures below 32°F and some problems with freezing in valves and lines. This run was made at a higher temperature than the ones with 3/1 H_2/CO, so in order to compare them the product yields from this run were adjusted to 826°F, by using the slopes of the yield-temperature relations established by the regression analyses of the runs with 1/1 H_2/CO. Thus, the implicit assumption was made that the slopes of the yield temperature relations are the same for operations with 1/1 and 1/3 ratios of H_2/CO. The run at 3/1 H_2/CO was made at 826°F, so no temperature adjustment was required for that yield period. However, the yields of major products from both runs were adjusted to the same average space veolcity and solvent/coal ratio by the slope of the relations found for the 1/1 H_2/CO operations at 826°F.

The results of these two runs are compared with the results calculated by the regression equations for operation

FIGURE 11 - Yields of Oil and Solvent Refined Lignite vs. Reactor Pressure (826-832°F, 1.3-1.4 LHSV, 1.8-2.2 S/C Ratio, 71-74% H_2 in Charge Gas, 11-13 SCF Gas/Lb Coal)

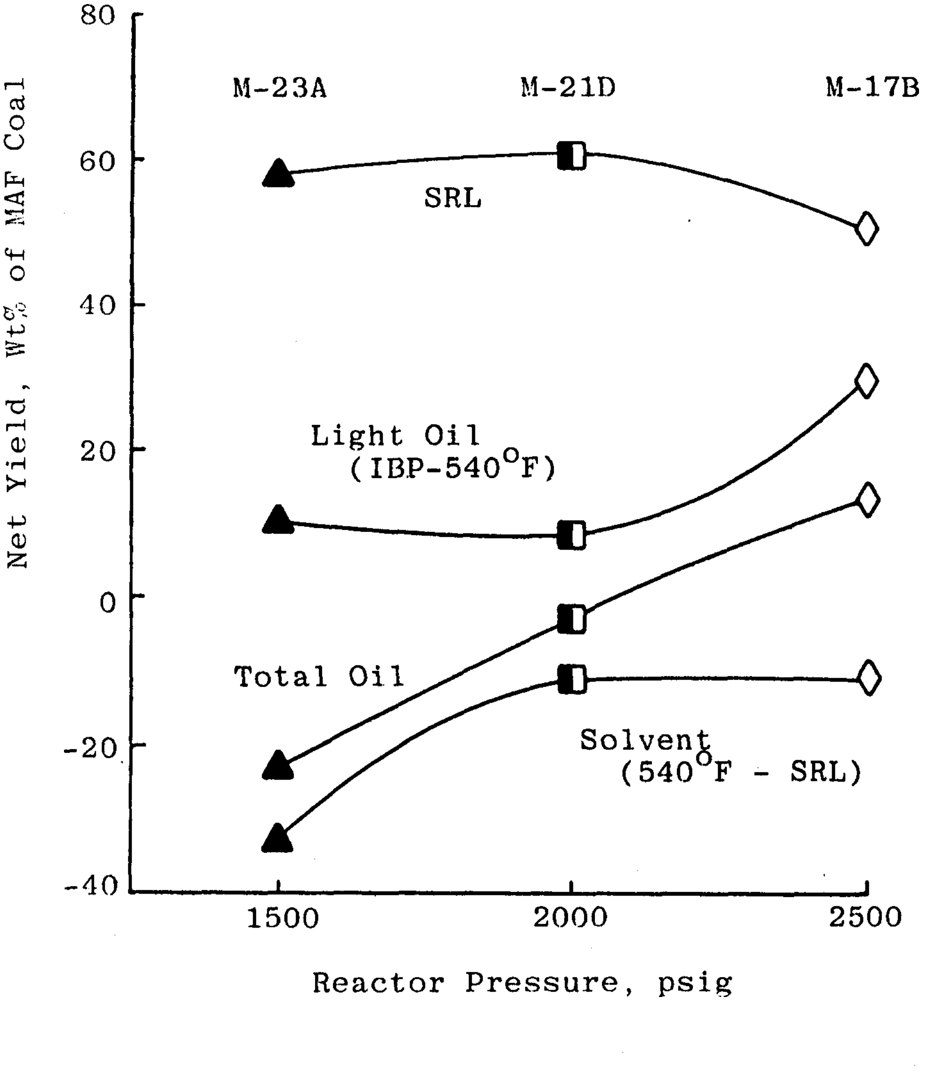
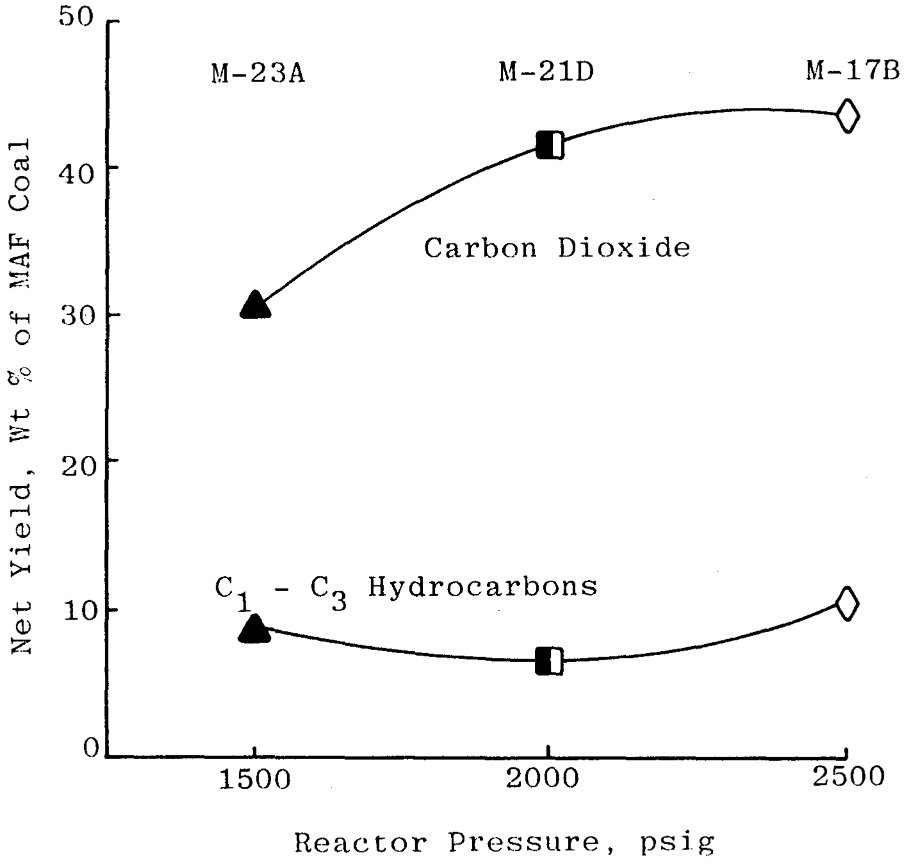


FIGURE 12 - Yields of Carbon Dioxide and Light Hydrocarbons vs. Reactor Pressure (826-832°F, 1.3-1.4 LHSV, 1.8-2.2 S/C Ratio, 71-74% H_2 in Charge Gas/Lb. Coal)



with 1/1 H_2/CO at the same temperature, space velocity, and solvent/coal ratio in Figures 13, 14, and 15.

In Figure 13, the yields of the major products are plotted against mol percent hydrogen in the charge gas. There are only small apparent effects of gas composition. The net product, the conversion, and the water consumption decrease slightly with increasing hydrogen concentration, while the net gas yield is little affected.

The product distribution is much more affected by gas composition as shown in Figure 14. The SRL yield is high at 25 percent hydrogen, decreasing to about the same yield at 50 and at 75 percent. Conversely, the oil yield is low at 25 percent and increases to about the same yield at 50 and 75 percent. The oil yield is primarily of light oil, and the light oil yield increases steadily with hydrogen concentration, while the solvent decreases and becomes a net loss at slightly above 50 percent. At 25 and 50 percent hydrogen there is a slight net make of solvent, but this becomes an appreciable loss at 75 percent.

In Figure 15 are plotted the yields of carbon dioxide and light hydrocarbons, and the consumption of hydrogen and carbon monoxide versus the percent hydrogen in the charge gas. As might be expected, the consumption of hydrogen and carbon monoxide (principally carbon monoxide) and the production of carbon dioxide are dramatically higher at the higher carbon monoxide concentrations.

The production of excessive quantities of carbon dioxide at high carbon monoxide concentrations and the failure to produce adequate quantities of solvent at high hydrogen concentrations led to the decision to concentrate on operations with hydrogen-to-carbon monoxide ratios of roughly one-to-one.

FIGURE 13 - Yields of Major Products vs. Mol % Hydrogen in Charge Gas (For 826°F, 2500 psig, 1.35 LHSV, 1.97 S/C Ratio)

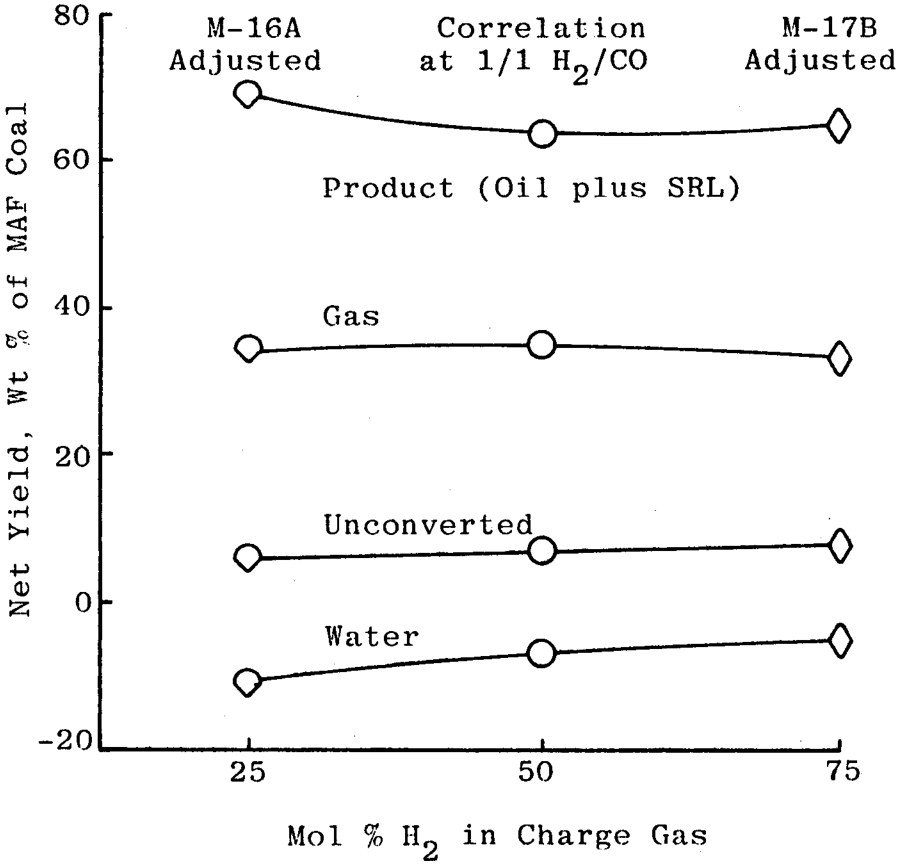


FIGURE 14 - Yields of Oil and Solvent Refined Lignite vs. Mol % Hydrogen in Charge Gas (For 826°F, 2500 psig, 1.35 LHSV, 1.97 S/C Ratio)

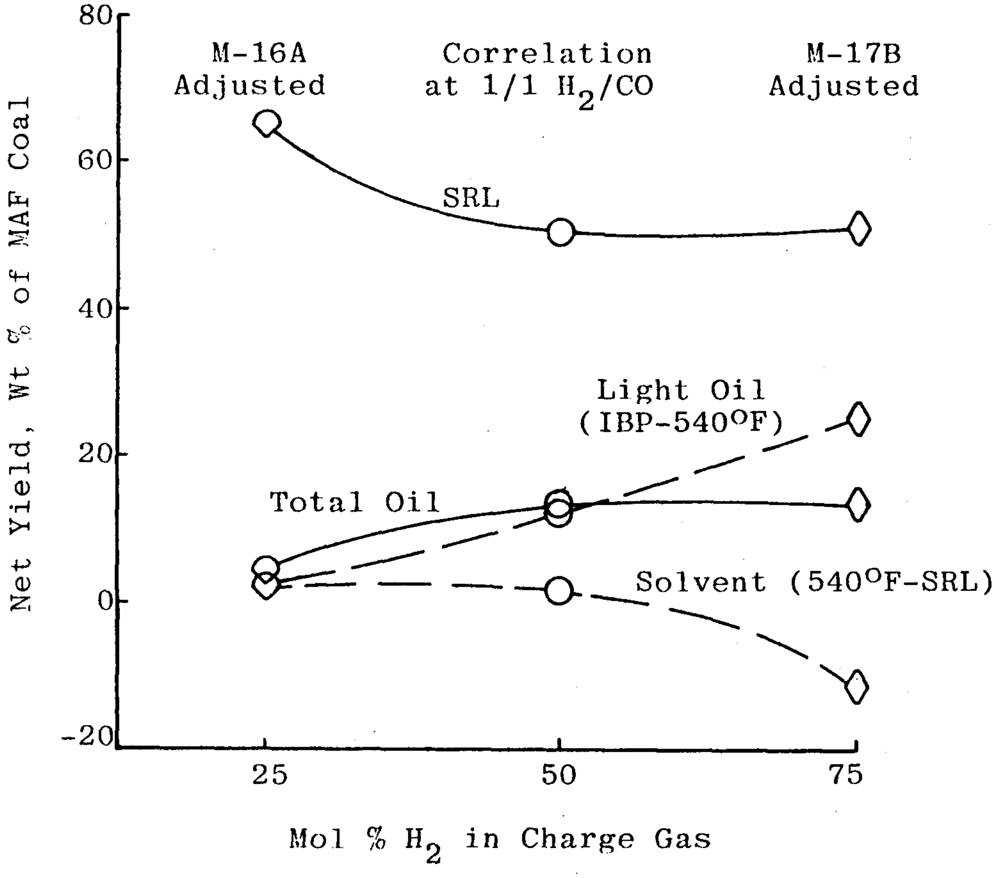
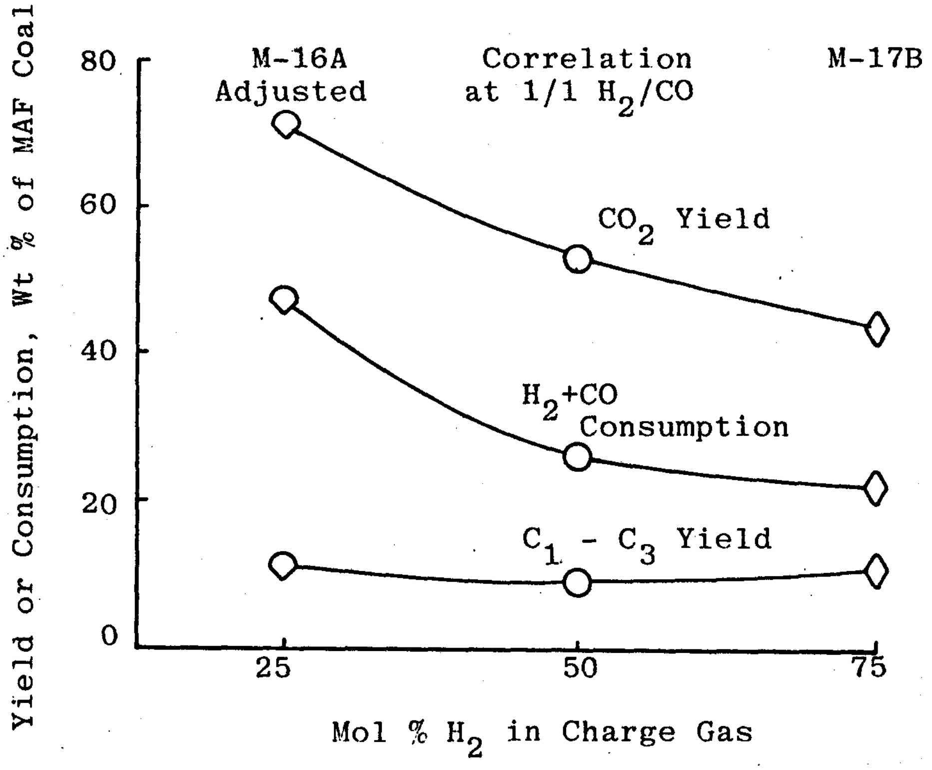


