SECTION 3

PROCESS INFORMATION EXTRACTS

INDEX

PROCESSES PAGE NO.

FIGURES

INDEX (Contd)

INDEX (Contd)

INDEX (Contd)

PROCESS INFORMATION EXTRACT

PROCESS 1.1.1 Solvent Refined Coal (SRC): SRC I and SRC II

Process Type: Hydroliquefaction

Main Products:
SNG
LPG - C_3, C_4
Light Distillate Oils
Heavy Distillate Oils
SRC

Development Status: Pilot Plant

PROCESS DEVELOPERS(1)

(1) Pittsburgh and Midway Coal Mining Company (PAMCO), a subsidiary of Gulf Oil Co.:
PAMCO operates a 48-TPD pilot plant located at Fort Lewis, Washington and a bench scale PDU located at Merriam, Kansas.

(2) Catalytic, Inc. (Southern Company Services, Manager):
Catalytic operates a 6-TPD pilot plant Located at Wilsonville, Alabama.

(3) Colorado School of Mines (CSM):
CSM operates laboratory-scale units at Golden, Colorado.

(4) Hydrocarbon Research, Inc. (HRI):
HRI operates laboratory-scale units at Trenton, N.J.

PROCESS SPONSORS

DOE sponsors development work performed by PAMCO and CSM.

DOE and EPRI cosponsor development work performed by Catalytic, Inc.

EPRI sponsors development work performed by HRI.

PROCESS DESCRIPTION

The SRC process can be described as having two different modes of operation: SRC I and SRC II. Each mode is described as follows:

(1) SRC I

Figure 1 shows the SRC I process. Dried and pulverized coal feed and a filtered process solvent recycle (typically a 500 - 850°F boiling-range, coal-derived liquid) are mixed to form a slurry in the slurry mixing vessel. The solids content is kept at a maximum level, usually less than 50 wt%, so that the slurry remains pumpable. The slurry is pumped from the mixing vessel and the recycle plus make-up hydrogen is injected in the stream ahead of the slurry heater. The entire mixture is heated to about 700°F in the heater and then enters the dissolver. The reaction is highly exothermic and heats the reactor contents to over 800°F, depending on the degree of hydrogenation. The reactor effluent is then let down in pressure and cooled in several flash separations in the vapor-liquid separator section. The gases separated are sent to an acid gas removal section to remove CO_2 and H_2S gases that have been formed by the reaction. The cleaned gas is sent to H_2 purification where the C_1 - C_3 vapors are separated from the recycle H_2 stream. The bottoms of the vapor-liquid separation are sent to the filtration section to remove the mineral residue, which contains the ash, and the undissolved organic material of the coal. The filtrate is sent to the vacuum fractionation section to vacuum-distill the distillates overhead from the SRC. The distillates are further fractionated to a light oil (C_4 - 400°F); a light distillate (400 - 500°F) or wash solvent, and a heavy distillate fuel or filtered process solvent recycle (500 - 850°F). Wash solvent is recycled to the filtration section and the filtered process solvent is recycled to the slurry mixing vessel.

(2) SRC II

Figure 2 shows the SRC II process. Dried and pulverized coal is fed to the slurry mixing vessel, where it is mixed with a slurry recycle stream and a filtered process solvent recycle. The slurry recycle is a flashed stream from the vapor-liquid separation section containing dissolved and undissolved coal, including the coal ash. The mineral contents of the coal act as a catalyst in the dissolving, hydrocracking, and hydrogenation reactions; hence, for the same residence times, the degree of reaction, as measured by the hydrogen consumption, is substantially increased in the SRC II mode, or slurry recycle mode, over the SRC I mode. The entire slurry is pumped from the mixing vessel and the recycle plus make-up hydrogen is injected in the stream ahead of the slurry heater. The entire mixture is heated to about 700°F in the slurry heater and then enters the dissolver.

Reaction heat increases the temperature of the reactants to over 800°F. The reactor effluent is then let down in pressure and cooled in several flash separations in the vapor-liquid separation section. The gases separated are sent to an acid-gas removal section for CO_2 and H_2S removal and a H_2 purification section to recover C_1 - C_3 and purify the recycle H_2. Part of the slurry stream is recycled to the slurry mixing vessel; the rest is filtered to separate the mineral residue from the coal solution. The filtrate is vacuum-distilled to distillate fuels overhead and SRC bottoms. The distillates are fractionated into the light oil, light distillate, and heavy distillate streams, including the wash solvent and filtered process solvent recycle.

The downstream process configuration for the SRC II process could be adjusted to selectively eliminate the SRC as a product.(2) This process scheme is shown on Figure 2, with the filter-bypass indicated as a dotted line. This process scheme eliminates the filtration and vacuum-fractionation unit operations by completely bypassing the filtration section. The unfiltered SRC plus mineral residue, after distilling off the distillate liquids, would be sent as a liquid slurry to an oxygen-blown gasifier and the synthesis gas generated would be shifted and purified to produce the make-up hydrogen required for the dissolver section.

Hence, this operation would produce only gases and liquids:

SNG, LPG : C_3, C_4
Light Oils : IBP - 400°F
Light Distillate Fuel: 400 - 500°F
Heavy Distillate Fuel: 500 - 800°F

OPERATING CONDITIONS

Temperature:	700 - 900°F
Pressure:	1,000 - 2,500 psi
Coal Residence Time:	15 - 150 min
Coal Space Velocity (defined to be equal to lb/h coal feed per ft^3 reactor):	15 - 100 $lb/h/ft^3$
Conversion (MAF conversion is defined to be equal to $\left[\frac{100 - \text{lb unreacted organic solids}}{100 \text{ lb MAF coal feed}}\right] \times 100\%$):	80 - 95%

Coals processed:	Kentucky No. 9 and No. 14 Coals Illinois No. 6 Coal Pittsburgh No. 8 Coal Wyodak, Wyoming Coal
Catalyst:	None*
Vehicle:	Hydrogen (for hydrogenation) Solvent (for coal dissolving) Solvent (as H_2 donor solvent) Coal ash (as pseudocatalyst)
Reaction variables (affecting the MAF conversion and/or hydrogen consumption:	Reaction temperature Reaction pressure (H_2 partial pressure) Unfiltered-slurry-recycle to coal ratio Filtered-solvent-recycle to coal ratio Reactor residence time Type of coal

PRODUCTS

Bases:	SRC I Mode: Using Kentucky Coal	SRC II Mode: Using Illinois No. 6 Coal
	Fort Lewis Pilot Plant (3)	Oil/Gas Plant Design Basis (4)
Feed Coal Ultimate Analysis:		
C	72.43	69.07
H	5.21	4.74
N	1.57	1.32
S	3.73	3.78
O	7.74	8.96
Ash	9.32	12.13
	100.00	100.00
Gross Heating Value, dry (Btu/lb):	N/A	12,444

---

*Although no catalyst is used, it is believed that the minerals in coal (and the slurry recycle if used) have a pseudocatalytic effect; slurry recycle, or SRC II, increases hydrogenation of the coal presumably due to this pseudocatalytic effect.

	SRC I Mode: Using Kentucky Coal	SRC II Mode: Using Illinois No. 6 Coal
	Fort Lewis Pilot Plant (3)	Oil/Gas Plant Design Basis (4)
Moisture with Feed, wet (wt%):	N/A	2.7
Proximate Analysis:		
Fixed Carbon	N/A	45.8
VM	N/A	39.7
Ash	N/A	11.8
Moisture	N/A	2.3
		100.00
Operating Conditions:		
Reactor Outlet Temperature (°F):	859	850
Reactor Pressure (psig):	1,478	2,025
Dissolver Space Rate (lb/h, coal feed per ft^3 reactor volume):	91.0	35
Recycle Wt Ratios:		
Slurry to Coal	1.54	2:1
Solvent to Coal	1.54	1:1
Yields (wt% MF Coal Feed):		
C_1	2.1	4.7
C_2	1.2	2.93
C_3	1.1	3.87
C_4	0.5	1.88
CO	0.1	-
CO_2	1.0	1.06
H_2S	1.3	2.65
NH_3	-	0.24
H_2O	4.6	2.67

	SRC I Mode: Using Kentucky Coal	SRC II Mode: Using Illinois No. 6 Coal
	Fort Lewis Pilot Plant (3)	Oil/Gas Plant Design Basis (4)
Light Oil (IBP - 400°F)	6.3	6.50
Wash Solvent (400 - 500°F)	3.4	7.34
Process Solvent (500 - 850°F)	0.7	15.25 (500 - 950°F)
SRC 850 °F+	64.2	36.70
Ash	9.7	12.13
Unreacted Coal	5.9	6.78
Total (including H_2 consumption)	102.1	104.7
Characteristics:		
1) Light Oil Composition (wt%)		
C	81.53	78.92
H	11.40	14.31
N	0.35	0.31
S	0.39	0.31
O	6.33	6.15
Ash	-	-
	100.00	100.00
Gross Heating Value (Btu/lb)	N/A	19,890
Boiling Range	IBP - 400°F	IBP - 400°F

	SRC I Mode: Using Kentucky Coal	SRC II Mode: Using Illinois No. 6 Coal
	Fort Lewis Pilot Plant (3)	Oil/Gas Plant Design Basis (4)
2) Wash Solvent Composition (wt%)		
C	83.47	81.61
H	8.90	10.90
N	0.59	0.68
S	0.27	0.27
O	6.77	6.54
Ash	-	-
	100.00	100.00
Gross Heating Value (But/lb)	N/A	18,134
Boiling Range:	400 - 500°F	400 - 500°F
3) Process Solvent Composition (wt%)		
C	87.87	83.41
H	7.57	9.18
N	0.87	1.11
S	0.47	0.33
O	3.22	5.97
Ash	-	-
	100.00	100.00
Gross Heating Value (Btu/lb)	N/A	17,376
Boiling Range	500 - 850°F	500 - 950°F

	SRC I Mode: Using Kentucky Coal	SRC II Mode: Using Illinois No. 6 Coal
	Fort Lewis Pilot Plant (3)	Oil/Gas Plant Design Basis (4)
4) SRC Composition (wt%)		
C	87.22	86.70
H	5.34	7.41
N	2.33	1.61
S	0.73	0.44
O	4.21	3.84
Ash	0.17	-
	100.00	100.00
Fusion Point:	380°F	>300°F(4)
Boiling Range:	850°F+	950°F+
Gross Heating Value (Btu/lb):	16,030	16,926

INTENDED PRODUCT USE/MARKET

(1) SNG & LPG: The hydrocarbon gases produced by the SRC process (I and II) constitute a high Btu fuel gas. After recovery and purification, these gases may be used as plant fuel gas and/or sold as pipeline quality SNG. LPG is also a marketable product available from the SRC process.

(2) Light Oil (IBP - 400°F): The light oil product must be hydrotreated for heteroatom (N, S, and O) removal; then it is valuable as a good reformer feedstock, as a precursor to lead-free gasoline due to its high aromatic and naphthenes content.

(3) Wash Solvent (400 - 500°F) and Process Solvent (500 - 850°F): The wash solvent and process solvent are low-sulfur fuels that could be used as a boiler fuel, produced from a high-sulfur coal.

(4) SRC: SRC, as produced from the SRC I process, is a solid at room temperature. However, it has a melting point of about 300°F and can be fired alternately as a solid or liquid fuel with burner modifications. Preliminary investigations of SRC as a pulverized solid fuel have indicated some problems with agglomeration and

plugging in pulverized coal burners(5). However, recent results were favorable for combustion tests of solid SRC in a 22.5-MW boiler(6).

The use of SRC (defined as 950°F+) is being investigated for its compatibility with petroleum fuels and use as a gas-turbine fuel.(7) SRC was found to be completely insoluble in No. 2 distillate fuel, No. 6 fuel oil, and heavy coker gas oil; hence, SRC is incompatible for use with petroleum liquid fuels. Its use as a heavy fuel oil would have to be in a captive market situation.

SRC as produced from SRC I has been analyzed to have a high percentage of undesirable asphaltenes.(8) This also indicates that it would require a captive market.

On a conceptual SRC II design of a commercial plant,(4) the 400 - 500°F wash solvent and the 500 - 950°F process solvent are combined with the SRC 950°F+ material. This combined fuel is a liquid fuel oil and may be marketable to the utilities industry as a No. 6 residual fuel oil.

PROCESS EFFICIENCY

Thermal Efficiency:

Clean Boiler Fuels from Coal(9):	63.5%
Oil/Gas Conceptual Commercial Plant(5):	77.6%

Utilities (including slurrying, dissolving, and filtration only)(4):

Basis: 1 ton moisture-free coal

Fuel Gas (internal consumption):	780 MBtu
Cooling Water :	250 gal
Power :	66 kWh
High-pressure Steam :	310 lb generated
Low-pressure Steam :	80 lb generated

Hydrogen Consumption : 4.7 lb per 100 lb dry coal

UNIQUE FEATURES OF PROCESS

- The process can produce a liquid or solid boiler fuel, depending on the amount of hydrogenation and the amount of lighter organic liquids (wash and process solvents) included in the boiler fuel blend.
- The process can use synthesis gas ($CO+H_2$) or hydrogen for liquefaction; also the solvent is believed to act as a donor of hydrogen to the coal.

- The reaction is a three-phase reaction: gas, liquid, and solids.
- Although the process is noncatalytic, it is believed that the minerals in the ash act as a pseudocatalyst in promoting hydrogenation.
- There is an optimal band of operating temperatures for good conversion (90%) of MAF coal to gas, liquids, and SRC. Below this band, conversion is limited by the Arrhenius reaction rate; above the band, thermal cracking and coke formation limit conversion.

PROCESS STATUS

Development Status:

Two pilot plants are operational. Also, there have been several conceptual designs of commercial-size coal liquefaction plants using the SRC I and II processes. At the present time, DOE has defined funding requirements and is making plans for the design, construction, and operation of a demonstration plant.(10)

The process has been patented (U.S. Patent 3,341,447).(11)

History:

Solvent refining of coal dates back to the Pott-Broche process developed in Germany in the 1920s. The process was utilized by Germany in World War II for the production of a raw material for carbon electrodes for aluminum plants. In the 1950s, R&D work on a modified Pott-Broche process was performed by the Spencer Chemical Company. In 1962, the U.S. Office of Coal Research (OCR) awarded a research contract to Spencer for technical evaluation of the SRC process. The process was demonstrated in a 50-lb/h, continuous-flow PDU in Merriam, Kansas. At that time, Gulf Oil Corporation acquired Spencer and reassigned the SRC project to The Pittsburgh and Midway Coal Mining Company. In 1966, OCR awarded PAMCO a contract for a study of the commercial feasibility of the process to include design, construction, and operation of a 50-TPD pilot plant. Stearns-Roger Corporation completed design of the plant in 1969 and after a delay due to lack of funds, detailed engineering and construction were awarded to Rust Engineering Company. The plant started up in late 1974 and has been in operation since then, along with the PDU in Merriam, Kansas. A 6-TPD SRC pilot plant was built in 1974 by Catalytic, Inc., which operates it now under the sponsorship of DOE and EPRI. Southern Company Services, Inc. serves as project manager.

There are many supporting studies for the SRC process, including product testing, product upgrading, process evaluations, and mechanical equipment testing.(1)

REFERENCES

1. Energy Research and Development Administration, *Fossil Energy Program Report, 1975-1976.*

2. Schmid, B.K. and D.M. Jackson, *Recycle Processing for Liquid and Solid Fuels,* presented at Fourth Annual International Conference on Coal Gasification, Liquefaction and Conversion to Electricity, University of Pittsburgh, August 1977.

3. The Pittsburgh and Midway Coal Mining Company, *Quarterly Report for the Period July - September, 1975,* Energy Research and Development Administration Contract No. E(49-18)-496, October 1975.

4. O'Hara, J.B. et al, *Oil/Gas Complex - Conceptual Design/ Economic Analysis - Oil and SNG Production, R&D Report No. 114, Interim Report No. 4,* Energy Research and Development Administration Contract No. E(49-18)-1775, March 1977.

5. Greskovich, E.J., "Chemical Characterization, Handling, and Refining of SRC to Liquid Fuels," *Quarterly Progress Report,* Energy Research and Development Administration Contract No. E(49-18)-2003, prepared by Air Products and Chemicals, Inc., September 1975.

6. *Chemical Engineering*, October 10, 1977, pp. 77-80.

7. Bendoraitis, J.G. et al, *Upgrading of Coal Liquids for Use as Power Generation Fuels - Phase I,* Electric Power Research Institute Contract No. RP 361-1, prepared by Mobil Research and Development Corp., January 1976.

8. Schwajer, I. and T.F. Yen, *Preliminary Examination of Coal Liquefaction Products,* presented at Symposium on Coal Liquefaction, 172nd National Meeting of the American Chemical Society, Division of Fuel Chemistry, San Francisco, California, August, 29 - September 3, 1976.

9. O'Hara, J.B. et al, *Demonstration Plant - Clean Boiler Fuels from Coal - Preliminary Design - Vol. I - Summary Report, R&D Report No. 82, Interim Report No. 1,* prepared for the United States Department of Interior, Office of Coal Research.

10. *Energy Research Digest,* Vol. III, No. 9, April 25, 1977, pp. 1, 2.

11. Bull, W.C. et al, *Solvation Process for Carbonaceous Fuels,* United States Patent No. 3,341,447, September 12, 1967.

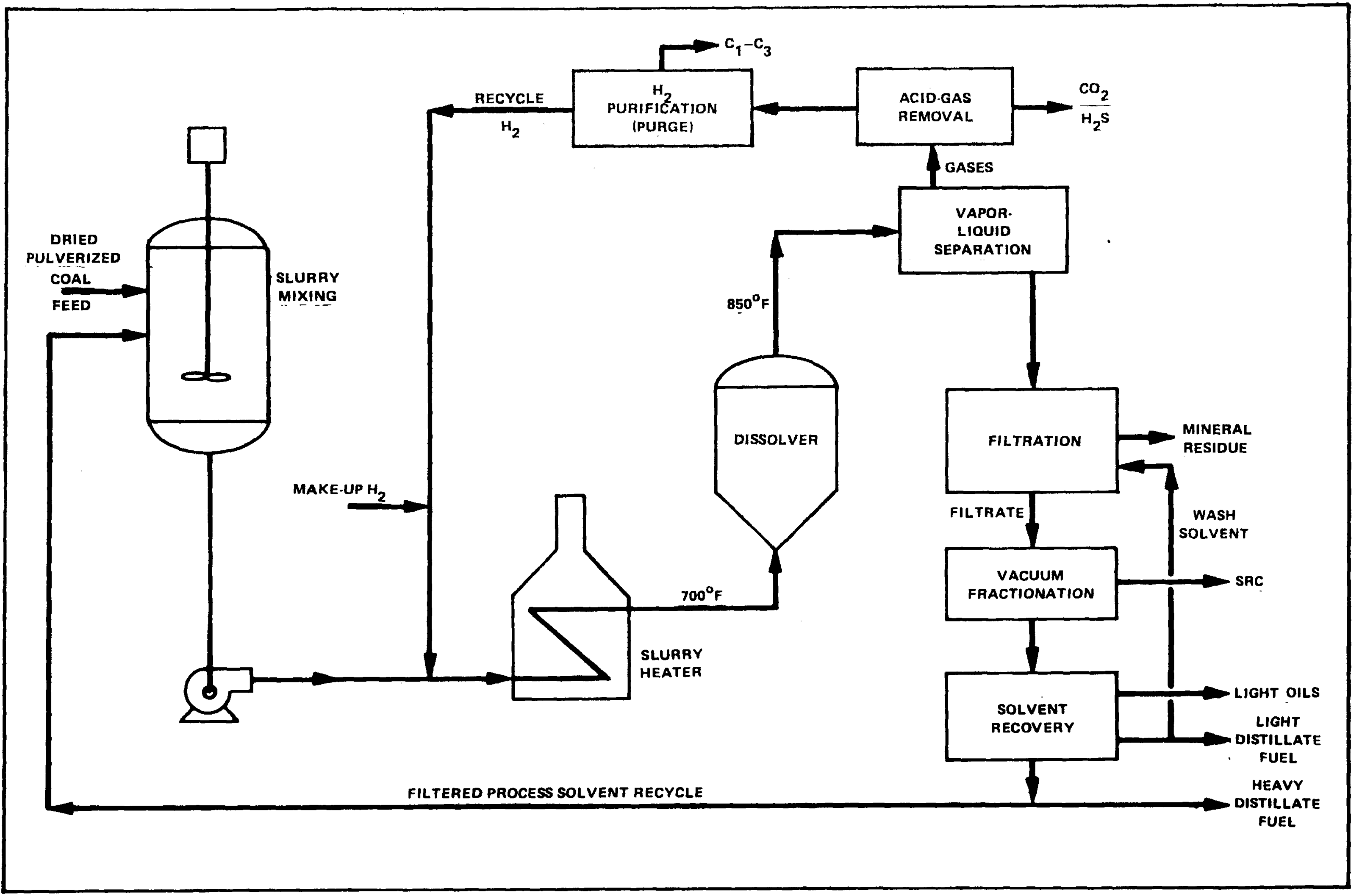

Figure 1 - SRC I Process (Ft. Lewis Pilot Plant)

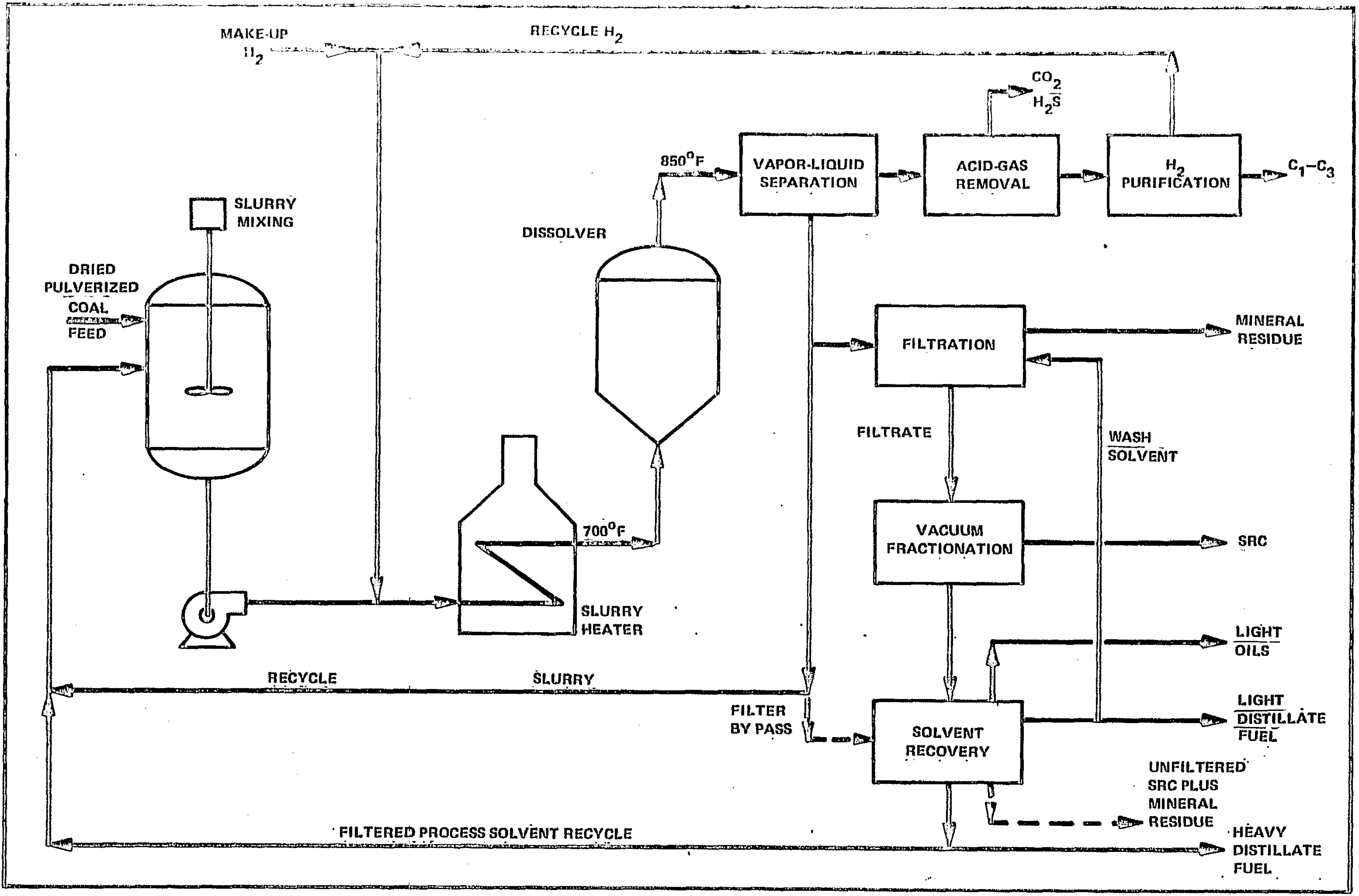

Figure 2 - SRC II Process (2)

PROCESS INFORMATION EXTRACT

PROCESS 1.1.2	UOP Extraction Process
Process Type:	Hydroliquefaction, Direct, noncatalytic
Main Products	Gas Coal Extract Liquids
Development Status:	Bench scale and pilot plant
PROCESS DEVELOPER	Universal Oil Products (UOP) Corporation Des Plaines, Illinois
PROCESS SPONSOR	Universal Oil Products Corporation

PROCESS DESCRIPTION

The UOP process has been described in the literature$^{(12), (77)}$ as is shown in Figure 1 and as follows:

Coal and solvent are charged to the reactor at sufficient velocity to cause degradation of the coal by impact against a fixed plate inside the reactor. Hydrogen is passed upwards through the slurry to cause hydrogenation of the coal and solvent. A conventional hydrogenation catalyst may be added to the reactor to promote hydrogenation.

The slurry is sent to a gas-liquid separator to recover gas for recycling to the reactor. The liquid coal extract and mineral residue is passed through an extraction zone where solids residue is removed. The liquid extract is then fractionated to produce light and heavy hydrocarbon liquid products (gasoline, jet fuel, diesel oil, and heavy fuels) and recycle solvent.

The light hydrocarbons are catalytically hydrotreated to produce a synthetic crude oil.

OPERATING CONDITIONS(13 - 18)

Temperatures:	700 - 800°F
Pressures:	70 - 2,000 psig
Residence time:	30s to 5h
Conversions:	50 - 90 wt% of MAF coal to liquids
Catalyst:	Typical hydrogenation catalyst, if used. An example is cobalt-molybdenum catalyst.
Reaction variables:	The effect of reaction variables has not been disclosed.
Coals processed:	Illinois coal

PRODUCTS

- Product yields are not given in the patents except to indicate the liquid coal extract weight percent yield. This varied from 50 to 60 weight percent of MAF coal. A syncrude yield of 4 bbl per ton was estimated.(18)
- Characteristics of the extract are not available, except that the syncrude has a sulfur content of less than 0.15 wt%.
- Intended product use/markets is briefly treated in the patents by saying the extract liquids are valuable for chemical syncrude, or fuel production.

PROCESS EFFICIENCY

UOP has not disclosed any information.

UNIQUE FEATURES OF PROCESS

- Use of impact plate and high velocities to promote hydroliquefaction
- Liquid-solid separation step, apparently without filtration.

Table 1 - UOP Coal Extraction Patents

Reference Number	13	14	15	16	17	18
U.S. Patent Number	3,477,941	3,488,278	3,505,202	3,583,900	3,598,718	3,867,275
First Stage Extraction						
Solvent	Donor	Donor	Tetralin	Tetralin	Tetralin	Aliphatic + Aromatic
Solv.:Coal wt Ratio	0.2-10	.2-10	1:1	3:1	3:1	1:1
Temperature (°F)	750	750	700	800	800	750
Pressure (psi)	500	500	70	2000	2000	1470
Residence Time	30 sec-5 h	30 sec-5 h	1 h	30 min	30 min	4 h
Second Stage Extraction						
Solvent	-	-	Hexane	Heptane	Benzene	Benzene
Solv.:Coal wt Ratio	-	-	-	5:1	5:1	-
Temperature (°F)	-	-	-	300	250	175
Pressure (psi)	-	-	-	500	500	14.7
Residence Time	-	-	-	30 min	1 h	4 h
Third Stage Extraction						
Solvent	-	-	-	Benzene	-	-
Solv.:Coal wt Ratio	-	-	-	5:1	-	-
Temperature (°F)	-	-	-	300	-	-
Pressure (psi)	-	-	-	500	-	-
Residence Time	-	-	-	30	-	-
Total Liquid Coal Extract - wt % of MAF Coal	50-90%	50-90%	>50%	>50%	>50%	
Unique Feature	Impact plate in reactor	Counter-current extraction	Filtration to separate solids and liquids	Three-stage solvent extraction	Two-Stage solvent extraction	Two-Stage extraction using aliphatic and aromatic solvents

PROCESS STATUS

Development Status:

Although UOP has over 20 patents on coal liquefaction, their research in the field is exploratory and has not evolved to the point where a definitive design can be said to exist.(19) The process description and operating conditions have been taken from reference No. 13. Table 1 presents a sampling of some other patents that have been issued to UOP including operating conditions.

Location of Development Work: Des Plaines, Illinois

History:

UOP has been investigating coal liquefaction since the 1960s. Patents were granted in 1969. Bench and pilot plant scale work has been done.

Problem Areas:

- Erosion of impact plate
- Large vessel size due to possible high residence times
- Complete process undefined

REFERENCES

12. Nelson, E.F., *Method of Treating Coal (Solvent Extraction)*, U.S. Patent No. 3,477,941, November 11, 1969.

13. Nelson, E.F., *Process for Treating Coal (Countercurrent Solvent Extraction)*, U.S. Patent No. 3,488,278, January 6, 1970.

