

ASSESSMENT OF FOREIGN COAL CONVERSION TECHNOLOGIES

Volume III

Appendix B

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CHICAGO, ILLINOIS 60616

ASSESSMENT OF FOREIGN COAL CONVERSION TECHNOLOGIES

Volume III
Appendix B

by

Martin Novil
Christopher F. Blazek
Edward J. Daniels



STAT

July 1982

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OVERVIEW

Coal-synfuels commercialization forecasts have traditionally been based on the assumption that the technology for implementation does or will exist. The processes under development worldwide represent an extension of existing coal synfuels technologies in their potential for greater process efficiency, tolerance of a variety of coals, or production of a specific product range of greater interest to the anticipated commercial market. Because of technical, economic or marketing problems most of the approximately 40 processes under development outside the United States will not succeed beyond the demonstration phase. The economic and market related issues have been presented as constraints in Volume II of this report. In this appendix, the technical and development status of emerging coal synfuels technologies is presented. Tables OB1 and OB2 summarize the status of processes analyzed. Technical details are provided in the text of this volume.

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Table OBI. COAL GASIFICATION AND COMBINED CYCLE PROCESSES

<u>Country/Process</u>	<u>Largest Pilot Plant Size (metric tons of coal/day except as noted)</u>	<u>Largest Pilot Plant Location</u>	<u>Commercial Plans</u>	<u>Proposed Commercial Sites</u>	<u>Proposed Commercial Plant Size (metric tons of coal/day except as noted)</u>	<u>Official Status</u>
<u>West Germany</u>						
Shell Koppers	150	Hamburg, FRG	✓			Continued in-house research feasibility studies continue, no commercial plant announcements. Intended plans have been cancelled for the FRG.
High Temperature Winkler	24	Frecher, FRG	✓	Huerth-Berhenrath, FRG*	6700	No government support, commercial plant under construction by Rheinbraun.
Saarberg-Otto	264	Volklinger-Furstenhausen, FRG				Government support ending soon, no commercial plans announced.
RAG/RCH Texaco	144	Oberhausen/Holtzen	✓	Muscle Shoals, US* Cool Water Project, US* Tennessee Eastman, US*		Government interested in supporting commercial demonstration plant in the FRG.
Ruhr-100 High Pressure Lurgi	135	Dorsten, FRG				Government support ending soon, no commercial plans announced.
KGN Fixed Bed	2	Huchelhoven, FRG	✓	Huchelhoven, FRG	5500	State government of Nordrhein, Westfalia, is expected to fund project through demonstration phase.
VEW Partial Gasification (Combined Cycle)	360	Stockum, FRG	✓	Gersteinwerke, FRG	5500	Commercialization prospects are low with anticipated government support ending.
Bergbau-Forschung Nuclear Assisted	10.8	Essen, FRG				Research continues, no commercial plans announced.
Rheinbraun Nuclear Lignite Hydrogasification	25	Wesseling, FRG				Research continues, no commercial plans announced.
Kloekner Molten Iron	240	Ruhr region, FRG	✓		1900	Government interested in supporting commercial demonstration plant in the FRG.
Steag/Lurgi (Combined-Cycle)				Lunen, FRG*	1700	Commercial size facility shut down for economic reasons.

* Already under construction or completed.

Table OB1. COAL GASIFICATION AND COMBINED CYCLE PROCESSES, (Cont.)

<u>Country/Process</u>	<u>Largest Pilot Plant Size (metric tons of coal/day except as noted)</u>	<u>Largest Pilot Plant Location</u>	<u>Commercial Plans</u>	<u>Proposed Commercial Sites</u>	<u>Proposed Commercial Plant Size (metric tons/day except as noted)</u>	<u>Official Status</u>
<u>Great Britain</u>						
British Gas Slagging Lurgi	800	Westfield, Scotland	✓			British Gas is trying to find a sponsor for commercialization.
British Gas Composite						British Gas has dropped this project.
Eso Chemical Active	3.6	Abington, England	✓		20MW	Proposed plant does not appear to be ready for commercialization.
National Coal Board PFB (Pressurized Fluidized Bed Combustion of Coal)**	5MW	Leatherhead, England	✓	Grimethorpe, England	20MW*	Testing of the demonstration size unit will last through 1982.
<u>India</u>						
Central Fuel Research	24.0	Dhambad, India				Experimental research facility in operation since 1962.
Bharat Heavy Electrical	144.0	Trichy, India	✓	Tiruchirapalli, India*	5MW	Demonstration unit undergoing testing.
<u>Japan</u>						
Sumitomi Molten Iron	60	Kashima, Japan	✓			Future plans call for the construction of a demonstration plant in Japan and Australia.
Mitsui M. Gas		Kitakyushu, Japan				Still in early phases of pilot plant testing.
Coal Mining Research (Combined Cycle)	40	Yubari, Hokkaido, Japan	✓		1000	Pilot plant still in testing phase.
Hitachi Coal Oil Mixture	4	Iawki City, Japan				Future plans call for construction of a 40 MTPD pilot plant.

* Already under construction or completed.

** Noted here because of potential competition with combined cycle coal gasification for electric power generation.

Table OBl. COAL GASIFICATION AND COMBINED CYCLE PROCESSES, (Cont.)

<u>Country/Process</u>	<u>Largest Pilot Plant Size (metric tons of coal/day except as noted)</u>	<u>Largest Pilot Plant Location</u>	<u>Commercial Plans</u>	<u>Proposed Commercial Sites</u>	<u>Proposed Commercial Plant Size (metric tons of coal/day except as noted)</u>	<u>Official Status</u>
<u>Soviet</u>						
Power Engineering High Speed Pyrolysis (Fixed Bed)	100-200	Kaliningrad (FB)	✓	Kansk Achinsk, USSR	175 mt/hr (demonstration plant)	Demonstration plant at Karsnoyarsk still in the construction phase. Proposed commercial plant at Kansk Achinsk has capacity of 25 X 10 ⁶ metric tons/yr.

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Table OB2. COAL LIQUEFACTION PROCESSES

<u>Country/Process</u>	<u>Largest Pilot Plant Size (metric tons of coal/day) except as noted</u>	<u>Largest Pilot Plant Location</u>	<u>Commercial Plans</u>	<u>Proposed Commercial Sites</u>	<u>Proposed Commercial Plant Size (metric tons of coal/day) except as noted</u>	<u>Official Status</u>
<u>West Germany</u>						
Ruhrkohle/Veba Oil Catalytic Hydrogenation	200	Bottrop, W.G.	✓		11,250	Commercial development in doubt pending decision on FRG support.
Saarbergwerke Catalytic Hydrogenation	6	Völklinger-Furstenhausen, W.G.	✓		7000	Commercial development in doubt pending decision on FRG support at end of pilot plant tests in 1983.
Rheinbraun	0.3	Wesseling, SG	✓		6000	Commercial development in doubt pending decision on FRG support.
<u>Great Britain</u>						
National Coal Board Supercritical Gas Solvent Extraction	2.7	Stoke Orchard, England				Commercial development unlikely in the near term due to National Coal Board R&D cutbacks.
National Coal Board Liquid Solvent Extraction	0.7	Stoke Orchard, England				Commercial development unlikely in the near term due to National Coal Board R&D cutbacks.
<u>Australia</u>						
CSIRO High Speed Flash Pyrolysis	0.5	North Ryde, England				Research is still underway but no commercial plans have been announced.
<u>Poland</u>						
Central Mining Institute Catalytic Hydrogenation	0.12	Tychy-Wyry, Poland				Still in the early phases of research.
<u>South Africa</u>						
Sasol (Lurgi/Fischer Tropsch)			✓	Sasolburg, (Sasol II)* Secunda (Sasol II + III)*	11000 60000	Commercial operation since 1955. Full commercial operation of both facilities by end of 1982.

* Under construction or already completed.

Table OB2. COAL LIQUEFACTION PROCESSES, (Cont.)

<u>Country/Process</u>	<u>Largest Pilot Plant Size (metric tons of coal/day) except as noted</u>	<u>Largest Pilot Plant Location</u>	<u>Commercial Plans</u>	<u>Proposed Commercial Sites</u>	<u>Proposed Commercial Plant Size (metric tons/of coal/day except as noted)</u>	<u>Official Status</u>
South African Explosives & Chemicals Koppers Totzek			/	Modderfontein*	1000	Commercial operation since 1972 for ammonia production.
South African Explosives & Chemicals/Mobil				Modderfontein	2400	Status of this development in doubt. Methanol production intended.
Sasol Direct Liquefaction	Bench scale	Sasolburg				Still in the early experimental phase of development.
<u>Japan</u>						
Mitsui Solvent Refined Coal	4.5	Ohmuta City, Japan	/	Victoria, Australia	5500	Recent announcements have indicated that construction of the demonstration facility has been postponed indefinitely.
Nippon Brown Coal "Kominic" Direct Hydrogenation	0.55	Osaka, Japan	/	Victoria, Australia	5000	Construction of a 50 MTPD demonstration plant in Australia should be completed in 1983.
Mitsui Eng. & Ship Building Direct	2.4	Kawasaki, Japan	/		500	Pilot plant still in early research stage.
Sumitomo Solvent Extraction	1	Ibaraki Prefecture, Japan	/		250	Pilot plant still in early research stage.
Mitsubishi Heavy Ind. Solvolysis	1	Nagasaki, Japan				Pilot plant planned at 40 MTPD input.
<u>Soviet Union</u>						
Inst. of Mineral Fuels Catalytic Hydrogenation	0.1	Moscow, USSR				Pilot plant in construction at Belkovskaya mine (10 MTPD). Future plans call for pilot plant construction at Kansk-Achinsk (75 MTPD).

* Under construction or already completed.

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INTRODUCTION

The following sections survey 39 foreign coal conversion processes. These processes are numbered consecutively and organized by country of origin:

- Sections 1 through 14 are West German processes.
- Sections 15 through 20 are British processes.
- Section 21 is an Australian process.
- Section 22 is a Polish process.
- Sections 23 through 25 are South African processes.
- Sections 26 through 28 are Indian processes.
- Sections 29 through 37 are Japanese processes.
- Sections 38 and 39 are Soviet processes.

WEST GERMAN COAL CONVERSION PROCESS

1. Shell-Koppers Coal Gasification ProcessProcess Description

The coal gasification reactor in the Shell-Koppers process is essentially an empty pressure vessel equipped with diametrically opposed diffuser guns. Crushed, ground, and dried coal sized to 90% $90 \mu\text{m}$ is fed to the reactor through a proprietary coal loading system on a series of lockhoppers that allow for continuous operation. The process will handle a wide variety of solid fuels, including all types of coal and petroleum coke. Tests indicate that fuels with ash contents of as high as 40 wt % and sulfur contents of up to 8 wt % can be used without trouble. Because of the feed preparation requirements, the entire mine output, including fines, can be utilized. In addition, the Shell-Koppers process seems less affected than other coal gasifiers by coal characteristics such as ash fusion temperature or coal caking properties.

The prepared coal, which is fed into the reactor through the diffuser guns, reacts with the blast of steam and oxygen in a flame-like manner. Although the outlet gas temperature does not exceed 2730°F, flame temperatures within the reactor can reach 3630°F. Wall temperatures in the reactor are presently being maintained by an intricate cooling system. An insulating brick wall reactor is also being investigated as an alternative to the current approach. Raw gas production is approximately 70,620 SCF/ton of bituminous coal. The gas produced has a heating value of roughly 300 Btu/SCF and has a CO-to-H₂ ratio of between 2 and 2.4 on a volume basis.

Much of the molten ash generated in the reactor is collected in a water-filled bottom compartment of the reactor. All ash leaving the system is in the form of solidified slag, which presents minimal environmental problems. Any ash droplets that are entrained in the gas exiting the reactor are solidified in a quench section located at the gasifier outlet. The hot raw gas is further processed in a waste heat boiler to produce superheated, high-pressure, process steam. This efficient heat recovery approach results in the production of 0.3 to 0.4 tons of high-quality steam (typically 1000°F at 1450 psia) per ton of moisture and ash free (MAF) coal. The recoverable heat content of the gas and steam represent 92 to 94% of the heat content in the

coal while the overall thermal efficiency, calculated in a lower heating value basis, for the production of a medium-Btu gas is 77% for a 10% ash and moisture content hard coal.

A proprietary particulate cleanup system which consists of a cyclone separator and scrubbers is an integral part of the process. The main advantage of this system, besides its claimed low cost and temperature independence, is that all solids are collected as a dry powder.

The cooled gas, which still contains sulfur compounds, is further processed to remove H_2 and COS. The recovered acid gases are processed in a Claus plant, which removes approximately 92% of the sulfur contained in the gas as elemental sulfur. For further conversion to SNG, the clean gas undergoes CO-shift conversion followed by CO_2 removal, methanation, and drying. After the removal of sulfur compounds and carbon dioxide, the dry synthesis gas contains roughly 69% volume CO, 30% volume H_2 , and 0.5% volume CH_4 . The absence of methane in the synthesis gas, a drawback in the production of SNG, is said to be outweighed by the relative simplicity of gas cleanup and the complete elimination of wastewater treating.

Process Goals

The Shell-Koppers process is a joint venture of Krupp-Koppers GmbH and Shell International Research Maatschappij B.V. The primary purpose of this research is to improve the efficiency and rate of coal gasification by elevating the pressure of the Koppers-Totzek process. Because of the high carbon conversion efficiency, a clean, particulate-free synthesis gas can be produced from almost any type of coal. An important element of this research is the union of Krupp-Koppers' experience in the design and operation of commercial scale Koppers-Totzek atmospheric coal gasifiers, including the preliminary pilot plant design of an elevated pressure gasifier, and Shell's experience in high-pressure oil gasification.

Relationship to Prior Technology

The Shell-Koppers gasification process is similar to the commercially available Koppers-Totzek process in that both of these coal gasification techniques are based on the principle of entrained-bed gasification under slagging conditions. In this type of high temperature (2250° to $2950^\circ F$) suspension process, the coal and hot blast (steam and oxygen) move con-

Table 1 Part 1 TECHNOLOGY FACT SHEET: Shell-Koppers Coal Gasification Process

NEW TECHNOLOGY (PROCESS DESCRIPTION AND GOALS): Shell-Koppers. Entrained-bed gasification under elevated pressure (20 - 40 atm) slagging conditions. Prepared coal feed through diffuser guns, reacts with blast of steam and O₂ (air for combined cycle). Target size: 50-100 TPH coal per reactor, 84.7-169.5 X 10⁶ SCFD raw gas.

RELATIONSHIP TO PRIOR TECHNOLOGY (INCLUDING STATE OF DEVELOPMENT OF PRIOR TECHNOLOGY): Koppers-Totzek process is based on entrained-bed gasification under slagging conditions.

CHARACTERISTICS OF THE TECHNOLOGY:

<u>CHARACTERISTICS</u>	<u>NEW TECHNOLOGY</u>	<u>PRIOR TECHNOLOGY</u>
PRIMARY OUTPUT (DESIGN CASE)...% vol. CO...H ₂	96%	
TYPE OF PROCESS.....	Pressurized entrained bed	
FEEDSTOCK REQUIREMENTS.....	All types of coal*	
OVERALL THERMAL EFFICIENCY (INCLUDING BY-PRODUCTS)..	92 - 94%	
CARBON CONVERSION EFFICIENCY.....	99%	
OPERATING TEMPERATURE.....	2550° - 2950°F	
OPERATING PRESSURE.....	290 - 590 psia	
BY-PRODUCTS.....(tons/ton/coal).....	Steam, slag	

* Ash content < 40%, sulphur content < 8%.

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Table 1, Part 2. **TECHNOLOGY FACT SHEET: Shell-Koppers Coal Gasification Process**

STATUS OF DEVELOPMENT:

OPERATING FACILITIES - 6 TPD pilot plant at Shell's Amsterdam lab. 150 TPD prototype plant
constructed by Krupp-Koppers at Harburg refinery of Dutch Shell.

MAJOR FUNDING AGENCY Joint venture between Shell International and Krupp-Koppers.

ANNUAL LEVEL OF FUNDING - _____

TECHNICAL PROBLEMS:

1. Intricate cooling system. May be replaced by insulating brick in reactor wall.
2. Solids recirculation.
3. Pressurized lock hoppers.
4. Refractories must withstand attack by molten coal mineral residue.

OTHER FACTORS AFFECTING OVERALL FEASIBILITY: Over \$2 X 10⁹ (1980) estimated to develop process to
commercial level.

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currently. In addition to producing a high-quality synthesis gas consisting primarily of carbon monoxide and hydrogen, this type of gasification method has the ability of processing a large variety of coal types. Coals with high ash or moisture content do not present a technical problem, although a slight process efficiency penalty may be encountered.

Numerous advantages are derived when operating the entrained-bed gasifier at elevated pressure. One of the advantages is an increased coal reaction rate that reduces residence time and hence reactor vessel size. The reduced reactor size facilitates reactor scale-up design and, as a result of decreased reactor surfaces area, a reduction in the heat loss rate through the reactor walls will occur.

Operating Facilities

Since December 1976, a 6 ton/day pilot plant has been in continuous operation at the Shell Research Laboratory in Amsterdam. This pilot plant is utilized to verify the process correlations. A larger prototype plant (150 TPD) has been constructed at the Harburg refinery of Deutsche Shell AG, near Hamburg, West Germany, to supply design and engineering data for a full-scale commercial plant design. A series of successful runs, made since November 1978, has demonstrated that hard coal can be gasified with a carbon conversion efficiency of 99% while producing a synthesis gas containing only 1% (by volume) of carbon dioxide. The construction cost of this prototype unit, which is owned and operated by Deutsche Shell AG, was approximately \$50 million.

The current development schedule calls for the completion of test runs in late 1980 followed by demonstration-scale design.

Funding Sources

Royal Dutch Shell and Krupp-Koppers have thus far been unable to gain government financial support for the construction of a 1,000 tonne/day commercial facility in both the Netherlands and West Germany. Higher coal subsidies in West Germany has prompted Krupp-Koppers to seek industrial participation for a proposed plant in Wilhelmshaven, West Germany, to produce methanol. These plans called for the construction of the commercial facility to start in 1983 with operation expected in 1985. Construction decision by

Shell of similar facility in Moerdijk, Netherlands, has been delayed due to an unexpected 25% increase in the construction cost and process technical problems in addition to the lack of government support.

In June of 1982 Shell Oil announced that it will not build either the Moerdijk or Wilhelmshaven demonstration facilities due to their lack of economic feasibility. However, Shell has not ruled out construction of a demonstration facility elsewhere in the world, possibly the U.S. Shell Oil has made it clear that they do not plan to stop research in their process. Continued efforts will be directed toward increasing the economic feasibility of the process.

In addition to possible European development plans, the Tokyo-based Shell member companies of Shell Sekiyu, Shell Kosan Co., and Showa Oil Co. are conducting a feasibility study on the construction of a Shell-Koppers coal gasification facility in Japan. Showa Oil Co. is trying to market this process to potential clients, which include electric and gas utilities as well as the government bodies on New Energy Development Organization (NEDO) and the Agency of Natural Resources and Energy (ANRE).

Technical Problems

Based on the composite questionnaire responses, the following technical problems confront researchers and developers of the Shell-Koppers gasifier:

- Refractory lining material tends to wear from slag erosion.
- The slag quench system produces steam, which interferes with slag removal and reduces product gas quality. Also, stalagmites in the quench bath are difficult to remove.
- Furnace slagging interferes with continuous operation. Accrued wall slag is difficult to remove.
- The heat recovery system must be demonstrated over time.
- The dust-treating system is unproven at high temperatures.
- Scale-up to commercial size must be proven over time.

The questionnaire responses, concerning technical barriers to commercialization, were basically in accord. Of the four respondents on the Shell-Koppers process, three cited both the refractory lining of the reactor and the slag-treating systems as critical to the commercial success of the

technology. One respondent stated that the refractory should be protected by the formation of a "skull" of solid slag on the inside wall of the reactor. This can occur only if the reactor cooling system removes enough heat to prevent the skull from melting and depositing a mineral residue (which causes erosion). The maximum solution would thus be a "permanent skull without the need to cool the reactor shell." This same expert suggests that the slag quench bath be replaced by "an internal grinder with a screw conveyor slag removal apparatus." In the maximum solution, "no erosion and pressure leaks" would occur with the screw conveyor. Concerning the system for dust treatment, three experts described the problem as moderately important to commercial success. The problem seems to be that the offgas contains high levels of particulate matter, which makes heat recovery via conventional heat exchangers difficult. The minimum solution, according to one expert, is a "steam producer exchanger with high temperature cyclones." The maximum solution is to "operate heat exchangers for all pressure steam needs of the plant with automated ash removal...eliminate cyclones/baghouses."

GERMAN COAL GASIFICATION PROCESS

2. High-Temperature Winkler Coal Gasification ProcessProcess Description

In the high-temperature Winkler process, crushed coal, typically 3/8 x 0 inches, is gasified in a fluidized bed. Coal-fired fluidized-bed dryers are utilized to reduce the moisture level of the coal below 18%, if required. The gasifier is cylindrical with a conical bottom. Oxygen or air and steam are introduced into the bottom, conical portion of the gasifier through water-cooled pipes that extend a short distance into the bed through the gasifier walls. Oxygen is injected not only at the bottom, but at various levels along the walls of the cone. At the lower part of the cone, a fluidizing gas, such as recycled product gas or CO₂, is introduced to ensure reasonable fluidization before the point of oxygen injection. The coal is fed via lockhoppers and a screw feeder into the lower cylindrical portion of the gasifier. This solid injection point is below the oxygen injection point, but above the inert fluidization ring. Because of the degree of solids mixing, very little short circuiting of the feed material into the discharge is anticipated. The heavy ash particles formed in the fluidized-bed are removed from the bottom of the gasifier by a water-cooled screw conveyor to an ash-lockhopper. The ash is then conveyed to a disposal hopper.

Entrained char and ash, along with raw gas, is subjected to further gasification in the large space above the fluidized-bed where additional oxygen or air and steam are introduced. The gasifier will be operated at temperatures on the order of 1650°F with additional injection of oxygen or air above the bed to achieve temperatures in the range of 1832°F. The ratio of oxygen to steam to coal can be controlled to maintain the required bed temperature. After the hot gases are processed in the cyclone, they pass through the heat recovery train where heat is removed to produce process superheated high-pressure steam and preheated boiler feedwater. An air preheater is utilized for air-blown operations, but insufficient sensible heat exists in the product gas to preheat the oxygen for oxygen-blown gasification (medium-Btu gas production). The hot gas is cooled to approximately 300°F in this section. The dust removal system is designed to remove a minimum of 85% of the entrained solids. Venturi scrubbers are further used to reduce the remaining char to a level of 1 grain/1000 SCF.

The cooled, cleaned gas is desulfurized by a combination of carbonyl sulfide hydrolysis and hydrogen sulfide removal using the Alkazid Process to a maximum total sulfur content of 100 ppm. The concentrated H₂ from the Alkazid unit is processed in a Claus plant to produce elemental sulfur. A technology fact sheet for this process is presented in Table 1.

Process Goals

The conventional Winkler process is limited to near atmospheric operation and relatively low temperatures, which limits the carbon conversion efficiency to 90%. The objective of this high-temperature Winkler process is to increase the reaction pressure and temperature to improve gasifier efficiency and design. However, the operation of the gasifier at elevated temperatures produces the problem of ash sintering and sticking, which will produce a negative effect on gasifier operation. Experiments at the Institut fur Eisenhutenkunde (Institute of Metallurgy) of the RWTH-Aachen (Technical University of Aachen) have indicated that the injection of limestone will increase the ash-softening temperature and melting point of the ash to form a granular slag. The addition of the lime or dolomite has the additional benefit of bonding with the sulfur compounds in the raw gas to reduce pollution control requirements. The increase in gasifier pressure also benefits the process viability for certain applications. For power station or direct iron-ore reduction, the gas can be delivered hot (with its latent heat) to increase efficiency and reduce compression requirements.

Relationship to Prior Technology

The high-temperature Winkler process is an offshoot of the commercially available Winkler coal gasification process. In 1922, while researching processes for the production of activated carbon, Dr. Fritz Winkler conceived the idea of gasifying coal in a fluidized-bed in order to avoid the expense of briquetting, which is required to convert German brown coal into a producer gas. The first commercial Winkler gasifier was put into operation at Leuna-Werke, Merseburg, Germany, in 1926. To date, 70 gasifiers at 24 plant locations throughout the world (including two pilot plants) have been designed, constructed, and commissioned.

The Winkler process is fully commercial for the production of a low- or intermediate-Btu gas when operated at near atmospheric pressure. Moreover,

Table 1 Part 2 TECHNOLOGY FACT SHEET: High-Temperature Winkler Coal Gasification Process

NEW TECHNOLOGY (PROCESS DESCRIPTION AND GOALS): Pressurized Winkler Coal Gasification Process.

This is a pressurized fluidized process that utilizes non-caking coal, steam, and oxygen or air to produce a synthesis gas. The objective of this process is to increase efficiency and output by increasing operating pressures and temperatures. Also feasible for combined cycle.

RELATIONSHIP TO PRIOR TECHNOLOGY (INCLUDING STATE OF DEVELOPMENT OF PRIOR TECHNOLOGY): Atmospheric pressure Winkler is a fully commercial process. To date, 70 gasifiers are in operation at 24 locations. Davy-McKee is marketing a Winkler gasifier with guarantees to 60 psia. Pressurized Winkler produces slightly more methane at higher efficiency. Rheinbraun is now entering the commercial phase of development with the High-Temperature Winkler process.

CHARACTERISTICS OF THE TECHNOLOGY:

<u>CHARACTERISTICS</u>	<u>NEW TECHNOLOGY</u>	<u>PRIOR TECHNOLOGY</u>
PRIMARY OUTPUT (DESIGN CASE).....	283 Btu/SCF	
TYPE OF PROCESS.....	Pressurized Fluidized Bed	Fluidized Bed
FEEDSTOCK REQUIREMENTS.....	Non-caking coals*	
OVERALL THERMAL EFFICIENCY (INCLUDING BY-PRODUCTS)..	70 to 85%	
CARBON CONVERSION EFFICIENCY.....	95%	91%
OPERATING TEMPERATURE.....	1652° → 2012°F	1400° → 1650°F
OPERATING PRESSURE.....	70 to 300 psia	22 psia
BY-PRODUCTS.....	steam, ash	steam, ash

* Will take mildly-caking coals if limestone injection is used.

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Table 1, Part 2. TECHNOLOGY FACT SHEET: High-Temperature Winkler Coal Gasification Process

STATUS OF DEVELOPMENT:

OPERATING FACILITIES - 14 atm HTW pilot plant at Brown Coal Works (24 metric tpd, 1500 Nm³/hr raw gas), Frechen, FRG. Started up in mid-1978.

MAJOR FUNDING AGENCY - FRG Federal Government (Rheinbraun).

ANNUAL LEVEL OF FUNDING - Funding by government through 1983 only.

TECHNICAL PROBLEMS: The technical problems to be overcome in the development of the High-Temperature Winkler appear to be relatively minor. The problems appear to be related to attempts to extend the technology to accept feedstocks beyond that for which it was originally conceived. Ash sintering, sticking at high temperatures - experimenting with limestone to prevent this.

OTHER FACTORS AFFECTING OVERALL FEASIBILITY:

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Davy McKee, Inc., of Lakeland, Florida (U.S. distributor of the Winkler process), will give normal process guarantees for pressure operation up to 4 atm. Apart from the development work carried out by Davy McKee, Inc., Union Rheinische Braunkohlen Kraftstoff, AG, an affiliate company of Rheinische Braunkohlenwerke AG, is developing an improved Winkler-type gasifier operating at a pressure of up to 10 bar (9.9 ATM) and a higher temperature than the process temperatures of the conventional Winkler gasifier. This development is a continuation of experiences gained from the year 1956 to the end of 1964 when Union Rheinische Braunkohlen Kraftstoff AG operated two Winkler fluidized-bed generators with a total capacity 17,000 m³/hr for the production of a synthesis gas.

Operating Facilities

The high-temperature Winkler gasifier is currently entering commercialization from the pilot plant stage of development. The pilot plant, which was started up in mid-1978, demonstrated the feasibility of operating a modified Winkler fluidized-bed gasifier at elevated temperatures and pressures to improve the gasifier efficiency. The ability of limestone to overcome the coal caking tendencies at higher temperatures, as well as limestone's sulfur scavenging effect, was monitored as part of this program.

The first commercial scale high-temperature Winkler plant will be built by the process developer. Rheinische Braunkohlenwerke AG (Rheinbraun). Groundbreaking for this \$165 million facility occurred in July 1981 at Huerth-Berkenrath, West Germany. The plant, designed to convert 2.2 million tons of brown coal to synthesis gas, consists of four modules, each with a capacity of 300 million cubic meters/year. The synthesis gas output will be pipelined to the Rheinbraun group's Wesseling refinery a few miles south of Cologne for conversion into methanol as chemical feedstock. The first module is expected to be on stream in 1984 with the remaining balance of plant in operation by 1987.

The West Germany firm of Uhde has been awarded the contract to supply design and construction services for the new facility by Rheinbraun. Uhde will also provide engineering services for future commercialization efforts with third parties.

Major Funding Agencies

Rheinbraun is expected to receive government support for the pilot operation from the Ministry of Research and Technology through 1983. At that time Rheinbraun will proceed without subsidies due to the commercial viability of the process.

Technical Problems

Based on the composite questionnaire responses, the following are the technical problems confronting researchers and developers of the high-temperature Winkler gasifier:

- Production of a high-carbon ash wastes a fuel source unless suitable recovery techniques are developed.
- The sintering of the ash at elevated temperatures could produce a negative effect on gasifier operations.

WEST GERMAN COAL GASIFICATION PROCESS

3. Saarberg-Otto Coal Gasification ProcessProcess Description

The Saarberg-Otto gasifier is an entrained flow, high-temperature gasifier which incorporates a slag bath. This slag bath is maintained in a rotating motion by the injection of feedstock and gasifying media through nozzles, which are oriented in a downward and tangential direction toward the surface of the molten slag bath. The slag bath acts as a heat shield and permits the use of a high flame temperature. The use of the slag bath and the injection system also increases the rate of the gasification reaction. This is made possible by the high-turbulence zone above the molten slag bath where rapid removal of the gas film surrounding the solid-matter particles occurs in conjunction with high temperatures. Another advantage that the slag bath ensures is the absolutely safe ignition of the feedstock as well as a stable flame. In addition, the quantity of slag in the raw gas that exits the gasifier is reduced because much of the slag is bonded to the slag bath at the bottom of the gasifier.

Some additional characteristics of the Saarberg-Otto gasification process that can also be attributed to the slag bath are as follows:

- It is reported to be suitable for all fossil fuels having an ash content of up to 40%, irrespective of their caking and crushing properties, and for residues from coal hydrogenation plants
- Solid, liquid, and gaseous fuels can be gasified simultaneously
- The raw gas is free from higher boiling hydrocarbons as tar and gum-forming substances
- The fly coke not gasified amounts to between 10% and 30% of the feedstock, depending on the type of fuel and the reaction conditions. It is removed by dedusters and returned to the gasifier, so the carbon conversion amounts to 99.5%.

The gasifier consists of three sections or stages. Stage I contains molten slag at 2700° to 3100°F. Stage II (the after-gasification zone) has an outer pressure shell and an inner shell of closely spaced, water-cooled tubes. The tube walls are initially protected by a castable refractory, but this is expected to gradually dissolve and be replaced by a layer of solid slag. In Stage III (also refractory lined), raw gas is cooled by recycled gas

to about 1500° to 1700°F to solidify slag droplets. Slag, char particles, and dust (amounting from 10% to 30% of the coal feed) are carried overhead with product gas and are recovered for recycle to the gasifier. Molten slag, which overflows to a bottom weir, contains less than 1% carbon. It is quenched with water and is discharged intermittently through a lockhopper.

The gasifier is reportedly capable of utilizing all types of coal having an ash content of up to 40%, regardless of caking or crushing properties. The coal feed preparation entails crushing in a gas-swept mill using impact hammers. The maximum feed size is 3 mm with 70% being minus 0.05 mm. The coal moisture requirements for the gasifier dictate that hard coals be dried to approximately 2% while brown coals are dried to approximately 12% in order to maintain free flowing of the feedstock. In the demonstration facility, the prepared coal is collected in a 150-ton hopper, which feeds directly into a single-line lockhopper system consisting of an upper lockhopper, double valve, and a lower feedhopper.

The reactants are fed through nozzles into Stage I of the gasifier, which is the primary reaction zone. The gasification reactions take place in a fraction of a second, partially in the nozzle flame, at the 1650° to 2400°C temperature in this zone. The larger particles impinge on the slag bath and, therefore, have longer residence time to complete the gasification flows down through a central overflow to a quench vessel where the slag is granulated.

The demonstration plant gasifier's internal dimensions are 1.4 meters in diameter by 16 meters high. The lower reaction zone, described above, is separated from the middle zone by straighteners designed to negate the whirling effect created in the bottom zone. This gas dewhirling section eliminates undesired slag buildup in Stage II. In Stage II, the gases from the bottom zone enter at a temperature from 1500° to 1700°C. This zone acts as a dampening zone for irregularities that may occur in Stage I and as a secondary reaction zone where another 10% of the carbon is reacted.

In Stage II, the raw gas is cooled from 1500° to 1200°C. This cooling is a function of the shaft height of Stage II and the gas throughput. In Stage III, recycled quench gas is then added to cool the solids entrained in the raw gas stream to 800° to 900°C. Cooling to this temperature is necessary to solidify any entrained slag, thus avoiding fouling of the zone. The gas leaving this zone at about 800°C is 94% free of carryover after processing in a

cyclone. A waste heat boiler follows the cyclone to produce steam at 570°F and 650 psia. After the waste heat boiler, the gases pass through a baghouse and a desulfurization unit. The solids collected in the baghouse and cyclone and are recycled to the feedhopper for mixing with the coal feed. A technology fact sheet for this process is presented in Table 3.

Process Goals

The construction of the Saarberg-Otto demonstration plant represents some areas of new development (first designs). Special examples of first designs in entrained-bed pressure gasification are —

- Coal intake system
- Cooling system
- Slag discharge system
- Removal of dust from product gas.

In the area of coal feeding, the precise metering of a continuously fed dry, fined-grained, and powdery coal into a pressurized system represents a considerable problem which has yet to be satisfactorily resolved. The second area of concern is the especially high temperatures, up to 2400°C, which are generated in the lower gasification section. The Saarberg-Otto slag bath gasifier is, therefore, equipped with a combined boiling/condensation cooling system to deal with this heat flux problem. The slag removal system followed by quenching will also be tested in this facility. In the area of hot gas particulate removal, the demonstration plant will test new technical developments for dust removal under the temperature and pressure conditions present (800°C, 370 psia).

Relationship to Prior Technology

The Saarberg-Otto gasifier is based on an earlier gasifier design known as the Otto-Rummel slag bath gasifier. This gasifier was capable of operating on liquid fuels, as well as coal. When operating on a German brown coal and an oxygen blast, the gasifier could produce 13 million SCFD of medium-Btu gas from 264 ton/day of coal.

In 1960, a new improved gasifier was installed in a Winkler gasification facility for the production of a syngas. Although the peripheral equipment associated with the Winkler gasifier was mismatched to meet the requirements

Table 1 Part I TECHNOLOGY FACT SHEET: Saarberg-Otto Coal Gasification Process

NEW TECHNOLOGY (PROCESS DESCRIPTION AND GOALS): Saarberg-Otto Gasification Process.

Improved version of Otto-Rummel slag bath gasifier. New process is an entrained flow, slag bath, three section gasifier which operates at elevated temperatures and pressures. Apparent goal is increased throughput. Also feasible for combined cycle operation. It is an oxygen blown system to produce synthesis gas.

RELATIONSHIP TO PRIOR TECHNOLOGY (INCLUDING STATE OF DEVELOPMENT OF PRIOR TECHNOLOGY): The Saarberg-Otto

process is related to the single-shaft Otto-Rummel gasifier developed in the 1950's and 1960's. A double shaft Otto-Rummel gasifier was also developed in the 1960's by the British Gas Corp.

CHARACTERISTICS OF THE TECHNOLOGY:

<u>CHARACTERISTICS</u>	<u>NEW TECHNOLOGY</u>	<u>PRIOR TECHNOLOGY</u>
PRIMARY OUTPUT (DESIGN CASE).....	~280 Btu/SCF (oxygen and H ₂ O)	275 Btu/SCF (oxygen and H ₂ O)
TYPE OF PROCESS.....	entrained flow, slag bath	entrained flow, slag bath
FEEDSTOCK REQUIREMENTS.....	all types of coal*	all types of coal*
	3mm max w/70% < 0.05 mm	0 → 5 mm
OVERALL THERMAL EFFICIENCY (INCLUDING BY-PRODUCTS)..	88 → 94%	90 → 94%
CARBON CONVERSION EFFICIENCY.....	99.5%	99%
OPERATING TEMPERATURE.....	2700°F → 3100°F	2900°F → 3300°F
OPERATING PRESSURE.....	370 psia	15 psia
BY-PRODUCTS.....	steam, slag	steam, slag

* All types of coal w/less than 40% ash content.

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Table 1 Part 2 TECHNOLOGY FACT SHEET: Saarberg-Otto Coal Gasification Process

STATUS OF DEVELOPMENT:

OPERATING FACILITIES - Demonstration plant operating at Volklingen/Furstenhauzen, West Germany.
Start-up of the 12.0 MMSCF/D, 264 ton/day coal, facility was in May of 1979.

MAJOR FUNDING AGENCY 75% by Federal Ministry for Research and Technology in Germany;
25% by the Dr. C. Otto GmbH and Saarbergwerke AG.

ANNUAL LEVEL OF FUNDING - 36 million DM initial funding and 12 million for first year of operation.
All funding by the Federal Ministry for Research and Technology will be terminated at the end of 1982.

TECHNICAL PROBLEMS:

Critical problems involve the critical slag tap/reactor diameter ratio for removing slag from the system.

Other technical problems involve reactor scale-up, refractory life, and hot gas particulate removal.

OTHER FACTORS AFFECTING OVERALL FEASIBILITY: Demonstration of process reliability and operating factor.

Large development cost ($\$1.0$ to 3×10^9) may hamper commercialization.

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of the slagging gasifiers, the new gasifier was run satisfactorily for approximately 18 months. Because of the introduction of steam-naphtha reforming in the year 1964, the entire coal gasification facility was shut down.

In addition to the development of the single-shaft Otto-Rummel gasifier, experimental work was performed on a double shaft slag bath gasifier in Wesseling in the early 1950's. The double-shaft gasifier, incorporating an annular space between two concentric shafts, produced a product gas with over 60% nitrogen. Despite this restriction, the British Gas Council (BGC) sponsored development of the double-shaft gasifier. In 1962, the BGC built a gasifier at Bromley, England, which produced 2 million SCFD of medium-Btu gas from Midland coal. This facility was soon shut down because of design and operating difficulties.

The gasifiers mentioned above were all operated at near atmospheric pressure. In January 1976, Dr. C. Otto and Company GmbH in Bochem, Germany, and the Saarbergwerke AG, Saarbrucken, entered into a joint effort to develop, test, and utilize a modified version of the old Otto-Rummel process.

Operating Facilities

The new Saarberg-Otto process is of the entrained flow, high-temperature, slag-bath type and will operate at 25 atmospheres. This demonstration plant at Volklingen/Furstenhausen started up on April 22, 1980, and utilizes 264 metric ton/day of coal with a corresponding gas output of approximately 22,800 m³/hr.

Major Funding Agencies

At present, three phases are involved: construction, operation with oxygen, and operation with air. Funding for the construction phase was estimated to be 36 million DM (1978 DM) and phase II, which is operation with oxygen, was funded for one year at a level of 12 million DM. Three-quarters of this money is supplied by the Federal Ministry for Research and Technology in Germany. Funding by the Federal Ministry for Research and Technology will be completely terminated at the end of 1982 for this process.

Technical Problems

Based on the composite questionnaire responses, the technical problems confronting researchers and developers of the Saarberg-Otto gasifier are as follows:

- Refractory life is adversely affected by corrosion because of the possible existence of molten iron in the slag
- The limitation of reactor diameter (8.5 ft maximum) could cause scale-up problems
- The plugging of the slag tap hole could cause excessive downtime
- The reactor wall cooling system is unproven
- The suitability of all coal types to Saarberg-Otto gasification is unproven.

One of the responses to the questionnaire on Saarberg-Otto gasification outlined some possible solutions to these technical problems. According to that particular respondent, the minimum acceptable solutions in terms of component lifetime are six months for the refractory lining and three years for the water-cooled walls; the maximum solutions are 5 years and 25 years, respectively. The minimum solution for the slag tap is a 70% operating factor (no maximum given). The minimum solution for the reactor diameter problem is to employ 8-ft reactors in multiples; the maximum solution is a 16-ft diameter reactor. The solution to the "coal type" problem is to simply run the gasifier on all coals for an extended period of time (two years). The scale-up problem is estimated to take five years to solve and the other problems can be minimally solved in three years or less. The total cost of resolving all technical problems is estimated to be between $\$1.5 \times 10^9$ and $\$3.0 \times 10^9$, according to this particular respondent.

WEST GERMAN COAL GASIFICATION PROCESS

4. RCH/RAG Texaco Coal Gasification ProcessProcess Description

The Ruhrkohle and Ruhrchemie (RCH/RAG) Texaco coal gasifier is a single-stage, pressurized, downflow, entrained bed reactor that operates under slagging conditions. This type of reactor can utilize both caking and non-caking coals to produce high throughputs of gas relatively free of tars and other by-products. The Texaco process contains four main features which the developer feels are essential for both technical and financial success. These features include high-temperature operation, high reactor pressures, the utilization of pulverized coal, and the slurry introduction of feed into the gasifier.

To prepare the slurry, wet run-of-mine coal is mixed with water before entering a grinding mill. During slurry preparation in the mill the coal is ground to 0-10 mm, and a high solids content, nearly homogeneous mixture, is produced. This nearly homogeneous slurry mixture is then sent to a slurry tank equipped with a stirrer where further homogenization of the slurry takes place. At this point additives may be added to improve slurry flow properties. The slurry that exits the mixing tank is preheated in heat exchangers and then pumped to a specially designed burner at the top of the gasifier.

The burner design and dimensions are based on the data available from Texaco's oil gasification units. Upon the addition of oxygen, and possibly a reaction temperature modulator, at the burner head, autothermal gasification occurs. The partial combustion proceeds within seconds in the resulting flame to supply energy to heat the reactants and drive the enthalpy requirements of the reaction. The reaction occurs at temperatures between 2300° and 2700°F.

The raw gas and slag that exit at the bottom of the refractory lined reactor vessel are deflected at the discharge end of the radiative waste heat boiler, which is located below the gasifier reactor vessel. As the raw gas is cooled to below the ash fusion temperature of the coal, the slag is separated and granulated in a water bath at the bottom of the radiative cooler. The ash that settles in the water bath is collected for removal in a programmable lockhopper. The radiative heat boiler is sized to take advantage of the high-

temperature syngas to the point where convective heat transfer is more economical. The partially cooled raw gas is then sent to the second stage convective waste heat boiler where additional process steam is generated.

The cooled syngas exits the waste heat boilers and passes through a venturi or orifice type scrubber where any remaining particulate matter, as well as ammonia, is scrubbed out by direct contact with water. The wet-scrubbing, which takes place at near gasifier pressures, is expected to remove over 99.9% of the entrained particulates and fly ash. The water removed from the quench section and scrubbing system is sent to a settling tank. The extracted particulate matter is then recycled to the wet grinding mill for reinjection into the gasifier. This step assures high process carbon conversion efficiencies. The gas leaving the scrubbing system has a particulate loading of typically less than 1 mg/Nm.³

The wet gas exiting the scrubber is further cooled in a heat exchanger by cooling water to condense water from the syngas. After this final cooling the gas is processed in a Selexol[®] unit where sulfur compounds, hydrogen sulfide and some carbonyl sulfide is removed. The sulfur removal step is expected to remove approximately 97% of the sulfur compounds. The concentrated hydrogen sulfide stream from the Selexol[®] unit will be converted to elemental sulfur for sale or disposal in a Claus[®] unit. Sulfur emissions for the Claus[®] plant will be reduced by a tail gas treating section. A technology fact sheet for this process is presented in Table 1.

Process Goals

The RCH/RAG variant of the Texaco gasification process results from cooperative agreement of RCH/RAG to improve the facility designs through process development and testing. The main process steps which RCH/RAG has sought to improve include:

- Start-up phase
- Grinding section (mill) and slurry preparation
- Gasifier
- Reactor lining
- Separation of slag/ash

Table 1 Part 1 TECHNOLOGY FACT SHEET: RCH/RAG Texaco Coal Gasification Process

NEW TECHNOLOGY (PROCESS DESCRIPTION AND GOALS): Rurchemie and Ruhrkohle 01 and Gas Texaco Gasifier.

The Texaco coal gasification process (TCGP) is an extension of their synthesis gas generating process (TSGGP) for converting high-sulfur residual petroleum fuels and tars into syngas. The Texaco coal gasifier is a single stage, pressurized, down flow, entrained bed reactor which operates under slagging conditions. The goal of the cooperative research agreement between Texaco and RCH/RAG is to improve the facility designs through process development and testing.

RELATIONSHIP TO PRIOR TECHNOLOGY (INCLUDING STATE OF DEVELOPMENT OF PRIOR TECHNOLOGY): Research on the TSGGP was started in 1949 and resulted in licensing agreements by 1953. More than 75 plants, in 22 countries, have been constructed since 1955 for the ammonia, methanol, and oxo-chemical industries. These plants use liquid feedstocks.

CHARACTERISTICS OF THE TECHNOLOGY:

<u>CHARACTERISTICS</u>	<u>NEW TECHNOLOGY</u>	<u>PRIOR TECHNOLOGY</u>
PRIMARY OUTPUT (DESIGN CASE).....	281 Btu SCF	syngas ~
TYPE OF PROCESS.....	entrained bed	entrained bed
FEEDSTOCK REQUIREMENTS.....	pulverized coal size 0-10mm	heavy oils, tars
OVERALL THERMAL EFFICIENCY (INCLUDING BY-PRODUCTS)..	92%	
CARBON CONVERSION EFFICIENCY.....	>98%	
OPERATING TEMPERATURE.....	up to 2,700°C	
OPERATING PRESSURE.....	600 psig	
BY-PRODUCTS.....	tars, oils	

* cold gas efficiency 74%.

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Table 1, Part 2. **TECHNOLOGY FACT SHEET: RCH/RAG Texaco Coal Gasification Process**

STATUS OF DEVELOPMENT:

OPERATING FACILITIES - Texaco pilot plant (15 tons/day) in Montebello, Cal. since early 50's. Demon-
stration plant built at Oberhausen-Holtzen, West Germany in 1978. Tests at this
6 ton/hr facility are still underway. TVA has recently started up a Texaco
gasifier at their Muscle Shoals facility.

MAJOR FUNDING AGENCY Two-thirds funded by Federal German Ministry for Research and Technology.

ANNUAL LEVEL OF FUNDING - Initial plant cost 29 MM DM, Annual operating cost offset by syngas sale to
Ruhrchemie. Government support for commercial development seems likely.

TECHNICAL PROBLEMS: From the tests to date, two areas of critical concern are the ash removal systems and
the refractory lining of the gasifier vessel. Molten ash removal has caused a shut-down at the Muscle Shoals
Texaco facility and equipment re-design is required. System re-design was also required at the RCH/RAG pilot
plant in 1978. The most suitable ceramic lining materials tested to date have expected operating lifetimes
of just over 10,000 hours. This relatively short lifetime may call for gasification vessel re-design.

OTHER FACTORS AFFECTING OVERALL FEASIBILITY:

- Testing of alternative components required
- Substitution of water as the suspension agent
- Optimization of heat transfer equipment
- Carbon recycle system needs further demonstration
- Testing of different types of coals required.

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Research on the RCH/RAG version of the Texaco coal gasification process also includes the following aspects:

- Test of alternative components and systems
- Investigation of fundamental relationships and further system optimization
- Substitution of water as the suspension agent
- Optimization of heat transfer equipment
- Optimization of recycle carbon and noncombustible materials
- Testing of different geographic coals

Relationship to Prior Technologies

The Texaco coal gasification process (TCGP) is an extension of their synthesis gas generating process (TSGGP) for converting high-sulfur residual petroleum fuels and tars into synthesis gas. Research on the TSGGP was started in 1949 and resulted in licensing agreements by 1953. More than 75 plants, in 22 countries, have been constructed since 1955 for the ammonia, methanol, and oxo-chemical industries.

The feedstock flexibility of the TSGGP suggested to Texaco that this concept could also be used for converting coal into synthesis gas. The abundance of coal in the U.S. and its potential role as a leading energy supply prompted Texaco to initiate process development research on the Texaco coal gasification process in 1984. In the early 50's Texaco's Montebello research laboratory, east of Los Angeles, started development work on a 15 ton/day coal gasification pilot plant. This resulted in the construction and two-year operation of a 100 ton/day demonstration plant in Morgantown, West Virginia, in 1956. This project was sponsored by the U.S. Government through the Bureau of Mines. Eastern coal was used in the gasifier to produce synthesis gas to make ammonia. This air-blown quench-type gasifier operated at 400 psig and incorporated a refractory lining and water jacket. Due to the increasing availability of low-cost oil and natural gas, this demonstration plant research was discontinued.

The Arab oil embargo in 1973 rekindled Texaco's interest in coal-gasification. During this same time period Ruhrchemie AG, a syngas consumer, and Ruhrkohle AG, a coal producer and processor (RCH/RAG) of West Germany began their own investigation of coal gasification technologies. At that time,

their conclusions resulted in the selection of the Texaco process for commercial applications. In 1975, RCH/RAG entered into a licensing agreement with Texaco Development Corp. to adapt the TCGP to a commercial scale. Ruhrchemie's and Ruhrkohle's experience in the commercial scale equipment aspects related to coal gasification, and the experimental results from Texaco's Montebello gasifier were combined to build a 150 ton/day demonstration plant at Oberhausen-Holtent, West Germany.

Operating Facilities

Ruhrkohle and Ruhrchemie have operated a 160 ton/day Texaco coal gasification pilot plant since 1977 in Oberhausen, West Germany, More than 50,000 tons/day of coal have been gasified in 10,000 hours of operation. Texaco, U.S. also operates two 15 ton/day gasifiers at its Montebello California research facility. Another Texaco gasification facility is undergoing testing at the Tennessee Valley Authority's Muscle Shoals ammonia facility.

The Texaco coal gasification process has thus far been licensed to a number of plants around the world. These plants and other organizations considering Texaco coal gasification technology include following:

- Tennessee Eastman project
- Cool Water Combined Cycle Coal Gasification project
- Tennessee Valley Authority project
- Dow Chemical (Texas) project
- Ruhrkohle and Ruhrchemie project
- Alsands project
- WyCoal Gas project
- SRC-II project
- Mitsubishi Heavy Industries and Central Research Institutes of the Electric Power Industry (Japan) Combined Cycle project
- Rotterdam Municipal Utility project
- Ube Industries (Japan) Ammonia project
- Nyanes Petroleum (Sweden) project
- Moers-Meerbeck (West Germany) Combined Cycle project

Major Funding Agencies

The total demonstration plant costs were 29 million Deutsche Marks (DM), with approximately two thirds of the funding from the Federal German Ministry for Research and Technology. This relatively minor project cost is the result of effective utilization of the Ruhrchemie plant infrastructure and the sale of the syngas to Ruhrchemie's syngas network, which helps to offset the annual operating cost expenditures. The companies of Carl Still, Rechlinsghausen, and Friedrich Uhde, Dortmund FDR, are monitoring the project. Ruhrkohle's project responsibilities are being carried out by its subsidiary "Gesellschaft fur Vergasung and Verflussigung von Steinkohle mbH."

In June of 1982 the West German Ministry of Economics requested additional environmental and economic information from RCH/RAG in order to make a final decision to assist in the construction of a demonstration facility. The Ministry can grant up to half of the 220 million Deutsch Mark (\$93 million DM) cost of the project. The Ministry is expected to make a final decision of the RAG/RCH Texaco demonstration project in September or October of 1982.

Technical Problems

From the test results to date, two areas of critical concern are the ash removal systems and the refractory lining of the gasification vessel. The present shut-down of the TVA Muscle Shoals Texaco plant due to ash removal problems is an illustration of those problems. Ash removal problems were also encountered at the RCH/RAG Texaco pilot plant. These problems resulted from the unexpectedly low density of the slag, which inhibited settling velocities in the slag bath. The low bulk density ash also created very high transport volumes. These problems were overcome at the RCH/RAG pilot plant through system redesign. Another ash removal problem centers on the necessity to use special valves which must be resistant to the strongly eroding slag solids. These valves are subject to failure if lockhopper operation is not closely monitored.

Another area of major concern at the onset of the RCH/RAG pilot plant operation was reactor lining life. A test program to determine the best refractory lining material was undertaken at the Holten pilot plant. In special test zones in the reactor jacket lining, some 50 different ceramic materials from various manufacturers were tested. Inferior refractory lining

life times were encountered in the early tests due to early failure of the weakest materials. The most suitable ceramic lining materials tested to date have expected lifetimes of more than 16,000 hours. Unfortunately, this is less than one year of commercial plant operation. Strict temperature control will be required for commercial operation.