FIGURE 15 - Yields of Carbon Dioxide and Light Hydrocarbons and Consumption of Hydrogen and Carbon Monoxide vs. Mol % Hydrogen in Charge Gas (For 826°F, 2500 psig)



D. OPERATIONAL PROBLEMS

With experience in operation, techniques were developed which enabled the PDU to be run for extended periods (5 days) although completely smooth operation was difficult to achieve because of process upsets. Any mechanical failure or plugging resulted in changes in product characteristics. Major operational problems were associated with plugging in reactors or in process lines. By switching to the alternate reactor, cleaning process lines, or replacing components "on the fly", run conditions could usually be reestablished and the runs continued. Some of the major difficulties are reviewed in the following sections.

1. Materials of Construction

Considerable effort was spent during the design of the PDU on selecting materials of construction so as to avoid major corrosion problems while still providing data suitable for scale up. Type 316 stainless steel was selected for areas where design conditions were 3000 psi and 950°F so as to avoid hydrogen embrittlement which was believed to be the major corrosion mechanism. Beyond the high pressure section, type 304 stainless steel was generally used. Type 316 stainless steel had been used for years in batch autoclave work with lignite. Further, no major corrosion problems had been noted at other locations in continuous liquefaction operations using bituminous coal.

After 400 hours of preliminary operation, including approximately 125 hours with lignite in the system, the preheater coil constructed from 9/16" OD x 5/16" ID, type 316 stainless steel tubing plugged. During removal of the plug, the coil fractured near the slurry inlet. Two replacement coils of 316 stainless steel failed during later operation. One failed after 250 hours of operation and the other after only 28.

Samples cut from the coils were examined and branched-transgranular stress-corrosion cracks were found that initiated at the inside of the tube. Metallurgical examination diagnosed the failures as classical examples of chloride-induced stress corrosion. Little evidence of other corrosion was evident. In the course of additional operation and investigation, stress corrosion cracking was found in the dissolvers, and in tubing and fittings in high-pressure, high-temperature areas.

Samples of the type 316 stainless steel were analyzed and found to meet specifications. Chloride concentrations of 536 and 379 ppm were found by water extraction of the inside of a preheater coil section. Sodium ion concentrations were approximately a tenth of the chloride concentration. It was believed that the chloride at the extract pH of 5 to 6 would be sufficient to initiate the stress corrosion cracking. Possible sources of chloride were investigated. The Gulf FS-120 feed solvent contained about 90 ppm chloride, the recycle solvent near 200 ppm, and the lignite feed 40 ppm. Such concentrations are not high by comparison to those in similar liquefaction processes using coal of high rank where this type of corrosion has not been a problem. It was suggested that perhaps the chloride would be concentrated at the interface where the moisture was evaporated from the lignite which would be not far down from the inlet to the coil preheater. A chlorinated solvent had been used to clean various components in the low pressure sections of the PDU, including the slurry-mix tank. Although care was taken to remove the solvent, 1,1,1-trichloroethane, after cleaning, the design of the mix tank was such that complete residue removal was difficult, and this was probably the source of the chlorine causing the problems. The slurry mix tank was replaced with a trough mixer and cleaning with chlorinated solvent was discontinued.

Because of the major stress corrosion problems with the stainless steel, the entire high-pressure, high-temperature section of the PDU was replaced with Incoloy 800. This replacement resulted in about a six month delay in operations, and the wall thickness of the Incoloy 800 tubing available limited the maximum operating pressure to 2500 psig.

The PDU has been operated from January of 1976 through June of 1977 without appreciable corrosion of the Incoloy. Microexamination of tubing samples after one month service indicated the presence of shallow surface cracks. However, an unused section of tubing exhibited the same appearance.

One other experience with corrosion necessitated a change in materials. The overhead line from the primary separator to the condenser just ahead of the second high-pressure separator was penetrated by a pin hole apparently resulting from acid attack where water and light oils condensed. The original type 304 tubing was replaced by type 316 tubing and there have been no more apparent problems in this area.

Possible corrosion problems in the PDU continue to receive considerable attention. Metal samples are periodically removed for miscroscopic examination. No stress corrosion attack has been noted in the stainless steel (type 316) fittings replaced in January, 1976. However, corrosion and erosion of thermocouple wells and at piping bends have been noted.

2. Solid-Liquid Separation

Removal of the mineral and unreacted components from the SRL is a difficult operation for all SRC processes, most of which used a filtration system at high pressures and temperature. Filtration did not appear to be the answer to separation, and when the laboratory batch tests showed that adequate separation could be obtained, a secondary solvent-settling scheme was designed and installed using toluene as the secondary solvent.

The toluene separation system has not yet been operated satisfactorily despite modifications to improve operation. The initial design involved blending the undeashed vacuum bottoms at 500°F and 400 psi with toluene. Pumping the ash-containing vacuum bottoms and handling the resulting mixture proved extremely difficult.

The system was then modified to perform the solids-liquid separation ahead of the vacuum flash. This greatly improved the handling characteristics of the slurry, but since the solvent was now included in the material for separation, the amount of material dissolved in the toluene was greatly increased, and the design capacity of the system was exceeded. A partial flash was introduced between the liquid-gas separators and the toluene system. Some process solvent was thus removed, reducing the amount handled in the toluene system.

The flow systems have been operating satisfactorily but the separation in the original tower has been successful only for short periods. Some ash-free SRL has been obtained, though the unit has produced ash-free SRL for only a short time before the ash content started increasing. Apparently the bottoms from the settling tower are not being completely withdrawn from the system, and the tower fills with solids in a few hours, preventing proper settling. The modifications attempted within the framework of the present system have not been successful. Redesign and replacement was not within the scope of the project. It is still believed that this mode of separation would be a viable process, but much additional work is required.

3. Accumulation of Solids in Reactor

The major unresolved problem affecting continuous smooth operation of the PDU was related to accumulation of solids in the reactor. With the upflow configuration of the reactors, some of the more difficult-to-liquify portions of the lignite material had a tendency to remain in the reactor and, in fact, had a tendency to agglomerate. Other liquefaction projects with the same uplow reactor design have reported similar build-up.

The accumulation influenced operation by two modes. First, the less reacted material had higher melting points and included even essentially non-reacted material, which when discharged into the downstream process lines resulted in plugging of the lines and separation difficulties of the material in subsequent steps. When one step in the process did not operate properly, the following steps also experienced difficulties and complete shut-down often resulted. Second, the material which remained for longer periods in the reactor reduced the effective reactor volume and resulted in increased liquid space velocities. This reduced liquefaction, resulting in similar operational problems as in the first case. Additionally, the material remaining in the reactor for periods at elevated temperature had a tendency to coke and form deposits which at times plugged the reactor and had to be drilled out following shutdown.

With the reactor design employed, the build-up of solids could not be eliminated. Modication or redesign of the reactors could not be accomplished within the scope of the research contract.

4. Miscellaneous

As is common with new systems operating under severe conditions of temperature and pressure, and handling abrasive solids and viscous liquids, initial difficulties were encountered with pumping, separation, and with the control system. Most of these problems were eliminated or reduced by replacing components including valve trims and pumps as well as modifying the flow systems. However, when process upsets resulted in very viscous liquids or off-control conditions, troubles were still encountered. Under good operating conditions, the system did function satisfactory. Some problems still remain such as separating oil-water mixtures when the components are of nearly the same density.

V. LABORATORY STUDIES IN BATCH AUTOCLAVES

A. INTRODUCTION

Investigation of lignite liquefaction by solution-hydrogenation at the University of North Dakota in bench-scale using batch autoclaves was started under private sponsorships and continued with Federal sponsorship (Office of Coal Research, Energy Research and Development Administration and Department of Energy). The purposes of the laboratory work were initially to develop process conditions to prove lignite liquefaction at high conversions to a Solvent Refined Lignite (SRL) product, develop design parameters for a continuous Process Development Unit (PDU), to serve a supporting role during operation of the PDU, and test process variables not run in the PDU in a small scale. Results have been reported in several publications (2-11).

Special note should be made of the work done in the Department of Chemistry at the University of North Dakota under a subcontract (11); additionally, a series of Master's Theses were done on phases of the laboratory development investigation, some of which were written prior to Federal sponsorship (12-15). These theses are available from the Chester Fritz Library, University of North Dakota. Complete details concerning the batch autoclave work are in the Quarterly Technical Progress Reports which are available through the National Technical Information Service (16).

This section summarizes the major findings in the bench-scale investigations.

B. EQUIPMENT AND PROCEDURES

The laboratory studies were done generally in two one-gallon stainless steel stirred autoclaves having maximum working pressures of 5000 psi at temperatures to 900°F. Lignite was slurried with an organic solvent and processed in a hydrogen, a hydrogen plus carbon monoxide (synthesis gas) or a carbon monoxide atmosphere under selected process conditions.

The organic solvent and pulverized lignite were charged to the autoclave in a 2:1 ratio of solvent to moisture-ash-free (MAF) lignite. Approximately 320 grams of lignite (200 grams MAF) was used for each batch. Water was added in some instances to maintain a constant water content in the autoclave. The autoclave was pressured with gas to give the atmosphere desired, brought to reaction temperature, and kept at temperature for the desired reaction time. After completion of a test, the autoclave was cooled to about 400°F, and the gases discharged through a series of cold traps and collected in a balloon receiver. The collected gases were mixed, analyzed, and metered. After discharge of the gases, the autoclave was repressurized with nitrogen, and the slurry, still at 400°F, was discharged through a dip tube onto a heated Buchner funnel where solvent and coal-derived liquids were separated from unreacted lignite and mineral matter by filtration. Detailed equipment arrangement, test procedures, analytical procedures, product work-up, and calculation methods were presented in Interim Report No. 2 (2).

In evaluation of process variables a bulk sample of a "standard" lignite was used as a constant feed source of known analysis and variability. Proximate and ultimate analyses were made of the sample used for each test, and the analytical results were reported with the test data. During pulverization care was taken to see that lignite moisture was retained to the greatest extent possible. The pulverized lignite was liquefied under controlled conditions in a standardized test sequence except for special tests when modifications of the standard procedure were used. Complete material balance, yield and analytical data have been reported in Interim Reports Nos. 2 and 4 (2,3). Selected data are included in the individual presentations. Comparisons are usually with average data obtained from baseline conditions. Modified test conditions for special tests are noted in the discussion section.

Yield data are reported on the basis of net conversion of MAF lignite to products. Changes in liquids, gases and water from processing are assigned to the lignite. Reported are the solvent refined lignite (SRL), net liquid, and net gas yields. Net liquid is that of lower boiling point than the SRL in excess of the amount of solvent charged and includes changes in solvent inventory. Mineral matter is reported as ash and the small change in the mineral matter inventory is reported with the change in water as "net water plus ash". The water yield is usually negative because moisture in the lignite is used in the liquefaction reactions. This consumption is considered a negative production.

The fraction obtained from 100° to 230°C at 1.6 mm Hg in distilling the gross solvent sample is the standard solvent. Light oil is the fraction from the initial boiling point to 100°C. Heavy oil is that obtained above 230°C. As presented in the tables, light oil and SRL yields are presented separately as well as together as "net liquids". Yields are normalized to the basis of 100 percent recovery of total input.

Details of the autoclave mechanism are illustrated in Figure 16. The arrangement of the autoclave and accessories are shown schematically in Figure 17. Figure 18 presents a summary of the basic analytical scheme employed.