14. Nelson, E.F., *Solvent Extraction Method (Hydrogen-Donor Solvents)*, U.S. Patent No. 3,505,202, April 7, 1970.

15. Gatsis, J.G., *Coal Liquefaction Process by Three-Stage Solvent Extraction Process*, U.S. Patent No. 3,583,900, June 8, 1971.

16. Gleim, W.K.T. et al, *Solvent Extraction of Coal (Two-Stage Extraction System)*, U.S. Patent No. 3,598,718, August 10, 1971.

17. Gleim, W.K.T. and M.J. O'Hara, *Two-Stage System Using Hydrogenated Bottoms Fraction and Benzene*, U.S. Patent No. 3,867,275, February 18, 1975.

18. *Chemical Engineering Progress*, Vol. 71, No. 4, April 1975, pp. 98-99.

19. Personal Telephone Communication to M.J. O'Hara and W. Page (UOP) and J.F. Ferrall (The Ralph M. Parsons Company), March 17-18, 1977.

77. Howard-Smith, I. and G.J. Warner, *Coal Conversion Technology: A Review, May 1975.*

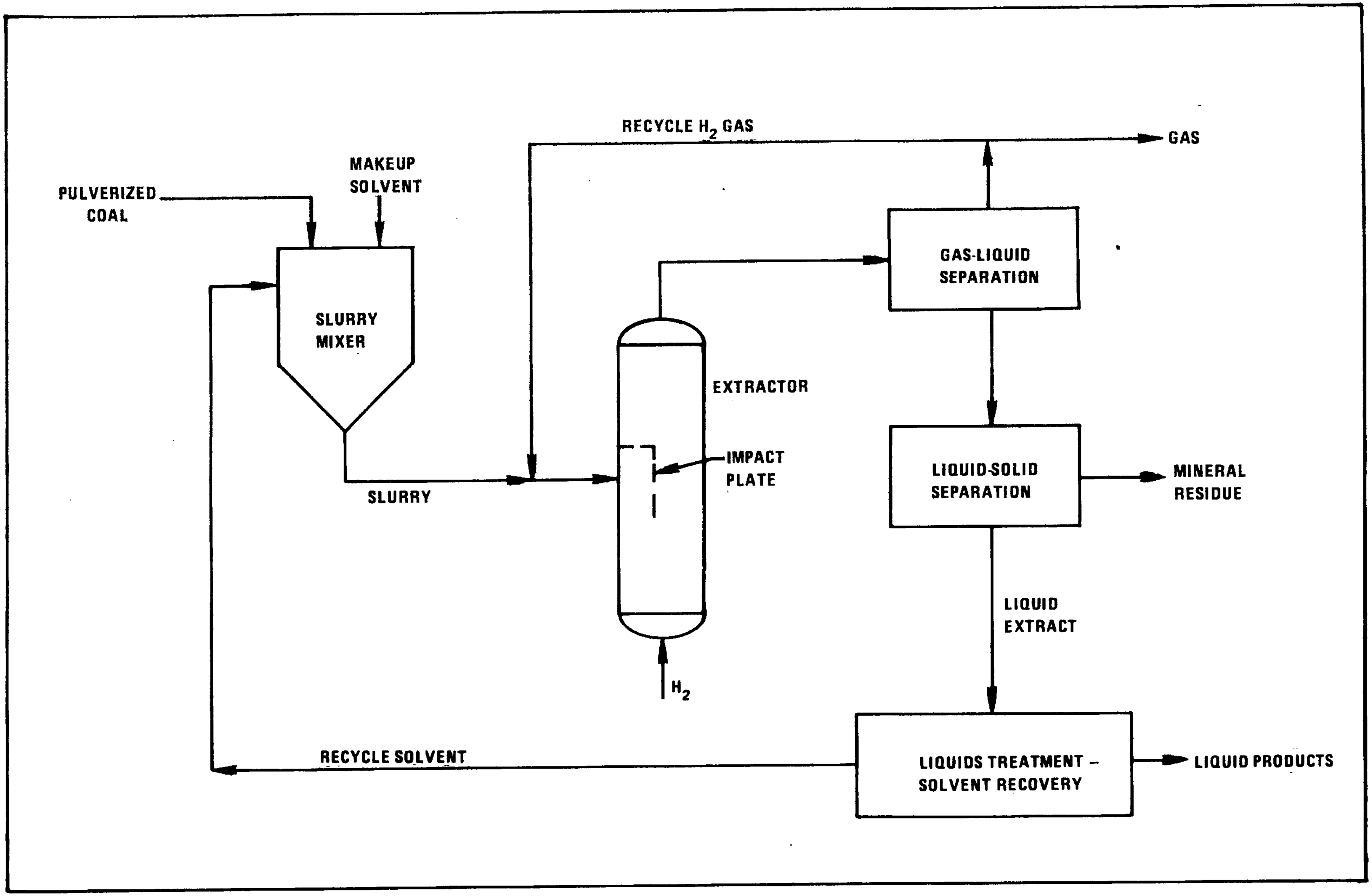

Figure 1 - UOP Extraction Process (12), (77)

PROCESS INFORMATION EXTRACT

PROCESS 1.2.1	Gulf Catalytic Coal Liquids (CCL)
Process Type:	Hydroliquefaction, Direct, Catalytic
Main Products:	Low-sulfur fuel oil
Development Status:	Pilot Plant
PROCESS DEVELOPER	Gulf Research and Development Company Pittsburgh, Pennsylvania
PROCESS SPONSOR	Gulf Oil Corp.

PROCESS DESCRIPTION(21) (Figure 1)

The CCL process contains four major process unit operations:

(1) Liquefaction

(2) Separation (solvent recovery/removal of insoluble solids)

(3) Coking

(4) Hydrogen manufacture

In the liquefaction unit, hydrogen and powdered coal, conveyed in a portion of the product oil, are combined and preheated to reaction temperature. The hydrogen stream consists of recycle hydrogen and makeup hydrogen. The reactor is of a unique proprietary design which permits the use of a fixed-catalyst bed.

The total reaction product is cooled by heat exchange and flows to a hot vapor-liquid separator. The liquid/coal residue slurry is reduced in pressure and hydrocloned to produce a low solids overhead stream, which is recycled for slurrying with the feed coal, and a high solids bottom product, which is sent to a solids removal process.

Solids are removed from the primary liquid product by filtration, distillation, and other processes. After solids removal, the liquid is ready for storage and shipment. The solids are sent to a coking unit for recovery of occluded solvent and any volatile matter remaining in the undissolved coal.

Separated gases are processed to remove NH_3, H_2S, H_2O, and light hydrocarbons. The purified hydrogen is combined with fresh hydrogen and recycled to the reactor.

Light hydrocarbons are steam reformed to balance process hydrogen requirements.

OPERATING CONDITIONS

Liquefaction:

Pressure, atm	>136
Temperature, °F	>750
Solvent/coal, wt/wt	1.5

Space Velocity/Residence time:

Preheater residence time, min	<2
pound dry coal/cf reactor/h (2-1/4 in. ID reactor)	45

Conversions:	Sub-bituminous	Bituminous
H_2, wt% MAF coal	6.23	4.58
H_2, SCF/bbl oil (filtrate)	8137	4299
Coal converted, wt%	91	90

Catalyst: Gulf proprietary catalyst

Reaction Variables:

Reaction variables studied have not been published. A 120-lb/day pilot unit can operate at a pressure up to 680 atm, and at a temperature as high as 1000°F. Gulf studies indicate highest conversion yields are obtained by high-ash, high-volatile bituminous coals containing high proportions of vitrinite and exinite.

Coals Processed:

Lignite, sub-bituminous, and high volatile bituminous.

PRODUCTS(21)

Coal Feed Basis:	Big Horn Sub-bituminous	Pittsburgh Seam Bituminous
Coal moisture, wt%:		
To unit	20	2.9
As received	22.0	2.9
Coal Analysis (dry)		
Proximate, wt%:		
Ash	4.4	8.3
Volatile matter	42.6	34.1
Fixed carbon	53.0	57.6
Ultimate, wt%:		
Carbon	69.34	76.84
Hydrogen	4.60	5.06
Nitrogen	1.23	1.61
Sulfur	0.54	1.49
Oxygen	19.90	6.72
Ash	4.39	8.28
HHV, Btu/lb (dry basis)	11,192	13,913

Yields (MAF coal basis, excluding hydrogen manufacture & utilities)

	Sub-bituminous	Bituminous
CO_2 + HC gas, wt%	20.7	8.8
Light ends, wt%	11.5	7.5
Light ends, bbl/ton	0.78	0.50
Filtrate, wt%	52.1	75.1
Filtrate, bbl/ton	2.91	4.04
Solids, wt%	12.2	16.4
(% Ash in solids)	55.9	50.5
Characteristics:		
Filtrate:		
Distillation, °F[(21)]		
IBP	130	294
20 vol %	450	517
40	495	590
60	570	680
70		900
80	870	N.A.
90	900	N.A.
Ultimate Analysis, wt% (dry):		
Hydrogen	9.08	8.18
Nitrogen	0.41	0.45
Sulfur	0.044	0.145
Oxygen	0.91	1.41
HHV, Btu/lb	17,930	17,373
Sp. Gr. (60°F/60°F)	1.007	1.073
Viscosity, CS @ 100°F	7.1	N.A.
Ash, wt%	0.02	N.A.

INTENDED PRODUCT USE/MARKETS (21)

(1) Low sulfur fuel oil: as a substitute for No. 6.

(2) Gas turbine fuel oil: precoat filters have been successfully used at room temperature to produce a product of very low ash content suitable for firing gas turbines.

PROCESS EFFICIENCY (21)

Thermal Efficiency:	Sub-bituminous	Bituminous
Product only %	77	74
Overall, including net utilities, %	67	63
Utilities, per ton dry coal liquefied:		
Fuel , mm btu	2.71	3.74
Hydrogen Consumption:		
Wt% MAF coal liquefied	6.23	4.58
SCF/bbl net oil	6048	3825

UNIQUE FEATURES OF PROCESS

Coal slurry hydrogenation in a fixed bed catalytic reactor, reactor oil slurry clarified by hydroclone to produce oil to slurry coal, addition of molecular hydrogen to liquefaction reactor, solids removed from product by filtration/distillation, and coking recovered solids.

PROCESS STATUS

Development Status:

One-ton-per-day continuous pilot plant began operation January 1975.

Joint research program between Gulf and Pennsylvania State University studying effects of coal characteristics on liquefaction behavior.

Location of Development: Gulf R&D, Hammaville, PA.

History:

Research on catalytic conversion of coal began with autoclave experiments, about 1968, and then developed into a research program on a continuous 0.5 to 5 lb/h bench scale reactor.

January 1975 a 1-TPD pilot plant began operation. Operation of the pilot plant was to provide design data for a large 50 to 500-TPD pilot plant in the design stage. A 15,000-TPD demonstration plant to produce about 50,000-BPD liquids is planned.

Problem Areas:

No problems reported. A six week continuous run was completed in the bench scale tests.

REFERENCES(21)

21. Chun, S.W., *Gulf Catalytic Coal Liquids (CCL) Process*, Paper presented at Workshop on Material Problems and Research Opportunities in Coal Conversion, Columbus, Ohio, April 16, 1974.

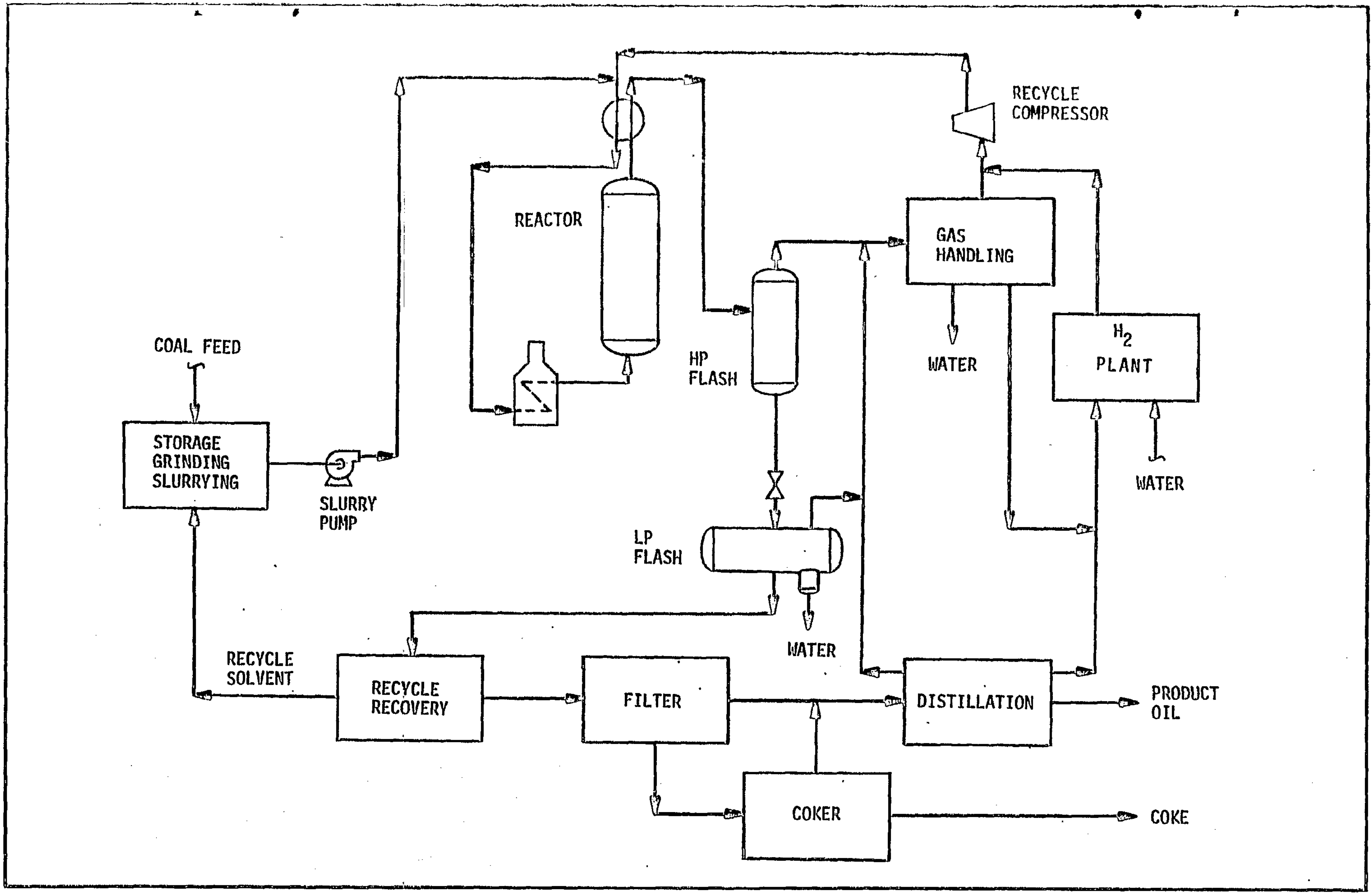

Figure 1 - Flow Diagram Gulf CCL Process (21)

PROCESS INFORMATION EXTRACT

PROCESS 1.2.2 SYNTHOIL

Process Type: Hydroliquefaction, Direct, Catalytic

Main Products: Low-sulfur fuel oil/Gas turbine fuel

Development Status: Process Development Unit (PDU)

PROCESS DEVELOPER

Pittsburgh Energy Research Center (PERC)
Pittsburgh, Pennsylvania

PROCESS SPONSORS

(1) Energy Research and Development Administration (ERDA)

(2) Bethlehem Steel Company was a participant by a cooperative agreement.

PROCESS DESCRIPTION (22), (23), (24)

The Synthoil process contains four major process unit operations:

(1) Liquefaction

(2) Separation (solvent recovery/removal of insoluble solids)

(3) Pyrolysis

(4) Hydrogen Manufacture

The integration of the units is depicted in the Synthoil block flow diagram, Figure 1. A simplified flowsheet for a commercial Synthoil plant is shown in Figure 2.

In the liquefaction unit, hydrogen and powdered coal, conveyed in a portion of the product oil, are combined and preheated to reaction temperature. The hydrogen stream consists of recycle hydrogen containing some light hydrocarbons and makeup hydrogen. Preheat furnace tubes are packed with ceramic pellets to promote heat transfer. The heated feed stream flows upward through a fixed-bed of hydrogenation catalyst in the liquefaction reactor. Turbulent flow prevents plugging or fouling of the catalyst. Reactor temperature rise is controlled by quench oil (cold recycle oil product).

The total reactor product is cooled by heat exchange and condensed liquids and unreacted solids separated. The liquid/coal residue slurry is reduced in pressure and solids are separated from the liquid by centrifuge. The liquid is divided into recycle, quench, and product fuel oil. Solids are pyrolyzed to yield additional fuel oil and a high ash content char.

Separated gases are processed to remove NH_3, H_2S, H_2O, C_1-C_5 hydrocarbons, and other gaseous reaction products. The purified hydrogen is combined with fresh hydrogen and recycled to the reactor.

Char from the pyrolyzer is gasified to produce fresh makeup hydrogen for the process. Some coal may also be fed to the gasifier, or C_1-C_5 hydrocarbons steam reformed, to balance process hydrogen requirements.

OPERATING CONDITIONS (22), (23)

	Kentucky Coal (22),(23)	
Liquefaction:		
Pressure, atm.	136	272
Temperature, °F	806-842	842
Solvent/Coal, wt/wt	1.86	1.86
H_2/Coal, SCF/lb Coal	148.6	148.6
Space Velocity/Residence Time pound dry coal/ f^3/h	91	91
	(~2 minutes based on slurry)	
Pyrolysis:		
Temperature, °F	700-760	590-650
Conversions:		
H_2, wt% MAF coal,	3.43	4.75
SCF/bbl oil	3450	4375

Catalyst:	Cobalt-molybdenum (1/8 in. x 1/8 in. pellets)

Reaction Variables:

The following variables have been tested in the bench scale and PDU plants:

	Range		
Pressure, psig	2000	and	4000
Temperature, °F			
Reactor	842		
Preheater Outlet	806	and	842
Space Velocity, lb Coal/ft^3/h	42	-	92
Solvent/Coal, wt/wt	1.22	-	2.33
Catalyst Age, lb Coal/lb Catalyst	Up to	795	

Catalyst Age: (24)

As catalyst age varies from start-of-run (SOR) to 500 hours, 795 lb coal/lb catalyst, for Kentucky coal:

% Sulfur varied			
@ 4000 psig	0.15	to	0.5
@ 2000 psig	0.20	to	0.7
Viscosity, SSF @ 180°F varied			
@ 2000 psig	10	to	30
@ 4000 psig	14	to	98
% Ash in de-ashed oil varied			
@ 2000 psig	0.1	to	1.1
@ 4000 psig	0.16	to	2.9

Pressure (24):

For similar run conditions, operating at 4000 psig rather than 2000 psig results in increased oil hydrogenation and a small increase in coal liquefaction.

Coal	West Virginia		Kentucky	
Pressure	2000 psig	4000 psig	2000 psig	4000 psig
Yield bbl/ton coal	3.2	3.3	3.0	3.2
Oil HHV, Btu/lb	16,700	17,000	16,600	17,400
Viscosity (EOR), SSU at 180°F	580	40	98	30

Temperature (22):

For similar run conditions with reactor at 4000 psig decreasing the preheater outlet temperature reduced the degree of oil hydrogenation and coal liquefaction.

Coal	Kentucky	
Preheat temperature	842°F	806°F
Yield, bbl/ton coal	3.27	3.20
Wt% S	0.2	0.45

Space Velocity (24), (25)

Space velocities, lb coal/cf reactor/h, of 42 and 63 were reported for a 5/16-inch reactor. A space velocity of 92 is sufficient for the 1 in. PDU reactor due to less wall effect and greater catalyst packing. Residence time in the feed preheater is about 30 minutes based on coal slurry.

Coal Processed: High volatile A and B bituminous.

PRODUCTS (23)

Coal Feed Basis:	Kentucky (hvAb)
Coal moisture, wt%:	
As received	4.2

Coal Analysis (dry)

Proximate, wt%:	
Ash	17.2
Volatile matter	37.8
Fixed carbon	45.0
Ultimate, wt%:	
Carbon	63.4
Hydrogen	4.5
Nitrogen	1.3
Sulfur	5.7
Oxygen	7.9
Ash	17.2
HHV, Btu/lb (dry basis):	11,503

Yield: MAF coal, at 4000 psig, excluding hydrogen manufacture and utilities.

H_2S, NH_3, wt%	6.3
Hydrocarbon gases (C_1-C_5)	
wt%	7.6
SCF/ton	3745
Light oil	
wt%	2.3
bbl/ton	0.15
Synthoil heavy oil	
wt%	61.5
bbl/ton	3.37
Pyrolysis oil	
wt%	11.1
bbl/ton	0.63
Carbonaceous residue (74.5% ash)	26.9

Characteristics:

(1) Synthoil heavy oil(22)

Solvent analysis of centrifuged oil, wt%

Run Operating Pressure:	2000 psig	4000 psig
Oil	65.9	88.3
Ashphaltenes	27.6	10.8
Organic benzene insolubles	4.4	0.8
Ash	2.1	0.1
HHV, Btu/lb	16,640	17,400
Sulfur, wt%	0.6	0.2
Viscosity oil, SSF SOR-EOR	13.5-97.6 180°F	26-440 77°F
Ash, wt% SOR-EOR	1.3-2.9	0.1-0.2
(2) Pyrolysis oil(22)		
Sulfur, wt%	0.2	0.4
Ash, wt%	0.0	0.0

INTENDED PRODUCT USE/MARKET

(1) C_1-C_5 Hydrocarbons: May be sold as SNG or steam reformed to hydrogen.

(2) Low Sulfur Fuel Oils: Viscosities and pour points ranging from No. 2 to No. 6 fuel oil. Total oil sulfur content less than 0.7 wt%.

(3) Gas Turbine Fuel: Centrifuged synthoil can be filtered to an ultra-low-ash oil, containing less than 0.05 wt% ash, potential for firing gas turbines.

PROCESS EFFICIENCY[23] (@ 4000 psig, with Kentucky coal)

Thermal Efficiency:	
Product only, %	78.1
Overall, including net utilities, %	74.9
Utilities, per ton dry coal liquefied:	
Power, kWh	110
Fuel (process), MMBtu	0.42
Hydrogen Consumption:	
Wt% MAF coal liquefied	5.04
SCF/bbl net oil	4,559

Catalyst:

Catalyst life tests not completed. PDU unit Synthoil sulfur level reaches 0.7 wt% after 500 hours at 2000 psig or 795 lb coal/lb catalyst.

UNIQUE FEATURES OF PROCESS

Coal slurry hydrogenation in a fixed bed catalytic reactor, product oil used to slurry coal, addition of molecular hydrogen to liquefaction reactor, centrifuge separation of solids and unreacted coal from product oil, and pyrolysis of solids recovered.

PROCESS STATUS

Development Status:

One-half-ton-per-day continuous pilot plant constructed March 1973, and put into productive operation. A 10-TPD pilot plant under design and construction. Preliminary design for a 700-TPD demonstration plant to begin April 1977.

Patents: U.S. - 3,840,456

Location of Development: Pittsburgh Energy Research Center of DOE

History:

Synthoil process developed in the early 1970s by the Bureau of Mines, Pittsburgh Energy Research Center (now DOE PERC).

Initial development work used a 5/16-inch I.D. reactor capable of processing 48-lb coal/day or 120-lb coal slurry/day. Reactor operated from February 1971 to December 1972.

Current work uses a 1.1-inch I.D. reactor capable of processing 400-lb coal/day or 1/2-ton coal slurry/day.

Problem Areas:

None reported.

REFERENCES

23. Akhtar, S. et al, *The Synthoil Process -- Material Balance and Thermal Efficiency*, Paper presented at 67th Annual Meeting, The American Institute of Chemical Engineers, Washington, D.C., December 1-5, 1974.

24. Yavorsky, P.M., *ERDA Coal Liquefaction Programs for Synthetic Fuel-Oil Development*, Paper presented at Symposium on Air Quality Management in the Electric Power Industry, Austin, Texas, January 28-30, 1976.

25. Yavorsky, P.M. et al, *High Turbulence and Fixed Bed Catalytic Desulfurization*, U.S. Patent No. 3,840,456, October 8, 1974.

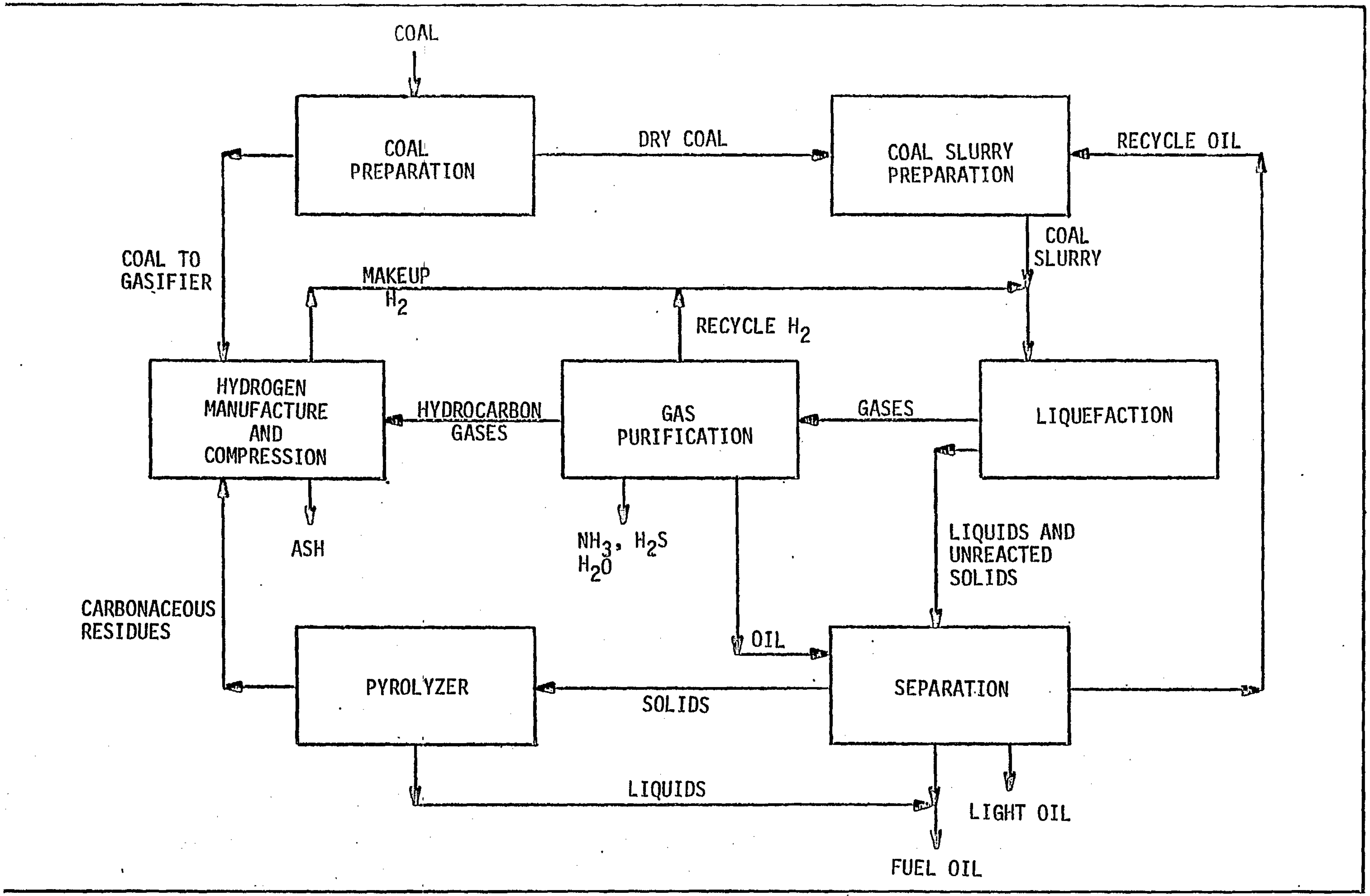

Figure 1 - Block Flow Diagram of Synthoil Process (23)

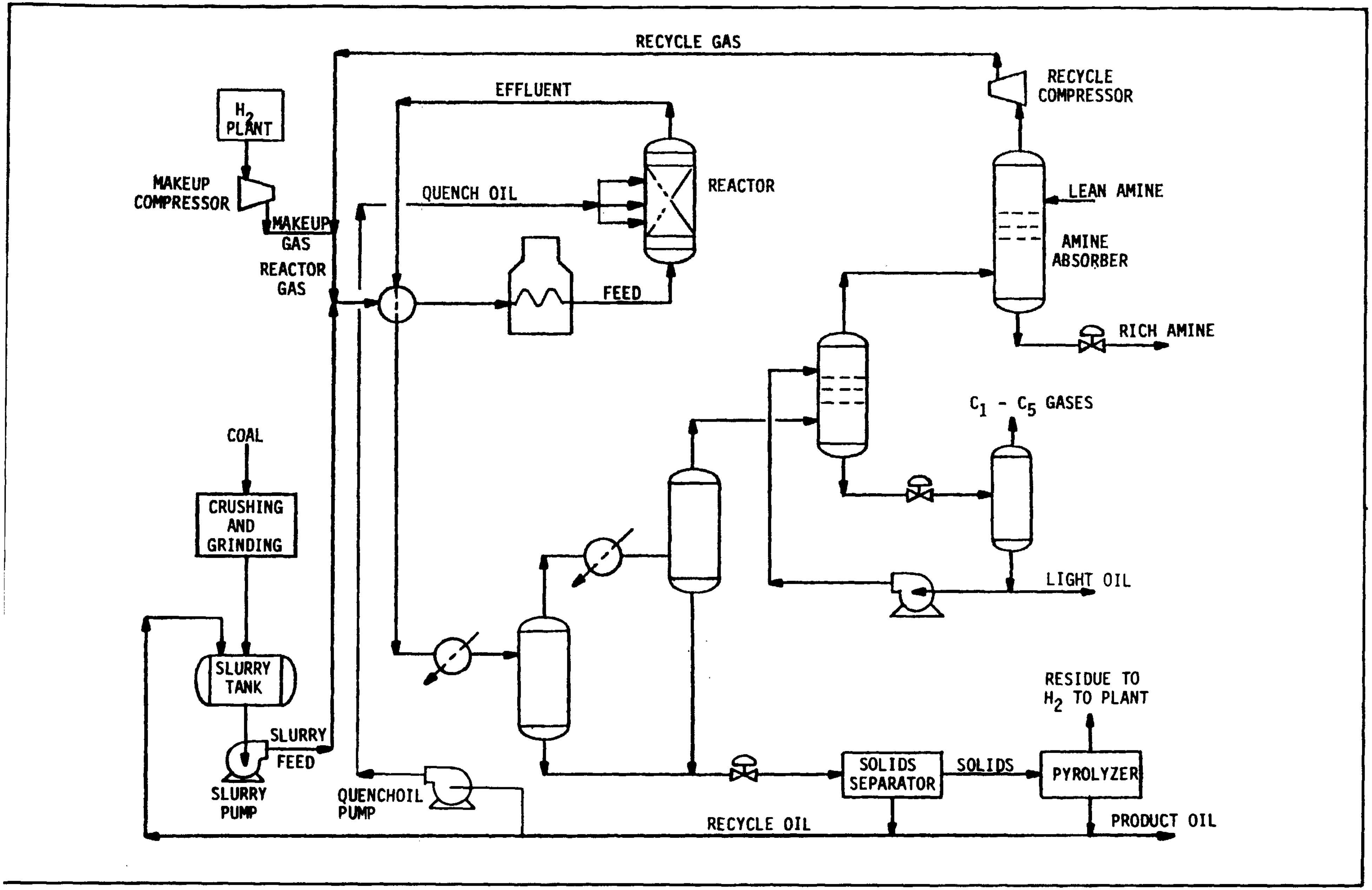

Figure 2 - Synthoil Plant Flow Diagram (23)

PROCESS INFORMATION EXTRACT

PROCESS 1.2.3 H-Coal

Process Type: Hydroliquefaction, Direct, Catalytic

Main Products: Naphtha and wide boiling range low-sulfur fuel oil, or synthetic crude oil.