Another problem that was mentioned in the questionnaire responses is that because the RCH/RAG Texaco is an entrained gasifier, the inventory of coal in the gasifier is small. Consequently, the stability of the process becomes very dependent upon the capability to adequately monitor the conditions of the process. Measurement of the reactor temperature has been a particular problem of the RCH/RAG pilot plant as the temperature monitoring devices apparently degrade in their environment of slag and hydrogen.

WEST GERMAN COAL GASIFICATION PROCESS

5. Ruhr 100 High Pressure Lurgi Coal Gasification ProcessProcess Description

The Ruhr 100 gasifier is designed to operate between 25 and 100 bars (360 to 1450 psi) at a coal feed rate of 3 to 7 metric tons per hour. The 135 metric ton/per day pilot plant gasifier has a 1.5 meter inside diameter and a height of 11.5 meters, excluding the coal lockhoppers. The total reactor height including the ash and coal lockhoppers is more than 20 meters. Coal feed at the Ruhr 100 pilot plant is stored in bunkers at the neighboring Furst Leopold mine for transportation to the gasifier by a 350 meter conveyor belt system. The coal arriving at the gasifier plant is then dedusted and screened to 6.3 mm before being stored in an 85 metric ton coal bin. Coal from the bin is fed to a weighing system before injection into the coal lockhoppers. Two alternating mode lockhoppers are used to feed coal into the gasifier to reduce lockhopper gas losses by 40%.

The pressurized Ruhr 100 reactor vessel consists of three sections with a cooling water jacket surrounding the middle and bottom sections. The outer walls of the gasifier vessel are made of a special steel alloy, formerly used only on a nuclear reactor, to help minimize the wall thickness. The inner shell of the reactor is comprised of a number of different alloys in order to collect data for future designs of larger units. Special temperature measurement equipment is also installed in the pilot plant gasifier in order to monitor the gasification reactions at various points within the gasifier.

As the coal enters the gasifier via the lockhoppers, it descends through a drying zone, carbonization zone, gasification zone and combustion zone. The slowly descending coal is evenly distributed within the gasifier by an externally driven rotating grate. A special feature of the Ruhr 100 gasifier is the addition of an extra raw gas outlet located below the carbonization zone. This second outlet makes it possible to influence the temperature and flow rate of the hot raw gases into the devolatilization zone. The effect of this control mechanism will be to reduce the amount of fines carryover by a reduction of gas velocity through the topmost gas outlet. The product gas exits the "clear gas" outlet below the carbonization zone at a temperature of approximately 800°C and the carbonization zone outlet is rich in methane and

hydrocarbon liquids which were formed in the carbonization zone. A technology fact sheet for this process is presented in Table 1.

Process Goals

The basic objective of the Ruhr 100 project is to increase the operating pressure of the Lurgi gasifier from its normal operating pressure range of 20 to 28 bars (290 to 400 psi) to a maximum operating pressure of 100 bars (1450 psi). One positive outcome in this increased pressure operation will be an increase in gasifier throughput without increasing the gasifier diameter. In addition bench scale and theoretical research by G. Baron has indicated that due to the higher partial pressure of hydrogen of the carbonization products results in a 60% to 80% (by volume) increase in methane formation over 25 bar operation. The increase in methane production within the gasifier reduces coal and oxygen consumption due to the exothermic reaction of methane formation and the resulting reduction of the partial combustion of coal within the gasifier to produce heat. The results of tests made by Sustmann and Ziesche also indicate a 75% reduction of tars in the raw gas stream at the 100 (1450 psi) bar operating pressure. This is due in part to the hydrogasification of some of the tars in the gasifier. In addition to less tar production, fewer higher hydrocarbons and phenols are formed as a result of the 100 bar (1450 psi) gasification pressure.

The increase in methane production within the gasifier is of particular importance to SNG plant design and economics and of obvious concern to the Ruhrgas AG. The increase in the methane content of the raw gas will reduce the size of the downstream balance plant, especially the methanation plant, to further reduce plant costs. An added benefit to increased pressure operation is the production of SNG above the typical pipeline operating pressure of 70 bars (1015 psi) which will eliminate the need for recompression equipment.

Another special feature of the Ruhr 100 gasifier is the addition of a second raw gas outlet just above the gasification zone. This second gas outlet will reduce fines carryover by reducing the gas velocity before the devolatilization zone. The gas exiting this second outlet will also contain far fewer condensible products because these condensible products are formed predominantly in the devolatilization zone and withdrawn from the topmost (1st) gas outlet. The reduction of fines carryover will enable the Ruhr 100 gasifier to accept a wide range of coal particle sizes including possible run-of-mine coals.

Table 1 Part 1 TECHNOLOGY FACT SHEET: Ruhr 100 High-Pressure Lurgi Coal Gasification Process

NEW TECHNOLOGY (PROCESS DESCRIPTION AND GOALS): The Ruhr 100 gasifier is a fixed bed, countercurrent flow reactor consisting of three sections with a cooling water jacket around the middle and bottom sections.
The goals of the project are to increase throughput and methane production by increasing reaction pressures to 1450 psia. Other goal include operation of the gasifier on run-of-mine coals and "clear" gas removal from novel side port.

RELATIONSHIP TO PRIOR TECHNOLOGY (INCLUDING STATE OF DEVELOPMENT OF PRIOR TECHNOLOGY): The "Ruhr 100" project is an extension of the continuous research effort to improve the Lurgi coal gasification process. This research has led to the development of the Mark IV gasifier, the British Gas Slagging Lurgi gasifier and the Ruhr 100 gasifier. The Lurgi gasifier has been commercially available for more than 40 years. The Ruhr 100 gasifier is currently in the pilot plant stage of development with a 135 metric ton/day plant operating in the FRG.

CHARACTERISTICS OF THE TECHNOLOGY:

<u>CHARACTERISTICS</u>	<u>NEW TECHNOLOGY</u>	<u>PRIOR TECHNOLOGY</u>
PRIMARY OUTPUT (DESIGN CASE).....	340 Btu/SCF	309 Btu/SCF
TYPE OF PROCESS.....	Fixed Bed	Fixed Bed
FEEDSTOCK REQUIREMENTS.....	Screened to \leq 6.3 mm	--
OVERALL THERMAL EFFICIENCY (INCLUDING BY-PRODUCTS)..	79.7% ^{**}	62.6%
CARBON CONVERSION EFFICIENCY.....	98.9%	--
OPERATING TEMPERATURE.....	800°C	Below coal's ash fusion temperature
OPERATING PRESSURE.....	1450 psia	370 psia
BY-PRODUCTS.....	tar, oils, ash	Ash, steam, tars, oils, phenols

* Represents efficiency for high Btu gas production
 ** Cold gas efficiency for syngas

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Table 1 Part 2

TECHNOLOGY FACT SHEET: Ruhr 100 High-Pressure Lurgi Coal Gasification Process

STATUS OF DEVELOPMENT:

OPERATING FACILITIES - The Ruhrgas/Ruhrchemie Lurgi coke-oven gas facility at Dorsten, FRG, was selected for the 135 metric ton/day Ruhr 100 pilot plant site: this decision was based on the availability of offsite facilities and experienced operators that were still available from the former Lurgi operations that closed in 1967.

MAJOR FUNDING AGENCY 75% funding by German Ministry of Research and Technology until the end of 1982.

ANNUAL LEVEL OF FUNDING - Four year funding program at 150 million DM level until the end of 1982.

TECHNICAL PROBLEMS: The areas of critical concern include lock hopper operation at high pressures, fouling due to fines carryover, utilization of caking coals, and plugging of downstream processes. Of these problems lock hopper operation at high pressures for coal feeding and ash removal and operation which involves large amounts of gas, high speed operation, erosion of valves, control system reliability and sealing are areas of great concern.

OTHER FACTORS AFFECTING OVERALL FEASIBILITY: Ruhrkohle is an owner/parent company of Ruhrgas. Incentive for development is to provide SNG for Ruhrgas while providing a market for Ruhrkohle's coal.

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Relationship to Prior Technology

The "Ruhr 100" project is an extension of a continuous research effort to improve the Lurgi coal gasification process. This research, which started with the theoretical work of Drawe and Danulat, produced the first generation Lurgi gasifier for the gasification of lignite under pressure in the early 1930's.

Due to the loss of availability of lignites from Central Germany after World War II, Lurgi and Ruhrgas continued research to adapt the Lurgi process to the gasification of hard coals such as the mildly caking subbituminous variety available in the Ruhr district of West Germany. This renewed research led to the construction of a pilot plant in 1950 which was equipped with a 1 meter diameter gasifier at the Ruhrchemie site in Oberhausen-Holtens, West Germany. The pilot plant tests resulted in two different second generation Lurgi designs.

The Dorsten site acted as a proving ground for technical improvements to the Lurgi process. These improvements were in the area of corrosion control, grate water cooling systems, raw gas treatment, and coal and ash lockhopper design. Although the second generation designs represented a marked improvement to gasifier throughput, further increases in gasifier throughput were still desirable. Based on the operating experience gained in the Dorsten facility, a third generation design with a larger internal diameter was introduced in 1969.

In the wake of the Arab oil embargo of 1973, a sense of urgency developed to further improve the Lurgi gasification design. This research effort has centered on increasing gasifier capacity, especially for SNG plants, and increasing the range of gasifiable coal type and coal size distributions, including run of mine coals. This has led to the development of the Mark V gasifier, the British Gas slagging Lurgi gasifier, and the Ruhr 100 gasifier, all based in part on the basic Lurgi design philosophy. Work on the Ruhr 100 project started in 1973 when Ruhrgas AG, Ruhrkohle AG, and Steag AG formed a joint venture to explore a high pressure Lurgi option for hard coal gasification to increase gasifier throughput and methane conversion rates.

Operating Facilities

The Ruhrgas/Ruhrchemie Lurgi coke-oven gas facility at Dorsten, West Germany was selected as the Ruhr 100 pilot plant site. From the first test in September of 1979 to September of 1981 a total of 15 tests were conducted. Over 2360 hours of operation were logged which consumed about 16,000 tons of coal and produced nearly 9.6 million m³ of gas. The tests lasted from 3 to 20 days. During the most recent test conducted at 96 atms, 60 to 100% tar recycle was achieved over a ten day period. Previous tests proved gasifier operation with run-of-mine coal having an ash content of up to 50%.

After the pilot plant stage of development which is scheduled to be partially funded by the German Government through 1982. Plans call for the possible construction of a 3-million tonne/year commercial facility to produce 1.5 billion m³/yr of SNG.

Major Funding Agencies

The total engineering design and cost of the pilot plant in 1977 was about DM 60 x 10⁶. An additional DM 90 x 10⁶ was spent to cover four to five years of operating costs. The West German Ministry of Research and Technology has funded 75% of this cost to date. Funding is expected to last through 1982. It does not appear that the Ministry will fund this project after this period.

Technical Problems

Although pilot plant testing is still underway, areas of critical concern have surfaced. These areas include lockhopper operation at high pressures, fouling due to fines carryover, utilization of caking coals, and plugging of downstream processes. Of these problems, lockhopper operation at high pressures for coal feeding and ash removal and operation with caking coals are the most important. Due to rapid pressurization and depressurization which involves large amounts of gas, high speed operation, erosion of valves, control system reliability and sealing are areas of great concern. The utilization of caking coals will involve better control and operation of the devolatilization section in order to avoid coal caking in the lower sections of the gasifier.

The Ruhr 100 high pressure Lurgi gasifier is, in general, a more complex gasifier with regard to control systems because of the high pressure

However, the Ruhr 100 operates at temperatures below the coal's ash fusion temperature and some of the problems expected with the British gas slagging Lurgi type gasifier are avoided. The most critical problems avoided are the reactor lining corrosion/erosion problem and the ash removal (slag tap) design problem.

During testing in the fall of 1981, the coal feed preparation was changed from 6 mm to 35 mm feedstock, to a feed containing 40 to 45% by weight below 6 mm material. Plugging of the clear gas outlet occurred and all gas was withdrawn from the carbonization gas outlet.

WEST GERMAN COAL GASIFICATION PROCESS

6. KGN Fixed Bed Coal GasificationProcess Description

The Kohlegas Nordrhein GmbH (KGN) fixed bed gasifier consists of a conventional revolving grate with a carbonization shaft as shown in Figure 1. The gasifier has been designed to operate in both a cyclic and continuous mode. However, synthesis gas (medium Btu-gas) can be only produced in the continuous mode of operation. In the continuous mode the gasifier can be operated between atmospheric and 30 bars of pressure depending on end use application.

Due to the nature of the counter flow fixed bed design this process can only operate on non caking or slightly caking coals. In addition, coal feed size requirements are also critical. To overcome coal feed size requirements and the ever increasing production of fines in modern mining techniques, the KGN process utilized anthracite briquettes of approximately 40 grams in weight produced from fines. These briquettes are introduced into the gasifier via a coal-lockhopper.

The coal which enters the gasifier passed through a rotating distributor grate at the top into the carbonization zone where carbonization gas is produced. This crude gas containing tars and higher hydrocarbon gases is drawn into the recycle carbonization shaft by steam injection. By drawing the tar and soot laden gases into the gasification zone through the carbonization shaft, all tars and higher hydrocarbons are thermally cracked. In addition, this technique also guarantees that only coke reaches the gasification zone as it is formed in its downward movement in the gasifier.

The oxygen and part of the steam required for the gasification reaction are injected into the bottom of the gasifier under the revolving ash grates. These grates separate the ash which is then collected in an ash lockhopper underneath the gasification vessel.

The superheated steam (580°C) required for the process is partially produced by heat exchange with the exiting gas and from heat collected by the gasifier cooling jacket. The oxygen and the steam required for gasification are injected at 35 bars. As the tar-free synthesis gas exits the gasifier at 800°C and 30 bars, most of the soot in the gas is removed in hot cyclones. The gas is then cooled to 280°C before entering a final soot removed stage using venturi scrubbers.

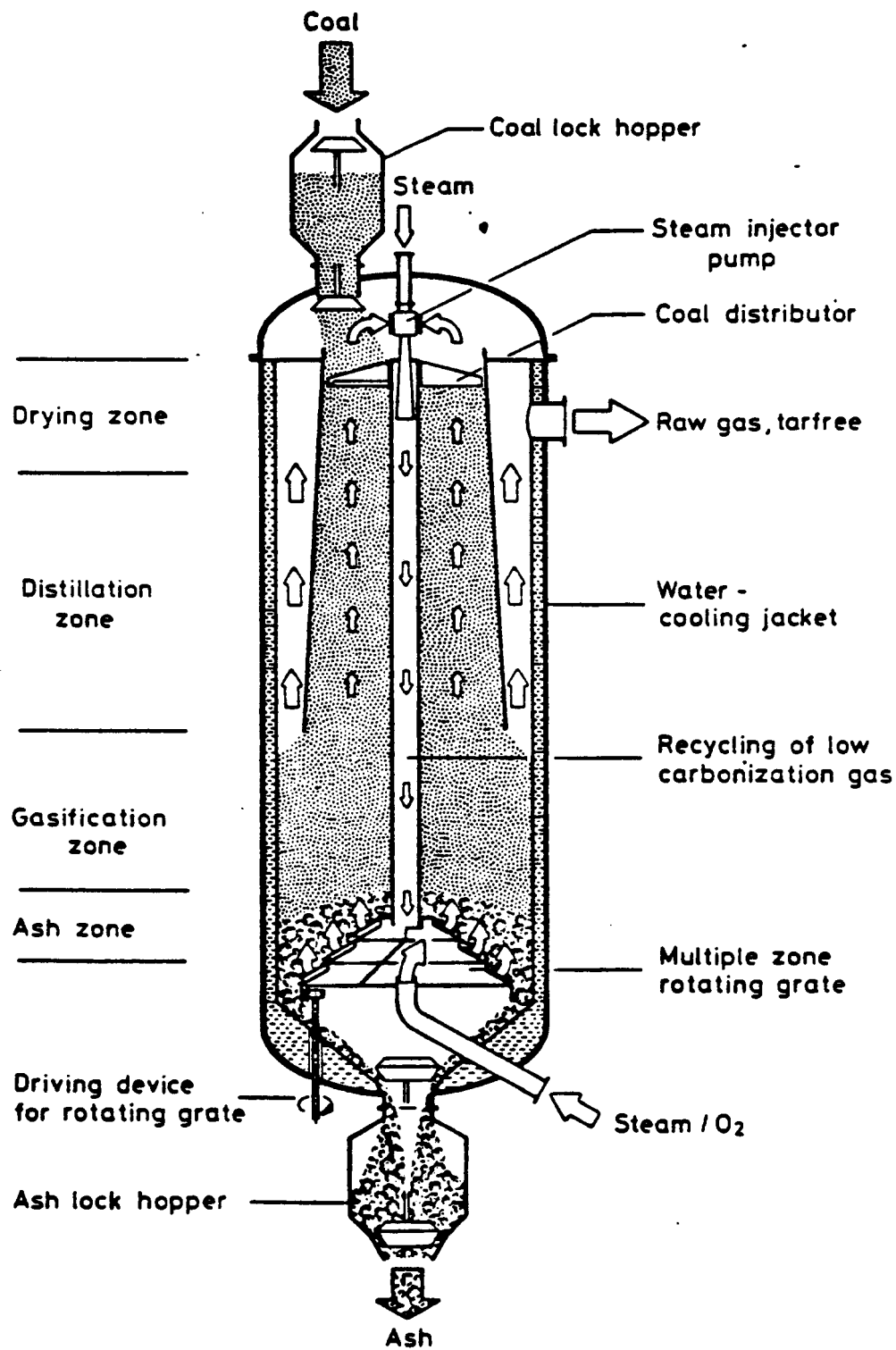


Figure 1. KGN FIXED-BED GASIFIER

Table 1 Part 1 TECHNOLOGY FACT SHEET: KGN Fixed Bed Coal Gasification

NEW TECHNOLOGY (PROCESS DESCRIPTION AND GOALS): The KNG gasifier consists of a conventional revolving grate fixed bed gasifier which incorporates a carbonization shaft (Figure 1). Coal, fed through lockhoppers, descends slowly through the integrated pyrolysis/char bed to the gasification zone. Raw synthesis gas containing tar is recycled to the combustion zone for thermal cracking. Project goals include the production of a tar-free gas which would simplify gas conditioning. Other goals include the use of fines with a high ash content, the development of a briquetting stage, and the development of a suitable gas cleaning system.

RELATIONSHIP TO PRIOR TECHNOLOGY (INCLUDING STATE OF DEVELOPMENT OF PRIOR TECHNOLOGY): _____

Based on a conventional counter flow fixed bed principle.

CHARACTERISTICS OF THE TECHNOLOGY:

<u>CHARACTERISTICS</u>	<u>NEW TECHNOLOGY</u>	<u>PRIOR TECHNOLOGY</u>
PRIMARY OUTPUT (DESIGN CASE).....	234 Btu/SCF	
TYPE OF PROCESS.....	Pressurized fixed bed	
FEEDSTOCK REQUIREMENTS.....	Non-caking coals	
OVERALL THERMAL EFFICIENCY (INCLUDING BY-PRODUCTS)..	80%	
CARBON CONVERSION EFFICIENCY.....	850°C	
OPERATING TEMPERATURE.....	30 bars	
OPERATING PRESSURE.....	Steam, ash, sulfur	
BY-PRODUCTS.....		

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Table 1 Part 2 TECHNOLOGY FACT SHEET: KGN Fixed Bed Coal Gasification

STATUS OF DEVELOPMENT: Pilot Plant

OPERATING FACILITIES - Two ton/hr pilot plant capable of operating in the cyclic or continuous mode. Pilot Plant is located near Huchelhoven in the colliery of Sophia-Jacoba

MAJOR FUNDING AGENCY West German State Nordrhein/Westfalia

ANNUAL LEVEL OF FUNDING - Total cost approximately 19 million DM.

TECHNICAL PROBLEMS: Early operating problems with the grate drives and lockhoppers were reported but have since been fixed.

OTHER FACTORS AFFECTING OVERALL FEASIBILITY: Briquetting requirements may prove uneconomical unless inexpensive source of coal fines is available.

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After further cooling the 75°C the gas can be cleaned of CO₂ and sulfur compounds in a Stretford unit. The final gas processing step involves gas drying in a Glycol unit.

Process Goals

The objective of the Kohlegas Nordrhein project is to develop a commercial size fixed bed gasifier which is easy to operate and which can produce tar-free gases. In addition to the production of tar free gas the sensible heat of the crude gas will also be available.

Relationship to Prior Technology

The KGN process is based on the commercially proven fixed bed counter flow gasifier principle.

Operating Facilities

Construction of a 2.0 tonne/hour pilot plant was completed in February 1979. The reactor, which has an internal diameter of 2.1 meters is capable of operating at pressures of up to 7 bars. Over 200 tons of briquette coal have been processed in the facility with a turndown ratios of nearly 20%. The longest run of 1000 hours was achieved without problems. Only two operators per shift are required for this fully automatic pilot plant design. Briquettes for the process are produced on site by the colliery in which the pilot plant is located. This facility, located near Huchelhoven, West Germany, is operated by Gewerkschaft Sophia-Jacoba. In actuality Kohlegas Nordhein GmbH is a joint venture company of Gewerkschaft Sophia-Jacoba and Projektierung Chemische Verfahrenstechnik GmbH. This site was selected due to the availability of nearly 1.8 million tons/year of coal fines from the colliery operation.

Major Funding Agencies

The project is sponsored by the Ministry of Economics of the state of Nordrhein Westfalia, West Germany. Project cost is estimated to be approximately 19 million DM. Operation of the pilot plant phase of development is expected to continue through 1982. The State Ministry of Economics is expected to continue funding of this process through the demonstration phase.

Technical Problems

During early pilot plant operation mechanical difficulties were encountered with the rotating grate drive units and the coal-lockhoppers. Operating problems with the carbonization recycle tube also occurred. However, all of these problems were overcome and the pilot plant has since sustained smooth operation.

WEST GERMAN COAL GASIFICATION PROCESS

7. VEW Coal Conversion ProcessProcess Description

The Verinigte Elektrizitatswerke Westfalen (VEW) coal conversion process has been developed for combined cycle power generation applications. However, it can also be applied to synthesis gas and reducer gas production when operated with oxygen.

In this process, shown in Figure 1, coal is partially gasified in the entrained flow gasifier. Prior to gasification the coal must be pulverized to a partical size of 90 μm or less. In the pilot plant operation, the coal is pneumatically transported to a heat exchanger where it is heated to a temperature of 350 to 400°C. Next the coal undergoes pre-treatment with steam for 2 to 3 seconds in a reaction tube to reduce its caking tendencies. Coal pre-treatment may not be necessary since pilot plant operation seems to indicate that the coal's caking properties do not affect gasifier operation.

The pulverized coal is injected through a gasification burner at the top of the reactor with preheated air or oxygen and 600°C steam (see Figure 2). Since the gasifier operates at atmospheric pressure, the use of a lockhopper feed system is not required. The coal entering the burner undergoes rapid devolatilization and partial combustion to form a tar free gas and coke. Due to the rapid reaction rates the coke which is formed has a large surface area. This large surface area promotes desulfurization. The resulting crude gas and coke exit the bottom of the gasifier at 1200 to 1300°C and the coke is separated out for use as a boiler fuel. Waste heat is recovered from the hot crude gas at a level of 100 to 150°C. Heat is also recovered from the hot exiting coke stream. The gas can then undergo further treating depending upon application. At a coal conversion rate of 50% to 60%, approximately 80% of the gasifier steam requirements can be produced from waste heat recovery.

Coke formed in this process has been tested as a boiler fuel in separate tests. It's combustion characteristics show very good ignition and burning qualities comparable to hard coal firing.

A technology fact sheet for this process is shown in Table 1.

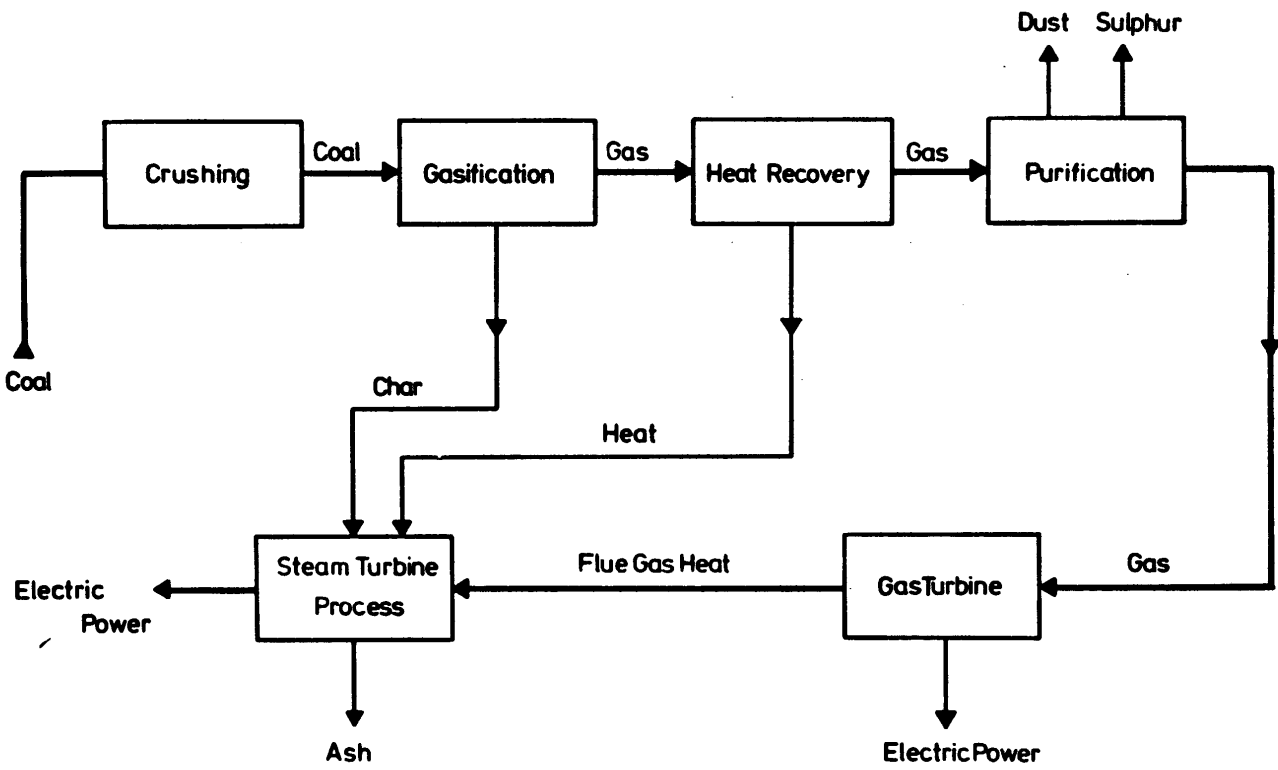


Figure 1. VEW-COAL CONVERSION PROCESS

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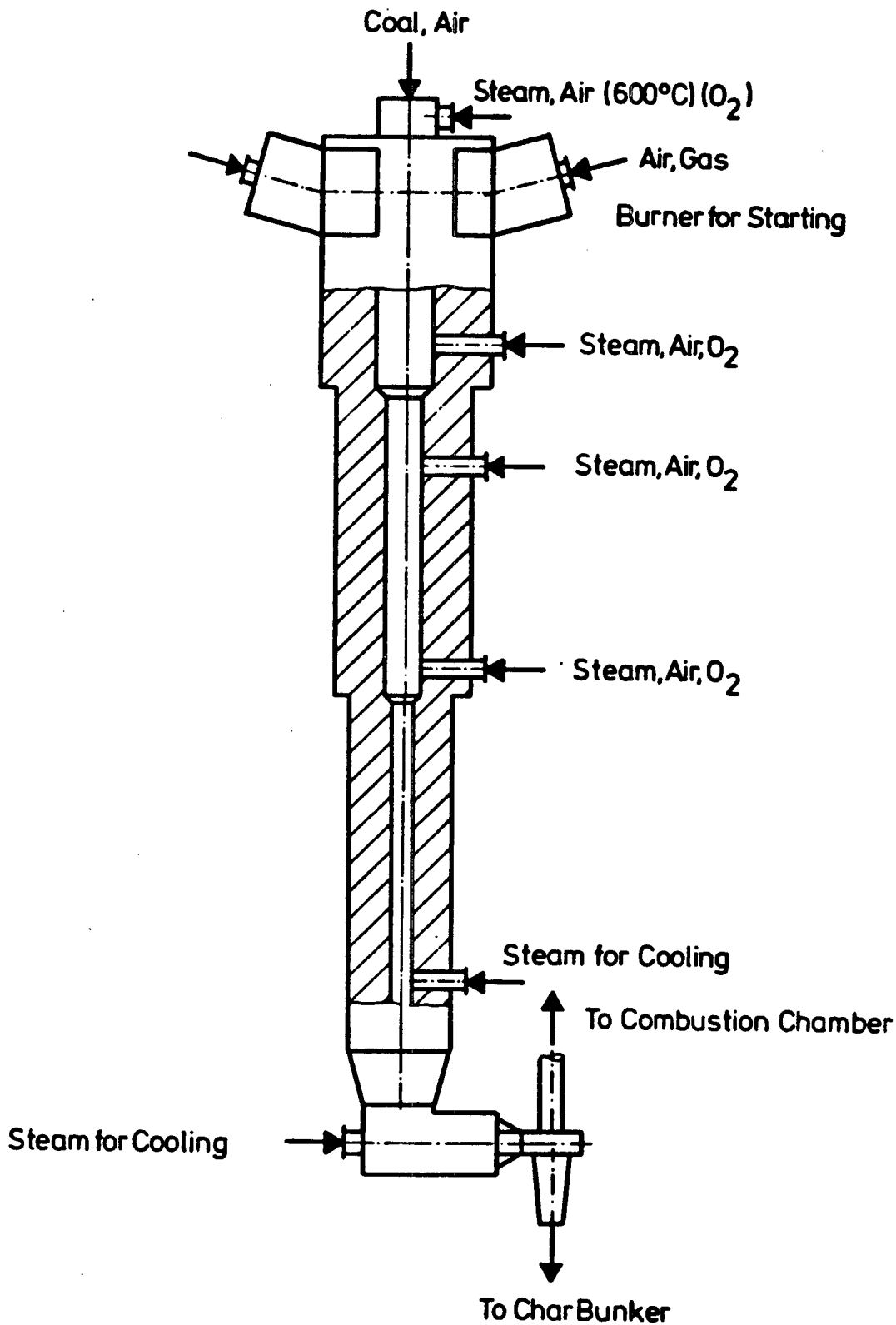


Figure 2. VEW-COAL CONVERSION PROCESS REACTOR

Table 1 Part 2 TECHNOLOGY FACT SHEET: VEW Coal Conversion Process

NEW TECHNOLOGY (PROCESS DESCRIPTION AND GOALS): The VEW process has been developed primarily for combined cycle applications. This partial gasification, entrained flow gasifier is claimed to significantly reduce the sulfur content of the by-product coke thereby making it more attractive for direct combustion applications.

RELATIONSHIP TO PRIOR TECHNOLOGY (INCLUDING STATE OF DEVELOPMENT OF PRIOR TECHNOLOGY): _____

CHARACTERISTICS OF THE TECHNOLOGY:

<u>CHARACTERISTICS</u>	<u>NEW TECHNOLOGY</u>	<u>PRIOR TECHNOLOGY</u>
PRIMARY OUTPUT (DESIGN CASE)..... syngas	@30 to 60 coal conversion	44.4 to 48.5 Vol.% H ₂ , approx. 44.6 Vol.% CO, 1.1 to 8.3 Vol. % CH ₄
TYPE OF PROCESS.....	partial gasification - entrained bed	all types of coal
FEEDSTOCK REQUIREMENTS.....	41% coal to electricity	
OVERALL THERMAL EFFICIENCY (INCLUDING BY-PRODUCTS)...	designed operation 30 to 60%*	
CARBON CONVERSION EFFICIENCY.....	1300°C	
OPERATING TEMPERATURE.....	1 atm	
OPERATING PRESSURE.....	coke, sulfur	
BY-PRODUCTS.....		

* low carbon conversion efficiency due to partial gasification

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Table 1

TECHNOLOGY FACT SHEET: VEW Coal Conversion Process

STATUS OF DEVELOPMENT:

OPERATING FACILITIES - A one tonne/hr pilot plant has been in operation at Stockum, West Germany since 1977. A 15 tonne/hr demonstration plant began operation in 1981.

Plans call for the construction of a 1.8 million tonne/yr commercial plant by 1985.

MAJOR FUNDING AGENCY - The Federal Ministry for Research and Technology, West Germany.

ANNUAL LEVEL OF FUNDING - The 1 tonne/hr pilot plant cost 9 million DM to construct and the 15 tonne/hr demonstration plant cost 37 million DM to construct.

TECHNICAL PROBLEMS:

Coal caking tendencies and heat recovery from dust laden gases were originally seen as problem areas. However, these areas posed no technical problems during pilot plant operation.

OTHER FACTORS AFFECTING OVERALL FEASIBILITY:

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Process Goals

The main objective of the VEW development work is to apply this process to combined cycle power generation. In addition, the partial gasification principle is being tested for the production of low sulfur coke. The favorable sulfur scavenging effect in the reactor during gasification produces a 70 to 80% sulfur removal rate based on a 50 to 60% coal conversion efficiency.

Relationship to Prior Technology

The VEW gasification process is based on the entrained flow principle of coal gasification. However, the VEW process is not an extension of a commercially available gasification process (i.e., Koppers-Totzek).

Operating Facilities

The VEW gasification process is being tested in a 9 million DM 1 tonne/hr pilot plant in Stockum, West Germany. The pilot plant gasifier which has been in operation since 1977 consists of a refractory lined vertical shaft of about 12 meters in length with a diameter of 0.5 meters. The plant does not have gas purification facilities. Operation of this facility with air and different coals thus far has proved satisfactory. VEW is all being developed by the company of Stein Miller, Gummersbach, which is responsible for the gasification plant. The company of Still, Recklingshausen, is responsible for the gas cleaning system.

In 1981 construction was completed on a 15 tonne/hour demonstration plant which cost 37 million DM. The plant also began operation in 1981. Future plans call for the construction of a 1.8 million tonne/yearly VEW combined cycle plant in 1983. This plant will be located at Gersteinwerke, Lippe and Emstand. Operation is scheduled for 1985.

Major Funding Agencies

The Federal Ministry of Research and Technology in West Germany is the major funding source of the VEW process. It does not appear that government support of this process will continue.

Technical Problems

None reported to date.

WEST GERMAN COAL GASIFICATION PROCESS

8. Bergbau-Forschung Nuclear Assisted Coal Gasification Process

Process Description

Bergbau Forschung GmbH (BF) is developing a nuclear heat assisted coal gasification [Prototypanlage Nukleare Prozesswärme (PNP)] process for the production of synthesis gas or SNG. Based on the study of coal kinetics it was determined that the heat produced from a high-temperature gas-cooled nuclear reactor (HTGR) is sufficiently high (950°C) to gasify coal. With the gasifier heat requirements being met by nuclear energy, a substantial reduction in oxygen requirements and coal could be accomplished.

The heat produced in the HTGR is transferred by a helium circuit at a temperature of 950°C. For safety reasons, the heat is transferred to a secondary helium loop which flows through the gas generator and the power plant. The gas generator which is shown in Figure 1, is a fluidized bed reactor with a submerged helium heat exchanger. The hot helium flows through this immersed heat exchanger to supply the heat of reaction. All gas loops and the gasifier reactor are operated at a pressure of 40 bars.

The gasifier itself is a horizontally mounted cylindrical pressure vessel. Feed coal below 0.5 mm is fed into the top of the reactor through lockhoppers. The bed is designed with a perforated trough where steam enters to provide fluidization. The hot (900°C) helium heat exchanger tubes drop from above into the fluidized bed. The coal as it is introduced into the gasifier moves longitudinally through the bed as it is gasified. The ash which accumulates at the other end as well as entrained solids which are removed overhead in a cyclone are removed through another set of lockhoppers.

In the pilot plant noncaking coals are fed to the free board while caking coals are fed directly into the fluidized bed. The difficulty of feeding into the bed is probably a reason for using this technique only when absolutely necessary. In large scale commercial operation a pretreatment step may be required for caking coals. A schematic of the pilot plant process is shown in Figure 2. A summary of this process is presented in the technology fact sheet of Table 1.

Process Goals

Conventional coal gasification processes consume 30 to 40% of the input coal for process energy needs. The remaining 60 to 70% is then available for

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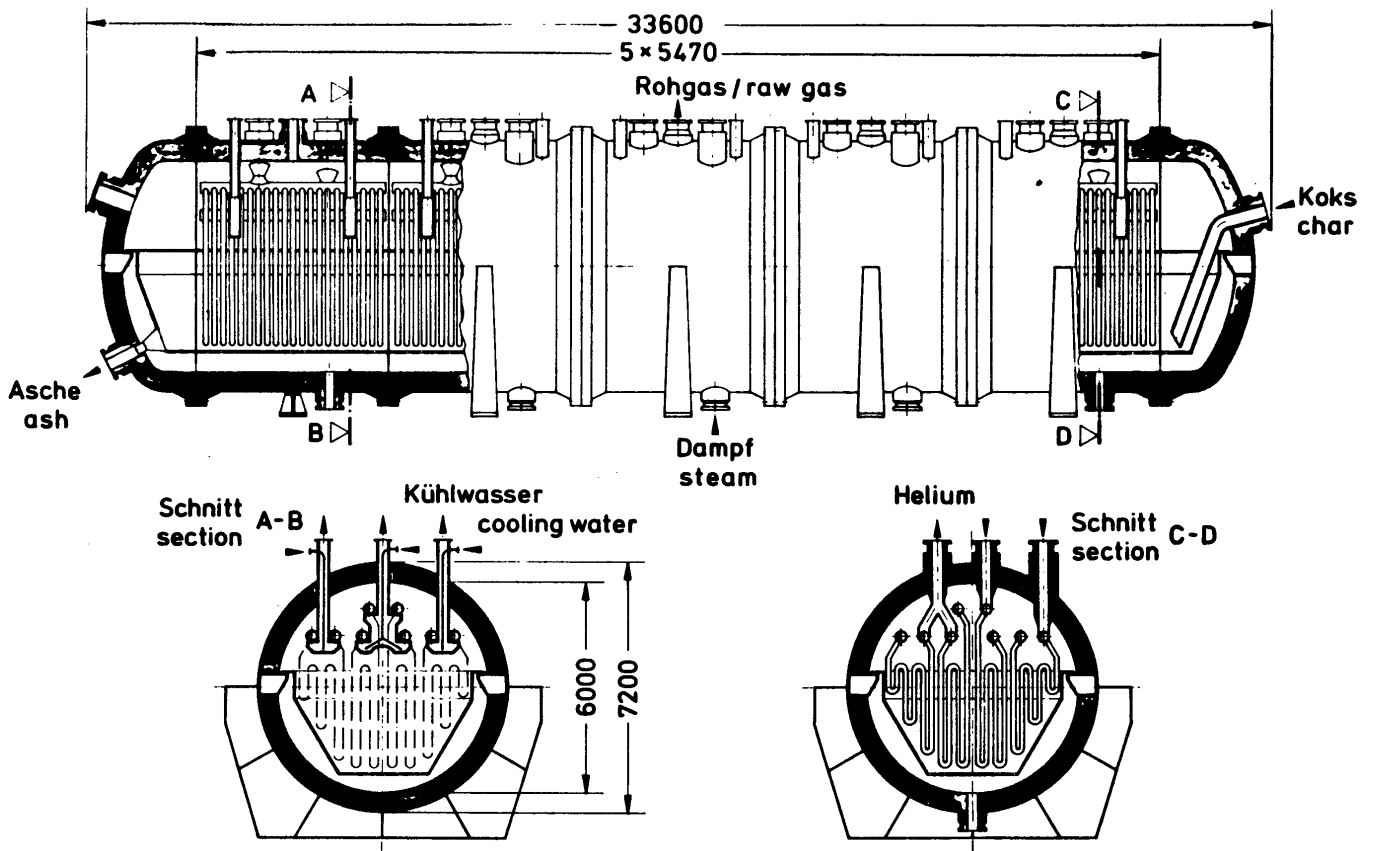


Figure 1. GAS GENERATOR

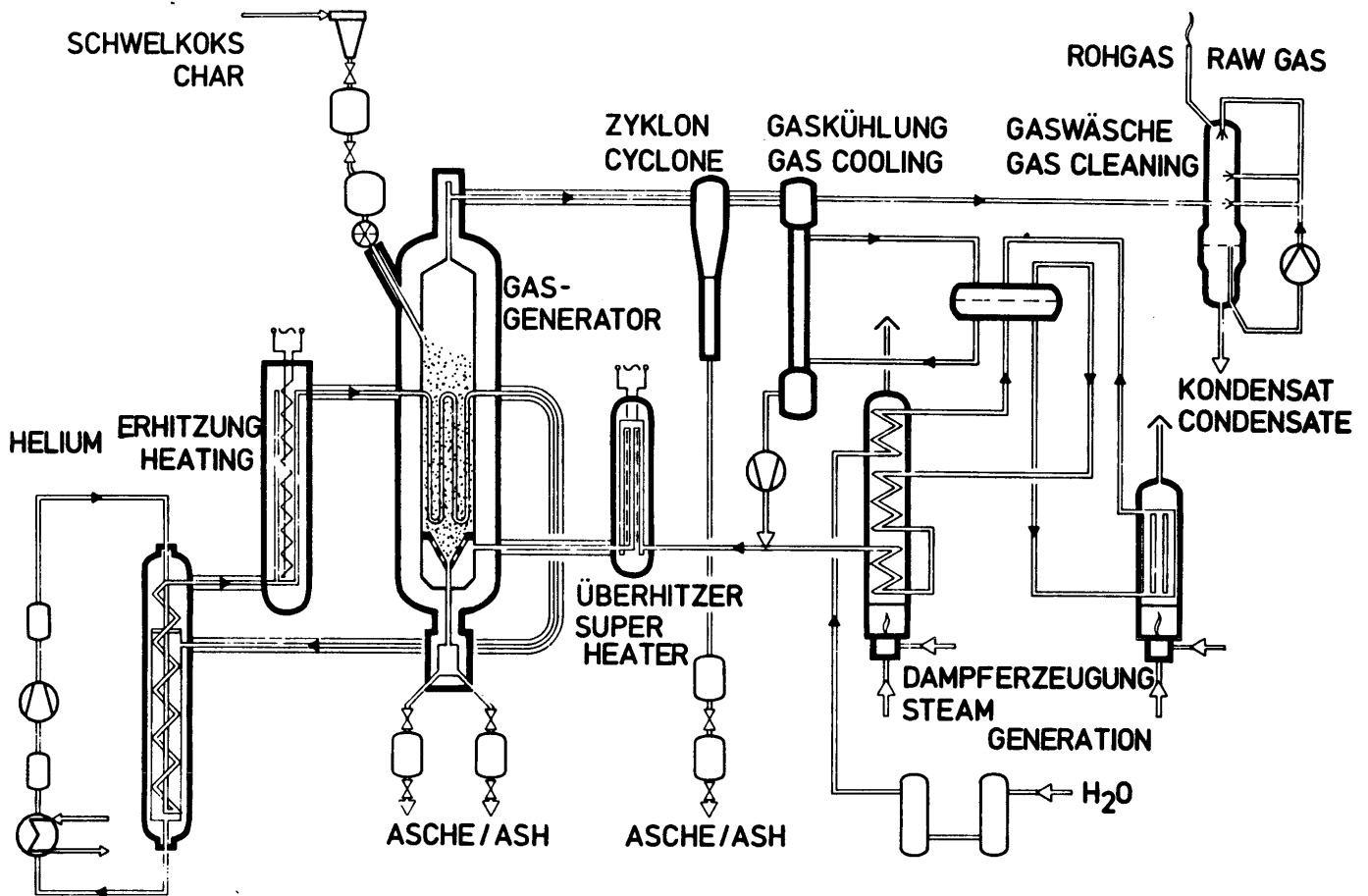


Figure 2. GASIFICATION PILOT PLANT

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Table 1 Part 2 TECHNOLOGY FACT SHEET: Bergbau-Forschung Nuclear Assisted Coal Gasification Process

NEW TECHNOLOGY (PROCESS DESCRIPTION AND GOALS): The Bergbau-Forschung process is being developed to use the energy from a high temperature gas cooled nuclear reactor to supply process heat to a steam coal gasification process.
The process utilizes a fluidized bed gasifier which has an internal heat exchange that circulates 900°C helium from the HTGR. The use of an outside heat source will lower oxygen demand and coal use as well as reduce gaseous pollutant emissions.

RELATIONSHIP TO PRIOR TECHNOLOGY (INCLUDING STATE OF DEVELOPMENT OF PRIOR TECHNOLOGY): _____
No relation to prior technology.

CHARACTERISTICS OF THE TECHNOLOGY:

<u>CHARACTERISTICS</u>	<u>NEW TECHNOLOGY</u>	<u>PRIOR TECHNOLOGY</u>
PRIMARY OUTPUT (DESIGN CASE).....syngas (vol.%).....	H ₂ =53%, CO=11%, CO ₂ =26%, CH ₄ =10%	
TYPE OF PROCESS.....	Nuclear assisted fluidized bed	
FEEDSTOCK REQUIREMENTS.....	Coal less than 0.5 mm	
OVERALL THERMAL EFFICIENCY (INCLUDING BY-PRODUCTS)..	95%	
CARBON CONVERSION EFFICIENCY.....	900°C	
OPERATING TEMPERATURE.....	40 bars	
OPERATING PRESSURE.....	None	
BY-PRODUCTS.....		

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Table 1 Part 1

TECHNOLOGY FACT SHEET: Bergbau-Forschung Nuclear Assisted Coal Gasification Process

STATUS OF DEVELOPMENT:

OPERATING FACILITIES - 450 kg/hr pilot plant at Bergbau Forschung Facility in Essen, West Germany.

MAJOR FUNDING AGENCY West German Federal Ministry of Research and Technology/The state of Nordrhein-Westfalia.

ANNUAL LEVEL OF FUNDING - 3.0 x 10⁶ DM/yr

TECHNICAL PROBLEMS: Ability to utilize caking coals without pretreatment in question. Long residence time in reactor will require large reactor size.

OTHER FACTORS AFFECTING OVERALL FEASIBILITY: Coupling of HTGR heat source with fluidized bed coal gasification process still unproven. Also, production of low cost process heat with an HTGR relative to coal still need analysis.

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conversion into gas. To overcome the use of feed coal to supply process energy, the goal of this development work is to use process heat from an HTGR as a source of process energy. This offers the following process advantage:

- Better use of coal supplies
- Higher process efficiency as well as nuclear cycle efficiency
- Decrease of gaseous pollutants per unit of gaseous energy produced
- Lower production cost if coal prices are regionally high and nuclear energy is inexpensive.

Relationship to Prior Technology

This technology represents a novel approach to coal gasification and does not have any relationship to prior technology.

Operating Facilities

Bergbau-Forschung has been developing this nuclear assisted coal gasification process since 1969. This research is being conducted at BF's Essen facilities. Besides coal gasification research, BF is also involved in coal liquefaction, production of active carbon, flue gas treatment, water purification, sewage treatment, gas separation by molecular sieves, and coke and electrode carbon production. The support research laboratories for coal gasification were predominately built in the last six years. These facilities include a high pressure thermogravimetric analyzer, a high pressure wire screen reactor, a curve-point pyrolysis reactor, a high-pressure hot-stage microscope, and an externally heat steam-char gasification PDU.

The development of the nuclear assisted coal gasification unit has proceeded in a step wise manner since 1969. The reaction kinetics of steam and hydrogasification of different coals and chars using particle sizes smaller than 2 mm has been investigated since 1969 in a fixed-bed differential sweep gas reactor at temperatures up to 1000°C, total pressures up to 70 bar ($70 \times 10^5 \text{ N/m}^2$), and various steam-hydrogen mixtures. A small-scale pilot plant has been operated since 1973 at 40 bar ($40 \times 10^5 \text{ N/m}^2$) using an internally heated fluidized bed. This unit processes up to 5 kg/h and gives result concerning reaction kinetics, gas composition, reaction heat, fluidized bed density, and heat transfer under quasi-realistic conditions.

In the mid-70's BF started construction of a 450 kg/hr pilot plant which started operation in 1979. The pilot plant gasifier is 0.9 m in inside

diameter and approximately 4 meters in height. The overall pilot plant structure is about 60 ft tall by 40 ft square. Since the beginning of pilot plant tests in 1979, they have logged over 7525 hours of coal gasification. The longest run lasted for 40 days in 1979. A typical raw gas is reported to contain $\text{CH}_4=14\%$, $\text{CO}=14\%$, $\text{H}_2=51\%$, and $\text{CO}_2=21\%$. BF claims 95% carbon conversion but with very long residence times (in excess of two hours). The steam distributor is made up of an incolloy 800 pipe with holes pointing downwards.

Major Funding Agencies

The nuclear assisted steam coal gasification process is being jointly developed by the following companies:

- Bergbau-Forschung GmbH, Essen, in cooperation with:
- Gesellschaft für Hochtemperaturreaktor-Technik GmbH
- Hochtemperatur-Reaktorbau GmbH
- Kernforschungsanlage Jülich GmbH
- Rheinische Braunkohlenwerke AG

The major funding agency for this project is the West German Federal Ministry of Research and Technology as well as the State of Nordrhein-Westfalia. The pilot plant was designed and built at a cost of $\text{DM } 13.5 \times 10^6$ and the current annual operating cost is $\text{DM } 9 \times 10^6$. The project manager is Dr. van Heek who also directs what is called the pyrolysis laboratories (including devolatilization and char gasification work) and the materials testing laboratory. The annual costs for these two support research activities are about $\text{DM } 2.0 \times 10^6$ and $\text{DM } 4.0 \times 10^6$, respectively. This research group has a total of 30 to 35 people out of the $\text{DM } 15 \times 10^6$ annual budget. The group spends nearly $\text{DM } 4.68 \times 10^6$ per year towards salaries and the rest towards equipment, materials, and supplies and utilities.

Technical Problems

The fluidized bed gasifier used in this process is very sensitive to the caking properties of the coal feedstock. In addition, the required long residence time in this reactor will require a large reactor size to attain the desired coal throughput. Finally, it has yet to be proven whether or not two different processes such as the HTGR and the fluidized bed coal gasification process can operate in harmony given their inherently different operating conditions.

WEST GERMAN COAL GASIFICATION

9. Rheinbraun Nuclear Assisted Lignite Hydrogasification Process

Process Description

The Rheinbraun process is being developed as part of the West German Prototype Plant Nuclear Process Heat Project (PNP). In this process, a fluidized bed coal gasifier is used to hydrogasify coal. Unlike conventional hydrogasification techniques that react a part of the coal input to produce the required hydrogen, the Rheinbraun process reforms part of the end-product methane to produce hydrogen. The heat for the endothermic reforming reaction is produced by a high temperature gas-cooled nuclear reactor (HTGR). A flow diagram of this process is shown in Figure 1.

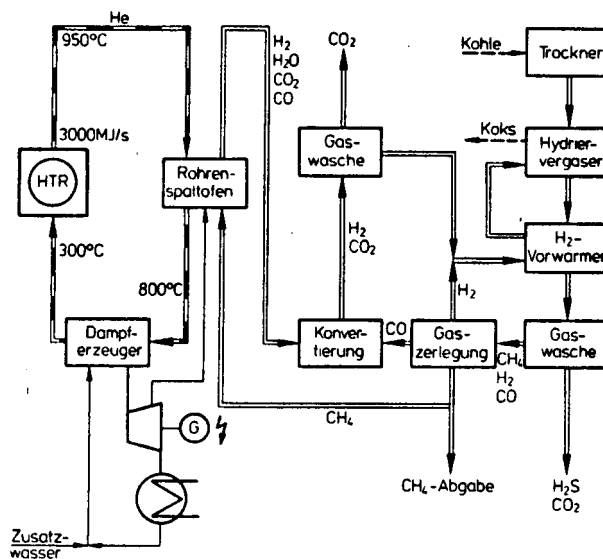


Figure 1. RHEINBRAUN HYDROGASIFICATION PROCESS

In the pilot plant arrangement shown in Figure 2, low sulfur coal is crushed and fed into the fluidized bed gasifier via a lockhopper. Prior to entering the gasifier the coal powder is also dried and pretreated to reduce caking properties. The hydrogasifier reactor operates at a pressure of 65 to 100 bars and a temperature of 1000°C with a coal throughput capacity of up to 400 kg/hr of raw brown coal. The gasifier consists of a steel pressure vessel with an alumina lining.