FIGURE 16
BATCH AUTOCLAVE APPARATUS

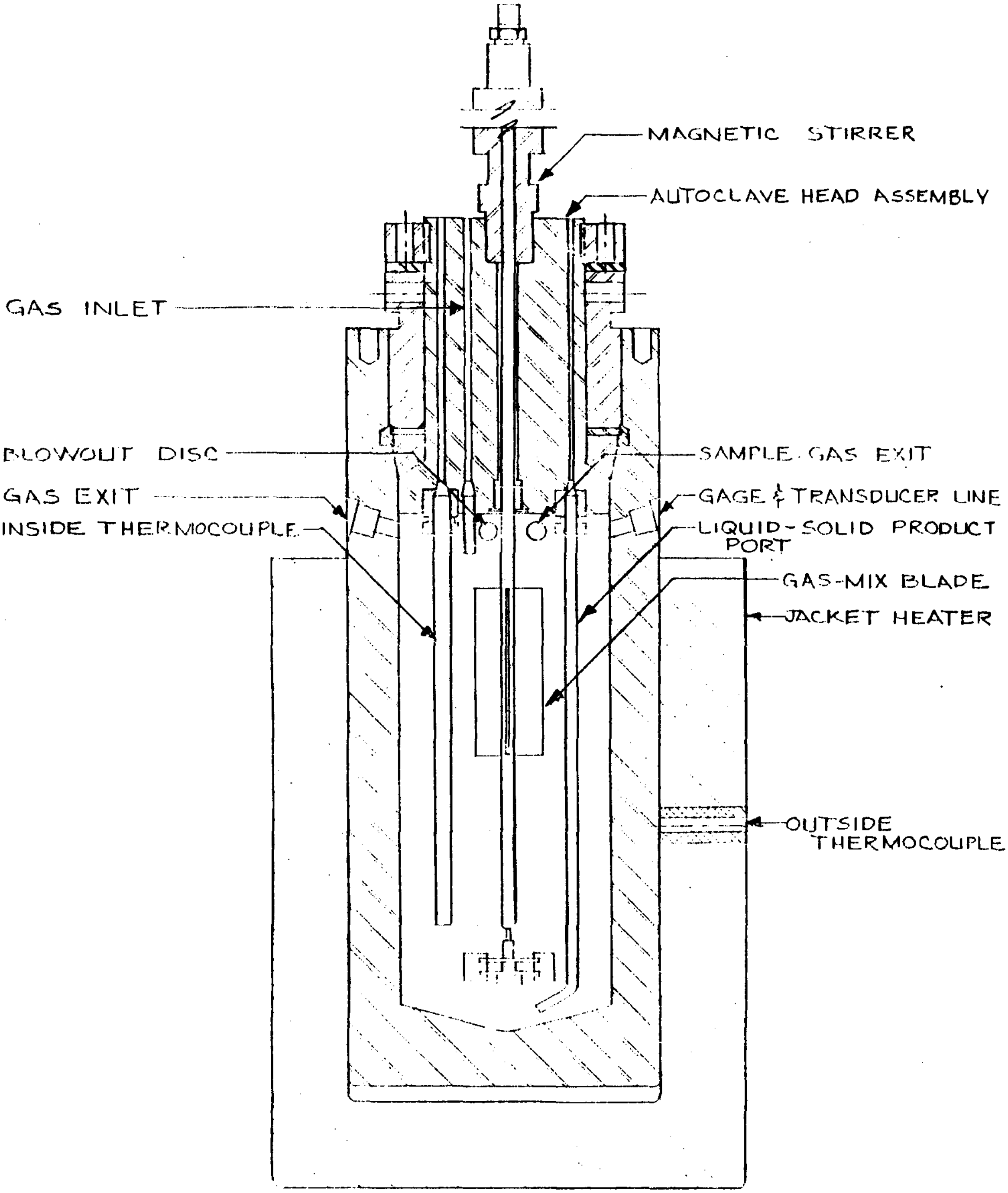
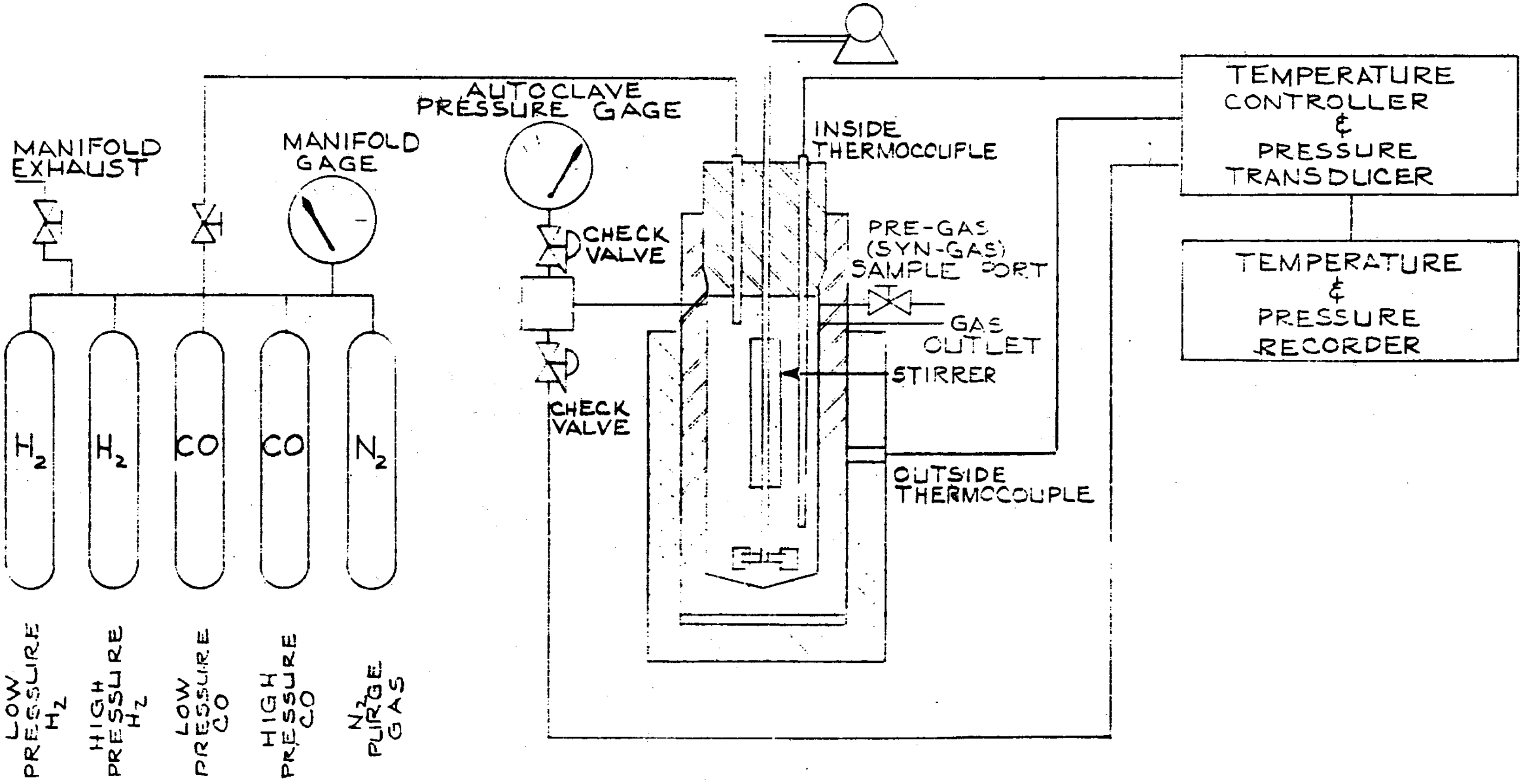


FIGURE 17
AUTOCLAVE APPARATUS/ACCESSORY SCHEMATIC



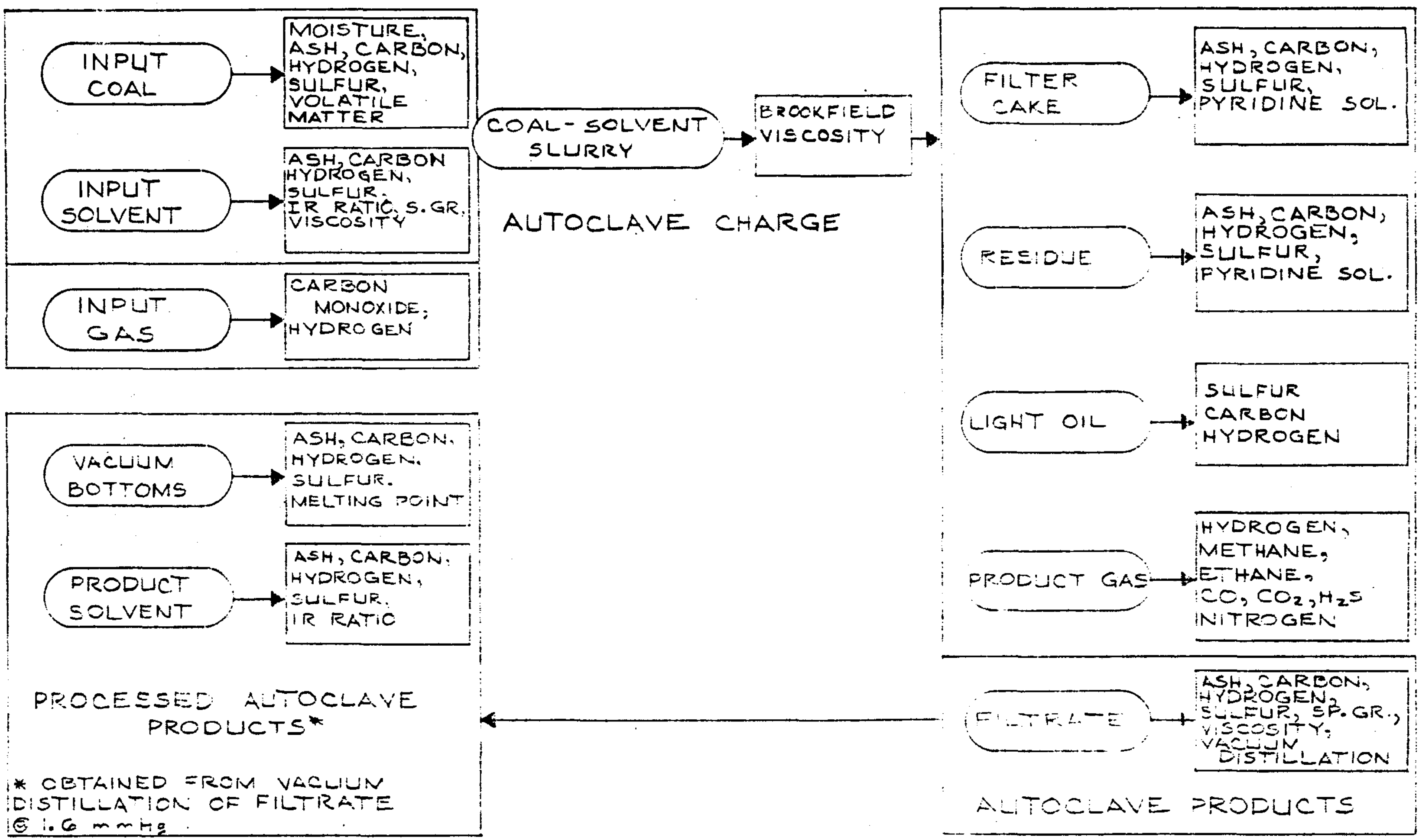


FIGURE 18
ANALYTICAL TEST SCHEMATIC
BATCH AUTOCLAVE LIGNITE LIQUEFACTION STUDIES

C. RESULTS FROM PARAMETRIC STUDIES

1. General

Prior to Federal funding, experiments had shown that lignite could be liquefied to produce a SRL at high total conversion and yields in a variety of organic solvents and in a carbon monoxide, hydrogen atmosphere and hydrogen alone (5,6,11,12,13). In the batch autoclaves, good results were obtained by heating the mixed slurry of full moisture lignite and organic solvent initially at a pressure of 1000 psi in the selected atmosphere to 400°C (752°F) at a rate of 3°/min. and holding at reaction temperature for a half hour, the minimum time required for cycling of the batch process. Major activities in the batch autoclave investigations included investigating process variations to define conditions for design of the continuous 50 lb/hr unit and to provide special testing in support of the continuous operations. Results from the parametric investigations are summarized under the appropriate headings in the following subsections.

2. Evaluation of Start-Up Solvents

Although a variety of solvents had been used, anthracene oil had been the solvent of choice for most of the runs in the batch autoclave. Anthracene oil, a coal-derived solvent, was satisfactory and reasonably priced in comparison to most other solvents initially investigated, but insufficient quantities were immediately available for start-up of the process demonstration unit. Consequently, other solvents were investigated for possible use in start-up and initial operation of the PDU until process-derived solvent would become available to sustain the process. Details of this phase of solvent evaluation and other solvent studies have been reported (1,2,3,7,10).

In the survey of solvents, both coal and petroleum-derived solvents were used in standard batch autoclave tests. Evaluation was on the basis of product yields, characteristics of the solvent refined lignite product, characteristics of recovered recycle solvent, ease of handling, and the percentage of a "standard cut" in as-received solvent. The standard cut in solvent distillation is the fraction obtained in the temperature range of 100 to 230°C at a pressure of 1.6 mm Hg. Light oil is the fraction distilled from IBP to 100°C, and heavy ends is that distilling between 230 and 255°C. For some tests, the raw or "as-received" solvents were used directly. In others the as-recieved solvents were topped to 350°F before use, but in most the standard solvent cut was used. Some of the anthracene oil and creosote oil solvents were hydrogenated before use. Descriptions and distillation data on the solvents are given in Table 3, as well as the numbers of the runs in which they were used.