Development Status: Process Development Unit (PDU)

PROCESS DEVELOPER
Hydrocarbon Research, Inc. (HRI)
Trenton, New Jersey

PROCESS SPONSORS

(1) Continental Oil Co.

(2) Mobil Oil Company

(3) Ashland Oil

(4) Standard Oil of Indiana (Amoco)

(5) Electric Power Research Institute (EPRI)

(6) Department of Energy (DOE)

PROCESS DESCRIPTION(26), (27)

The H-coal process contains three major process operations.

(1) Liquefaction.

(2) Separation (solvent recovery/removal of insoluble solids).

(3) Hydrogen Manufacture

The integration of the units is depicted in the H-Coal block flow diagram, Figure 1.

A simplified flowsheet of the H-Coal PDU is shown in Figure 2.

In the liquefaction unit, crushed coal is slurried with recycle solvent, and the coal slurry then mixed with plant makeup hydrogen. The coal slurry hydrogen mix is preheated in a co-current radiant type fired heater and fed to the reactor. Recycle hydrogen is also preheated and fed to the reactor. The coal slurry and hydrogen flow upward through an expanded bed of hydrogenation catalyst. Internal and/or external liquid recycle may be used to maintain the catalyst in an ebullated state. Two reactors in series may be required for coals of low rank.

The reactor product liquid and coal residue is withdrawn as a slurry and reduced in pressure by means of a high-pressure drop control valve. Flash gases are separated and sent to an atmospheric distillation unit. Flash liquid is sent to a vacuum distillation unit for recovery of solvent and syncrude. For low-sulfur fuel oil production, solids are removed from the liquid stream prior to vacuum distillation.

A proportion of the flash liquid is clarified by hydroclones and recycled to the reactor.

Hydrogen-rich gas is withdrawn from the reactor and cooled at reactor pressure to remove heavy hydrocarbons and water, then scrubbed to recover light hydrocarbons. A proportion of the scrubbed gas is purged from the process and used as fuel and for hydrogen manufacture. The remainder is recycled to the reactor. Separated hydrocarbons are fed to the atmospheric distillation unit.

Recovered solvent from the distillation units is slurried with fresh coal to repeat the cycle. Heavy bottoms and coal residue from the vacuum unit go to a gasifier for hydrogen production.

OPERATING CONDITIONS

Coal Processed:	Illinois No. 6			Brown Coal
Case:	Synthetic Crude (26)	Low Sulfur Fuel Oil (26)	Low Sulfur Fuel Oil (27)	Two Stage (28)
Pressure, atm:			184	153
Temperature, °F:			850	850
Solvent/Coal wt/wt:			2.27	1
H_2/Coal - SCF/lb coal:			14	35
Space Velocity/Residence Time: (pound dry coal/ft^3/h)	33.4	93.6	72.8	31.2

	Illinois No. 6			Brown Coal
	Synthetic Crude (26)	Low Sulfur Fuel Oil (26)	Low Sulfur Fuel Oil (27)	Two Stage (28)
Conversions:				
H_2, wt% MAF coal	5.44	3.57	3.38	5.03
H_2, SCF/bbl oil (C_4-975°F+)	4247	2824	N.A.	4760
Coal converted, wt%	94.2	92.4	91.1	99.0
Catalyst:	Cobalt-molybdenum			

Reaction Variables:

The following variables have been tested in the PDU.(29)

	Range
Pressure, atm	109 - 204
Temperature, °F	800 - 900
Space velocity, lb coal/h/lb catalyst	0.5 to 2.0
Slurry oil H/C atomic ratio	0.8 to 1.3
Catalyst age, lb/coal/lb catalyst	up to 2300

Catalyst age:(27)

PDU operating results show that as the catalyst ages:

% coal reacted declines slowly

% hydrogen consumed declines to an approximate constant value

C_1 - C_3 gas yield declines slowly

C_4 - 400°F liquid yield declines linearly

400°F+ fuel oil yield increases

Sulfur content of 400°F+ fuel increases

Reactor residuum content:

- It has been found that the concentration of residuum in the reaction zone has a significant effect on the conversion of coal to distillate products. Increasing the residuum content of the reactor slurry from 21.1 to 33.0 wt% resulted in a 30% reduction in required reactor residence time, and increased the C_4-975°F distillate yield by 11%.[27]

Coals Processed:

Lignite, Brown Coal, sub-bituminous, bituminous C, high volatile bituminous.

PRODUCTS

Coal Processed:	Illinois No. 6			Brown Coal
Case:	Synthetic Crude (26)	Low Sulfur Fuel Oil (26)	Low Sulfur Fuel Oil (27)	Two Stage (28)
Coal moisture: wt%				
To unit:	N.A.	N.A.	2.29	3.28
As received:	N.A.	N.A.	9.22	64.2
Coal Analysis (dry basis):				
Proximate, wt%:				
Ash	9.9	9.9	11.17	8.31
Volatile matter	42.0	42.0	38.68	48.98
Fixed Carbon	48.0	48.0	50.15	42.71
Ultimate, wt%:				
Carbon	70.7	70.7	69.66	62.64
Hydrogen	5.4	5.4	4.86	4.50
Nitrogen	1.0	1.0	1.28	0.53
Sulfur	5.0	5.0	3.52	1.22
Oxygen	8.1	8.1	9.51	23.20
Ash	9.9	9.9	11.17	8.31

HHV, Btu/lb (dry basis):	N.A.	N.A.	12,522	N.A.
Yield, wt% MAF				
$C_1 - C_3$	10.7	5.4	5.3	8.0
C_4 - 400°F	17.2	12.1	17.0	19.4
400 - 650°F	28.2	19.3	22.4	39.0
650 - 975°F	18.6	17.3	22.4	39.0
975°F+ liquids	10.0	29.5	37.6	5.7
Unconverted coal	5.8	7.6	8.9	1.0
CO_2	N.A.	N.A.	N.A.	15.9
CO	N.A.	N.A.	N.A.	0.3
H_2O	15.0	12.8	11.5	14.2
NH_3	N.A.	N.A.	N.A.	0.6
H_2S	N.A.	N.A.	N.A.	1.1
Characteristics C_4+ liquid:				
API Gravity	15.0	4.0	N.A.	N.A.
Hydrogen, wt%	9.48	8.43	N.A.	N.A.
Sulfur, wt%	0.19	0.43	0.70	(0.38)
Nitrogen, wt%	0.68	1.05	N.A.	N.A.
Oxygen, wt%	N.A.	N.A.	N.A.	(1.37)

The products are highly aromatic and relative to petroleum stocks contain high levels of nitrogen.

INTENDED PRODUCT USE/MARKET

HRI has prepared a process design for a hydrogen refinery to upgrade the coal liquids to gasoline and furnace oil.(29) Naphtha, middle distillates, and heavy gas oil are hydrotreated to reduce sulfur and nitrogen levels. Middle distillates are hydrocracked to light and heavy naphtha. Crude naphthas and hydrocrackates are upgraded by catalytic reforming to high octane gasoline blending stocks.

Vacuum bottoms from the H-Coal process are coked to yield additional heavy gas oil and a coal residue (Char-Ash-Coke).

8616 tons/day of Illinois No. 6 coal are converted to 8649-BPSD crude naphtha, 13,124-BPSD distillate, 6677-BPSD heavy gas oil, and 3636-TPD vacuum bottoms in an H-Coal process. These products are converted by hydrorefining to 20,000-BPSD gasoline, 10,000-BPSD furnace oil, and 2274-TPD coal residue.

PROCESS EFFICIENCY(26) (Illinois No. 6 coal feed)

	Synthetic Crude	Low Sulfur Fuel Oil
Thermal Efficiency:		
Product only, %	74.7	62.6
Overall, including net utilities, %	62.7	56.0
Utilities, per ton dry coal		
Power, kWh	201	157
Fuel (coal), MM Btu	0.96	0.
Raw water, gal.	661	501
Hydrogen Consumption:		
wt% MAF coal liquefied	5.44	3.57
SCF/bbl net oil	6210	4354
Catalyst: (27)		
Average life of cobalt molybdenum catalyst for low sulfur fuel, lb coal/lb catalyst	N.A.	1000

UNIQUE FEATURES OF PROCESS

Main features of the process are:

Coal slurry hydrogenation in a catalytic ebullating bed reactor, recycle of residuum to the reactor, product oil used to slurry coal, and addition of molecular hydrogen to reactor.

PROCESS STATUS

Development Status:

Three-ton-per-day continuous process development unit began operation 1971.

Design for a nominal 600-TPD pilot plant began September 1974 for construction in Catlettsburg, Kentucky.

HRI Patents:

U.S.-3,519,553
U.S.-3,519,555
U.S.-3,540,995
U.S.-3,617,474
U.S.-3,700,584

Location of Development Work:

PDU located at HRI laboratory, Trenton, New Jersey. Scale pilot plant to be located adjacent to the Ashland Oil Refinery at Catlettsburg, Kentucky.

History:

The H-Coal process is a related application of the ebullated bed H-Oil process, developed by HRI and Cities Service Oil Company to convert heavy petroleum oil residues into lighter fractions. First H-Oil commercial installation put into operation in 1963.

Early development of the H-Coal process began mid 1960s in 25 lb per day bench-scale units sponsored by the Office of Coal Research (OCR) until September 1967. From 1968 to 1970 the program was supported by ARCO, and from 1971 to 1972 by a consortium of six companies. A 3-TPD PDU began operation 1971. From 1973 to 1974 program supported by a consortium of four companies. Since 1974 four oil companies, EPRI, and DOE have sponsored the program.

Approximately 1200 days of operation have been obtained in the bench-scale units, and 147 days in the PDU.

PDU tests since 1974 have been directed towards confirming the design basis for a 600-TPD pilot plant. Liquid solid separation schemes are also currently being studied.

Proposed pilot plant designed to process:

- 633-TPD coal to 1920-BPSD 0.7 wt%S fuel oil
  or,
- 210-TPD coal to 740-BPSD synthetic crude.

Problem Areas:

Separation of solids from 975°F plus oil in low sulfur fuel oil production mode.

REFERENCES

26. Johnson, C.A. et al, *Present Status of the H-Coal Process*, Paper presented at The Institute of Gas Technology Symposium: "Clean Fuels from Coal," Chicago, Illinois, September 10-14, 1973.

27. Johnson, C.A. et al, *H-Coal Process Development*, Paper presented at 68th Annual Meeting, The American Institute of Chemical Engineers, Los Angeles, California, November 16-20, 1975.

28. Johnson, E.S., *Hydrogenation of Low Rank Coal*, U.S. Patent No. 3,700,584, October 24, 1972.

29. Hendrickson, *Synthetic Fuels Data Handbook*, Cameron Engineers, Inc., 1975, pp. 215-222.

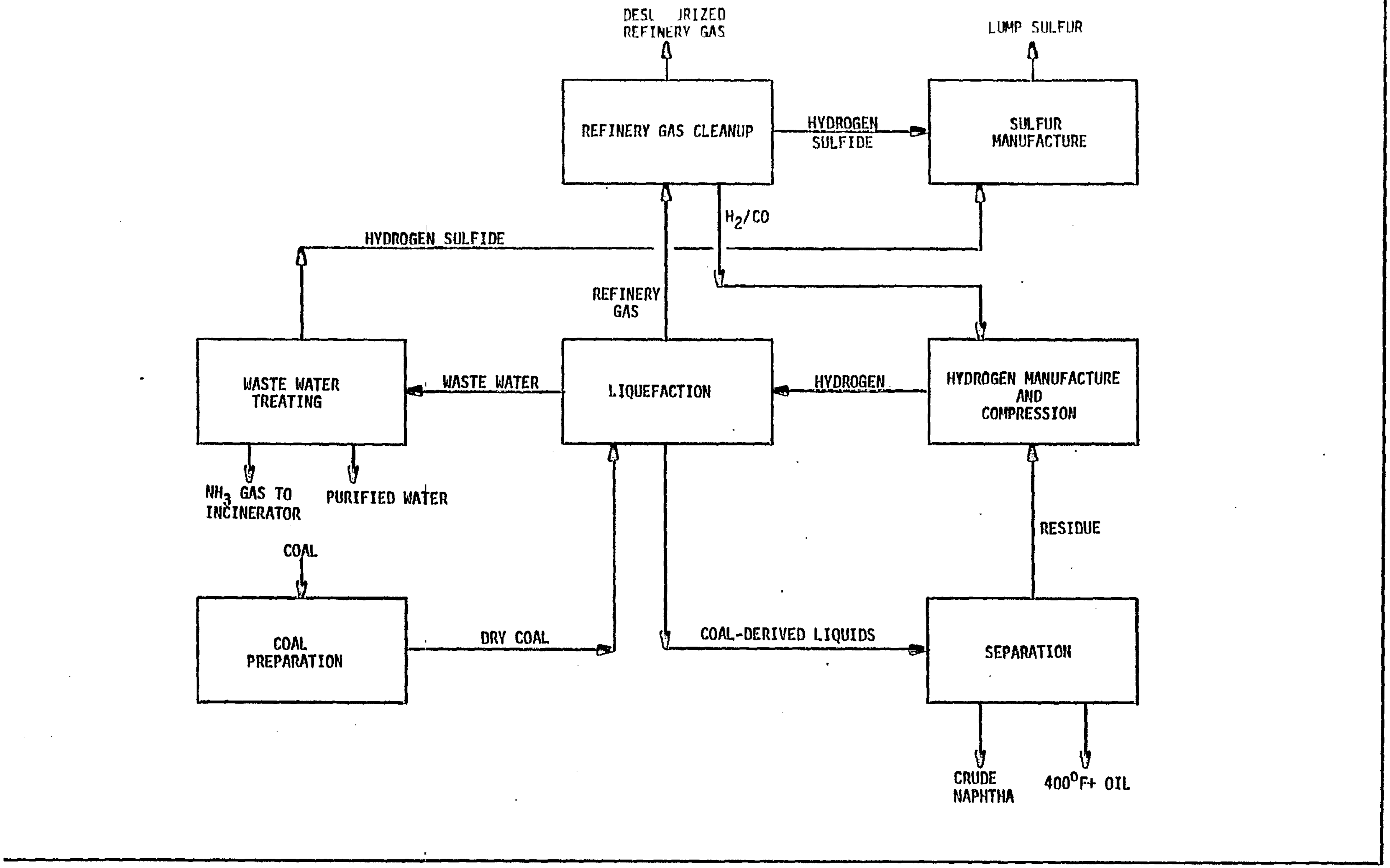

Figure 1 - Block Flow Diagram of H-Coal Plant (26)

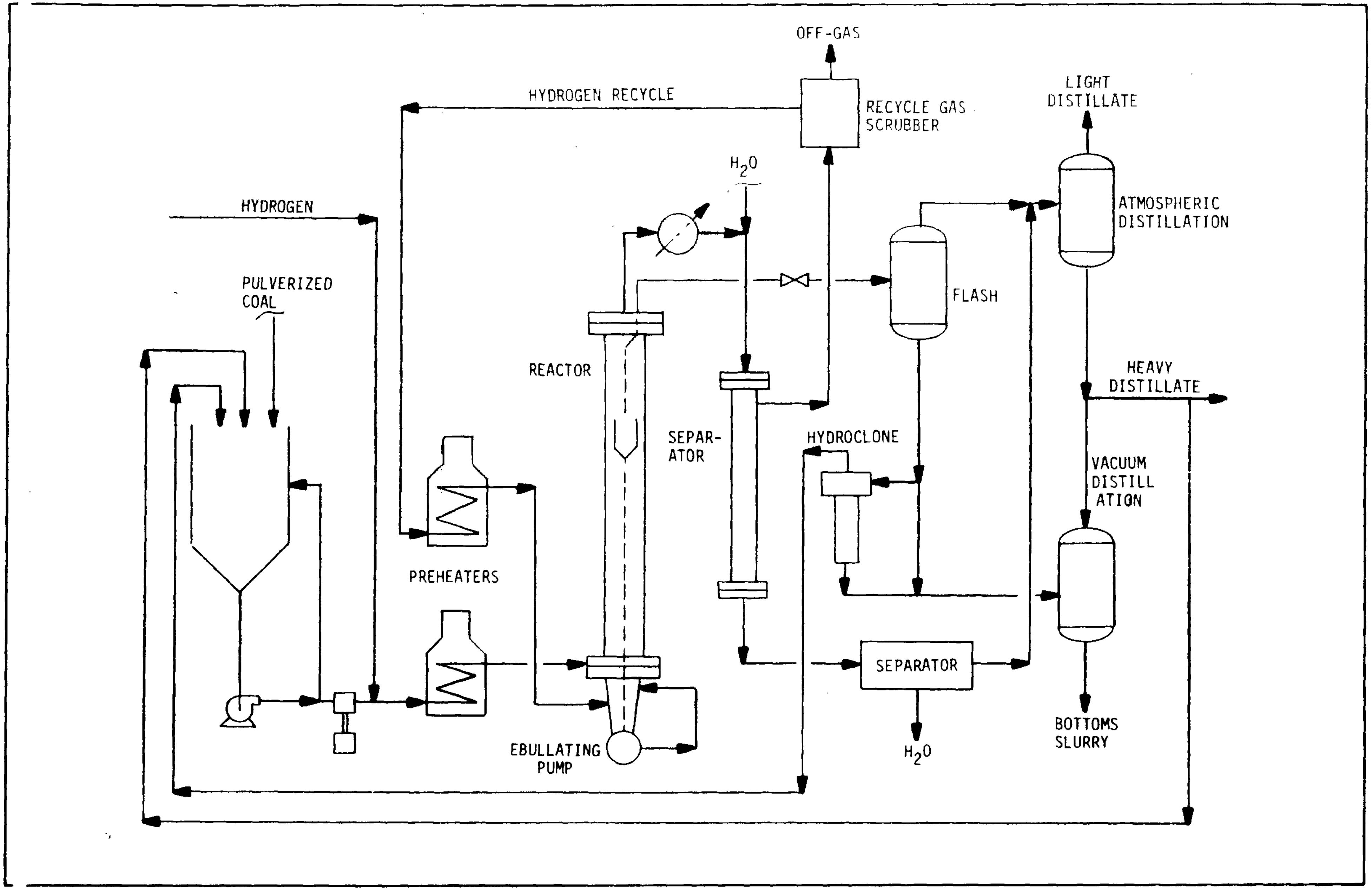

Figure 2 - H-Coal Process Development Unit (27)

PROCESS INFORMATION EXTRACT

PROCESS 1.2.4 Clean Fuel from Coal (CFFC)

Process Type:	Hydroliquefaction, Direct, Catalytic
Main Products:	Gas Naphtha Fuel Oil
Development Status:	Pilot Plant

PROCESS DEVELOPER

C-E Lummus (Subsidiary of Combustion
Engineering, Inc.)
Bloomfield, New Jersey

PROCESS SPONSORS

(1) Combustion Engineering, Inc.
(2) DOE (30)

PROCESS DESCRIPTION (31), (32), (33), (34), Figure 1

After crushing, sizing, and drying, coal is slurried with a pasting solvent (boiling range of 600° – 900°F). The solution enters the catalytic hydrodesulfurization section where it is liquefied and desulfurized in an upflow, ebullating reactor. Reactor effluent is let down in pressure and flash separated (atmospheric or vacuum flash) to remove materials boiling up to 550°F. The flash vapors are separated and cleaned to remove H_2S and product naphtha. The flashed coal solution enters the de-ashing section where it is mixed with an anit-solvent (boiling range about 400° – 500°F). This anti-solvent, or de-ashing promotor liquid, has a Watson characterization factor which is at least 0.25 above that of the pasting solvent. The separation is accomplished in a gravity settler under sufficient pressure to prevent vaporization at medium temperatures (300°F – 500°F). Underflow from the settler is stripped to recover tars from the undissolved coal and ash residue. The tars and overflow liquids are fractionated to the following:

- Anti-solvent recycle
- Pasting solvent recycle
- CFFC liquid fuel

Hydrogen is generated by gasification of the ash plus residue from the stripped underflow and subsequent CO shifting.

OPERATING CONDITIONS(33)

	Hydro-Desulfurizer	Gravity Settler
Temperature, °F	750 – 850	300 – 500
Pressure, psia	1000 – 4000	0 – 300
H_2 Partial Pressure, psia	800 – 3000	N.A.
Liquid Hourly Space Velocity, h^{-1}	1 – 4	N.A.
Residence Time, h	N.A.	0.5 – 3
Catalyst	Co-Mo	None
Solvent Weight Ratios:		
Solvent to Coal	1.5 – 5 to 1	N.A.
Anti-solvent to Coal Solution	N.A.	0.3 – 1.5 to 1

PRODUCTS(31)

Basis: Illinois No. 6 Coal Feed

	wt% Dry
C	68.4
H	5.0
N	1.2
S	4.0
O	11.4
Ash	10.0
	100.0

Moisture with feed, wet wt%	8.0
HHV, Btu/lb as received =	11,600

Yields:

	wt% MAF Coal Feed
Water, CO, CO_2, NH_3	13.33
Hydrocarbon gases	2.88
H_2S	3.61
C_5 - 400°F naphtha	4.55
400 - 900°F distillate	23.30
Residual solids-free product (+900°F)	33.51
Unconverted coal (ash free basis)	5.00
Residual product associated with solids (unconverted coal + ash)	16.82
	103.00

Characteristics:

Naphtha

Boiling range	C_5 — 400°F
Gravity	48.7° API
HHV	19,200 Btu/lb

CFFC (pilot plant blend)

Distillation (Vacuum Engler):

vol%	
IBP	392°F
1	418°F
2	421°F
5	446°F
10	469°F
20	505°F
28.2	536°F
30	538°F
40	615°F
50	790°F
60	857°F
66.5	900°F
Residue (+900°F Content)	39.1 wt%

Viscosity	
@ 140°F	5900 SUS
@ 180°F	864 SUS
@ 210°F	325 SUS
Elemental Analysis (Ash free basis), wt%	
% Carbon	88.78
% Hydrogen	6.84
% Sulfur	0.34
% Nitrogen	1.21(a)
% Oxygen (by diff.)	2.79
% Chlorine	0.025
Trace Metal Analysis	
Sodium	5.7 PPM
Vanadium	1.0 PPM

NOTE

(a) Using standard high activity hydrodesulfurization catalyst used in the CFFC process. Expected product nitrogen content would be <0.5% using new Lummus catalyst with enhanced denitrification capability.

Sulfur Content	0.34 wt%
Ash Content	0.05 wt%
Pour Point	65°F
Sp. Gr. 170/60°F	1.100
Sp. Gr. 268/60°F	1.060
Conradson Carbon	25.0 wt%
Flash Point Pensky Martin Closed Cup	215°F
Fire Point Cleveland Open Cup	265°F
Higher Heating Value	16,800 ± 200 Btu/lb

INTENDED PRODUCT USE/MARKET

(1) Gas: for fuel within coal conversion plant.

(2) Naphtha: May be sold as a high-aromatics blending stock.

(3) CFFC Fuel: May be sold as a low-sulfur fuel oil. It also might be upgraded to a synthetic crude or to a gasoline boiling range material by the addition of hydroprocessing steps.

(4) Mineral Residue: May be used internally in a CFFC plant for hydrogen generation.

PROCESS EFFICIENCY(31)

Yields:

	% of Coal Feed HHV
Integrated plant	
• excluding purchased power	65.6%
• including purchased power at 9030 Btu/kW/h	63.8%

Utilities:

Basis: 1 ton MAF coal

Coal pasting, slurrying, and preheat (excluding coal crushing and drying)

Cooling water	384 gal.
Fuel	0.5 MMBtu
Brake horsepower (turbines plus motor drive)	20 hp

Hydroliquefaction (including H_2S removal from off gases)

Steam	324 lb
Cooling water	888 gal.
Fuel	0.42 MMBtu
Brake horsepower	16 hp

Fractionation and deashing (including solvent recovery)

Steam	(16) - net production
Cooling water	1,872 gal.
Fuel	0.82 MMBtu
Brake horsepower	10 hp

Hydrogen Consumption: 2 – 3 wt% MAF

Catalyst Consumption: 1.5 tons of coal per lb of catalyst, based on 3 wt% moisture coal feed

UNIQUE FEATURES OF PROCESS

- The gravity settler is claimed to be a major breakthrough for coal hydroliquefaction processes (catalytic or non-catalytic). The C-E Lummus de-ashing technique using the anti-solvent eliminates the need for filtration separations. The total cost of anti-solvent de-ashing has been projected to be significantly lower than pressure precoat filtration. It is believed that the anti-solvent used promotes agglomeration of ash and undissolved coal particles to effect faster settling.(32) (34)
- The hydrodesulfurizer is similar to the H-Coal process in that it is an ebullating catalytic reactor using Co-Mo catalyst. Hydrogen consumption (2 – 3%) is lower than H-Coal (3 – 5%).
- It is claimed that hydrogen utilization efficiency for CFFC fuel is better than that for SRC fuel.(31)
- C-E Lummus claims a new catalyst under development(31) that selectively denitrogenates the coal to low values without substantially increasing the hydrogen consumption.

PROCESS STATUS

Development Status:

The process is being investigated by C-E Lummus under a DOE contract. The process as described had three coal liquefaction patents (U.S. Patent No. 3,852,182; 2,852,183; and 3,856,675) awarded in 1974 and one (U.S. Patent No. 3,974,073) in 1976. C-E Lummus claims the process is based on proven equipment, hence no equipment development is necessary.

Location of Development Work:

C-E Lummus Engineering Development Center Annex
New Brunswick, New Jersey

History:

C-E Lummus has been involved with coal liquefaction and patents dating back to 1965. Work on the present process started with bench scale work in 1971. Currently, the small scale pilot plant is operational under a DOE contract.

Problem Areas:

- Catalyst attrition

REFERENCES

30. Personal Telephone Communication to A.A. Simone (C-E Lummus Company) from J.F. Ferrall (The Ralph M. Parsons Company), March 7, 1977.

31. Simone, A.A., "Clean Fuel from Coal Process," *Combustion*, May 1976, pp. 15-19.

32. Sze, M.C. and G.J. Snell, "A New Process for Removing Ash from Coal Liquefied by Hydrogenation," *Proceedings of the American Power Conference*, Vol. 37, 1975, pp. 315-321.

33. Snell, G.J., *Coal Liquefaction*, U.S. Patent No. 3,852,183, December 3, 1974.

34. Sze, M.C. and G.J. Snell, *Application of the C-E Lummus Deashing Technique to Coal Liquefaction Processes*, Paper presented at ACHEMA, Frankfurt, West Germany, June 20-26, 1976.