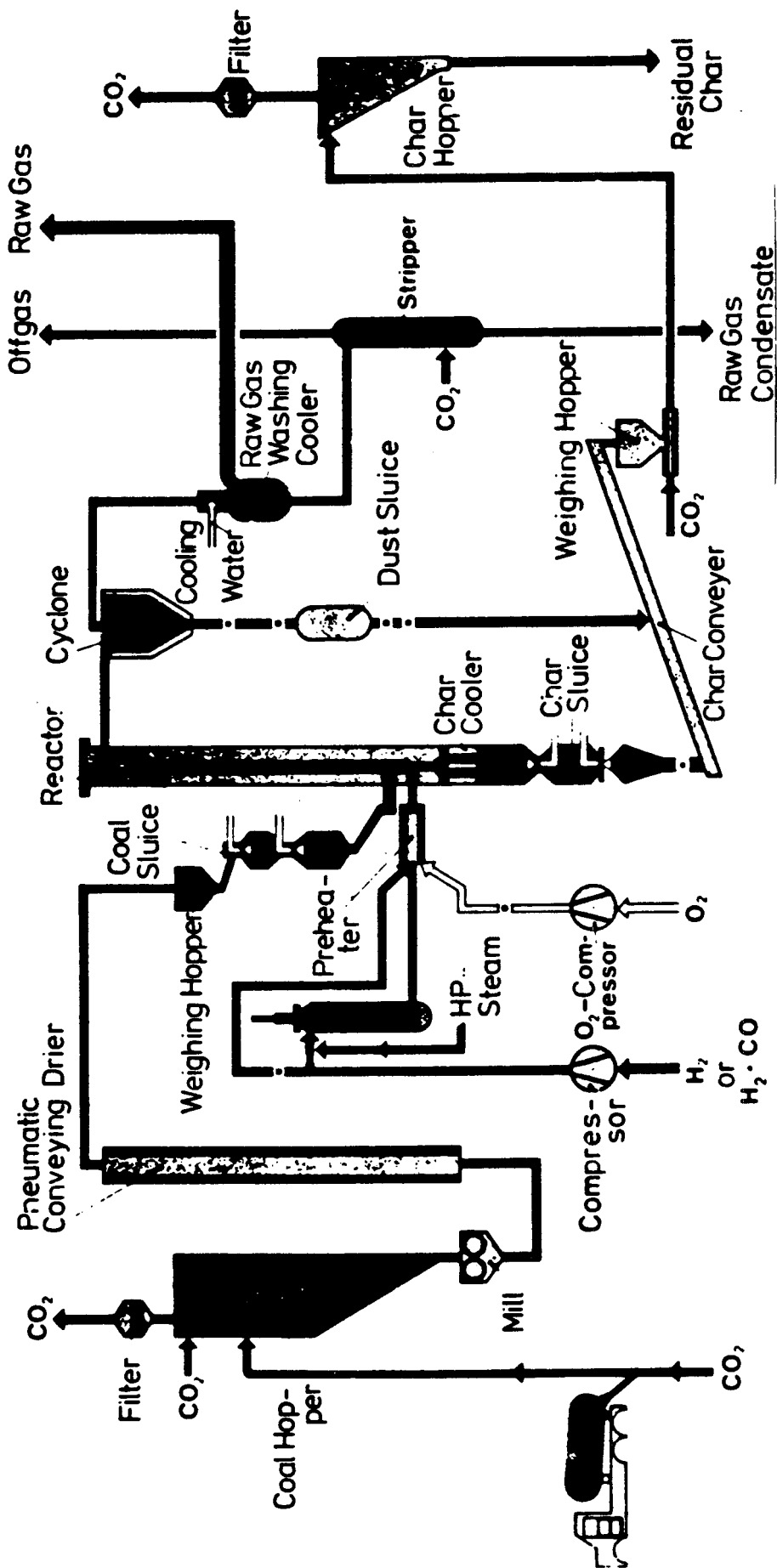


Figure 2. SEMI-TECHNICAL PLANT FOR HYDROGASIFICATION OF COAL

Because of the long residence time required for complete gasification, only a fraction of the coal is gasified. This fraction of gasified coal is in the range of 50 to 70% of the coal input. The resulting char is removed from the gasifier via lockhoppers and can be used as a boiler fuel to raise steam. Hydrogen for the hydrogasification reaction along with steam and oxygen are injected into the bottom of the gasifier. These reaction products also provide the fluidization in the bed. A synthesis gas with a methane content of 30 to 50% can be produced in this gasifier.

In a commercial operation, the hot raw gas exiting the gasifier is cooled and separated into its prime constituents. The carbon dioxide and hydrogen sulfide are scrubbed out and processed while the hydrogen is recycled back into the gasifier. Part of the captured methane exits the plant as product while the remainder is sent to a catalytic steam reformer. The heat required for the endothermic steam reforming reaction is supplied by an HTGR via heat exchange with a 900°C loop of circulating helium. This step produces the additional hydrogen required by the hydrogasification reaction. The resulting synthesis gas is then mixed with the carbon monoxide from the gasifier and reacted in a water gas shift reactor to produce additional hydrogen. This hydrogen is then sent to the gasifier and the carbon dioxide is scrubbed and released to the atmosphere.

The above described process can be modified by the addition of a High-Temperature Winkler gasifier. This gasifier would eliminate the use of nuclear heat by consuming the unreacted char from the hydrogasifier to produce the additional hydrogen required by the process. A technology fact sheet describing the HTGR coupled process is shown in Table 1.

Process Goals

The goal of this project is to produce SNG with an integrated HTGR and hydrogasification process. This offers the advantage of reducing gaseous pollutants and conserving coal reserves. This project will demonstrate the use of a novel gasifier, methane steam reformers, and integrated HTGR.

Relationship to Prior Technology

Although the concepts of each major component are not new, the designs and application of these concepts have not been demonstrated by any previous process.

Table 1 Part 2 TECHNOLOGY FACT SHEET: Rheinbraun Nuclear Assisted Lignite Hydrogasification Process

NEW TECHNOLOGY (PROCESS DESCRIPTION AND GOALS): The Rheinbraun Hydrogasification process is being developed

to prove the commercial viability of coupling an HTGR to a hydrogasification process to provide process heat.

Part of the SNG from the fluidized bed gasifier is routed to a methane steam reforming reactor where

hydrogen is produced from the hydrogasification process. Heat for the endothermic reforming reaction is

obtained via heat exchange from a 950°C helium loop from an HTGR.

RELATIONSHIP TO PRIOR TECHNOLOGY (INCLUDING STATE OF DEVELOPMENT OF PRIOR TECHNOLOGY):

This project is part of the West German Prototype Plant Nuclear Process Heat (PNP) Project. This process is not directly derived from any previous concept.

CHARACTERISTICS OF THE TECHNOLOGY:

CHARACTERISTICS	NEW TECHNOLOGY	PRIOR TECHNOLOGY
PRIMARY OUTPUT (DESIGN CASE).....	SNG	
TYPE OF PROCESS.....	nuclear assisted fluidized bed	nuclear assisted fluidized bed hydrogasification
FEEDSTOCK REQUIREMENTS.....	pulverized coal	pulverized coal
OVERALL THERMAL EFFICIENCY (INCLUDING BY-PRODUCTS)...	50 + 70%	
CARBON CONVERSION EFFICIENCY.....	65 + 100 bars	
OPERATING TEMPERATURE.....	1000°C	
OPERATING PRESSURE.....		
BY-PRODUCTS.....	char	

Table 1 Part 1 TECHNOLOGY FACT SHEET: Rheinbraun Nuclear Assisted Lignite Hydrogasification Process

STATUS OF DEVELOPMENT:

OPERATING FACILITIES - A 400 kg/hr process development unit has been in operation since 1975 at the Rheinbraun facility in Wesseling. A 25 ton/day unit is currently under construction as well as a 30 tube steam reformer.

MAJOR FUNDING AGENCY 75% of the funding comes from the West Germany Federal Ministry of Research and Technology

ANNUAL LEVEL OF FUNDING - Total Funding is 150 million DM

TECHNICAL PROBLEMS: Problem with fouling has occurred when operating on caking coals. The concept

has yet to be proven with heat from an HTGR.

OTHER FACTORS AFFECTING OVERALL FEASIBILITY: The economics of building an HTGR to supply heat at a lower unit cost than coal has not been proven.

Operating Facilities

Successful operation of this concept has been demonstrated at the 0.4 metric ton/hr process development unit operated by Rheinbraun since 1975 at their Wesseling facilities. The nuclear heat steam reformer concept has been tested since 1972 in a single tube design by Kernforschungsanlage (KFA) in Julich. Construction is underway of a one-tenth scale, 25 ton/day, pilot scale gasification plant and a 30 tube nuclear assisted methane steam reformer reactor.

Major Funding

The nuclear assisted lignite hydrogasification process is being developed by Union Rheinische Braunkohlenwerke AG (Rheinbraun) in association with the Gesellschaft fur Hochtemperaturreaktor — Technik mbH, Hochtemperatur — Reactorbau GmbH, Kernforschungsanlage Julich GmbH, and Bergbau-Forschung GmbH. Funding for this project is being sponsored by the Federal Ministry of Research and Technology (FMRT) as well as the State of Nordrhein-Westfalia. Total cost of the project is 150 million DM through the semi-commercial stage of development. Seventy-five percent of this cost is sponsored by the FMRT.

Technical Problems

Blockages occurred in the upper gasifier section when agglomerates formed during operation on caking coals. This problem can be overcome by the selective use of coals or the pretreatment of caking coals.

Capital Costs

The capital cost for a nuclear assisted lignite hydrogasification facility consuming 17.4×10^6 metric tons/yr of brown coal is shown in Table 2. This facility would cost 3,615 million DM in 1976 DM. This cost includes a 3000 MW HTGR to supply process heat.

Table 2. CAPITAL COST OF THE RHEINBRAUN INTEGRATED HTGR-HYDROGASIFICATION PROCESS (1976 Basis)

HTGR Thermal Output	3000 MW
Utilization Factor	7500 hr/yr
Coal Input	17.4×10^6 tonne/yr
Gas Output	2.96×10^9 m ³ /yr
Char Output	2.13×10^6 tonne/yr
Electricity Production	877×10^6 kWhr/yr
Project Cost	3,615 million DM
Coal Price	7 DM/G cal

WEST GERMAN COAL GASIFICATION PROCESS

10. KHD Kloeckner-Humbolt-Wedag Molten Iron Coal Gasification Process

Process Description

The Kloeckner process is based on the use of a molten iron bath for gasification. The gasifier is a refractory lined vessel equipped with gas cooled tuyeres. Crushed coal sized up to 3 mm is injected into the bath through the bottom tuyeres which are similar to the Q-BOP technology proven by the steel-making industry. The coal which has been dried to a moisture content of 1.5% is also mixed with lime of the same partical size before being pneumatically injected with nitrogen into the molten bath. Additional gasifying agents such as oxygen, air steam, or CO₂ can also be injected simultaneously into the bath through the special tuyeres. A diagram of the tuyeres design is shown in Figure 1.

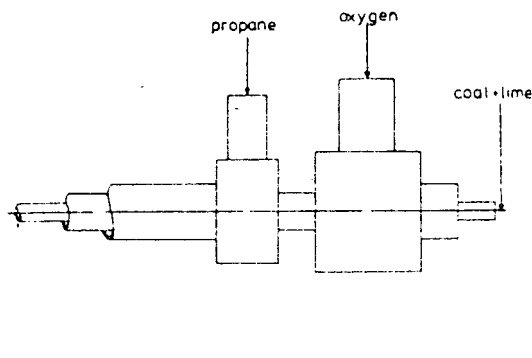


Figure 1. TRIPLE-FLOW TUYERES DESIGN

To protect these tuyeres which are made of a high grade steel, from the excess heat, a cooling gas is blown through the outermost annular gap. This gas can be propane, methane, carbon dioxide, steam or purified recycled synthesis gas. A simplified flow diagram of the Kloechner process is presented in Figure 2.

As the coal enters the 1350 to 1400°C iron bath it undergoes rapid devolatilization and cracking of the hydrocarbons. The carbon in the coal is dissolved in the bath and the coal ash rises to the bath surface. Any sulfur

in the coal reacts with the iron to form FeS. This FeS reacts further with the injected lime to form CaS. The calcium sulfide and ash are removed from the gasifier as liquid slag. The dissolved carbon in the iron bath reacts with the injected gasification agents such as oxygen and steam to produce a synthesis gas consisting of CO and H₂. The high process temperature produces a gas which is relatively free of carbon dioxide, volatiles, and sulfur compounds. When the gasifier is operated under continuous conditions the carbon content of the iron bath is approximately 3.5%. The carbon conversion efficiency of the process is 98% with a raw gas production of 2100 m³ per ton of coal. Table 1 shows the operating characteristics of the Kloechner pilot plant tests. The technology fact sheet for this process is shown in Table 2.

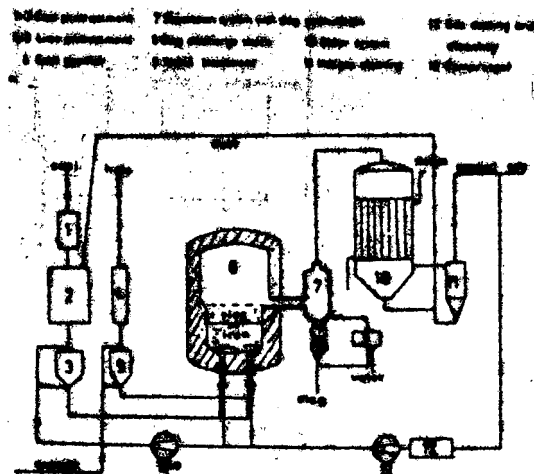


Figure 2. FLOW DIAGRAM OF THE KLOEKHNER MOLTEN IRON GASIFICATION PROCESS

Table 1. OPERATING CHARACTERISTICS OF THE KLOECKNER PROCESS

Coal	250 - 400 kg/hr • tonne Fe
Oxygen	0.58 m ³ /kg of coal
Propane	0.1 m ³ /m ³ O ₂
Transporting Gas	(N ₂) 0.1 m ³ /kg of solids (coal & lime)
Coal Composition:	Fixed carbon 67.50%; sulfur 1.0%; volatile matter 22.0%; ash 8.0%; moisture 1.5%, net heating value 7,5000 kcal/kg
Raw Gas Composition	CO 65 +; H ₂ 25-30%; CO ₂ > 0.3% S < 20 ppm; CH ₄ > 1%.

Process Goals

The objective of the Kloeckner-process is to produce a synthesis which is relatively gas free of unwanted components for use in the chemical industry or as a combined cycle fuel. This is accomplished by the use of a molten iron gasifier equipped with specially designed tuyeres similar to those used in the Q-BOP technology of the steel making industry. This process also offers the stated advantage of being economical due to the high gasifier throughput and the dramatic reduction of gas cleanup facilities.

Relationship to Prior Technology

The concept of molten iron gasification was first patented and tested by the Applied Technology Corporation (USA) in the early seventies. However, the Applied Technology Corp. concept introduced the coal and oxygen feedstock through lances from the top into the molten iron bed. Cooling, material problems, and mechanical stability of these lances made this approach technically unfeasible. In 1978 Kloeckner acquired the patent rights and modified the process to use the bottom tuyeres feed arrangement.

Operating Facilities

A 2.4 tonne/hr process development unit with an iron bath capacity of 6 tonnes has been tested at the Maxhutte Research facilities in Bavaria (West Germany). A larger 10 tonne/hr facility is scheduled to begin operation at the end of 1982 in the Ruhr region. Testing in this 20,000 m³/hr capacity

Table 2 Part 1 TECHNOLOGY FACT SHEET: KHD Kloeckner-Humbolt-Wedag Molten Iron Coal Gasification Process

NEW TECHNOLOGY (PROCESS DESCRIPTION AND GOALS): The Kloeckner process is based on the use of a molten iron bath for gasification. The gasifier is a refractory lined vessel equipped with gas cooled tuyeres for coal steam and oxydent injection. The objective of this process is to produce a synthesis gas which is relatively clean for use in the chemical industry or as a combined cycle fuel. The production of a clean fuel will greatly reduce the cost of gas cleanup facilities.

RELATIONSHIP TO PRIOR TECHNOLOGY (INCLUDING STATE OF DEVELOPMENT OF PRIOR TECHNOLOGY):
 The concept of molten iron gasification was first patented and tested by the Applied Technology Corp. in the early 70's. Due to materials and design problems this process was not commercialized. In 1978 Kloeckner

CHARACTERISTICS OF THE TECHNOLOGY:

CHARACTERISTICS	NEW TECHNOLOGY	PRIOR TECHNOLOGY
PRIMARY OUTPUT (DESIGN CASE).....synthesis gas.....	CO = 65 + 70%, H ₂ = 25-30%, CO ₂ < 0.3%, CH ₄ < 0.1% S < 20 ppm	
TYPE OF PROCESS.....	molten iron bath	
FEEDSTOCK REQUIREMENTS.....	3 mm sized coal (all types)	
OVERALL THERMAL EFFICIENCY (INCLUDING BY-PRODUCTS)...		
CARBON CONVERSION EFFICIENCY.....	98%	
OPERATING TEMPERATURE.....	1350 + 1400 °C	
OPERATING PRESSURE.....	10 bars	
BY-PRODUCTS.....	none	

Table 1 Part 2 **TECHNOLOGY FACT SHEET:** KHD Kloeckner-Humbolt-Wedag Molten
Iron Coal Gasification Process

STATUS OF DEVELOPMENT:

OPERATING FACILITIES - A 2.4 tonne/hr (coal) PDU has been tested at the Maxhitte Research facility
in Bavaria. A larger 10 tonne/hr (coal) pilot plant is scheduled to begin
operation in 1982 in the Ruhr region.

MAJOR FUNDING AGENCY State of Nordrhein Westfalia

ANNUAL LEVEL OF FUNDING - \$15 million U.S. funding for 10 tonne/hr (coal) pilot plant

TECHNICAL PROBLEMS: None

OTHER FACTORS AFFECTING OVERALL FEASIBILITY:

gasifier will end in 1984. This facility is capable of operating at 10 bars and at a temperature of 1350 to 1400°C.

Major Funding Agencies

The initial process development unit testing was funded by the Maxhutte Research Institute. The 15 million U.S. \$ funding for the 10 tonne/hr pilot plant is being sponsored by the State of Nordrhein Westfalia. Future plans call for the construction of a molten iron coal gasification commercial size plant in 1985 in Bremen, West Germany. This plant will have a 200 tonne molten iron reactor with a rated capacity of 200,000 m³/hr of synthesis gas. The Federal Ministry of Research and Technology will fund 40% of the 750 million D.M. construction cost.

Technical Problems

Much of this or similar technology has been commercially proven in the steel industry and as such no major technical problems have been reported.

Capital Costs

The capital costs for a direct reduction, iron ore facility incorporating a Kloeckner molten iron gasification process has been published by KHD Humbolt Wedag AG. Figure 3 presents the layout of the iron ore reduction/gasification system. The operating and cost specifications for this plant are shown in Table 3. The gasification plant investment cost of 60 million D.M. was assumed to be on a 1980 basis. However, no basis was presented in the literature. Based on varying coal costs and the information presented in Table 2, Kloeckner has calculated the cost of producing reduction gas. These costs are presented in Figure 4, and vary from a gas cost of approximately 4¢/m³ at a coal cost of \$25/tonne to a gas cost of approximately 8.74¢/m³ for a coal cost of \$125/tonne.

Table 3. OPERATING AND COST DATA FOR A KLOECKNER MOLTEN IRON GASIFICATION PLANT CONNECTED TO A IRON ORE REDUCTION FACILITY

Plant Capacity	450000 t/a Fe
Reducing Gas Demand	1500 m ³ N/t by coal gasification -750 m ³ N/t by gas recycling
Operating Time	8000 h/a
Gasification Plant Investment	60 mil. DM
Capital Change	14 %/yr

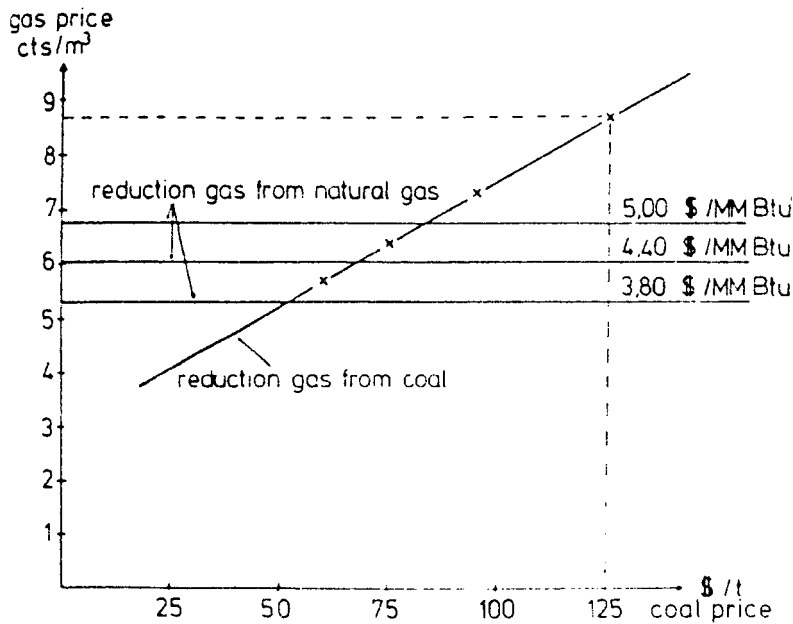


Figure 3. REDUCTION GAS COSTS USING THE KLOECKNER PROCESS

WEST GERMAN COAL LIQUEFACTION PROCESS

11. Ruhrkohle/Veba Oil Catalytic Hydrogenation Coal Liquefaction ProcessProcess Description

This catalytic hydrogenation process is a modification of the Bergius-Pier (IG Farben process) hydrogenation technology which was a commercial reality in Germany prior to 1945. Based on this working experience which was documented by BASF, Ruhrkohle (RAG) and Veba Oil in cooperation with KRONIG made the following modifications to the IG process:

- A mixture of heavy oil and middle distillate oil from the process is used as a solvent
- Separation of solids and asphaltenes is accomplished by distillation
- The residual material containing solids and asphaltenes is used to produce hydrogen.

These changes are expected to improve the IG process as follows:

- Reduce the process pressure from 700 bars to 300 bars
- Raise the specific coal throughput by 50%
- Improve heat recovery
- Raise thermal efficiency by 25%
- Reduce process capital costs.

Based on a renewed interest in the IG process by RAG and Veba Oil in 1974, a process development unit was designed in 1975 and completed in 1976 by Bergbau-Forschung in its Essen Laboratory. Test runs are reported to last a minimum of 4 days with an estimated total of over 21,000 hours operating experience gained since startup.

The process development unit has two 11 liter reactors, designed for 400 atm and 500°C with a rate coal throughput of 250 kg/day. About 600 m³/day of hydrogen is supplied from a huge bank of (about 200) 140 psig cylinders. The recycle gas flow rate is about 1000 m³/day. A wide variety of coals are tested in the pilot plant with carbon conversions ranging up to 95% and hydrogen consumption estimated at about 6% of wt. by d.a.f. coal.

The PDU involves mixing ground coal (less than 0.2 mm) with recycled oil, and a disposable catalyst (red mud) of 2% to 3% by weight of coal, resulting

in a 45% solids content slurry. The slurry is pressurized in stages (using Moyno pumps to 6-8 atm and high pressure piston pumps to 300 atm), with high pressure hydrogen, preheated to 400 to 450°C, and transferred to the hydrogenation reactor. The isothermal reactor contains an agitator oscillating axially. The resulting products are a mixture of oils of a wide boiling range, gases, water and residual char. These are separated in a series of vacuum flash distillation columns to produce ash and sulfur-free distillate (50% by weight of d.a.f. coal), byproduct gases (20%), sour water (10%), and residual char. Part of the distillate oil and hydrogen, separated by scrubbing the product gases, are recycled to the front end. BF observed that some H₂ in the recycled hydrogen aids liquefaction, possibly by activating the catalyst by sulfurization. However, use of pyrite in catalyst resulted in scale deposition in the slurry preheater.

A typical catalyst composition is given at 30% Fe₂O₃, 40% Al₂O₃, 2-6% TiO₂, and the rest silica and other inerts.

In November 1977, RAG and Veba Oil decided to jointly sponsor the construction of a 200 tonne/day pilot plant in Bottrop close to RAG's "Prosper" coking plant which supplies some of the support facilities. The flow diagram of this 200 tonne/day pilot plant, which is similar to the PDU operated by BF, is shown in Figure 1. Construction of the pilot plant started in 1979. The plant output will be processed in a oil refining pilot plant at Veba Oil's Scholven facility nearby. The fresh hydrogen required by the process will be obtained from a nearby hydrogen pipeline operated by Chemische Werke Huls AG.

In the pilot plant operation coal is delivered by train or truck and stored in the raw coal bins. From here the coal is sent to the grinding/drying mill where the coal is reduced to a grain size of less than 1 mm and a residue moisture of 0.5% by weight. Any airborne coal dust is recovered by a cyclone and electrostatic filter. All dried and sized coal is sent to the dry coal storage bin at a temperature of 120°F. The catalyst (red mud) is also stored in bins before it undergoes drying and sizing.

From the dry coal bins the coal is sent to the mixing section where it is admixed with the solvent, which is a 40 to 60 weight percent ratio of medium oil (boiling temperature range 200 - 325°C) and heavy oil (boiling temperature greater than 325°C) which is recycled from the process. At this point the coal is wet ground with the catalyst to less than 0.2 mm size. This slurry, which

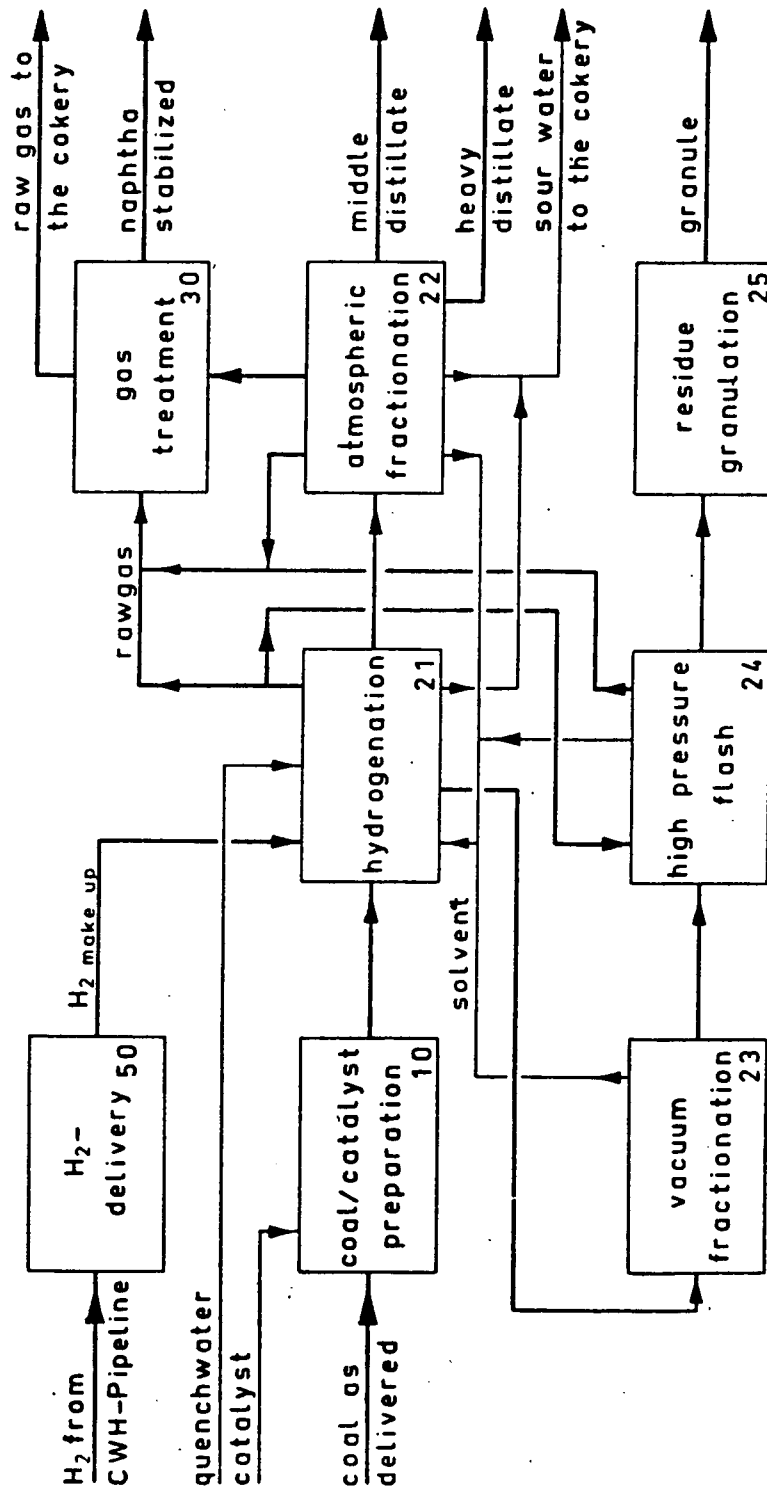


Figure 1. BLOCK FLOW DIAGRAM OF THE RAG/VEBA OIL PILOT PLANT AT BOTTROP

contains 40 weight percent solids, is pumped to a pressure of 300 bars and mixed with recycle gas, make up hydrogen, and recycled reacted slurry. This mixture is then heated to 380°C by heat exchange and then to the 425°C reactor inlet temperature by a gas fired heater.

The hydrogenation reactions are performed in a series of three reactors. The slurry is introduced in the topmost reactor and flows downward through the remaining two reactors. Due to the exothermic nature of these reactions the reactors are maintained at 475°C through the injection of cooled recycle gas at various levels of the reactors.

Upon leaving the last reactor at 450°C, the slurry gas mixture enters a separator where hot gases and vapors are drawn off the top and liquids and solids are removed from the bottom. The gaseous and vapor section is cooled by exchange with the incoming feed and then fed to the distillation section where naphtha, middle oil, and heavy oil are extracted. The resulting gas phase is oil washed to remove gaseous hydrocarbons, H₂S, CO₂, CO, and N₂. The remaining process gas is partially recycled to the feed/product heat exchanger and the rest is sent to the flash evaporation unit or sold as product to the cokery.

The liquid and solids separator bottoms are flashed to separate the gas and vapor before it is sent to the vacuum distillation unit. The slurry contains about 50 weight percent solids after distillation. If this weight percent ratio cannot be maintained the slurry is heated and compressed to 50 to 100 bars with hydrogen and further processed in a high pressure flash unit. The residue is sent to a granulation units where it is shaped into tablets for further processing outside the plant. The excess oil which is not recycled is then sent to a refinery for further processing. The feed and projected product slate for this pilot plant are shown in Table 1. The technology fact sheet for this process is presented in Table 2. The refined product specifications after coal-oil processing are shown in Table 3.

Process Goals

The objective of this process is to improve the efficiency, operating characteristics, and product slate of the old IG Farben Process. New equipment which will be tested in the pilot plant includes the slurry feed piston pumps, feed/product preheaters, reactors, bottoms separation flash units, and process oil upgrading.

Table 1. FEED AND PROJECTED PRODUCTS OF BOTTROP PILOT PLANT

Feed		
Coal, m.a.f.	t/d	200
Hydrogen H ₂ , makeup	m ³ /d (ft ³ /d)	220,000 (7.8x10 ⁶)
Process water	t/d	41.2
Catalyst (Fe ₂ O ₃)	t/d	4.0
Power	kWh/d	108,000
Projected Products		
Gas	t/d	61.3
Naphtha (stab.) I.B.P. 200°C (390°F)	t/d	24.4
Middle oil 200-325°C (390-620°F)	t/d	69.0-74.7
Heavy oil >325°C (620°F)	t/d	-
Residue	t/d	68.6-74.3

Table 2 Part 1 TECHNOLOGY FACT SHEET: Ruhrkohle/Veba Oil Catalytic Hydrogenation Coal Liquefaction Process

NEW TECHNOLOGY (PROCESS DESCRIPTION AND GOALS): The Ruhrkohle/Veba Oil Process is a Catalytic hydrogenation process which uses an inexpensive iron catalyst. Coal is ground and mixed with recycled solvent and catalyst

before entering the hydrogenation reactors. The hydrogenation step consists of three reactors where

liquefaction occurs at 100 bars and 475°C. Liquefied products are separated by a combination of flash evaporation and multistage distillation. Goals include reduced operating pressure, increased coal throughput and thermal efficiency and a reduction in plant cost relative to the 1st generation IG Farben process.

RELATIONSHIP TO PRIOR TECHNOLOGY (INCLUDING STATE OF DEVELOPMENT OF PRIOR TECHNOLOGY):

This process is a modified version of the IG Farben process which was in commercial operation from 1936 to 1945 in Germany.

CHARACTERISTICS OF THE TECHNOLOGY:

<u>CHARACTERISTICS</u>	<u>NEW TECHNOLOGY</u>	<u>PRIOR TECHNOLOGY</u>
PRIMARY OUTPUT (DESIGN CASE).....	LPG, gasoline, fuel oils	LPG, gasoline, fuel oils
TYPE OF PROCESS.....	Catalytic Hydrogenation	Catalytic Hydrogenation
FEEDSTOCK REQUIREMENTS.....	Coal, catalyst, hydrogen	Coal, catalyst hydrogen
OVERALL THERMAL EFFICIENCY (INCLUDING BY-PRODUCTS)..	55%	30 → 40%
CARBON CONVERSION EFFICIENCY.....		
OPERATING TEMPERATURE.....	475°C	550°C
OPERATING PRESSURE.....	300 bars	700 bars
BY-PRODUCTS.....	residual bottoms	residual bottoms

Table 2 Part 2 **TECHNOLOGY FACT SHEET:** Ruhrkohle/Veba Oil Catalytic Hydrogenation Coal Liquefaction Process

STATUS OF DEVELOPMENT:

OPERATING FACILITIES - Bergbau Forschung has operated a 250 kg/day PDU since 1976 at its Easin facility. A 200 tonne/day pilot plant operated in a 60/40 joint

venture of Ruhrkohle and Veba Oil started operation at Bottrop in 1981.

The Ministry of Economics of the State of Nordrhein-Westfalia.

MAJOR FUNDING AGENCY

ANNUAL LEVEL OF FUNDING - BF receives 3 million DM on an annual basis and the Bottrop plant will receive 200 million DM for its three years of testing.

TECHNICAL PROBLEMS: No technical problems have been reported. However, a number of new approaches will be tested in the pilot plant stage. These include slurry feed pump design, feed preheaters, reactor vessel design, slurry separation and coal-oil upgrading techniques.

OTHER FACTORS AFFECTING OVERALL FEASIBILITY: Both Veba Oil and Ruhrkohle have published studies that indicate that this process is not economically competitive in today's market.

Table 3. SPECIFICATIONS OF CRUDE AND REFINED COAL-OIL FRACTIONS

Specifications	Light oil		Middle oil	
	Crude	Refined	Crude	Refined
Gravity at 15°C g/cm ³	0.865	0.827	0.990	0.993
Sp. g. (59°F)	0.865	0.827	0.990	0.993
Aromat. C, wt %	46	31.6	64	50.0
Naphth. C, wt %	10	27	28	31.3
Paraff. C, wt %	43	41.4	28	18.7
Carbon, wt %	85.25	87.80	87.40	88.5
Hydrogen, wt %	11.15	12.25	9.10	10.75
Sulfur, wt %	146 ppm	2 ppm	0.60	0.23
Nitrogen, wt %	0.24	2 ppm	0.60	0.23
Oxygen, wt %	3.5	0.1	3.0	0.4
Base value, mg NH ₃ /l g/cc)	1100 1.1x10 ⁻³	3 3x10 ⁻⁶	8800 8.8x10 ⁻³	2900 2.99x10 ⁻³
CFPP, °C (°F)			-26 (-15)	-47 (-53)
Flash point, °C (°F)			93	67
Viscosity, mm ² /S Centipoise			3.1 3.1	2.1(50°C)(12°F) 2.1
Heating value, kJ/kg Btu/lb	41,000 17,625	43,000 8,485	38,500 16,550	40,900 17,585
Fuel value, kJ/kg Btu/lb	43,000 1,455	46,000 19,775	40,500 17,410	43,250 18,595
Phenols, wt %	15.4		16.2	0.29
Bases, wt %	0.25		6.5	2.32
Benzene, wt %		3.9		
Toluene, wt %		5.3		
Ethyl benzene, wt %		2.0		
Xylene, wt %		4.4		
ROZ (clear)		79.2		
Boiling range				
Initial boiling point, °C(°F)	76 (169)	43 (109)	212 (414)	169 (336)
10 vol %, °C(°F)	102 (216)	81 (178)	225 (437)	205 (401)
50 vol %, °C(°F)	158 (316)	149(300)	253 (487)	244 (471)
End point, °C(°F)	206 (403)	211(412)	324 (615)	303 (579)

Relationship to Prior Technology

The Ruhrkohle/Veba Oil Process is based on the Bergius Pier Process which was further developed into the IG Farben process in the 1930's. The first IG Farben Process plants started in 1936 and 1939 with an output capacity of 680,000 tons per year of gasoline and middle distillates. Production capacity using this process reached 4 million tons/year before complete shutdown in 1945.

Operating Facilities

In 1974 RAG and its subsidiary STEAG AG started evaluating coal liquefaction technologies with the goal of selecting the best process for further development. In the same year Veba-Chemie AG began a research program which involved the hydrogenation of coal in a continuously operating bench-scale autoclave unit. A larger process development unit with a capacity of 250 kg/day was designed in 1975 and completed in 1976 by Bergbau Forschung at its Essen facility. In 1977 Ruhrkohle and Veba Oil formed a joint venture to construct a 200 tonne/day pilot plant in Bottrop. The design and construction of this plant, which was completed in 1981, was based on the operating data collection by BF.

Major Funding Agencies

Early work by both Ruhrkohle and Veba was sponsored by the Ministry of Economics of the State of Nordrhein-Westfalia. The Bergbau Forschung process development unit was completed for a total cost of DM 3 X 10⁶. The operating costs for this PDU have been around DM 3 X 10⁶ per year. This research was also partially funded by the State of Nordrhein-Westfalia. The construction investment cost for the 200 tonne/day Bottrop pilot plant amounts to nearly 200 million DM. The operating cost for this plant is expected to cost another 200 million DM during the three-year demonstration testing period. The State of Nordrhein-Westfalia sponsored 90% of the construction phase costs and will sponsor approximately 80% of the operating phase costs. A decision to proceed with a larger demonstration size plant will be made by the West German Ministry of Science and Technology by the end of 1982.

Technical Problems

Although no technical problems have been reported, a number of new approaches being tested in the pilot plant still need to be proven. These new

technologies are the slurry feed pumps, preheaters, reactors, vacuum distillation units, and upgrading coal-oil equipment.

The slurry pumps will be driven by SCR controlled AC motors and operated in parallel. For test purposes the pumps will be operated in the piston vertical and horizontal position. These pumps will be equipped with remote valve boxes with inlet and outlet valves of different design and materials to test for performance in regard to corrosion and erosion. The external valve box will also provide for abrasion free action, although this will result in oil loss which will be made up by clean oil on each stroke.

The preheater section will consist of two different units that are each rated to handle the full plant capacity. The first heater is a shell and tube design which is similar to the ones used in the IG Farben process until 1945. At the expense of size and cost this heater provides for gradual feed heating to prevent coking. The second preheater utilizes a radiant furnace design which is frequently used in oil refineries. The higher heat transfer characteristics of this design increase the possibility of coking but decreases the unit size and cost.

The next new area to be tested at the Bottrop plant is reactor construction. The old IG Farben process coal hydrogenation reactors were made from forgings whereas the new reactors will be of a new multilayer design. Insulation will be accomplished using a layer of refractory brick which will be protected from abrasion by a stainless steel liner. A new type of reactor is in the design phase which will utilize the multilayer concept but the required insulation will become an integral part of the multilayer wall.

The fourth area of testing will focus on slurry bottoms separation using a combination of flash evaporation and vacuum distillation. These methods will be employed to maximize volatile matter separation from the hot bottom slurry which contains 65 to 75% oil.

The last area of investigation will determine the best upgrading approach for the coal derived oil. The raw oil from the coal liquefaction process varies from mineral oil in many respects. These include the distribution of boiling range fractions, sulfur, nitrogen and oxygen contents, aromatic and density properties. A comparison of the coal-oil properties to "Arabian light" as shown in Table 4.

Table 4. COMPARISON OF SOME CHARACTERISTIC PROPERTIES OF OIL FROM COAL AND ARABIAN LIGHT

		<u>oil from coal</u>	<u>Arabian light</u>
spec. gravity at 15°C	g/ml	0.950	0.856
API gravity		17.5	34
carbon	% wt	86.6	85.5
hydrogen	% wt	9.05	12.6
sulfur	% wt	0.1	1.7
nitrogen	% wt	0.75	0.2
oxygen	% wt	3.50	--
H/C ratio		1.26	1.77
gasoline, ibp-200 °C	% wt	22	23
middle distillate, 200-325°C	% wt	70	23
vacuum gasoil, 325-500°C	% wt	8	28
vacuum residue, 500 °C	% wt	-	25

Capital Costs

Veba Oil has published the cost of a liquefaction plant capable of both liquefying coal and for processing heavy refinery oil residues into light oil products such as gasoline. The plant would have a coal capacity of 3.7 million tonnes per year with a rated output of 1.95 million tonnes per year of liquid product. The overall efficiency would probably be nearly 55%. The liquid product would consist of 250,000 tonnes per year of LPG, 850,000 tonnes per year of gasoline, and 850,000 tonnes per year of heating oil. According to a Veba study, this plant would require a capital investment of 6 billion DM on a 1981 basis. Design and construction of this facility would take eight years which with interest charge, would bring the final plant costs to 7.8 billion DM.

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WEST GERMAN COAL LIQUEFACTION PROCESS

12. Saarberg Catalytic Hydrogenation Coal Liquefaction Process

Process Description

The Saarberg coal liquefaction process is a modification of the commercially proven IG Farben technology. The main differences are a lower pressure, reduced hydrogen consumption, improved heat transfer techniques and more efficient products separation equipment. A process flow diagram of the 6 tonne/day pilot plant operated by Saarbergwerke AG in Volkingen-Furstenhausen, West Germany is shown in Figure 1.

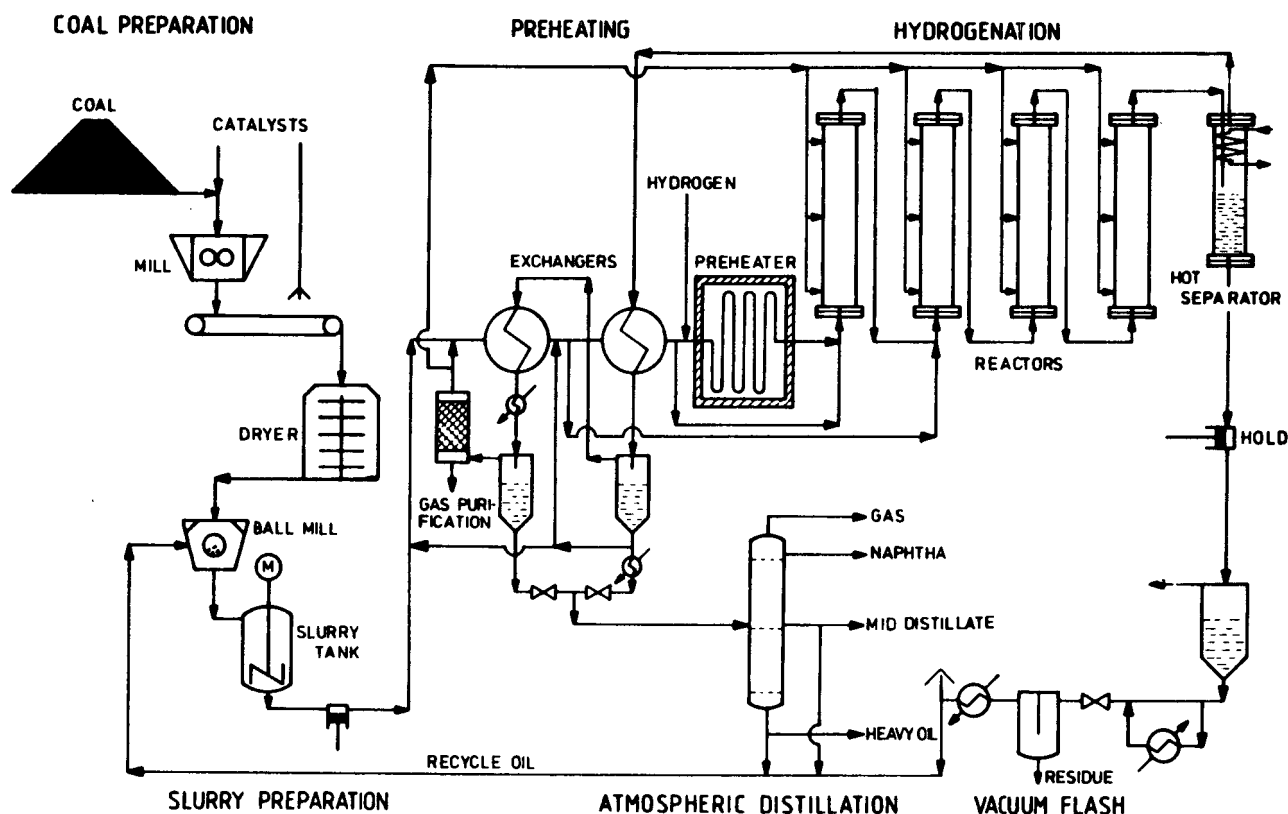


Figure 1. FLOW DIAGRAM OF THE 6 TONNE/HR SAARBERG PILOT PLANT

In this pilot plant the coal is first crushed and dried to a particle size of less than 3 mm before it is mixed into a slurry. Only high quality coal with an ash content of less than 15% can be used in this process. The dried coal powder is then fed into a ball mill where it is wet ground to a maximum particle size of 0.1 mm with recycled solvent, and catalyst. The catalyst consists of a mixture of iron based sulfates (FeSO_4), red mud (Fe_2O_3), and sodium sulfide (Na_2S). The catalyst cost was quoted as approximately 5¢/lb

in 1981 dollars. The catalyst mixture composition and quantity depend on the hydrogen pressure used in the process as well as the hydrogenation reactivity of the coal feedstock. With some coals that have a sufficient autocatalytic effect, the additional amount of catalyst that must be added may be negligible. This may be true of many non-German coals that have a high pyritic sulfur content. This feature makes the Saarberg coal liquefaction process flexible since the optimal hydrogenation conditions can be adjusted for by changes in the catalyst composition and process pressure. The slurry paste contains approximately 60 weight percent coal, FeSO_4 (1.21% of maf coal), red mud (2%), sodium sulfide (1%) and the remainder is chiefly distillate oil which is recycled from the vacuum distillate section.

This thick slurry paste is then sent to a high pressure injection pump before it is mixed with hydrogen and heated by a unique preheating section. The preheat section is designed to heat the paste by injecting hot condensed oil, which is obtained from the hydrocarbon vapors leaving the top of the hot separator feed section. The hot diluent oil, which was collected in an intermediate catchpot before injection into the preheat section, helps to heat the slurry and reduced the coal content to 50 weight percent. The paste is further diluted and heated by a second hot oil injection preheat section. By this means much of the exothermic heat generated in the hydrogenation section can be captured in the preheat injection oil and recycled in the process. The slurry which leaves the preheat section has a temperature of 400°C and a coal weight of 38%.

The initial boiling point of the diluent intermediate entering the preheat section is above 200°C . The remaining oil vapors which enter the preheat section and are condensed in heat exchange with the slurry has a boiling point of about 400°C . This internal hot oil recycle is about the same weight as the incoming fresh coal and helps to eliminate the need for atmospheric distillation of the oil fraction. One benefit of this method is that the oil recycle fraction is kept at a high temperature and pressure. Although this method reduces heat exchanger requirements, a bundle heat exchanger is still necessary for slurry preheat. A diagram of this technique is shown in Figure 2.

Further attempts to simplify the process led to another idea which eliminates the heat exchangers needed for preheating the slurry. In this method the coal slurry is mixed directly with the overhead vapors from the hot separator. For this purpose the coal slurry is passed to a mixing zone which is

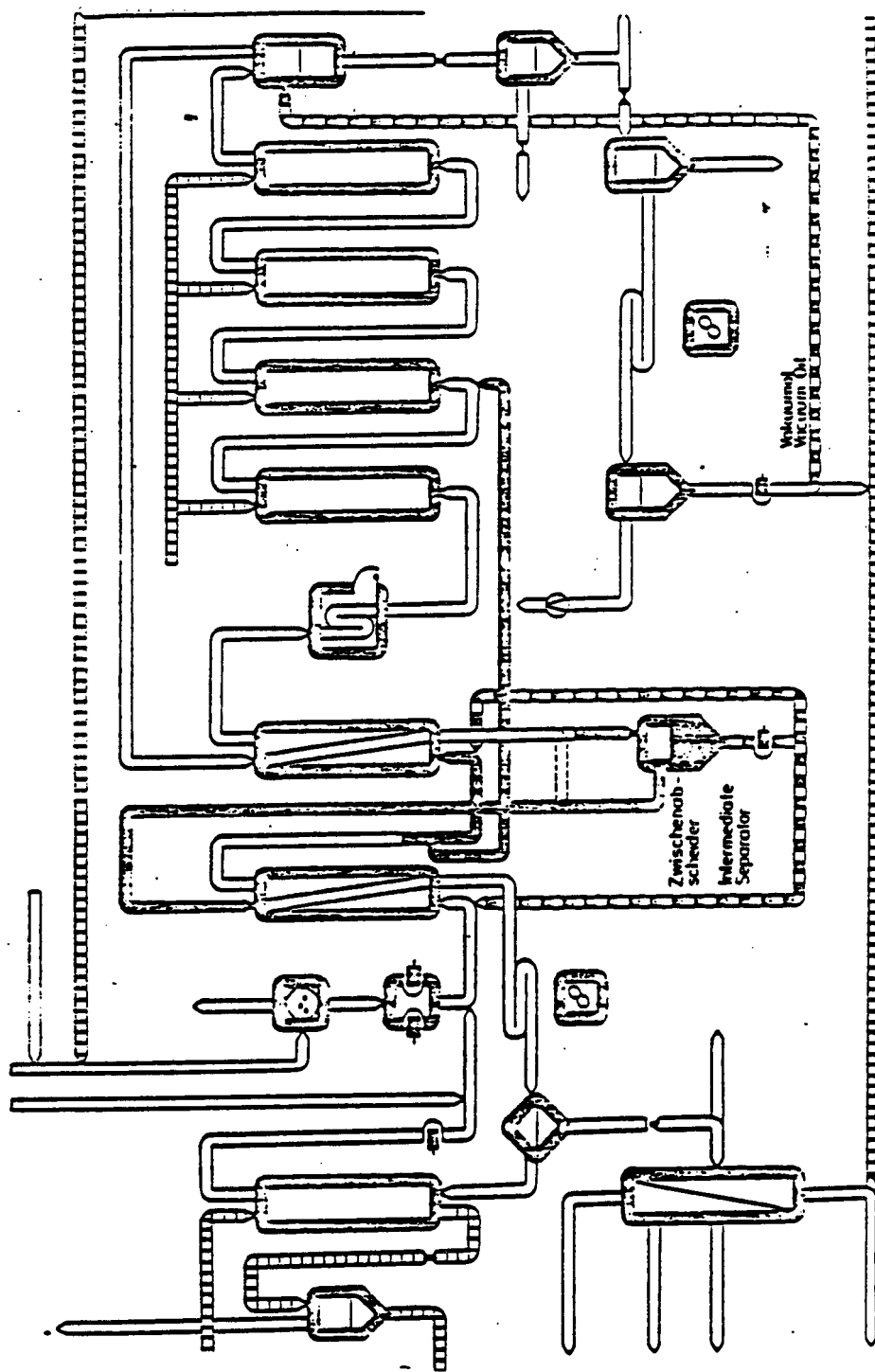


Figure 2. COAL SLURRY PREHEAT TECHNIQUE

also entered by the hot separator vapors. The vapors are cooled by the coal slurry, the heavy oils are condensed, and the slurry is preheated and diluted by the condensed oil. This also permits the use of coarser coal. Since the coal slurry is being transported only in tubes, there is little danger of sedimentation.

Coal particle diameters up to 1 mm (0.04 in.) are possibly applicable. The effluent of the mixing zone is separated into the hot coal slurry which is fed into the first reactor, and into the overheat vapors containing the products and the excess hydrogen. Application of this intermediate catchpot results in another advantage. The coal while being heated normally splits off CO_2 , H_2O , CH_4 , etc. These compounds enter the reactor and increase the total pressure by their partial pressure. Now these compounds are eliminated from the coal slurry prior to entering the reactor. Also the physically adsorbed water is stripped off and withdrawn from the intermediate separator overhead. This preheating system has the advantage that there is a reduction of the pressure resulting from a higher concentration of the hydrogen in the reactor and a lower pressure drop of the slurry compared with the use of heat exchangers. This new method is pictured in Figure 3. Saarberg believes that this arrangement can help make coal liquefaction simpler and more economic.

The final slurry preheat output is sent at a temperature of 430°C to the reaction section which consists of four reactors in series which operate at a pressure of 300 bars. Oil or quench gas is injected into these reactors to maintain an operating temperature range of 470 to 475°C . The reactor product is then sent to a separator which produces a bottom containing oil, ash, catalyst and unreacted coal. These bottoms are then reduced in pressure to 50 bars and fed to a vacuum distillation unit. An attempt will be made to recover energy from this pressure letdown stage via a piston engine device. The residual material from the vacuum distillation section which contains 50% solids and 50% bitumen with a melting point of 80°C can be gasified to generate process hydrogen. Gases which exit the top of the separator are used to supply part of the feed preheat energy via a concentric tube heat exchanger.