TABLE 3: EVALUATION OF LIQUEFACTION SOLVENTS

DISTILLATION DATA ON THE SOLVENTS

UND Designation (See Notes)	RAO	HRAO	CAO	HCAO	RCO	HRCO	CCO	HCCO
Vacuum Distillation @ 1.6 Torr								
IBP - 100°C, Wt%	6.2	7.0	3.9	6.0	26.8	16.3	20.5	16.3
100°-230°C, Wt%	85.4	83.6	84.7	85.6	68.5	78.6	72.9	78.3
230°+Bottoms, Wt%	8.4	9.4	11.4	8.4	4.7	5.1	6.6	5.4
ASTM D-246 Distillation								
IBP, °C	232		234		194		193	
IBP - 210°C, Wt%	0.0		0.0		0.6		0.7	
210°-235°C, Wt%	0.1		0.1		7.5		9.1	
235°-270°C, Wt%	2.2		2.6		27.9		27.9	
270°-315°C, Wt%	21.7		22.1		24.4		23.2	
315°-355°C, Wt%	35.7		36.6		22.7		18.0	
355°C+Bottoms, Wt%	40.3		38.6		16.9		21.1	
Atmospheric Distillation to 350°F								
IBP - 350°F, Wt%	0.6		0.7		1.2	3.0	1.0	
Topped 350°F + Cut, Wt%	99.4		99.3		98.8	97.0	99.0	
ASTM D-246 Distillation of Topped 350°F + Cut								
IBP, °C	251		254		---	---	---	
IBP - 210°C, Wt%	0.0		0.0		0.0	0.3	0.0	
210°-235°C, Wt%	0.0		0.0		5.0	6.5	7.1	
235°-270°C, Wt%	1.8		0.8		26.8	29.7	29.4	
270°-315°C, Wt%	10.7		13.1		24.6	24.5	20.6	
315°-355°C, Wt%	34.3		32.7		22.7	23.8	19.0	
355°C + Bottoms, Wt%	53.2		53.4		20.9	15.2	23.9	
Vacuum Distillation @ 1.6 Torr of Topped 350°F + Cut								
IBP - 100°C, Wt%	6.6		7.1		29.2	32.9	14.5	
100°-230°C, Wt%	86.8		84.0		66.2	60.5	72.7	
230°C + Bottoms, Wt%	6.5		8.9		4.6	6.6	12.8	
Used in Runs Nos								
As Received								
Topped to 350°F	511		510		463	464	462	
Standard 100°-230°C @ 1.6 Torr Cut	455	468	465,483 484	471,480	447,448 491	479	475	474

Notes: RAO, Raw Anthracene Oil; HRAO, Hydrogenated Raw Anthracene Oil; CAO, Chilled Anthracene Oil; HCAO, Hydrogenated Chilled Anthracene Oil; RCO, Raw Creosote Oil; HRCO, Hydrogenated Raw Creosote Oil; CCO, Chilled Creosote Oil; HCCO, Hydrogenated Chilled Creosote Oil;

RAO, CAO, RCO, and CCO from Reilly Tar and Chemical Co.

TABLE 3 Cont: EVALUATION OF LIQUEFACTION SOLVENTS

DISTILLATION DATA ON THE SOLVENTS

UND Designation (See Notes)	LCO	MHCO	FS120	FO-6	FO-5	HAN	ATS-2	AC
Vacuum Distillation @ 1.6 Torr								
IBP - 100°C, Wt%			0.5	6.9	0.8	47.2	0.3	0.0
100°-230°C, Wt%			65.2	24.2	46.3	52.8	55.4	23.2
230°C + Bottoms, Wt%			34.3	66.8	52.9	0.0	44.3	76.8
ASTM D-246 Distillation								
IBP, °C	238	339	316	218	276	214	271	355
IBP - 210°C, Wt%	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
210°-235°C, Wt%	0.0	0.0	0.0	0.5	0.0	9.2	0.0	0.0
235°-270°C, Wt%	48.4	0.0	0.0	3.0	0.0	70.3	0.0	0.0
270°-315°C, Wt%	43.8	0.0	0.0	8.0	0.5	19.0	20.6	0.0
315°-355°C, Wt%	6.4	9.9	2.4	6.9	3.0	1.0	24.7	0.0
355°C + Bottoms, Wt%	1.4	90.1	97.6	81.6	96.5	0.5	54.7	100.0
Used in Runs Nos								
As Received	503	507,508	514		513	519	496,509	500,506
Topped to 350°F								
Standard 100°-230°C @ 1.6 Torr Cut			485,497	473,478	486		495,499	494,501

Notes: LCO Light Creosote Oil; MHCO, Middle Heavy Creosote Oil; FS120, FS120 Carbon Black Feed Stock; FO-6, No. 6 Fuel Oil; FO-5, No. 5 Fuel Oil; HAN, Heavy Aromatic Naphtha; ATS-2, Aromatic Tar S-2; AC, Aromatic Concentrate.

LCO and MHCO from United States Steel Co.

FS120 from Gulf Oil Co.

FO-5 from Standard Oil Company

FO-6, HAN, ATS-2, and AC from EXXON Company, USA.

The ranges for unconverted lignite, yields of gas, and the water consumption in weight percentage on an MAF basis of lignite charged for the coal-derived solvents were:

Item	Weight Percent
Unconverted lignite:	5.7 to 14.6
Gas yield:	35.0 to 46.5
Water consumption:	5.0 to 17.3

Yields of light oil when the standard distillation cut was used were higher than when the heavy ends or as-received solvents were used. Hydrogenating creosote oil gave enhanced light oil yields; however, hydrogenation of anthracene oil had no apparent effect on light oil yields.

Filtration characteristics of the product slurries, rated from best to worst, for various solvents, follows: creosote oil, light creosote oil, anthracene oils, middle-heavy creosote oils. Ease of handling of as-received solvents, from best to worst, are: anthracene oil, light creosote oil, creosote oils, middle-heavy creosote oil.

The approximate weight percentage of standard cut solvent recoverable from the as-received solvents was:

Creosote oils	70
Anthracene oils	85
Light creosote oils	30

The product SRL had H:C atomic ratios greater than 0.9 only when hydrogenated creosote oil was the solvent. Use of middle-heavy creosote oil resulted in low H:C ratios < 0.7, and a high sulfur content ($\sim$0.4 weight percent) in the SRL.

IR ratio (IRR) is used to measure the relative intensity of the infra-red absorbances of aromatic and aliphatic hydrogens in liquefaction solvents at wave lengths of 3.28 and 3.41 microns, respectively. Thus, the degree of hydrogenation (or dehydrogenation) of a solvent during a liquefaction test can be approximated by comparing IR measurements before and after the liquefaction test. When the solvent is hydrogenated, the absorption intensity of the aliphatic hydrogen (3.41μ) increases and that of the aromatic hydrogen (3.28μ) decreases. Thus an increase in IRR for fractions of the same boiling range indicates an increase in aromatic content. Product that was recovered in the solvent boiling range exhibited an IRR lower than that of the charged solvent indicated hydrogenation of the solvent during liquefaction. Sulfur content of the recovered solvent was also lower except when hydrogenated creosote oil was used. Standard cut solvent recovery for possible recycle was nearly sufficient to meet recycle requirements for the anthracene, but was low for the creosote solvents.

Petroleum-derived solvents exhibit similarities in yield and conversion as follows:

Item	Weight Percent
Unconverted (MAF basis)	8.3 to 12.8
Gas yield (MAF basis)	∿40
Water consumption	7.4 to 14.0

Exceptions were #5 fuel oil which resulted in lower overall conversion and the aromatic concentrate which gave higher gas yields.

Low light oil yields which may be associated with more stable solvents were obtained with carbon black feedstock, aromatic concentrate, and #5 fuel oil when distilled to the standard cut. Some as-received solvents also gave low light oil yields. These were heavy aromatic naphtha, carbon black feedstock, and aromatic concentrate. Yields of standard cut solvents obtained from the as-received solvents ranged from 42.2 to 55.4 weight percent except for the carbon black feedstock where the yield was 65.9 weight percent. Ease of solvent handling from best to worst was:

1. Carbon black feedstock and heavy aromatic naphtha.
2. Aromatic concentrate and the fuel oils.
3. Aromatic tar.

Separation of the unconverted material from SRL by filtration was superior with carbon black feedstock, aromatic tar, and aromatic concentrate solvents to that obtained using heavy aromatic naphtha and the fuel oils as solvents.

SRL with the highest H:C atomic ratio of about 0.9 was produced using the fuel oils as solvent. However, the initial H:C ratio of solvent was also the highest with the fuel oils, about 1.5. The petroleum-derived solvents, except heavy aromatic naphtha and aromatic tar, resulted in relatively higher sulfur content of the SRL (∿0.9 weight percent) when used as-received. When the solvent was distilled to the standard cut, low sulfur content SRL (∿0.35 - 0.40 weight percent) was produced using aromatic tar and the carbon black feedstock.

The IRR's of initial and recovered solvents were not appreciably different, although the IRR of the recovered solvent was slightly lower than that of the initial solvent in the case of aromatic tar. During the liquefaction process, sulfur content of the solvent fraction was reduced. All solvents used as-received showed a net increase in standard solvent boiling range after one pass through the liquefaction step. Recovery of solvent was sufficiently high for the standard cut solvents to indicate that the process might be self-sustaining, except when using aromatic tar and aromatic concentrate.

Of the various solvents investigated, carbon black feedstock, anthracene oil and creosote oil appear the most promising for liquefaction of lignite. Because of availability and low cost, carbon black feedstock is the present candidate for use as the start-up solvent in the Process Development Unit.

Several other general conclusions regarding the various solvents are:

1. On the as-received basis, the fraction of standard cut solvent present was higher for the coal-derived than for the petroleum-derived solvents.
2. Hydrogenation of coal-derived solvents prior to use significantly changed the properties of creosote oils but not of anthracene oils.
3. During processing, coal-derived solvents were more highly hydrogenated than were the petroleum-derived solvents, as indicated by IRR.
4. Some hydrodesulfurization of all solvents, except the aromatic tar, was evident during processing.
5. A net increase in standard cut solvent was noted when processing as-received or heavy ends solvents indicating that some heavy ends distillable from the SRL should be included as recycle solvent.
6. Gas yields, water consumption, and the fraction of unconverted material were nearly the same for most solvents although standard solvent cuts from coal-derived solvents gave higher light oil yields than did the petroleum-derived solvents.
7. SRL prepared using petroleum-derived solvent had a higher sulfur content than did product obtained using coal-derived solvent. The higher sulfur content of the petroleum-type solvents apparently retards desulfurization of the SRL.

Use of the Gulf carbon-black feedstock, FS-120, derived from petroleum was successful as solvent in initial operation of the continuous unit. Because of operating problems associated with halide-induced stress-corrosion cracking of stainless steel components exposed to high pressures and high temperatures, a search was made for a solvent with a low chloride content that might be suitable. The highest concentration of chloride in process feed to the PDU was found to be in the solvent. One solvent listed was a petroleum based LSX-745 from Neville Chemical Co. Other solvents tested were recycle solvent obtained from the Pittsburg & Midway Coal Mining Company (PAMCO) pilot plant at Fort Lewis, Washington, and a recycle solvent from Project Lignite's PDU. Standard liquefaction procedures were followed.

a. Neville Chemical Co. Sample LSX-745

Solvent	Average Results with FS-120	LSX-745
Chloride Content, ppm	229.0	94.0
Yields, % MAF Lignite		
Gas	34.0	32.3
Liquid	64.6	66.0
(Light Oil)	(6.6)	(6.3)
(Net SRL)	(58.0)	(59.8)
Unconverted	11.4	6.6
H_2O and Ash	-10.0	-4.9

No significant difference is apparent in performance of the solvents as regards liquefaction. LSX-745 solvent appears to show satisfactory activity and could be used in place of the FS-120. Though the chloride content is less than one-half that of FS-120, it was not considered low enough to justify changing solvents.

b. PAMCO Process Solvent

Two tests under standard liquefaction conditions were made with the PAMCO process solvent. Yield data are summarized in the following together with average yield data from 19 tests using FS-120.

Yields, Wt % MAF Coal	Baseline Avg. of 19 runs	Run 611	Run 612
		Normal Liquefaction, using PAMCO process solvent & added water	
Gas	34.0	39.7	38.5
Liquid	64.6	67.6	56.8
(Light Oil)	(6.6)	(31.0)	(33.0)
(Net SRL)	(58.0)	(36.6)	(23.8)
Unconverted	11.4	3.1	13.7
Water/Ash	-10.0	-10.4	-9.2
Total	100.0	100.0	99.9
Solvent Recovery	102.0	70.9	87.1

Yields of light oil were much higher and recovery of solvent significantly lower using the PAMCO solvent which suggests that this solvent is particularly susceptible to cracking when used for liquefaction of lignite. SRL recovery was also reduced when the PAMCO solvent was used.

c. PDU Recycle Solvent

Start-up and recycle solvent from the PDU operations were used as solvent in standard batch autoclave tests on PDU coal. Results for Runs 626-628 are compared with those obtained in Run 618 which used the normal solvent fraction (100-230°C @ 1.6 Torr) of UND FS-120-75 in the following:

Run No.	618	626	627	628
Solvent	Standard	M-26 Start-up	M-26B Recycle	
Yields, Wt % MAF Coal				
Net Gas	32	41	47	42
Net Liquid	66	53	43	49
(Light Oil)	(8)	(5)	(20)	(27)
(SRL)	(58)	(48)	(23)	(22)
Unconverted	8	11	10	11
Net Water & Ash	-6	-5	0	-2
Solvent Recovery	91	94	99	97
Solvent Distillation @ 1.6 Torr				
IBP-100°C	-	11	16	
100-230°C	100	82	67	
230°C +	-	7	17	
Solvent Sp.Gr. d 60/60	1.032	1.037	1.014	

Conclusions which may be drawn from the comparisons are:

- Conversions were slightly lower with both fresh PDU solvent and recycle PDU solvent.
- Gas yields are higher and oil yields are lower with the PDU solvents, especially with recycle solvent.
- Net light oil yields were much higher with the recycle solvent.
- Water consumptions were appreciably less with the PDU recycle solvents, and this contributes to the lower liquid yields.

Generally, these results confirm the observations obtained with the recycle solvent from the Pittsburgh and Midway Coal Mining Company (PAMCO) pilot plant at Fort Lewis, Washington. It appears that lighter, lower boiling recycle solvents give relatively poorer results in batch autoclave tests than the fresh heavier solvents. Recycle solvents, however, seem to be adequate in continuous plants. The major difference, other than static versus flowing conditions, are the relative long heating-up and cooling-down periods in the batch operations even though the maximum temperatures are less than in the continuous operations.