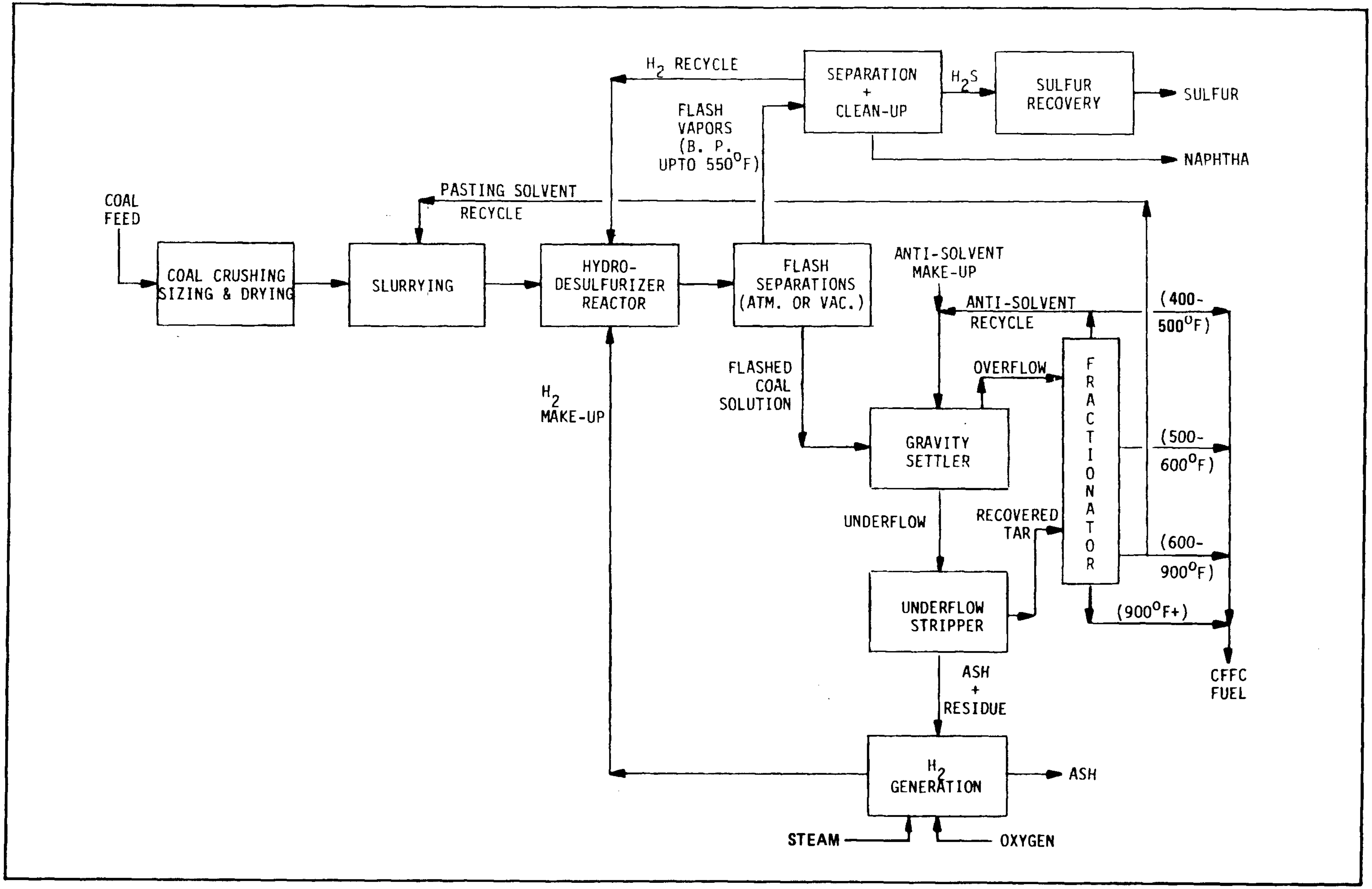

Figure 1 - Block Flow Diagram C-E Lummus CFFC Process (31)

PROCESS INFORMATION EXTRACT

PROCESS 1.2.5	Conoco Zinc Halide Hydrocracking
Process Type:	Hydroliquefaction, Direct, Catalytic
Main Products:	Naphtha
Development Status:	Bench scale
PROCESS DEVELOPER	Conoco Coal Development Company Library, Pennsylvania
PROCESS SPONSOR	DOE

PROCESS DESCRIPTION(35) (36)

The bench-scale continuous zinc chloride ($ZnCl_2$) hydrocracker consists of a hydrocracker section (Figure 1) and a catalyst regeneration section (Figure 2). In the hydrocracker, dry coal, ground to less than 150 microns, is mixed with oil and fed as a slurry, along with zinc chloride melt (catalyst) and hydrogen, to the first of two reactors arranged in series. These reactors are of the stirred autoclave type. (Reactors fitted with hydrogen sparger rather than a stirrer will be tested in a PDU.) The reaction mix passes from the first reactor through the second reactor to the melt-vapor separator. The spent melt, containing char, ash, and most of the zinc halide catalyst, flows by gravity from the separator to the spent melt collection system. The hot gaseous reaction products pass through the melt-vapor separator, and are cooled to recover condensable products.

In the catalyst regeneration unit spent melt from the hydrocracker is fed to an air fluidized bed (silica sand), along with a stream of anhydrous hydrogen chloride gas. The spent melt contains sufficient carbon for the adiabatic regeneration of the melt via combustion of the carbon with excess air.

The $ZnCl_2$ vapor leaves the reactor along with the combustion gases and entrained ash. The entrained ash is removed by a hot gas cyclone, and the gas is cooled to recover regenerated $ZnCl_2$ melt. The cooled combustion gases are then passed through a caustic scrubber where $ZnCl_2$ fog, water, and HCl are removed.

Zinc losses to the cyclone ash are due to the formation of ZnO, by hydrolysis of $ZnCl_2$.

$$ZnCl_2 + H_2O \rightleftharpoons ZnO + 2HCl$$

Ash from the cyclone is treated at cyclone temperature with hydrogen chloride in an inert gas stream for recovery of zinc from the cyclone ash.

OPERATING CONDITIONS (36)

Hydrocracker:	
Pressure, atm	238
Temperature, °F	775
Slurry oil/coal, wt/wt	1.5
Hydrogen/coal, SCF/lb	30
$ZnCl_2$/coal	1.5
(operation at a ratio of 1.0 has also been demonstrated)	
Coal, $lb/ft^3/h$	38
Catalyst melt, minutes	95
Conversions:	
H_2 wt% MAF coal	7
Coal converted -750°F product, wt%	75.3
Carbon in ash, wt%:	6.4
Catalyst:	Zinc chloride melt
Regenerator:	
Pressure, psig	3
Temperature, °F	1800
Superficial fluid velocity, fps	1

Melt Composition, wt%	
Ash	6.1
Char	6.0
Inlet Gas Composition, vol%:	
Air	94.5
Anhydrous HCl	5.5
Air (% Stoichiometric)	115
Zinc Recovery, wt%	98.77

Zinc recovery can be increased to 99.98% by secondary treatment of cyclone ash using 10% HCl/90% nitrogen at cyclone temperature for 2.25 hours.

Bed Solids:	28 - 48 micron silica sand

Reaction Variables, batch autoclave study:

- Increasing the hydrogen pressure from 2000 to 3000 psig at 725°F results in higher coal conversion, higher C_4-390°F distillate yield, lower gas yield, lower +890°F MEK-insolubles.
- Increasing the temperature from 725°F to 750°F, at 2000 psig and 60 minutes residence time, increases gas yield and +890°F MEK-insolubles even though yield of distillate, hydrogen consumption, and conversion remain constant.
- The yield of distillate and gases increases with increasing residence time.

Coals Processed:

Sub-bituminous coal (Colstrip) from the Rosebud Seam in Montana.

PRODUCTS (36)

Coal feed basis:	Colstrip (Kiln dried)
Coal moisture, wt%:	
To Unit	3.59

Coal analysis (dry):

Proximate, wt%:	
Ash	10.69
Volatile matter	41.58
Fixed carbon	47.58
Ultimate, wt%:	
Carbon	66.93
Hydrogen	4.39
Nitrogen	1.01
Sulfur	0.88
Oxygen	16.10
Ash	10.69
HHV, Btu/lb (dry basis): %MAF	11,570
Neville Solvent, wt%:	
445 - 620°F	60.4
620 - 890°F	28.2
+ 890°F	11.4

Organic products, excluding water, % MAF:

	Continuous*	Rocking Autoclave 3000 psi H_2/750°F/15 min $ZnCl_2$/Coal = 2.5
CO	1.22	1.2
CO_2	1.96	2.5
CH_4	1.57	0.4
C_2H_6	3.56	0.7
C_3H_8	16.62	3.8
iC_4H_{10}	15.09	6.8

*Only 26 - 27% of these products are derived from the coal.

	Continuous*	Rocking Autoclave 3000 psi H_2/750°F/15 min $ZnCl_2$/Coal = 2.5
nC_4H_{10}	1.65	0.8
C_5 - 392°F	108.84	39.3
392 - 446°F	33.01	
446 - 617°F	33.56	
617 - 887°F	29.06	
+887°F	16.46	28.3
MEK Insolubles	8.18	5.3
	270.78*	89.1

*Only 26 - 27% of these products are derived from the coal.

Characteristics (corresponds to autoclave results):

+482°F Oil, wt%:

Hydrogen	12.48
Carbon	86.79
Nitrogen	0.0
Oxygen	0.69
Sulfur	0.04
Aromatics	34.3
Saturates	65.2
Olefins	0.5

392 - 752°F Oil, wt%:

Hydrogen	9.8
Carbon	85.25
Nitrogen	0.89
Oxygen	3.96
Sulfur	0.10

+887°F MEK-Soluble Residue, wt%:

Hydrogen	8.09
Carbon	87.64
Nitrogen	0.66
Oxygen	2.95
Sulfur	0.13
Ash (essentially $ZnCl_2$)	0.53

INTENDED PRODUCT USE/MARKETS

The objective of this process is the production of clean gaseous and liquid fuels from coal, with emphasis on the production of high-octane gasoline.

PROCESS EFFICIENCY

Hydrogen:	
wt% MAF Coal	7.0
Catalyst:	
$ZnCl_2$, lb/lb coal	0.03
(99.98% catalyst recovery, $ZnCl_2$/Coal = 1.5)	
Chemicals:	
Hydrogen chloride, lb/lb coal	Negligible

UNIQUE FEATURES OF PROCESS

Main features of the process are: Direct hydrocracking of coal in a melt of zinc chloride, and recovery of the zinc chloride from the solid residue by combustion in a fluidized bed in the presence of hydrogen chloride.

PROCESS STATUS

Development Status:

2 lb/h bench scale hydrocracker and 4 lb/h bench scale operational since 1975. Run periods of up to 12 hours duration achieved. 100 lb/hr PDU being engineered.

Patents:

U.S. - 3,355,376

U.S. - 3,629,159

Location of Development Work:

Conoco Coal Development Company Research Division
Library, Pennsylvania

History:

The zinc halide hydrocracking process was developed during the Office of Coal Research work on Project Gasoline (1963-1968) and used for the hydrocracking of solvent refined coal to gasoline. The bench scale zinc halide hydrocracker and regenerator used for Project Gasoline were rebuilt and modified in 1975 for the use of coal feed and coal extract.

A 100 lb/hr process development unit was designed in 1975 and long-lead-time items ordered. Engineering design is underway.

Problem Areas:

Corrosion resistance and reliability have to be demonstrated for extended run times. Most runs have been less than 12 hours duration.

Zinc chloride inconel condenser coil corroded through after 33 hours operation.

REFERENCES

35. CONOCO Coal Development Company, "Zinc Halide Hydrocracking Process for Distillate Fuels from Coal," (FE-1743-1), *Annual Technical Progress Report for 1975, including Quarter ending January 31, 1976,* February 15, 1976. ERDA Contract No. E(49-18)1743

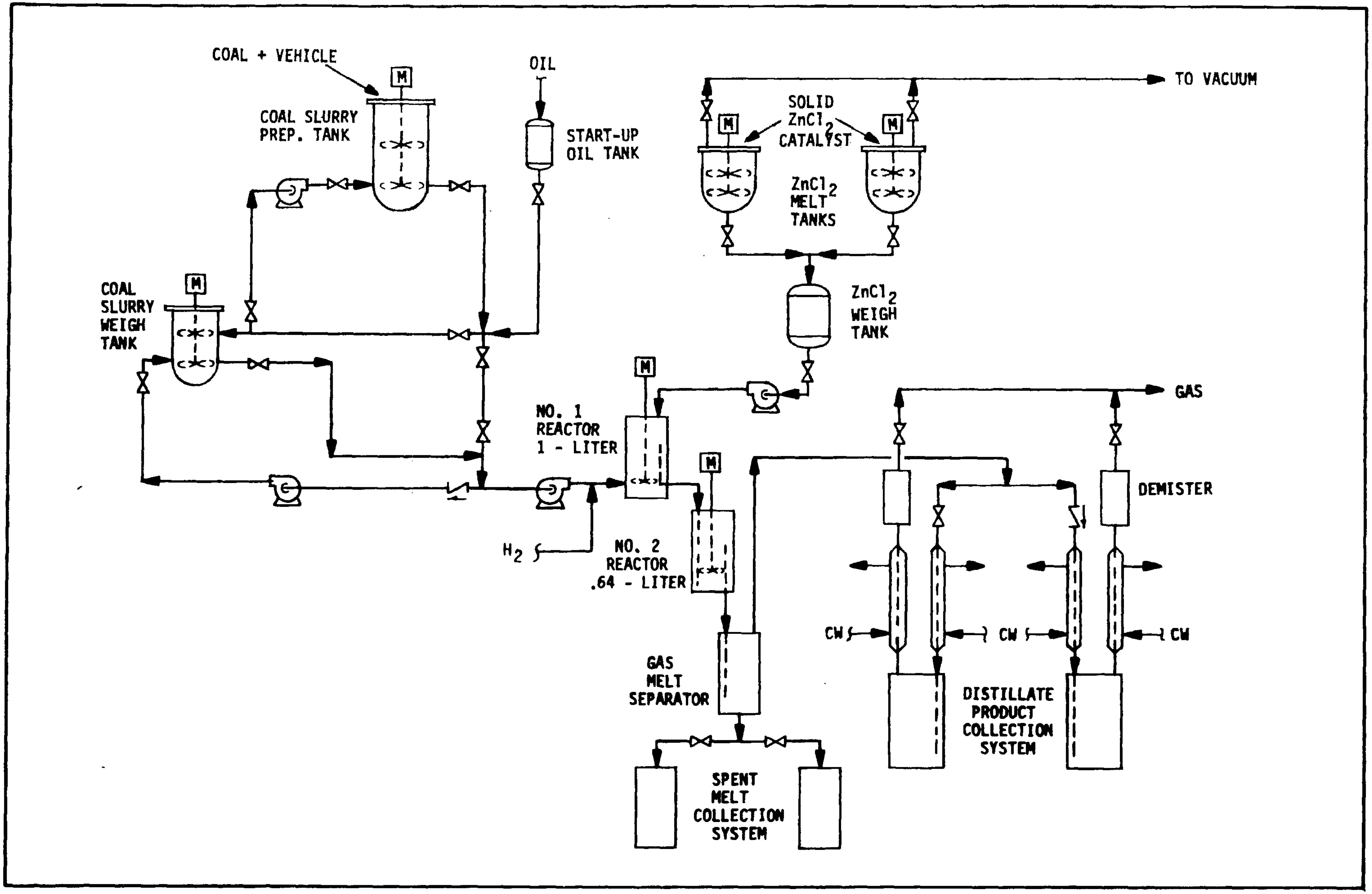

Figure 1 - Simplified Flow Diagram Bench-Scale (2 lb/h) $ZnC1_2$ Hydrocracker (35)

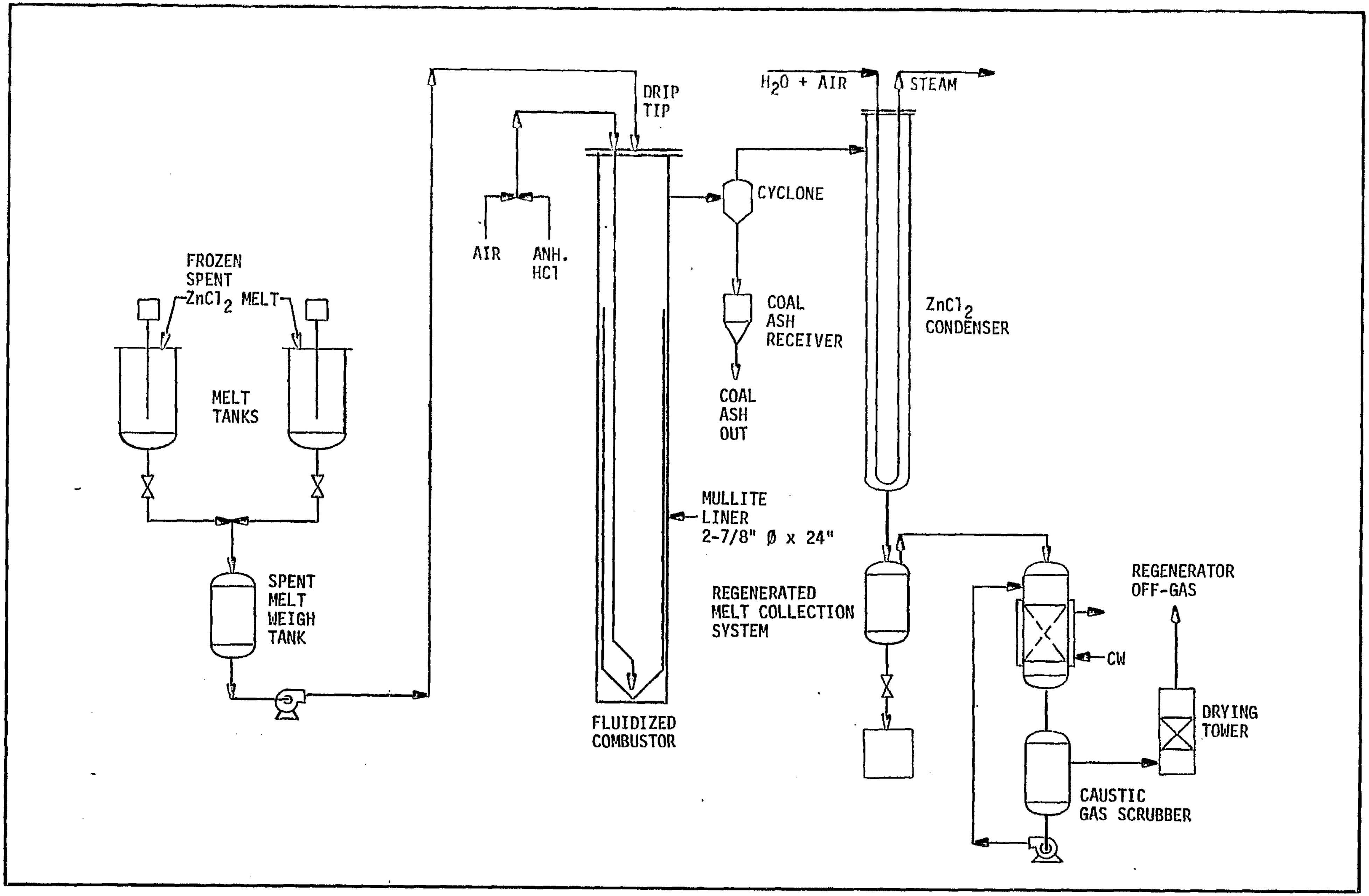

Figure 2 - Simplified Flow Diagram Bench-Scale (4 lb/h) $ZnCl_2$ Regenerator (35)

PROCESS INFORMATION EXTRACT

PROCESS 1.3.1	Exxon Donor Solvent (EDS)
Process Type:	Hydroliquefaction, Indirect (Donor Solvent)
Main Products:	Naphtha and wide boiling range low-sulfur fuel oil
Development Status:	Pilot plant.
PROCESS DEVELOPER	Exxon Research and Engineering Company Florham Park, New Jersey
PROCESS SPONSORS	Exxon Corp. Electric Power Research Institute (EPRI)

PROCESS DESCRIPTION(37)

The EDS process contains four major process unit operations:

(1) Liquefaction

(2) Separation (solvent recovery)

(3) Solvent Hydrogenation

(4) Hydrogen Manufacture

The integration of these units is depicted in the EDS block flow diagram (Figure 1). A simplified flowsheet of the EDS liquefaction and separation units is shown in Figure 2.

In the liquefaction unit, crushed coal is slurried with recycle solvent and preheated to reaction temperature. The hot coal slurry is mixed with preheated hydrogen recycle and the combined stream fed to the liquefaction reactor.

The product from the liquefaction reactor is separated into gas and liquid/residue streams in the hot high-pressure separator. The gas stream is cooled to remove heavy hydrocarbons and water, and then scrubbed to remove acid gases.

The liquid/residue stream is reduced in pressure and flash gases separated. The flash-gas is processed to recover heavy hydrocarbons. The slurry product is sent to the separation unit where gas, naphtha, recycle solvent, distillate, and heavy bottoms (containing all the solid residue from the liquefaction, and some very high boiling hydrocarbons are separated by distillation. Liquids boiling up to 1000°F are recovered from the product slurry by vacuum distillation, and then further fractionated into light vacuum gas oil, boiling up to 700°F, and a heavy vacuum gas oil product. The light vacuum gas oil is combined with other recovered liquid hydrocarbon streams and this material is fed to the hydrotreating system.

The liquid product is fractionated to remove lighter and heavier boiling range components from the recovered solvent. The excess solvent is removed from the system as a product.

In an earlier alternative version of this process only recycled solvent is hydrotreated.(38)

Fixed-bed catalytic reactors are used for solvent hydrogenation. The reactors are provided with quench hydrogen for temperature control. High pressure hydrogen-rich gas is cooled to remove heavy hydrocarbons and water, and then scrubbed to remove acid gases. In an earlier alternative version of the process, solvent was hydrogenated with gas from the HT/MP separator, and liquid feed was used for quench temperature control.(39)

Scrubbed gases from the hydrotreater and liquefaction reactors are compressed and recycled to the liquefaction unit.

Hydrotreated liquid is reduced in pressure and flash gases separated. The flash-gas is processed to recover heavy hydrocarbons. Flashed liquid is fractionated, as described above, for solvent recovery.

Recovered solvent is slurried with fresh coal to repeat the cycle. Heavy bottoms and solid residue go to a gasifier for hydrogen production.

Gas generated from the process is used as fuel and for hydrogen manufacture.

OPERATING CONDITIONS

Liquefaction:	(37)	(39)
Coal Processed:	Bituminous/ Sub-bituminous	Illinois No. 6
Pressure, atm	100 - 175	123
Temperature, °F	700 - 900	825
Solvent/Coal, wt/wt	1.2 - 2.6	0.5

	Bituminous/ Sub-bituminous	Illinois No. 6
H_2/Coal, wt% MAF coal	N.A.	5.75
SCF/lb coal		8.2
Residence Time, min	N.A.	36
Conversions		
H_2, wt% MAF coal	N.A.	1.74
H_2, SCF/bbl oil	N.A.	630
Catalyst	none	none
Solvent Hydrogenation:		
Pressure, atm	80 - 210	122
Temperature, °F	500 - 840	725
H_2/Oil, SCF/bbl	N.A.	4730
Space Velocity, wt/hr/wt	0.2 - 4.0	4.0
Conversion		
H_2, wt% MAF coal	N.A.	1.30
H_2, SCF/bbl oil	N.A.	630
Catalyst	commercial multi-metallic catalysts	cobalt molybdate on silica-alumina

Reaction Variables:

A wide range of process conditions have been studied by Exxon. Results of these studies have not been published. Coal conversion has been correlated with a solvent quality index.(37) The quality index varies with the degree of solvent hydrogenation. Details of the index are proprietary. These correlations show that the addition of molecular hydrogen to the liquefaction reactor significantly reduces the solvent quality requirement.

Coal Processed: Bituminous and sub-bitiminous coals.

---

*Partially hydrogenated hydrocarbons produced by the coal liquefaction process process (boiling range 40 - 700°F).

PRODUCTS(37)

Coal Feed Basis:

Coal Analysis:	Illinois No. 6	
Proximate, wt% (dry)		
Ash	9.41	
Volatile matter	41.45	
Fixed Carbon	49.15	
Moisture, wt%		
To unit	0	
As received	16.5	
Ultimate, wt% (dry):		
Carbon	69.67	
Hydrogen	5.05	
Nitrogen	1.85	
Sulfur	4.19	
Oxygen	9.67	
Ash	9.58	
HHV, Btu/lb (dry basis):	12,814	
Yield, MAF coal:		
Total liquid yield, wt%		51 - 60
Total liquid yield, bbl/ton		3 - 3.4
Naphtha, wt%		5 - 25

Characteristics:

	Heavy Naphtha*		392° + Fuel Oil	
	Raw Liquid	Hydrotreated Liquid	Raw Liquid	Hydrotreated Liquid
Nominal Boiling Range, °F:	158/392	158/392	392/1004	392/1004
Distillation, °F:				
10 wt%	223	198	477	462
50 wt%	356	315	694	657
90 wt%	390	360	811	774
Density (g/cm³):	0.87	0.80	1.08	1.01
Elemental Analysis, wt%:				
C	85.60	86.80	89.40	90.80
H	10.90	12.90	7.70	8.60
O	2.82	0.23	1.83	0.32
N	0.21	0.06	0.66	0.24
S	0.47	0.005	0.41	0.04
Higher Heating Value, Btu/lb:	18315	19304	17110	18900

INTENDED PRODUCT USE/MARKETS

(1) Naphtha

The (C_6 - 158°F) and (158 - 392°F) fractions are identified as potentially valuable naphtha blending stocks.

(2) Low-Sulfur Fuel Oil (LSFO)

The LSFO consists of a mid-distillate and a heavy gas oil. Economic studies have shown that these cuts are potentially valuable fuel oil blending stocks.

(3) Chemicals

Specialty product chemicals can be separated from the naphtha and LSFO.

---

*Excludes C_6/158 °F naphtha cut

PROCESS EFFICIENCY

Thermal Efficiency(37)

Product only, %:	80 - 90
Overall, including net utilities, %:	65 - 75

Hydrogen Consumption(39)

wt% MAF coal to liquefaction unit	3.04
wt% liquid yield	6.2 - 7.3

Catalyst:(39)

Cobalt molybdate catalyst life (between regenerations) - over one year

UNIQUE FEATURES OF PROCESS

Main features of the process are: Noncatalytic proprietary liquefaction reactor, production of the donor solvent in a fixed-bed catalytic hydrogenation reactor, addition of molecular hydrogen to the liquefaction reactor, and vacuum distillation to separate unconverted coal, ash, and liquids boiling over 1000°F from the reaction product.

PROCESS STATUS

Development Status:

One-ton-per-day continuous pilot plant constructed July 1965 and put in productive operation. Basic design specifications for a 250-TPD pilot plant made.

Exxon Patents:

U.S. - 3,645,885
U.S. - 3,726,784

Location of Development Work:

Large scale pilot plant to be located adjacent Exxon's Baytown, Texas, refinery.

36. CONOCO Coal Development Company, "Zinc Halide Hydrocracking Process for Distillate Fuels from Coal," (FE-17430-20), *Quarterly Technical Progress Report, February 1 - April 30, 1976,* May 14, 1976. ERDA Contract No. E(49-18)1743

History:

Exxon research and development work on donor solvent coal liquefaction began in 1966. Basic flow plan and reactor design established by 1973 in units ranging from 100 cc batch reactors to a 0.5-TPD integrated pilot plant. A 1-TPD pilot plant put into operation in 1975 to better define operating conditions and arrangement of process steps for the EDS process. Exxon has obtained federal government financial support for the construction and operation of a 250-TPD plant.

REFERENCES

57. Furlong, L.E. et al, Exxon Research and Engineering Company, *Coal Liquefaction by the Exxon Donor Solvent Process*, Paper presented at 68th Annual Meeting, The American Institute of Chemical Engineers, Los Angeles, California, November 16-20, 1975.

58. Harris, G.W. and F.G. Snow, *Turbulence-Free Up-flow Liquefaction Zone,* U.S. Patent No. 3,645,885, February 29, 1972.

59. Correer, J.J. and J.M. Hockman, *Liquefaction and Hydrotreating Zones Operated at Moderate Pressures,* U.S. Patent No. 3,726,784, April 10, 1973.