The pilot plant is designed for two different operations to form a distillate synthetic crude oil. In the first technique coal is processed only by high pressure hydrogenation. The expected results are shown in Table 1 as compared to that of the IG Farber process. In the second mode of operation the feed will also undergo carbonization after hydrogenation. The milder

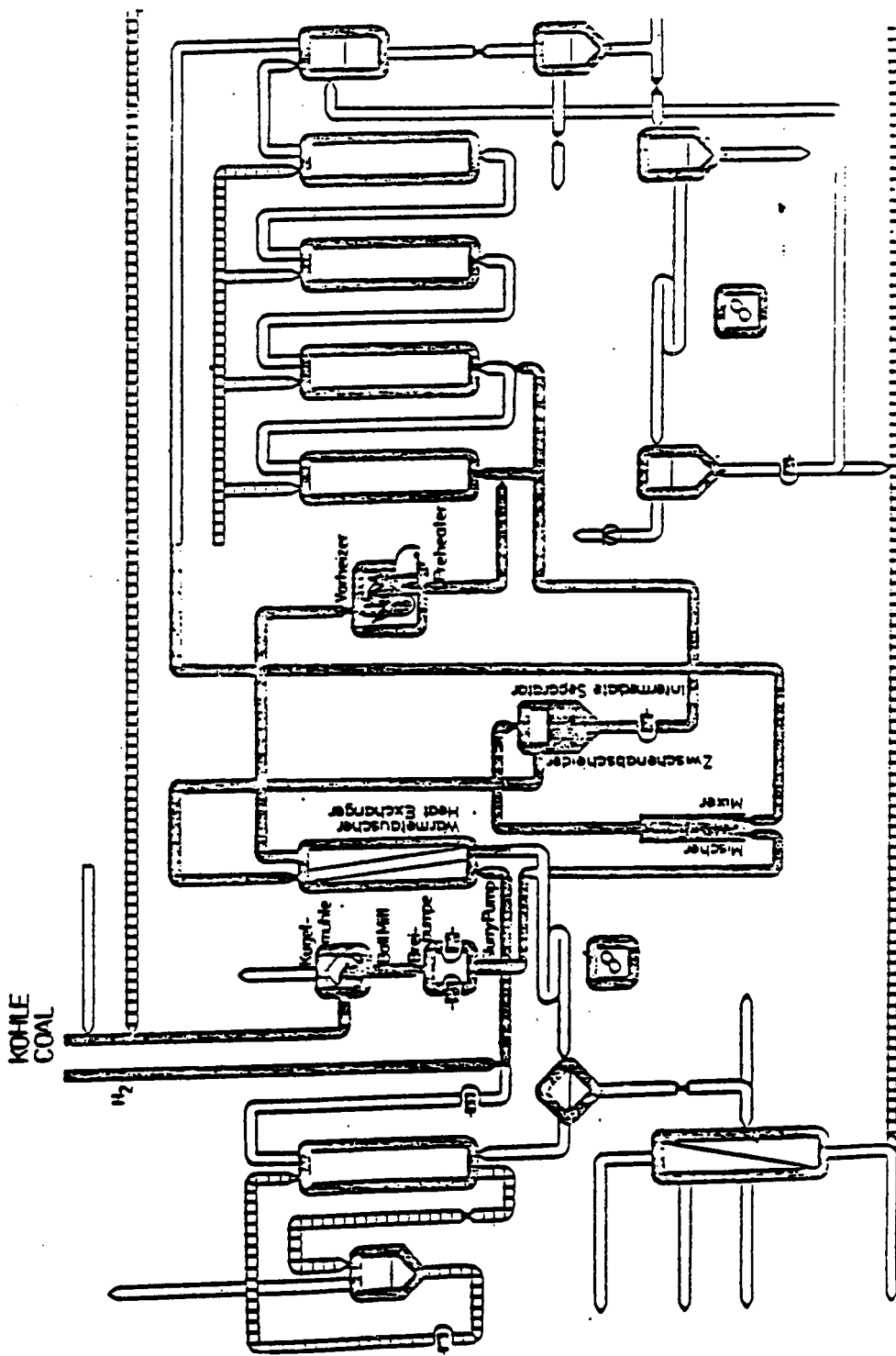


Figure 3. COAL SLURRY PREHEAT METHOD THAT ELIMINATES HEAT EXCHANGER

hydrogenation conditions of this second mode of operation seems more attractive from the standpoint of catalyst and hydrogen consumption. The byproduct char can then be gasified to produce hydrogen.

In the bench scale PDU the cut point of the atmospheric distillation was increased from 325°C in the old IG Farben process to 400°C.

Table 1. COMPARISON OF PRODUCTS FROM THE SAARBERG AND CLASSICAL IG PROCESS

<u>Conditions</u>	<u>Classical I.G. Process</u>	<u>Saarberg Modified (Bench Scale)</u>
Pressure, bar	700	300
Temperature, °C	490	475
Catalyst	iron	iron
CHSV(x)	0.62	0.65
<u>Product Distribution wt. % of M.A.F. coal)</u>		
C ₁ - C ₄ hydrocarbons	20.0	15.0
C ₅ - 200°C distillate	12.1	14.6
200 °C - 325°C distillate	31.4	30.2
325°C +	17.4	9.4
H ₂ O, H ₂ , NH ₃ , CO, CO ₂	13.9	11.7
Carbonization:		
Coke and Gas	12.5	--
Vacuum distillation:		
Residual oil	--	19.0
Unreacted coal;	(3)	5.6
H ₂ reacted	<u>(7.3)</u>	<u>(5.5)</u>
Total	107.3	105.5
(x) coal hourly space velocity		

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This increase reduces the cracking which occurs in the sump phase which is only slightly selective compared to a fixed-bed catalyst. This lowers the reaction temperature 10°C which results in the suppression of undesirable light hydrocarbons which have a higher hydrogen content. These changes have led to an improvement in liquefaction operating conditions. Hard coal has been liquefied at a pressure of 285 bars and a hydrogen consumption rate of 5.5 weight percent of the m.a.f. coal. Oil yields have increased to more than 56%. A technology fact sheet for this process is presented in Table 2.

Process Goals

The Saarberg process is a modification of the commercially proven IG Farben (Bergus-Piers) process. The major project goals of the Saarberg process are to reduce the operating pressure to a maximum of 30 bars compared to the previous 600 bars, reduced hydrogen consumption, and improved heat economy. This will be accomplished by employing novel slurry preheat concepts, the use of vacuum distillation, novel energy recovery equipment and a new recycle solvent approach.

Simultaneously with the experimental program, Saarberg has followed the upgrading of the sump phase oil to marketable products through cooperation with BASF. The main objective is the production of gasoline, particularly of high-octane blending components. For this purpose the coal oil is first refined to eliminate nitrogen, oxygen and sulphur. In a hydrocracker the refined middle distillate is cracked to naphtha. In a subsequent power former, both naphtha from the hydrocracker and sump-phase naphtha are transformed into high octane gasoline. The gasoline which was produced from the bench scale oils in the laboratories of BASF had a research octane number (clear) of about 104.

Relationship to Prior Technology

The Saarberg process is a modification of the IG Farben process. In this process, crushed and dried coal was mixed with a combination of red mud, FeSO_4 and Na_2S and recycle oil to form a paste. This was done in rotating mills with steel balls as grinding elements in the presence of oil to prevent oxidation. The homogenized paste was then fed to high pressure pumps for injection into the slurry feed heat exchangers. The discharge pressure in the injection pumps was 700 bars (10,500 psi). In the heat exchangers, slurry and hydrogen

Table 2 Part i TECHNOLOGY FACT SHEET: Saarberg Catalytic Hydrogenation Coal Liquefaction Process

NEW TECHNOLOGY (PROCESS DESCRIPTION AND GOALS): The Saarberg process is a modification of the commercially proven IG Farben process (Bergus-Piers). The main differences are a lower process pressure, reduced hydrogen consumption, improved heat transfer technique and more efficient produce separation techniques.

RELATIONSHIP TO PRIOR TECHNOLOGY (INCLUDING STATE OF DEVELOPMENT OF PRIOR TECHNOLOGY): This process is a modification of the commercial proven IG Farben process. The IG process produced nearly 4 million tonne of products per year by 1943. All capacity was shut down in 1945.

CHARACTERISTICS OF THE TECHNOLOGY:

<u>CHARACTERISTICS</u>	<u>NEW TECHNOLOGY</u>	<u>PRIOR TECHNOLOGY</u>
PRIMARY OUTPUT (DESIGN CASE).....	distillate oil, gas	distillate oil, gas
TYPE OF PROCESS.....	catalytic hydrogenation	catalytic hydrogenation
FEEDSTOCK REQUIREMENTS.....	coal with ash content > 15%	ash content > 15%
OVERALL THERMAL EFFICIENCY (INCLUDING BY-PRODUCTS)..	56%	~40%
CARBON CONVERSION EFFICIENCY.....	944%	97%
OPERATING TEMPERATURE.....	475°C	700 bar
OPERATING PRESSURE.....	300 bar	490°C
BY-PRODUCTS.....	residual oil	coke, gas

Table 2 Part 2 TECHNOLOGY FACT SHEET: Saarberg Catalytic Hydrogenation Coal Liquefaction Process

STATUS OF DEVELOPMENT:

OPERATING FACILITIES - 6 tonne/day pilot at Volklingen-Furstenhausen, West Germany operated by

Gesellschaft fur Kohleverflussigung, at joint venture of Saarbergwerke AG

and Gelsenberg AG which is a subsidiary of Deutsche BP.

MAJOR FUNDING AGENCY

75% funded by West German Ministry of Research and Technology State of Saarband also provides funds.

ANNUAL LEVEL OF FUNDING - Project total cost of approx. \$21 million.

TECHNICAL PROBLEMS:

(Empty lines for technical problems)

OTHER FACTORS AFFECTING OVERALL FEASIBILITY:

(Empty lines for other factors affecting overall feasibility)

flowed up outside the tubes with overhead vapors from the hot separator flowing countercurrently inside the tubes. Next, the coal paste and hydrogen were preheated to about 430°C (806°F) in a gas-fired heater. The gas/slurry mixture was then passed through four vertical reactors in which coal liquefaction took place at about 480°C (896°F). The exothermic reaction was controlled by the addition of cold recycle hydrogen. The effluent from the last reactor was separated into an asphaltene-free overhead product and a bottom product containing the unconverted coal, ash, catalyst, heavy distillate and residue oil. The gases and oil vapors from the top of the separator passed through the tubes of the slurry feed heat exchangers and were finally cooled. While the remaining gas was scrubbed to recycle purified hydrogen back into the process, the liquid was withdrawn from the cold catchpot and passed to an atmospheric distillation tower. All of the 325°C+ (617°F+) distillate was recycled as pasting oil. The main product was middle distillate oil which normally was converted to naphtha by hydrocracking. The hydrocracked naphtha and refined sump-phase naphtha were then reformed to high octane gasoline by the IG DHD (German acronym for pressure hydrogen dehydrogenation) process.

In the old system, processing of the hot separator bottoms consisted of two operations, centrifugation and carbonization. Both asphaltene-containing centrifuged filtrate and carbonizer oil were used as pasting media. The application of more severe hydrocracking conditions meant that the asphaltenes produced had to be recycled to the reactors since they could not be used elsewhere. In one German plant asphalt was taken out of the sump phase and used as a binder for weakly caking coals.

This process was first developed in Germany by Friedrich Bergius. In 1910 he received a Nobel Prize for his discovery of the hydrogenation process. The process was further developed and commercialized by the IG Farben chemical conglomerate. The first commercial size plant started operation in 1927 using brown coal as the feedstock at Leuna. By 1943, 12 plants were in operation with a combined capacity of 4 million metric tons of production.

Operating Facilities

In 1974 Saarbergwerke AG started the development of coal liquefaction technologies. This organization is a conglomerate owned by the Federal and State Governments (Federal Republic of Germany 74%, State Government of the

Saar 26%) with interests in mining, industrial and commercial operations. These include the mining of coking and steam coals, operation of coal preparation facilities, coking plants, electric power plants, oil and tar refineries, the manufacture of tools and machines, plastic and plastic products, packaging and building materials, construction and operation of central heating plants, sales, distribution, engineering, consulting, insurance, mortgages, and travel-agency activities.

In 1975 a continuous 10 kg/hr bench scale unit was constructed by Saarberg for initial testing of improvements to the IG Farben Process. This work led to the construction of a 6 tonne/day pilot which began operation in July of 1981. This pilot plant located at Volklingen-Furstenhausen is a 50-50 joint venture of Saarbergwerke AG and Gelsenberg AG, a subsidiary of Deutsche BP. The new joint venture firm, Gesellschaft fur Kohleverflussigung (GFK), will operate the pilot plant for a total of three years from commission to gather data for a full scale commercial plant. The Saarberg liquefaction plant is located adjacent to the Saarberg/Otto coal gasification pilot plant and near the Furstenhausen coke works, The Saarland oil refinery, the Fenne power plant, and the Saarbergwerke AG central laboratories.

Future plans call for the construction of a large demonstration scale plant that will convert 2.3 million tonne/year of coal into one million tonne/year of synthetic gasoline and other liquids. This would represent nearly 5% of West Germany's current gasoline consumption. A final decision in the construction of this plant will be made in 1983 when pilot plant testing will be nearly complete. This plant will take four or five years to construct.

Major Funding Agencies

The 6 tonne/day pilot plant at Volklingen-Furstenhausen was constructed at a cost of approximately \$21 million. The West German Ministry for Research and Technology sponsored 75% of this cost and the State of Saarland contributed an additional DM 1.5 million.

Technical Problems

No technical problems have been reported.

Capital Costs

The Saarberg liquefaction process and the Ruhrkohle/Veba Oil liquefaction process are both modifications of the commercially proven IG Farben Process. Both processes will operate at similar temperatures and pressures, and both will have similar efficiencies. Based on equipment modification of both these processes it would be logical to conclude that they would have similar capital cost requirements.

WEST GERMAN COAL LIQUEFACTION PROCESS

13. Rheinbraun Brown Coal Liquefaction ProcessProcess Description

The Rheinische Braunkohlen Werke AG (Rheinbraun) direct coal liquefaction process (also known as the HVB process — Hydrierende Verflüssigung von Braunkohle) is based on the IG Farben (Bergius-Piers) process.

In the HVB process, liquefaction of coal is carried out in two stages. In the first stage (called sump-phase hydrogenation), dry brown coal is catalytically converted into coal oil. In the second stage (gas-phase hydrogenation), the coal oil is converted into motor fuels by conventional oil refining techniques such as hydrocracking and reforming, or into feed materials for the chemical industry. Development work has been centered on improvements to sump-phase hydrogenation stage.

In the sump-phase hydrogenation (see Fig. 1), coal with a grain size of <1 mm (0.25 in.) and a water content of 10 to 12% by weight is used. The coal is slurried with sulfur and iron salts (as catalysts) in a recycle oil produced in the process. The slurry is heated to 350°C (662°F). In this process, water is evaporated from the slurry and humic acids are destroyed to such an extent that calcium carbonate does not stick in the hydrogenation reactor — a problem that occurred with the old technology.

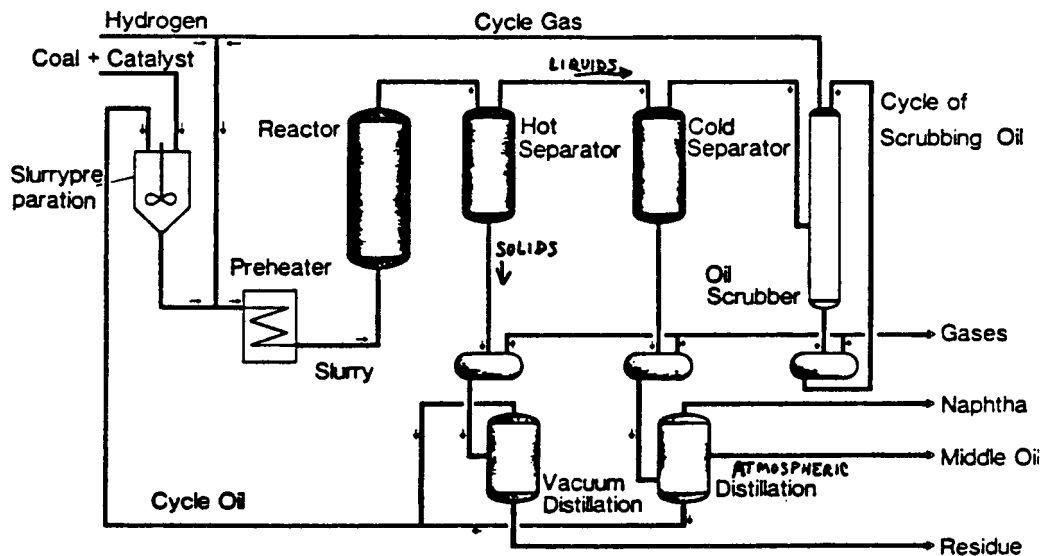


Figure 1. FLOW DIAGRAM OF THE SUMP-PHASE HYDROGENATION STEP

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Following this thermal pretreatment, the slurry is pumped into the high-pressure section. Before the slurry enters the preheater, hydrogen (contained in a mixture of fresh gas and recycle gas) is added. The mixture enters the hydrogenation reactor at a temperature of approximately 420°C (788°F). Under a reaction pressure of some 300 bars (4500 psi), the major part of conversion from coal into coal oil takes place.

The average retention time of coal in the reactor is about 30 min. The products of hydrogenation are separated in a hot separator at about 400°C (752°F) into a bottom product containing solids and a top product free of solids. The top product is cooled to about 50°C (122°F). This is accomplished by water cooling in the test plant. In future plants this heat will be largely used for heating up the coal oil reactor feed slurry. In the cold separator the condensed oils are separated from the gases. The gases are scrubbed with oil to remove hydrocarbons. The scrubbed gas is recycled to the hydrogenation reactor.

After depressurization, the bottom product from the hot separator which contains heavy oils, catalysts, and ash is distilled under vacuum. The vacuum distillate is used as a component of the recycle oil. In future plants the vacuum residue will be used for generating fuel gas or hydrogen for the process.

The bottom product from the cold separator is subjected to a multistage distillation.

The distillate fractions, i.e., the naphtha and middle oil, are the oil yields of the HVB process. The distillation residue is used as a further component of the recycle oil. Gases separated at different depressurization stages will be processed in future plants for the production of liquid gas and/or for the generation of hydrogen or as fuel gas.

Various catalysts, including iron mud from aluminum processing and iron salts, have been tried. Sulfur and iron salts are currently being used, but the exact form of the catalysts has not been divulged.

About 5 parts by weight of hydrogen per 100 parts of maf coal are added in the sump-phase hydrogenation stage (see Fig. 2).

Vacuum distillation is used to separate the liquid product from the solids which consist of ash, catalyst, and unconverted coal. The solids are

gasified to generate hydrogen. Part of the distillate is recycled for slurring the coal feed. Vacuum overheads are subjected to further multistage distillation. The resulting distillate fractions, i.e., naphtha and middle oil, constitute the oil yield of the HVB process.

Nearly 50% of the organic material in the coal is converted into syncrude which has a boiling range between 35°C (95°F) and 350°C (660°F). Ten tons of coal (with a moisture content of 60 wt %) are required to produce 1 ton of liquid fuel. This includes coal for steam and power generation and for hydrogen generation. A total thermal efficiency of about 50% is claimed, but it is not clear if this includes product upgrading requirements.

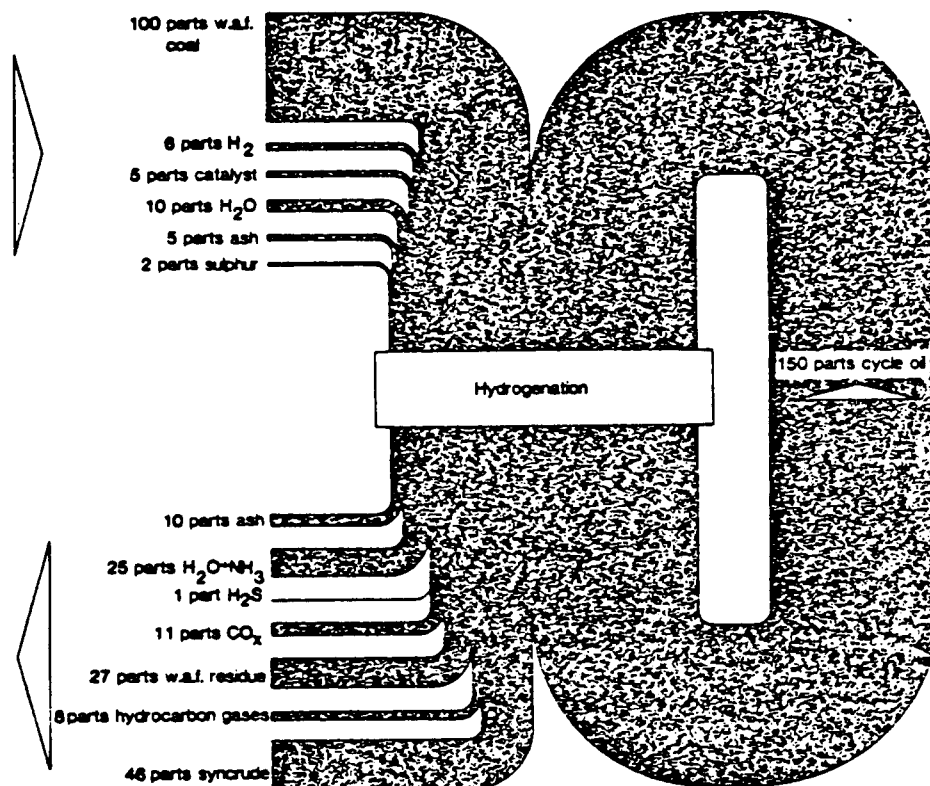


Figure 2. SUMP-PHASE HYDROGENATION MATERIAL BALANCE

The syncrude contains approximately 4 wt % oxygen and about 0.2 wt % nitrogen and sulfur each. Because of these concentrations of heteroatoms the syncrude must be further hydrogenated; this is accomplished by means of a catalytic middle-oil refinement process. The ratio of naphtha to middle oil shifts from approximately 2:1 to about 1:1 after refining. While the resulting middle oil, with a cetane number of 50, meets the requirements for diesel fuel without any further treatment being required, the naphtha must be catalytically reformed to meet this specification.

After catalytic reforming of the heavy naphtha fraction (80-180°C = 180-360°F), the combined total gasoline fraction has a research octane number (RON) of 97 and is considered to be premium gasoline. The properties of the middle distillate and gasoline fraction are presented in Table 1.

Table 1. MIDDLE DISTILLATE AND GASOLINE PRODUCT QUALITY

	<u>Middle Oil</u>	<u>Gasoline</u>
After Hydro-Treating		
— Cetane Number	50	--
— Research Octane Number	--	66
After Gasoline Reforming		
— Research Octane Number (unleaded)	--	97
— Research Octane Number (leaded)	--	99

Due to the properties of the HVB process products — high contents of parafins and naphthenes in the refined gasoline and middle oil and high content of aromatic substances in the reformed heavy naphtha — it is possible to produce the basic chemicals of organic chemistry such as olefins and aromatic BTX (benzene, toluene, xylene) substances from brown coal. A technology fact sheet for this process is presented in Table 2.

Process Goals

The objective of the HVB process is the production of chemical raw materials and/or motor fuels. Due to their high content of hetero-atoms the products from the sump-phase hydrogenation of brown coal have to be post-treated by hydrogenation before being used for other purposes. Syncrude from coal of the sump-phase hydrogenation contains approx. 70 per cent of middle oil and approx. 30 per cent of naphtha. By means of a catalytic middle oil refinement, the phenolic fractions in the middle oil are converted into hydrocarbons the major part of which will boil in the naphtha-range. Thus, the ratio of naphtha to middle oil shifts from approx. 2:1 to approx. 1:1 after the refinement of the middle oil and the naphtha fractions from the sump-phase hydrogenation.

Table 2 Part 1 TECHNOLOGY FACT SHEET: Rheinbraun Brown Coal Liquefaction Process

NEW TECHNOLOGY (PROCESS DESCRIPTION AND GOALS): The Rheinbraun process liquefies coal in a two stage manner.

In the first stage (called the sump-phase hydrogenation), dry brown coal is catalytically converted into coal liquids. In the second stage (gas-phase hydrogenation), the coal liquids are converted into motor fuels by conventional refining. Development work has been centered on improvements to the sump-phase hydrogenation stage.

RELATIONSHIP TO PRIOR TECHNOLOGY (INCLUDING STATE OF DEVELOPMENT OF PRIOR TECHNOLOGY): This process is

based on the I.G. Farben process which was developed in the 1920's.

CHARACTERISTICS OF THE TECHNOLOGY:

CHARACTERISTICS	NEW TECHNOLOGY	PRIOR TECHNOLOGY
PRIMARY OUTPUT (DESIGN CASE).....	gasoline, middle distillates	gasoline, middle distillates
TYPE OF PROCESS.....	catalytic hydrogenation	catalytic hydrogenation
FEEDSTOCK REQUIREMENTS.....	Lignite	Lignite
OVERALL THERMAL EFFICIENCY (INCLUDING BY-PRODUCTS)...	50%	~40%
CARBON CONVERSION EFFICIENCY.....	98%	--
OPERATING TEMPERATURE.....	460°C	478°C
OPERATING PRESSURE.....	300° bars	710 bars
BY-PRODUCTS.....	--	--

Table 2 Part 2 TECHNOLOGY FACT SHEET: Rheinbraun Coal Liquefaction Process

STATUS OF DEVELOPMENT:

OPERATING FACILITIES - A 0.3 ton/day process development unit has been operated by Rheinbraun at
Wesseling since 1978.

MAJOR FUNDING AGENCY West German Ministry of Research and Technology

ANNUAL LEVEL OF FUNDING - Over 6 million DM

TECHNICAL PROBLEMS: No major problems reported to date.

OTHER FACTORS AFFECTING OVERALL FEASIBILITY: Rheinbraun has estimated that the cost of gasoline from this process
will be 50% more expensive than gasoline from crude oil.

The development work is centered on the improvement to be made on the sump-phase hydrogenation of brown coal. In contrast to this, the gas-phase hydrogenation of syncrude on the basis of brown coal can be largely performed by means of the conventional techniques applied by crude oil refineries, for the coal oil on the basis of brown coal, except for its high phenol content, may be to a certain extent compared with the corresponding fractions from crude oil.

Process improvements include a reduction in reactor operating pressure from the wartime level of 700 bars (10,500 psi) to the present 300 bars (4500 psi), and an increase in the space-time yield of some 50%. Reaction temperature was also reduced from the previous value of 478°C (892°F) to 460°C (860°F).

Relationship to Prior Technology

The development on hydroliquefaction of brown coal is based on the work of Bergius and Pier in the 20's and on the know-how gained in the operation of the IG Farber full-industrial scale plant at UK-Wesseling. In 1943 this plant had an output of approx. 250,000 metric tons of motor fuel produced on the basis of Rhenish brown coal. The relationship of the new process compared to that of the old IG Farben process is shown in Table 3.

Table 3. COMPARISON OF OPERATION CONDITIONS OF THE IG FARBEN PROCESS AND THE HVB PROCESS

	<u>oil (1943)</u>	<u>new (1980)</u>
Operating Pressure (bars)	710	300
Partial H ₂ Pressure (bars)	510	270
Reaction Temperature (°C)	478	460
Processing of Residue	Centrifuging	Distilling
	Carbonizing	Gasifying
Specific Coal Throughput (t/m ³ h)	0,4	0,6
Space Time Yield (t/m ³ h)	0,16	0,28
H ₂ Consumption (in terms of w.a.f. coal) (wt %)	6,5	6,0

Operating Facilities

Rheinbraun has operated a test plant with a throughput of 0.25 tonnes (0.3 tons) per day at Wesseling near the old IG Farben-facility since 1978. The test plant is being used to develop basic data for the construction of an HVB pilot plant which is planned to start operating in 1985. The pilot plant will have a coal throughput of 15 metric tons/hr (16.5 tons/hr) and will constitute the final development step prior to construction of a commercial HVB plant in the Rhenish area.

The first line of the commercial plant, which will serve as a demonstration plant, will have a coal throughput of approximately 250 metric tons (275 tons) of raw coal per hour. Expected to start operation in the early 1990's, the demonstration plant will produce 400,000 metric tons (440,000 tons) of motor fuels and/or chemical feedstocks per year (440,000 tons/yr of motor fuel is equivalent to about 9000 barrels/day). The development schedule for this process is presented in Table 4. A decision by the West German Ministry of Research and Technology as to whether or not the process will continue to the demonstration stage will be made in late '82 or early '83.

Table 4. DEVELOPMENT SCHEDULE FOR THE RHEINBRAUN HVB PROCESS

<u>Status of the Plant</u>	<u>Working Period</u>	<u>Coal Consumption*</u> t/a	<u>Product Quantity</u> t/a
Commercial Plant	1941-1944	3.75×10^6	250×10^3
1. Test Plant**	1978-1986	75	18
2. Pilot Plant**	1985-1993	110×10^3	28×10^3
3. Demonstration Plant	1992-2000	$3,5 \times 10^6$	400×10^3

1st line of a full-scale industrial plant

* Raw coal with 60 wt% H₂O.

** Coal for power and H₂ excluded.

Major Funding Agencies

The West German Ministry of Research and Technology is supporting 80% of the nearly 6 million DM development effort.

Technical Problems

Areas in which further development is necessary include the following: coal hydrogenation, vacuum distillation of the raw liquid product, and gasification of the vacuum distillation residue.

No major technical problems have been reported.

Capital Costs

It has been reported by Rheinbrown that the cost of producing gasoline by means of the HVB process is estimated to be approximately 50% higher than from crude oil.

WEST GERMAN COMBINED CYCLE COAL GASIFICATION PROCESS

14. Steag/Lurgi Combined Cycle ProjectProcess Description

Steag AG has developed a combined cycle power generation process (known as the KDV-KOMBIBLOCK System) based on the conventional Lurgi gasification process. In 1972 Steag started operation of a 170 MW ce) demonstration facility at Lunen, West Germany. The plant consists of the following:

- five Lurgi pressure gasifiers
- two half-capacity supercharged boilers
- one 96 MW steam turbine
- one 74 MW gas turbine
- and accessories

as shown in Fig. 1.

Coal is gasified by means of air and steam at 20 bar pressure. A lean, raw gas is produced. Each gasifier has a primary wet scrubber for the removal of dust and tar, followed by a common, secondary scrubber. After the cleaning, the pressure of the gas is let down to 10 bar in an expansion turbine, then the gas is fired in the burners of the supercharge boilers. The combustion products formed at 1400°C first give off heat by raising steam for the steam turbine, then expand in the gas turbine to near atmospheric pressure at about 400°C temperature. Flue gas temperature is further reduced to about 170°C in a recovery type feedwater heater.

Air is provided by the main compressor of the gas turbine at 10 bar pressure. About 90% of the compressed air is used for firing the gas in the burners of the boilers, the balance is further compressed to 20 bar pressure for the gasifier in a booster compressor, which is driven by the expansion turbine. Steam for the gasification is taken from the steam turbine.

The gasifiers are of full commercial size, each rated to process about 13 t/h coal. More than sixty LURGI gasifiers have been built for the production of town gas and synthesis gas in chemical plants, using oxygen/steam mixture for gasification. This application, with air/steam mixture for the production of low-Btu gas, was a first and required some extrapolation beyond experience. The same applies to the gas purification, as the standard method

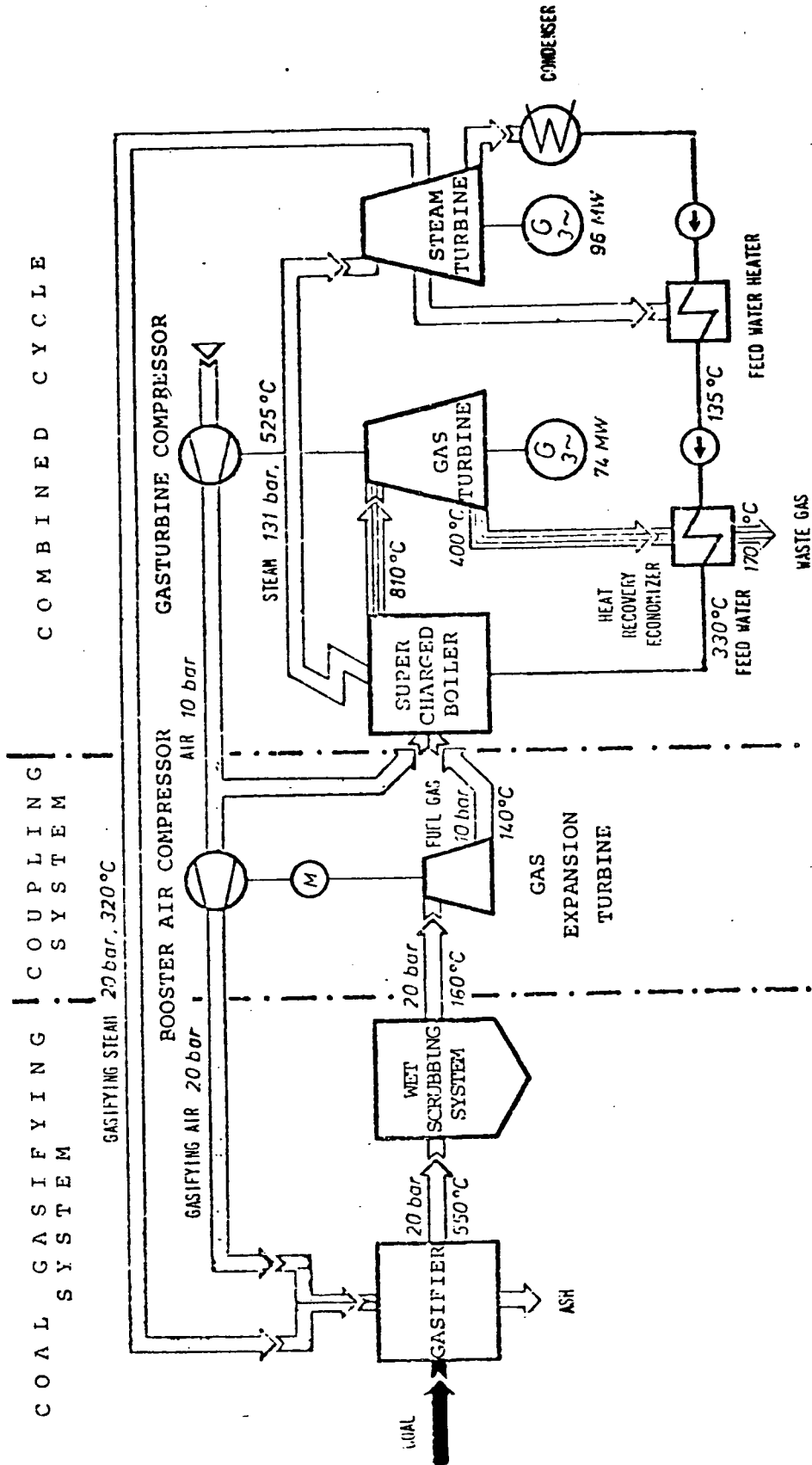


Figure 1. STEAG KDV-DOMBIBLOCK 170 MW (e) DEMONSTRATION PLANT

involves the condensing-out of the impurities from the raw gas at ambient temperature. This is associated with loss of sensible heat, which is undesirable for power plant application.

Turning to the power plant, in the STEAG cycle, unlike in most combined cycles, the upper region of the available temperature spread is utilized for the steam cycle and the lower for the gas turbine cycle. Here, the gas turbine is powered by the flue gases of the steam generator, which has to be pressurized for that purpose. The supercharged boiler also represents a first and required extrapolation beyond experience.

The Lunen boilers are designed for conventional velocity and low draft loss. However, a tenfold increase in the pressure of the combustion products results in the same increase in the heat transfer rate, as a tenfold velocity increase would cause, yet without undue pressure drop, thus preserving the power generation potential of the flue gas. Owing to very high heat transfer rate and volume reduction, both caused by the 10 bar combustion pressure, the boiler is light, compact and fast responding. It is lighter than the typical package boiler, requires only one-tenth of the space taken up by an atmospheric boiler and a lance type burner provides 180 Gcal/h (about 714×10^6 Btu/hr) heat input with low-Btu gas. The two, half capacity boilers were completely shop fabricated by the Balcke-Dürr company. Field erection was minimal.

At the time of the plant design, the largest gas turbine available in the German market had a net rating of about 50 MW. The VF-93 type gas turbine developed by Kraftwerk-Union (KWU) was selected. It consists of a 17-stage compressor section and a 4-stage turbine section. In the STEAG cycle, the gas turbine delivers 74 MW net output at reduced specific cost. The reason is the increase in the mass flow rate through the turbine and the decrease in the mass flow rate through the compressor, as compared to natural gas operation. At Lunen, the cycle is operating with 2.6 excess air ratio. In the large plants, the excess air ratio will be reduced to 1.08. The steam turbine is of standard KWU design, with three uncontrolled and one controlled bleed point.

For technological reasons, the coal gasification plant and the combined cycle plant operate at different pressure levels, of about 20 bar and 10 bar, respectively. The pressure gap is usefully bridged by a 4 MW control aggregate.

The operating data for this combined-cycle process is presented in Table 1. A technology fact sheet is shown in Table 2.

Process Goals

The system is the result of investigations made by STEAG in 1968, with the purpose of finding a clean and economic way of generating electricity from coal. In the era of cheap oil and growing environmental concern it became apparent to STAEG, that coal could not remain competitive with oil, unless three conditions were met:

- improved environmental effects
- higher efficiency of conversion
- lower investment cost

The conclusion reached was that these aims could not be simultaneously achieved with conventional methods. Conventional technique has been pushed to its limits and, moreover, is burdened by increasingly stringent requirements to minimize pollution of the environment.

Table 1. OPERATING DATA FOR THE 170 MW(e) STEAG/LURGI DEMONSTRATION FACILITY

Operating Data

Coal Consumption	70 t/h
Fuel gas	
Production	290 t/h
Pressure	20 bar
Temperature	160°C
Calorific value	5000 kJ/kg
Supercharge boiler	
Steam capacity	340 t/h
Steam pressure	130 bar
Steam temperature	525°C
Combustion chamber pressure	10 bar
Flue gas outlet temp.	820°C
Gas turbine capacity	74 MW
Steam turbine capacity	96 MW
Plant output	170 MW
Auxiliary power	7 MW
Net Plant output	163 MW
Start-up time	35 min.
Overall efficiency	$\frac{\text{electric power}}{\text{net CV coal}} = 36\%$

Raw Gas Analysis (Vol. %)

	<u>Design</u>	<u>Analysis</u>
N ₂	40.0	42-45
CO ₂	14.0	9-11
H ₂	25.0	19-21
CO	15.8	20-23
CH ₄	5.0	3-3.5
C _n H _m	0.2	0.5-1

The objectives of a stage program, which STEAG consequently decided to pursue, were to find solution for:

- test under actual operation conditions
- demonstrate on large commercial scale

Table 2 Part 1 TECHNOLOGY FACT SHEET: Steag/Lurgi Combined Cycle Project

NEW TECHNOLOGY (PROCESS DESCRIPTION AND GOALS): Steag AG has developed a combined cycle power generation process (known as the KDU-KOMBIBLOCK SYSTEM) which is based on conventional Lurgi and gas turbine technologies. The goal of this 170 MW(e) project was to increase electrical generation efficiency, improve environmental effects, and lower investment costs.

RELATIONSHIP TO PRIOR TECHNOLOGY (INCLUDING STATE OF DEVELOPMENT OF PRIOR TECHNOLOGY): This system is based on Lurgi coal gasification technology.

CHARACTERISTICS OF THE TECHNOLOGY:

<u>CHARACTERISTICS</u>	<u>NEW TECHNOLOGY</u>	<u>PRIOR TECHNOLOGY</u>
PRIMARY OUTPUT (DESIGN CASE).....	170 MW(e)	
TYPE OF PROCESS.....	Combined Cycle	
FEEDSTOCK REQUIREMENTS.....	Non-slugging coals	
OVERALL THERMAL EFFICIENCY (INCLUDING BY-PRODUCTS)..	36%	
CARBON CONVERSION EFFICIENCY.....	99%	
OPERATING TEMPERATURE.....	1400°C	
OPERATING PRESSURE.....	20 bars	
BY-PRODUCTS.....	--	

Table 2 Part 2 TECHNOLOGY FACT SHEET: Steag/Lurgi Combined Cycle Project

STATUS OF DEVELOPMENT:

OPERATING FACILITIES -- A 170 MW (e) demonstration plant was commissioned in 1972 at Lunen. This facility operated to 1977.

MAJOR FUNDING AGENCY State of Kordrhein-Westfalen

ANNUAL LEVEL OF FUNDING - Construction costs were 80 million DM (1971 DM)

TECHNICAL PROBLEMS: All technical problems were resolved in the demonstration program

OTHER FACTORS AFFECTING OVERALL FEASIBILITY: The Lunen facility was shut down in 1977 due to economic reasons.

— implement for future generating capacity

a new, coal based power generation technology which could meet the three conditions. As a built-in constraint, the technology was to be based on the use of proven components and processes to the largest extent practicable, in order to minimize system development time and risk and maximize reliability. This stipulation was, however, not to preclude an innovative approach, neither in system engineering, nor in component design. Extrapolation beyond current experience, but within good engineering judgement, was anticipated and eventually resulted in many successful "firsts" as described later. In short, the approach was to be that of an innovative system user, rather than that of an interested process developer.

Working along these lines, STEAG investigated various technologies and opted for combining two known technologies — each proven in its own right — into one system, namely: (1) the gasification of coal under pressure for the production of a clean fuel gas palatable to the gas turbine, and (2) the use of standard gas and steam turbines in a combined cycle, specially adapted to the gasification process selected. This concept was implemented in the 170 MW prototype plant.

Relationship to Prior Technologies

The Steag KDV-Bombiblock system is based on the commercially proven Lurgi coal gasification process and conventional turbine technology.

Operating Facilities

The development of the STEAG coal gasification-combined cycle concept antedates the energy crisis. Conceptual studies were completed in 1968 and decision to build a 170-MW prototype was made in the spring of 1969. The unit was ready for commissioning in February 1972.

The prototype plant is located at the KELLER-MANN Generating Station of STEAG, in Lunen, West Germany, using space, buildings and structures available in this existing coal fired generating station. KEL-LERMANN is the first power plant in the world, in which electricity is commercially produced from coal by a combined cycle unit in conjunction with a LURGI coal gasification plant. The plant is designed as a medium-load peak load facility and is suitable for daily start-ups. To date, around one thousand million kWh power have been generated in approx. 10,400 operating hours, using 120,000 t light fuel oil and 230,000 t coal.

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Major Funding Agencies

The State of Nordrhein-Westfalen sponsored the research and construction of the 80 million DM (1971 DM) demonstration plant.

Operating Experience

Operation of the Lünen plant encountered difficulties from the outset due to the Lurgi gasification system. Owing to initial difficulties with the gas generating plant and in order to meet contractual obligations for energy supply, light oil burners were added to the gas burners for the boilers, rated for 75% capacity.

The combined cycle plant then operated for six months with oil firing on peaking and mid-range duty and reached the expected performance at 125 MW output. It was possible to fully demonstrate the operating capability of all components. It is worth mentioning the extremely high reliability of the supercharged boilers, in spite of their practically new design.

Based on early experience, various modifications and reconstruction had to be made to the gasification plant so that it could be matched to the varying load conditions imposed by the combined cycle plant. Operational testing of the whole plant continued in October-November 1973. Deficiencies noted during this operation, especially with respect to gas purity and tar separation, led to the decision to implement extensive additions and improvements to the gas purification plant and the tar separation system.

This work was completed by September 1974 and the testing with coal gas resumed. Gas purity improved from 100 mg/m³, particulates, so that in December 1974 the expansion gas turbine and the booster air compressor could be put in operation, thereby making the entire plant operational for the first time. Shortly thereafter the testing objectives of the "KOMBIBLOCK" were achieved in most respects and its development was deemed to be free from problems. However, the gas generating plant proved to require further development and more thorough testing.

Important and ultimately decisive engineering know-how and operating experience were obtained in four, long duration test periods between 1975 and 77. Table 1 contains statistics. The experience gained is summarized in Table 3.

The 170 MW prototype plant at Lunen has, over the past few year, achieved-over 10,000 operating hours with the combined cycle plant, — over 7,000 operating hours with the gasification plant and-over 4,000 operating hours with the H₂S removal pilot plant. More than 200,000 tons of coal, of various origin and properties, were used for the testing. This facility was closed down in 1977 due to economic reasons. Future plans call for the construction of a 500 MW plant. However, no formal commitment has been made to date.

Capital Costs

A summary of the capital costs for a Steag combined cycle facility with roughly 1000 MW of generating capacity is presented in Table 4. The total capital costs for this size facility is roughly 850 million dollars (1979 basis).

Table 3. 170 MW KDU-KOMBIBLOCK DEMONSTRATION PLANT-OPERATIONAL TESTING

<u>Period</u>	<u>March- April 1975</u>	<u>August- Novemb. 1975</u>	<u>May- July 1976</u>	<u>January April 1977</u>	
Number of days	37	116	58	75	
Number of Gasifiers together in Operation for days	One Two Three Four Five	-- 2 20 15 --	4 10 31 37 34	-- 3 29 25 1	-- 1 13 56 5
Cumulative Operating Hours since Commissioning by end of period					
Gasification Plant	3,100	4,900	5,850	7,300	
Boiler & Gas Turbine	4,900	7,300	8,600	10,430	
Steam Turbine	4,600	6,980	8,290	10,090	
Amount of Coal Used t	34,674	75,085	39,917	51,853	
Max. Output MW	148	120	110	114	
Electricity Produced 10 ⁶ kWh	91,44	192,06	109,75	168.2	
Supporting oil firing m ³ /h	7	0.0 to 1.1	2.4	7	
Air Used for:					
Coal Gasification m ³ h	90,000	84,000	84,000	82,000	

Table 4. SUMMARY OF CAPITAL INVESTMENT COSTS FOR A 1000 MW STEAG COMBINED CYCLE FACILITY

Gasification Process	Lurgi
Oxidant	Air
Combined-Cycle Type	STEAG
Case Designation	<u>MASW</u>
<u>PLANT INVESTMENT — \$1000</u>	
Coal Handling	26,039
Oxidant Feed	20,544
Gasification and Ash Handling	103,269
Gas Cooling	20,765
Acid Gas Removal and Sulfur Recovery	39,204
Lurgi Process Condensate Treating	60,213
Steam, Condensate and BFW	37,078*
Combined Cycle	244,946
General Facilities	60,542
Process Contingencies	24,092
Project Contingencies	<u>91,890</u>
TOTAL PLANT INVESTMENT	728,582
<u>CAPITAL CHARGES</u>	
Prepaid Royalties	2,760
Preproduction Costs	19,203
Inventory Capital	7,088
Initial Catalyst and Chemical Charges	1,472
Allowance for Funds	91,000
During Construction	
Land	<u>1,000</u>
TOTAL CAPITAL CHARGES	122,523
TOTAL CAPITAL REQUIREMENTS	851,105

* Cost includes tar boiler

BRITISH COAL GASIFICATION TECHNOLOGY

15. British Gas Slagging Coal Gasification ProcessProcess Description

The BG/Lurgi slagging gasifier is a modification of the commercially available "dry bottom" Lurgi process. This slagging gasifier, shown in Figure 1, will accept caking coals and operate at higher temperatures than the conventional Lurgi gasifier. The slagging gasifier is basically a semi-continuous, slowly moving bed gasifier of vertical, cylindrical construction. Crushed and screened coal, sized 3/8-inch to 1-1/2-inch is fed into the unit by a lockhopper mounted atop the gasifier. Recent tests have indicated that the gasifier will accept a fines content ($> 1/4"$) of 35%. The gasifier, which operates at 350 to 400 psig, is surrounded by a water jacket that utilizes boiler feedwater to recover heat escaping from the gasifier shell.

The steam and oxygen that are introduced through tuyeres near the bottom of the slagging gasifier pass upward through four distinct zones. The topmost portion of the gasifier acts as a preheat and coal drying zone where the coal comes into contact with the hot crude gases leaving the reactor at about 1100°F. As the coal travels down the reactor, devolatilization starts where the bed temperature ranges from 1150° to 1600°F. After devolatilization, the resulting char undergoes gasification.

The combustion zone is the fourth and last zone located at the bottom of the gasifier. Heat for the endothermic devolatilization and gasification reaction is produced in this zone by the combustion of carbon and oxygen, which produces mostly carbon dioxide. Molten slag, at about 2700°F, runs through a slag tap hole at the bottom of the gasifier. This slag drops into a quench vessel where it is solidified in quench water. The solid slag is then removed from the system through a slag lockhopper. Carbon conversion to products is better than 99.9%.

Crude gases that exit the gasifier at the top are scrubbed and cooled to remove the entrained tar and oils. These gases are cooled further to about 300°F by a waste-heat recovery boiler. The resulting washwater from the scrubbing unit is mixed with the condensate from the waste-heat boiler. Tar in the washwater also contains a mixture of coal and ash dust which is returned to the gasifier for cracking and gasification after processing is

FIXED BED SLAGGING GASIFIER

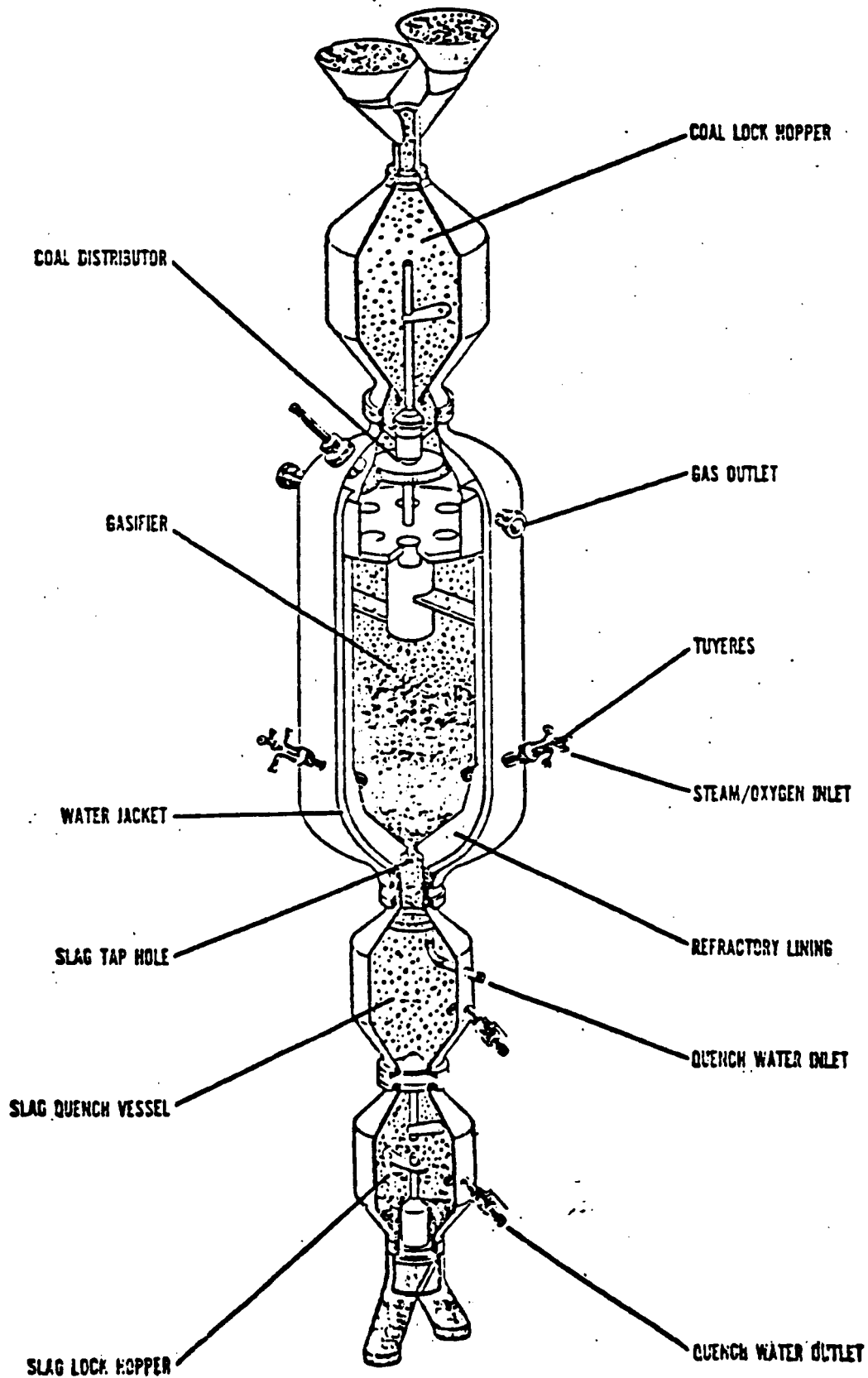


Figure 1. BRITISH GAS SLAGGING LURGI-TYPE GASIFIER

shown in Figure 2. Performance data for the Slagging Lurgi producing syngas is presented in Table 1.

Further processing of the off-gas for SNG production (if desired for the user's application) requires that part of the scrubbed crude gas be sent to a shift conversion unit where the following reaction takes place:



This adjusts the H_2 -to- CO ratio of the gas mixture to an optimum of 3.6 for the methanation reaction. Before the methanation reaction, removal of H_2S , CO_2 , and COS is accomplished in a Lurgi Rectisol unit. Because the physical absorption process uses low temperature methanol (down to -50°F), all hydrocarbons having two or more carbon molecules are removed. The Lurgi Phenosolvan process is used to extract phenols from the process water. Ammonia is removed from the gas by the CLL process aqueous ammonia.