3. Simulated Solvent Recycle

Although petroleum-derived carbon black feedstock was indicated to be suitable as an initial solvent for the continuous unit in place of anthracene oil, the question remained as to the effect of changing from the petroleum based solvent to one derived from lignite. To investigate this effect, a series of runs was made in which recovered solvent from a previous batch run was used in a subsequent test. Such a sequence of batch runs simulates a continuous process in that the characteristics of the solvent with successive passes should approach those obtainable in continuous operation. Because of losses and the necessity of sampling together with the inability to recover solvent quantitatively from the filter cake, an average of one and a half batch runs were required to obtain solvent for the subsequent pass. Standard test conditions were used for all tests.

A total of 40 tests were necessary for five passes, and complete details have been reported, (2,3,16). Averaged data for the complete series considering all runs at each level to be replicates are presented in the following summary:

Pass Number	1	2	3	4	5
Number of Tests	19	11	6	3	1
Average yields Wt% MAF lignite					
Net Gas	34.0±1.7*	34.1±3.5*	34.2**	33.2**	34.2**
Net Liquid	64.6±1.4	64.1±2.1	63.1	64.2	64.4
Unconverted	11.4±0.9	10.9±0.7	10.3	11.2	10.7
H_2O plus ash	-10.0±1.0	-9.1±2.1	-10.6	-8.6	-9.3
Net SRL, Wt % MAF Lignite	58.0±1.6	58.2±1.5	56.4	57.9	57.7
Solvent Recoverable % of charged	102.0±1.1	102.3±2.4	100.5	99.9	101.9

* The 95 percent confidence range of average.

** Confidence range not calculated because of small number of tests.

A 95 percent confidence range of the average was calculated for each dependent variable for which sufficient tests were available for adequate degrees of freedom. No significant difference is evident that depends on the number of cycles completed, indicating that the solvent does not degenerate during the process and that the recovered coal derived solvent is at least adequate in regards to solution characteristics and recoverability. No adverse effects were noted in other solvent characteristics in the batch-autoclave tests that would negate use of carbon black feedstock as initial solvent or would indicate that insufficient solvent was regenerated to sustain a continuous process. In fact, solvent recycle appeared qualitatively to have a favorable effect on product handling quality as indicated by improved filterability of the reactor slurry in later passes. Although filtration improves as the pass number increased, viscosities of the reaction slurry measured at room temperature tended to increase as follows:

Pass No.	Brookfield Slurry Viscosity, cp
1	2580-4170
2	3570-4080
3	3770-3890
4	3575-4500
5	5864

As observed in actual hot filtration, this apparent increase in ambient viscosity had no adverse effect.

4. Effect of Lignite Storage

Coals are known to have some alteration of properties including in part caking, heating value, and moisture content during storage, with the effects usually adverse. Liquefaction of lignite in a synthesis gas atmosphere is dependent on properties which may be influenced during stockpiling. In most processing applications there is delay between mining and utilization and often, a considerable amount of coal is stockpiled in dead storage for use in case of interruption in deliveries.

To determine if protracted storage would change the liquefaction properties of lignite, a bulk sample of lignite was subdivided into representative samples, and the samples stored under the following conditions for a total of 70 weeks.

a. Lump lignite under distilled water at ambient conditions. After storage and just prior to liquefaction, the sample was pulverized in a single pass through the mill.

b. Lignite, pulverized by one pass through the mill, in air at 50 percent relative humidity, ambient conditions.

c. Lignite, pulverized by one pass through the mill, in nitrogen at 50 percent relative humidity, ambient conditions.

After predetermined times in storage, samples from each of the three storage conditions were liquefied using the standard procedures. A duplicate test was made on one of three samples in each set.

Results have been reported including the intermediate data (2,3,16). Of major interest are the liquefaction characteristics after 70 weeks of storage. Pertinent data are presented below along with baseline data obtained at the start of the series.

Storage	None	Under N_2	Under Air	Under H_2O
Net Yields, Wt% MAF Coal				
Gas	35.4	31.3	35.1	33.8
Liquid	66.0	64.5	57.6	66.7
(Light Oil)	(20.7)	(18.1)	(20.5)	(22.3)
(SRL)	(45.3)	(46.4)	(37.1)	(44.4)
Unconverted	8.9	9.2	12.4	7.4
Water	-10.3	-5.0	-5.1	-7.9
Conversion, Wt% MAF Coal	91.1	90.8	87.6	92.6

Air storage appears to have a significant adverse effect on lignite liquefaction, i.e., lower net liquid and net SRL yields and lower conversion. Lignite stored in an air-free environment, either in nitrogen or under water, for 70 weeks has about the same reactivity as fresh lignite, as indicated by net liquid and net SRL yields and conversions. Results from liquefaction of samples having intermediate lengths of storage were close to those obtained in baseline tests. The tests series was terminated after 70 weeks.

5. Catalytic Effect of Native and Added Cations

Ash components and naturally occurring cations in lignite are believed to have some catalytic effect on conversion and yields during liquefaction. A series of tests were made in which various cations--Na, K, Fe, Ca, Co, Ni, Mo, and a Ni-Mo catalyst (HDS-3A)--were added to the lignite before liquefaction. Prior extraction of the naturally occurring cations using HCl allowed the added cations to be investigated individually.

The characteristic ability of lignite to function as an ion-exchanger was used to control the concentration of the added cations. Initial HCl extraction reduced the dry-content ash of the lignite from 10 to about 3 percent, most of which was alumina, silica, and iron oxide. Reverse ion exchange allowed the cations in soluble form to be added to the extracted lignite. By adjusting contact time and concentration of the cation in the contacting solution, concentrations of individual cations associated with the lignite could be controlled with possible concentrations in excess of those initially present. Standard test procedures were used in liquefaction. Details of the exchange process, test conditions and results were reported in Interim Report No. 2 (2). A summary of yield data for this series of tests follows:

Cations Present	Lignite, Natural	Extracted Lignite None	Alkali Metals	Alkaline Earths	Transition Metals
Yields, Wt% of MAF Lignite:					
Net Gas	38	26	41	30	25
Net Liquid	68	64	67	62	68
(Light Oil)	(15)	(13)	(17)	(19)	(20)
(SRL)	(53)	(51)	(49)	(43)	(48)
Net Water	-13	-1	-14	0	-1
Unconverted	7	11	6	8	8
Solvent Recovery, %	93	91	93	89	90

Most tests with added alkali metal or transition metal cations gave liquid yields between 67 and 71 percent. The highest yields of SRL were obtained with the raw lignite, along with relatively high gas yields and water consumption. The catalyst ions generally produced more conversion to lighter products, i.e., light oil and gas.

Sodium and potassium, which are water gas shift catalysts, gave high consumption of water during the reaction, and the untreated lignite had high water consumption due to its alkali metal content. Water consumption with calcium and with the hydrogenation catalysts cobalt, nickel, and iron was much lower.

The effect of a given cation was quite insensitive to the amount added in the ranges studied, which for the alkali metals were of the same order of magnitude as the amounts naturally present in the coal.

Solvent recovery was less than 100 percent of solvent charged for all of these experiments. The natural cations and the shift catalyst additions gave closest to self-sufficiency of solvent together with good liquid yields.

The average conversions of lignite to gases and liquids was about 89 percent for extracted lignite, and 92 to 94 percent with cations present. Addition of cations increased the production of light oil at the expense of heavy fuel components.

Apparently, the naturally occurring cations present in the as-mined lignite provide sufficient catalytic action to give high yields of liquid products in a relatively short reaction time. This is a most favorable result since the expense and operational difficulty encountered from catalyst addition are avoided. Ash and unreacted coal can be separated from the polymerization products which removes some of usual hydrogenation catalysts. Additionally, some sulfur and nitrogen are removed. If lower molecular weight liquids are desired, the ash-free SRL can be directly hydrogenated using specific catalyst and process conditions that would optimize product characteristics and distribution.

6. Effect of Lignite Source

High rank coals do not respond well to liquefaction procedures employing either a CO or a synthesis gas (CO + H_2) atmosphere. To find if low rank coals (lignites) from different sources would have a varying response, some lignites from outside the Fort Union Formation of North Dakota were treated using standard liquefaction procedures in a syngas atmosphere. Lignite from the Denver Basin, Thailand, Chile, and North Dakota are compared in the following: (see (2) and (3) for details)

Lignite	North Dakota*	Denver Basin	Thailand	Chile
Yields, Wt% MAF Lignite:				
Net Gas	34.0	34.2	31.9	12.6
Net Liquid	64.6	65.4	67.0	71.2
Unconverted	11.4	6.5	2.1	6.6
Water	-10.0	-6.1	-1.0	9.6

* Average of 19 tests.

All of the lignites responded adequately to the liquefaction process with no significant difference noted between the North Dakota, Denver Basin and Thailand lignites although the Thailand lignite may be marginally subbituminous. The Chilean lignite responded somewhat differently, not reacting to produce as much gaseous products. This difference appears to be related to the reduced utilization of inherent moisture for a source of hydrogen, probably because the minerals in the Chilean lignite lack the ability to catalyze the water gas shift reaction to the extent exhibited by the minerals in the other lignites.

7. Influence of Atmospheric Composition

Three different atmospheres, carbon monoxide, hydrogen, and synthesis gas (syngas), were used in otherwise standard batch-liquefaction tests. Syngas was 50-50 mol percent hydrogen and carbon monoxide. Initial pressure was 1000 psig in each case. Both as-received and acid-extracted lignite were tested. Acid extraction (with HCl) lowers the mineral content of the coal and reduces any catalytic effect of the minerals. A test in an inert atmosphere (He) was also run. Yields for these tests are summarized in the following: (see (2) and (3) for additional details)

As-Received Lignite

Atmosphere	He	CO	Syngas	H_2
Net Yields, Wt% of MAF Coal				
Gas	19.5	66.8	35.2	13.6
Light Oil	4.1	9.3	13.1	14.5
SRL	-	33.3	55.3	43.2
Unconverted	69.6	17.2	10.0	17.8
Water	6.8	-26.6	-13.6	10.9

Acid-Extracted Lignite

Atmosphere	CO	Syngas	H_2
Net Yields, Wt% of MAF Coal			
Gas	24.9	27.1	13.6
Light Oil	11.3	12.4	14.4
SRL	48.7	47.2	43.4
Unconverted	22.6	12.1	15.8
Water	-7.5	1.2	12.8

Conversion of lignite was generally higher in the syngas atmosphere, 88 to 90 percent, as compared with 77 to 84 percent for the other tests except for the test in the inert atmosphere where essentially no liquefaction took place.

Yields of liquid products were appreciably higher using the syngas atmosphere and the as-received lignite with its natural moisture content, 68 percent as compared with 43 to 60 percent in all the other tests. With acid-extracted lignite the liquid yields were not appreciably different regardless of the atmosphere.

For both extracted and unextracted lignite, yields of gaseous components were higher with more carbon monoxide in the charge gas. The highest gas yield was obtained with carbon monoxide and as-received lignite because of the catalytic effect of the mineral matter.

The syngas atmosphere with as-received lignite gave the best liquefaction, i.e., highest conversion and highest yield of liquid products.

8. Effect of Drying and Carbonization on Liquefaction

Several advantages have been suggested for drying or carbonizing before liquefaction. Reducing the moisture content would result in lower transportation costs if the liquefaction plant were not at the mine mouth. A lower moisture content of the charged lignite would decrease the generation of carbon dioxide resulting in a reduced load on the gas purification system. The same level of hydrogen-carbon monoxide partial pressure could be maintained at a lower total operating pressure. Similar advantages could be attributed to carbonization with the additional removal of oxygen, concentration of methane production and possible improvement in liquefaction, and increased carbon content.

The effect of prior dehydration on liquefaction was measured in a test series in which the solvent and lignite were heated at 215°C in the autoclave to remove the moisture. The lignite-solvent mixture was cooled, and a standard liquefaction run made. Tests were made using anthracene oil and FS-120 as solvents. Details and experimental results are reported in (2) and (3). It was found that reducing the moisture content adversely influenced the extent of liquefaction, especially in a synthesis gas atmosphere. Percentage of unconverted lignite for the dehydrated slurry ranged from 23.9 to 41.6 weight percent on an MAF basis compared with 8.0 to 9.8 weight percent for the baseline tests. Conversion was higher in a hydrogen atmosphere than in the synthesis gas atmosphere. Yields of liquids were low, ranging from 36 to 54.2 weight percent of the MAF lignite charged, compared to base test liquid yields from 61.0 to 63.4 weight percent. Filtration rates for the product slurry were reduced significantly starting with dehydrated lignite. However, yields of carbon dioxide were drastically reduced.

When liquefaction is the primary concern, the gain resulting from reduced carbon dioxide generation will not offset the loss in liquefaction activity resulting from the moisture loss.

In other tests, lignite was dried externally to the autoclave, the dried lignite slurried with the solvent, and water added to the same total water content as when full moisture lignite was charged. Yields of light oil was much lower when using dried lignite, and the liquid-solid product slurry was more viscous and difficult to filter, indicating reduced conversion to the lower molecular weight liquids than was found when using full moisture lignite. Tests were made in which lignite was mildly carbonized in the presence of the liquefaction solvent or carbonized separately before liquefaction. Mild carbonization or heat-treatment of the lignite in the presence of the liquefaction solvent show slight deleterious effects on the overall yields and conversion as indicated by the following comparison:

Pretreatment Temperature °C	None	260	315	370
Yield, Wt% MAF Lignite				
Net Gas	38	44	40	33
Net Liquid	68	64	61	57
(Light Oil)	(15)	(25)	(10)	(12)
(SRL)	(53)	(39)	(51)	(45)
Net Water	-13	-15	-10	-8
Unconverted	7	7	9	18

Although somewhat lower yields of carbon dioxide were achieved and small quantities of methane were produced by carbonization, the undesirable effects of prior carbonization on the liquefaction step make the combined process unattractive and was the case with any prior thermal treatment attempted.

9. Severe Dehydration in Oil

Although prior thermal treatment was found to be disadvantageous in conjunction with liquefaction, the possibility of sufficient liquid generation suitable as vehicles for drying was suggested. Additionally, under the severe drying conditions any sulfur removal would be advantageous in subsequent processing. If such an oil regenerative process proved feasible, an oil-based drying system could be devised that would dehydrate and condition the lignite in one operation.