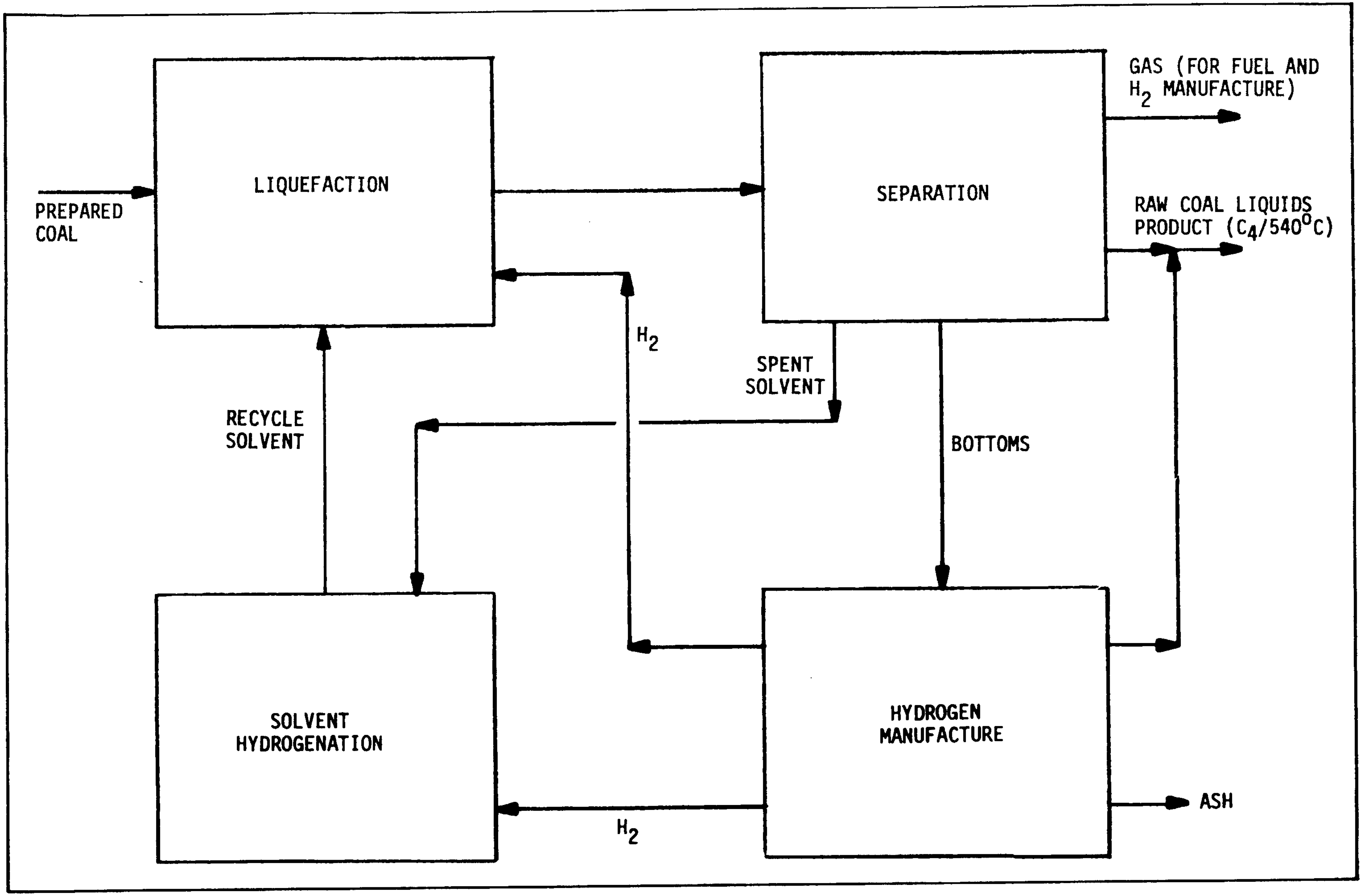

Figure 1 - EDS Simplified Block Diagram (37)

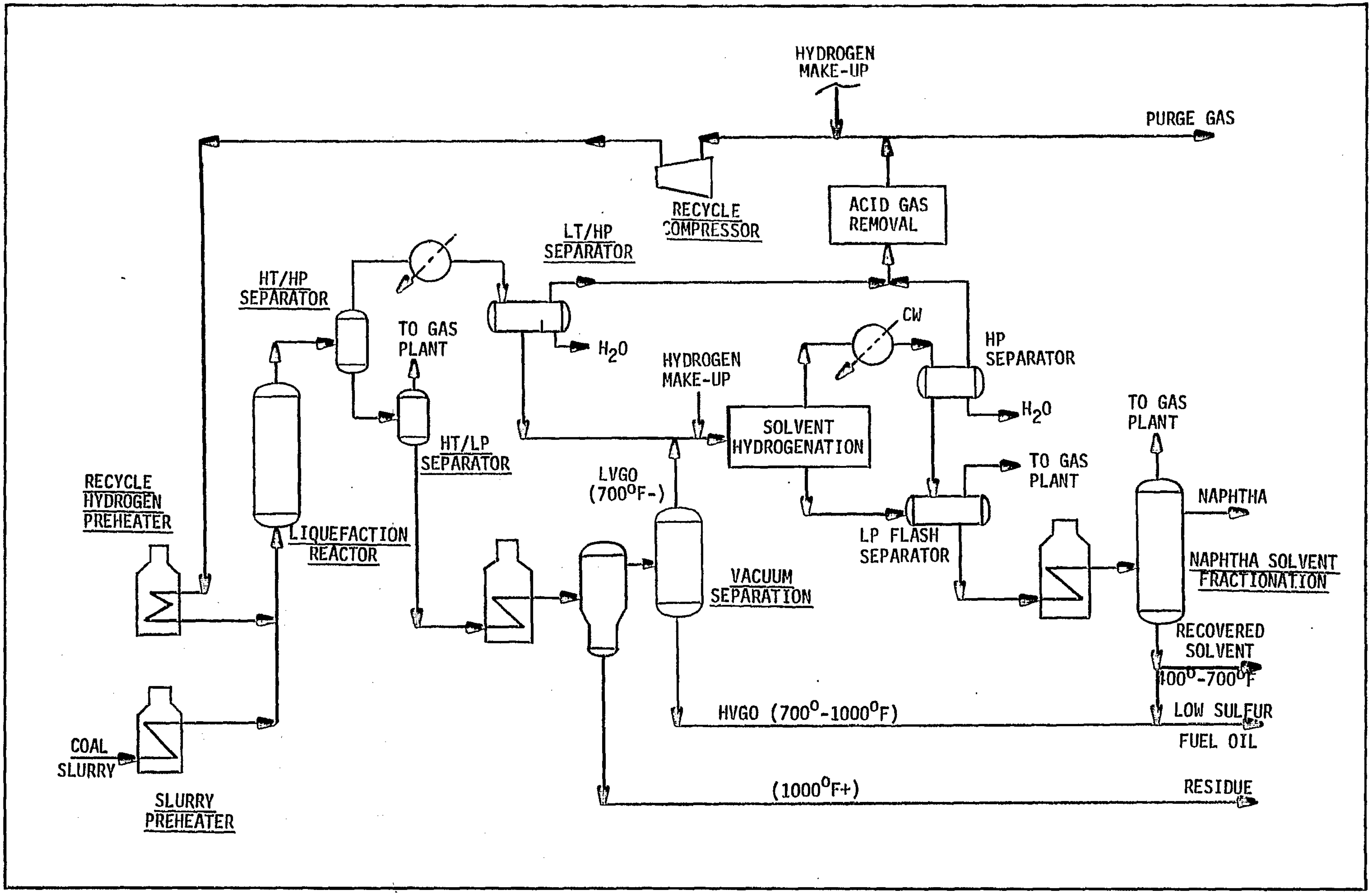

**Figure 2 - Flow Diagram
EDS Liquefaction and
Product Separation (37)**

PROCESS INFORMATION EXTRACT

PROCESS 1.3.2	ADL Extractive Coking
Process Type:	Hydroliquefaction, Indirect (Donor Solvent)
Main Products:	Syncrude, Coke
Development Status:	Bench scale and pilot plant tests.

PROCESS DEVELOPER

Arthur D. Little Inc.
Acorn Park, Cambridge, Mass.

(Experimental program to verify basic concepts is a joint venture among ADL, Foster-Wheeler and the Pittsburgh Energy Research Center (PERC).)

PROCESS SPONSOR   DOE

PROCESS DESCRIPTION

The ADL Extractive Coking process (Figure 1 Block Diagram) contains four major process unit operations:

(1) Liquefaction

(2) Separation (solvent recovery/solids removal)

(3) Solvent hydrogenation

(4) Hydrogen manufacture

The process involves the use of a hydrogen-donor solvent to achieve liquefaction and the use of cracking and coking to separate the product as an overhead vapor from the coal ash and the heavy portion of the coal extract.

Crushed and ground coal and mildly-hydrogenated recycle solvent are added to the coke drum. The coal is extracted by the solvent in the drum for approximately one to two hours, under sufficient pressure to maintain most of the solvent as a liquid. During this extraction, hot solvent vapor passes through the drum continuously to supply heat and agitation. After the extraction, the drum pressure is gradually reduced to flashoff solvent and light ends, with incoming hot solvent vapor supplying the latent heat of vaporization. The drum contents are subsequently heated with more hot vapor and allowed to coke. The drum is cooled and decoked hydraulically while a second drum goes through the extraction-coking cycle.

The vapors from the coke drum are continuously fractionated to separate recycle solvent from the products. A portion of the recycle solvent is hydrotreated and stored for use as hydrogen-doner solvent. The other portion is revaporized and superheated to provide heat to the coke drum. The fractionation tower also separates the products into gases, light extract (C_4 - 450°F), middle boiling range material (450 - 750°F), and heavy extract (750 + °F). A portion of the high-ash char is gasified to produce hydrogen and medium-Btu fuel gas.

OPERATING CONDITIONS

Operation	Cycle Time Hours	Pressure psig	Temperature °F
First Slurry Charge	0.7		
Extraction	2.0	100	750
Solvent Flash	4.0		
Second Slurry Charge	0.4		
Extraction	1.2	100	750
Solvent Flash	2.5		
Heatup and Coking*	5.2	30	850
Switching Drums	0.5		
Steaming Out	2.0		
Water Cooling	2.0		
Draining Water	2.0		
Hydraulic Decoking	4.0		
Headup and Test	1.0		
Warmup	4.5		
	32.0		

*Note: Second drum is preheated with a portion of the solvent vapors during this step.

Conversions:

H_2, wt% MAF coal: 2

Coal converted (gas + liquids) wt% MAF: 45 - 55

Catalyst: None

Reaction Variables:

- The experimental program has only obtained a limited number of runs so far. All these runs were conducted using solvent obtained from a SRC pilot plant, and used batchwise or on a once-through basis. The limited amount of data collected indicates, for both PDU and autoclave runs, that conversion passes through a maximum as the solvent hydrogen content is increased by hydrogenation. The largest conversions have been obtained using a 8.7 weight percent hydrogen content solvent, hydrotreated solvent with 1.1 weight percent hydrogen added.
- Liquid yield varies inversely with coking pressure, but the products are lighter as pressure increases.

Coals Processed:

Pittsburgh seam coal.

PRODUCTS

Coal - Pittsburgh seam (10% ash)

Yield, extractive coker, MAF coal basis - Excludes hydrogen manufacture and utilities.

	Autoclave	PDU
	(8.7 wt% H solvent)	
Gases, wt%	N.A.	3-5
Liquids, wt%	44 - 55	44 - 52

Characteristics:

Yield predictions indicate the following H:C atomic ratios:

Gases	2.4 - 2.8
Liquids	1.2 - 1.25
Coke	0.45 - 0.5
Sp. gr (60°F/60°F)	0.96

INTENDED PRODUCT USE/MARKETS

(C_4 - 450°F) / (450 - 750°F)

(1) The light and medium boiling range material requires hydrocracking or hydrotreating to convert to a low-sulfur distillate that can be processed into gasoline, jet fuel, and household heating oil.

(2) Middle distillates, being rich in polycylics, are not suitable for olefin production.

Heavy Extract (750 + °F):

Heavy extract may be blended with lighter fractions to meet the viscosity specifications for residual fuel oil, recycled to the coker, or coked to ash-free coke in a fluid coker.

PROCESS EFFICIENCY

Thermal Efficiency:

Products only, including char, %: 67

Overall, including net utilities, %: 56

Utilities, per ton dry coal liquefied:

Steam, lb:

	Generated	Consumed
600 psig	1095	1095
250	76	76
52	94	94
10	5	5
	1270	1270

Power, kWh:	54.18
Fuel, MMBtu:	3.776
Cooling Water, gal.:	20.78
Hydrogen Consumption:	
wt% MAF coal liquefied	2
SCF/Bbl net oil	2732

UNIQUE FEATURES OF PROCESS

Main features of the process are: Hydrogen donor solvent extraction of coal in a coking drum, cracking and coking to separate liquids from ash and unreacted coal, and production of the donor solvent in a fixed-bed catalyst hydrogenation reactor.

PROCESS STATUS

Development Status:

Laboratory scale experiments being conducted by ADL and PERC. Foster-Wheeler has performed experiments in a 20-40 lb batch extractive coker.

ADL Patents:

Patent applied for based on original concept of A. M. Schutte.

Location of Development Work:

Foster-Wheeler Laboratories, Livingston, New Jersey.

History:

Experimental study to verify original concepts began under ERDA sponsorship about 1975.

Problem Areas:

None reported, experimental studies are at an early stage. Extractive coking process uses proven petroleum refining equipment and hardware.

REFERENCES

40. Reber, S.A. et al, *The Arthur D. Little Extractive Coking Coal Liquefaction Process -- Economics and Potential for Chemicals Production*, Arthur D. Little publication.

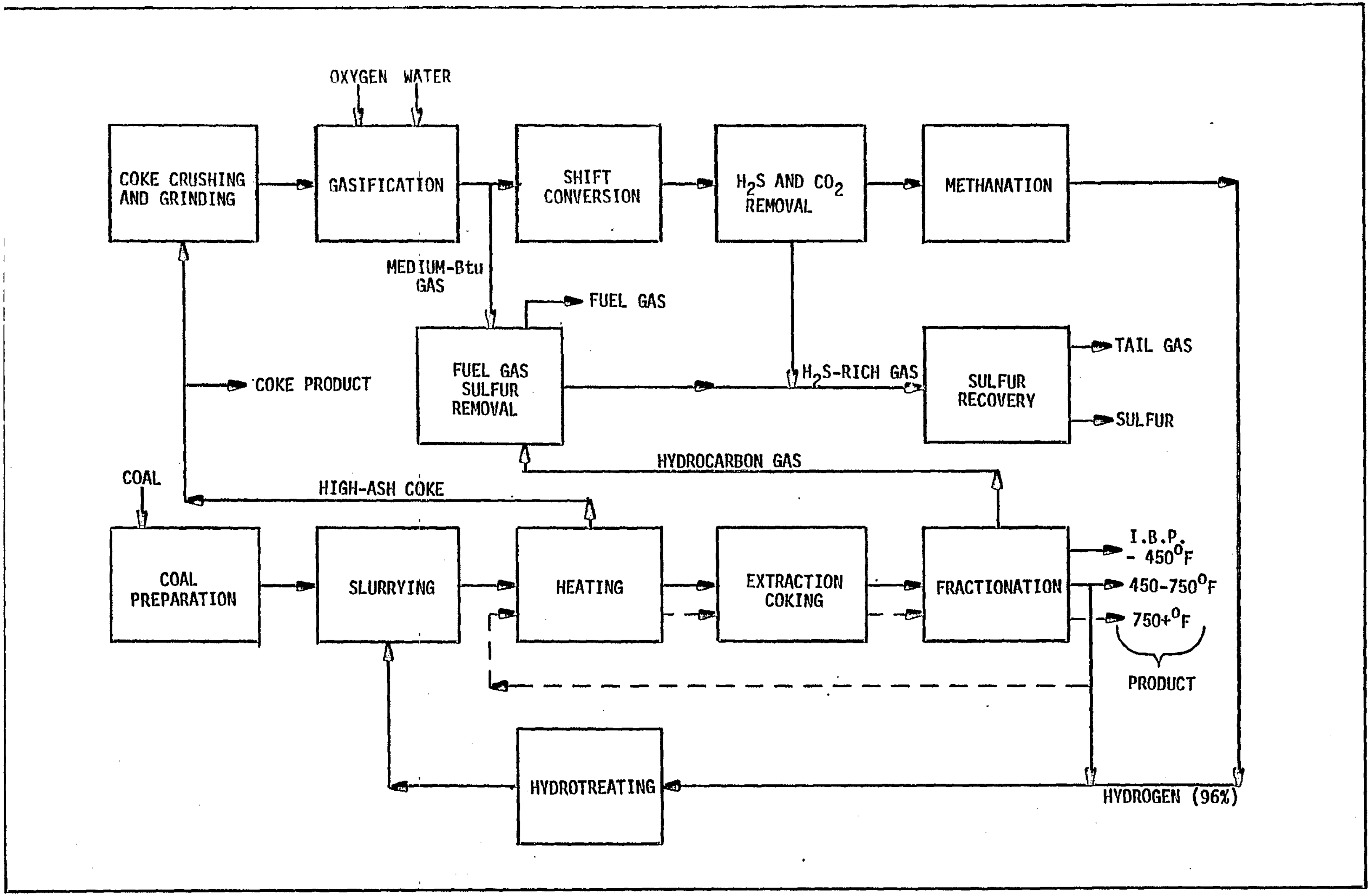

Figure 1 - Block Diagram
ADL Extractive
Coking Process (40)

PROCESS INFORMATION EXTRACT

PROCESS 1.3.3	Consol Synthetic Fuel (CSF)
Process Type:	Hydroliquefaction, Indirect, Donor Solvent
Main Products:	Gas, Naphtha, Fuel Oil, Char
Development Status:	Pilot Plant

PROCESS DEVELOPERS(42)

Pilot Plant Renovation: The following companies are under an ERDA contract to renovate and operate the CSF Cresap pilot plant as a multi-purpose test facility and to prove the plant can be operated as an integrated coal liquefaction process:

(1) Fluor Engineers & Constructors, Inc.

(2) Coal Conversion Development Corporation

(3) Allegheny Power Service

(4) American Electrical Power Service

PROCESS SPONSOR: DOE

PROCESS DESCRIPTION

The CSF process can be described as a combination of two major steps: Coal Extraction and Extract Hydrogenation.

Coal Extraction (refer to Figure 1):

Prepared coal is fed to a slurry mix tank and mixed with fresh donor solvent and recycled solvent. The slurry is pumped to the heater and enters the stirred solvent extractor reactor at 750°F and 465 psia. Vapors evolved in the extractor are taken overhead, condensed, and separated into sour gases and condensed liquids. A liquid level is maintained on the reactor and extract slurry is let down in pressure by a level control valve. The slurry is then fed to the primary feed tank, which is maintained at about 150 psia and 650°F by recycling slurry after heating to 700°F, where it is mixed with wash solvent from the Solvent Recovery Section, recovered solvent, and tar recycle from the Low Temperature Carbonization section. The solids are separated in a two-stage hydroclone system. The material is handled at an elevated temperature (650°F) to avoid precipitation of high molecular weight asphaltenes. Extract liquids removed from the primary hydroclone are sent to the Solvent Recovery where, under vacuum distillation (2.5 psia), gases, naphtha and middle distillates, solvent, and extract solution are separated. Solids from the Solids Separation section are fed from the underflow receiver as a slurry to a Low Temperature Carbonization unit. The extraction residue is pyrolyzed at 800°–900°F and low pressure to recover the solvent left in the slurry, additional distillate, and extract, and to reject a solid, high-ash char. Air is supplied to partially oxidize the char for the heat source.

Extract Hydrogenation (refer to Figure 2):

Extract from the Solvent Recovery section is hydrogenated in a multi-reactor-stage unit. The stages (two or more) operate at about 3000 psi and 800°F. The reactors are ebullating, catalytic, upflow reactors in which liquid is recirculated to suspend the catalyst in the reactor liquids. The ebullating reactor is a three-phase system: a gas phase, a liquid phase, and a fluidized catalyst plus liquid phase. Co-Mo-Ni on alumina base catalyst is used. High pressure gases are flashed off in the reactor itself and sent for gas separation and purification to recover recycle hydrogen, purge excess gas, and remove acid-gases. Liquid overflow from the reactor is sent to a Fractionator section to separate naphtha, heavy fuels, and donor solvent, which is recycled to the Extraction section to hydrogenate the coal.

OPERATING CONDITIONS(41),(44),(46)

	Extraction	Extract Hydrogenation
Temperature, °F	650 - 750	800
Pressure, psia	150 - 500	3000
H_2 Consumption, wt% MAF Coal Feed	0.3	3.8 (6.0% extraction feed)
Space Velocity	$\left(\frac{0.4\ \text{ft}^3\ \text{Reactor}}{\text{ft}^3/\text{h dry coal feed}}\right)$	N.A.

	Extract	Extract Hydrogenation
Residence Time	30-40 minutes	N.A.
Solvent to Coal (wt% Ratio):		
Recycle	1.12	N.A.
Donor	0.38	N.A.
Total	1.5	N.A.
Catalyst	-	Co-Ni-Mo

Reaction Variables:

EXTRACTION - Increasing temperature and residence time in the extractor has been shown to increase the wt% MAF coal conversion.(46)

PRODUCT(41)

Basis: West Virginia Coal

(1) Analyses of coal as feed to extraction:

Ultimate Analysis, dry wt%

C	69.0
H	4.9
N	1.3
S	4.3
O	7.3
Ash	13.2
	100.0

Moisture with feed to Extraction (wet wt%)	0
MF Gross Heating Value, Btu/lb	12,640
Size Range	3/8 inch x 0

(2) Analysis of extract and residue as feed to extract hydrogenation;

C	84.5
H	6.2
N	1.5
S	1.9
O	4.5
Ash	1.4
	100.0%

Yields:

(1) Extraction	wt% of MF Coal
Solids	
MAF Residue Solid	26.0
Ash in Solids	13.2
	39.2
Liquids	
Light Oil	0.7
Extract	53.6
	54.3
Gases	
H_2O	3.1
H_2S	1.4
CO	0.1
CO_2	0.8
H_2	-
C_1	0.4
C_2	0.3
C_3	0.2
C_4	0.5
	6.8
Total	100.3 (0.3% H_2 transferred)

(2) Extract Hydrogenation:

	wt% of MF Coal to Extraction	wt% of Extract to Hydrogenation from Solvent Recovery
Gases		
H_2S	1.0	1.8
NH_3	0.6	1.2
H_2O	2.6	4.8
C_1	2.0	3.7
C_2	1.6	2.9
C_3	1.7	3.2
Gas Subtotal	9.5	17.6
Liquids		
C_4 – 400°F	7.5	13.8
400° – 750°F	28.9	53.2
750°F+ (MAF)	10.9	20.0
Ash in liquids	0.7	1.4
Liquids Subtotal	48.0	88.4
Total	57.5%	106.0% (6.0% H_2 Consumed)

(3) Total Integrated CSF Coal Conversion Plant (Refer to Figure 4):

	wt% dry coal to extraction
Fuel Gas	9.0
Naphtha	8.0
Fuel Oil	30.9
Ammonia	0.6
Sulfur	4.2
Ash	12.5
Subtotal	65.2

Characteristics:

(1) Extraction	Solids Extract Residue (wt%)	Liquids Light Oil (wt%)	Liquids Extract (wt%)	Gases (wt%)
C	53.7	90.8	85.8	19.5
H	2.5	8.2	6.4	10.4
N	1.3	0.1	1.5	-
S	5.0	0.8	1.8	20.1
O	3.8	0.1	4.5	50.0
Ash	33.7	-	-	-
	100.0	100.0	100.0	100.0

Gases	vol%
H_2O	59.8
H_2S	14.8
CO	1.0
CO_2	6.4
H_2	1.9
C_1	8.4
C_2	3.8
C_3	1.2
C_4	2.7
	100.0%

(2) Extract Hydrogenation	Liquids C_4-400°F (wt%)	Liquids 400°-750°F (wt%)	Liquids 750°F+ (wt%)	Gases (wt%)
C	85.2	88.2	84.6	43.8
H	14.6	11.1	6.4	16.6
N	0.1	0.4	1.1	5.5
S	0.1	0.1	0.8	9.9
O	-	0.2	0.7	24.2
Ash	-	-	6.4	-
	100.0	100.0	100.0	100.0

Gases	vol%
H_2S	6.9
NH_3	8.8
H_2O	33.7
C_1	29.2
C_2	12.1
C_3	9.3
	100.0

(3) Total Integrated CSF Coal Conversion Plant (Refer to Figure 4):

Gas	vol%
H_2	29.3
N_2	3.5
CO	7.3
C_1	39.6
C_2	12.0
C_3	8.3
	100.0
Gross heating value (Btu/SCF)	933
Naphtha	
°API	58.0
Sulfur Content (wt%)	0.056
Gross Heating Value (Btu/lb)	19,566
Fuel Oil	
°API	10.3
Sulfur Content (wt%)	0.128

Fuel Oil

Gross Heating Value (Btu/lb)	17,969

INTENDED PRODUCT USE/MARKET

(1) Fuel gas: May be sold as a High-Btu, pipeline quality gas.

(2) Naphtha: May be sold as a gasoline-pool feedstock.

(3) Fuel oil: May be sold as a low-sulfur fuel oil or crude oil substitute.

(4) Char from Low-Temperature Carbonization: As an intermediate product, the intended use is to gasify the char to a syngas for hydrogen generation.

PROCESS EFFICIENCY(41)

Yields:

(1) Extraction (excluding utilities):

	HHV of Products as a % of Coal Feed HHV
Gases (C_4 and lighter)	3.3
Liquids	
Light oil	1.1
Extract	68.6
Solids	28.7
	101.7

(2) Extract Hydrogenation (Excluding Utilities):

	HHV of Products as a % of Coal Feed HHV
Gases (C_3 and lighter)	15.6
Liquids	
C_4–400°F	18.7

	HHV of Products as a % of Extract Feed HHV
400–750°F	66.1
750°F (MAF)	20.5
	120.9

Thermal Efficiency:

Total Integrated CSF Coal Conversion Plant:

Overall Thermal Efficiency (including utilities):

	% of Coal Feed HHV
Gas	14.7
Naphtha (C_4-400°F)	12.4
Product Fuel Oil	43.9
Sulfur	1.3
Ammonia	0.5
	72.8%

Utilities: see Table 1

Hydrogen Consumption:

	% of MF Coal Feed
Extraction	0.3
Extract Hydrogenation	3.3
Total	3.6%

Catalyst Consumption:

Extraction Hydrogenation: 1,860 lb extract feed per lb Co-Mo-Ni catalyst on alumina base.

UNIQUE FEATURES OF PROCESS

- The coal extraction reaction in a stirred-tank reactor
- Minerals separation by hydroclones
- Ebullating catalytic hydrogenation reactor containing three phases (gas, liquid, and suspended catalyst in liquid)

PROCESS STATUS(42), (43), (44)

Development Status:

A 20 TPD-pilot was operated in the late 1960s for 33 months. The pilot plant was shut down in 1970 after having defined major mechanical problems. It is expected to restart in 1977. A block flow diagram of the pilot plant is shown on Figure 3.

The CSF process was part of Project Gasoline which was terminated in the early 1970s. Also involved in the project was development of a zinc halide catalyst for hydrocracking extract to a higher-value gasoline product. Work in this area is continuing under a contract to Conoco Coal Development Company and Shell Development Company.(42)

Several patents involving the CSF process have been issued. The latest patent(43) was issued in 1969 jointly to Consolidation Coal Company and the U.S. Secretary of the Interior.

Location of Development Work:

CSF Process - Cresap, West Virginia

Zinc Halide Catalyst - Library, Pennsylvania

History:

Consolidation Coal Company, a subsidiary of Continental Oil Company, has been involved in coal extraction development work at least since the early 1960s. A patent on the early version process was filed in 1960 and granted in 1962.(44) In 1963, Consolidation Coal Company and the Office of Coal Research (OCR) entered into a contract for further development of the CSF process. A 20-TPD pilot plant was designed using data from extensive bench-scale testing by Consolidation Coal. In 1967, the plant started up including a 13-TPD extract hydrogenation section. The plant was shut down in 1970 due to severe mechanical problems. In 1974, Fluor Corporation was awarded an OCR project for the revamping of the Cresap pilot plant. Included in the contract is operation of the plant as an integrated CSF process and use of the plant as a multi-purpose test facility.

Problem Areas:(41), (45), (46)

Severe mechanical problems preceded a shutdown and re-evaluation. Among the more serious mechanical problems experienced at the pilot plant were:(45)

- filtration (later abandoned for hydroclones)
- hydrogen compressor
- high pressure reactor recycle pumps
- high pressure closures and piping joints
- hot oil pumps
- extraction reactor, agitator-shaft sealing
- The extract hydrogenation consumes more hydrogen than required for donor solvent extraction and uses a high amount of catalyst.
- Hydroclones separation of solids is not the most efficient method for removing solids from extract and may reduce catalyst life.

REFERENCES

41. Foster Wheeler Corporation, *Engineering Evaluation and Review of Consol Synthetic Fuel Process, R&D Report No. 70*, Office of Coal Research, U.S. Department of the Interior Contract No. 14-32-0001-1217, February 1972.

42. Energy Research and Development Administration, *Fossil Energy Program Report, 1975-76*, pp 26-28.

43. Gorin, E. et al, *Process for Making Liquid Fuels from Coal*, U.S. Patent No. 3,523,886, August 11, 1970.

44. Gorin, E., *Production of Hydrogen-Rich Liquid Fuels from Coal*, U.S. Patent No. 3,018,214, January 23, 1962.

45. The Ralph M. Parsons Company, *Final Report to the Office of Coal Research*, Department of the Interior Contract No. 14-01-0001-225, May 15, 1970.

46. Phinney, J.A., *Coal Liquefaction at the Cresap, West Virginia Pilot Plant*, Paper presented at Coal Conversion Pilot Plant Symposium. The American Institute of Chemical Engineers, Pittsburgh, Pennsylvania, June 2-5, 1974.

Table 1 - Utilities For Integrated Coal Conversion Plant(41)
Basis: 1 ton MF West Virginia Coal to Extraction

Major Units	Cooling Water (M gal)	Power (kWh)	Fuel Use (MMBtu)	Steam - (lb)			
				25 psi	150 psi	600 psi	1250 psi
Coal Preparation	-	14.6	0.802	-	-	-	-
Extraction	-	2.5	0.881	-	-	-	-
Solids Separation	0.23	1.5	-	-	-	-	-
Solvent Recovery	0.14	0.6	-	(454)*	(154)*	-	-
Low Temp. Carbonization	0.33	2.1	-	-	-	-	-
Tar Distillation (Vacuum)	-	0.3	0.216	-	-	-	-
Extract Hydrogenation	0.66	1.4	0.211	-	-	(346)*	-
Hydroletdown + Absorption	1.11	4.0	-	-	-	112	-
Hydrodistillation	0.01	1.8	0.180	-	-	-	-
Gas Recovery + Cleanup	1.15	1.2	0.113	200	22	79	-
H_2 Manufacture	6.38	27.7	-	120	31	600	476
H_2 Compression	1.14	-	-	-	2	256	-

*Numbers in ( ) indicate lb steam generated.