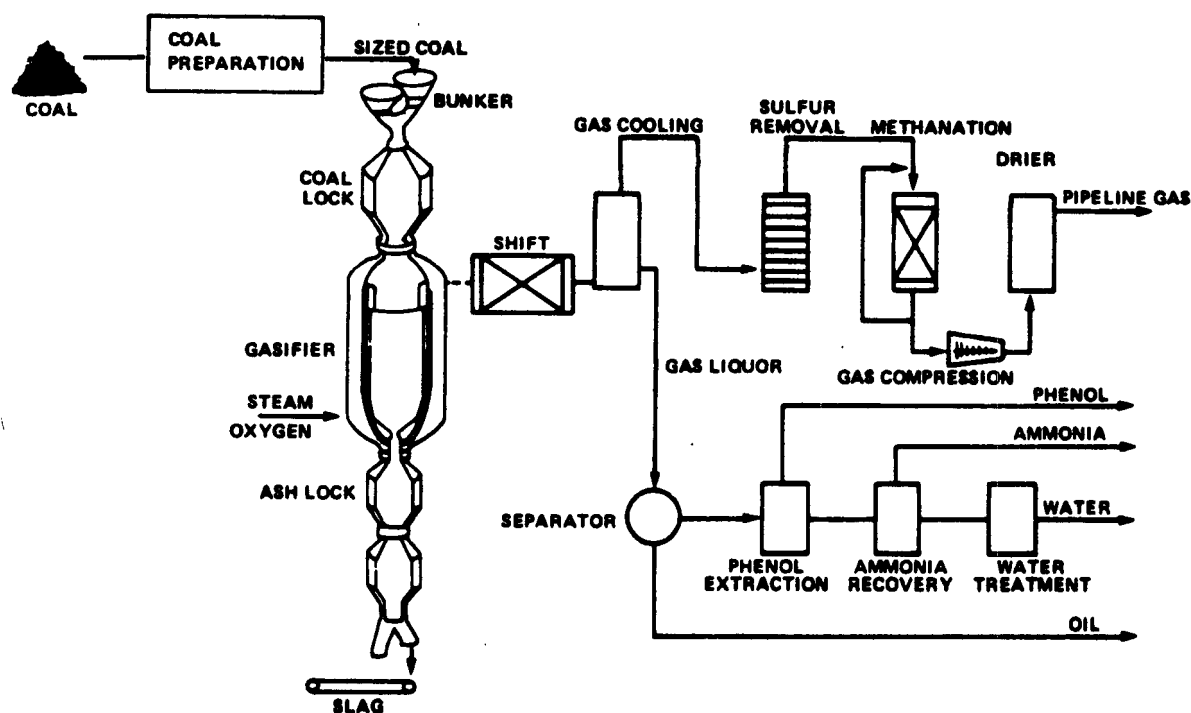


Figure 2. BRITISH GAS/SLAGGING LURGI PROCESS

Table 1. PERFORMANCE DATA FOR THE BRITISH GAS/LURGI SLAGGING AND LURGI GASIFIER AT WESTFIELD

Coal Origin	Princes Scotland	Rossington England	Ohio 9 USA	Pittsburgh 8 USA
Size (ins)	1-1	1-1	1-1	1/8-1 1/2
<u>Proximate Analysis, (% w/w)</u>				
Moisture	8.7	9.5	6.1	4.2
Ash	4.4	4.6	18.9	7.2
Volatile Matter	32.9	31.2	33.6	35.4
Fixed Carbon	54.0	54.7	41.4	53.2
<u>Ultimate Analysis (% w/w)</u>				
Carbon	83.0	83.5	79.6	82.4
Hydrogen	5.5	4.9	6.1	5.3
Oxygen	9.2	7.7	7.4	9.1
Nitrogen	1.4	1.7	1.2	1.5
Sulphur	0.5	1.7	5.6	1.6
Chlorine	0.4	0.5	0.1	0.1
B.S. Swelling No.	1 1/2	1 1/2	4 1/2	7 1/2
Caking Index (Gray King)	B	B	C	CB
<u>Operating Conditions</u>				
Gasifier Pressure, (aba)	24	24	24	24
Steam/Oxygen ratio (v/v)	1.3	1.3	1.3	1.3
Outlet Gas temperature (*F)	896	896	770	950
<u>Crude Gas Composition, (% v/v)</u>				
H ₂	28.6	27.2	28.7	28.9
CO	57.5	58.1	53.2	54.9
CH ₄	6.7	6.8	6.9	7.1
C ₂ H ₆	0.4	0.5	0.3	0.6
C ₂ H ₄	0.2	0.2	0.2	0.2
N ₂	4.2	3.9	4.0	4.4
CO ₂	2.3	2.9	5.5	3.4
H ₂ S	0.1	0.4	1.2	0.5
HHV, (Btu/scf)	375	375	362	375
<u>Derived Data</u>				
Coal Gasification Rate(lb/ft ² h)	852	848	664	666
Steam Consumption, (lb/lb coal)	0.405	0.398	0.390	0.407
Oxygen Consumption, (lb/lb coal)	0.539	0.549	0.555	0.547
Liquor Production, (lb/lb coal)	0.20	0.21	0.16	0.21
Gasifier Thermal Output, (therms/ft ² /h)	106	106	78	83
Gasifier Thermal Efficiency **, (%)	83.4	82.1	82.3	79.9
Coal expressed "moisture and ash free"				

** Defined as total product gas thermal output (based on HHV, including tar, oil, naphtha) divided by corresponding thermal input of coal feedstock and the fuel equivalent of the steam and oxygen used.

The resulting CO and H₂ are combined catalytically in a series of fixed-bed reactors using a pelleted nickel catalyst to produce CH₄ and H₂O. Heat, generated by this exothermic reaction is used to generate most of the process steam for the gasification reaction. In addition to these process steps, the plant will include support services, such as steam production, water purification, air separation plant, incineration, waste treatment, as well as product loading, coal storage, product tankage, and buildings.

The primary material balance, using Ohio No. 9 coal, for a proposed pipeline gas demonstration plant incorporating the BG/Lurgi slagging gasification process is shown in Figure 3. This gasification process is expected to be technically feasible for all U.S. coals with the possible exception of those coals having a moisture content greater than 35%. The cold gas efficiency of a commercial size facility producing SNG is expected to be 62.3%, which includes fuels requirements for steam and power generation. The overall efficiency of this process producing SNG, including all saleable energy products, is 66.8%. The technology fact sheet for the process is presented in Table 2.

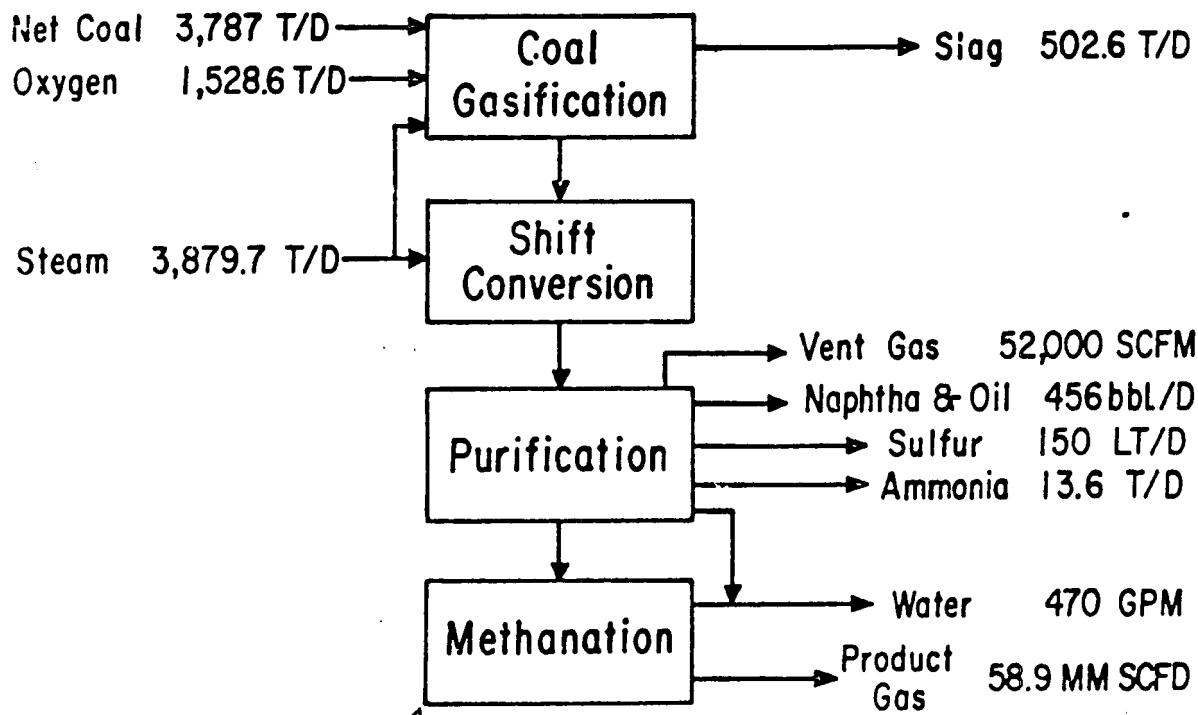


Figure 3. PIPELINE GAS DEMONSTRATION PLANT FIXED-BED SLAGGING GASIFICATION

Table 2 Part 1 TECHNOLOGY FACT SHEET: British Gas Slagging Lurgi Gasification Process

NEW TECHNOLOGY (PROCESS DESCRIPTION AND GOALS): Slagging Lurgi Coal Gasification Process.

Slagging Lurgi is a slowly-moving bed gasifier of vertical, cylindrical construction and operated in the slagging mode. Goals include higher gas output rate, reduced steam consumption, and higher thermal efficiency. Design case is oxygen blown for synthesis gas.

RELATIONSHIP TO PRIOR TECHNOLOGY (INCLUDING STATE OF DEVELOPMENT OF PRIOR TECHNOLOGY): Lurgi dry-bottom

process developed in 1930's and commercialized in the 1940's. Slagging Lurgi can accept all types (with few exceptions) of coal where conventional Lurgi cannot.

CHARACTERISTICS OF THE TECHNOLOGY:

CHARACTERISTICS	NEW TECHNOLOGY	PRIOR TECHNOLOGY
PRIMARY OUTPUT (DESIGN CASE).....	375 Btu/SCF	309 Btu/SCF
TYPE OF PROCESS.....	Fixed Bed	Fixed Bed
FEEDSTOCK REQUIREMENTS.....	Coal sized 1/4 to 1-1/2 inch	62.6%
OVERALL THERMAL EFFICIENCY (INCLUDING BY-PRODUCTS)...	66.8%	
CARBON CONVERSION EFFICIENCY.....	99.9%	
OPERATING TEMPERATURE.....	2700°F	Below coal's ash fusion temperature
OPERATING PRESSURE.....	350 psia	370 psia
BY-PRODUCTS.....	Steam, slag	Ash, steam, tars, oils, phenols

* Represents efficiency for high-Btu gas production.

Table 2 Part 2 TECHNOLOGY FACT SHEET: British Gas Slagging Lurgi Gasification Process

STATUS OF DEVELOPMENT:

OPERATING FACILITIES - Pilot plant operated at Westfield, Scotland with a 6 foot internal diameter

and capable of processing 300 to 350 tons/day of coal. Tests were completed

in 1981 with a successful 90 day run which converted 30,000 tons of coal to 1,750 million ft³ of syngas.

MAJOR FUNDING AGENCY Westfield plant funded by British Gas, 15 U.S. companies headed by Conoco, Lurgi, and EEC.

ANNUAL LEVEL OF FUNDING - Total 8 year demonstration cost of 6 ft gasifier totaled \$60 million.

TECHNICAL PROBLEMS: Molten iron (reduced) has a tendency to form when certain types of coal are used.

This molten iron is very corrosive to slag tap and refractory lining, and adversely affects slag flow characteristics. Also, reduced volume of raw gas to upper section of reactor makes, drying and pyrolysis of cold coal feed more difficult. The use of tuyeres to inject steam and oxygen at high velocities will also be demonstrated in this process. Maintaining slagging conditions that affect slag tap (i.e., plugging of tap is apparently a problem).

OTHER FACTORS AFFECTING OVERALL FEASIBILITY:

1. Slightly less CH₄ produced in raw gas than in conventional Lurgi process.
2. Consumes 10% more O₂ than dry bottom Lurgi.
3. Heat is not directly recoverable from molten slag leaving reactor zone.

Process Goals

The Slagging Lurgi development effort is currently entering into the final phase of gasifier development before commercialization. This phase will involve the construction of an 8 foot diameter gasifier which is capable of gasifying 600 to 800 tons/day. Aspects of the testing program for this larger gasifier include:

- Performance testing on a range of British Coals
- Refinements of the fine coal injection system into the gasifier
- A three month continuous test run
- The production of SNG via the BGS's HCM process.

Construction of the 8 foot gasifier is currently underway and is scheduled to be on-line by early 1983. Following Phase III, gasifier development, British Gas will concentrate on the development of downstream technology. British Gas will probably not construct a commercial facility in the near future. They will focus their efforts on selling this technology to other countries.

Relationship to Prior Technology

Prior to the 1930's, town gas was manufactured from caking coals via the simple process of coal carbonization. In the 1930's, research on alternative methods was being explored with the objectives of utilizing cheap non-caking coals and eliminating the formation of byproduct coke to produce an inexpensive high-quality gas. During this period, the Lurgi dry-bottom gasification process was under development in Germany. This early work was the basis for a long term research program conducted by Dr. F. J. Dent for the Gas Council in Great Britain.

From the Lurgi gasifier's conception, it was recognized that the main disadvantage of this technique was the necessity to supply a large excess of steam to gasify the coal's carbon. This was done in order to reduce the combustion zone temperatures below the coal's ash fusion temperature to avoid clinker formation. The higher the ash fusion temperature, the lower the steam requirements were to maintain the ash in a friable condition so that it could be extracted through the mechanical grates. In 1938, while investigating steam and oxygen effects on gasification, Dr. Dent recognized the potential of slagging operation to reduce steam and oxygen requirements.

These studies led to the design of a pilot Lurgi-type gasifier in the year 1949. The pilot slagging gasifier was developed and operated at the British Gas Midlands Research Station (MRS) at Solihull during the period from 1955 to 1964. This work was terminated when it was felt that a full scale demonstration plant was needed in order to prove its commercial feasibility. The Lurgi Company also recognized the advantages that a slagging gasifier could offer. Paul Rudolf or Lurgi experimented with a pilot unit at Holten in the early 1950's.

In 1952, a large deposit of easily mined coal was discovered in Scotland. After successful trials of this coal in Lurgi's test plant in Germany, two Lurgi plants were constructed. One plant was located at Westfield in Scotland and the other at Coleshill in the Scottish Midlands. Construction on the Westfield plant started in the year 1958 and town gas production began in the year 1960. These plants were competitive with the old coal carbonization plants, but could not compete with the emerging oil gasification technology based on naphtha steam reforming. The Catalytic Rich Gas (CRG) process replaced coal carbonization for the production of town gas by the early 1960's. In 1964, the discovery of vast oil and gas deposits in the North Sea further diminished the importance of coal in Britain. The Coleshill facility was shut down in the year 1969 for economic reasons, but the availability of cheaper coal allowed the Westfield plant to remain in operation until the year 1974. During that time, the arrival of natural gas from the North Sea replaced the town gas market.

Because of the Arab oil embargo in the year 1973, the Westfield facility was not closed down entirely. It was recognized that this facility represented a unique opportunity for the western world to develop and demonstrate processes based on Lurgi technology. Further tests were also conducted on a wide range of coals in order to measure their influence on gasifier operation. Because of the relatively low ash fusion temperature of many American coals, research was sponsored to convert one of the Lurgi gasifiers at Westfield for operation under slagging conditions. This development was sponsored by 15 American companies headed by Conoco. The first successful run of a 6 foot diameter gasifier capable of processing 300 to 350 tons per day of coal was completed by April 1975. Further testing and development was successfully completed in the year 1977. At that time Conoco received funds from ERDA/DOE for the design of a commercial demonstration plant.

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I N S T I T U T E O F G A S T E C H N O L O G Y

During this test period British Gas began a formal co-operation agreement with Lurgi, the engineering division of Metallgesellschaft of West Germany. In this agreement, Lurgi offered to help in the design of the top half of the gasifier and British Gas would concentrate its efforts on the design of the bottom half. This teaming effort will lead to the construction of an 8 foot diameter gasifier.

Operating Facilities

British Gas has conducted testing of the Slagging Lurgi gasifier at its Westfield, Scotland, research facilities since 1974. This facility contains three conventional Lurgi gasifiers and one Lurgi gasifier that has been converted to slagging operation. This modified gasifier has an inside diameter of 6 feet and a coal processing capacity of 300 tons per day. Extensive testing of this gasifier has been performed since 1975. Testing of this gasifier was completed in 1981 with a successful trial run of 90 days in which 30,000 tons of coal was gasified to produce over 1,750 million cubic feet of synthesis gas. Construction is currently underway for the modification of one of the other conventional Lurgi gasifiers. This gasifier will have an 8 foot diameter and will be capable of gasifying 600 to 800 tons/day of coal. Construction of the gasifier will be completed in 1983, at which time a 90 day test run will be attempted.

Major Funding Agencies

Experimental testing of the 6 foot diameter Slagging Lurgi gasifier at Westfield has required nearly \$60 million in funding. This money has been obtained from British Gas as well as a U.S. Consortium headed by Conoco, the Electric Power Research Institute (ERPI-USA), Lurgi and the European Economic Community (EEC). Funding for the next stage of development which involve the construction of an 8 foot diameter gasifier at Westfield is estimated to cost nearly \$28 million. The EEC has already committed \$11.2 million to this latest demonstration phase. In addition, British Gas has announced a 20 year development plan, which started in 1978, that includes the demonstration of a range of processes to produce SNG. This program is estimated to cost roughly \$630 million. However, this program has recently been downsided by British Gas.

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Technical Problems

Based on the composite questionnaire responses, the following are the technical problems confronting researchers and developers of the British Gas Slagging Lurgi gasifier:

- Fines (coal particles >0.25 inch in diameter) cannot be gasified. Fines pass through the gasifier and cause downstream tar and deposit problems. Caking coal feed which contains fines seems to be a problem to gasify (when certain types of coal are used).
- The molten slag tends (when certain types of coal are used) to produce molten metallic iron, which is highly corrosive to the metallic elements of the refractory lining of the gasifier and the slag tap. The separation of molten iron from the slag also adversely affects the slag flow characteristics.
- The gasifier operating pressure should be increased, to improve methane production and process efficiency (this affects SNG thermal efficiency).
- The use of tuyeres to inject steam and oxygen at high velocities must be demonstrated over time.
- Plugging of the slag tap must be prevented. This can occur if the slag solidifies due to a temperature drop. The tap hole would then have to be reopened.
- Reduced volume of raw gas to the upper section of the reactor may make drying and pyrolysis of the coal feed more difficult.

In several instances, there were questionnaire responses that conflicted either among respondents or with statements made in the literature. One instance is the question of acceptability of caking coals, and of fines plus coals. At a recent coal technology conference, a BGC official stated that the slagging Lurgi pilot plant is successfully gasifying fines plus coals, both caking and non-caking, for prolonged periods of time. However, questionnaire respondents identified this as a major problem. One respondent stated that, "Operation of the unit for prolonged period on caking coals with fines included, is of 'great' importance to commercial success."

Capital Costs

The projected gas cost for a demonstration plant and a commercial facility producing 250 MMCFD are shown in Table 4 in 1975 dollars. The demonstration plant economics for the production of 60 MMCFD of SNG includes a spare gasifier and does not include all the support facilities required for a self-sufficient operation. For these reasons, a scaling factor should not be

calculated using these two sets of numbers. The installed cost of the gasification reaction section is expected to be less than 10% of the total plant investment. Calculations were based on an 8% DCF rate of return and a 20-year life. Coal cost was \$15.50 per ton of coal for both demonstration and commercial plants. Coal costs for the demonstration plant is slightly less than the commercial facility because of the purchase of off-site power. A byproducts credit was calculated which assumed a naphtha price of \$12 to \$14/bbl and an oil price of \$12/bbl.

Table 4. SUMMARY OF GAS COSTS IN 1975 DOLLARS

<u>Cost Elements</u>	<u>Commercial Plant</u>	<u>Demonstration Plant</u>
	<u>Cost in \$/MM Btu</u>	
Coal	1.185	1.029
Operations & Maintenance	0.714	1.310
Administration	<u>0.065</u>	<u>0.163</u>
Subtotal	1.955	2.502
Depreciation and Return On Investment	1.085	1.529
Income Taxes	<u>0.661</u>	<u>0.998</u>
Total Costs	3.701	5.029
Byproduct Credits	<u>(0.243)</u>	<u>(0.243)</u>
Net Cost of Gas	3.458	4.786

32(3)/process/ER

BRITISH COAL GASIFICATION PROCESS

16. British Gas Composite Coal Gasification ProcessProcess Description

The composite gasifier is being developed by British Gas Corp. as an extension of their research on the Slagging Lurgi gasifier. This research is being motivated by the higher production of fines (up to 60%) in modern underground mining techniques and the historic inability of conventional Lurgi gasifiers to accept a high fines content feedstock. In order to overcome this fines content limitation British Gas has proposed the integration of a pressurized entrained-flow gasifier for coal fines coupled in a proprietary way in series below the lump coal Slagging Lurgi gasifier. In this manner the large sensible heat content of the hot raw gas leaving the entrained stage would provide sensible heat to the endothermic gasification reactor in the slagging stage. This direct coupling approach would minimize overall oxygen consumption in both stages as well as efficiency penalties resulting from the use of two gasification zones.

Process Goals

The goal of this project is to increase the utilization of coal fines in the Slagging Lurgi Process through the coupling of an entrained-flow gasifier to the slagging gasifier. This composite gasifier process could then accept run-of-mine coal feedstock.

Relationship to Prior Technology

This technology is a combination of conventional fixed-bed and entrained-bed design which incorporates the Slagging Lurgi gasification process which British Gas Corporation has been developing over the past 10 years.

Operating Facilities

The concept for this composite gasifier is based on tests that were carried out at the Coal Utilization Research Laboratory (CURL), Leatherhead, in 1962. A pilot plant with a 20 ton/day capacity has been designed, but recent decisions by British Gas to stop research on the composite gasifier has cancelled plans for its construction.

Major Funding Agencies

The British Gas Corporation has funded the preliminary research on the composite gasifier concept. However, recent tests of the Slagging Lurgi gasifier have indicated that fines content of the coal feedstock can be increased to above 30% without affecting gasifier performance. For this reason research on the composite gasifier has been terminated.

BRITISH COAL LIQUEFACTION PROCESS

17. Natural Coal Board Supercritical Gas Solvent Extraction Coal Liquefaction ProcessProcess Description

The supercritical gas extraction process is a two stage process designed for the production of transportation fuels and chemical feedstocks. This process is based on the ability of compressed gases, acting as a solvent, to dissolve large quantities of high molecular substrates in coal. In theory the solvent property of the gas increases with its density. This will be at a maximum at the gas' minimum reduced temperature, for a given pressure. Solvent gas selection is based on the gases critical temperature which should be slightly below the liquefaction reactor operating temperature. High extraction rates can be achieved when the substrate has a low vapor pressure and the solvent gas has a density and viscosity which is significantly lower than a corresponding liquid phase solvent.

With proper selection of the gas phase solvent the following advantages will occur:

- Liquid products can be extracted from coal in the temperature range of 673°K to 723°K without undergoing secondary repolymerization reactions
- Separation of undissolved solid residue is less difficult in a gas phase reaction than when using a liquid phase reaction
- Separation of liquid product and solvent is simple and efficient due to the large differences in their volatility.

The preferred gas to use for solvent extraction of coal should have a critical temperature in the range of 573°K to 673°K.

A pilot plant with a capacity of 5 kg/hr was built in 1977 to test the performance of this process. Based on this testing a 25 ton/day facility was designed. A simplified flow diagram of this facility is presented in Figure 1. In this facility coal will be received by truck and stored uncrushed on the ground. Storage capacity will be roughly 600 tons of m.a.f. coal. The coal is then crushed to below 3 mm before being dried with hot flue gases from a fired boiler. Coal preparation in this plant differs from that of the 5 kg/hr PDU which utilized crushed coal with a nominal — 210 μ m coal feed due to plant pipework size. In the proposed 25 ton/day pilot plant the

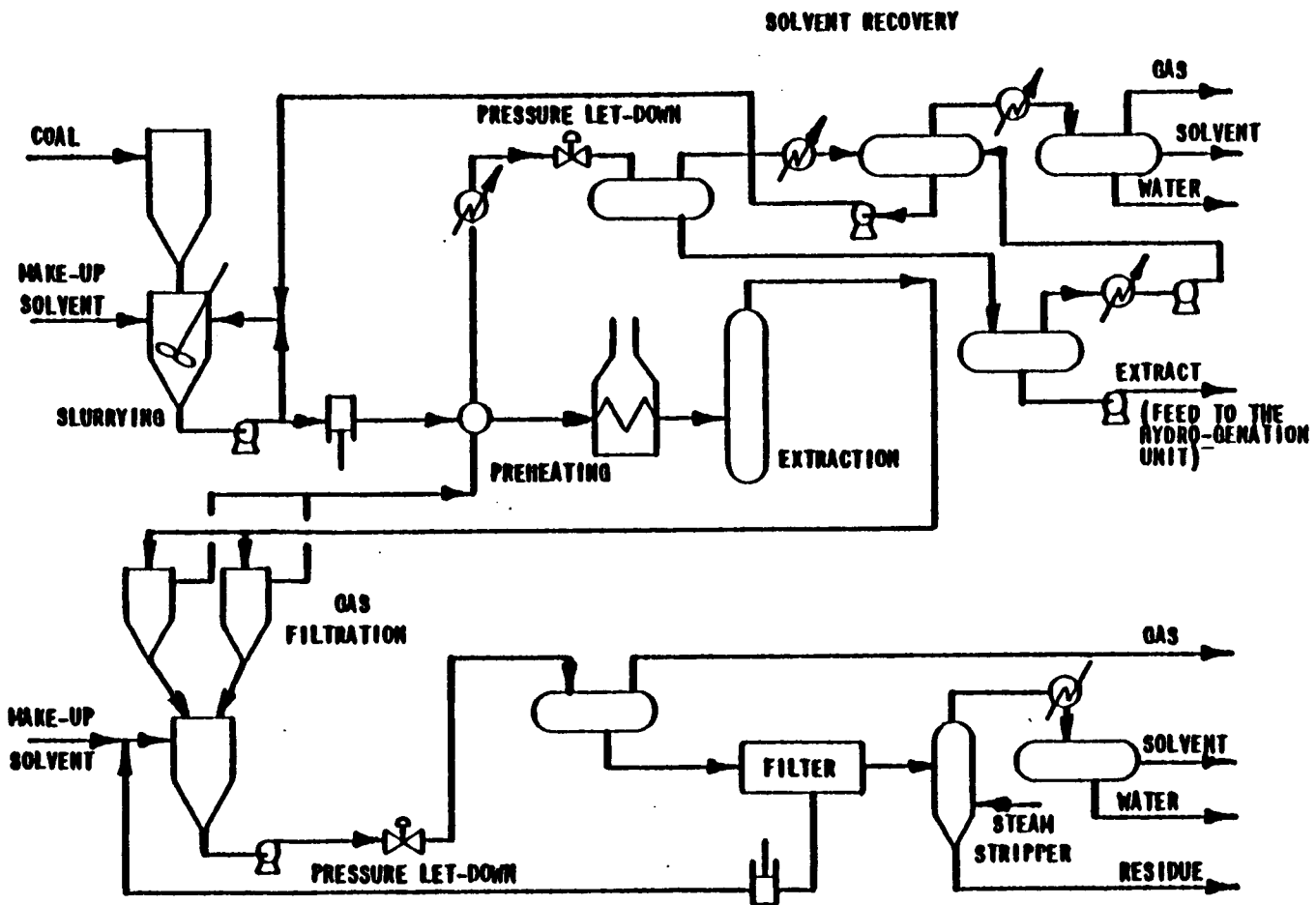


Figure 1 SUPERCRITICAL GAS EXTRACTION PROCESS

coal is then screened and fed into a surge bin before being pneumatically transported to a storage silo. This silo will be sized to contain a three day supply of coal.

From the silo, coal will be continuously fed to a slurry mixing tank where it will be mixed with 5 parts of hot recycled solvent (toluene was used in the PDU). From the slurry tank centrifugal circulating pumps feed the coal to a high pressure metering pump. This slurry is then heated to 750°F before it enters a fluidized bed reaction vessel at a pressure of 2900 psig. At this point the solvent in the slurry is above its critical temperature. Both a fluidized bed and entrained flow reactor are incorporated in the plant design, as was the case for the 5 kg/hr PDU. Mean residence time within these vessels is on the order of minutes.

In the reactor vessels the coal is desolved to form a lower molecular weight liquid (coal extract) and a solid particulate residue. The extract formed in the reactor is completely desolved in the supercritical gas solvent. Approximately 40 to 50% of the m.a.f. coal is desolved by the solvent. The extract leaving the top of the reactor vessel is passed through a sintered filter to remove particulates and then sent to a slurry feed heat exchange. Upon exiting the heat exchanger the extract passes through a pressure letdown valve.

Solvent extraction studies by British Gas indicate that approximately half of the extract has an average molecular weight of 300 to 600. This portion of the extract can be obtained at relatively mild conditions and increased in yield with longer reaction times. The remainder of the extract consists of benzene-insoluble fractions with an average molecular weight of 600 to 1200. This fraction increases in yield with reaction time but passes through a maximum as reaction temperature increases. A peak is reached due to the recombination of larger molecular weight molecules which form insoluble species. For this reason all of the benzene-insoluble fraction cannot be removed from the coal. In bench scale and PDU tests little variation in the yield or composition of the minor paraffin fraction were noticed as extraction conditions were varied. Over 95% of the extract consists of aromatic fractions. These fractions are predominately open structures with single aromatic rings joined by methylene and ether bridges, by hydroaromatics, aromatic and indene rings and by biphenyl linkages. Short alkyl groups occupy

about 1/4 of the available positions on the aromatic structure and another 1/10 are occupied by phenolic hydroxyl groups. The proportion of these linking structures remain nearly constant regardless of the extracting conditions.

The solid residue which settles in the bottom of the extraction reactor is continuously removed as a free-flowing slurry and sent to a slurry vessel. The residue is then depressurized and steam stripped to remove the remaining solvent which is recycled to the process. The dried residual char is maintained as discreet particulates and is porous in nature. Tests have indicated that the reactivity of this char is the same as that of the coal feedstock and therefore can be used as an energy source for hydrogen or power generation.

The hot filter extract exiting the top of the reactor is flashed to a low pressure in a flash drum. A majority of the solvent is recovered in the overhead vapor stream. The coal extract which still contains some residual solvent is removed in the underflow. This underflow then passes to a vacuum unit where the remaining solvent is substantially removed. The overheads, contain solvent is condensed to the liquid state and recycled, at just below the boiling point, to the slurry preparation unit. The non-condensed vapors are further cooled in a cold water condenser where gases and liquids are formed. The gaseous portion is flared and liquid condensate is sent to the waste solvent tank or a waste water treatment facility.

After solvent recovery the coal extract is mixed with recycle oil, pressurized, and sent to the second stage hydrotreating section. A diagram of this section is shown in Figure 2. The extract entering the hydrogenation section is heated to 750°F to 840°F and pressurized to greater than 2900 psi. Recycled impure hydrogen is added to the high pressure liquid, resulting in two-phase flow through the heat exchange and furnace heating zones. The hydrogen is scrubbed to remove some of the sulfur, nitrogen and chlorine compounds prior to injection. Makeup hydrogen for the proposed 25 ton/day pilot plant will be obtained by steam reforming natural gas. However, a commercial size facility will probably generate hydrogen by gasifying a portion of the solid residual char.

The hot extract solution and hydrogen gas are passed through a guard reactor to remove hydrocracker catalyst poisons. Two or more of these units will be operated in parallel to allow for continuous operation when one unit is regenerated.

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I N S T I T U T E O F G A S T E C H N O L O G Y

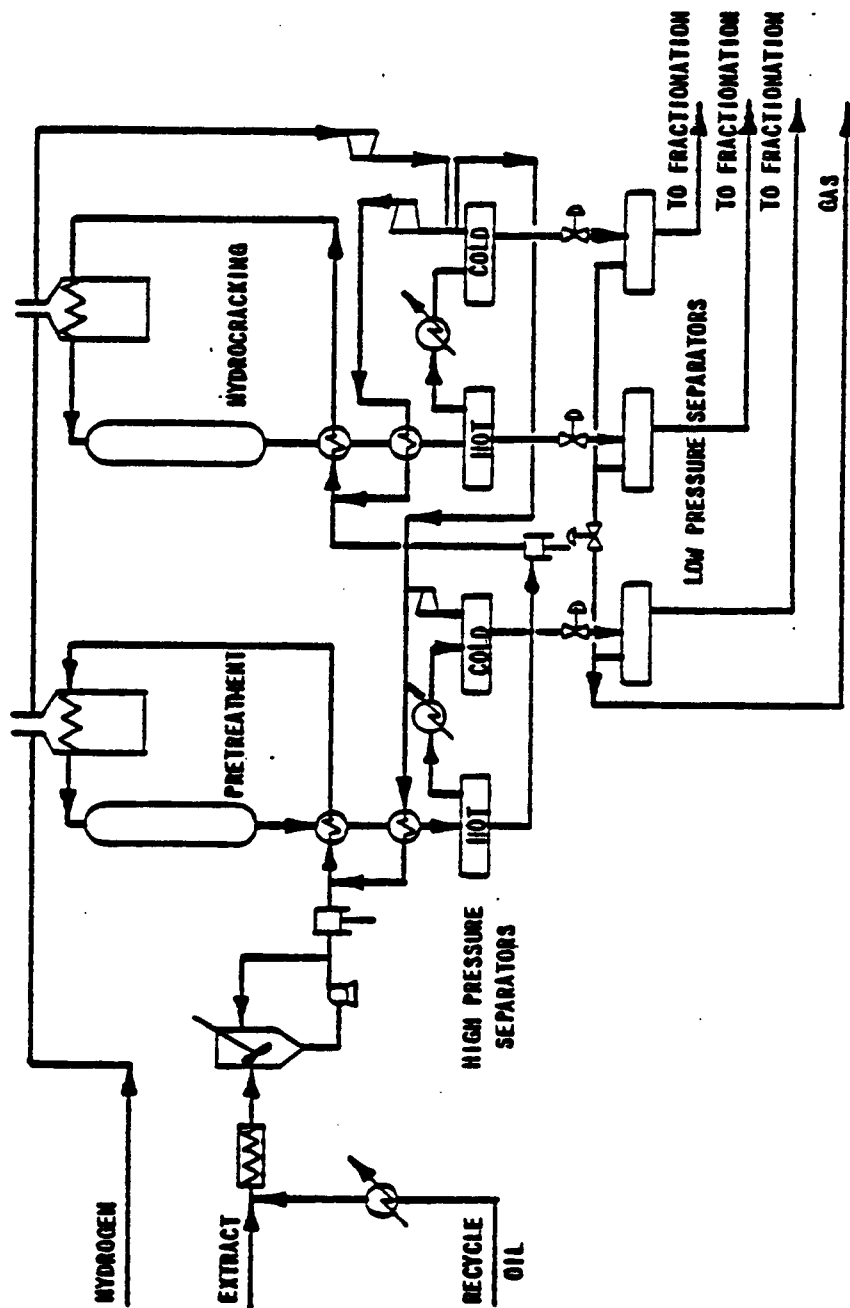


Figure 2. HYDROGENATION FOR SUPERCRITICAL GAS EXTRACTION PROCESS

The primary hydrocracking reactors are of conventional trickle bed design. A number of catalyst beds are used to limit temperature rise to a maximum of 45°F in each bed. Intercooling between beds with recycle hydrogen is also used to limit the temperature rise. Facilities are also included to regenerate the catalyst material.

Effluents from the hydrocracker are cooled by heat exchange with extract feed and are sent to a high pressure separator. Hydrogen rich gases containing some methane and other gases are recycled to the hydrocracker units after repressurization and scrubbing. Liquid bottoms are sent to the fractionation system. This system contains a vacuum column as well as normal atmospheric pressure crude fractionation units. The principle cuts include gases, C₅ (345°F) liquids, 345 to 480°F boiling range liquids and 480°F plus liquids. The primary product cuts are the two lower boiling point liquids. An additional cut will also produce makeup solvent. A block flow diagram and material balance for a 10,000 tonne/day coal feed commercial plant is shown in Figure 3. This plant is expected to have a thermal efficiency of 67.9%. A technology fact sheet for this process is presented in Table 1.

Process Goals

The supercritical gas extraction process is being developed as part of a two-stage coal liquefaction process which involves extraction and product upgrading for the production of transportation fuels and chemical feedstocks. Testing in a continuous 5 kg/hr PDU has been performed to determine operating parameters and performance. Different supercritical fluids are being investigated for their extraction capabilities. These include water, toluene, benzene and other solvents which are supercritical in the temperature range of 573 to 673°K. Continued research is also being conducted in extract upgrading. The overall goal of this program is to establish the potential of this process to operate on a commercial basis in an integrated/self-sufficient mode. Future plans which called for the construction of a 25 tonne/hr pilot plant have been cancelled by British Gas. However, research on this process will continue on a small scale.

Relationship to Prior Technology

The ability of compressed gases to act as solvents was first noticed by Hanney and Hogarth as far back as 1897 in an article appearing in the

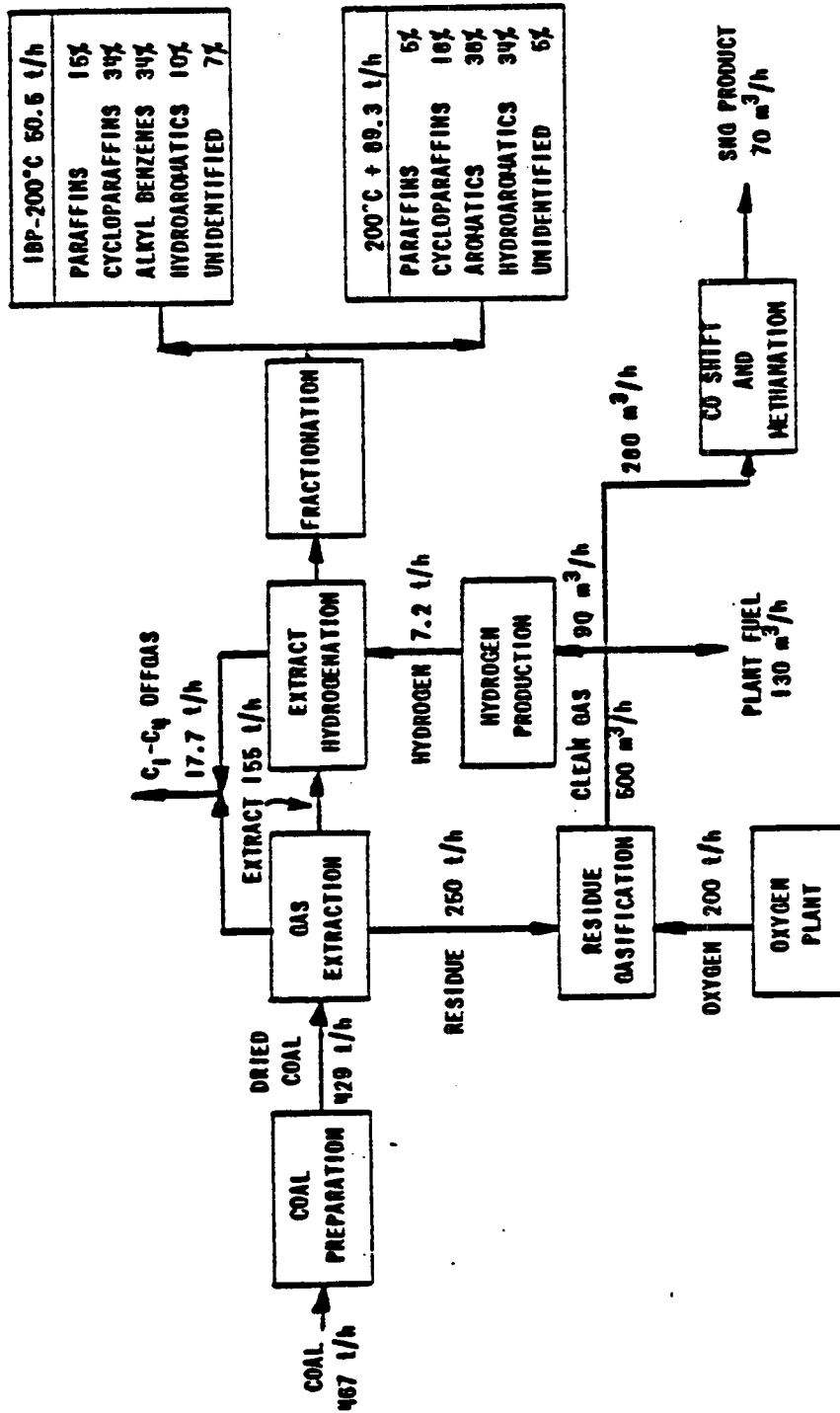


Figure 3. OVERALL FLOW DIAGRAM AND MATERIAL BALANCE FOR SUPERCritical GAS EXTRACTION PROCESS

Table 1 Part 1 TECHNOLOGY FACT SHEET: Extraction Coal Liquefaction
 NCB Supercritical Gas Solvent

NEW TECHNOLOGY (PROCESS DESCRIPTION AND GOALS): The SCGE process is a two stage process designed for the production of transportation fuels and chemical feedstocks. This process is based on the ability of compressed gases, acting as solvents, to dissolve large quantities of high molecular substrates in coal.

RELATIONSHIP TO PRIOR TECHNOLOGY (INCLUDING STATE OF DEVELOPMENT OF PRIOR TECHNOLOGY): Based on supercritical gas extraction techniques applied to deasphalting of petroleum fractions.

CHARACTERISTICS OF THE TECHNOLOGY:

<u>CHARACTERISTICS</u>	<u>NEW TECHNOLOGY</u>	<u>PRIOR TECHNOLOGY</u>
PRIMARY OUTPUT (DESIGN CASE).....	liquid fuels	
TYPE OF PROCESS.....	supercritical gas extraction	
FEEDSTOCK REQUIREMENTS.....	coal < 210 μm	
OVERALL THERMAL EFFICIENCY (INCLUDING BY-PRODUCTS)..	69.7%	
CARBON CONVERSION EFFICIENCY.....	40 to 50%	
OPERATING TEMPERATURE.....	750°F	
OPERATING PRESSURE.....	2900 psia	
BY-PRODUCTS.....	gas, pitch, char	

**Table 1 Part 2 TECHNOLOGY FACT SHEET: NCB Supercritical Gas Solvent
Extraction Coal Liquefaction**

STATUS OF DEVELOPMENT:

OPERATING FACILITIES - A 120 kg/hr continuous feed PDU started operation in 1977 at the
NCB's Coal Research Establishment's Stoke Orchard facility.

MAJOR FUNDING AGENCY National Coal Board

ANNUAL LEVEL OF FUNDING -

TECHNICAL PROBLEMS: Scale up from the current 0.1 tonne/day PDU to a proposed 25 tonne/day.

OTHER FACTORS AFFECTING OVERALL FEASIBILITY: Low extract production.

Proceedings of the Royal Society of London. The enhanced solvent properties of polar gases in the supercritical region was demonstrated by Hagenback in Leipzig in 1901. Work by Frank (1956) investigated the solvent properties of supercritical water and the investigation of Eisenbeiss (1964) and Weale (1967) demonstrated the solubility of solids in compressed nonpolar and polar gases. The solvent properties of supercritical fluids was applied to the deasphalting of petroleum fractions even before 1950. Work at the National Coal Board's Coal Research Establishment at Stokes Orchards in the mid-1960's focused on liquids recovery from coal when heated above 750°F. In 1971 Paul and Wise in London described the principles of gas extraction as a means of liquefying coal. This technique has undergone further research by the National Coal Board in various size test units since the early '70's.

Operating Facilities

All work on the supercritical gas extraction process is being carried out at the National Coal Board's coal research Establishment facilities, Stoke Orchard, near Cheltenham. This research has progressed from a benchscale unit to the construction and operation of a 5 kg/hr continuous feed process development unit. This unit started operation in 1977. Design of a 25 tonne/day pilot plant have been completed which was to be located at the Point of Ayr in North Wales. The National Coal Board has recently made the decision not to construct this pilot plant. It is not certain as to whether or not research on this process will continue.

Major Funding Agencies

The National Coal Board has sponsored all funding for this process. However, the NCB has recently announced that it will shelve this process as well as their Liquid Extraction Process.

Technical Problems

The major technical problems facing this technology is scale-up of the current 0.1 tonne/day PDU to the proposed 25 tonne/day pilot plant.

Capital Costs

Capital costs for a commercial scale facility have not been published. However, the capital costs for the 25 tonne/day pilot plant have been estimated at £ 14.8 m in 1978.

BRITISH COAL LIQUEFACTION PROCESS

18. National Coal Board Liquid Solvent Extraction Coal Liquefaction ProcessProcess Description

The National Coal Board's Liquid Solvent Extraction (LSE) process utilizes anthracene and coal derived solvents to liquefy coal at a temperature of 420 to 450°C and at 20 bars pressure. The process actually consists of two stages, the liquefaction stage, and the hydrotreating stage. In the liquefaction stage the solvent also acts as a mild hydrogen donor. A process flow diagram for the liquefaction and hydrotreating sections is presented in Figures 1 and 2.

In this process run-of-mine coal is fed to the coal preparation section where it is cleaned and dried. From here it is sent to a pulverizer where it is reduced to less than 0.2 mm in size. All coals, except for anthracite, can be used as feed regardless of caking properties. However, extract yields can vary widely depending on the coal feed. A cleaned fraction of coal containing approximately 6% ash is then fed to the slurry preparation section. The remaining diameter coal is sent to a gasifier for hydrogen generation or to the boiler for steam and power generation.

The pulverized coal entering the slurry preparation section is mixed with a high boiling solvent, recycled from the hydrocracking unit, in the ratio of 3.5 ton of solvent to one ton of coal. This slurry is then pumped to a pressure of less than 20 bars and sent to the preheat section. This section consists of a heat exchanger where the slurry is heated by the liquefaction digester effluent, and a fired heater. The slurry enters the digester at 400°C where it is reacted for 30 to 90 minutes. Under optimum conditions as much as 95% of the coal's non-mineral matter can be dissolved. During reaction the solvent acts as a mild hydrogen donor to increase the extract hydrogen content as much as 2 percent. The effluent from the digester is then sent to the feed preheat exchanger for cooling and then into a gas/liquid separation vessel.

In this flash vessel the pressure of the slurry is reduced and the lighter products are separated. The overhead stream is then cooled to condense out the C₅-250°C fraction and water. The gaseous product can then be recycled or sold while the liquid bottoms from the condenser are separated to

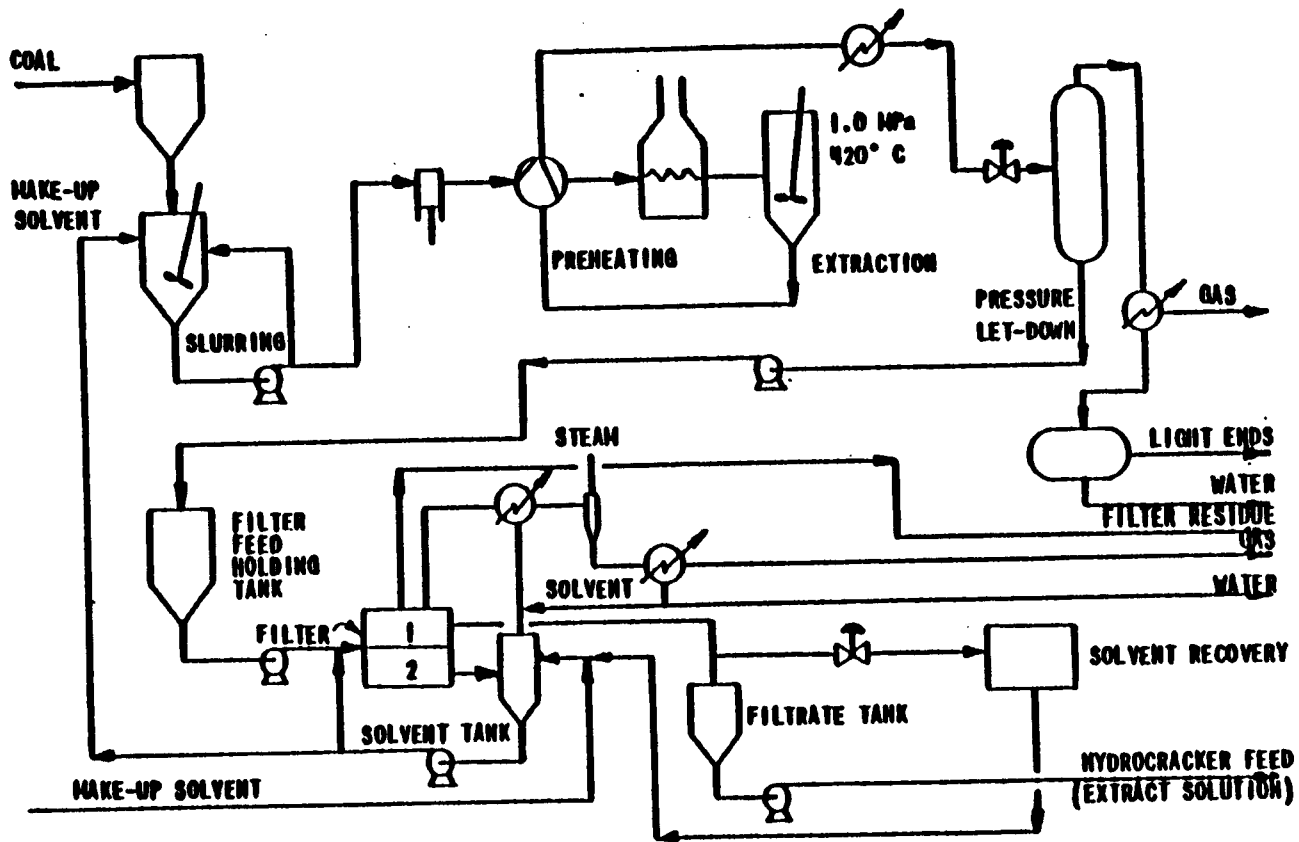


Figure 1. LIQUID SOLVENT EXTRACTION PROCESS SECTION

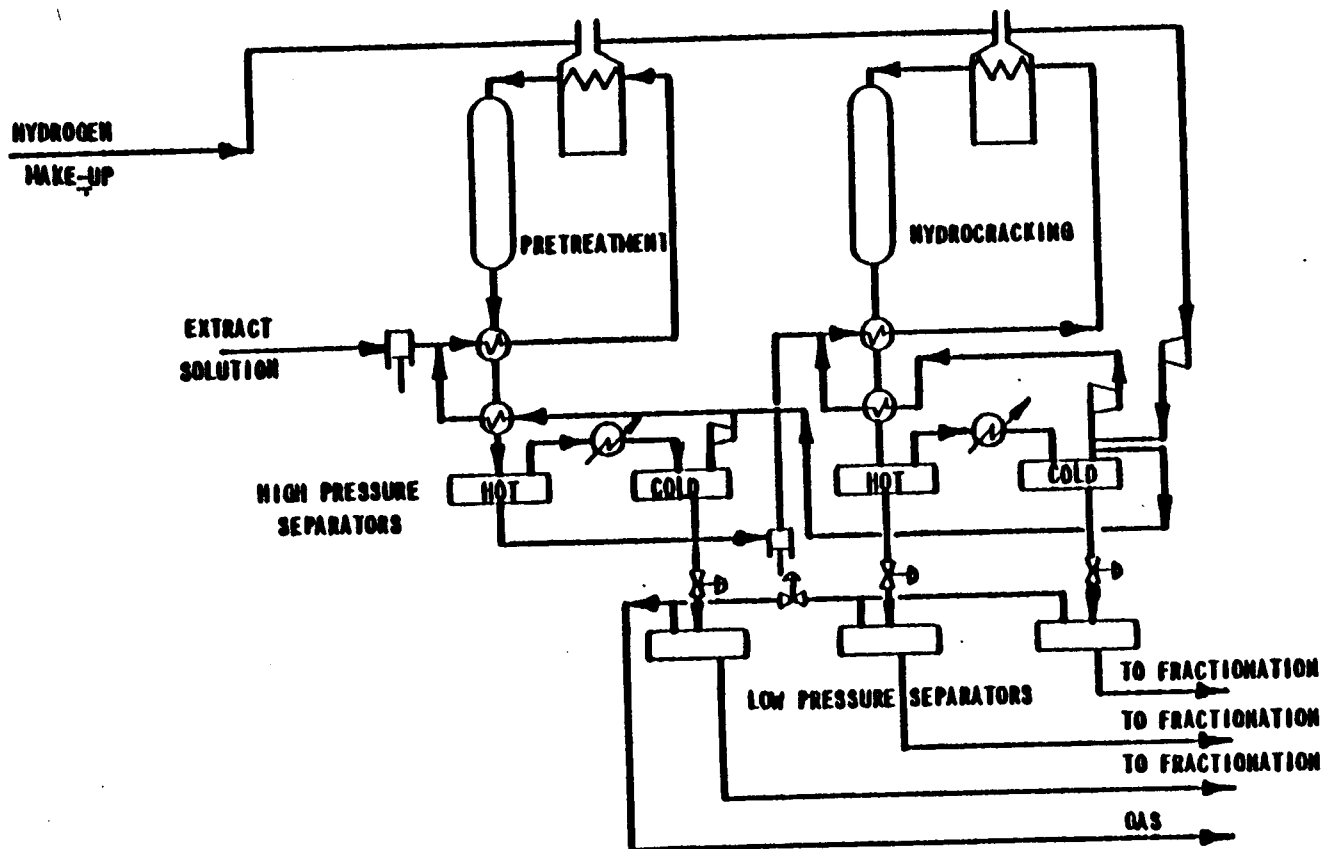


Figure 2. HYDROGENATION UNIT FOR LIQUID SOLVENT EXTRACTION PROCESS

remove the water. The liquid bottoms from the flash vessel, which contain ash, are sent to a holding tank where they are fed batch-wise into a commercially available filter. The dried filtrate is then sent to the gasifier for hydrogen generation and the filtered coal extract is sent to storage.