Chilled anthracene oil and fuel oil were used as the drying media in separate tests. A 2.6 weight ratio of drying medium (solvent) to full moisture content lignite was charged to the autoclave and a nitrogen atmosphere was maintained in the autoclave. Drying temperature was 400°F. Modifications to standard liquefaction procedure were made primarily in product work-up. Details of procedures and data have been reported (2). In summary format, the process results were:

Drying Agent	Chilled Anthracene Oil	#2 Fuel Oil
Yield, Wt%		
Gas	0.5	2.9
Liquid	-7.5	-13.3
Unconverted	101.0	106.5
Water	6.0	3.9
	100.0	100.0
Percent of Total Filtrate in Residue	13.7	6.5

The negative liquid yields indicate that in both cases the solvents have either polymerized or reacted with components of the lignite. The percentages of unconverted material greater than 100 indicate that the organic content of the coal increased because of some reaction with the solvent. In addition, the positive yields of water may be due to the thermal removal of chemically-bound water or hydroxyl groups from the coal. It seems that No. 2 fuel oil is more reactive in this regard than chilled anthracene oil. Removal of sulfur from the lignite was negligible for each test.

As there was appreciable loss of drying medium with the solid material, it appears there was no net generation of liquids so the process is not self-sustaining. The extent of solvent loss resulted in high make-up requirements that would make the process unattractive because no improvement other than moisture reduction was apparent.

10. Direct Hydrogenation of Dried Lignite

Stress-corrosion cracking of the stainless steel preheat tube and reactors in the PDU was found to be serious after limited service. The high moisture content of lignite and the presence of chlorides have been suggested as part of a possible mechanism for the rapid corrosion in the PDU operation although no evidence of accelerated stress-corrosion cracking in autoclave work had been noted. It was suggested that using dried lignite would eliminate the corrosion problem by eliminating moisture. However, with dried lignite a hydrogen atmosphere would be necessary for the hydrogenation because previous tests had shown that the carbon monoxide-hydrogen gas mixture was not effective with dried lignite. Preliminary tests were made in the batch autoclave using moisture-free lignite, solvent and a hydrogen atmosphere under otherwise standard test conditions to indicate whether sufficient conversion would occur to insure operability of the PDU. Yield and conversion data for the tests in a hydrogen atmosphere are summarized in the following together with the average baseline data for standard tests with a syngas atmosphere. (See (3) for complete details and experimental data.)

Atmosphere	H_2/CO*	H_2	H_2
Moisture	Yes	No	No
Yield, % MAF Lignite			
Gas	34.0	13.3	11.5
Liquid	64.6	59.5	52.3
Unconverted	11.4	16.0	23.7
H_2O + Ash	-10.0	11.2	12.5
Total	100.0	100.0	100.0
Light Oil	6.6	11.0	10.6
Net SRL	58.0	48.5	41.6

*Base line data.

Conversion was lower in the hydrogen atmosphere tests using dried lignite than when moist lignite and a syngas atmosphere were used. Yields of SRL were lower and light oil higher. More gas was produced in the syngas atmosphere because of carbon dioxide reacting with moisture. Although conversion of lignite was reduced in the hydrogen atmosphere with dried lignite, conversion is thought to be sufficient for operation of the PDU, especially since light oil production increased.

11. Liquefaction at Lowered Pressure

In runs in the PDU at the lower pressure of 1500 psig, there seemed to be a net production of water rather than consumption. Replicated batch autoclave tests were made at reduced process pressure to confirm this observation using a sample of the lignite charged to the continuous units (3). Results are summarized in the following:

	Standard Test	Average of Duplicates
Maximum Pressure, psig	3050	1650
Yields, Wt% of MAF Coal		
Net Gas	32	36
Net Liquid	66	31
(Light Oil)	(8)	(11)
(SRL)	(58)	(20)
Unconverted	8	26
Net Water & Ash	-6	7
Solvent Recovery %	91	85

(continued)

	Standard Test	Average of Duplicates
Product Gas Composition, Mol %		
H_2	46.3	45.2
CO_2	26.5	21.7
CO	25.1	28.0
CH_4	1.4	3.0
C_2H_6	0.4	1.2
C_3H_8	-	0.5
H_2S	0.3	0.4
Water Gas Shift Relation $\frac{(H_2)\ (CO_2)}{(H_2O)\ (CO)}$	1.00	0.60

Overall recovery was lower than usual because of the smaller quantities charged. However, the tests were generally valid. At the lower pressures, total conversion and net liquid yields and solvent recovery were significantly reduced.

The extent of the water gas shift reaction was altered at the lower pressure in that water was produced rather than consumed. Perhaps at the reduced pressure, the rate of the reaction is reduced. Changing the total pressure should not alter the equilibrium of the reaction $CO + H_2O \rightleftarrows H_2 + CO_2$. However, a lower reaction rate would reduce the hydrogen partial pressure relatively more than that caused by the reduction in total pressure, in view of the same amount of water in the gas phase.

12. Deashing of SRL

Separating mineral matter and unreacted coal from the solvent refined product is a continuing major problem. High temperature-high pressure filtration has been used in the large pilot plants. Mixing undeashed SRL (vacuum bottoms) with a light solvent at temperatures slightly lower than the critical temperature of the solvent and at a pressure above the solvent vapor pressure, followed by settling, has been suggested as an alternate deashing method and this method was investigated to a limited extent as the thesis project of a M.S. graduate (12). It was found that with benzene as a solvent at temperatures approaching the critical, and with superimposed pressures above the critical, SRL and liquefaction solvent may be dissolved and unreacted lignite and mineral matter settled out. A few experiments were conducted over a wide range of variables with the following results:

Temperature, °F	500	500	200	500	500
Pressure, psig	400	400	400	400	400
Settling Time, hrs	2	0.5	2	2	2
Benzene/Slurry Ratio	1:1	1:1	1:1	1:2	1:1
SRL Recovered, Wt %	86.5	88.1	72.4	83.8	84.7
Ash Content of SRL, Wt %	0.74	1.80	3.04	2.29	0.66

The first and last experiments above were replicates at the same conditions and the others evaluated, in turn, the effects of decreasing the settling time, decreasing the temperature, and decreasing the benzene/slurry ratio. All of these resulted in increasing the ash contents, though SRL recoveries were comparable at all but the low temperature. Complete results of these runs, taken from the thesis are included in Interim Report 4 (3).

Additional laboratory experiments were made in the autoclave using undeashed SRL or SRC as feedstocks rather than the total reactor slurry which contained liquefaction solvent as well as the solvent refined material with the mineral matter and unconverted coal. Toluene and methyl isobutyl ketone (MIBK) were investigated as solvents in addition to benzene. The results of these deashing tests are also presented in the Interim Report (3).

In the deashing tests, the light solvent and the undeashed feedstock were mixed in a batch autoclave at selected pressure and temperature and then allowed to settle. A nitrogen atmosphere was maintained in the autoclave. Mixing was a nominal half hour followed by a nominal 2.5 hours settling. Four dip tubes in the autoclave allowed the settled contents to be removed under operating pressure and temperature at various heights. Including the material below the lower tube (No. 4) termed "autoclave bottoms", five samples representative of a vertical cross section of the autoclave contents could be obtained. The sampling technique employed resulted in withdrawal of material through the sampling tubes somewhat greater or less than the actual volume above the particular withdrawal tube.

A comparison of the results with the three solvents all at 2.5 hours settling time, 4/1 solvent/SRL ratio, and 550 psig follows:

Solvent	Benzene		Toluene		MIBK
Temperature, °F	500	450	500	450	500
SRL Recovered, Wt %	46.0	59.1	65.5	65.0	58.4
Ash Content of SRL, Wt %	0.82	0.78	0.51	0.29	0.35

From these limited data, toluene appears to be the best of the three solvents, with the lower temperature giving a lower ash content. The amounts of ash are higher than desirable, but are thought to be the results of some residue being sampled with the solution because of the sampling tube locations. Generally ash contents of the SRL from the two upper sampling tubes ranged from zero to 0.2 weight percent. Toluene at 400-450°F was selected for the deashing solvent in the PDU because it seemed to be slightly more effective than benzene, and is much preferred from the viewpoint of health and safety. The MIBK is better than benzene and has about the same selectivity as toluene, with both yield and ash content being lower than with toluene.

The undeashed SRL or SRC is the vacuum bottoms remaining after flashing off the recycle liquefaction solvent. The amount of SRL in the vacuum bottoms is determined by pyridine extraction, the extracted material being considered the SRL. However, as SRL is extracted from the vacuum bottoms with benzene or toluene, there appears to be relatively even less remaining. That is, the amount of SRL extracted plus the pyridine solubles remaining in the extraction residue is less than the pyridine solubles determined on the original vacuum bottoms. In the preceding tabulation the fraction of SRL recovered is based on the pyridine solubles in the charge (input basis). If the fraction of SRL recovered is based on the amount recovered plus the pyridine solubles remaining in the residue (output basis), a higher recovery is calculated. This is indicated in the following table for the same runs summarized previously:

Solvent	Benzene		Toluene		MIBK
SRL Recovered, Wt %					
Input Basis	38.3	59.1	65.5	65.0	58.4
Output Basis	39.6	65.9	75.9	73.7	55.4

This desolubilization effect with benzene and toluene is quite interesting, and should be kept in mind because it could result in somewhat misleading data and erroneous conclusions. It is also interesting that it apparently did not occur with MIBK, which, however, was not as effective a solvent as toluene.

Samples of undeashed solvent refined coal (SRC) obtained from pilot plants at Tacoma (Fort Lewis), Washington and Wilsonville, Alabama, were extracted with toluene in the autoclave. Results with the Tacoma SRC were so favorable that an additional experiment was made at lower temperature and pressure. The results are compared with those of SRL in the table following.

Feed	F-1 Btms	Tacoma SRC		Undeashed Wilsonville SRC
Temperature, °F	500	500	300	500
Pressure, psig	550	550	100	550
% SRL or SRC in Feed				
Input Basis	68	73	73	76
Output Basis	60	65	63	63
% of SRL or SRC Recovered				
Input Basis	66	74	68	71
Output Basis	76	83	78	86
% of Original Ash in Product	1.61	0.06	0.75	1.71
Ash Content of Product, %	0.51	0.02	0.27	0.38

Both samples of undeashed SRC resulted in higher yields of deashed product of lower ash content than was achieved with undeashed SRL. With the undeashed Tacoma SRC at the lower temperature and pressure of 300°F and 100 psig, respectively, the yield of deashed product was almost as high, but the ash content increased from 0.02 percent to 0.27 percent, which still may be satisfactory. Further studies to optimize this deashing process for SRC might result in as high yield and as good quality as is realized by filtration.

Samples of undeashed vacuum bottoms from the PDU were occasionally worked up by dilution with liquefaction solvent followed by hot filtration through filter paper in a Buchner funnel. One study was made of the effect of dilution, that is, of the ratio of solvent to vacuum bottoms employed. There appears to be a maximum in yield at about 3:1 ratio of solvent to vacuum bottoms.

A comparison of the results obtained by extraction-settling and by hot filtration is shown in the table following:

Operation	Extraction		Hot Filtration		
Solvent	Benzene	Toluene	←FS-120→		
Solvent/VB Ratio	4:1	4:1	2:1	3:1	4:1
Temperature, °F	450	450	400	400	400
% SRL Recovered					
Input Basis	59	65	70	86	84
Output Basis	66	74	75	86	87
% Ash in SRL	0.78	0.29	0.04	Nil	0.25

Too much attention should not be given to the ash contents of the filtered SRL, because these are functions of the laboratory filtration techniques more than of the type of operation. Further, there is little room for increasing recovery by filtration, though more thorough washing and blowing in commercial filtration might reduce the oil content of the filter cake from the 60 to 75 percent realized in the laboratory to perhaps 50 percent. On the other hand, the solvent extraction and settling operation has the potential of being greatly improved by employing extraction and countercurrent stripping in a continuous unit.

13. Miscellaneous Studies

Several miscellaneous investigations associated with batch autoclave work are summarized under the following headings:

a. Reaction Time

In operation of the autoclave, the fastest that the batch operation can be cycled includes a minimum of 0.5 hr at reaction temperature. To reconfirm that 0.5 hr was sufficient time for the hydrogenation-solution reactions, a comparison was made with runs under otherwise identical standard conditions except for the time the reactor was at reaction temperature.

Run	422	418
Time, hr	0.5	2.0
Yields, Wt % of MAF Coal		
Net gas	35.2	49.3
Net liquids	68.7	56.8

These data indicate that longer time at reaction temperature results in more decomposition of the lignite which increases gas yield and reduces liquid yield. Decomposition of water allows total yield of gases and liquids to exceed 100 percent on an MAF lignite basis for both reaction times. The hydrogenation-solution process is apparently rapid with high liquid yields occurring at the shortest reaction time practical with the equipment used. Thus the 0.5 hr at reaction temperature was used as the standard test time at reaction temperature.

b. Particle Size

The standard testing procedure established required that the lignite be pulverized to -100 U.S. mesh before liquefaction as it had been postulated that such a small particle size would be necessary to insure rapid reaction. To check this assumption, a test was made with a larger top size, 0.25 inches, under the otherwise standard liquefaction procedure. The results of this run are compared with the average of three standard runs in the summary following:

Lignite size	-100 mesh*	-0.25 in.
Yields, Wt % of MAF Coal		
Net Gas	43	45
Net Liquid	62	61
(Light Oil)	(14)	(17)
(SRL)	(48)	(44)
Net Water	-13	-11
Unconverted	8	5

*Average of three tests.

These data showed that liquefaction is essentially the same under the employed test conditions either with an initial particle size of -100 mesh or -1/4 inch. Therefore, in pilot plant operation, it should not be necessary to pulverize the lignite completely to -100 mesh to have satisfactory liquefaction.

c. Liquefaction of Phenol Pretreated Lignite

Coals when pretreated with some organic solvent-type compounds undergo a partial dissolution indicated by swelling. Phenol acts in this fashion with lignite. To find out if this pretreatment would be beneficial in liquefaction of lignite, some phenol treated lignite was tested under otherwise standard conditions (3).