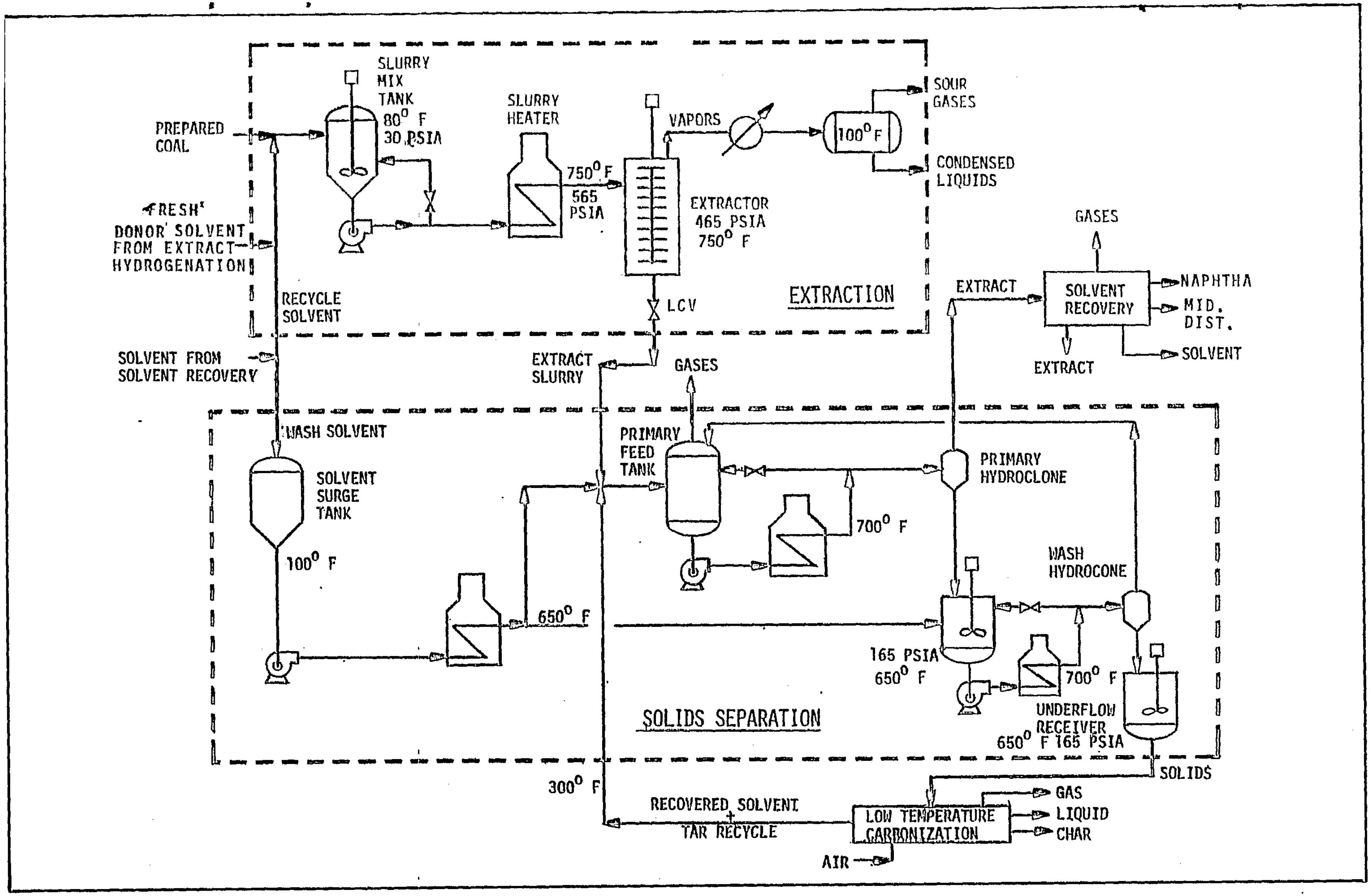

Figure 1 - Flow Diagram
CSF Process — Extraction
and Solids Separation (46)

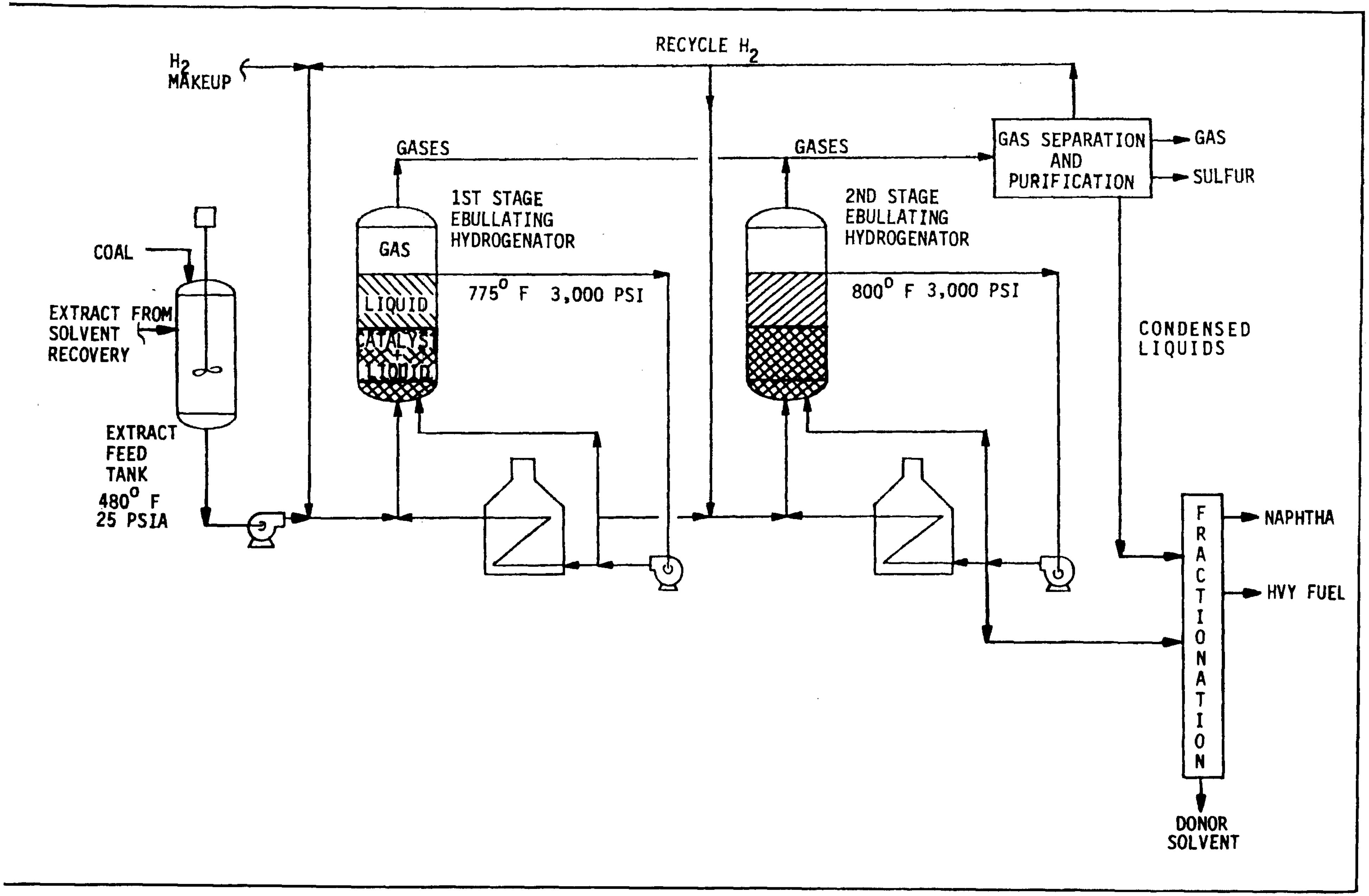

Figure 2 - Flow Diagram
CSF Process – Extract
Hydrogenation (46)

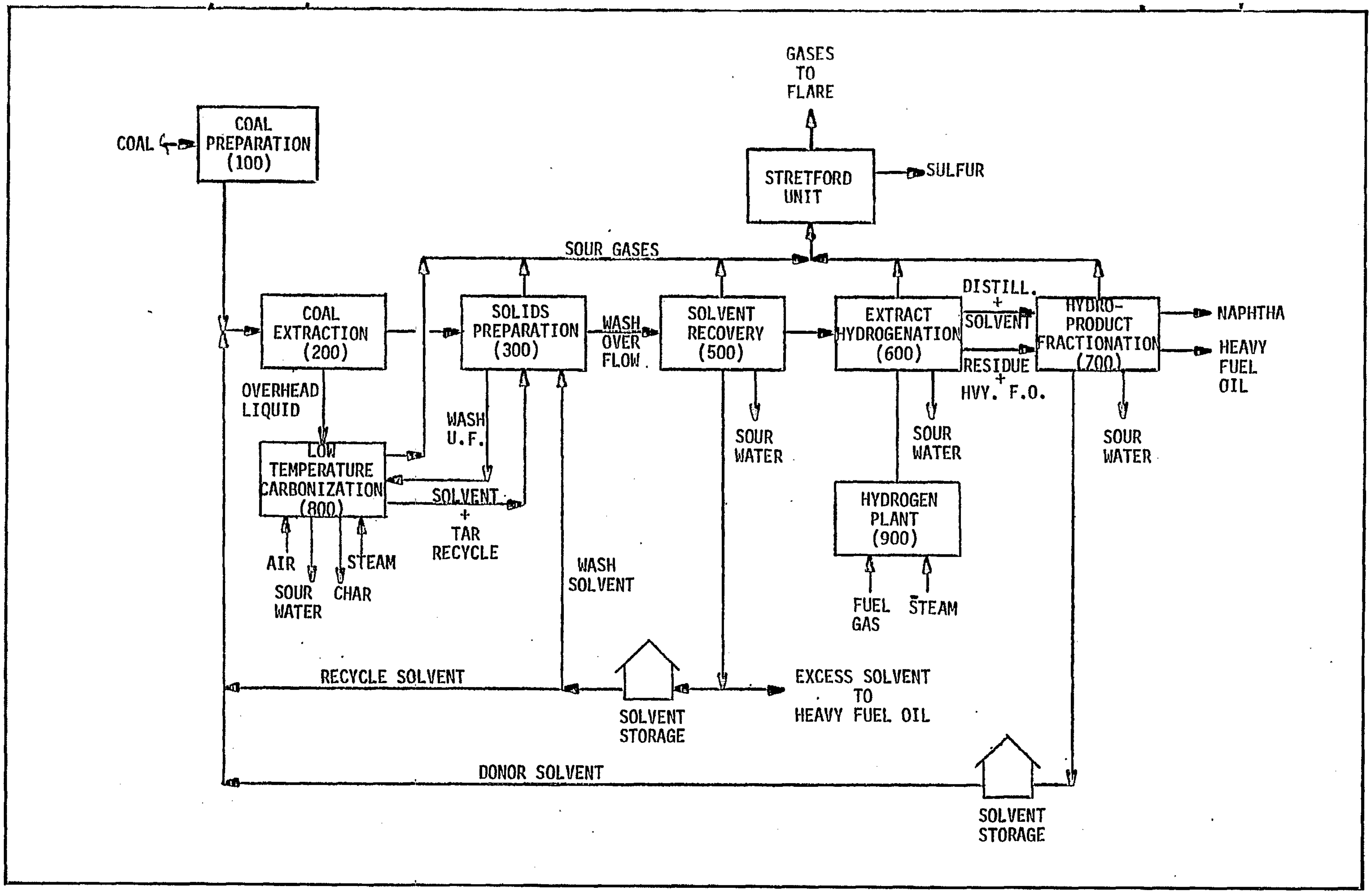

Figure 3 - Block Flow Diagram
20 TPD CSF Pilot Plant

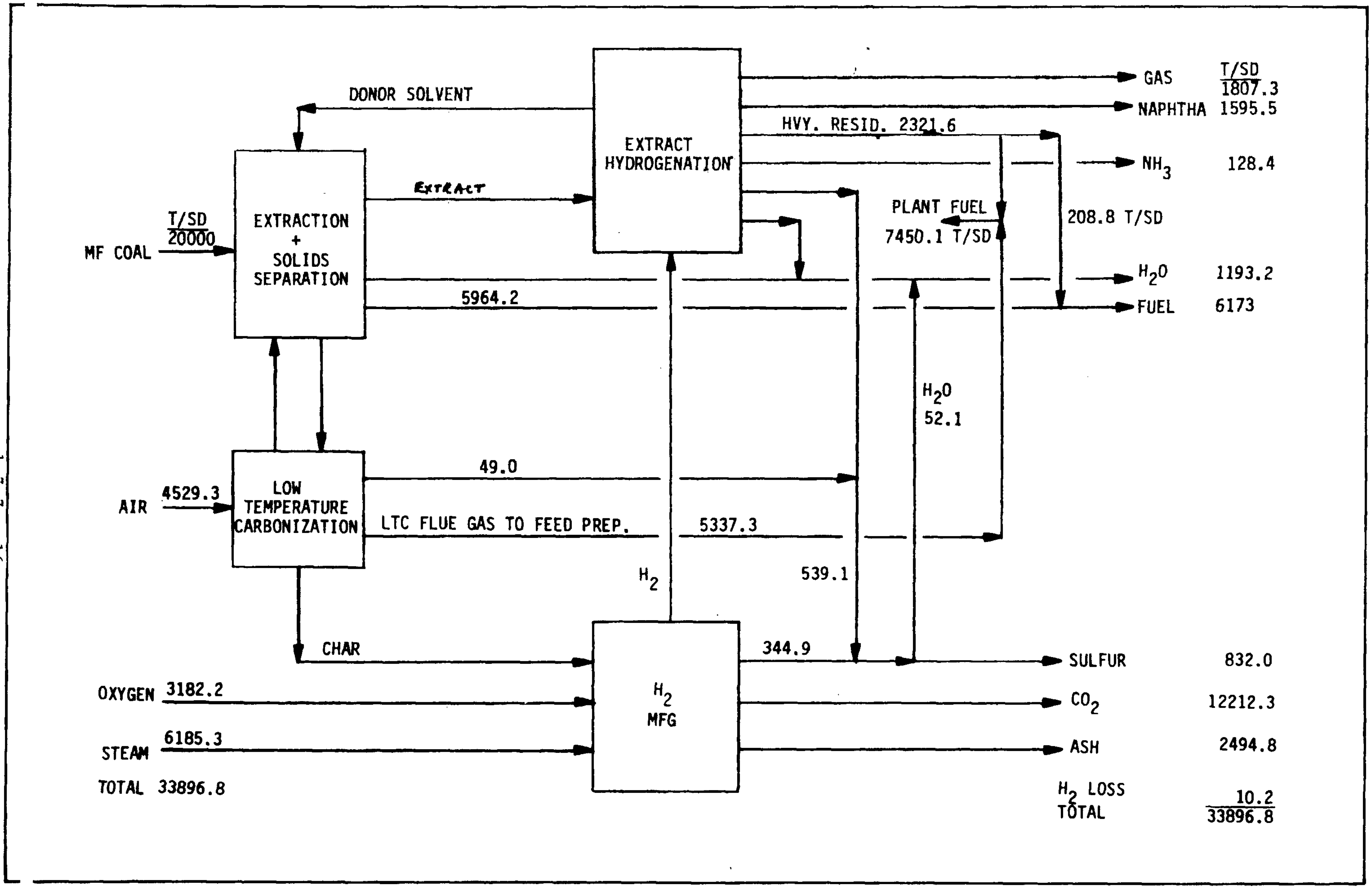

Figure 4 - Block Flow Diagram
Integrated CSF Coal
Conversion Plant (41)

PROCESS INFORMATION EXTRACT

PROCESS 2.1.1	Char-Oil-Energy-Development (COED) Ralph M. Parsons Conceptual Design
Process Type:	Pyrolysis + char gasification, Direct
Main Products:	High-Btu pyrolysis gas } used to generate Low-Btu combustion gas } power for sale Pyrolysis oil
Development Status:	Conceptual

PROCESS DEVELOPERS

(1) The Ralph M. Parsons Company,
Pasadena, California

(2) FMC Corporation
Chemical Research and Development Center
Princeton, New Jersey

PROCESS SPONSOR — Energy Research and Development Administration (ERDA)

PROCESS DESCRIPTION (Refer to Figure 1)(47)

Wet coal is fed to lift-pipe driers where it is contacted with a hot, low-Btu gas stream generated from char gasification. The driers also serve to size the dried coal by returning oversize particles to the crushers. Minus 6 to plus 325 mesh coal is fed to three fluidized-bed pyrolyzers and heated to 575°F, 815°F, and 1050°F in the first, second, and third stages, respectively. The heat for pyrolysis is supplied by: (1) recycle char from the gasifiers at 1600°F and (2) the circulating char heaters, which take a char stream from each pyrolyzer and contacts it with the hot, low-Btu gas from the gasifiers in a lift pipe. The char, after separation from the gas, is returned to the pyrolyzer. Heat for the process is generated by reacting char, in three fluidized-bed gasifiers, with steam, oxygen, and waste water. Each pyrolysis vessel is fluidized by the vapors evolved from the coal and a recycle pyrolysis gas. Each gasifier is fluidized by the vapors evolved from char gasification and the steam and oxygen feeds.

OPERATING CONDITIONS

	Coal Driers	Pyrolyzers			Char Burners (Average)
		1st Stage	2nd Stage	3rd Stage	
Temperature Range (°F)	200–400 (outlet)	550-850	815–860	925-1,060	1,500–1,650
Pressures (psia)	19.2 (outlet)	29.7	29.7	29.7	29.7
Average Solids Residence Time (min)	0.05	5	4	4	95
Average Fluidizing Gas Velocity (fps)	100	4	3	3	3
Average Freeboard Velocity (fps)	100	4	4	4	5

Reaction Variables: The primary reaction variable affecting liquid yields in the COED process is the volatile matter content of the coal feed. The conceptual design allows for a 44 to 49 wt % MAF variation in VM content of coal feed, reflected in the temperatures above.

Coal Feed Particle Size: 1/8 in. x 0

PRODUCTS(48)

Basis: Illinois No. 6 Coal Feed

Proximate Analysis, dry wt%

VM	40.7
FC	47.1
Ash	12.2
	100.0

Moisture with Feed, (wet wt%)	10.5

Ultimate Analysis, dry wt%

Carbon	69.0
Hydrogen	4.7
Nitrogen	1.3
Sulfur	3.8
Oxygen	9.0
Ash	12.2
	100.0

HHV, Btu/lb Coal Feed	11,139

Yields (pyrolysis only):

	wt% of dry Illinois No. 6 Coal Feed
Char (as net feed to char burners):	64.0
Pyrolysis oil:	21.4
High-Btu pyrolysis gas (C_4 + lighter):	9.1
H_2O:	5.5
	100.0

Characteristics:

(1) Net Char Composition to Char Burners:

Proximate Analysis, dry wt%

VM	4.7
FC	73.5
Ash	19.1

Ultimate Analysis, dry wt%

C	72.8
H	1.7
N	1.4
S	4.0
O	1.0
Ash	19.1
	100.0

(2) Pyrolysis Oil (raw, unfiltered, and untreated):

Solids, wt% dry basis 5

Ultimate Analysis	wt%
C	81.50
H	7.10
N	1.15
S	2.00
O	8.25
	100.00

°API gravity -4

ASTM Distillation (vol%)	(°F)
IBP	320
2	400
5	450
10	500
20	620
30	730
40	820
50	910
60	1000

(3) High-Btu Pyrolysis Gas	vol%
N_2	0.8
H_2	20.9
CO	6.4
CO_2	7.7
CH_4	49.2
C_2H_4	1.9
C_2H_6	4.7
C_3H_6	1.2
C_3H_8	0.8
C_4s	1.7
H_2S	4.7
	100.0

INTENDED PRODUCT USE/MARKET

(1) Char: the char from pyrolysis is nearly completely gasified to form a low-Btu fuel gas, which is used for hydrogen generation and power generation after gas treatment. The power may be sold (see Figure 2).

(2) Pyrolysis oil: after filtration and hydrogenation, the COED oil may be sold as a Syncrude for petroleum refinery feedstock.

(3) High-Btu Pyrolysis gas: after gas treatment, the pyrolysis gas is also used to generate power along with the low-Btu gas.

PROCESS EFFICIENCY

Yields (estimated from References 47 and 48):	HHV in Products as a % of Coal Feed
Char (as net feed to char burners)	60.0
Pyrolysis oil	26.0
High-Btu pyrolysis gas	11.0

Thermal efficiency:	Btu in Products as a % of Coal Feed-HHV
Pyrolysis process (excluding utilities):	97%
Parsons COED Coal Conversion Plant:	
export power	39.7%
export fuel gas	58.4%

Utilities:

Basis: 1 ton MF Illinois No. 6 coal

	Pyrolysis and Gasification	Oil-Vapor Recovery	Oil Filtration
• Steam (net used)			
600 psia/825°F, lb	87	304	32
165 psia/sat'd, lb	N.A.	6	1
Low pressure, lb	336	N.A.	2
• Cooling water, gal.	4,230	1,650	180
• Power, kW	2.9	1.4	0.59

Hydrogen Consumption: The pyrolysis step itself consumes no hydrogen. However, to hydroconvert raw pyrolysis oil to a suitable syncrude requires 3000 - 4000 SCF H_2 per bbl.

Catalyst Consumption: The pyrolysis step is noncatalytic.

UNIQUE FEATURES OF PROCESS

- The process completely segregates the high calorific vapors of pyrolysis from the low-Btu gases of char burning.
- Each pyrolyzer operates in parallel with respect to vapors volatilized but in series with respect to coal feed.
- The fluidized pyrolyzers and entrainment driers enable the process to handle agglomerating coals.
- The use of lift pipes for transferring most of the heat required for pyrolysis.
- The use of lift pipes for size classification in the pyrolysis/gasification unit.
- The full utilization of the char from the pyrolysis unit.

PROCESS STATUS

Development Status:

Parsons has completed a conceptual design and an economic analysis for a 25,000-TPD plant using a variation of the COED process. The pyrolysis yields were obtained from the FMC 36-TPD pilot plant using Illinois No. 6 coal.

Patent Status: None

History:

Parsons started the conceptual design in 1973 originally for the Office of Coal Research. The process design was published in 1975.

Problem Areas:

- Low process thermal efficiency
- High capital investment ($1 billion in 1974 $ for a 25,000-TPD coal conversion complex)

REFERENCES

47. The Ralph M. Parsons Company, *Oil and Power by COED-Based Coal Conversion, R&D Report No. 114, Interim Report No. 9*, Commercial Complex Conceptual Design/Economic Analysis, Energy Research and Development Administration Contract No. E(49-18)-1775, September 1975.

48. FMC Corporation, *COED Databook*, Compiled on Pilot Plant Operation Using Illinois No. 6 Seam Coal for Project COED, U.S. Government Contract No. 14-01-0001-1212.

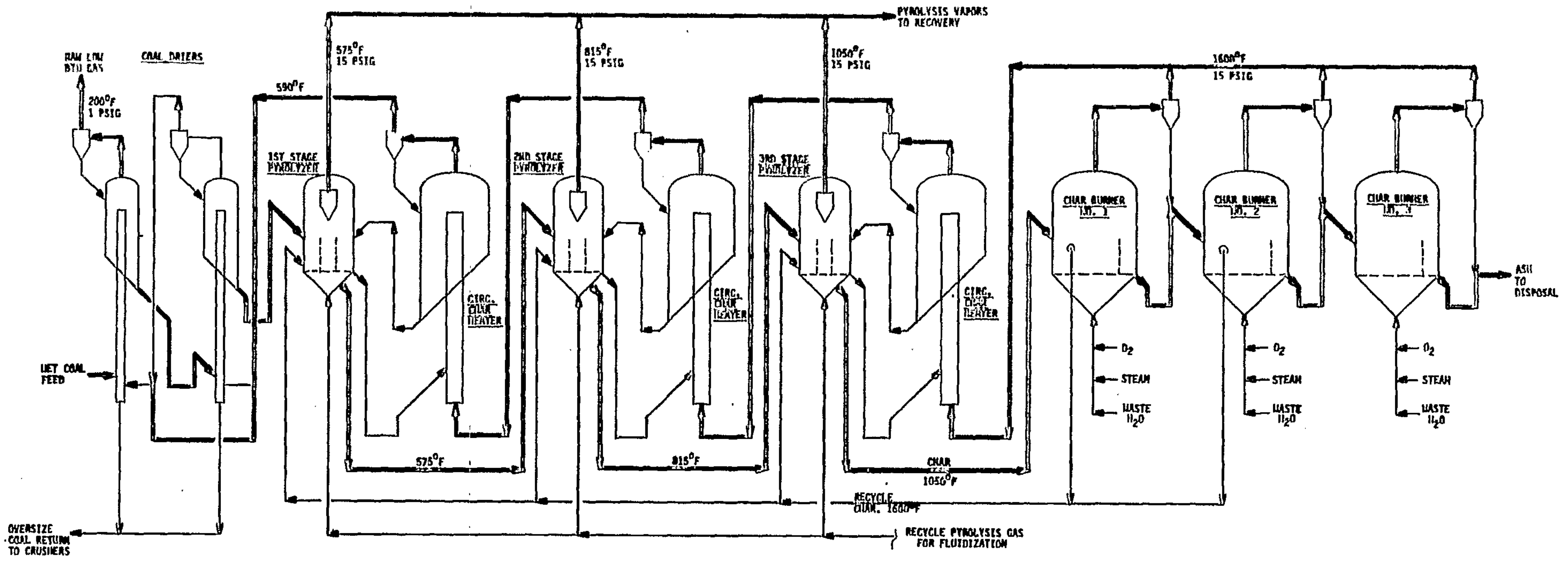

Figure 1 - Coed Pyrolysis and Char Gasification Process (RMP) (47)

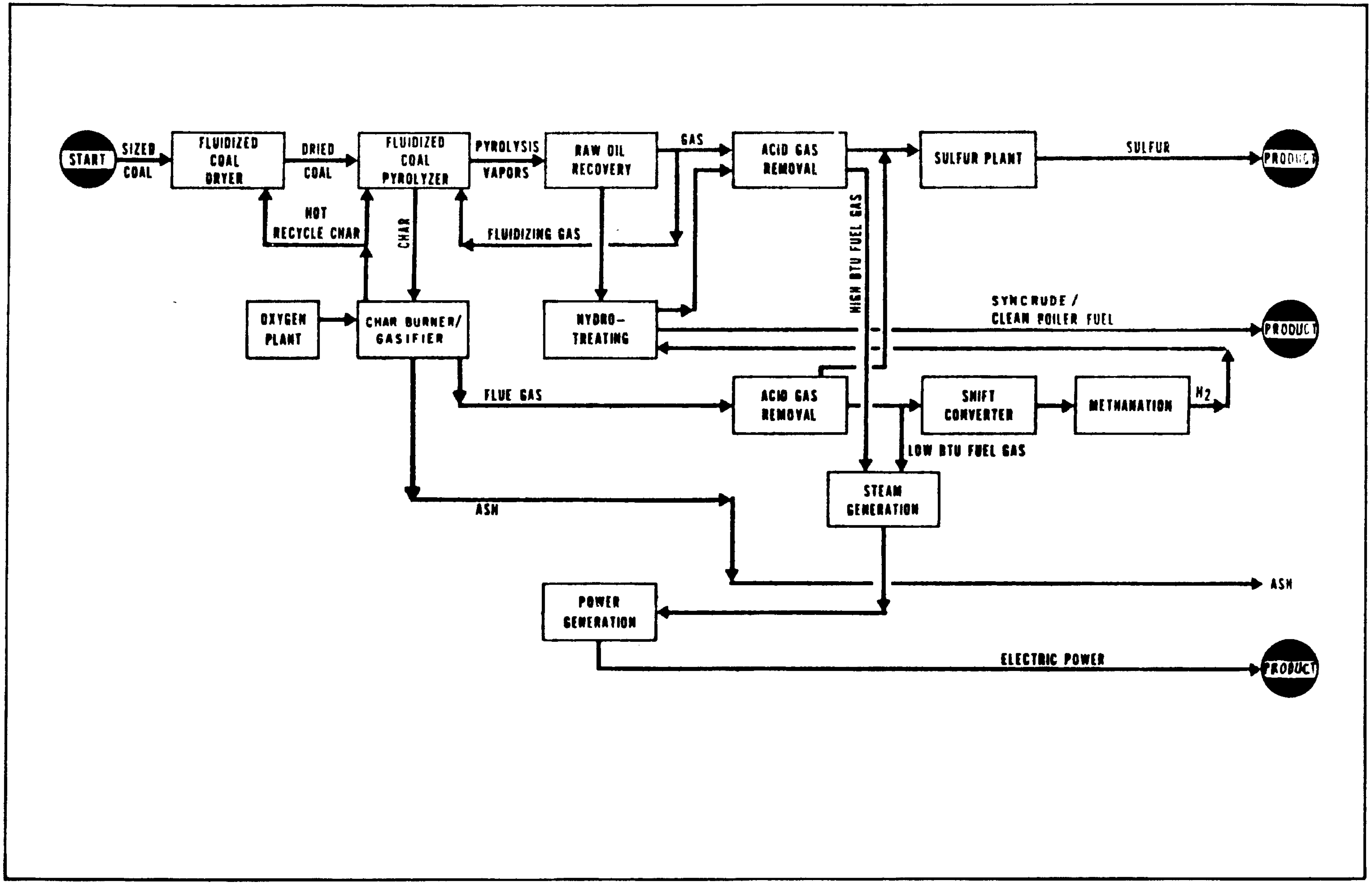

Figure 2 - COED Process
Clean Energy Plant Design
Combined Clean Oil/Electric Power Facility (47)

PROCESS INFORMATION EXTRACT

PROCESS 2.1.2	Char-Oil-Energy-Development (COED) FMC
Process Type:	Pyrolysis, Direct
Main Products:	High-Btu pyrolysis gas Pyrolysis oil Low-Btu combustion gases Char
Development Status:	Pilot plant - to 1974
PROCESS DEVELOPER	FMC Corporation
PROCESS SPONSOR	Office of Coal Research, U.S. Department of of Interior - to 1974

PROCESS DESCRIPTION (Refer to Fig. 1)(49)

Dried, crushed coal is heated in a series of fluidized-bed reactors. The first reactor is a pretreater where the coal is heated with recycle char and fluidizing gas to 300°F; the second and third stages are the true pyrolysis stages, where the coal is heated to 550°F and 1050°F, respectively. Heat for pyrolysis comes from partial combustion of the char in the fourth stage. Steam and oxygen reacts with the char to produce a low-Btu gas at 1600°F. The sensible heat of the char from the fourth stage is transferred to the second and third stages by recycling the char.