From storage the coal extract is mixed with recycled oil to adjust its concentration before it is pressurized and sent to the second stage hydro-treating section shown in Figure 2. This mixture entering the hydrogenation section is heated to 750 to 840°F and pressurized to greater than 2900 psi. Recycled impure hydrogen is added to the high pressure liquid, resulting in two-phase flow through the heat exchange and furnace heating zones. The hydrogen is scrubbed to remove some of the sulfur, nitrogen and chlorine compounds prior to injection. Make-up hydrogen for the proposed 25 ton/day pilot plant will be obtained by steam reforming natural gas. However, in a commercial size facility much of the required hydrogen will be generated by a gasifier from recycled filtrate, pitch and dirty coal.

The hot extract and hydrogen gases are passed through a guard reactor to remove hydrocracker catalyst poisons. Two or more of these units will be operated in parallel to allow for continuous operation when one unit is regenerated.

The primary hydrocracking reactors are of conventional trickle bed design. A number of catalyst beds are used to limit temperature rise to a maximum of 45°F in each bed. Intercooling between beds with recycled hydrogen is also used to limit the temperature rise. Facilities are also included to regenerate the hydrocracker catalyst.

Effluents from the hydrocracker are cooled by heat exchange with extract feed and are sent to a high pressure separator. Hydrogen rich gases containing some methane and other gases are recycled to the hydrocracker units after repressurization and scrubbing. Liquid bottoms are sent to the fractionation system. This system contains a vacuum column as well as atmospheric pressure crude fractionation units. The principle cuts include gases, C₅(345°F) liquids, 345 to 480°F boiling range liquids and 480°F plus liquids. The primary product cuts are the two lower boiling range liquids. The 480°F + cut is used as recycle solvent. Alternatively, 700°F + cut can be produced and fed to a thermal cracker for production of a premium cut. The material balance for this process is shown in Figure 3. A process technology fact sheet summarizing the process is shown in Table 1.

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I N S T I T U T E O F G A S T E C H N O L O G Y

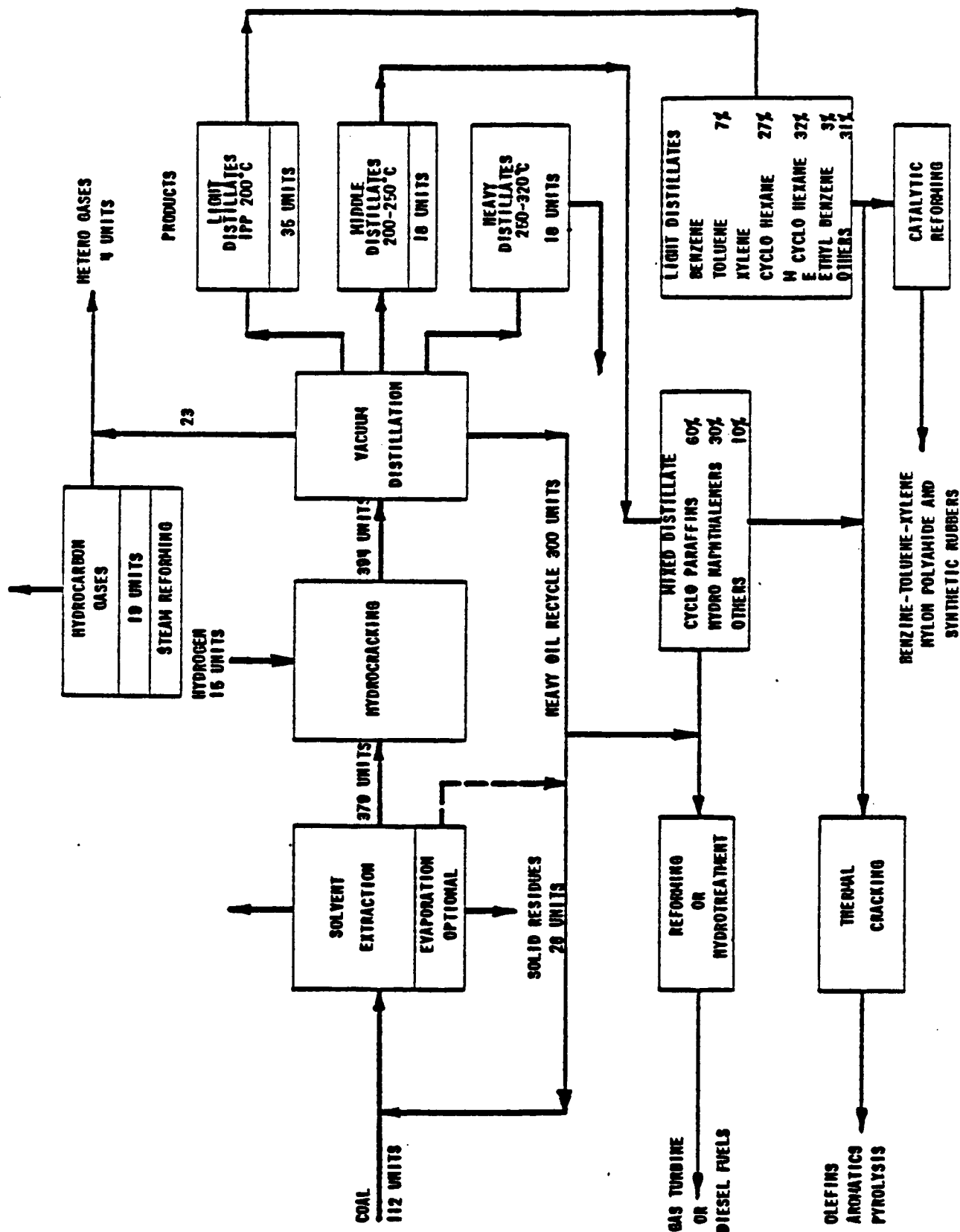


Figure 3. OVERALL FLOW DIAGRAM AND MATERIAL BALANCE FOR THE LIQUID SOLVENT EXTRACTION PROCESS

National Coal Board Liquid Solvent Extraction
Coal Liquefaction Process

Table 1 Part 1 **TECHNOLOGY FACT SHEET:**

NEW TECHNOLOGY (PROCESS DESCRIPTION AND GOALS): The National Coal Board's Liquid Solvent Extraction(LSE)

Process utilizes anthracene and coal derived solvents to liquefy coal at a temperature of 420 to 450°C and at 20 bars pressure to produce a liquid transportation fuel. The process actually consists of two stages, the liquefaction stage and the hydrotreating stage.

RELATIONSHIP TO PRIOR TECHNOLOGY (INCLUDING STATE OF DEVELOPMENT OF PRIOR TECHNOLOGY): This process

has developed from studies carried out at the NCB's Coal Research Establishment's Stoke Orchard facilities in the 1960's. This work concentrated on the production of electrode coke from carbon products.

CHARACTERISTICS OF THE TECHNOLOGY:

<u>CHARACTERISTICS</u>	<u>NEW TECHNOLOGY</u>	<u>PRIOR TECHNOLOGY</u>
PRIMARY OUTPUT (DESIGN CASE).....	liquid fuels	
TYPE OF PROCESS.....	liquid solvent extraction	
FEEDSTOCK REQUIREMENTS.....	coal sized < 0.2 mm	
OVERALL THERMAL EFFICIENCY (INCLUDING BY-PRODUCTS)..	60 to 68%	
CARBON CONVERSION EFFICIENCY.....	95%	
OPERATING TEMPERATURE.....	420 to 450°C	
OPERATING PRESSURE.....	20 bars	
BY-PRODUCTS.....	gas, pitch	

Table 1 Part 2

TECHNOLOGY FACT SHEET: National Coal Board Liquid Solvent Extraction
Coal Liquefaction Process

STATUS OF DEVELOPMENT:

OPERATING FACILITIES - A 66 lb/hr PDU has undergone testing at the NCB's Coal Research Establishment laboratories in Stoke Orchard near Cheltenham, U.K.

A 25 tonne/day pilot plant is proposed but no plans exist to build this facility.

MAJOR FUNDING AGENCY National Coal Board, International Energy Agency, Phillips Petroleum

ANNUAL LEVEL OF FUNDING - 25 ton/day pilot plant will cost \$100 million.

TECHNICAL PROBLEMS: Technical problems include solid/liquid separation material, and hydrocracking catalyst development.

OTHER FACTORS AFFECTING OVERALL FEASIBILITY:

The overall process efficiency and product slate varies with coal feed type and end product requirements. Tables 2 and 3 present the product mix when using a bituminous coal or lignitic coal. Table 2 also presents process efficiencies for different plant configurations when pitch is used as a gasifier feed, SNG byproduct is recycled for process gas, and when both pitch and SNG are recycled. Overall process efficiencies can vary from 68% in the base case using bituminous coal to 60.2% when operating on a lignitic coal with 23% moisture and 20% ash.

Process Goals

The liquid solvent extraction process is being developed as part of a two-stage coal liquefaction process which involves extraction and product upgrading for the production of transportation fuels and chemical feedstocks. Testing in a 66 lb/hr PDU has been performed to determine operating parameters and performance. Continued research is also being conducted in the extract upgrading stage. The overall goal of this program is to establish the potential of this process to operate on a commercial basis in an integrated self-sufficient mode. Future plans call for the construction of a 25 tonne/day pilot plant. However, construction of this pilot plant has recently been cancelled by the National Coal Board.

Relationship to Prior Technology

Development of the Liquid Solvent Extraction (LSE) process started in the early '60's at the National Coal Board's (NCB) Coal Research Establishment (formerly called the British Coal Utilization Research Laboratories) in Stoke Orchard, near Cheltenham. This work centered on the extraction of coal liquids by anthracene and other tar oils to produce carbon products and electrode coke. A 66 lb/hr process development unit was constructed in the early '70's to make electrode cokes to be used in the steel and aluminum smelting furnaces. This PDU is the basis for the proposed construction of a 25 tonne/day pilot plant. In general this process is similar to the Solvent Refined Coal (SRC) process which also utilizes a coal derived solvent to dissolve coal at high temperatures and moderate pressures.

Operating Facilities

All research on the Liquid Solvent Extraction process has been carried out at the NCB's Coal Research Establishment facilities in Stoke Orchard near

Table 2. EXAMPLE OF INTEGRATED LIQUEFACTION PLANT AND REFINERY
PROCESSING A BITUMINOUS COAL

	Basic case	Pitch used as gasifier feedstock	SNG used for process gas	Both pitch and SNG used in process
<u>Product Slate</u> (ton/100 ton d.a.f. coal)				
Gasoline	9	9.5	10	11
Diesel and jet fuel	23	25	27	30
Pitch	6	0	7	0
LPG	4	5	5	6
Substitute natural gas	9	9	0	0
<u>Coal input</u> (ton d.a.f./barrel transport fuels produced)				
- to process	0.25	0.25	0.25	0.25
- to gasifier	0.14	0.10	0.08	0.04
- to boiler	0.03	0.03	0.03	0.03
Total:	0.42	0.38	0.36	0.32
Barrels liquid fuels/ ton d.a.f. coal	2.4	2.6	2.8	3.1
<u>Overall thermal efficiency</u> (%)	68	66	65	63

Table 3. EXAMPLE OF INTEGRATED REFINERY PROCESSING A LIGNITIC COAL
(23% moisture, 20% ash)

<u>Product Slate</u> (ton/ton d.a.f. coal)	
Gasoline	16
Diesel and jet fuel	19
Pitch	0
Substitute natural gas	0
LPG	2.3
<u>Coal input</u> (ton d.a.f./barrel transport fuels produced)	
- to process	0.27
- to gasifier	0.07
- to boiler	0.05
	0.39
Barrels liquid fuels/ton d.a.f. coal	2.6
<u>Overall thermal efficiency</u>	60.2

Cheltenham. This research has progressed from bench scale studies in the '60's and early '70's to the construction of a 66 lb/hr PDU in the mid-'70's. Photographs of the 1st stage PDU liquefaction unit and the 2nd stage hydrotreating section are shown in Figures 4 and 5, respectively. Future plans called for the construction of a 25 tonne/day pilot plant to be located at Point of Ayr, North Wales. This project has recently been cancelled.

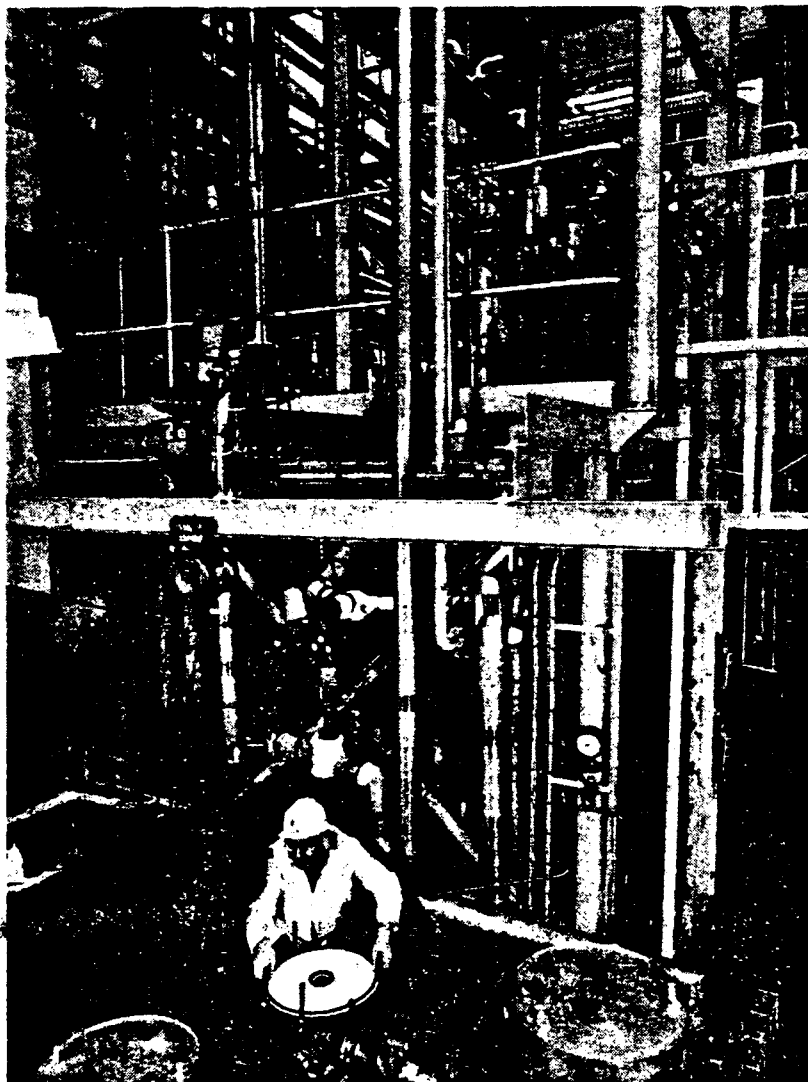


Figure 4. SOLVENT EXTRACTION PLANT AT THE COAL RESEARCH ESTABLISHMENT

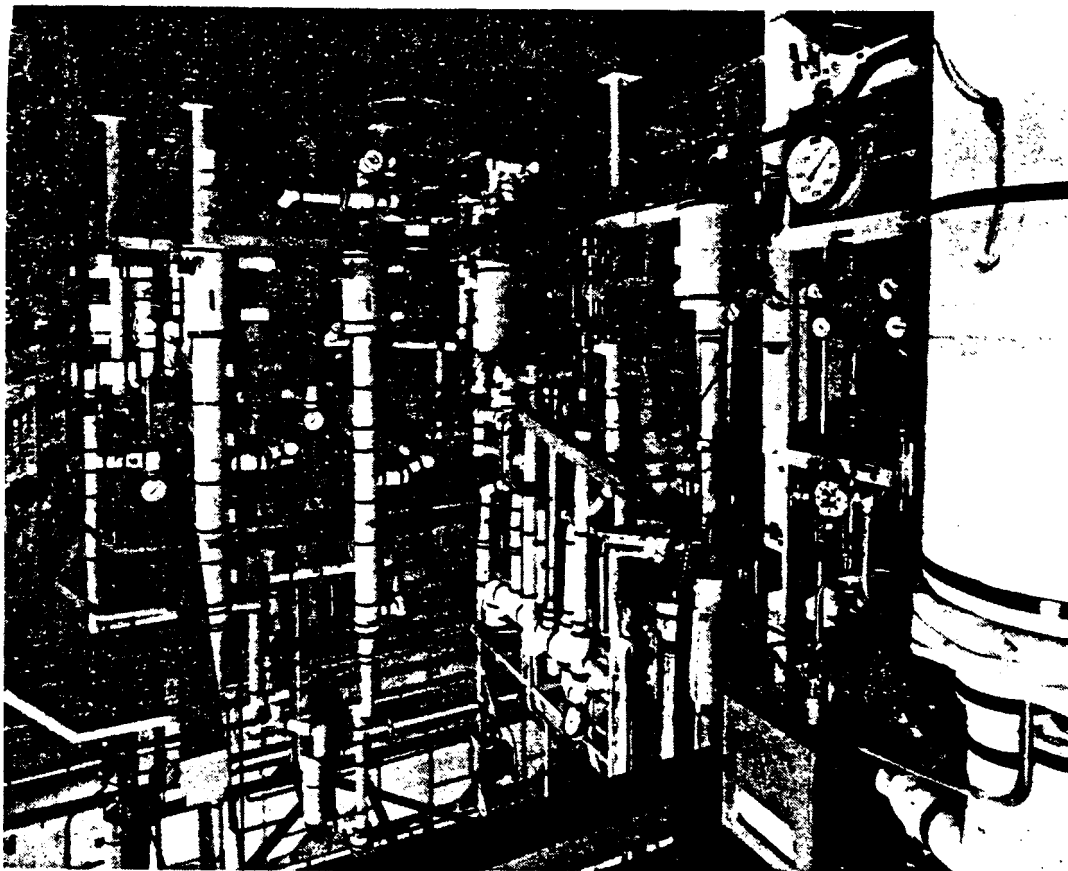


Figure 5. HYDROCRACKING PILOT PLANT OF THE COAL RESEARCH ESTABLISHMENT

Major Funding Agencies

The National Coal Board has sponsored all funding for this process to date. Funding for the \$100 million 25 tonne/day pilot plant would have come from the National Coal Board, Commission of the European Communities (\$10 million) and Phillips Petroleum. Recently British Petroleum which was also a co-sponsor withdrew their support to the project. If pilot plant testing had been successful, future plans called for the construction of a 1,000 ton/day semi-commercial plant in an overseas market in the late '80's.

Technical Problems

The two areas of most concern are the solid/liquid separation section and the catalytic hydrocracking/extract upgrading section. In the filtration section hot batch filtration has been tested in a 1100 lb/day unit used for electrode coke production. However, a continuous unit has not been tested. The batch unit was able to reduce ash content in the liquid extract to below 0.1 percent. Although this type of hot filtration is more expensive than other separator techniques it is anticipated that the filtration section will still account for less than 10% of the capital costs. In the PDU a glass fibre cloth with a particle size retention of 0.5 μm was used successfully, but its weakness and lack of rigidity make it unsuitable for the continuous high temperature operation required in a commercial facility. An alternative precoated wire screen method has shown promise in experimental runs but screen size and precoat material selection has yet to be optimized. Alternative precoat materials to Celite are being investigated which are made of carbonaceous materials. The spent precoat containing ash can then be sent to the gasifier or combustion section for disposal, thereby decreasing plant waste.

Research on the hydrocracking section has focused on catalyst selection. Over 50 different catalysts have been tested in a stirred autoclave for their lifetime, resistance to contaminants, and kinetic properties. Alternatives to catalyst poisoning include designing an extract cleaning system to remove deactivating agents before they reach the catalysts.

Capital Costs

Capital costs for a commercial scale LSE process plant have not been published. The capital cost of the 25 tonne/day pilot plant was expected to be \$100 million.

BRITISH DIRECT COAL COMBUSTION PROCESS

19. National Coal Board Pressurized Fluidized Bed Coal Combustion ProcessProcess Description

The Pressurized Fluidized Bed Coal Combustion (PFBC) process is being developed for combined gas/steam cycle utility power generation. Research in this area is being conducted by the National Coal Board (NCB) in a 5 MW test facility at Leatherhead. A typical flow diagram of a PFBC combined cycle utility power plant is shown in Figure 1. The concept involves burning coal in a fluidized-bed of calcium-containing mineral such as limestone to absorb the polluting sulfur in the coal. The sulfated limestone is removed from the combustor and discarded for land fill applications or regenerated for elemental sulfur recovery and reuse in the combustor. In the pressurized combustion concept, the generated hot flue gases are expanded through a turbine to generate electricity.

The fluidized-bed boiler furnace section consists of an enclosed space with a perforated base to admit combustion air. Part of the enclosed space directly above the air distribution plate is occupied by a layer of granular material such as sand or limestone. The air, which is forced through the supply plenum is sufficient to lift (fluidize) the bed and suspend it in the airstream. This promotes violent boiling (mixing) and agitation of the bed material. After fluidization has occurred the combustible fuel is introduced and ignited. The bed material absorbs the heat of reaction and transfers as much as 50% of it to immersed water tubes and waterwalls. The waterwall tubes, which surround the inner fluidized-bed walls, maintains the wall temperatures within a safe operating range. The optimum operating temperature is between 1500° and 1700°F. When a high moisture content fuel such as wood wastes or municipal refuse is burned, the heat required to evaporate the moisture in fuel maintains the bed temperature within acceptable limits without the use of waterwall cooling tubes.

Because of the enhanced heat transfer characteristics of the fluidized bed, the unit is smaller than a conventional boiler of the same output. In order to take advantage of the heat transfer mechanism and to limit the temperature, the bed may be divided into a number of cells each surrounded by waterwalls. The heat of the gas leaving the combustion zone is removed by conventional convection heat-recovery equipment.

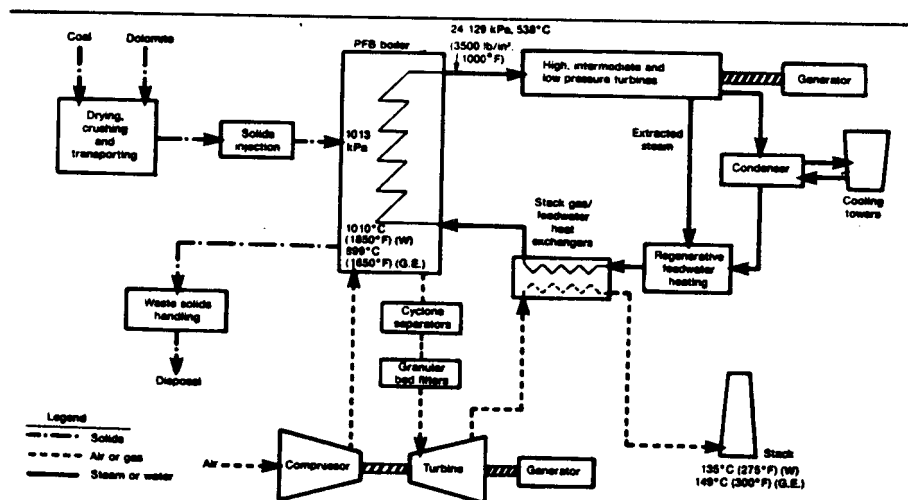


Figure 1. SUPERCHARGED BOILER COMBINED CYCLE

The 5 MW PFBC facilities of the NCB in Leatherhead has recently completed a 1000 hour testing phase. This testing program also involved Stal-Laval, Combustion Systems Ltd., and Babcock International. The fluidized bed combustor configuration used in the Leatherhead facilities is shown in Figure 2. This unit incorporates a range of superheater and reheater tube alloys for testing at bed temperatures typical to those that would be encountered in a conventional boiler. The bed has a cross-sectional area of up to 0.84 m^2 and can operate at pressures of up to 6 bars at heat inputs of 5 MW.

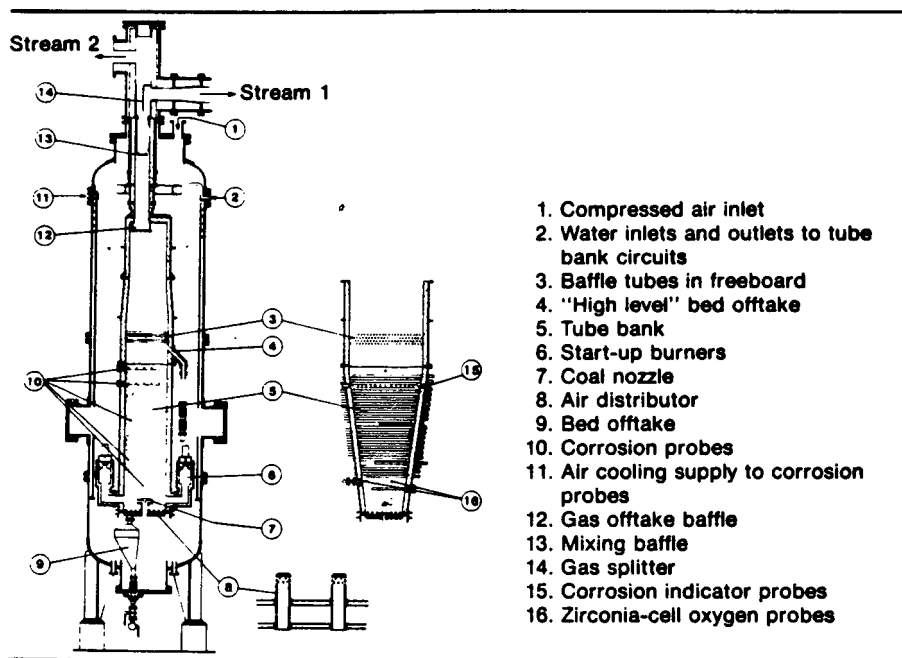


Figure 2. ARRANGEMENT OF FLUID BED COMBUSTOR Mk VI

The combustion gases leaving the 5 MW PDU are split into two streams as shown in Figure 3. In the initial testing period each stream had 3 cyclone dust collectors in series for particulate removal. Each stream then proceeded to a cascade of turbine blades that serves as a turbine blade test unit. One cascade was supplied by Stal-Laval and the other by GE to test gas turbine alloys exposed to combustion gases with relative velocities of up to 525 m/s. The gas cleanup cyclones were effective at removing most particles larger than 10 microns. The particles that did not reach the cascade blades were relatively soft. After 650 hours of operation the Stal-Laval stream was modified

by removing one of the cyclones to increase particulate loading. After 1000 operating hours the cascade systems were dismantled to check for wear and corrosion. With the exception of minor ash buildup which did not affect performance, none of the coated blades or specimens in either streams showed signs of corrosion attack.

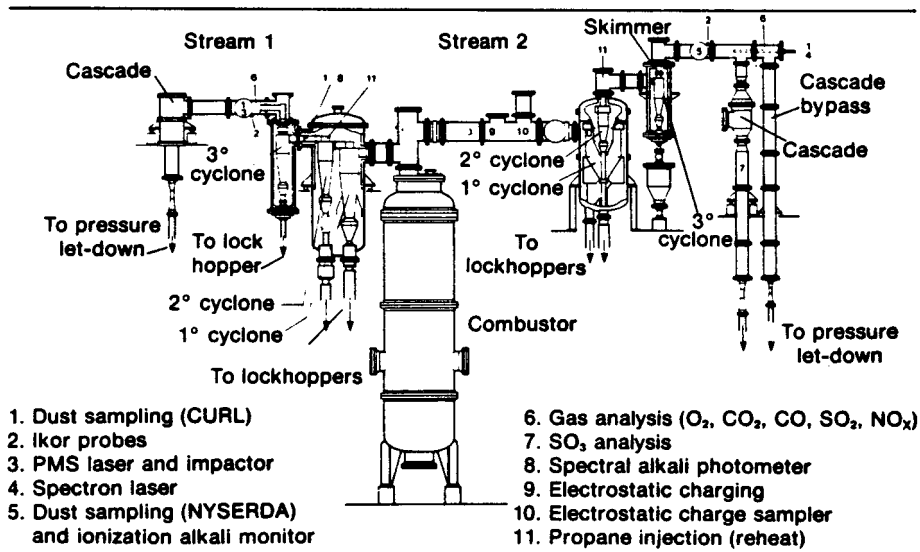


Figure 3. TEST FACILITY

The 3 foot by 2 foot cross-section bed is usually operated with an 8 foot deep fluidized-bed. The bed normally operates in the temperature range of 700°C to 950°C and at about 6 bars pressures. Combustion efficiencies as high as 99% have been achieved when operating with 30% excess air and a fluidizing velocity of 5 feet per second. When operating on 3% sulfur coal over 85% of the sulfur was retained in the bed. This bed contained 1.5 times the stoichiometric quantity of dolomite required to retain the sulfur.

Due to the small size of the Leatherhead facility scale up to a 500 MW pilot plant was deemed too risky. Therefore, an 80 MW experimental pilot plant was constructed at Grimethorpe for further testing. The overall flow diagram for this facility is shown in Figure 4. The combustor (Figure 5) is designed to operate at 12 bars and has a 14 meter height and 4 meter outside diameter. When operating at maximum capacity this 2 m x 2 m cross-sectional area combustor operates with a fluidizing velocity of 2.5 m/s, a bed temper-

ature of 850°C, and an air mass flow of 31 kg/s. This unit can handle 10 tons/hr of coal with a top size of 6.4 mm. Coal mixed with the limestone or dolomite SO₂ sorbent is fed into the bed via a Petrocarb feeder at nine injection nozzles. The calcium to sulfur mole ratio to achieve 90 removal of the SO₂ is 2.6.

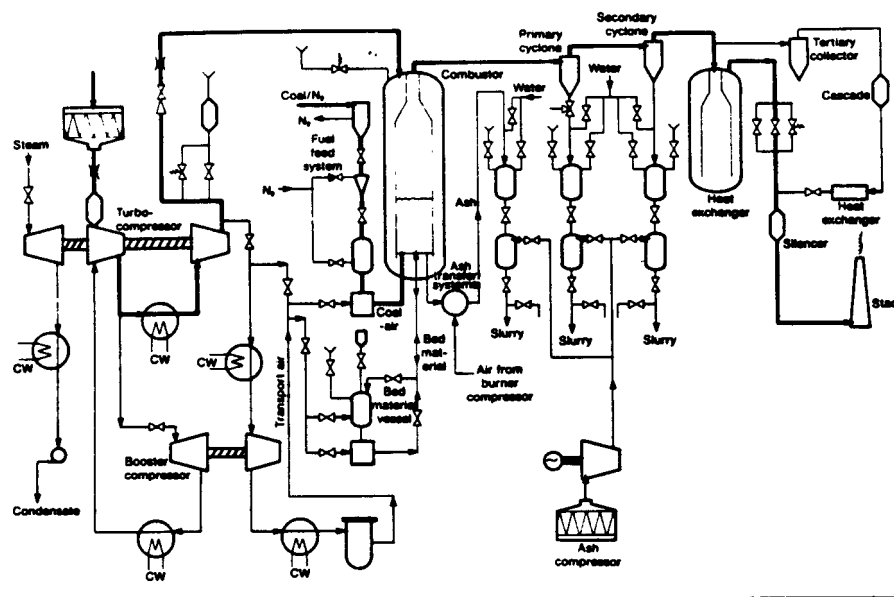


Figure 4. OVERALL FLOW DIAGRAM

The fluidized bed has a maximum depth of 4.5 meters and contains a bank of evaporator tubes. The water-cooled walls extend an additional 4.5 meters above the expanded bed height. This 4.5 meter freeboard is designed to provide space for larger entrained particles to settle out and return to the bed. The freeboard area also contains a small band of heat exchanger tubes for combustion gas cooling in the event of excessive above bed burning. The combustion product leaving the top of the PFBC unit is split into four streams for feeding into four primary cyclones arranged radially around the combustor. Each primary cyclone is connected to a secondary cyclone where the cleaned gas finally exits into a common header. From here the gas is cooled to below 300°C before pressure letdown, silencing and exiting into a 90 meter tall flue stack.

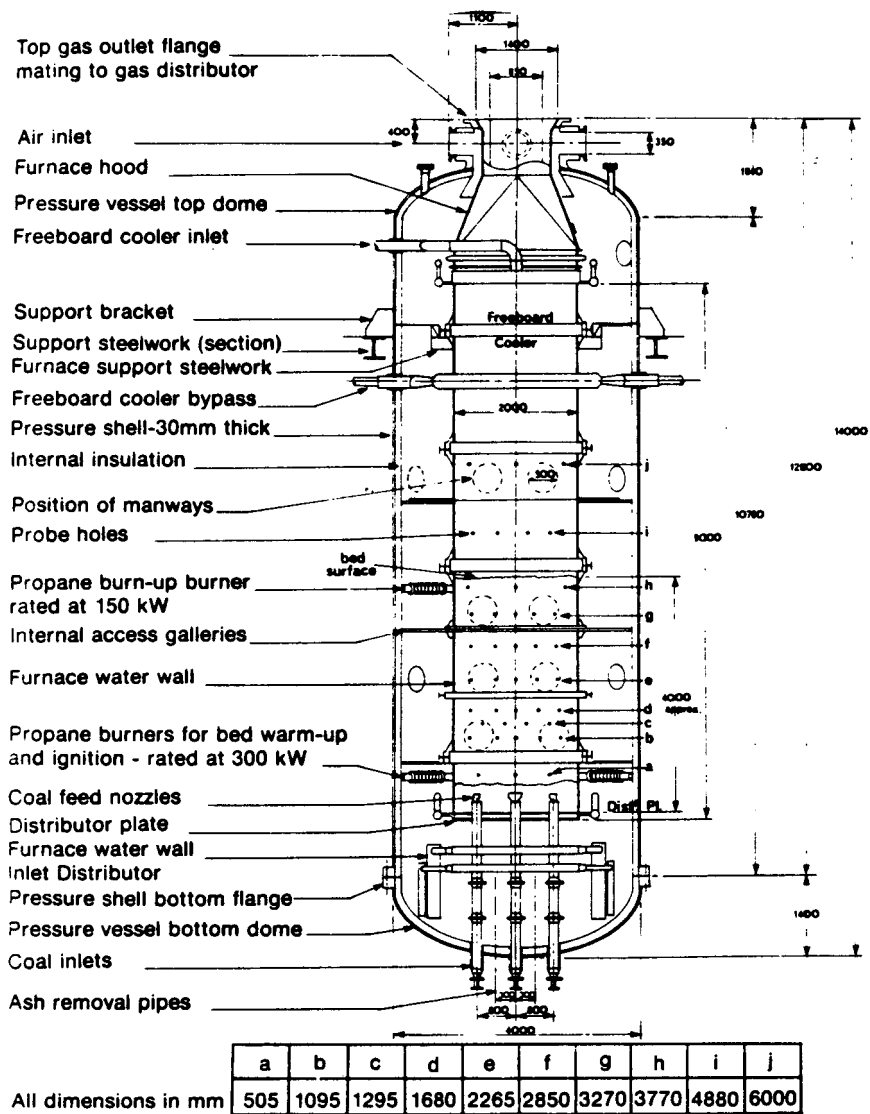


Figure 5. COMBUSTOR PRESSURE VESSEL

During testing tube bundle configurations and dust collection equipment will be modified to record system performance. Part of this testing will involve passing combustion gas through a tertiary cleaning system and then expanding it through a cascade of gas turbine blades to obtain additional performance data. These gases leaving the cascade will be cooled and depressurized in a separate system. The first phase of testing is envisioned to require 2900 hours and take 24 to 30 months to complete. Testing started in April 1981. A technology fact sheet for this process is shown in Table 1.

Table 1 Part 1 TECHNOLOGY FACT SHEET: NCB Pressurized Fluidized Bed Coal Combustion Process

NEW TECHNOLOGY (PROCESS DESCRIPTION AND GOALS): This concept involves burning coal in a fluidized bed of

calcium-containing minerals such as limestone to absorb the polluting sulfur in the coal. When operating at elevated pressures hot combustion gas can be used for combined cycle power generation.

RELATIONSHIP TO PRIOR TECHNOLOGY (INCLUDING STATE OF DEVELOPMENT OF PRIOR TECHNOLOGY): PFBC is directly related to conventional fluidized-bed technology which was invented in the 1920's.

CHARACTERISTICS OF THE TECHNOLOGY:

<u>CHARACTERISTICS</u>	<u>NEW TECHNOLOGY</u>	<u>PRIOR TECHNOLOGY</u>
PRIMARY OUTPUT (DESIGN CASE).....	electricity	
TYPE OF PROCESS.....	fluidized-bed combustion	
FEEDSTOCK REQUIREMENTS.....	dried and sized coal and limestone	
OVERALL THERMAL EFFICIENCY (INCLUDING BY-PRODUCTS)..	40% to electricity	
CARBON CONVERSION EFFICIENCY.....	99%	
OPERATING TEMPERATURE.....	950°C	
OPERATING PRESSURE.....	12 bars	
BY-PRODUCTS.....	--	

Table 1 Part 2 TECHNOLOGY FACT SHEET: NCB Pressurized Bed Coal Combustion Process

STATUS OF DEVELOPMENT:

OPERATING FACILITIES - An 80 MW PFBC test facility is located at Grimethorpe and 5 MW pilot plant is operated by the NCB at Leatherhead.

MAJOR FUNDING AGENCY

The International Energy Agency provided funds for the Grimethorpe facility.

ANNUAL LEVEL OF FUNDING -

The Grimethorpe facility costs approximately 10,000 to construct in 1978.

TECHNICAL PROBLEMS:

No major technical problems have been reported.

OTHER FACTORS AFFECTING OVERALL FEASIBILITY:

Increase in coal size feedstock requirements, a better coal feed system and higher combustor operating temperatures can make this process more commercially viable.

Process Goals

Testing in the 5 MW PFBC reactor at Leatherhead has established the steady state operating performance of this technology at 6 bars pressure. The 80 MW pilot plant at Grimethorpe is designed to provide a wider range of operating conditions for pressures up to 12 bars. This testing program is designed to:

- Determine combustor performance and pollution emission levels of a wide range of operating conditions at elevated pressures
- Establish system performance for different coal feedstocks; two different U.S. and West German coals will be tested in addition to U.K. coals
- Provide part-load data and dynamic response data for systems control design.
- Assess system performance effects when design changes are made, for example, elimination of some of the coal feed nozzles or changes in bed or freeboard heights.
- Determine corrosion behavior of tubes submerged in the fluidized bed as well as those subjected to particulates in the freeboard.
- Determine heat transfer distribution in the bed.

Relationship to Prior Technology

Though fluidized-bed combustion is often divided into the two distinct areas of atmospheric and pressurized technology, the two technologies have common development roots and several common research concerns. Because of the similarities of the technologies, in this report they will be treated in an intermingled manner.

The concept of the fluidized-bed was invented in the 1920's as a means to promote chemical reactions. By the early 1940's, the fluidized-bed concept was in commercial use for petroleum cracking. By the late 1950's the fluidized-bed technique was commercially successful for metallurgical heat treatment and ore roasting. Hundreds of these units have been sold. In the late 1940's, several U.S., British, German, and French companies began development of fluidized-bed combustion systems. A French design, which features two-staged coal combustion, was not a market success.

In the late 1950's, the British National Coal Board's (NCB) Coal Research Establishment operated by the British Coal Utilization Research Association

(BCURA) continued earlier work by Douglas Elliott of Britain's Central Electricity General Board (CEGB). General lack of interest in coal due to low cost oil and gas slowed research. By the late 1960's, however, interest in fluidized-bed coal combustion was prompted in Britain by the NCB's desire to sell coal worldwide and by the emergence of future oil supply problems. In 1968, Elliott developed the concept of pressurized fluidized-bed combustion. Under his direction, a 5 MW (thermal) unit was commissioned in 1969. Though the NCB proposed a 20 MW pilot facility in 1971, the British Government and the CEGB refused funding.

In the United States, Michael Pope of Pope, Evans, and Robbins (PER) won support from the U.S. Office of Coal Research (OCR) in 1965 to build three atmospheric fluidized-bed test units. The largest of these units, a 0.5 MW (thermal) unit, was first operated in 1965 at Alexandria, Virginia. Foster Wheeler Corporation and Combustion Power Company were carrying out limited fluidized-bed combustion research by 1970. The phenomenon of greatly reduced sulfur emissions was first reported as a result of PER's work with OCR in 1968 using the Alexandria, Virginia test facility,. In 1972, OCR provided funds for PER to build three prototype atmospheric fluidized-bed boilers. The first was built at Alexandria, Virginia. Environmental concern and the EPA led to a joint EPA/NCB program aimed at sulfur dioxide and nitrogen oxides control using fluidized-bed combustion. The program indicated that up to 95% of the sulfur in high sulfur coal could be captured by fluidized-bed combustion. With OCR funding, PER built a 30 MW unit at Rivesville, West Virginia in 1972. The formation of ERDA after the OPEC oil embargo resulted in \$19.8 million in U.S. Government funding for fluidized-bed combustion in 1975.

Recent development in Europe has had a more commercial orientation than in the United States. In 1972, Douglas Elliott of the NCB and a partner founded Fluidfire Development, Ltd. The company has developed and successfully marketed heat treatment furnaces, refuse burning boilers, and flue heat recovery units using fluidized-bed designs. The company has been aggressive in concept development; one development has been a fluidized-bed boiler with a bed depth of only a few inches. In one configuration, a novel method of controlling the boiler was developed by varying the position of the fluidized-bed around the heat exchangers.

Similarly, with commercialization intended, the NCB sponsored converting the boiler at the Babcock and Wilcox, Ltd. factory in Renfrew, Scotland to fluidized-bed firing in 1974. By August 1975, the 40,000 lb/hr unit was in operation. The 10 ft² bed operates at 400 psi and 560°F. The unit which is started up by oil-fired overhead burners now operates on oil. However, its original testing program included coal firing; up to 98% sulfur retention was noted with 3.6% sulfur coal and a limestone bed. After testing, and to date, no loss in bed tube material has been observed. The unit's turndown ratio is 4:1. In 1976, Babcock and Wilcox, Ltd. announced that units would be made available with a warranty for production of up to 500,000 lb/hr of steam.

The NCB has sponsored other small conversions as well as including the conversion of an 80,000 lb/hr boiler operated by British Steel Corporation. This unit was to be commissioned in 1978.

Operating Facilities

The National Coal Board has operated a PFBC unit with a 5 MW rated capacity at its Coal Utilization Research Association Laboratories (BCURA, now CURL) in Leatherhead. Over 3000 testing hours have been logged on this unit with operating pressures of up to 6 bars. Initial bench scale studies at the NCB's Coal Research Establishment (CRE) labs provided the data for the Leatherhead facility.

In 1977 the International Energy Agency started construction of the 80 MW Grimethorpe facility which is based on the research data from the Leatherhead facility. Cold flow commissioning began in October 1979 and hot commissioning started in September 1980. Cold flow tested required an additional two month period due to mechanical problems in the ancillary equipment. The first phase of the experimental program at Grimethorpe started in April 1981 which will involve 2900 hours of testing over a 2 to 2-1/4 year period.

Major Funding Agencies

PFBC studies which started in 1969 and much of the laboratory work since 1972 has been sponsored by the U.S. DOE in cooperation with other U.S. organization. These include the Electric Power Research Institute, American Electric Power (AEP) and General Electric. Funding for the 80 MW PFBC facility at Grimethorpe in Yorkshire was provided by the International Energy Agency.

Technical Problems

Although much of the experimental work to date has concentrated on the reactor design, material problems and turbine blade performance, little research has been done on coal feeding problems. In the current approach, coal is dried and crushed to a compatible size dependent on the pressurized fluidizing gas velocity. For example, for a fluidizing velocity of 1.2 m/s a coal and dolomite mixture with a top size of 3 mm would be required as feed. Based on the current use of nozzles to inject the coal into the PFBC reactor, one nozzle is required for each 1.5 m² of bed area. Therefore, for a 200 to 250 MW(e) plant over 50 coal nozzles would be required to feed the coal into the reactor. Larger nozzles or a less complicated feed system is required to make large commercial plant operation feasible.

In addition to reducing the number of nozzles required, it would be advantageous to increase the maximum allowable coal size range to 30 mm. Large coal of this size has been experimentally combusted in a fluidized bed with no problems. However, if noncombustible materials accumulate within the bed, performance can be degraded. A larger coal size distribution will require new nozzle design, especially when handling wet run of mine coals. This large size distribution will also require better fluidizing bed control. It may be desirable to use a circulating bed which will recycle the dust overflow back into the bed.

The final area of concern is increases to overall plant efficiency for the PFBC combined cycle plant. Current efficiency is estimated to be in the range of 39 to 40 percent with reheat of the steam system and a gas turbine inlet temperature of 800°C. This performance is 5 percent better than a conventional coal burning power plant with flue gas desulfurization to reduce SO₂ while burning a 3 to 4 percent sulfur coal. Higher efficiencies of 45 percent can be obtained by increasing gas turbine inlet temperatures to 1100°C. However, slag formation which will bond to turbine blades is likely at this temperature. A secondary combustor may be required to reach this temperature by heating the PFBC reactor output. Fuel for this after burner stage may be produced in a preliminary pyrolysis/gasifier stage of the PFBC reactor.

Capital Costs

Reduction of capital costs of 12 percent and a decrease of operating costs when compared with conventional coal combustion power generation have been predicted. The 80 MW pilot plant at Grimethorpe was constructed at an estimated cost of $\$10 \times 10^6$ in 1978.

BRITISH COAL GASIFICATION PROCESS

20. Esso Chemically Active Fluidized Bed Coal Gasification ProcessProcess Description

The Esso Chemically Active Fluidized Bed gasification process (CAFB) has been tested in a 150 kg/hr reactor. A schematic of this pilot plant is shown in Figure 1. The pilot plant consists of a fluidized bed reactor where fuel is partially oxidized at 900°C within a bed of lime particles fluidized by air. A top coal size of 3000 microns can be fed into the gasifier. Over 80% of the sulfur in the fuel, either oil, gas or coal, is retained by the lime when it is converted to CaS. In addition to sulfur, metals within the coal structure such as V, N, and Na are also retained in the lime bed. The spent lime material is then transferred from the gasification reactor to the adjoining regeneration fluidized bed reactor. In this reactor the CaS which was formed in the gasification reactor is oxidized back to lime and SO₂ at 1050°C.

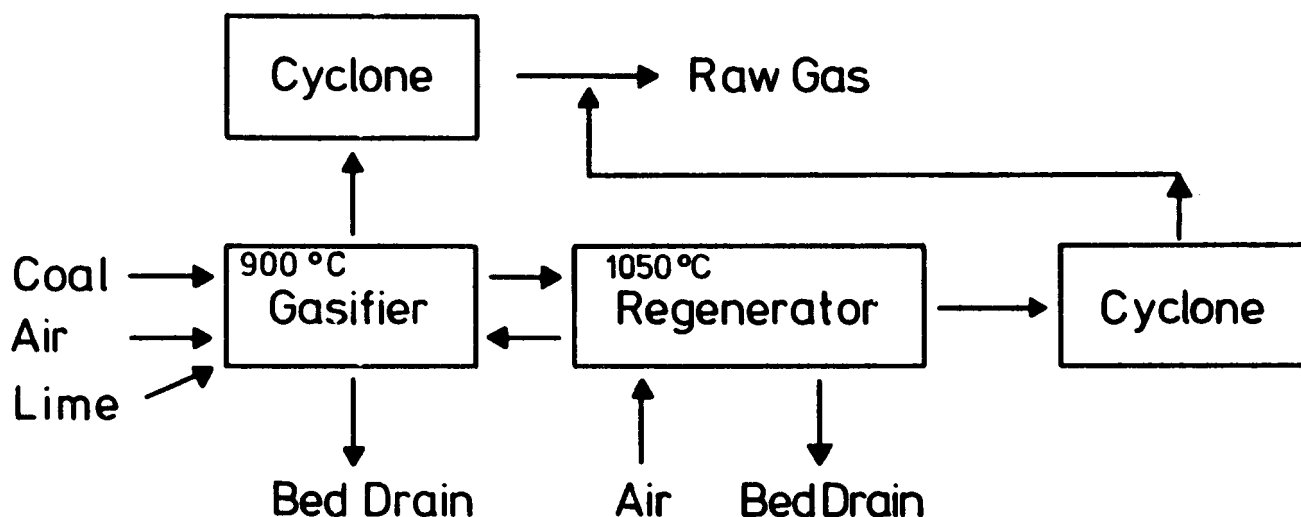


Figure 1. FLOW DIAGRAM OF ESSO CHEMICALLY ACTIVE FLUIDIZED BED GASIFICATION PROCESS

During the reaction in the gasification vessel the coal undergoes pyrolysis to release large concentrations of condensable hydrocarbons. In actuality only partial gasification occurs and large quantities of low sulfur char is produced as a by-product. Tests results using Illinois No. 6 and the Texas lignite coals shown in Table 1 are presented in Table 2. A summary of the sulfur removal effectiveness of this process when operating on the Illinois and Texas coals is shown in Table 3. A technology fact sheet summarizing this process is presented in Table 4.

Table 1. ANALYSIS OF ILLINOIS NO. 6 AND TEXAS LIGNITE
FED TO CONTINUOUS UNIT

	<u>Illinois No. 6</u>		<u>Texas Lignite</u>	
	<u>Actual</u> %	<u>Dry Basis</u> %	<u>Actual</u> %	<u>Dry Basis</u> %
Moisture	4.63	-	14.02	-
Ash	7.36	7.80	18.50	21.50
Carbon (corrected)	70.3	73.64	51.20	59.50
Hydrogen (corrected)	4.61	4.83	3.60	4.20
Sulphur (total)	1.78	1.90	0.82	0.95
Nitrogen	1.44	1.50	1.00	1.20
Oxygen errors (by difference)	9.85	10.40	10.90	12.60
Gross calories/GM	6936	7260	5004	5817
Gross Btu/lb	12486	13068	9007	10470
CO ₂ (%)	0.16	-	0.51	-

Table 2. COMPARISON OF GASIFIER GAS QUALITY FROM FUEL OIL AND COAL

<u>Gasifier Fuel</u>	<u>Heavy Fuel</u> <u>Oil</u>	<u>Illinois No.6</u> <u>Coal</u>	<u>Texas</u> <u>Lignite</u>
Nitrogen + inerts	58.4	59.2	59.0
Carbon monoxide	10.2	12.2	12.2
Carbon dioxide	10.2	9.9	12.1
Methane	7.7	4.2	2.2
Ethylene	5.0	0.8	0.7
Ethane	0.1	0.1	0.1
Hydrogen	8.4	13.6	13.7

Table 3. SUMMARY OF DESULPHURIZING PERFORMANCE DURING
MINI-RUN ON COAL

<u>Coal (type and size)</u>	<u>Mean % SRE on Coal</u>	<u>Expected Result on Oil (% SRE)</u>	<u>SO₂ Emission (lb SO₂ per 10⁶ Btu)</u>
Illinois No. 6 (1405 μ down)	74.8	73.8	0.73
Texas Lignite (800 down & 1405 μ down)	64.0	68.5	0.76
Texas Lignite (1/8" down)	82.4	67.2	0.42

Process Goals

The objective of this development is to develop a commercial coal or oil burning gasifier which can remove sulfur from pollutants from the fuel and can be easily integrated with existing gas-fired boiler plants. Research on the operating range and performance of this gasifier is underway. Also being examined in the long term affect of coal ash on the sulfur absorbing properties of the line bed material.

Relationship to Prior Technology

The concept of the Catalytically Active Fluidized Bed process was first developed in 1967 and was originally applied to the desulfurization of fuel oils. In 1975 this concept was extended to the use of coals in a batch test at Abingdon, U.K. However, the concept of removing sulfur compounds from gas using a limestone absorbent is not new. This approach has been applied to sulfur removal from stack gases as a pollution abatement process.

Operating Facilities

Esso Petroleum Company Ltd., Abingdon, Oxon, U.K. has operated a 150 kg/hr process development unit at the Abingdon test facilities since 1975. A 20 MW pilot plant unit has been proposed for operation at the Texas Central Power and Light Company which will operate on Texas lignite.

Table 4 Part 1 TECHNOLOGY FACT SHEET: Esso Chemically Active Fluidized Bed Coal Gasification Process

NEW TECHNOLOGY (PROCESS DESCRIPTION AND GOALS): The ESSO CAFB process utilizes two fluidized bed reactors

to pyrolyze and partially gasify coal at 900°C in the presence of a sulfur absorbing lime bed. Spent lime, containing CaS, is transferred from the fluidized bed gasification vessel to the fluidized bed lime regeneration reactor. Sulfur is removed as SO₂ at 1050°C and the lime is recycled to the gasifier. This process is designed for use with a gas-fired conventional boiler facility.

RELATIONSHIP TO PRIOR TECHNOLOGY (INCLUDING STATE OF DEVELOPMENT OF PRIOR TECHNOLOGY): Process was first developed in 1967 to gasify high sulfur fuel oil.