Full moisture lignite was treated with aqueous phenol, ranging from 30 to 100 percent phenol. In one instance, phenol was added directly to the liquefaction autoclave just prior to liquefaction. Generally, following the phenol treatment, the lignite was washed with warm water and followed by a light rinse with ambient temperature water to remove excess phenol. After the rinse, the lignite was processed using normal liquefaction procedures. A standard liquefaction test serves as baseline data for the comparison. Experimental conditions and yield data on a MAF lignite basis are summarized in the following:

Run No.	PTR-1	PTR-2	PTR-3	PTR-4	PTR-5	PTR-6	PTR-7
Yields, Wt % MAF Lignite							
Net Gas	47.9	39.4	40.4	42.1	56.3	37.0	41.4
Net Liquid	64.2	52.0	62.2	56.2	62.9	68.6	77.4
Unconverted	6.1	10.1	8.8	6.2	5.5	7.1	6.7
Net H_2O + Ash	-18.2	-1.5	-11.4	-4.5	-24.7	-12.7	-25.5

Conditions: PTR-1, standard test
PTR-2, 100% phenol treat, 212°F wash, light rinse
PTR-3, 100% phenol treat, 140°F wash, light rinse
PTR-4, 100% phenol treat, ambient wash, light rinse
PTR-5, phenol treat, crystals added
PTR-6, 100% phenol treat, light rinse
PTR-7, 30% phenol treat, light rinse

Data indicate little or no improvement in basic yields as a result of phenol treatment except in Run PTR-7. Total conversion as indicated by the extent of unconverted lignite is similar to the differences obtained in replicate runs. The test with phenol pretreatment using a 30 percent slurry of phenol followed by a light rinse did indicate a higher yield of net liquids and higher loss of net water plus ash. However, values may not be comparable because of the problems of analyzing for phenol in aqueous and organic media.

d. Variability in Standard Lignite

Analytical data have been accumulated on the lignite charged to the batch autoclaves in the laboratory liquefaction program. Lignite source was North American Coal Company's Indian Head Mine near Zap, North Dakota, in Mercer County. Bulk samples were obtained and carefully stored with small portions removed, pulverized, and used in liquefaction tests. Partial proximate and ultimate analyses were made for each run. Data from 67 analyses were used to calculate the variability introduced by different samples and different chemists. Results are summarized in the following:

Proximate Analysis, Wt %	Average	Standard Deviation
Moisture	28.5	1.82
Volatile matter	31.4	1.70
Ash	6.67	0.683
Ultimate Analysis, Wt % MAF		
Carbon	64.5	1.45
Hydrogen	4.27	0.261
Sulfur	0.873	0.234
Nitrogen*	0.798	0.129

*Only 12 values used in calculation.

The results indicate a relatively limited variability in the analyses. The standard deviations reported are for a single value, not of the average. Moisture content is low because there was loss during pulverization to -100 mesh and preparation of the sample. Both moisture and ash contents are generally considered to be the more variable of the analyses. More uniformity is exhibited in the moisture-ash-free ultimate analyses reported. Of these, sulfur is usually the most variable.

D. CONVERSION OF SOLVENT REFINED LIGNITE TO LIGHT LIQUIDS

Under a subcontract to Project Lignite, faculty of the Department of Chemistry of the University of North Dakota investigated the catalytic conversion of SRL to light liquids in a two phase program consisting of characterization of SRL and SRC and of their conversion. Details are given in Interim Report No. 3 (11).

In the conversion studies, nearly ninety batch autoclave hydrogenation runs were performed to determine the effects of solvents, catalysts, temperature and pressure on conversion of SRL and SRC to distillable liquids and gases. NMR, elemental analysis, titration, and solubility of vacuum bottoms were used to evaluate conversions and give insight into the hydrogenation reactions. The following general conditions were established for the catalyst comparisons: an initial hydrogen pressure of 2500 psi, 50 percent catalyst by weight, 150 ml tetralin solvent, and 375°C reaction temperature. Under these conditions, a total of eight catalysts were tested with SRL. Studies were also conducted of the effects of catalyst pretreatment, temperature, pressure, type and amount of solvent, and of the hydrogenation of solvent refined lignites and solvent refined coals from various sources.

The hydrogenation experiments revealed several interesting results. One of these is that the initial ratio of solvent (tetralin) to SRL used influenced the overall conversion. Maximum conversion of 93 percent was obtained with a solvent to SRL ratio of 1:1. The conversion decreased slightly when the ratio was increased or decreased from 1:1. Catalyst conditioning in a hydrogen atmosphere before use promoted hydrogenation and increased overall conversion slightly. Also, it was demonstrated that SRC could be converted with almost equally high conversion under experimental conditions developed for SRL conversion.

More specific conclusions from the conversion phase studies are:

1. The catalysts Ni-Mo-Al_2O_3 (Harshaw HT-100) and $SnCl_2$ gave the highest conversions of SRL to distillable liquid products, though the $SnCl_2$ removed little nitrogen as ammonia.

2. Presulfided catalysts gave generally high conversions. Presulfided Ni-Mo-Al_2O_3 was more effective in removing heteroatoms of nitrogen and oxygen in addition to giving a high conversion, and was judged the best catalyst overall of those tested.

3. The conversion of SRL to gases and distillable liquids increased almost linearly with reaction temperature over the range studied and reached 88 percent at 450°C.

4. The conversion of SRL increased as the initial reactor pressure was increased from 1000 psi to 1750 psig, but further increases to 2500 psig were not beneficial.

5. Tetralin is a good solvent for SRL conversion, but a relatively high conversion is obtained in the absence of solvent.

6. Solvent refined coals, regardless of their sources, were converted to gases and distillable liquids in relatively high yields using reactor conditions developed for SRL conversion.

7. The experimental conditions for the most effective conversion are: initial hydrogen pressure of 2500 psig, 75g of SRL, tetralin solvent with a solvent to SRL ratio of unity, presulfided HT-100 catalyst, 450°C, and two hours reaction time.

The solubilities of SRL in about 40 organic solvents were determined during the characterization work. The solvents possessing both polar and aromatic groupings resulted in higher solubilities.

From determinations of basic nitrogen, it was inferred that some of the nitrogen content of the SRL came from the solvent used in its preparation, and that, to produce SRL with the lowest nitrogen content, the solvent for SRL formation must also be low in nitrogen. Chemical and physical characteristics determined for SRL and SRC indicated some definite differences in chemical composition.

Conclusions concerning an average molecule of SRL are that: the average molecule contains several aromatic rings, some condensed, an average of 0.8 acidic groups, mainly as phenols, and an average of 0.4 basic nitrogen atoms. Fixed in the non-acidic or non-basic structures are 1.8 atoms of oxygen and 0.37 atom of nitrogen per average molecule as determined by elemental analyses and titration data. In this matrix of aromatic rings, oxygen and nitrogen functions are connected by a smaller but significant number of aliphatic carbons.

VI. ECONOMIC STUDIES

A. INTRODUCTION

Concurrently with experimental work, a series of economic studies was made to estimate the selling price of products in conceptual commercial installations. Estimates were made of plant investment and annual operating costs including a unit cost sensitivity analysis. Cost comparisons were made with other coal technologies and the economics for principal uses of SRL were also investigated. The general purpose of the relative product prices was for comparison with current fuel costs. The following sections are summaries of the various reports, most of which were previously reported in quarterly or monthly reports of the project and are indicated by title in the list of references (5,16-23).

B. METHODOLOGY

Initially, the technological requirements for the process were determined and sizes of capital equipment necessary estimated based in part on published OCR, ERDA and other reports describing similar technologies. Material and labor requirements were determined by scaling to the desired capacity from a base output level capital. Operating costs were estimated based on the technological requirements with process area costs scaled by the six-tenths power law of capacity when unit capacities were changed without an increase in number of units. Scaled capital costs were adjusted to those at the time of the estimate using an escalation factor taken from the CE plant cost index. Labor and material costs were those prevailing in North Dakota during the year of the estimate.

Estimated plant costs were processed using a discounted cash-flow (DCF) computer program to arrive at a base unit cost. Sensitivity of the unit cost to variations in the DCF rate, lignite costs and labor costs were made.

C. A COMPARATIVE ECONOMIC EVALUATION OF TWO PLANTS FOR THE SOLVENT REFINING OF LIGNITE

The purpose of this study, one of the first completed, was to obtain product costs for liquefaction of lignite to produce approximately 9000 tons of SRL per stream day plus nearly 3000 tons of light oil. One conceptual design was a scaling of a standard solvent refined coal plant (SSRL) to operation with lignite at the capacity specified from data presented in OCR R&D Report No. 53, Interim Report No. 1.

The design was additionally modified in that the reactant gases were produced from gasification of lignite in a Lurgi plant (SRLL) rather than from reforming of methane (natural gas). Data for Lurgi gasification were taken from published U.S. Bureau of Mines reports. A comparison of cost for the two processes is for mid-1974 prices.

Item	$000 (1974 Prices) SSRL	 SRLL
Total Investment	236,711	236,713
Total Direct Production Costs	63,924	40,434
Total Operating Costs	80,635	57,307
Total By-Product Credits	3,506	2,413
Total Operating Costs after Credits	77,129	54,844

Total investment for the two plants is essentially the same. Despite the requirement of additional lignite (35,000 tons per stream day for the SSRL plant vs. 39,000 tons/ stream day for the SRLL unit), total operating costs are lower for the SRLL unit because natural gas did not have to be purchased at an assumed cost of 60¢/MM BTU. This translates into a lower selling price for products.

Discounting the cost data using the DCF method at a rate of return of 12 percent resulted in required composite selling price for products of 82.9¢/MM BTU ($28.51/ton) for the SSRL plant and 65.1¢/MM BTU ($22.46/ton) for the SRLL unit. Assuming a product distribution ratio of 3:1 for SRL and light oil gives a selling price of $26.53/ton for SRL and $34.82/ton for light oil vs. $20.83/ton for SRL and $27.34/ton for light oil, respectively, for the SSRL and the SRLL units.

Sensitivity analysis showed the required selling price for these capital intensive processes to be more influenced by the required rate of return and by the price for lignite than by labor costs. The effect of rate of return on selling price is:

Required DCF Rate Percent	¢/MM BTU (1974 Prices) SSRL	 SRLL
10	76.2	58.6
12	82.9	65.1
14	90.0	72.1

While for lignite cost the effect is:

Lignite Cost, $/Ton	¢/MM BTU (1974 Prices) SSRL	 SRLL
0	63.1	43.7
2.025	80.9	63.0
2.25	82.9	65.1
2.475	84.9	67.3

In all cases, for otherwise similar assumptions, significant savings are possible if the required make-up process reducing gas is produced from gasification of lignite rather than from reforming purchased natural gas. Product costs are also indicated to be higher than those for petroleum products or natural gas at the time of the estimates.

D. AN ECONOMIC EVALUATION OF A LURGI GASIFICATION SYSTEM FOR PRODUCING LOW-BTU GAS FROM LIGNITE

The carbon monoxide plus hydrogen content of product gas (synthesis gas) from coal gasification is suitable for use as the reacting gases in the liquefaction of lignite. The objective of this economic analysis was to evaluate the costs associated with a slagging fixed-bed Lurgi gasification system to produce 121 million SCFD of 350 BTU/SCF gas. A slagging Lurgi would have a gas making capacity advantage over a dry ash unit.

Plant requirements and operating data were estimated from U.S. Bureau of Mines reports, R.I. 7408 (1970) and Special Report 70-21 Case III (1970). Approximately 3,000 tons of lignite would be gasified per stream day.

Total investment was estimated at $21 million (1974 prices), and it was assumed to be 100 percent equity financed with a 12 percent required rate of return. Feed lignite and labor were the major operating expenses with the total annual operating costs being $5.6 million before taking credit for by-products ($65 thousand a year). Application of the DCF method for the base case of a 12 percent rate of return results in a selling price of 68¢/million BTU both for the product gas and by-product tar.

Sensitivity of product costs to variation in rate of return, price of lignite, and labor cost at 1974 prices are illustrated in the following tabulations:

DCF Rate Percent	Product Cost ¢/MM BTU
10	61.3
12	68.4
14	76.0

Lignite, $/Ton	Product Cost ¢/MM BTU
0	49.0
2.025	66.5
2.25	68.4
2.475	70.4

Labor Cost*	Product Cost ¢/MM BTU
0.90	67.7
1.00	68.4
1.10	69.2

*Base Case (1.0) is $935.7 thousand including direct maintenance, and indirect labor plus supervising and technical services.

Typical of high investment operation, the required selling price of product is influenced most by the necessary rate of return followed by the price of the feed lignite. While labor cost does affect the selling price, the change is not as pronounced. While higher than the regulated price of natural gas at the time of the economic evaluation on a heating value basis, the product price is low (see following section).

E. REVISED ESTIMATE OF THE COSTS OF A PLANT FOR PRODUCING LOW-BTU GAS FROM LIGNITE

Additional data for gasification of lignite became available with the FPC filing for the ANG Coal Gasification Company which allowed better estimates of plant requirements including necessary controls to meet environmental regulation and for recovery of sulfur. Using this information together with that obtained in the first economic evaluation of production of low BTU gas, the original estimates were revised.

The major change was a large increase in total investment from \$21 to \$90 million for the same size of installation. Annual operating costs were \$15.8 million before taking a by-product credit of \$20 thousand. Assuming 100 percent equity financing and 12 percent DCF rate of return for the base case, the estimated selling price of the joint product was \$2.34/MM BTU. The capital and annual operating costs were over three and a half times that of the earlier estimate which resulted in the increase in unit costs from 68¢ to \$2.84 per million BTU. Determining the response of product price to variations in the basic parameters of DCF rate, lignite price and labor costs showed that the required rate of return had the greatest effect as illustrated in the following:

DCF Rate Percent	Selling Price \$/MM BTU (1974 Prices)
10	2.09
12	2.34
14	2.62
20	3.54

Changing lignite price from zero to \$2.475/ton increased the cost by 21¢/MM BTU while a 20 percent variation in labor cost varied the selling price by only 6¢/MM BTU.

F. AN ECONOMIC EVALUATION OF A PLANT FOR THE SOLVENT REFINING OF LIGNITE

A study was made of a plant for processing 30,000 tons/stream day of North Dakota lignite by liquefaction to 8,850 tons of SRL and 2,200 tons of light oils to determine the effects of rapidly increasing inflation and of environmental control requirements. Major plant components were liquefaction and filtration, gasification, effluent control and oxygen production. Basis for the plant was OCR R&D Report No. 82, Interim Report No. 1 (1973) together with process data from Project Lignite.