OPERATING CONDITIONS(55)

	Coal Dryer	1st Stage Pyrolyzer (pretreater)	2nd Stage Pyrolyzer	3rd Stage Pyrolyzer	4th Stage Char Combustion
Temperatures (°F)	300	550	820	1050	1600
Pressures (psia)	17	21	22	24	27
Residence Time (min)	12.3	10.0	7.0	3.0	16.0

	Coal Dryer	1st Stage Pyrolyzer (pretreater)	2nd Stage Pyrolyzer	3rd Stage Pyrolyzer	4th Stage Char Combustion
Fluidizing Velocity (fps)	1.9	2.4	1.5	1.0	3.0
Vapor-Space Velocity (fps)	2.5	3.1	2.2	2.5	6

Reaction Variables:

The primary reaction variables affecting liquid yields in the COED process are volatile matter content of the feed and the severity of pretreatment to prevent agglomeration in the beds, i.e., the temperatures and residence times used for coal drying and pretreating.

Coal Feed Particle Size minus 1/8 in.

PRODUCTS

Basis: Illinois No. 6 coal feed

Typical analyses:

Proximate analysis, dry wt%

VM	36.6%
FC	52.6
Ash	10.8
	100.0%

Moisture with feed, wet wt% 5.5%

Ultimate analysis, dry wt%

Carbon	71.2%
Hydrogen	4.8
Nitrogen	1.4
Sulfur	2.9
Oxygen	9.5
Ash	10.2
Chlorine	0.1

HHV, Btu/lb coal feed	12,210
Bulk density, lb/ft^3	46.5
Packed density, lb/ft^3	56.7
Particle density, lb/ft^3	62.4
Pore volume, cc/gm	0.19
Pore size, micron	8.6

Yields:

		wt% of dry Illinois No. 6 coal feed
(1)	char* (as net feed to 4th stage)	61.7
(2)	pyrolysis oil	21.2
(3)	High-Btu pyrolysis gas (C_4 + lighter)	13.7
(4)	H_2O	3.4
		100.0%

*Product char from 4th stage = 35.6 wt% of dry coal feed.

Characteristics:(48)

(1) Char - as product char from 4th stage:

Proximate analysis, dry wt%

VM	4.2 %
FC	78.6 %
Ash	17.2 %
	100.0 %

Ultimate analysis, dry wt%

C	76.9 %
H	0.6 %
N	1.0 %
S	2.0 %
O	2.3 %
Ash	17.2 %
Chlorine	-
	100.0 %

Gross Heating Value, Btu/Lb	11,030
Bulk density	28.7
Packed density	32.3
Particle density, lb/ft^3	69.3
Pore volume, cc/gm	0.29
Pore size, micron	5.1

(2) Raw pyrolysis oil (unfiltered, untreated) from Illinois No. 6 seal coal:

Moisture, wt%	1.2
Solids, wt% dry basis	

Ultimate analysis

C	80.5
H	6.8
N	1.2
S	2.1
O	8.0
Ash	1.4

°API gravity	-4
Pour Point, °F	100
HHV, Btu/Lb	14,920

ASTM Distillation

Recovery (vol%)	Temp. (°F)
IBP	320
3	435
5	450
10	500
15	550
20	620
30	730
40	820
50	905
55	955
60	1005
65	-
E.P.	1045

Total Recovery vol% = 64%

(3) High-Btu pyrolysis gas from Illinois No. 6 seam coal

	vol%		vol%
N_2	1.2	C_2H_6	9.8
H_2	17.2	C_3H_6	0.8
CO	6.8	C_3H_8	7.5
CO_2	5.9	C_4+	5.6
CH_4	36.8	NH_3	0.1
C_2H_4	0.2	H_2S	8.1
	68.1		31.9
			68.1
			100.0

INTENDED PRODUCT USE/MARKET(51),(53)

(1) Char

COED char may conceivably be used as a utility fuel provided stack gas clean-up is used for environmental restrictions. However, a low-Btu gasifier would be another use of the char to either produce low-Btu gas as a product and use the low-Btu gas by shifing and methanation to pipeline quality medium- or high-Btu gas.

(2) Raw pyrolysis oil

The raw pyrolysis oil may be sold as a syncrude after hydrogenation and filtration (see Figure 2 and Figure 3).

(3) High-Btu pyrolysis gas

The high-Btu gas may be purified and separated into SNG, LPG, and other light hydrocarbons (see Figure 2 and Figure 3).

PROCESS EFFICIENCY

Yields:

	Btu: Products
(1) Pyrolysis oil	25%
(2) High-Btu pyrolysis gas	12%
(3) Char* (as net feed to 4th stage)	-

*Product char from 4th stage = 30% of coal feed GHV.

Thermal efficiency - (HHV in products as a % of HHV in feed coal adjusted for coal equivalent of utilities):

COED pyrolysis process - 90%

Integrated plant - 57%

Utilities:

The following utilities, taken from reference (50) include coal pyrolysis, product recovery, and oil filtration.

Process steam (excluding steam for drivers) - 34.3 lb per ton coal to pyrolysis

Net Driver hp - 3.4 hp per ton coal to pyrolysis

Fuel - 0.87 MMBtu per ton coal to pyrolysis

Hydrogen consumption:

The COED pyrolysis process itself consumes no hydrogen. However, to hydroconvert raw pyrolysis oil to a suitable syncrude requires 3000 - 4000 SCF H_2 per bbl.

Catalyst consumption: Noncatalytic

UNIQUE FEATURES OF PROCESS

- The COED process as a staged coal conversion step attempts to separate the volatile matter from the fixed carbon and ash for the coal without destroying the chemical characteristics of the oil fraction. The aromatic structure of the gasoline cut is a potential feedstock to a gasoline-type refinery.
- The flow diagram (Figure 1) shown keeps all combustion gases separate from pyrolysis vapors. This is different from the original pilot plant flow scheme.
- The staged preheater, pyrolyzers, and char combustor enable the process to maintain fluidization in spite of the agglomerating tendencies of the feed coal.

PROCESS STATUS(52),(54),(56)

Development Status:

A 36-TPD pilot plant was operational from 1970-1974. The pilot plant included pyrolysis, partial gasification of char, filtration, and hydrotreating of pyrolysis oil (refer to Figure 4). Over 20,000 tons including seven different coals have been processed. In November 1973, 17,000 gallons of hydrotreated pyrolysis oil was successfully burned as fuel on the USS Johnston. Gasification tests for optimum char utilization are being made. The process was patented in March 1968 (U.S. Patent 3,375,175) and in April, 1971 (U.S. Patent 3,574,065).

Location of Development Work:

(1) Pyrolysis, oil filtration hydrogenation FMC R&D Center - Princeton, N.J.

(2) Char gasification (COGAS process) COGAS Process - British Coal Utilization Research Assoc., Ltd., Leatherhead, England

History:

FMC Corporation became interested in coal research in 1956 when they had a need for coke in an electric furnace plant in Idaho. The coke was formed by joining char and tar produced in fluidized-bed pyrolyzers.

In 1962 Project COED came under the sponsorship of the Office of Coal Research (OCR) to develop a process to convert coal into fuels. Several years of operation at the bench scale and later in a 100 lb/h process development unit (PDU) were followed by operation of a 36-TPD pilot plant in Princeton, New Jersey in 1970. Several commercial-size designs have been completed using the COED process.

Problem Areas:

- Low pressure pyrolysis in an integrated coal conversion plant leads to low thermal efficiency.
- Char utilization: selling of char is questionable; conversion to power leads to selling power at an uneconomical price if it is sold as a byproduct.
- Fines recovery and handling: the counter-current flow of gas and coal from stage-to-stage leads to a buildup of fines as experienced in the pilot plant.

REFERENCES

48. FMC Corporation, *COED Databook*, Compiled on Pilot Plant Operation Using Illinois No. 6 Seam Coal for Project COED, U.S. Government Contract No. 14-01-0001-1212.

49. FMC Corporation, *Project COED - Monthly Report No. 20*, U.S. Government Contract No. 14-32-0001-1212, April, 1973.

50. Shearer, H.S., "The COED Process Plus Char Gasification," *Chemical Engineering Progress*, Vol. 69, No. 3, March 1973, pp. 43-49.

51. Bonner and More Associates, *Economic Evaluation of Coal Based Synthetic Crude*, Energy Research and Development Administration Contract No. E(49-18)-1506, August 1973.

52. Sacks, M.E. and R.T. Eddinger (prepared by the editors of *Chemical Engineering Progress*), "Development of the COGAS Process," *Coal Processing Technology*, Vol. 2, 1975, pp. 94-97.

53. Terzian, H.D., et al, *Processing Coal to Produce a Synthetic Crude Oil and a Clean Fuel Gas*, Paper presented at Meeting, The American Institute of Chemical Engineers, Southern California Section, April 16, 1974.

54. Merrill, R.D., et al, (prepared by the editors of *Chemical Engineering Progress*), "Clean Fuels from Eastern Coals by COED," *Coal Processing Technology*, Vol. 2, 1975, pp. 88-93.

55. *COED Pilot Plant Design; Pyrolysis Heat and Material Balances*, Letter to J.B. O'Hara (The Ralph M. Parsons Company) from H.D. Terzian (FMC Corporation), April 19, 1973.

56. Eddinger, R.T., et al, *Pyrolysis of Coal*, U.S. Patent No. 3,375,175, March 1968.

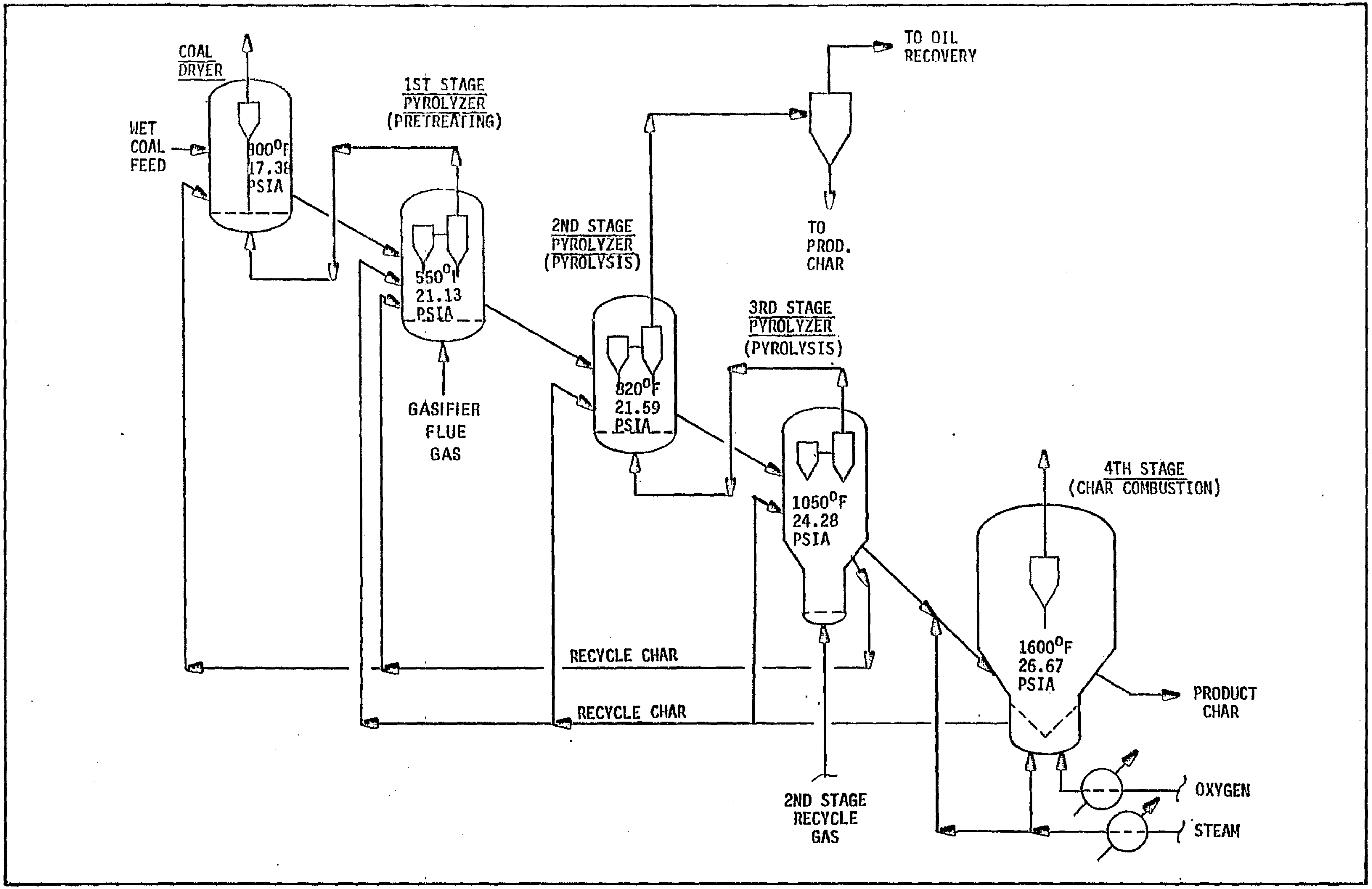

Figure 1 - FMC Pyrolysis Process (49)

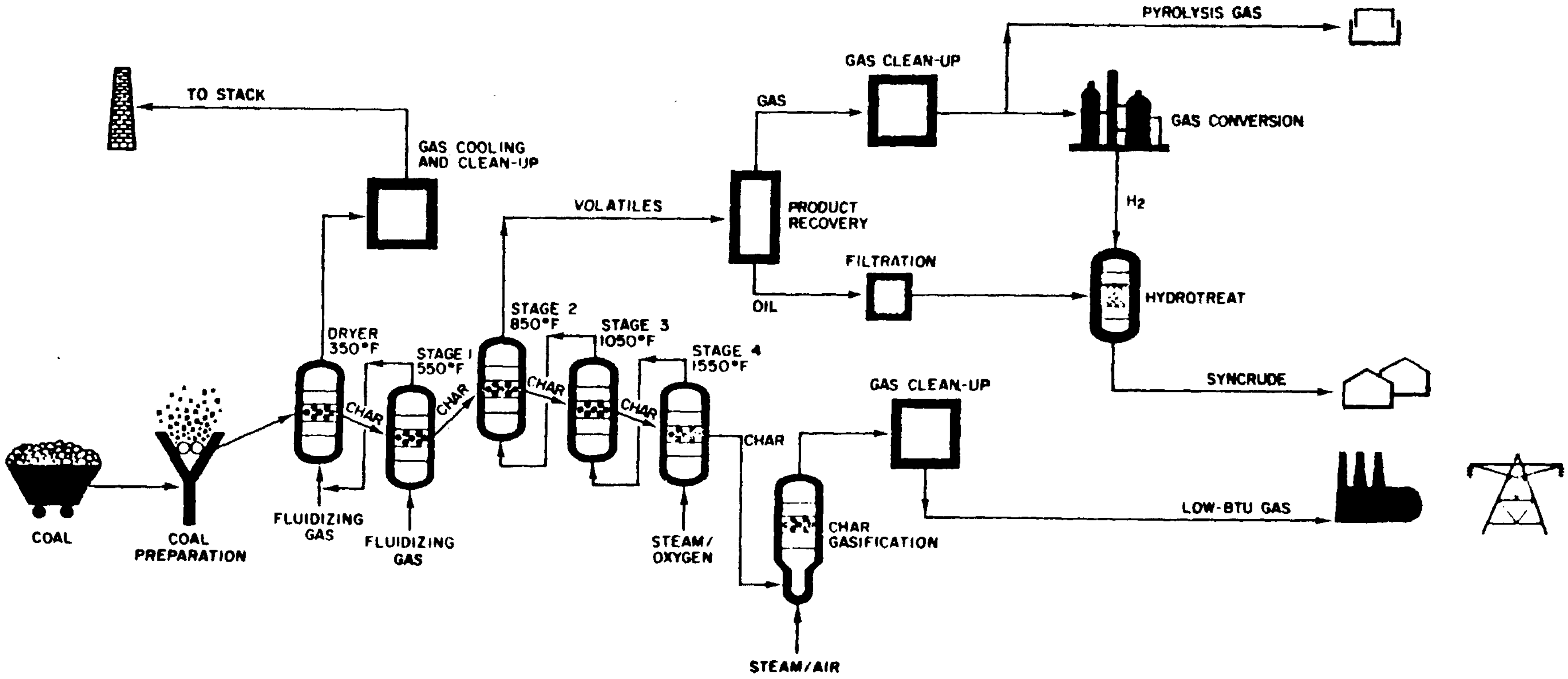

Figure 2 · COED Process
with Added Char Gasification
FMC – OCR (53)

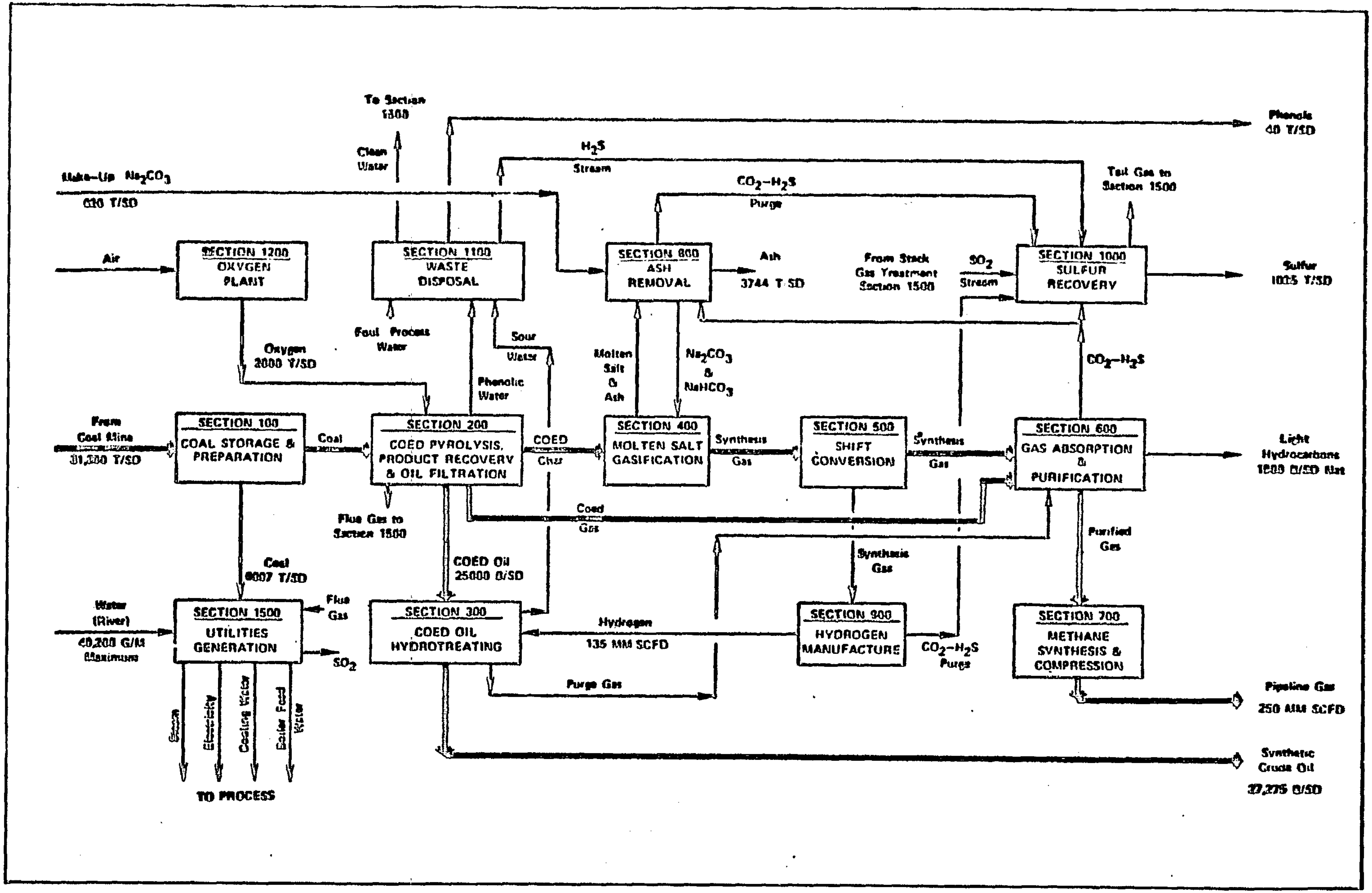

Figure 3 - The COED Char Gasification Process (50)

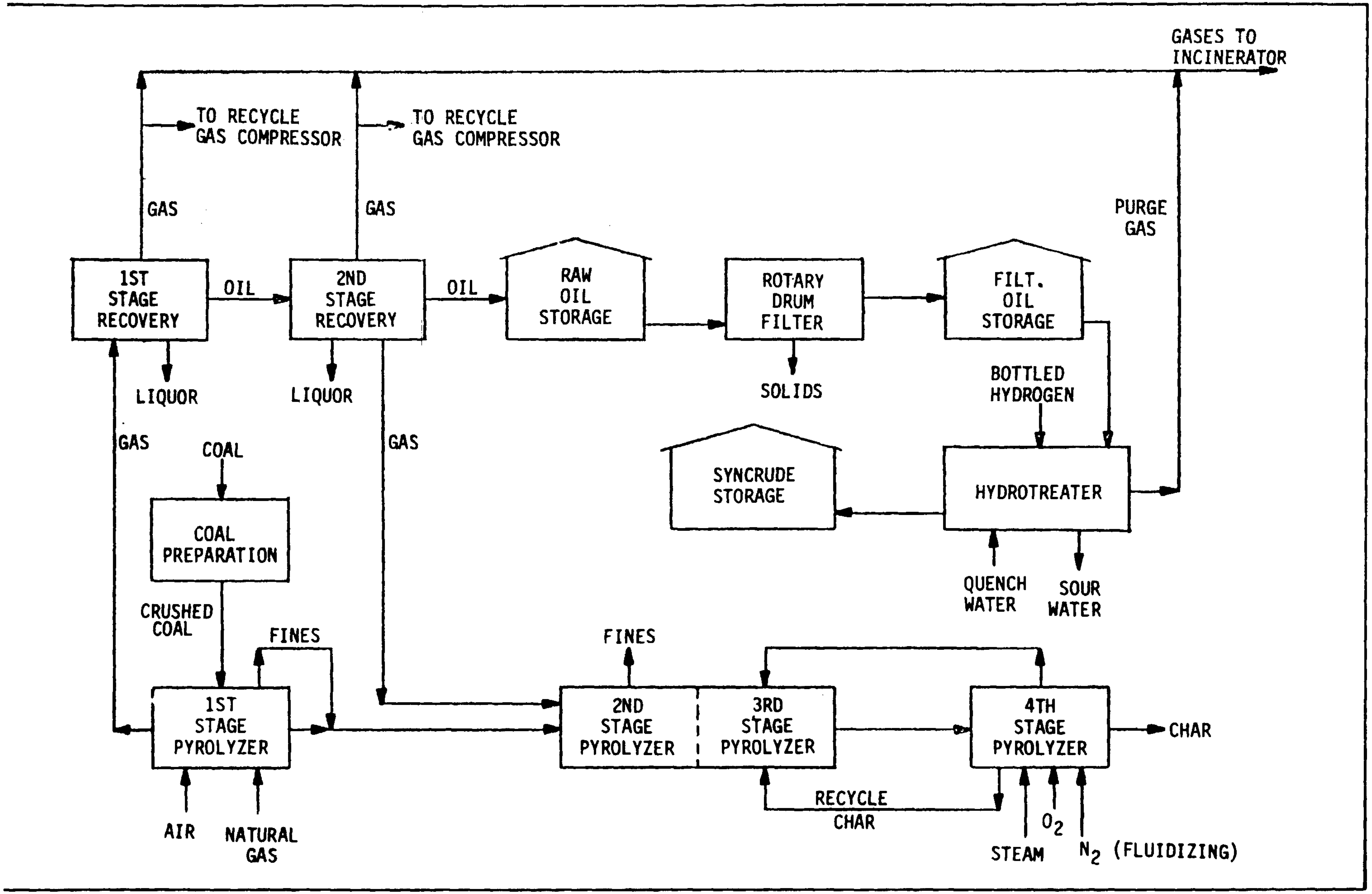

Figure 4 - Block Flow Diagram
36 TPD COED Pilot Plant

PROCESS INFORMATION EXTRACT

PROCESS 2.1.3	Occidental Coal Flash Pyrolysis
Name:	Pyrolysis, Direct
Main Products:	High-Btu Pyrolysis gas Pyrolysis oil Char
Development Status:	Bench-scale and PDU
PROCESS DEVELOPER	Occidental Research Corporation La Verne, California
PROCESS SPONSOR	DOE

DESCRIPTION OF PROCESS (Refer to Figure 1)

Dried, pulverized (-200 mesh) coal is contacted with hot recycle char in a flash pyrolyzer. The coal is devolatilized in less than three seconds and the pyrolysis vapors are separated from the solids (recycle and pyrolytic char) and then condensed by scrubbing with a cooled recycle oil. The pyrolytic char is partially gasified to heat up the recycle char from 1100°F to 1600°F in an entrained-bed char heater. Recycle char is separated from the low-Btu gas from the partial combustion of char and inventory of hot char is left in the hot char storage vessel. Recycle char is transported with an inert gas back to the flash pyrolyzer for pyrolysis heat.

OPERATING CONDITIONS

Temperatures	flash pyrolyzer - 900 - 1200°F char heater -1200 - 1800°F

Space Velocity	(~15 ft/sec)
Pressure	< 65 psia
Residence Time	< 3 s (flash pyrolyzer)
Vehicle	Recycle (or inert) fluidizing gas
Reaction Variables:	Reactor temperature is the primary reaction variable affecting liquid yields; max liquid yields have been observed around 1075°F
Feed coal particle size	minus 200 mesh

PRODUCTS

Bases:

	West Kentucky Coal	Wyoming Coal	
	Coal as Feed to 1 in. x 10 ft bench scale reactor (Ref. 58)	Coal as Feed to 1 in. x 10 ft bench scale reactor (Ref. 59)	Coal as Feed to 3.6-TPD PDU (Ref. 57)
	wt%	wt%	wt%
Proximate Analysis, (dry)			
VM	41.0	N.A.	N.A.
FC	50.5	N.A.	N.A.
Ash	8.5	N.A.	N.A.
Total	100.0		
Ultimate Analysis, (dry)			
C	73.5	68.84	68.6
H	5.0	4.28	4.7
O	8.7	15.20	18.8
N	1.5	0.97	1.1
S	2.8	0.83	0.6
Ash	8.5	9.88	6.2
Total	100.0	100.00	100.0

	West Kentucky Coal	Wyoming Coal	
	Coal as Feed to 1 in. x 10 ft bench scale reactor (Ref. 58) wt%	Coal as Feed to 1 in. x 10 ft bench scale reactor (Ref. 59) wt%	Coal as Feed to 3.6-TPD PDU (Ref. 57) wt%
Fischer Assay	500°C		
Tar + Light Oil	16.3	N.A.	10.2
Char	69.3	N.A.	68.8
Gas	5.0	N.A.	11.9
Water	9.4	N.A.	9.1
Total	100.0		100.0
HHV (Btu/lb coal as fed)	N.A.	9,200	9,400
Moisture with Feed (wet wt%)	N.A.	18.9	13.8

Yields:

	Wt.% of Moisture free West Ky. Coal (Ref. 58; 1100°F)	Wt.% of Moisture free Wyoming Coal (Ref. 59; 1600°F)	Wt.% of Moisture free Wyoming Coal (Ref. 57; 1100°F)
Pyrolytic char	56.7	56	62.4
Pyrolysis oil (tar)	35.0	12	15.3
High-Btu pyrolysis gas	6.6	25	11.1
H_2O	1.7	7	11.2
	100.0	100	100.0

Characteristics:

(1) Char

a. Pyrolytic char

Ultimate Analysis (dry)	Wt%	Wt%	Wt%
C	74.5	74.0	74.8
H	2.4	1.9	3.3

Ultimate Analysis (dry)	Wt%	Wt%	Wt%
O	4.6	3.9	10.6
N	1.4	1.0	1.4
S	2.6	0.6	0.5
Ash	14.5	18.6	9.4
	100.0	100.0	100.0
Higher Heating Value, (Btu/lb)	12,100	11,700	11,610

(b) Product char	Moisture free West Kentucky Coal (Ref. 58; 1100°F)	Moisture free Wyoming Coal (Ref. 59; 1600°F)	Moisture free Wyoming Coal (Ref. 57; 1100°F)
Ultimate Analysis (dry)	Wt%	Wt%	Wt%
C	N.A.	N.A.	82.1
H	N.A.	N.A.	1.5
O	N.A.	N.A.	2.2
N	N.A.	N.A.	1.0
S	N.A.	N.A.	0.5
Ash	N.A.	N.A.	12.7
			100.0
Higher Heating Value (Btu/lb)	N.A.	N.A.	12,000(approx)

(2) Pyrolysis oil

Elemental Analysis	Wt%	Wt%	Wt%
C	75-80	92.7	75–80
H	6-7	4.3	6–7.5
O	10-14	0.8	9–15

	Wt%	Wt%	Wt%
N	1–1.5	1.6	1–1.5
S	1.5–2.5	0.6	1.5–2.5
		100.0	
Benzene insolubles	5–20%	N.A.	N.A.
Asphaltenes	20–30%	N.A.	N.A.
Density (gm/cc)	1.1	1.1 (100°C)	1.1 (25°C)

3. High-Btu Pyrolysis Gas

	Moisture free West Kentucky Coal (Ref. 58; 1100°F)	Moisture free Wyoming Coal (Ref. 59; 1600°F)	Moisture free Wyoming Coal (Ref. 57; 1100°F)
N_2, H_2S free gas composition	Vol%	Vol%	Vol%
H_2	25.0	35.3	N.A.
CH_4	42.3	18.8	N.A.
C_2H_6	5.6	14.4 (C_2+)	N.A.
C_3H_8	4.0		N.A.
CO	4.9	22.4	N.A.
CO_2	10.6	9.1	N.A.
HHV (dry, H_2S free) Btu/SCF	700	646	N.A.