CHARACTERISTICS OF THE TECHNOLOGY:

<u>CHARACTERISTICS</u>	<u>NEW TECHNOLOGY</u>	<u>PRIOR TECHNOLOGY</u>
PRIMARY OUTPUT (DESIGN CASE).....		
TYPE OF PROCESS.....	fluidized bed	
FEEDSTOCK REQUIREMENTS.....	coal sized <3000 micron	
OVERALL THERMAL EFFICIENCY (INCLUDING BY-PRODUCTS)..		
CARBON CONVERSION EFFICIENCY.....		
OPERATING TEMPERATURE.....	900 °C	
OPERATING PRESSURE.....		
BY-PRODUCTS.....	char	

Table 4 Part 2 **TECHNOLOGY FACT SHEET:** Esso Chemically Active Fluidized Bed Coal Gasification Process

STATUS OF DEVELOPMENT:

OPERATING FACILITIES - 150 kg/hr process unit started operation at Esso's Abingdon test facility in 1975.

MAJOR FUNDING AGENCY Esso and the U.S. Environmental Protection Agency.

ANNUAL LEVEL OF FUNDING -

TECHNICAL PROBLEMS:

OTHER FACTORS AFFECTING OVERALL FEASIBILITY:

Major Funding Agencies

Esso Petroleum Company Ltd. and the U.S. Environmental Protection Agency have provided the funding for this process.

Technical Problems

No technical problems have been reported in the literature.

Capital Costs

Capital costs for this process have yet to be published.

32(3)/process/ER

AUSTRALIAN COAL CONVERSION PROCESS

21. CSIRO Flash Pyrolysis Coal Liquefaction ProcessProcess Description

The Commonwealth Scientific and Industrial Research Organization -- Division of Fossil Fuels (CSIRO) high speed pyrolysis process utilizes a fluidized bed reactor to convert pulverized coal (0.2 mm in size) to tar, gas and reactive char. The reactions occur in an oxygen-free atmosphere at about 600°C and atmospheric pressure. Reaction times of about 1 second are maintained followed by rapid quenching of the product. The tar, which can amount to more than 30% of the m.a.f. coal feed, is then hydrogenated to lighter grade liquid fuels. Some of the product gas and char are used to produce hydrogen for the hydrogenation reaction. A plant layout chart and flow diagram of the flash pyrolysis process are shown in Figures 1 and 2, respectively.

Process Goals

The goal of this program is to determine the technical and economic feasibility of the flash pyrolysis process on Australian coals for the production of liquid fuels. The CSIRO process is also being tested for the pyrolysis of wood wastes, the production of olefins from coal pyrolysis, and the production of carbon anodes from coked flash pyrolysis tars.

Relationship to Prior Technology

The flash pyrolysis process has been studied by various groups for the last 30 years or more. Experiments have been performed in laboratory bench-scale units to nearly commercial scale-pilot plants. Electric power has been produced by electric utilities in the U.S. and West Germany using char produced by the pyrolysis process.

Operating Facilities

The CSIRO research effort on flash pyrolysis started in 1974. A 1 to 3 gram/hr fluidized-bed pyrolysis reactor and a 100 gram/hr entrained bed pyrolysis reactor have been in operation since 1975.

In 1977 a 20 kg/hr process development unit capable of operating in either the fluidized bed or entrained bed mode was commissioned. This unit was converted to an integrated facility in 1980/81 incorporating the pyrolyzer, a char-burning heat generator and an on-line tar hydrogenator.

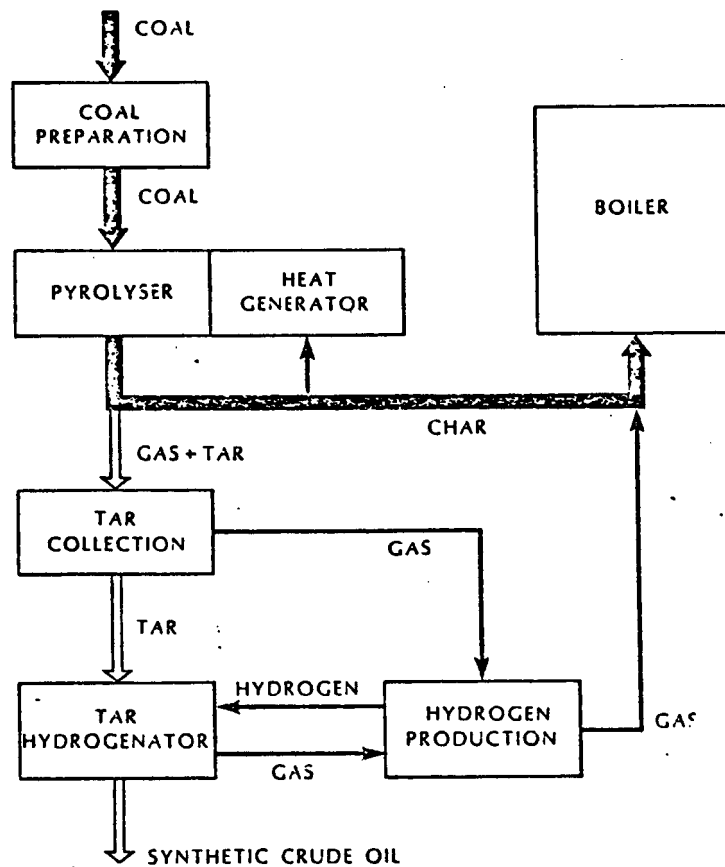


Figure 1. LAYOUT OF COUPLED PYROLYSER — POWER PLANT INSTALLATION

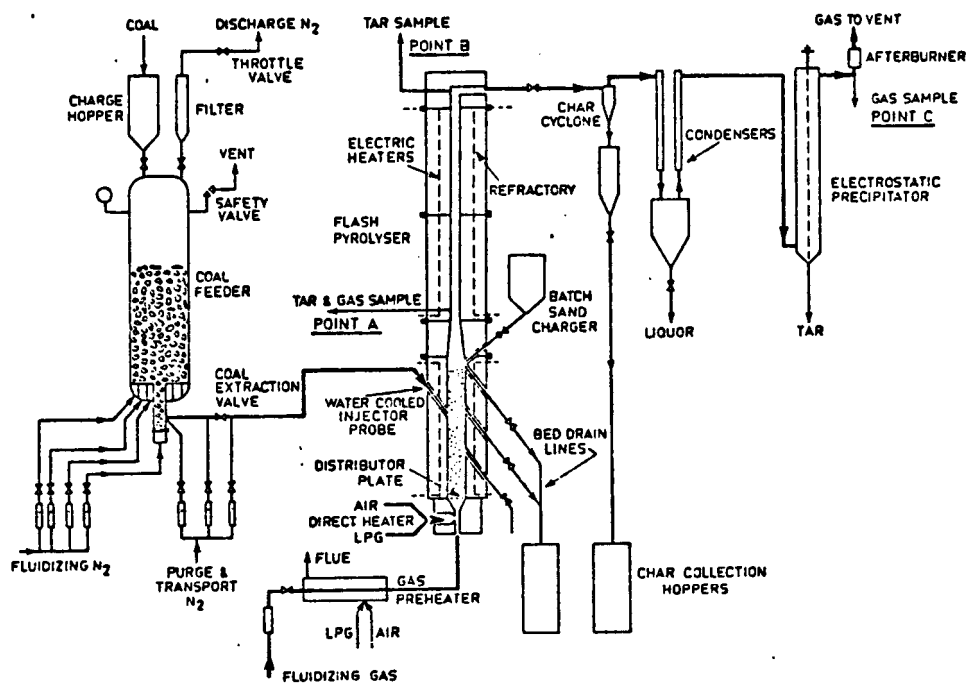


Figure 2. FLOW DIAGRAM OF 20 kg h⁻¹ FLASH PYROLYSER

Operation of this facility, which started in late 1981, will provide the necessary data for the construction of a 1 tonne/hr pilot plant.

The CSIRO work has been carried out within the Organization mainly by the Division of Fossil Fuels and and Division Applied Organic Chemistry. Outside CSIRO, support has come from the Australian Mineral Development Laboratories, and the Universities of Sydney and Queensland. A version of the CSIRO laboratory-scale reactor has been built at the University of Waterloo, Ontario, for the pyrolysis of wood-waste. Another version has been built by DuPont at Wilmington, Delaware, to study the production of olefins by flash pyrolysis of coals at temperatures around 900°C. In the CSIRO laboratories a second PDU-scale rig has been operated for the Aluminum Development Council, by COMALCO, to determine if flash pyrolysis tars can be coked to produce a substitute for petroleum coke used in carbon anodes for aluminum smelting. COMALCO are also building a version of the laboratory-scale rig as part of the carbon anode project.

Major Funding Agencies

Funding on all synthetic fuels programs in Australia amounted to about $\$10 \times 10^6$ in 1979-80. The CSIRO spent $\$5.07 \times 10^6$ in 1979-80 in synthetic liquids fuels research. Part of this money ($\$420,000$) came from the National Energy Research, Development and Demonstration Council (NERDDC) which itself had a total budget that year of $\$5.05 \times 10^6$ for synthetic liquid fuels research.

Technical Problems

Agglomeration of coal in the fluidized bed reactor was a problem in the small scale reactor. Nearly all coals show some plastic behavior during flash pyrolysis. Oxidative pre-treatment will reduce agglomeration but will also reduce tar yields. However, agglomeration did not appear to be a problem in the larger size reactors. In addition to caking tendencies, the coals cation content effects yields. In general, for a given coal, as the cation content increases, the tar yield decreases. Acid washing of the coal can be used to decrease cation content.

Capital Costs

The capital costs and operating costs for the CSIRO flash pyrolysis process are shown in Table 1. All costs are in late 1980 dollars. An Australian Millmerran coal was assumed to be the feedstock.

Table 1. CAPITAL AND OPERATING COSTS FOR 40,000 BARRELS PER DAY
CSIRO FLASH PYROLYSIS COAL LIQUEFACTION PLANTS

<u>Capital Costs (\$10⁶)*</u>	
1. Process units	
Feed preparation	60
Pyrolysis and product recovery	274
Primary liquid hydrogenation (1)	448
2. Offsites, utilities, engineering and design	312
3. Estimating contingency, work capital, initial catalysts and chemicals, and start up costs	--
4. Total capital investment	<u>1483</u>
<u>Operating Costs (\$10⁶ p.a.)*</u>	
1. Raw materials	
Feedstock	137 (2)
Water	4
2. Other	
Maintenance	44
Taxes, insurance and overheads	33
Operating labor and supervision	11
Catalysts and chemicals	12
Purchased fuel gas and electricity	- (3)
3. Credits for ammonia and sulphur byproducts	-4
4. Net operating costs	<u>237</u>

* End 1980 US\$

Notes.

- (1) Includes H₂ production
- (2) Cost at \$1/GJ, based on \$28/tonne dry, opportunity cost.
Cost shown net after allowing byproduct char credit at \$1/GJ.
- (3) All requirements produced from pyrolysis gas and coal char.

Table 2 Part 1 TECHNOLOGY FACT SHEET: CSIRO Flash Pyrolysis Coal Liquefaction Process

NEW TECHNOLOGY (PROCESS DESCRIPTION AND GOALS): The CSIRO high speed flash pyrolysis process converts pulverized coal (sizes < 0.2 mm) to ton, gas, and reactive char. The tar, which can amount to more than 30% of the m.a.f. coal feed by weight, is then hydrogenated to lighter grade liquids.

RELATIONSHIP TO PRIOR TECHNOLOGY (INCLUDING STATE OF DEVELOPMENT OF PRIOR TECHNOLOGY): Flash pyrolysis has been studied by numerous countries over the last 30 years.

CHARACTERISTICS OF THE TECHNOLOGY:

CHARACTERISTICS

YIELDS FROM PYROLYSIS OF VARIOUS COALS AT 600°C

Coal	Liddell		Millmerran	Pittsburgh		South African
	A	B		No. 8		
Yield at 600°C (% w/w daf coal)	25.4	30.0	32.0	31.0	22.1	
Tar	61	52	54	60	41	
Total volatile matter	4.3	4.2	5.6	5.6	2.2	
Total C ₁ -C ₃	2.0	2.3	2.2	3.0	1.8	
CH ₄	0.6	0.5	1.2	0.6	0.4	
C ₂ H ₄	0.8	0.8	0.8	1.0	0.5	
C ₂ H ₆	0.5	0.4	0.8	0.4	0.2	
C ₃ H ₆	0.3	0.3	0.4	0.4	0.2	
Char retained in bed (%)	80	92	80	92	90	

Table 2 Part 2 TECHNOLOGY FACT SHEET:

STATUS OF DEVELOPMENT:

OPERATING FACILITIES - The CSIRO research effort on flash pyrolysis started in 1979. A 1 to 3 gram/hr fluidized-bed pyrolysis reactor and a 100 gram/hr entrainment reactor have been operated since 1975, ca 1977 a 20 kg/hr reactor began operation.

MAJOR FUNDING AGENCY CSIRO and Australian NERDDC

ANNUAL LEVEL OF FUNDING - CSIRO spent \$5.07 x 10⁶ in 1979-80 on synthetic liquids research.

TECHNICAL PROBLEMS: Agglomeration of coal in the fluidized bed reactor was a problem in the small scale reactor but did not appear in the larger PDU. Coal acid washing may be required on certain coals to decrease cation content.

OTHER FACTORS AFFECTING OVERALL FEASIBILITY:

POLISH COAL LIQUEFACTION PROCESS

22. Polish Central Mining Institute Catalytic Hydrogenation Coal Liquefaction ProcessProcess Description

The Central Mining Institute, Division of Carbochemistry, has developed a catalytic hydrogenation coal liquefaction process. In this process, coal is first crushed and dried before entering a ball mill where it is ground to less than 0.5 mm. The prepared coal is then fed to a mixing tank where recycled process solvent is mixed with the coal to form a slurry. The slurry contains approximately 28.6 wt % coal and 71.4 wt % recycled solvent. The recycled solvent is actually a mixture of .44 vol. % anthracene oil and 56 vol % high boiling range recycled oil. The properties of this solvent are presented in Table 1.

From the slurry preparation tank the mixture is heated and sent to the extract mixer column where the coal is reacted with the solvent. Coal reaction time in this reactor is about 50 minutes. The reaction proceeds at 410°C and at 45 atm. The mixture exiting this reactor is then hot filtered to remove mineral matter and unreacted coal residue. After filtration the coal oil enters a vacuum distillation unit where the more volatile oils are separated. Bottoms from the distillation unit and filtrate cake are sent to a carbonizer.

Table 1. PROPERTIES OF RECYCLED OIL SOLVENTS (Hydrogen Donor)

Specific gravity at 20°C, g/ml	1.02
Ultimate analysis (maf), wt%	
Carbon	90.92
Hydrogen	8.64
Sulfur	0.31
First drop boiling/initial boiling point °C	187
Fraction boiling to 360°C, vol%	96
Fraction boiling to 400°C, vol%	97.8

The low-ash extract produced in the vacuum distillation unit is then processed in a catalytic hydrogenation unit. The coal extract entering this unit contains about 50% liquids with a boiling range above 400°C. Hydrogenation occurs in the presence of a nickel-molybdenum catalyst at 240 atm and at 400°C. The hydrogenated liquids are then sent to an atmospheric distillation unit where the low and middle boiling range liquids are separated from the high boiling range (>400°C) liquids. The high-boiling range hydrogenation product is then vacuum distilled. Recovered oil from the vacuum distillation unit is recycled to the slurry preparation section for use as the hydrogen donor solvent, and the bottom residue is carbonized. The property of the coal used in pilot plant testing is shown in Table 2. When operating on this coal the properties of the low-ash extract and liquid products from the hydrogenation unit that can be expected from this process are shown in Table 3. These properties reflect operation of the process under the conditions in Table 4. The technology fact sheet for this process is in Table 5.

The light liquids products are reported to be suitable for further refining into motor fuel products.

Process Goals

This process is being developed in Poland by the Central Mining Institute's division of Carbochemistry at Tychy-Wyry. Research is being performed to determine the operating parameters of the process under various operating conditions. Particular attention has been focused on the selection of a sulfur-tolerant catalyst for use in the hydrogenation reactor. The following process steps are also being investigated.

- Coal/solvent slurry preparation
- Coal extraction with and without hydrogen
- Extract residue separation
- Extract solvent recovery/distillation
- Extract residue low-temperature carbonization
- Extract hydrogenation
- Hydrogenation product distillation.

Research has also been conducted on the production of electrode coke, binders, and carbon absorbents from the coal liquids products.

Table 2. PROPERTIES OF THE COAL USED IN PILOT PLANT TESTING

Moisture, wt%	4.55
Ash (dry basis), wt%	8.23
Volatile matter (maf basis), wt%	41.57
Elements (maf basis), wt%	
Carbon	79.33
Hydrogen	5.52
Sulfur	0.58
Petrographical composition, wt%	
vitrynite	73.0
Egzynite	9.0
Inertynite	13.0
Mineral matter	
Heating value, kcal/kg	7998

Table 3. THE PROPERTIES OF THE COAL EXTRACT AND LIQUID PRODUCTS AT THE 1200 kg COAL/DAY PLANT

<u>Property</u>	<u>Products</u>		
	<u>Coal Extract</u>	<u>Light Liquid Product</u>	<u>Heavy Liquid Product</u>
Water content, wt%	trace	0.3	trace
Ash (dry basis) wt%	0.10	trace	0.16
Specific gravity at 20°C, g/ml	1.22	1.08	1.13
Ultimate analysis (maf basis), wt%			
Carbon			
Hydrogen	6.50	7.56	6.95
Sulfur			
Benzene insolubles, wt%	14.6	1.2	9.5
Asphaltene, wt%	5.6	1.3	12.3
Oils, wt%	79.8	97.5	78.2
Initial b.p., °C	262	93	247
Fraction boiling to 200°C, vol%	--	3.0	--
Fraction boiling to 320°C, vol%	1.5	46.4	7.5

Table 4. PROCESS CONDITIONS OF THE CENTRAL MINING INSTITUTE
CATALYTIC HYDROGENATION COAL LIQUEFACTION PROCESS

1. <u>Extraction</u>		
1.1. Coal content in the slurry	wt %	28.6
1.2. Feed rate of the slurry	l/hr	14.8
1.3. Reaction time	min.	50
1.4. Pressure	atm.	45
1.5. Temperature	°C	400
2. <u>Filtration</u>		
2.1. Temperature	°C	233
2.2. Filtration rate	kg/m ² /hr	287
3. <u>Carbonization</u>		
3.1. Max. temperature	°C	550
4. <u>Filtrat Distillation</u>		
4.1. Pressure	mm	40
4.2. Max. temperature column top	°C	265
5. <u>Hydrogenation</u>		
5.1. Catalyst		Ni-Mo
5.2. Pressure	atm	250
5.3. Reactor temperature	°C	430
5.4. Through-put	kg feed/ liter of catalyst/ hour	0.37

Relationship to Prior Technology

This process is based on the Bergius-Piers process (IG Farben Process) which was developed in Germany. However, hydrogenation of the coal without the addition of a catalyst or hydrogen in the first reaction step represents a significant deviation from the original concept. It was not stated directly in the literature, but one can conclude that hydrogen generation for the catalytic hydrogenation step of this process will be produced by the gasification of coal residue from the process. This has not been technically demonstrated and is another different modification of this process.

Table 5 Part 1 TECHNOLOGY FACT SHEET: Polish Central Mining Institute Catalytic Hydrogenation Coal Liquefaction Process

NEW TECHNOLOGY (PROCESS DESCRIPTION AND GOALS): This process utilizes both a hydrogen donor reaction and a catalytic hydrogenation step to produce a liquid product from coal. In the first step a hydrogen donor solvent which is produced in the process is reacted with the coal. The liquid product is then catalytically hydrogenated in a separate step prior to product recovery. Research has concentrated on catalyst testing and use of the liquid product for electrode coke, binder and carbon absorbent production.

RELATIONSHIP TO PRIOR TECHNOLOGY (INCLUDING STATE OF DEVELOPMENT OF PRIOR TECHNOLOGY):

This process is a modification of the IG Farben Process

CHARACTERISTICS OF THE TECHNOLOGY:

CHARACTERISTICS	NEW TECHNOLOGY	PRIOR TECHNOLOGY
PRIMARY OUTPUT (DESIGN CASE).....	middle and low distillate fuel	
TYPE OF PROCESS.....	hydrogen donor/catalytic hydrogenation	
FEEDSTOCK REQUIREMENTS.....	crushed/dried coal	
OVERALL THERMAL EFFICIENCY (INCLUDING BY-PRODUCTS)..	--	
CARBON CONVERSION EFFICIENCY.....	--	
OPERATING TEMPERATURE.....	400°C/430°C	
OPERATING PRESSURE.....	45 atm/250 atm	
BY-PRODUCTS.....		

Table 5 Part 2 TECHNOLOGY FACT SHEET: Polish Central Mining Institute Catalytic Hydrogenation Coal Liquefaction Process

STATUS OF DEVELOPMENT:

OPERATING FACILITIES -- A 120 kg/day PDU and a 1200 kg/day pilot plant are located at the Central Mining Institutes' Division of Carbochemistry at Tychy-Wyry.

MAJOR FUNDING AGENCY Polish Government

ANNUAL LEVEL OF FUNDING --

TECHNICAL PROBLEMS: None reported

OTHER FACTORS AFFECTING OVERALL FEASIBILITY: Hydrogen production from liquefaction residue has not been proven both technically and economically feasible.

Operating Facilities

A bench-scale unit with a coal capacity of 120 kg/day was constructed at the Carbochemistry Institute of Tychy-Wyry in the mid-70's. This unit was used to gain operating information for the construction of a 1200 kg/day pilot plant in 1977. This unit is also located at the Carbochemistry Institute.

Major Funding Agencies

This process is supported by the Polish Government.

Technical Problems

No technical problems have been reported in the literature.

Capital Costs

The capital costs for this process have not been published.

SOUTH AFRICAN COAL LIQUEFACTION PROCESS

23. SASOL (Lurgi/Fischer-Tropsch) Coal Liquefaction Process

Process Description

Sasol I

SASOL (South African Coal, Oil, and Gas Corporation, Ltd.) was founded in 1950 for the production of synthetic liquids from coal. Their first synthetic liquids plant was completed in 1955 and is located at Sasolburg, 50 miles southwest of Johannesburg, South Africa. In 1979 Sasol was reorganized into Sasol Limited with 70% of the government owned company sold to the public. The new corporate structure which includes the two new synthetic liquids plants and the South African government's Industrial Development Corps.' involvement is presented in Figure 1.

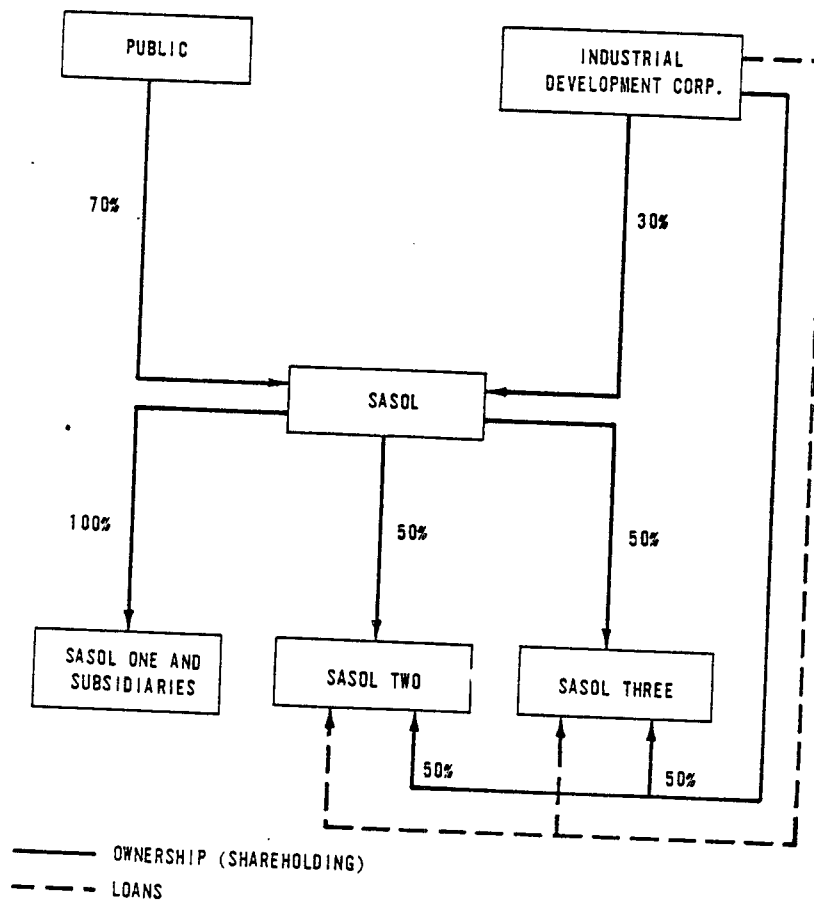


Figure 1. SASOL GROUP STRUCTURE

South Africa's first coal to liquids plant, Sasol I, utilizes Lurgi/Fischer-Tropsch technology to produce a broad range of synthetic products. A block flow diagram of this facility as it exists today, is shown in Figure 2. This facility contains 17 Lurgi gasifiers; 13 Mark III, 3 Mark IV units, and one experimental Mark V gasifier. The Mark III, IV, and V Lurgi gasifiers have a 3.68, 3.85, and 4.7 m internal diameter, respectively. The Mark IV gasifiers were installed in 1977/8 and the Mark V unit was commissioned in 1980.

In the Sasol facility coal is delivered from the adjacent Sigma mine in two coal sizes, -12 mm and 12-50 mm. This low grade bituminous coal, analysis shown in Table 1, is split about 40% fines and 60% coarse material. The fines are fed to two power plants and the remainder is gasified. The coal is delivered from the mine by conveyors and stored in bunkers before entering the gasifier lockhoppers. These lockhoppers located at the top of the gasifiers hold between 6.5 and 10 tonnes of coal depending on their age. After pressurization with raw gas the coal is fed by gravity through conical valves into the distribution grate of the Lurgi gasifier (Figure 3). Gasification is carried out at approximately 375 psig and 1200°C. Total facility gasifier inputs include 9,000 to 11,000 tonne/day of coal, 65,00 to 95,000 m³/hr of oxygen, and 329,000 to 438,000 kg/hr of high pressure steam. Total raw gas production ranges from 450,000 m³/hr to 600,000 m³/hr. Overall availability of the gasifier plant is nearly 85%.

The raw gas leaving the gasifier at about 500°C is scrubbed and cooled to 20 to 25°C to remove higher boiling point tars, oils and entrained ash dust. All tars and oils are filtered before undergoing further processing. The cooled raw gas is then sent to one of three Rectisol trains where carbon dioxide, hydrogen sulfide and organic sulfur are removed. Rectisol utility consumption averages 35.7 kWhr of electricity, mostly used for methanol recirculation pumps, and 79.2 kg of steam per 1000 m³ of pure gas produced. The pure gas leaving the Rectisol units is sent primarily to the Fischer-Tropsch sections; a Ruhrchemie-Lurgi Arge fixed bed design and the other and M. W. Kellogg Co. Synthol entrained bed design. A small fraction of the pure gas is also sent to a methane-reformer where the methane content is reduced before entering the Synthol reactors. The synthesis gas composition entering the two types of Fischer-Tropsch reactors is shown in Table 2.

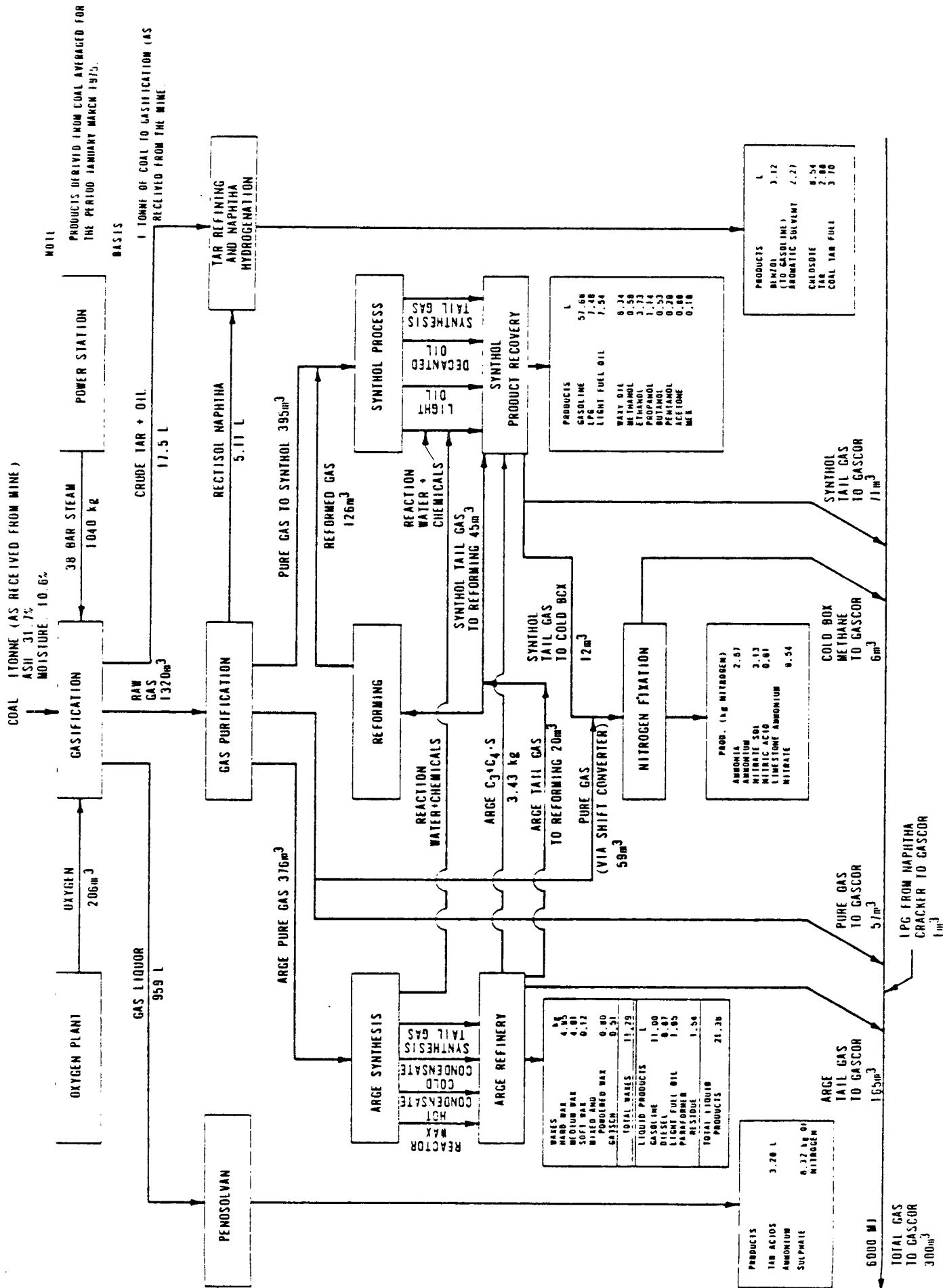


Figure 2. SASOL I FLOW DIAGRAM

Table 1. TYPICAL SIGMA COAL ANALYSIS

<u>Type</u>	<u>Bituminous</u>
Moisture (as received)	10.7 wt %
Volatiles (dry basis)	22.3 wt %
Ultimate Analysis (dry basis), wt %,	
Ash	35.9
Sulfur	0.5
Nitrogen	1.2
Carbon	50.8
Hydrogen	2.8
Oxygen	8.8
Ash fusion Temperature, °C:	
Softening point	1340
Melting point	1430
Fluid point	1475
Energy Content, MJ/kg (dry)	20-22

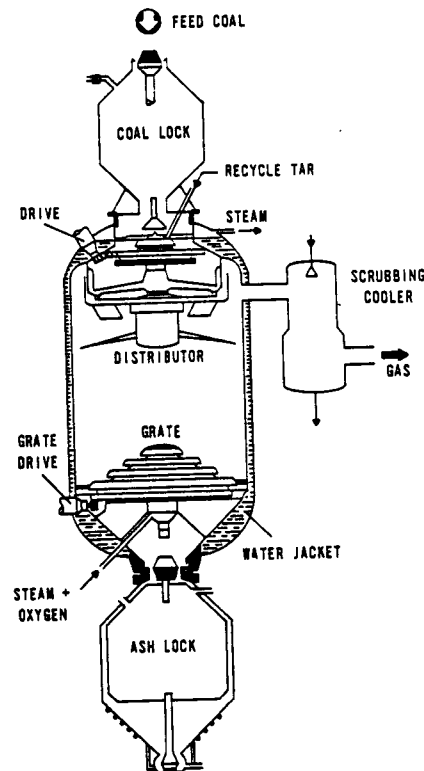


Figure 3. LURGI MARK IV GASIFIER

Table 2. RAW GAS COMPOSITIONS, VOLUME %

<u>Component</u>	<u>Arge</u>	<u>Synthol</u>
H ₂	39	40
CO	22	18
CO ₂	28	31
CH ₄	9	9
N ₂	1	1
H ₂ S	1	1

The Synthol reactor system has been developed to the commercial stage by Sasol, which today owns all the rights. This process produces predominantly lower-boiling point hydrocarbons in the gasoline and diesel oil range and the Arge process produces high-boiling point hydrocarbons, including a range of solid waxes. A breakdown of the product mix from these two reactors is shown in Figure 2. Both processes use an iron-based catalyst activated with certain promoters. These catalysts were manufactured in West Germany prior to 1969 when Sasol commissioned its own catalyst preparation plant. Production composition can be varied somewhat by charging catalyst properties and reactor conditions.

Part of the synthesis gas can be blended with industrial gas which is distributed by the Gascor subsidiary in a pipeline system. Some of the Synthol tail gas is also blended with synthesis gas as feed for an ammonia synthesis unit. Before entering the ammonia synthesis section the feed gas is shifted in a water-gas shift reactor and the carbon dioxide and hydrocarbons are removed. Nitrogen for the ammonia synthesis reaction is supplied as a byproduct of the gasification section air separation units. Ammonia produced can be further converted to ammonium nitrate, nitric acid and limestone ammonium nitrate.

Also linked to the Sasol One plant is an olefin section. Fuel gas, LPG and gasoline from Sasol I are the primary feedstock. Here, two naphtha crackers produce nearly 125,000 tonne/yr of 98% pure ethylene. In addition to

Approximately 25,000 tonne/yr of styrene and 30,000 tonne/yr of 1,3-butadiene are produced in this facility.

Sasol II and III

After the Arab Oil embargo South Africa re-evaluated its synthetic-oil production potential. This re-evaluation resulted in the construction of two new synthetic oil production facilities located at Secunda, 140 km northeast of Sasolburg and 130 km east of Johannesburg. Like Sasol I, Sasol II and Sasol III utilize Lurgi/Fischer-Tropsch technology and are mine-mouth facilities. Figure 4 is a simplified flow diagram of Sasol II which was commissioned in 1981. Sasol III is located adjacent to Sasol II and is a mirror image of this facility. Commissioning of Sasol III is scheduled for mid-1982. A view of Sasol II in the construction phase is shown in Figure 5.

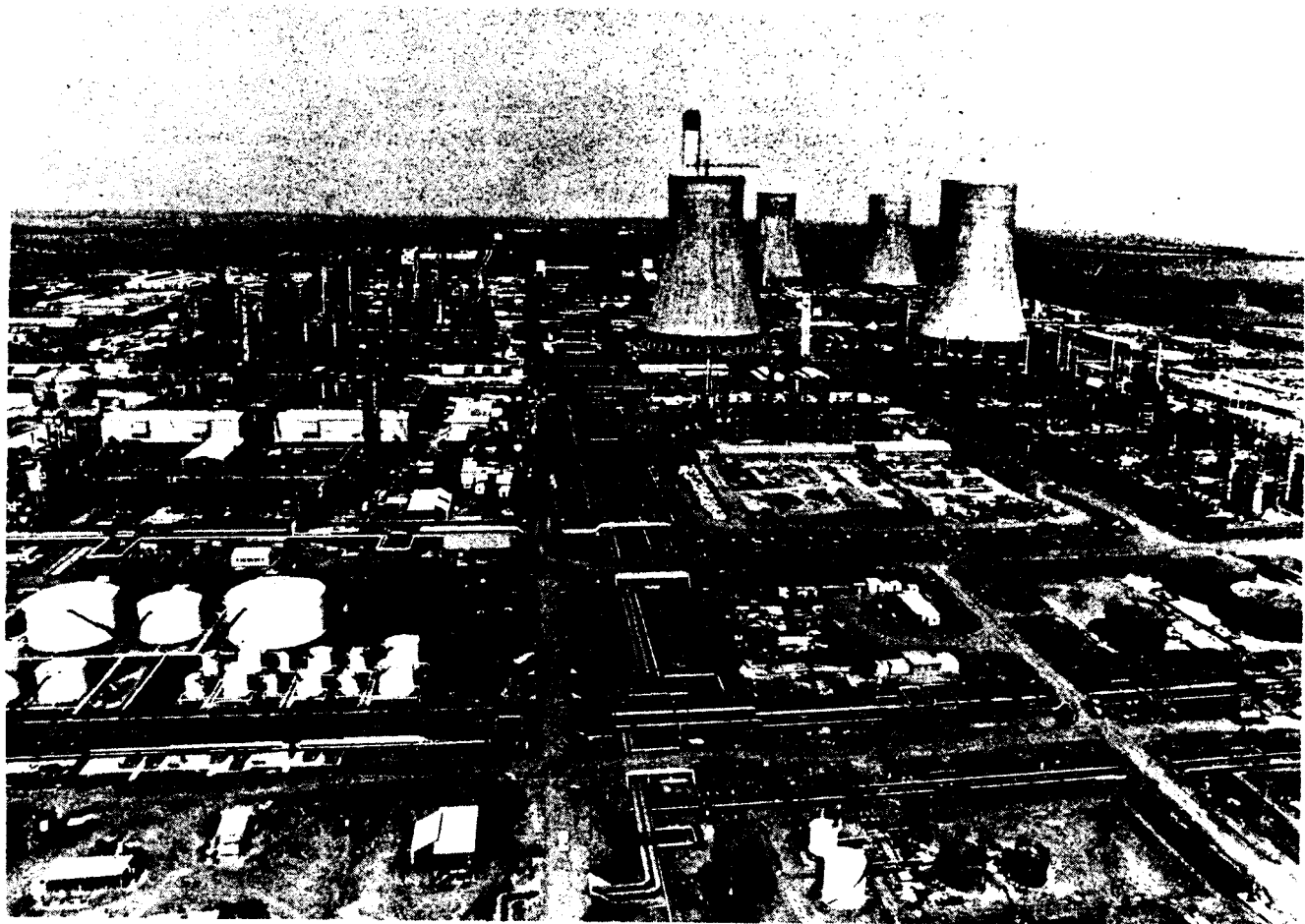


Figure 5. A VIEW OF SASOL II PLANT. TOGETHER, SASOL II AND III WILL OCCUPY 6 SQUARE MILES

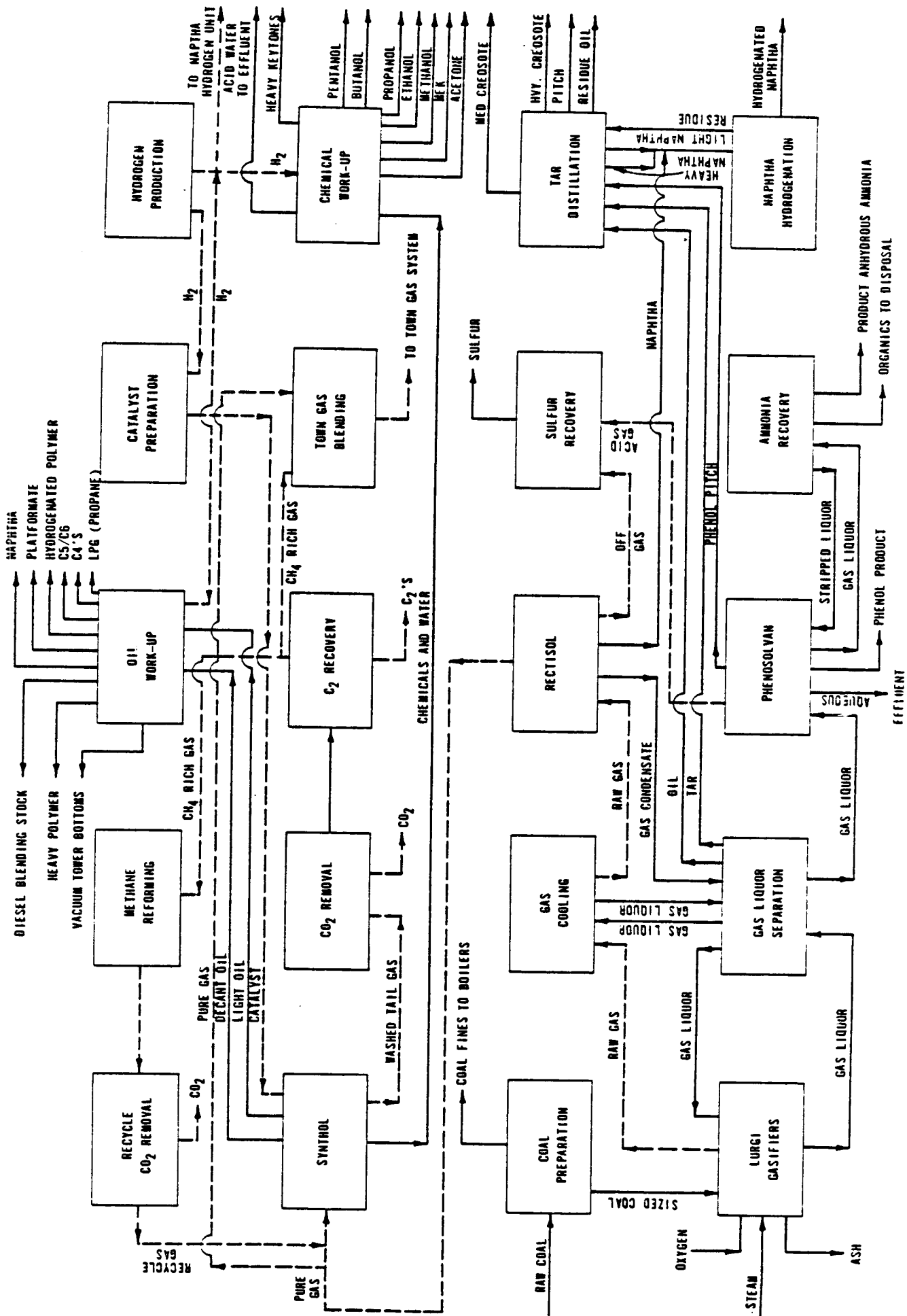


Figure 4. SOSOL II FLOWSHEET

Coal for the Sasol II and III facilities is being produced at the Bosjesspruit mine just south of the facilities. Total production of the four shaft mine which started operation in 1979 is estimated to be 27.5 million tonnes/yr. The mine, operated by Sasol I, is part of the Highveld coalfield and employs 6,600 people. The life of this mine at anticipated production levels has been estimated to be over 70 years. The typical coal properties for this relatively high quality bituminous coal are shown in Table 3. Coal from the mine is delivered to the synthetic fuels facility by conveyor.

Table 3. BOSJESSPRUIT COAL PROPERTIES

<u>Type</u>	<u>Bituminous</u>
Moisture (as received)	5.5% wt
Ash (moisture free)	22.5%
Volatiles (moisture free)	24.8%
Fixed carbon (moisture free)	52.7%
Heating value	23.9-24.5 Mj/kg
Carbon (daf)	76.9%
Hydrogen (daf)	4.3%
Sulfur (daf)	1.3%
Nitrogen (daf)	2.0%
Oxygen (daf)	13.6%
Ash properties	
Softening point	1290°C
Melting point	1330°C
Fluid point	1360°C

Sasol II is equipped with 36 Lurgi Mark IV high pressure gasifiers; 30 on line and 6 standby units. These gasifiers, which were tested for 3 years at Sasol I, weighed 140 tonnes each, have a 3.85 m internal diameter and operate at 27 bars pressure. A total of 1.65 million m³/hr of raw gas will be produced by these gasifiers with the composition shown in Table 4. Total gasifier section coal consumption will be from 25,000 to 30,000 tonne/day. In addition to coal these gasifiers will also require 30,000 to 36,000 tonne/day of high pressure steam and 8,000 to 9,000 tonne/day of 98.5% pure oxygen. Total oxygen requirements for Sasol II is nearly 12,000 tonne/day at approximately 500 psig (8,600 to the gasifiers and 3,400 to the reformers).

Table 4. RAW GAS FROM GASIFIERS AND PURE GAS FROM RECTISOL UNIT COMPOSITIONS

<u>Raw Gas Composition</u>		<u>Pure Gas Composition</u>	
H ₂	38.1 volume	H ₂	56% by volume
N ₂	0.3	CO	28%
CO	19.0%	CO ₂	1.5%
CO ₂	32.0%	CH ₄	13.5%
CH ₄	9.4%	C ₂ H ₄	0.02%
C ₂ ⁺	0.5%	C ₂ H ₆	0.20%
H ₂ S	0.7%	N ₂	0.08%
		A	0.46
		S	0.07 ppm

Gas produced in the gasification section will be scrubbed in a Phenosolvan plant to recover approximately 120,000 tonne/yr of anhydrous ammonia. Tars and oils separated from the raw gas stream before di-isopropyl treatment in the Phenosolvan plant (about 200,000 tonne/yr) will be used to produce creasotes, road tars, pitch and coal tar fuels. Naphthas will be recovered for further processing to high octane gasoline components.

After the raw gas has been scrubbed it is sent to the Rectisol section which consists of 4 process trains. These units produce about 1,100,000 m³ of pure gas with the composition shown in Table 4. Gases absorbed by the Rectisol units -70°C methanol wash are sent to a Stretford section where 90,000 tonnes/yr of elemental sulfur are recovered. Pure gas from the Rectisol section and reformed tail gas from the Synthol section are blended from the fresh feed stream to the Synthol section.

Based on Synthol and Arge operating experience at Sasol I it was decided that the Synthol Fischer-Tropsch technology was most appropriate for Sasol II and III. The Badger Companies scale up and designs were used to construct eight Synthol units at the Sasol II facility. Each unit has a capacity of about 300,000 to 350,000 m³/hr of raw gas. The Synthol reactors operate at 340°C and approximately 340 psia. It is estimated that these Synthol reactors

will consume 1.5 million m³/hr of pure gas. Catalyst for these reactors is prepared on site. The product slate produced by the Synthol section is presented in Table 5. Unreacted pure gas and tail gases from the Synthol unit are recycled to a cryogenic separation section before sending the 225,000 m³/hr of 90% pure methane output to eight methane reformer units. Methane reformer output is recycled into the Synthol reactor section.

Table 5. PRODUCT SELECTIVITY FOR THE SASOL TWO SYNTHOLS
(mass % basis)

Methane	11.0
Ethane/Ethylene	7.5
Propane/Propylene	13.0
Butanes/Butylenes	11.0
C ₅ to 195°C fraction	37.0
190°C to 400°C fraction	11.0
400°C to 520°C fraction	3.0
Heavier than 520°C fraction	0.5
Chemicals	<u>6.0</u>
	100.0

The liquid output of the Synthol reactors is sent to an oil upgrading section after oil stabilization. This section, shown in Figure 6, consists of a fractionation, vacuum distillation, naphtha hydrotreating, catalytic reforming (Platforming), catalytic condensation/polymerization, polymer gasoline hydrogenation distillate finishing and gasoline blending section. Process design for this section was done by Mobil, UOP and Linde. Total liquids production from Sasol II is estimated to be 2.1 million tonne/yr. This breaks down to 1.5 million tonne/yr of motor fuels, 160,000 tonnes/yr of ethylene, 200,000 tonnes/yr of tar product, 100,000 tonnes/yr of ammonia and 90,000 tonne/yr of elemental sulfur. Sasol III production will be similar, but emphasis will be placed on maximizing motor fuel production and minimizing other byproducts. A summary fact sheet of the Sasol technology is presented in Table 6.

Table 6 Part 1 TECHNOLOGY FACT SHEET: SASOL (Lurgi/Fischer-Tropsch)
Coal Liquefaction Process

NEW TECHNOLOGY (PROCESS DESCRIPTION AND GOALS): The Sasol technology uses commercially proven Lurgi and

Fischer-Tropsch (Synthol/Arge) processes to produce a broad spectrum of liquid and gaseous products from coal.

RELATIONSHIP TO PRIOR TECHNOLOGY (INCLUDING STATE OF DEVELOPMENT OF PRIOR TECHNOLOGY): Technology used

at Sasol is based primarily on processes developed in Germany prior to World War II.

CHARACTERISTICS OF THE TECHNOLOGY:

CHARACTERISTICS	NEW TECHNOLOGY	PRIOR TECHNOLOGY
PRIMARY OUTPUT (DESIGN CASE).....	Liquid motor fuels	
TYPE OF PROCESS.....	Indirect liquefaction	
FEEDSTOCK REQUIREMENTS.....	sized coal 12 to 50 mm	
OVERALL THERMAL EFFICIENCY (INCLUDING BY-PRODUCTS)..	55 to 60% (Sasol I)	
CARBON CONVERSION EFFICIENCY.....	98%	
OPERATING TEMPERATURE.....	1200°C (Lurgi), 340°C (Synthol)	
OPERATING PRESSURE.....	375 psi (Lurgi), 340 psi (Synthol)	
BY-PRODUCTS.....	tars, sulfur, ammonia, ethylene	

Table 6, Part 2. TECHNOLOGY FACT SHEET: SASOL (Lurgi/Fischer-Tropsch) Coal Liquefaction Process

STATUS OF DEVELOPMENT:

OPERATING FACILITIES - Sasol I at Sasolburg and Sasols II and III at Secunda

South Africa.

MAJOR FUNDING AGENCY

ANNUAL LEVEL OF FUNDING -

TECHNICAL PROBLEMS: Commercially proven for experiences gained in operation at Sasol I.

OTHER FACTORS AFFECTING OVERALL FEASIBILITY:

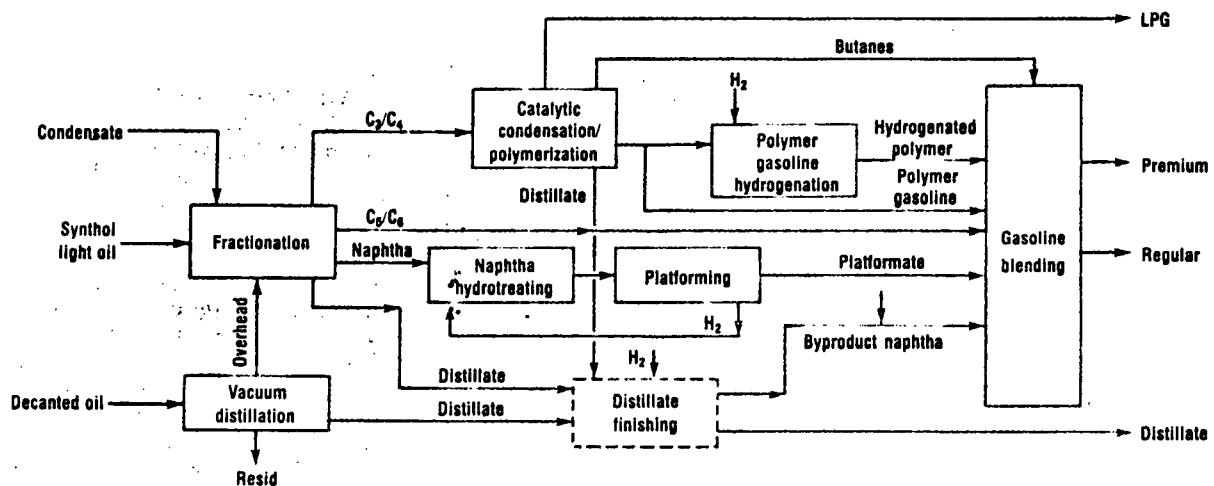


Figure 6. SASOL II OIL UPGRADING SECTION

Process Goals

The South African government has recognized the country's dependence on imports of transportation fuels as far back as 1927. Research on German indirect liquefaction technologies in the '30's and '40's culminated in the government support of Sasol I. This facility has been in operation since 1955 producing a broad range of chemical and transportation fuels. It is the government's goal to increase transportation fuel production from coal in South Africa to lessen the country's dependence on foreign imports. Sasol I, II, and III are expected to supply an estimated 40 to 50% of South Africa's liquid fuel needs by 1985.

Relationship to Prior Technology

Sasol Technology is based on Lurgi gasification and Fischer-Tropsch synthesis. The Lurgi gasification process was developed in Germany in the 1930's and the Fischer-Tropsch synthesis reaction was discovered in 1925. Within 10 years of the Fischer-Tropsch discovery the first plant was in operation. By 1944 nine F-T plants were producing 560,000 tonnes/yr of fuel in Germany. Total world F-T capacity at that time is estimated to be 1.1 million tonnes/yr.

South Africa has an abundance of minerals but in all the exploration activities no discoveries of important oil deposits have been found. In 1927, a South African White Paper was published discussing the available processes for production of oil from coal. Developments in Germany were closely followed with particular interest in the Fischer-Tropsch process. One of the

South African mining corporations, the Anglo Transvaal Consolidated Investment Co., better known as Anglo Vaal, acquired in 1935 the South African rights to the German Ruhrchemie/Lurgi Arge Fischer-Tropsch process and the American Hydrocarbon Research Inc. Hydrocol Fischer-Tropsch process.