Total investment was estimated at $560 million which was assumed to be 100 percent equity financed. Cost of lignite, labor and depreciation are the major items in the annual operating costs of $88 million. Using a DCF rate of 12 percent gave a joint product price of $1.55/MM BTU. A summary of costs using mid-1975 prices is:

Item for Costs	$000
Total Plant	469,000
Start-up	14,070
Depreciation Subtotal	483,070
Total Investment	559,570
Total Production	51,436
Total Operation	87,909

Variation of the basic parameters of rate of return, lignite price, and labor cost in the sensitivity analyses again indicated the major effect of the large investment required. Varying the rate of return from 10 to 20 percent increased the product selling price from $1.40 to $2.20/MM BTU. Assuming lignite prices from zero to $3.85/ton varied product cost from $1.27 to $1.58/MM BTU while a 20 percent change in labor cost increased the selling price by $0.02/MM BTU.

G. AN ECONOMIC COMPARISON OF THREE COAL LIQUEFACTION PLANTS FOR PRODUCING 9,000 TONS OF FUEL PRODUCT PER DAY

The processes evaluated were Synthoil using Wyodak coal, Clean Boiler Fuels from Coal (CBFFC) also using Wyodak coal, and Solvent Refined Lignite (SRL). Products from the processes differ with a heavy fuel oil the major product from the Synthoil process, two grades of fuel oil plus naphtha from the CBFFC, and SRL plus light oil from lignite liquefaction. Data were taken from ERDA Report No. 76-35, "Economic Analysis of Synthoil Plant Producing 50,000 Barrels per Day of Liquid Fuels from Two Coal Seams: Wyodak and Illinois No. 6," 1976 as a basis. Requirements were adjusted to the size of plant being considered.

Estimated total investment and annual operating costs are summarized in the following for mid-1975 prices:

	$000		
Item	CBFFC	Synthoil	SRL
Total Capital Investment Cost	686,283	640,868	569,432
Total Production Cost	102,966	96,096	79,327
Total Annual Operating Costs	162,042	151,054	127,823

Differences in capital investment are caused by variation in the number and size of the process areas in each plant. For example, the CBFFC plant employs hydrogenation, shift conversion, and methanation which are not necessary in a SRL plant. The technology of Synthoil process is quite different as hydrogenation and hydrogen production are more important process areas; they account for nearly half of the plant investment.

Annual operating costs are highest for the CBFFC facility and lowest for the SRL plant, chiefly because of differences in price of feed coal. All items in the annual operating cost figure for the Synthoil process are the highest, except for the feed coal cost which is approximately 32 percent higher than for SRL and 9 percent lower than for CBFFC plant. Price of lignite has a significant effect on annual operating cost.

Unit cost of the product is also significantly influenced by the required DCF rate as shown in the following tabulation:

	Product Unit Cost, $/MM BTU		
DCF, Percent	12	15	20
Plant			
SRL	2.33	2.56	3.14
CBFFC	2.59	2.93	3.56
Synthoil	2.51	2.80	3.45

Unit product cost for the SRL process, on a heating value basis, is honest; however, the major product is solid rather than liquid. Adding a hydrogenation unit to the SRL plant to give greater yields of liquid products would result in nearly the same product from the three processes. However, there is a possibility that the SRL process has an advantage in cost over the others for production of boiler fuels.

H. AN ECONOMIC ANALYSIS OF A POWER PLANT BURNING LIGNITE AND A POWER PLANT BURNING SOLVENT REFINED LIGNITE IN A NORTH DAKOTA AND A TWIN CITIES SETTING

The economic comparison is between the existing commercial process of electrical power generation using lignite for fuel and the emerging technology of liquefaction to produce a higher quality fuel that is more environmentally acceptable. Potential savings from reduced boiler size because of the higher heating value of SRL and lower gas clean-up costs due to the reduced sulfur and ash contents would aid in offsetting the higher price of the SRL.

The situations consisted of 474-MW power plants at a North Dakota mine with one burning as mined lignite and the other SRL. The output was to be transmitted by a 250 KV direct current line to the twin cities of Minneapolis, and St. Paul, Minnesota. The two other cases were 440 MW plants in the Twin Cities area with both the lignite and SRL transported by rail to the plants. The higher capacity of the North Dakota plants is necessary to compensate for transmission line losses and to provide the same quantity of energy at the load center. A 90 percent load factor was assumed, plant investment was depreciated by "sum of years digits" over the first 15 years of the 25 year plant life, and the plants were 100 percent equity financed. Costs are summarized in the following at mid-1975 prices:

	North Dakota		Twin Cities	
Total Costs, Thousands $	Lignite	SRL	Lignite	SRL
Capital Investment	333,608	267,247	213,491	142,993
Plant Production Cost	15,417	111,843	33,724	102,630
Transmission Cost	262	262	---	---
Fixed Charges	19,379	14,560	12,258	7,625
Annual Operating Charges	35,058	126,665	45,982	110,255

The higher investment for the North Dakota plants is cuased by the need for transmission lines and by the increased plant size. The differences between SRL and lignite plants is caused by increased environmental control necessary for the lignite-fired plants. Production costs for the SRL-fired units is due almost entirely to the cost of the fuel. Production costs for a lignite fueled power plant in the Twin Cities area is approximately twice that in North Dakota because of transportation costs.

Translating these data into units of cents per KWH using the DCF method gives the following results depending on the required rate of return.

Plant	Unit Price, Cents/KWH (1975)					
	Lignite			SRL		
DCF Rate, Percent	12	15	20	12	15	20
North Dakota	2.24	2.74	3.67	4.67	5.07	5.82
Twin Cities	2.12	2.44	3.03	4.00	4.21	4.61

In all cases a plant fueled by SRL produced electricity at a substantially higher cost than one using lignite. For 100 percent equity financing the lowest unit cost is realized when operations are near the load center because of the I^2R losses and transmission line costs. Use of SRL in an urban setting may be favored because of low sulfur and ash content if environmental restraints continue or are increased. Additionally, low sulfur fuels are limited in availability and may not fill increasing demands for environmentally acceptable fuels.

I. PROCESS DEVELOPMENT OF LIGNITE LIQUEFACTION

Cost estimates for the production of SRL updated to mid-1976 prices were presented at the 1977 Lignite Symposium with total plant investment being estimated at $500 million and total capital requirements at $602.5 million. Total annual operating costs excluding depreciation was $94.3 million. Cost for lignite at $4/ton was the largest operating item of $38.7 million. Investment was assumed to occur on a 11:33:56 ratio for a three year construction period, and were to be 100 percent equity financed. The sum-of-the-years digit method was used to depreciate investment over 16 years of the 20 years operation. On a DCF rate of 15 percent the unit cost of product was calculated as $2.46 per million BTU.

Sensitivity analysis was done to consider the effects of inflation on plant investment initially at $500 million with the following results:

	Millions of Dollars		
Year of Start-up	1980	1985	1990
Escalation Rate, Percent			
3	554.4	642.8	767.6
5	593.1	757.0	1,014.4
7	633.6	888.6	1,333.6

By 1980, the plant investment could be 11 to 27 percent higher than at present depending on whether the inflation rate was 3 or 7 percent. Even with a moderate inflation rate of 5 percent, the required plant investment would double in 14 years.

Combining inflation with variation in DCF rate of return, the following illustrates the effect for 5 percent increments on unit costs.

	Unit Cost, $/Million BTU		
Escalation at 5 percent to:	DCF Rate, Percent		
	10	15	20
1980	2.32	2.91	3.59
1985	2.97	3.72	4.58
1990	3.97	4.98	6.13

Thus, DCF rate of return and inflation are major items in higher costs for the product in capital intensive liquefaction operations.

J. ECONOMIC EVALUATION OF A HYDROCRACKING PLANT FOR UPGRADING SOLVENT REFINED LIGNITE

Increased interest in light products resulted in an economic evaluation of a hydrocracking unit for upgrading 7,200 tons of SRL per day to a liquid product equivalent to gasoline and fuel oil plus fuel gas products. Products would be similar to those from liquefaction processes such as Synthoil. The facility would consist of the two major operations--hydrogen production and hydrocracking.

Total plant costs at mid-1976 prices were estimated at $188.5 million of a total capital investment of $229.1 million. Total annual operating costs were $222.7 million excluding depreciation. Major operating costs were for purchase of SRL ($84/ton) and lignite ($4.00/ton) and totaled $204.8 million a year. Assuming plant construction over a three year period on a 11:33:56 schedule and a plant life of 20 years, the unit cost of products depending on the required rate of return were:

	Cost $/Unit		
Unit	12	15	20
Ton	113.15	118.16	127.38
10^6 BTU	3.57	3.73	4.02

A five percent increase rate of return results in approximately a 30¢/million BTU increase in unit cost.

This study has not been published previously, and is presented in the attached Appendix IX.

VII. CONCLUSIONS AND RECOMMENDATIONS

The following general conclusions resulted from this work:

1. Extended operation with recycle of lignite-derived solvent indicated some continued improvement in conversion with cumulative time of operation. Sufficient lignite-derived solvent was generated to support continuous operation. The longest period of operation completed was 150 hours on stream charging lignite.

2. Optimum conditions for operation of the liquefaction section of the PDU as constructed were 2500 psig reactor pressure, 823-830°F maximum reactor temperature, 1.4 to 1.5 superficial liquid hourly space velocity, about 350 superficial gaseous hourly space velocity, and a solvent-to-coal ratio of from 1.4/1 to 1.8/1 with the lowest practical ratio being desirable as limited by handling of the feed slurry and the product slurry or vacuum bottoms. In the early operations it appeared that higher temperatures gave higher conversions, but the later runs demonstrated that higher temperatures (above about 830°F) resulted in increased pyridine insoluble material because of polymerization of the solvent or condensation of the solvent with the coal matter.

3. Integrated operation of the deashing section with the liquefaction section was plagued with mechanical problems. Limited operation indicated separation was achieved and low ash content material was produced initially, though the ash content soon rose to undesirably high levels.

The following is a brief summary of the recommendations that have been made for a continuation of the PDU program:

The continuation should provide for extended runs in the liquefaction section of the Process Development Unit together with operation both of the solvent recovery and recycle section and of the gas cleanup and recycle section, the evaluation of the solid-liquid separation system, corrosion coupon testing under reaction condition, the determination of solids accumulation in the dissolvers, the production of clean solvent refined lignite, and data collection and analysis.

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IX. APPENDIX: ECONOMIC EVALUATION OF A HYDROCRACKING PLANT FOR UPGRADING SOLVENT REFINED LIGNITE

A. INTRODUCTION

This is an economic evaluation of a hydrocracking facility designed to upgrade 7200 tons of solvent refined lignite (SRL) per day into gasoline, fuel oil, and other HHV fuels. The facility consists of two major operations, namely hydrogen production and hydrocracking, and is entirely self-contained except for water and electric power requirements. Refining SRL in this fashion yields products similar to those produced by other liquefaction technologies like the Synthoil process developed by ERDA.

Total capital invested is estimated at $229 million and assumed to be 100% equity financed. Annual operating costs are $223 million before depreciation charges. SRL and raw lignite are principal operating cost items and are purchased at $84 per ton and $4.00 per ton respectively. Annual output is 2.4 million tons of fuels having a combined heating value of 75 trillion BTUs. When these data are processed by a discounted cash flow computer program (DCF) at a 15% rate of return on equity, a cost of $118 per ton or $3.73 per million BTU is obtained.

B. PROCESS DESCRIPTION

Crushed lignite is fed into a gasifier where it reacts with steam and oxygen producing a hydrogen-rich gas. The gas passes through two shift-conversion units and a purification unit which remove substances like CO_2 and COS converting the gas into pure hydrogen. The hydrogen is compressed and then sent to the hydrocracking plant where it reacts with SRL converting SRL into a variety of fuels ranging from CH_4 through heavy fuel oil.

C. CAPITAL INVESTMENT

Table 1 shows capital investment as $229 million. The investment figures include total plant costs, capitalized start-up costs, return on equity during construction, and working capital but exclude plant-site costs and contingencies.

D. ANNUAL OPERATING COSTS

Annual operating cost before depreciation is $223 million with the cost of SRL the largest item ($199 million). Over

Table 1

Economic Summary
(Mid-1976 Prices)

Investment	$MM
Total Plant Cost	188.5
Start-up Cost	5.7
Depreciation Subtotal	194.2
Return During Construction	15.9
Working Capital	19.0
Total Capital Requirement	229.1
Annual Operating Cost	
Lignite (178 tons/hr @ $4.00/ton)	5.6
SRL (300 tons/hr @ $83.84/ton)	199.2
Direct Labor (360 man-hours/day @ $6.00/hr)	.8
Maintenance Labor & Material	6.8
Supervision & Payroll Overhead	1.0
Other Direct Production Costs	1.8
Administrative & General Overhead	3.6
Taxes & Insurance	3.9
Total Annual Operating Cost (excluding depreciation)	222.7

1.4 million tons of lignite are purchased annually for the hydrogen plant at a cost of $5.6 million. The yearly direct-labor requirement is over 131 thousand man-hours involving an outlay of $800 thousand. It is assumed that 2 maintenance men per direct laborer are needed and are hired at $15,000 per year. The cost of maintenance material is taken as 3% of plant cost. Payroll overhead includes such items as payroll taxes and fringe benefits, and other direct production cost items are water, electricity, and chemicals. Property tax and insurance are considered fixed charges and are taken as 2% of plant cost. Depreciation is calculated by the sum-of-the-years-digit method over the first 16 years of plant life and is excluded from the operating cost items shown in Table 1.

E. UNIT COST

Plant construction is assumed to occur over three years on an 11:33:56 percent schedule. Also, the operating life of the plant is assumed to be 20 years. Given these assumptions, processing the above data by a DCF-computer program at 12, 15, and 20 percent rates of return yields the following unit costs:

Table 2

Costs per Ton and 10^6 BTU at DCF-Rates of 12, 15, & 20 Percent

Item	12	15	20
Ton	113.15	118.16	127.38
10^6 BTU	3.57	3.73	4.02

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