INTENDED PRODUCT USE/MARKET

(1) Char: Char is expected to be marketable as a fuel. The ignition characteristics are similar to the parent coal and a significant sulfur reduction from the parent coal was experienced. (Reference 59) For example, a Western coal, containing 0.7 lb sulfur per million Btu was reduced to a char containing 0.5 lb sulfur per million Btu.

(2) Pyrolysis Oil: The pyrolysis oil, after solids separation and hydrotreating, may be marketed as a synthetic crude oil or a low sulfur fuel oil.

(3) High-Btu Pyrolysis Gas: After gas processing and separation, this gas may be marketed as a pipeline gas.

PROCESS EFFICIENCY

Hydrogen Consumption:

The pyrolysis and char heating step does not consume hydrogen. However, to hydroconvert the raw pyrolysis oil will require a substantial amount of hydrogen.

Catalyst Consumption: Noncatalytic

UNIQUE FEATURES OF PROCESS

- The tar and gas yield is almost twice the Fischer assay yield. The high yield is attributed to the extremely short residence time thereby minimizing gas-phase cracking reactions. Yields are also higher than fluidized-bed pyrolysis processes such as COED.
- The entrained-bed reactor and char heater concept provides rapid devolatilization and char to coal heat transfer while still separating the high calorific value pyrolysis vapors from the low calorific combustion gases.
- The process can pyrolyze agglomerative coals.
- The process can produce a char with a lower sulfur content than the parent coal.

PROCESS STATUS

Development Status:

Evaluation and operation of the 3.6-TPD PDU is under the sponsorship of DOE. Several coals are expected to be run with monthly progress development reports available from DOE. A bench-scale pyrolysis reactor has been set up to produce tar for preliminary characterization and hydrotreating work. Two patents have been assigned to the process (References 60 & 61).

Location of Development Work:

Occidental Research Corporation
1855 Carrion Road
La Verne, California 91750

History:

Around 1969, the Garrett Research and Development Company initiated a coal research program to explore the feasibility of developing a coal conversion process to utilize the vast coal reserves of the Occidental Research Corporation. After literature review, a one-inch alloy tube, 10 feet long, was set up as an entrained flow pyrolysis reactor. It was observed that short residence times gave over 100% of Fischer assay yields. In 1971, a 4-TPD PDU was built in La Verne for further coal conversion study. However, the first two years of operation was devoted primarily to processing of municipal waste materials. (Based on PDU results, a 250-TPD municipal waste processing plant was built in San Diego County). In 1974, West Kentucky and Wyoming coals were charged. In 1976, Occidental Research Corp. (formerly Garrett R&D Co.) started up the PDU and bench-scale units under contract E(49-18)-2244 for DOE.

Problem Areas:

- Solids separation: the entrained flow reactor requires small particles (∿200 mesh) which will require extensive solids removal and separation equipment.
- Pyrolysis vapor quenching: an inherent disadvantage of rapid pyrolysis is rapid quenching; hence much of the high level heat of the pyrolysis vapors will be thrown away and contribute to the inefficiency of the process.
- Char utilization and/or marketing: the char, while lower in sulfur than the parent coal, will exceed sulfur emission standards for eastern coals. Marketability of chars from western coals is questionable.

REFERENCES

57. Sass, A., *The GR&D Pyrolysis Process - A Status Report*, Paper presented at 67th Annual Meeting, The American Institute of Chemical Engineers, Washington, D. C. December 3, 1974.

58. Sass, A., *The Garrett Research and Development Process for the Conversion of Coal into Liquid Fuels*, Paper presented at 65th Annual Meeting, The American Institute of Chemical Engineers, New York, New York, November 29, 1972.

59. McMath, H.G. et al, *Production of Gas from Western Sub-Bituminous Coals by the Garrett Flash Pyrolysis Process*, Paper presented at 66th Annual Meeting, The American Institute of Chemical Engineers, Philadelphia, Pennsylvania, November 1973.

60. Garrett, D.A., *Continuous Process for the Conversion of Carbonaceous Solids into Pipeline Gas*, U.S. Patent No. 3,698,882, October 17, 1972.

61. Sass, A. et al, *Process of Pyrolyzing and Desulfurizing Sulfur-Bearing Agglomerative Bituminous Coal*, U.S. Patent No. 3,736,233, May 29, 1973.

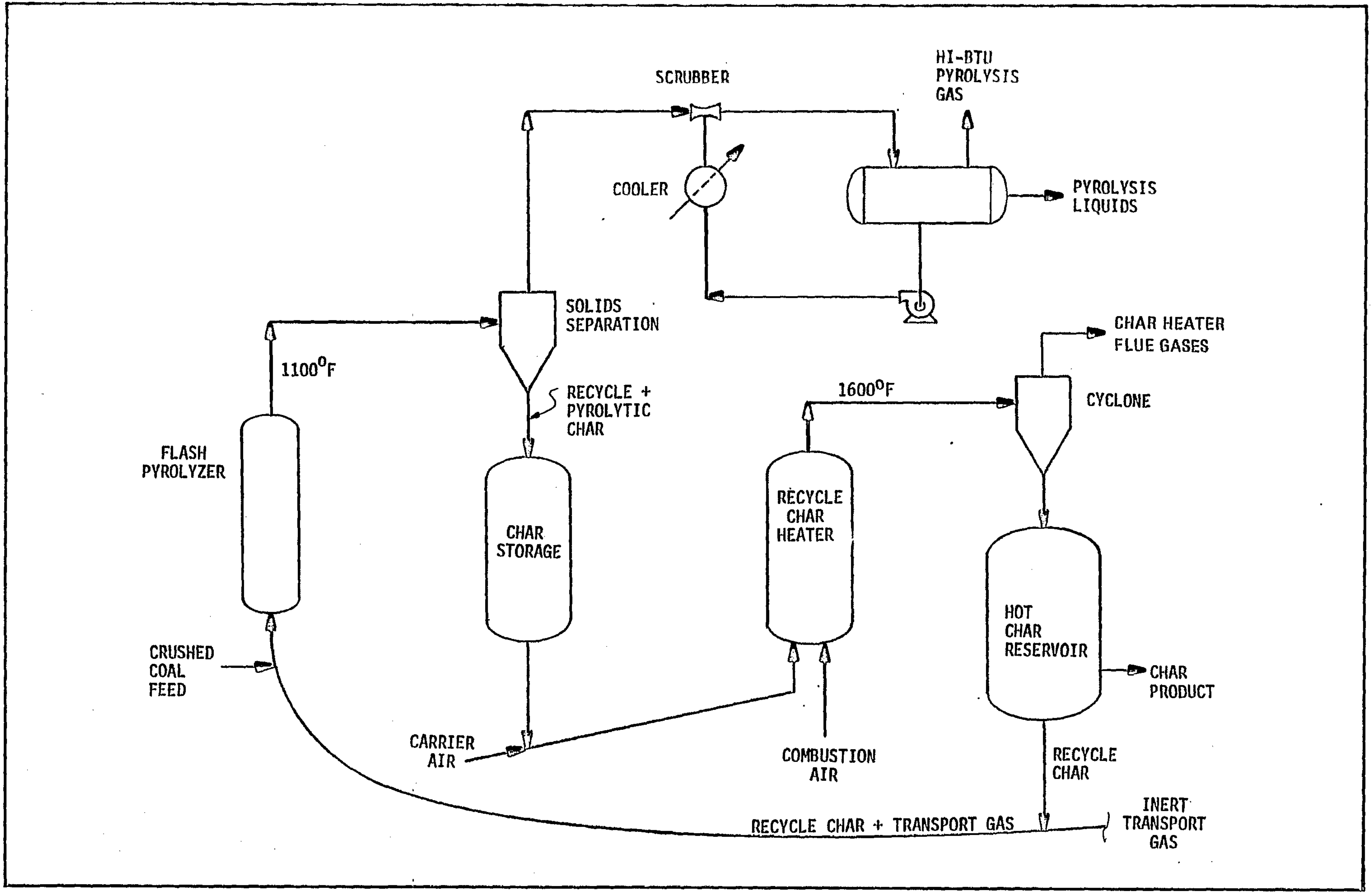

Figure 1 - Flow Diagram
Occidental Coal Flash
Pyrolysis Process

PROCESS INFORMATION EXTRACT

PROCESS 2.1.4	TOSCOAL
Process Type:	Pyrolysis, Direct
Main Products:	Char, pyrolysis oil, High-Btu pyrolysis gas
Development Status:	Pilot plant
PROCESS DEVELOPER	The Oil Shale Corporation
PROCESS SPONSOR	The Oil Shale Corporation

PROCESS DESCRIPTION (refer to Figure 1)

Shale (or coal) is heated and lifted by the hot flue gas from the ball heater in the lift pipe. The preheated feed is contacted with heated ceramic balls in a rotating drum retort. The ceramic balls are separated from the spent shale (char) in a trommel screen at the outlet of the retort. The spent shale (char) generates steam in the cooler. The ceramic balls are elevated and charged to the ball heater where they are heated to the required temperature by burning fuel gas. Pyrolytic vapors are cooled and the gas, tar, and water separated.

OPERATING CONDITIONS (63)

Temperature:	800 - 1000°F pyrolysis
Pressure:	atmospheric
Residence time:	5 - 10 min
Vehicle:	rotating drum
Reaction varibles:	reactor temperature, coal type (% VM)
Feed coal particle size:	minus 1/2 in.

PRODUCTS(62),(65)

Basis: Wyodak sub-bituminous coal:

Proximate Analysis, dry	wt%
VM	44.0
FC	48.5
Ash	7.5
	100.0

Moisture with feed, wet wt%:	30.0

Ultimate Analysis, dry	wt%
Carbon	66.1
Hydrogen	4.0
Nitrogen	1.0
Sulfur	0.4
Oxygen	21.0
Ash	7.5
	100.0

Gross Heating Value, Btu/lb of coal feed (HHV)	8,139
Net Heating Value, Btu/lb of coal feed (LHV)	7,570
Hardgrove grindability	56.0
Bulk density, -3/4 inch -	
poured, lb/ft^3	42.4
packed, lb/ft^3	47.7
$lb/SO_2/MM$ Btu	0.74
Free Swelling Index	0
Sulfur forms MF coal basis	
Pyrite	0.21
Sulfate	0.01
Organic	0.19
	0.41

Fischer Assay of Wyodak Coal (No. 3221 at 950°F):

	wt% of Dry Coal
Char	69.0%
Gas - C_3 and lighter	10.9%
(SCF/ton dry feed)	(2780)
Tar - C_4 and heavier	12.6%
(GPT dry feed)	(31.6)
Water	7.5%
	100.0%

Yields (wt% of dry coal feed):

	Retort Temperature		
	800°F	900°F	970°F
Char	74.9	72.3	69.2
Gas - C_3 and lighter	8.5	11.2	9.0
(SCF/ton dry feed)	(1786)	(2539)	(2321)
Tar - C_4 and lighter	8.1	10.2	13.3
(GPT dry feed)	(18.9)	24.9)	(31.0)
Water	8.5	6.3	8.5
	100.0%	100.0%	100.0%

Characteristics:

	Retort Temperature		
	800°F	900°F	970°F
(1) Char			
Proximate (wt%):	*		
Moisture	0.0	0.0	0.0
Ash	12.4	10.0	9.8
Volatile Matter	25.3	19.7	15.9
Fixed Carbon	62.3	70.3	74.3
Total	100.0	100.0	100.0

---

*Feed Wyodak coal was different from that used in runs C-2 and C-3.

Ultimate (wt%):			
Carbon	68.8	74.7	77.5
Hydrogen	3.4	3.0	2.9
Oxygen	13.3	11.8	8.3
Nitrogen	1.0	1.2	1.3
Sulfur	0.5	0.2	0.3
Chlorine	0.0	0.0	0.0
Moisture	0.0	0.0	0.0
Ash	12.4	10.0	9.8
Total	99.4	100.9	100.1
Other Data:			
Equilibrium Moisture (wt%)	10.0	10.8	9.9
Hardgrove Grindability	59.5	49.1	45.6
Heating Values			
Gross, Btu/lb	11,826	12,560	12,963
Net, Btu/lb	11,516	12,280	12,693
Bulk Density			
Packed, lb/ft^3	51.2	48.8	47.8
lb SO_2/MM Btu	0.85	0.32	0.46
(2) Pyrolysis Oil:	*		
Ultimate (wt%):			
Carbon	81.4	80.7	80.9
Hydrogen	9.3	9.1	8.7
Oxygen	8.3	9.4	9.3
Nitrogen	0.48	0.7	0.7

---

*Feed Wyodak coal was different from that used in runs C-2 and C-3.

Sulfur	0.43	0.2	0.2
Chlorine	0.0	0.0	0.0
Ash	0.0	0.2	0.1
Total	99.91	100.3	99.9
Heating Values			
Gross, Btu/lb	16,590	16,217	15,964
Net, Btu/lb	15,740	15,372	15,160
API Gravity			
Primary Tar	7.9	4.5	1.9
Calculated, with C_4 and heavier components of gas added	13.2	12.1	6.2
Pour Point (°F)	90	100	95
Conradson Carbon (wt%)	7.6	9.9	11.4
Distillation** (vol%)			
2.5	413°F	420°F	390°F
10	490	475	405
20	575	550	455
30	645	625	545
40	710	700	640
50	765	775	725
Viscosity (SSU)			
180°F	122	123	128
210°F	63	66	69

**Combination of TBP and D-1160 distillations.

(3) High-Btu Pyrolysis Gas

	Retort Temperature		
	800°F	900°F	970°F
	*		
Component (mole%)			
H_2	0.8	1.0	7.8
CO	18.0	17.3	18.4
CO_2	51.1	42.3	36.4
H_2S	1.7	1.3	0.3
C_1	16.9	22.0	24.9
C_2	3.6	4.7	4.4
C_2-	1.9	1.9	2.4
C_3	1.3	2.2	1.2
C_3-	1.6	3.7	1.6
iC_4	0.1	0.1	0.0
C_4	0.3	2.0	1.1
C_5	1.0	1.8	0.7
C_6	0.7	0.6	0.4
C_7	0.5	0.1	0.3
C_8+	0.2	0.0	0.1
Total	99.7	101.0	100.0
Average Molecular Weight:	35.9	35.0	30.6
Weight Percent Carbon:	40.5	45.9	44.7
Heating Values, Calculated			
Gross, Btu/SCF:	534	717	630
Net, Btu/SCF:	494	663	580

---

* Feed Wyodak Coal was different from that used in runs C-2 and C-3.

Calculated with CO_2 and H_2S removed			
Gross, Btu/SCF:	1,113	1,234	995
Net, Btu/SCF:	1,029	1,138	920

INTENDED PRODUCT USE/MARKET

(1) Char: The char is a suitable fuel in conventional coal-fired boilers according to evaluations by several boiler manufacturers after evaluating the grindability, ignition, and combustion characteristics. The char also represents a beneficiation from the parent coal in Btu/lb and moisture content, hence reducing the transportation cost on a $/MMBtu basis. Char could possibly be used, along with a binder, as form coke for blast furnaces. Char gasification to synthesis gas is another possibility.

(2) Pyrolysis Oil: The lowest boiling tar fractions contain a high concentration of commercially valuable cresols and cresylic acids. Although the sulfur level is low the oxygen content of the tar is very high reducing the heating value to approximately 16,000 Btu/lb. Hydrogenation would be required to increase the heating value to an acceptable fuel oil value (20,000 Btu/lb) for use as fuel or refinery feedstock. After thermal treatment, the oil could be used as electrode pitch and for the manufacture of coke.

(3) High-Btu Pyrolysis Gas: After CO_2 and H_2S removal, this gas is 900 - 1100 Btu/SCF and could be suitable for SNG marketing.

PROCESS EFFICIENCY(66)

Yields (retort at 970°F):	HHV in products
Utilities and fuels excluded	as a % of coal feed HHV
char:	75.9
pyrolysis oil:	17.9
High-Btu pyrolysis gas:	6.2
	100.0%

Thermal efficiency:	
pyrolysis process	95% (utilities excluded)
Utilities:	240 kWh/ton coal

Hydrogen Consumption: None

Catalyst Consumption: None

UNIQUE FEATURES OF PROCESS

- The use of ceramic balls - the balls, which are heated by direct firing of a fuel gas, act to transfer heat and grind up the coal in the pyrolysis drum and trommel
- A rotating pyrolysis reactor
- The apparent beneficiation of the parent coal to the produced char (heteroatoms, moisture, Btu/lb).

PROCESS STATUS

Development Status:

The TOSCOAL process is essentially the TOSCO II process as applied to coal, rather than oil shale. The Oil Shale Corp. (TOSCO) has retorted sub-bituminous, Wyoming coals in their 25-TPD oil-shale pilot plant at Rocky Flats, Colorado. A 1,000-TPD oil-shale, semi-works plant has operated at Parachute Creek, Colorado. Several 50,000-BPD oil-from-shale plants using TOSCO II have been proposed.(64)

Recently, TOSCO has extended pilot plant test work to coals with caking properties, including Utah and Illinois No. 6 coal. Short runs were made after the Illinois coal was pretreated to prevent agglomeration.

Gasification of char is being tested; also, char utilization tests as boiler fuel have been made by three boiler manufacturers.

The process and various equipment used in the process has several patents applied to it for the retorting of shale.

Location of Development Work:

The Oil Shale Corp., Rocky Flats Research Center, Golden, Colorado.

History:

TOSCO, in cooperation with other companies, had developed the TOSCO II process for the retorting of oil shale. The process has been demonstrated at the 1000-TPD semi-works plant at Parachute Creek, Colorado. At the present time, there have been several commercial size plants proposed to produce approximately 50,000-BPD of shale oil.

The TOSCOAL process is an adaptation of the TOSCO II process with similar flow sheets. The data for TOSCOAL was taken at the 25-TPD pilot plant built for TOSCO II development.

PROBLEM AREAS

- Ceramic ball attrition
- Process efficiency can be expected to be lowered due to low pressure operation and ceramic ball system
- Char utilization if high sulfur coals are used
- Market penetration if tars sold as phenolics

REFERENCES

62. Carlson, F.B. et al, *The Toscoal Process for Low Temperature Pyrolysis of Coal*, Paper presented at 65th Annual Meeting, The American Institute of Chemical Engineers, New York, New York, November 26-30, 1972.

63. *Synthetic Fuels Data Handbook*, Compiled by Dr. Thomas A. Hendrickson, Cameron Engineers, Denver, Colorado, 1975, Fig. 122, p. 228.

64. "Status of Synfuels Projects," *Synthetic Fuels*, Compiled by Cameron Engineers, Denver, Colorado, Vol. 13, No. 1, March 1976, pp. B-1, B-2.

65. Carlson, F.B. et al, "The TOSCOAL Process for Low Temperature Coal Pyrolysis," *Chemical Engineering Progress*, March 1973, p. 50.

66. Atwood, M.T. and B.L. Schulman, *The TOSCOAL Process-Pyrolysis of Western Coals and Lignites for Char and Oil Production*, presented to the ACS Division of Fuel Chemistry, New Orleans, Louisiana, March 1977.

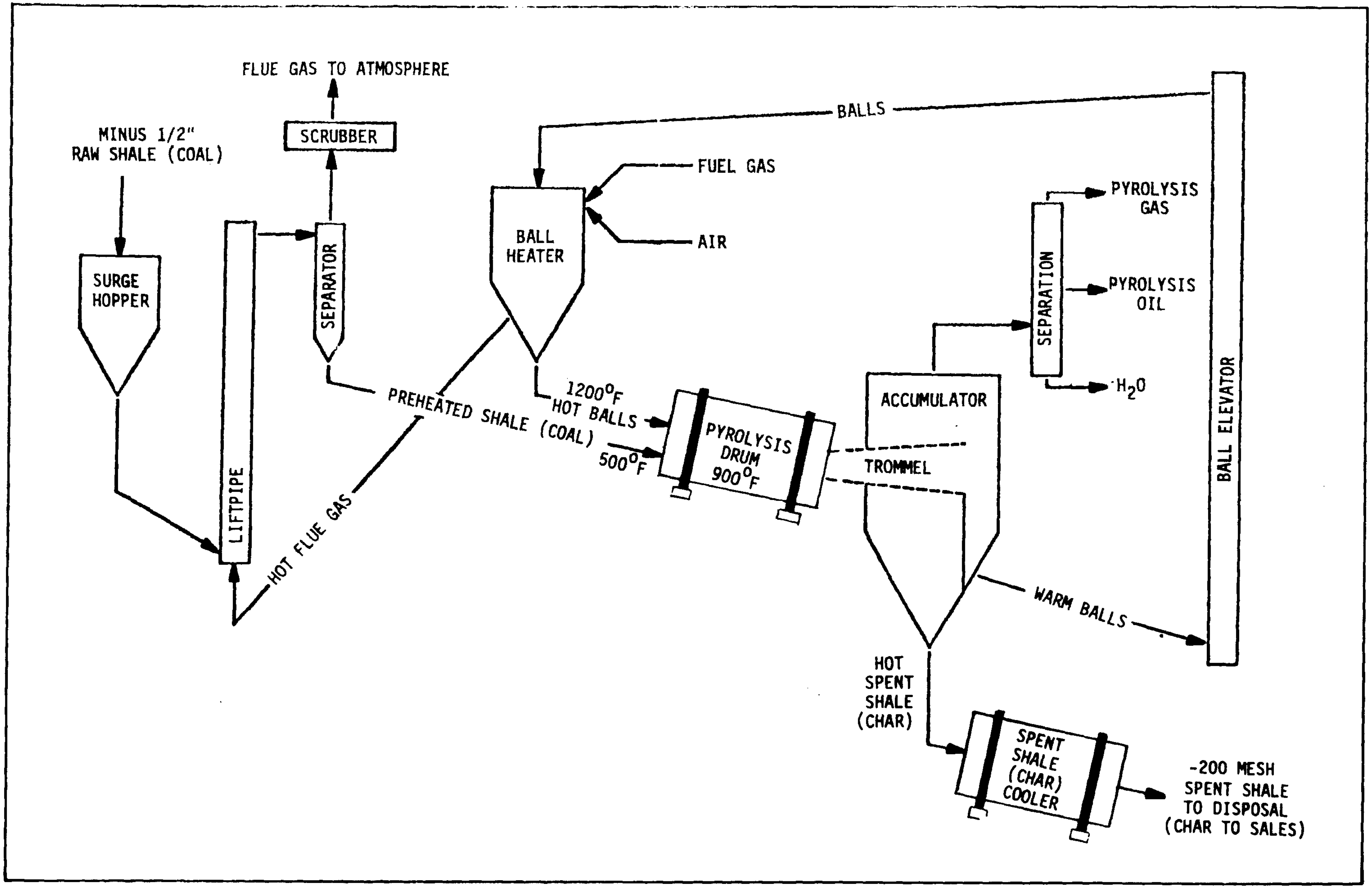

Figure 1 - Flow Diagram
TOSCO II (Toscoal) Process (63)

PROCESS INFORMATION EXTRACT

PROCESS 2.1.5	Lurgi-Ruhrgas
Process Type:	Pyrolysis, Direct
Main Products:	Char or semi-coke High-Btu Pyrolysis gas Pyrolysis Oil
Development Status:	Commercial

PROCESS DEVELOPER

Lurgi GmbH, a subsidiary of
Metallgesellschaft AG and Ruhrgas AG
6000 Frankfurt, Main
West Germany

American contact:
American Lurgi Corporation
377 Route 17
Hasbrouck Heights, N.J. 07604

PROCESS SPONSOR

Lurgi GmbH and Ruhrgas AG

DESCRIPTION OF PROCESS (Refer to Figure 1)

Crushed coal (0 to 5 mm range) is continuously fed to a mechanical mixer where it is uniformly mixed with hot recycle char. Temperature equalization and subsequent devolatilization is very rapid due to uniform mixing and high heat transfer rates of the recycle char and coal. Pyrolysis vapors are removed at the end of the mixer and subjected to dust collection and separation in the Hot Cyclone and Condensing systems. The char, including recycle and newly formed char, is allowed to further equilibrate in the carbonizer which also serves as a surge hopper. The char flows to the bottom of the lift pipe where it is contacted with combustion gases which heat and entrain the char. The combustion gases result from firing part of the pyrolytic tar or high-Btu gas or the char itself with preheated combustion air. The hot char is separated from the combustion gases in the collecting bin and cyclone and recycled to the mixer. The sensible and low-calorific value heat of the combustion gases are utilized in waste heat-recovery and coal drying.

OPERATING CONDITIONS

Temperatures	Mixer 800-1100°F
Pressure	Atmospheric
Residence time	In the order of a few seconds
Vehicle	Mechanical Mixer (pyrolysis) Combustion Gases (lift pipe char heater)
Reaction variables	Reactor temperature; maximum liquid yields at 1100°F
Feed coal particle size	In 0.1 - 3 mm range

PRODUCTS

Based on Feed Coal:	Sub-bituminous Coal (Reference 67)	Brown Coal Used in Yugoslavia Plant (Reference 68)
Proximate Analysis, dry		
VM	(45.4 MAF basis)	46.5
FC	N.A.	44.8
Ash	N.A.	8.7
	-	100.0
Tar Content (MAF wt%)	13.1	
Moisture with Feed, wet wt%	N.A.	7.9
Ultimate Analysis, dry	N.A.	
Carbon	N.A.	65.4
Hydrogen	N.A.	4.9
Nitrogen	N.A.	0.9
Sulfur	N.A.	1.0

	Brown Coal Used in Yugoslavia Plant (Reference 68)
Oxygen	19.1
Ash	8.7
	100.0
Bulk weight (lb/ft^3)	40.6
Fischer Assay	
Moisture	7.9
Water (decomposition product)	10.6
Tar	10.0
Coke	59.3
Gas + losses	12.2
	100.0

Yields

(1) Basis: Moisture free, sub-bituminous coal estimated in Reference 67.

	Wt% of Dry Coal Feed
Char	50%
Gas (MW=22)	11%
H_2O + Tar	39% (by difference)
	100%

NOTE

It was indicated that flash carbonization tests in continuous lab scale apparatus resulted in tar yields between 129% and 212% of Fischer assay (tar is all condensable hydrocarbons) using different coals between 19 and 45 MAF wt% VM. In the 12-TPD pilot plant, yields were 137 to 189% of Fischer assay (based on dust and water free tar from flash carbonization) using coals with 35-46% volatiles.

(2) Basis: Operating results reported from the semi-coke commercial plant in Yugoslavia. The yield basis is dry Brown Coal from the Kreka district, Reference 68.

	Wt% of dry, Brown Coal feed
Semi-Coke (char + heavy tar)	73
Middle Oil	1
Gas + Naphtha (used for heating char)	9
Gas Liquor	12
Miscellaneous	5
	100

NOTE

Operation of the plant was set to produce char with 15–20 wt% volatile matter; the char is then mixed with heavy tar to produce a semi-coke, which is used for coke-oven feedstock. However, the process is capable of devolatilizing the coal to less than 2% residual volatile matter content.

Characteristics:

(1) Pyrolysis Gas Analyses (vol%):

	Sub-Bituminous Coal	High Volatile Coal	Brown Coal
H_2S	1.7	3.3	N.A.
CO_2	11.9	6.7	48.0 (CO_2 + H_2S)
N_2	N.A.	1.1	0.7
CO	15.8	12.0	11.5
H_2	22.0	14.8	13.1
CH_4	31.5	44.1	23.0
C_2H_4	5.8	2.3	3.7 (C_2+)

	Sub-Bituminous Coal	High Volatile Coal	Brown Coal
C_2H_6	6.1	7.9	3.7 (C_2+)
C_3H_6	3.9	N.A.	3.7 (C_2+)
C_3H_8	1.3	7.8	3.7 (C_2+)
	100.0	100.0	100.0
Net calorific value, Btu/SCF	710	840	N.A.

(2) Liquids Analyses

Liquids from sub-bituminous coal:

	Heavy Tar	Middle Oil	Gas Naphtha
Wt% on Total	70.7	22.4	6.9
Ultimate Analysis:			
C	81.81	83.36	N.A.
H	5.98	9.76	N.A.
O	9.26	5.96	N.A.
N	1.01	0.33	N.A.
S	0.37	0.43	N.A.
Ash	1.57	0.16	N.A.
	100.00	100.00	
Density @ 20°C (kg/L)	1.259	0.95	0.738
Dust Content (wt%)	9.3	0.02	N.A.
Conradson Residue (wt%) (in dust free tar)	45.3	1.06	N.A.
Softening point (°F)	264	N.A.	N.A.
Pour Point (°F)	N.A.	-2	N.A.
Viscosity (°E/°C)	6.85/200	1.62/20	N.A.