During the next few years Anglo Vaal devoted much attention to the development of a scheme for the production of oil-from-coal. During the war and in the post-war years, Anglo Vaal remained in close contact with developments. In 1943, negotiations were held in America which led to the procurement of the rights of the American M.W. Kellogg Synthol variation of the Fischer-Tropsch process. In 1946 a new study was made and an application made to the government to create a suitable framework within which a long-term industry could be established. During 1947 the Liquid Fuel and Oil Act was passed and in agreement was reached between the South African government and Anglo Vaal in which the Anglo Vaal rights were taken over by the government. The South African Coal, Oil and Gas Corporation Ltd. was formed and incorporated under the companies' act as an ordinary public company.

Though it was clear that the plant would be based upon the synthesis of hydrogen and carbon monoxide as invented and developed by Fischer and Tropsch, it still had to be decided which processes to choose for the individual steps in this integrated complex. For gasification the Lurgi pressure gasification with steam and oxygen was selected because this process had already been demonstrated in gasifiers of a smaller size. It had the advantage of being able to work on the rather low grade, high ash coal available to Sasol. The fact that it operated at a pressure of approximately 350 psi which is also the desired operating pressure for the Fischer-Tropsch plant, was an additional advantage. This avoids cumbersome compression of large volumes of gas arising from low pressure gasification.

The raw gas from such a gasification system contains, of course, apart from the hydrogen and carbon monoxide, appreciable quantities of undesired components such as unsaturated hydrocarbons, sulphur compounds, etc. Moreover, the raw gas contains a large percentage of carbon dioxide which has to be brought down to a lower level. A number of possibilities to purify the gas existed, all involving at least two or three different process steps. However, in the late '40's Lurgi in cooperation with Linde of Germany, had developed a combined gas purification process (Rectisol) which used one solvent,

methanol, at low temperatures as the single absorption agent to remove all undesired components from the gas. A full size plant did not exist, but the pilot plant work was convincing enough to justify its selection for the Sasol plant.

The gasification process produces as a side-stream a gas liquor in which components such as ammonia and phenols are dissolved. Obviously, such a gas liquor cannot be disposed of before treatment and for this treatment another "first" was chosen, the Lurgi Phenosolvan process in which phenols are extracted from the water with a solvent such as butyl acetate. The ammonia can then be recovered by stripping with steam and converted into, for instance, ammonium sulphate. An additional advantage of having the gasification, gas purification and gas liquor treatment all from one process know-how supplier, was that the responsibility for the performance of these plants which are all to a certain extent inter-related, was concentrated with one company.

On the Fischer-Tropsch process itself the choice was not easy. On the one hand there was the German Arge design which used a fixed bed reactor system which was developed by Lurgi in Germany and known to work. The reaction took place in long tubes surrounded by a bath of boiling water for temperature control and the only difference between the small demonstration reactors and the proposed reactors for the Sasol plant, was in the number of tubes in one shell. This was not expected to give scale-up problems. On the other hand there was the American Synthol developed moving bed reactor type using a fluidized catalyst on which only pilot plant data was available but which offered the opportunity of building reactor units with a much higher capacity. Though the basic chemistry for both reactor types is the same, the fixed bed reactor produces in general straight chain hydrocarbons with a high average molecular weight and most of the production is in the range of diesel oil and paraffin waxes. The fluid bed process produces branched olefins of a low average molecular weight and the production is mainly in the range of LPG and gasoline. In view of the uncertainties the wise decision was taken to build two synthesis plants in parallel using both systems.

The original flow sheet made provision to send approximately two thirds of the pure synthesis gas to the fixed bed reactors and to send the tail gas of that system with the increased methanol content, to the reforming plant

where it was converted together with the remaining one third of the pure gas into feedgas of the right composition for the fluid bed plant. The tailgas of this plant is also recycled to the reforming units after removal of carbon dioxide. Detail design of the plants was done by M. W. Kellogg. Construction started towards the middle of 1952 and the first units were put into operation towards the end of 1954. By the end of 1955 all the main construction work was completed.

Operating Facilities

Sasol technology is being commercially utilized at Sasol I in Sasolburg and at Sasol II and III in Secunda, South Africa. Sasol I produces a host of products ranging from synthesis gas, chemicals, motor fuels to waxes, tars, and pitch. Total output is in the range of 14,600 to 18,000 bbl/day of liquid fuels and chemicals. Output for each of the Sasol II and III facilities is estimated to be 30,000 bbl/day of motor fuels 160,000 tonnes/yr of ethylene, 50,000 tonnes/yr of other chemicals, 200,000 tonnes/yr of tar products, 100,000 tonnes/yr of ammonia, and 100,000 tonnes/yr of elemental sulfur.

Major Funding Agencies

Anglo Transvaal Consolidated Investment Ltd. (Anglovaal) turned over the rights of the Sasol technology to the South African government in 1950. The government in turn formed the South African Coal, Oil and Gas Corporation Limited with funding coming from the treasury via the Industrial-Development Corporation (IDC). The IDC is the state corporation that has provided much of the venture capital to such high risk projects. The first Sasol facility at Sasolburg was constructed with this government funding at an estimated cost of \$230 million in 1952-1955. The IDC also provided the loan capital for expansion activities at Sasol I throughout the years. As sole owner of Sasol I the IDC received all profits.

The financing of Sasol II will come from the State Oil Fund (1.711 billion Rands), export credits (492 million Rands), and parliamentary grants (300 million Rands). Export credits will provide 20% of the financing of Sasol III; the balance will be provided by the State Oil Fund, parliamentary grants, and the proceeds of two stock issues in a newly formed company called Sasol Ltd. The issues were made in September and October of 1980. 245 million shares were offered to institutional investors and 17 million

shares to the public. Cost per share was 2 Rands. The response was tremendous, with more than \$1 billion offered for each of the two issues. These shares account for 70% of the stock in the new company Sasol Ltd.; the remaining 30% is assigned to the Industrial Development Corp. (IDC) and its subsidiary Konoil Ltd. Sasol Ltd. will have 100% ownership in Sasol I and 50% ownership in Sasol II and III; the balance will be controlled by the state through IDC/Konoil. Once Sasol II and III are operative, further shares will be offered to the public from time to time. This dilution of government involvement reflects policies aimed at limiting the government's role in the economy.

In 1979 when Sasol was restructured, Sasol Limited was created as the new holding company. The new corporate structure is shown in Figure 1 along with ownership percentages. The old Sasol is now known as Sasol I and is a wholly owned subsidiary of Sasol Limited. The Sasol I subsidiaries that have developed throughout the years to give the company its diversified energy business are shown in Table 7. This table presents the Sasol I subsidiaries and their principal activities. In addition to the companies presented in Figure 1 and Table 7, Sasol also oversees South Africa's strategic petroleum reserve which has been stored in underground coal mines.

Technical Problems

Sasol I has been commercially proven with over 25 years of operation. Sasol II which started full operation in 1981 did not experience any major startup problems. Sasol III which will be fully commissioned in 1982 is not expected to have any startup or operating difficulties since it is based entirely on technology used in Sasol II.

Capital Costs

See Major Funding Agencies section.

Table 7. SASOL ONE SUBSIDIARY COMPANIES

1. Sasol Marketing Company (SMC) — markets petroleum products of the Sasol group excluding road binder material, tar and bitumen.
2. South African Gas Distribution Corporation (Gascor) — Distributes industrial gas by pipeline from Sasol One to the industrial complex of the Witwatersrand and Vaal Triangle. It has also been designated as the distributor of any natural gas that may be discovered in South Africa.
3. National Petroleum, Refiners of South Africa (Natref) — Refines crude oil which is pumped by pipeline from the coast 600 km away for Sasol One into LPG, gasoline, diesel oil, kerosene, jet fuel, bitumen and other products. Sasol One owns 52.5%. The other partners are the National Iranian Oil Company (NIOC), 17.5% and Compagnie Francaise des Petroles (Total), 30%.
4. Sasol Dorpsgebiede (SDB) — Undertakes township development at Sasolburg and provides housing for the Sasol's group's employees.
5. Inspan Beleggings — Holds the major portion of the coal rights of Bosjesspruit's coal fields.
6. Leslie Coal Development Company — Holds Sasol One's longer-term coal rights.
7. Allied Tar Acid Refiners (Atar) — Refines tar acids in one of Sasol One's factories. Atar also markets phenols and cresylic acid through SMC.
8. Tosas — Holds a 50% interest in FTS Binders which in turn markets road binder material, tar, and bitumen.
9. Southern Oil Exploration Corporation (Soekor) — Sasol One has a 50% share of Soekor which is to lead and coordinate the search for oil in South Africa on behalf of the government with state funds.
10. Fedgas — Markets industrial gases such as oxygen nitrogen and argon some of which are supplied by Sasol One. Sasol One has a 20% interest in Fedgas.
11. Inspan Bedryf, Sasol Konstruksiemaatskappy and naftachem are dormant companies.

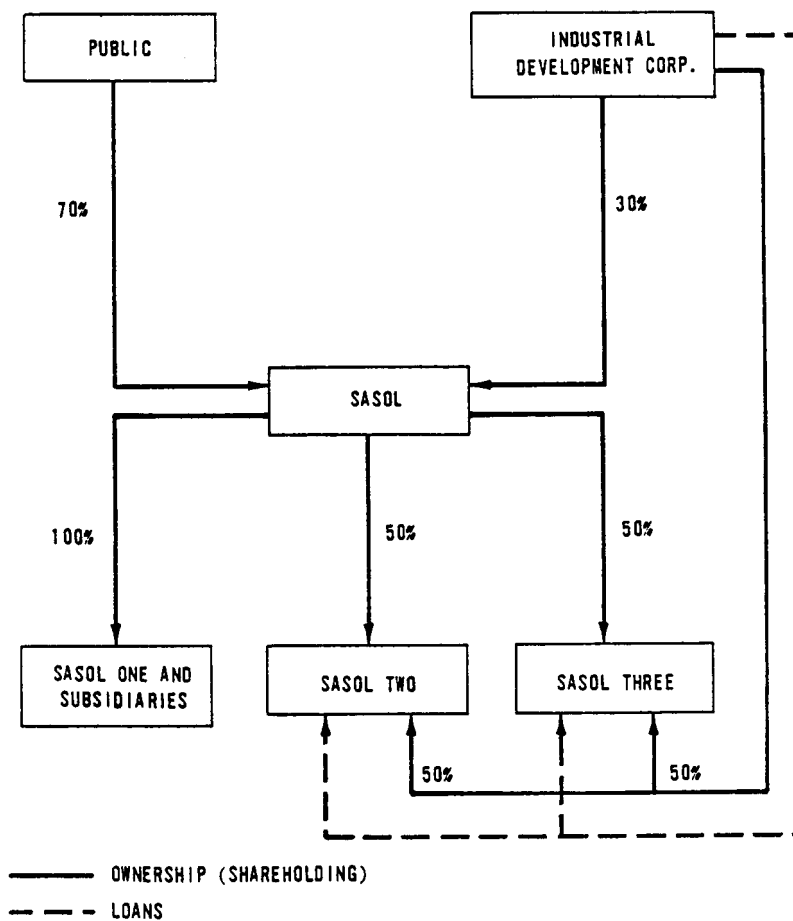


Figure 7. SASOL GROUP ORGANIZATION

SOUTH AFRICAN COAL CONVERSION PROCESS

24. Modderfontein (African Explosives and Chemicals Industries Ltd.)
Coal-to-Methanol ProcessProcess Description

The AECI Ltd. has proposed to produce 800,000 metric tons/yr of methanol from coal for blending into gasoline stocks. The approach now being employed at their Modderfontein facility to produce methanol uses commercially available Koppers-Totzek (K-T) and ICI technology. Current capacity of the Modderfontein facility is 1,000 metric tons/day of ammonia and approximately 90 metric tons/day of methanol.

The K-T gasifiers currently used at the Modderfontein facility are of the two-head design, but future expansion plans will probably employ the four-head design. Both designs are based on high-temperature, atmospheric-pressure, entrained-bed concepts which utilize pulverized coal. This process was first developed in Germany in the 1930's. The gasifier's characteristics enable it to use most types of coal to produce a clean synthesis gas consisting of chiefly CO and H₂, with few if any hydrocarbon contaminants.

The coal feedstock for the Modderfontein facility is delivered by rail from a mine 90 km away. A typical analysis of the feed coal is shown in Table 1. The coal is unloaded by one person from the incoming coal cars to a conveyor belt which is also operated by one person. Coal is stored in bunkers which is typically sufficient for two weeks of operation. During winter months the coal storage is larger due to increased regional demand. From coal storage the coal is pulverized and simultaneously dried to about 1.5% moisture in two ring and ball mills. The resulting coal dust particles are typically sized 90% less than 90 μ m. The pulverized coal is transported to a network of bunkers, and fed to the gasifier via screw feeders.

The prepared coal entering the gasifier is entrained into a stream of premixed oxygen and steam and the reaction mixture enters the gasifier via burner nozzles located in the gasifier heads. Very rapid exothermic reactions occur causing the temperature in the flame to exceed 2000°C. Subsequent endothermic reactions and cooling by the steam jacket gasifier wall reduces the overall temperature within the gasifier to 1600°C. Residue time of the coal in the gasifier is typically about 0.5 to 1 second. The Modderfontein facility currently utilizes six two-headed K-T gasifiers. A diagram of the K-T two headed gasifier is shown in Figure 1.

Table 1. TYPICAL ANALYSIS OF GASIFICATION FEED COAL

<u>Ultimate analysis (dry basis) % m/m</u>	
C	64.3
H	3.7
N	2.3
O	8.6
S	0.6
Ash	20.5
Inherent moisture, % m/m	1.9
Volatile combustible matter, % (air dry basis)	26.1

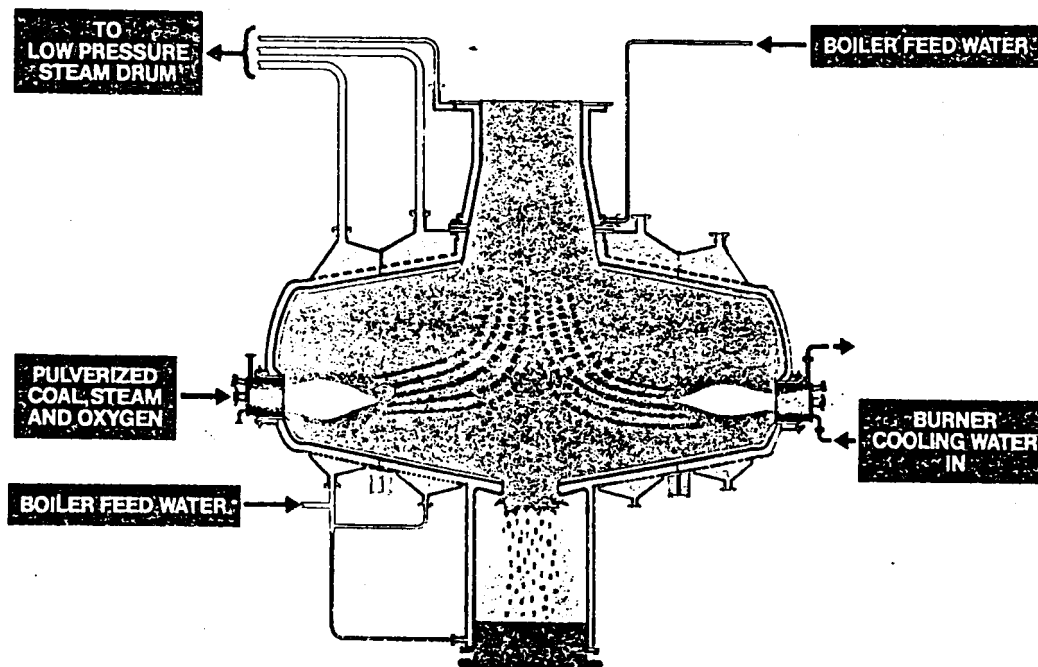


Figure 1. K-T GASIFIER

A portion of the mineral matter in the coal is slagged in the gasifier and impinges on the walls where it accumulates and flows down to a slag outlet in the base. The molten slag is then quenched and granulated in a water bath and removed by a continuous scraper system. The remaining mineral matter leaves the gasifier as fly ash with associated unreacted carbon. The exit temperature of 1600°C requires that the still molten fly ash be quenched with direct water injection to about 900°C. This avoids fouling in the waste heat boiler system where steam is raised at 55 bars. The gas is further contacted with water in a washing tower where most of the solids are removed. The synthesis gas is subjected to further dust removal before passing to a raw gas recompression section prior to gas purification. A typical composition of the raw gas at this point is shown in Table 2.

Table 2.

Typical Analysis of Raw Gas by Volume I (Dry Basis)

CO	58%
H ₂	27%
CO ₂	12%
CH ₄	100 ppm
H ₂	0.5%
COS	0.04%
SO ₂	0.1 ppm
HCN	100 ppm
NO	30 ppm
NH ₃	15 ppm
N ₂	0.9%
Ar	0.6%
O ₂	100 ppm

A block flow diagram of the whole coal to methanol process is shown in Figure 2. The dust-free raw gas from the gasification plant is compressed to 30 bars in 2 parallel stream turbine-driven raw gas compressors. The compressed gas is then sent to the first stage of the two-stage Rectisol gas purification unit. Gas entering this stage is first treated by a water absorber to remove HCN and then scrubbed with cold methanol to remove H₂S and

COS to less than 1 ppm. Significant amounts of CO₂ are not removed in this unit.

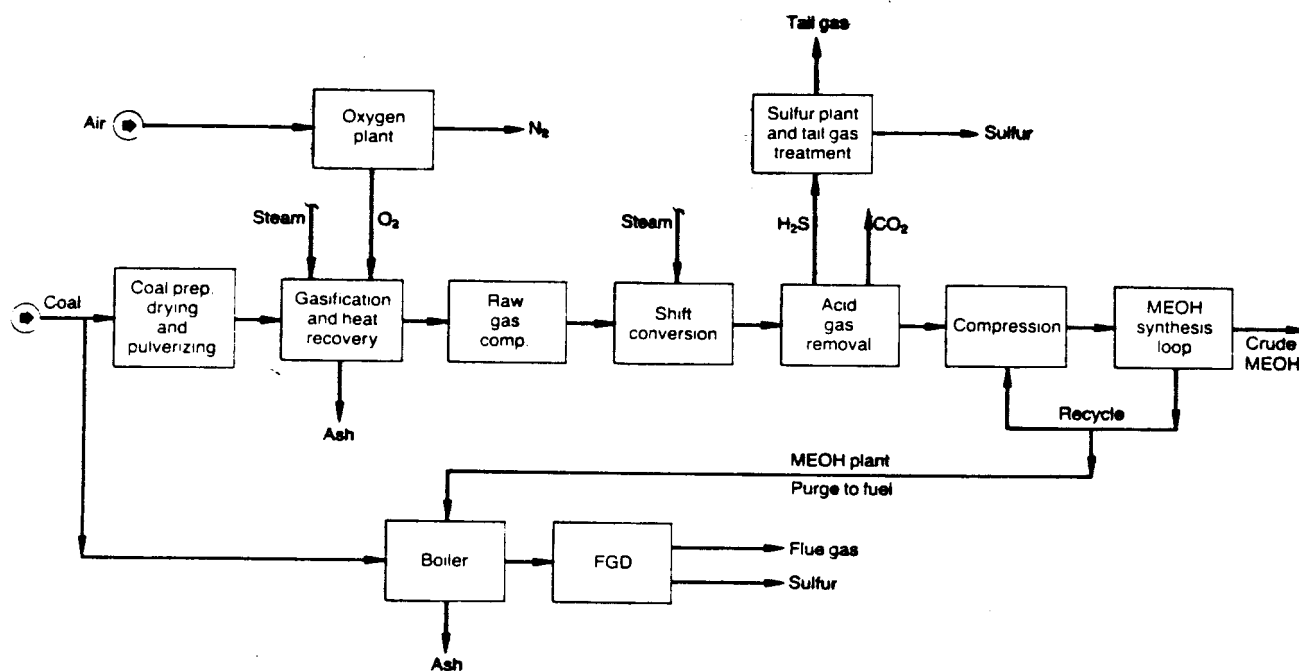
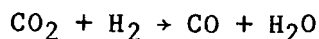
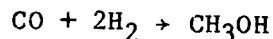


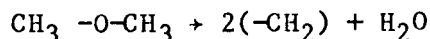
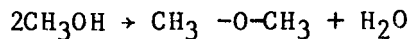
Figure 2. COAL-TO-METHANOL PROCESSING USING KOPPERS-TOTZEK GASIFICATION PROCESSING

The gas is then recompressed to 50 bars before injection into the water-gas shift unit where the CO-to-H₂ ratio is adjusted for the methanol synthesis reaction. The methanol synthesis process used in the Modderfontein plant is of the ICI design. This unit, shown in Figure 3, consists of compressing the makeup gas, mixing this makeup gas with recycle gas, and then feeding the mixture to the methanol converter. The two overall reactions in the catalyzed bed occur as follows:



The hot effluent is cooled by a waste heat recovery unit, heat exchange with incoming feed, and cooling water. The liquid-rich stream is finally flashed to remove gases, which are then recycled to the reactor. The catalytic methanol reactor uses a copper-based catalyst to operate at a temperature of 400° to 575°F and a pressure of 750 to 1,500 psig. The crude methanol requires further distillation to produce a pure product for more fuel blending or can be sent to a Mobil MTG reactor to produce gasoline.

In the Mobil process, shown in Figure 4, methanol is partially dehydrated to an equilibrium mixture of methanol, dimethyl ether, and water over a dehydration catalyst in a limited reactor. In a second reactor, a zeolite conversion catalyst is used to convert both methanol and dimethyl ether to high-octane gasoline. During this operation recycled gas is used as a heat sink to remove the exothermic reaction heat. In the overall process the reaction can be described as follows:



Typical yields of the raw product from this process are presented in Table 3. The hydrocarbon product is primarily gasoline which must undergo further treating to add butanes and alkylate the butenes and propylenes. The finished gasoline product in Table 4 typically has a Rvp of 9 psig and unleaded RON of 93.

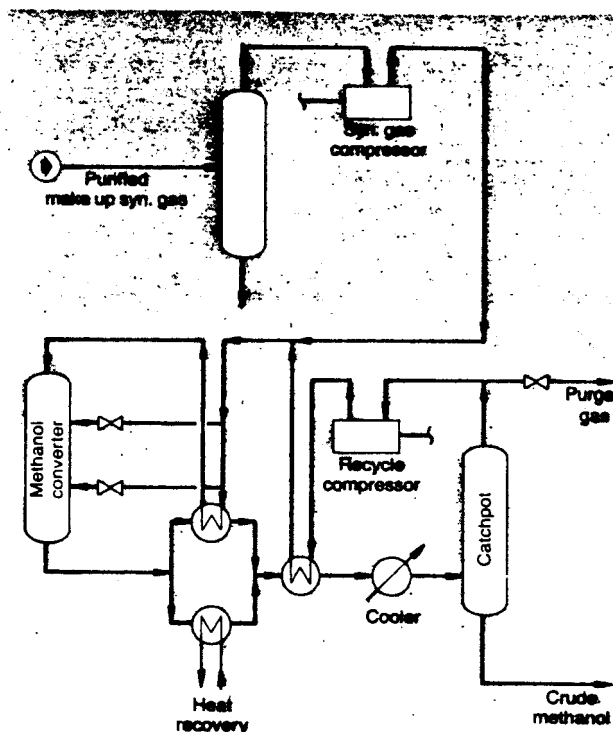


Figure 3. METHANOL SYNTHESIS

Table 4. TYPICAL YIELDS FROM CRUDE METHANOL USING
FIXED BED PROCESS

Yields, wt % of charge		Components, wt. %	
Methanol + ether	0.0	Butanes	2.7
Hydrocarbons	38.2	Alkylate	3.2
Water	63.2	C ₃ + synthesized gasoline	94.1
CO, CO ₂	0.3		100.0
Coke, other	0.2		
	<u>100.0</u>	Composition, vol %	
Hydrocarbon product, wt %		Paraffins	51
Light gas	1.4	Olefins	13
Propane	5.5	Naphthenes	8
Propene	0.2	Aromatics	28
i-Butane	8.6		100
n-Butane	3.3		
Butenes	1.1	Research octane	
C ₅ + gasoline	79.9	Clear	93
	<u>100.0</u>	Leaded, 3 cc TEL/U.S. gal	100
		Reid vapor pressure, psig	9.0
Gasoline (including alkylate)	85.0	Specific gravity	0.730
	93 RON clear, 9 Rvp)	Sulfur, wt %	NI
LP gas	13.6	Nitrogen, wt %	NI
Fuel gas	1.4	Corrosion, copper strip	1A
	<u>100.0</u>	ASTM distillation, °F	
		10%	114
		30%	145
		50%	198
		90%	330

*Methanol with 17 wt % H₂O

About 3000 stpd methanol (100% basis) are required to produce 10,000 bpd total liquid products (gasoline plus LPG)

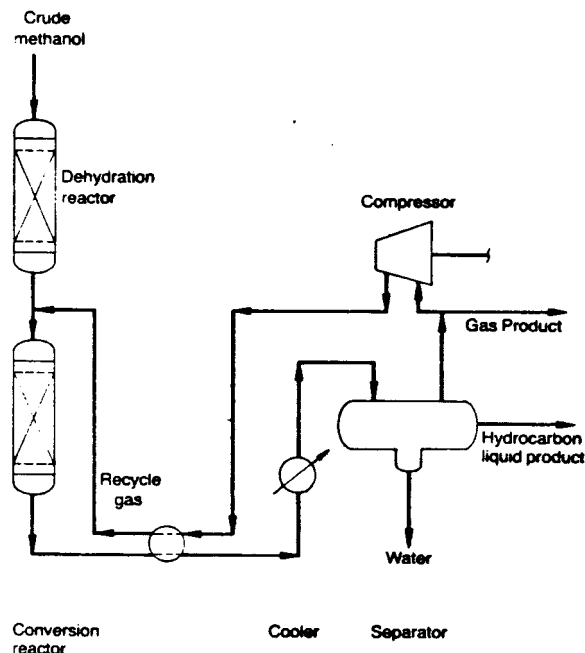


Figure 4. MOBIL METHANOL-TO-GASOLINE PROCESS — FIXED BED OPERATION

Process Goals

In the spring of 1980 AECI Ltd. announced plans to construct an 800,000 metric ton/yr methanol facility based on coal gasification technology. This decision stemmed from a South African government announcement to grant private-enterprise fuel-from-coal tax breaks similar to those granted SASOL. The methanol produced in the AECI Ltd. facility would be used as a blending feedstock (up to 10%) to stretch existing gasoline and diesel supplies. However, reports from the AECI Ltd. in 1981 indicated that methanol may not be more desirable. Plans to go ahead with this development have yet to be announced.

Relationship to Prior Technologies

The technology used to produce methanol has already been commercially developed. This includes the K-T gasifier which was developed in Germany in the 1930's and the ICI methanol process developed in England in the late 50's and early 60's. If gasoline is to be the final product, the Mobil MTG process will also be required. This process is currently in the pilot plant phase, but, a large commercial facility is being built in New Zealand using the Mobil MTG process.

Operating Facilities

AECI Ltd. currently operates a 1000 metric ton/day ammonia plant and a 90 metric ton/day methanol plant which uses coal as the feedstock. This

facility at Modderfontein, South Africa, started operation in 1972. This complex, which is the largest ammonia plant in South Africa, took 30 months to construct.

Major Funding Agencies

Privately financed by AECI Ltd.

Technical Problems

The Modderfontein plant can exceed its 1000 metric tons/day design capacity. The plant consists of six gasifiers, but five gasifiers are sufficient to reach the ammonia plant design capacity. The gasifiers are arranged in a pair of three. At start-up, the gasifiers ran at 70% of capacity, and eventually reached 130% of design capacity. They expect greater turndowns are possible with the four-headed gasifier. AECI personnel felt that a spare gasifier would have been a desirable feature. Of course, this is easier to justify on large plants. Gasifiers can easily be taken on and off stream for brief periods of time (say, one hour); they are kept hot during standby by means of integral burners. They said the gasification efficiency varies with the type of coal. They strongly recommend full-scale testing of coals prior to selection system. They found the gasifiers sensitive to the type of coal fed.

They estimate that the No. 4 plant has twice the amount of equipment a conventional natural gas or oil based ammonia plant would have. Thus one would expect more difficulty in maintaining high reliability. (They did not want to discuss on-stream factors.) A chart on the meeting room wall during a trip there in 1981 indicated that actual production was nearly equal to scheduled production for the previous few months.

Their main problems are associated with the stoker-type boiler. No redundancy was built into the system, hence they often have difficulty supplying adequate steam. There are also considerable time delays when additional steam is demanded, further complicating operations. Cycling the steam output is hard on the boiler components and contributes to increased maintenance. They reminded us that the pulverized coal alternative to a stoker is more difficult to operate. Another plant problem mentioned is erosion of process surfaces. These repairs are made during their overhaul period, which occurs every two years (standard practice for the chemical process industry).

They have solved the refractory lining problems that were encountered earlier. They stated that in general their plant experiences more corrosive contaminant than a natural gas-based ammonia plant. They have been able to slowly increase the intervals between certain maintenance procedures through the years as they became more familiar with the plant. In their opinion gas is easy to make compared to the remaining processes in the plant.

They strongly felt that all portions of the process should be highly instrumented because of the inherent possibility of fire, explosions, and toxicity dangers. They are currently in compliance with all environmental regulations, although they were having some recent dust problems from the coal milling section (that had been previously reported in the literature as well, indicating a possible chronic problem). They must obtain an operating permit from the Air Pollution Control Officer, who has the power to shut down the facility. They enjoy amiable relations with the environmental authorities. South African law requires that pressure vessels be tested every four years; this is done during one of the overhauls scheduled every two years. They currently burn the hydrogen sulfide removed from the process. They envision two alternatives to burning: production of sulfur from the gas via a Claus plant, or production of sulfuric acid. They warn that heavy metals in coal ash may be a problem in the U.S. (they are not experiencing problems). They see no reason not to recycle about 20% of their process water.

They store a minimum coal supply of two weeks. During winter, when demand is high, they prefer to have a larger supply. Coal supply problems have not occurred.

The plant has not suffered from labor problems. Labor is represented by unions according to race. The plant is a closed shop, but not all labor is unionized. The plant uses a four-shift system with three shifts per day. Gasification labor requires a part-time foreman, one supervisor, one operator in the control room, two outside laborers, and less than one auxiliary laborer.

The coal supply section is less sophisticated and requires one laborer to unload the coal cars, one laborer to oversee conveyor operations and one laborer in the coal mill. They estimate skilled labor receives a net salary of R12,000 (\$15,000) and unskilled, R6,000 (\$7,500). If labor were more expensive they postulated that they might increase automation in the coal

handling section to eliminate one or two positions, or simply require each worker to be responsible for more process operations. They feel that their labor productivity is typical for South Africa.

The major process industry differences they see between the U.S. and South Africa are coal and electricity prices. Typical coal prices are R6 and R9 per ton (this is about \$0.50 to \$0.70 per million Btu). Electricity in the vicinity of the plant costs about R0.01 per kWh (13 U.S. mills/kWh) and in the Cape about R0.021 per kWh (26 U.S. mills/kWh).

Capital Cost

Construction of the 800,000 metric ton/yr methanol plant was estimated to be \$549 million in 1980.

SOUTH AFRICAN COAL LIQUEFACTION PROCESS

25. Sasol Direct Coal Liquefaction ProcessProcess Description

Little, if any, information has been published on Sasol's direct liquefaction technology development program. Information published by the Nippon Brown Coal Liquefaction group of Japan has indicated that Sasol has developed an SRC-II type pilot plant process which utilizes catalytic hydrogenation technology. However, published verification of this facility has not been found. In addition, Sasol officials indicated in mid-1981 that direct coal liquefaction technologies were not superior to their indirect process, especially when taking South Africa's poor coal quality into consideration.

However, bench-scale experiments have been conducted by the Fuel Research Institute of South Africa on catalytic hydroliquefaction of South African bituminous coals. In these experiments four different liquefaction reactors were tested on one South African coal type. The proximate and ultimate analysis of the bituminous coal used in these experiments is shown in Table 1.

Table 1. ANALYSIS OF WATERBERG TRANSVAAL COAL USED^a

Moisture	(wt % air dried basis)	3.4
Ash	(wt % air dried basis)	12.7
Volatile Matter	(wt %, air dried basis)	34.8
C (wt % daf)		80.7
H (wt% daf)		5.5
N (wt % daf)		1.5
S (wt % daf)		1.0
O (wt %, daf) (by difference)		11.3
Vitrinite (vol %)		83.2
Exinite (vol %)		4.2
Inertinite (vol %)		5.7
R ₀ (mean maximum reflectance of vitrinite)		0.720

^a 0.5 — 0.25 μ m (30 — 60 mesh)

The first process is hydrolysis (dry hydrogenation) with a "hot-rod" reactor,²⁻⁵ operated in a semi-continuous mode. In this reactor the coal is

heated at about $200^{\circ}\text{C min}^{-1}$ while secondary reactions are minimized by removing and quenching the products. The second process was hydrolysis in a rotating autoclave with a heating rate of about $7^{\circ}\text{C min}^{-1}$ and retention of the product at reaction conditions for 1 hr. The other processes were supercritical gas extraction using toluene with and without hydrogen. These extractions were carried out in an autoclave with a lengthy heating-up period and in a modified "hot-rod" reactor with reasonably rapid heating. Identical temperatures and pressures were used either with the same concentration of catalyst or without catalyst. In all the experiments, sand was mixed with the coal to limit agglomeration.⁶ Thermolysis of supercritical toluene is a problem; however, the products of the thermal breakdown are known.⁷

Liquefaction Procedures

Method A (Runs 1 and 2). Hydrogenation was carried out in a "hot-rod" reactor.²⁻⁵ The coal (25 g), impregnated with stannous chloride catalyst (tin 1 wt % of the coal) for the catalyst run, was mixed with sand (1:2 by weight). The reactor was heated (about $200^{\circ}\text{C min}^{-1}$) to 450°C and maintained at this temperature for 15 min. Hydrogen (20 MPa; 221 min^{-1}) was passed through the fixed bed of coal/sand/catalyst. The products were condensed in high-pressure cold traps.

Method B (Runs 3 and 4). The reactor was a 1 liter rotating autoclave fitted with a glass liner. Coal (50 g) sand catalyst preparation procedures were the same as in Method A. The charge was heated (about $7^{\circ}\text{C min}^{-1}$) under hydrogen to 450°C and maintained at this temperature for 1 h (pressure 20 MPa).

The product was washed from the cooled reactor system with toluene. The solid residue was extracted with boiling toluene (250 ml) in a Soxhlet extractor for 12 h. The toluene solutions were combined and the toluene removed under reduced pressure. Hexane (250 ml) was added to the extract and it was allowed to stand for 24 hr with occasional shaking. The solution was filtered to leave a residue (asphaltene) and the hexane was removed from the filtrate under reduced pressure to give the oil. The residue in the Soxhlet thimble after toluene extraction was extracted with pyridine in a Soxhlet extractor to give pre-asphaltenes (toluene-insoluble pyridine-soluble product).

Method C (Runs 5-8). Supercritical gas extraction was carried out in a stirred 1 liter autoclave. Coal (100 g) sand catalyst mixtures were the same as that for Method A. In the runs without hydrogen, the autoclave was charged with coal/sand/catalyst and 600 ml of toluene and heated. It took about 1 h to reach the required temperature and pressure. On reaching the desired temperature (450°C) and pressure (20 MPa), toluene (about 2 liter h⁻¹) was pumped into the reactor through the coal bed via a dip tube. The toluene extract was cooled at atmospheric pressure in a water-cooled condenser. The runs with hydrogen were similar except that only 300 ml of toluene was loaded into the autoclave, which was then flushed and pressurized with hydrogen to a cold pressure of 5 MPa. After attaining the reaction temperature and pressure (450°C and 20 MPa), the autoclave was maintained at these conditions for 1 h before starting to pump the toluene.

The extraction condensate was filtered to remove material which precipitated on cooling. This residue was soluble in pyridine and designated as pre-asphaltene. The toluene was removed from the filtrate under reduced pressure, and this product was then treated with hexane as described to give the asphaltene and oil fractions.

Method D (Runs 9-12). The apparatus used was the same as for Method A except that provision was made for the introduction of toluene as well as hydrogen. Coal/sand/catalyst preparation procedures were as for Method A. In the runs without hydrogen, toluene (4 liter h⁻¹; 20 MPa) was passed through the reactor for 15 min on attaining the reaction temperature. In the runs with hydrogen, hydrogen (8 liter min⁻¹) was simultaneously passed through the reactor. The pressure was maintained at 20 MPa. The toluene extract was collected in the condenser system.

The conversions and product distributions are shown in Table 2. In the case of supercritical gas extractions without hydrogen (Runs 5, 6 and 9), the yield of extract was greater than the conversion due to the thermal breakdown of toluene. The gas yields (including water) were obtained by difference and in the experiments where supercritical toluene was used, these will be low owing to errors caused by formation of toluene pyrolysis products. G.I.C. analysis of the oils from supercritical amounts of bibenzyl and the other toluene pyrolysis products.

Molecular weights, viscosities, and sulphur analysis are given in Table 3. Sulphur analysis was not carried out on the supercritical gas oils (Runs 5-8) because of the large amount of toluene pyrolysis product in the oils. Thermogravimetry in nitrogen of the four hydrogenation oils (Samples 1-4) were very similar. About 60% weight loss had occurred at 200°C and essentially 100% weight loss had occurred by 400°C. Thermogravimetric analysis in nitrogen of the asphaltenes showed that the asphaltenes from the supercritical gas extraction without hydrogen assistance were less volatile than the other asphaltenes.

The fractions from the silica gel chromatographic separation were grouped as aliphatic hydrocarbons, aromatic hydrocarbons, and polar compounds. The results for the oils from the hydrogenation samples are summarized in Table 4. The samples from the rotating autoclave contain a lower percentage of polar compounds than those obtained from the equivalent "hot-rod" experiment. Because of the large quantity of toluene pyrolysis product in the oils from the supercritical extractions, these oils have not been grouped as aliphatic, aromatic, and polar compounds.

Table 2. PROCESS. CONDITIONS. CONVERSION AND PRODUCT DISTRIBUTION

Run	Process ^a	Conditions	Conversion Extract yield Oil Asphaltene Pre-asphaltene Gas					
			(wt % coal, daf)					
1	HR	H ₂	38.7	—	13.2	13.2	6.8	5.5
2	HR	H ₂ , SnCl ₂	90.6	—	29.4	10.9	5.9	44.4
3	RA	H ₂	56.1	—	13.0	3.5	3.1	36.5
4	RA	H ₂ , SnCl ₂	79.6	—	24.5	6.8	1.8	46.5
5	SCGE	—	32.4	39.6	14.7	13.8	10.2	—
6	SCGE	SnCl ₂	29.0	36.0	14.4	11.6	8.2	—
7	SCGE	H ₂	41.7	32.4	16.8	13.1	2.7	9.3
8	SCGE	H ₂ , SnCl ₂	69.5	52.1	19.9	25.1	4.4	17.4
9	SCGE/HR	—	31.2	32.6	8.6	15.4	8.6	—
10	SCGE/HR	SnCl ₂	32.6	31.4	7.3	15.6	8.5	1.2
11	SCGE/HR	H ₂	34.0	24.0	7.7	13.2	3.1	10.0
12	SCGE/HR	H ₂ , SnCl ₂	49.2	44.5	12.0	26.4	6.1	4.7

^a HR, Hot-rod reactor; RA, rotating autoclave; SCGE, supercritical gas extraction using autoclave; SCGE/HR, supercritical gas extraction using hot-rod reactor

Table 3. MOLECULAR WEIGHT, VISCOSITY AN SULPHUR ANALYSIS OR PRODUCT

Run	Molecular wt.		Viscosity of oil (mPa s)	Wt % S in oil
	Oil	Asphaltene		
1	221	398	62	0.40
2	197	330	40	0.33
3	194	352	33	0.30
4	194	351	17	0.24
5	214	487	15	
6	215	491	13	
7	222	427	34	
8	210	428	22	
9	237			0.52
10	235			0.51
11	248			0.46
12	230			0.32

Table 4. COMPOSITION OF THE HYDROGENATION OILS

<u>Run</u>	<u>Alphatics (wt % oil)</u>	<u>Aromatics (wt % oil)</u>	<u>Polar compounds (wt % oil)</u>
1	8	48	44
2	6	55	39
3	5	59	36
4	4	70	26

Process Goals

The conversion of coal to liquid and gaseous products was highest for the "hot-rod" reactor when a catalyst was used (Table 2). The conversion was much lower without a catalyst. Using the rotating autoclave the effect of the catalyst was far less pronounced presumably owing to the much longer residence time. As expected, when the product is held in the autoclave, gas production at the expense of liquid product increases. The amount of asphaltenes and pre-asphaltenes²² (or asphaltols²³) is higher for the "hot-rod" reactor experiments than for the autoclave experiments. With time, asphaltenes and pre-asphaltenes are converted to oil and gas. The higher conversion obtained with the "hot-rod" reactor with a catalyst compared to the equivalent rotating autoclave run may indicate that polymerization of part of the product does occur to give mainly a pyridine-insoluble char. However, it is possible that the higher rate of heating in the "hot-rod" reactor may produce more intense thermal fragmentation and thus higher conversion.

Different hydrogenation processes even when carried out at the same pressure and temperature on the same coal, give considerably different products. The liquid product from hydrogenation in a rotating autoclave contains less hetero-atoms and is more aromatic than the product from a short residence time semicontinuous reactor working at the same pressure and temperature. These differences may have important implications for the further processing of the product to transportation fuels and petrochemicals. The long residence time of batch autoclaves tends to "mask" the effect of a catalyst compared to short residence time reactors. Therefore, catalyst screening in batch autoclaves may only be of limited value when applying these results to short residence time reactors. Supercritical gas extraction gives a more aliphatic liquid product, and even in the presence of hydrogen and catalyst is a milder process than hydrolysis at the same

temperature and pressure. The work with supercritical gas extraction has shown the problem that solvent breakdown can have on the yield and the nature of the liquid obtained in a batch process.

Relationship to Prior Technology

Based on catalytic hydrogenation and other technologies; however, a process configuration has not been established.

Operating Facilities

Bench-scale studies are being carried out at the Fuel Research Institute Laboratories at Pretoria, South Africa.

Technical Problems

Technology is not mature enough to project technical problems.

Capital Costs

Technology is not mature enough to project capital costs.

INDIA COAL GASIFICATION PROCESS

26. Central Fuels Research Institute (Lurgi) Coal Gasification ProcessProcess Description

The conventional Lurgi process pilot plant was installed and commissioned in 1962. The plant has a capacity to gasify 1 tonne of coal per hour. The gasifier is a conventional rotary grate type gas producer designed to operate up to a pressure of 32 kg/cm². The gasifier shell, 1000 mm dia. and 5800 mm length, is a double-walled alloy steel vessel of welded construction. In the annular space between the walls, steam is raised at gasification pressure. The inside of the gasifier is partly lined with refractory bricks in the combustion zone. A rotating coal distribution (speed 7-45 rpm) is fitted inside the gasifier at its top for maintaining a fuel bed of constant height. The grate at the base of the generator can be rotated at speeds varying from 4 to 24 rpm. A coal lockhopper chamber for feeding the coal and an ash lockhopper for ash discharge are provided at the top and bottom of the gasifier, respectively. The tar-laden hot gases emerge out of the gasifier at a temperature of 400°-500°C. The plant is equipped with gas cleaning system, tar recovery system, and safety devices with adequate instrumentation wherever necessary. The flow diagram of the system is given in Figure 1. The performance of this gasifier on a number of Indian coals is presented in Table 1.

Process Goals

The goal of this research effort is to study the gasification characteristics of noncaking Indian coals with steam and oxygen. The primary application of this conventional technology is the production of ammonia. The gasification characteristics of the following noncaking coals were studied in detail:

- Talcher, bottom seam of Talcher coalfields in Orissa.
- King seam coal of Singareni (Kothagudem) field in A.P.
- Dobrana seam coal of Raniganj field in West Bengal.
- Burhar seam coal of Shahagpur in M.P.

Relationship to Prior Technology

This project is based on the commercially available Lurgi gasification process.

Table 1. SOME TYPICAL OPERATION DATA OF PRESSURE GASIFICATION TESTS

Name of Seam	Surkachar	Lower Kenda	Samla	Burhar	Singareni	Dobrana		Talcher
						Sample 'A'	Sample 'B'	
Gasifier pressure, kg/cm ² (g)	10.6	14.0	21.1	21.1	21.1	21.1	21.1	25.0
Dry coal feed rate, Kg/hr	363	486	518	453	556	390	359	450
Oxygen/coal (dry), Nm ³ /kg	0.201	0.217	0.278	0.281	0.257	0.346	0.322	0.284
Steam/coal (dry), kg/kg	1.276	1.306	1.834	1.668	1.552	1.720	1.995	1.747
Dry coal input on cross-sectional area of Gasifier, kg/m ² /hr.	462	619	660	577	708	497	457	573
Gas (raw) make rate, Nm ³ /hr.	617	648	830	709	917	635	582	794
Raw gas composition, % by volume								
CO ₂	27.8	27.3	30.4	31.2	29.3	29.9	31.2	30.9
CnHm	0.6	1.2	0.4	0.8	0.8	0.4	0.9	0.8
O ₂	0.0	0.0	0.0	0.0	—	0.0	0.0	0.0
CO	19.2	17.4	16.0	15.2	19.8	19.1	15.4	17.2
H ₂	42.7	41.5	39.6	40.5	37.9	38.1	38.9	38.7
CH ₄	8.8	12.2	13.1	11.9	11.7	11.8	11.9	11.7
N ₂	0.9	0.4	0.5	0.4	0.5	0.7	1.7	0.7
Calorific value of purified gas (CO ₂ =2%) K. cal/Nm ³	4207	4242	4228	4218	4159	4074	4178	4176
Thermal efficiency cold gas, %	73.8	75.5	75.1	78.3	78.1	75.7	76.0	74.4
Carbon gasified, %	76.4	75.4	79.7	81.7	83.1	86.3	82.9	82.8
Consumption per 1000 Nm ³ of raw gas								
Coal (dry), kg	587	750	624	639	606	615	617	567
Steam, kg	749	980	1171	1066	941	1057	1231	991
Oxygen, Nm ³	118	163	173	180	156	212	199	161
Consumption per 1000 Nm ³ of purified gas (CO ₂ =2%)								
Coal (dry), kg	799	1011	879	910	840	860	879	804
Steam, kg	1016	1321	1649	1519	1305	1478	1754	1406
Oxygen, Nm ³	160	220	244	256	216	296	283	228
Yield of tar/1000 kg of dry coal, litre	—	—	48.0	44.0	35.0	—	53.0	57.5

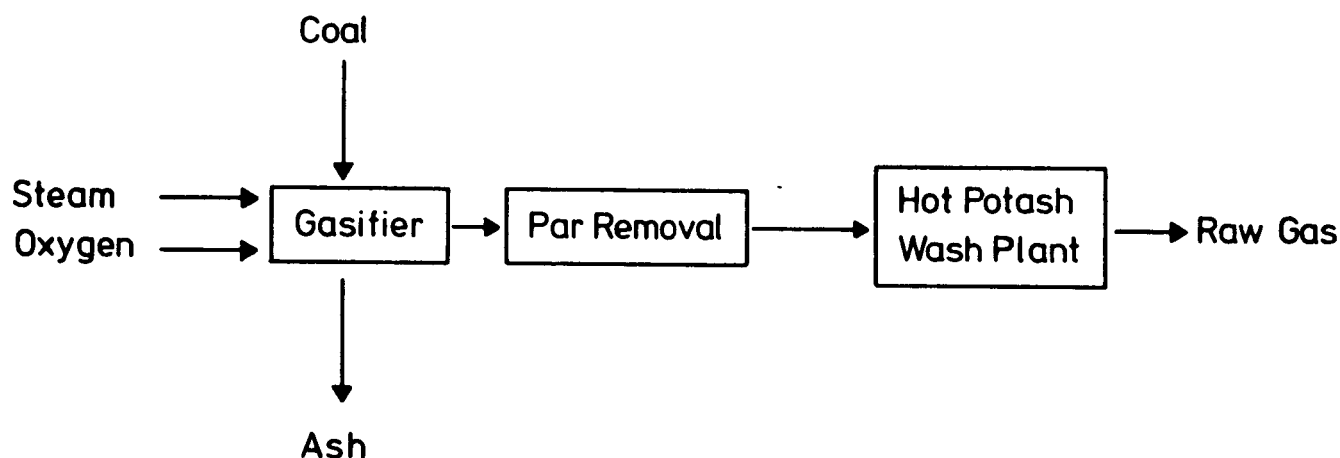


Figure 1. LURGI PRESSURE GASIFICATION PILOT PLANT

Operating Facility

The 1 tonne/hr Lurgi gasifier has been in operation since 1962 at the CFRI in Dhambad.

Major Funding Agencies

Council of Scientific and Industrial Research, Government of India.

Technical Problems

None.

Capital Costs

The project is not expected to be scaled up; therefore no large scale capital cost projections have been made.

INDIAN COAL LIQUEFACTION TECHNOLOGY

27. Central Fuels Research Institute (Bergius) Coal Liquefaction ProcessProcess Description

CFRI claims that they have completed experiments to liquefy coal by the Bergius process in a bench-scale unit at a capacity of 18 kg/hr. They are looking for foreign cooperation to scale up the process to 1 TPH capacity.

Solvent extraction of -5 mesh coal with aromatic oils was attempted in small batch reactors. The extraction was carried out for about 3 hours at 300 to 360°C (572° to 680°F) at the vapor pressure of the solvent (10 to 15 kg/cm² or 140 to 210 psi). No hydrogen or catalyst was added, and about 50% of the coal was extracted. CFRI claims that the coarse coal feed enables easier filtration of the extract. The extract could be used as a substitute petroleum feed stock. Delayed coking of the extract produces a low ash coke suitable for anodes (less than 0.5% ash) in electrometallurgy or a petroleum coke substitute.

Feedstock requirements for this process include lignites, subbituminous, and bituminous coals with high vitrian and fusain content, but with low ash content. Higher sulfur coals are an advantage for this process since the sulfur promotes catalyst activation. The primary output of this process is middle distallates, particularly diesel oil, which is the preferred product in India. In addition, hydrocarbon gases, naptha, and kerosene are also produced. The estimated process efficiency is in the range of 65% to 70%.

Process Goals

The goals of this program are to develop indigenous coal liquefaction technology within India to offset foreign oil imports.

Relationship to Prior Technology

This development is an offshoot of the Bergius-Piers process (I.G. Farben Process) developed in Germany prior to World War II.

Operating Facility

The CFRI has conducted tests in an 18 kg/hr bench scale unit in Dhambad since the mid-70's. A larger facility has not been built due to lack of funding support.

Major Funding Agencies

This small-scale development work is being supported by the Central Fuel Research Institute.

Technical Problems

Materials of construction, especially in vessel lining material, and valve life are considered the major problems. Hesser problems include separation of heavy oils from ash solids and the production of an economical source of hydrogen, which is required as a feedstock to this process. Oil loss from the process has also been indicated as a problem.

Capital Costs

Capital costs for a 1 million tonne/yr liquid product facility has been estimated to be R_g 11,000 million (Base year 1980-Rs 8.00 = \$ 1 U.S.). Operating costs for such a facility are estimated to be roughly R_g 1150 million/yr, excluding feedstock costs. Coal feedstock costs were estimated at R_g 675 million/yr.

INDIAN COAL GASIFICATION PROCESS

28. Bharat Heavy Electricals Ltd. (BHEL) Combined Cycle Coal Gasification ProcessProcess Description

The BHEL process utilizes a high-pressure, Lurgi-type fixed-bed reactor in a combined-cycle layout to generate electric power. In this process lumpy coal of graded size is gasified with air and steam at a pressure of 10 bars (10^6 N/m^2) to yield a low calorific value gas of 4960 KJ/Nm^3 . In addition to the gas, tar, oil, and liquor are obtained as by-products.

The process flow diagram, shown in Figure 1, comprises the gasifier, mixing vessel, gas cooler, and scrubber. Coal of graded size is fed to the gasifier through a suitable feeding system. Air and steam mixed in the mixing vessel are led into the gasifier through the tuyeres located in the rotating grate of the gasifier. Coal coming in contact with air and steam undergoes successive processes of carbonization, gasification, and combustion yielding a low calorific value gas and ash. The ash is continuously withdrawn from the gasifier through the rotating grate and ash discharge chamber. The gas leaves the gasifier through the gas exit line.

As the gasifier operates continuously under pressure it is necessary to have some means of introducing coal and discharging the resultant ash from the process at atmospheric pressure. The coal feeding system and ash discharge system provide these means, respectively.

The grate and coal distributors are two other parts in the gasifier which regulate a uniform distribution of gas, air, steam, and coal in the gasifier. The gasifier is also provided with a feedwater jacket where part of the steam needed for gasification is produced.

The raw gas leaving the gasifier at about 540°C traverses through the gas cooler and drops down to a temperature of 350°C . In the process it superheats the steam circulated from the main waste heat boiler by indirect contact, and in addition dust particles and part of the tar vapors are condensed and collected. Part of the superheated steam is led to the mixing vessel where it admixes with air fed from a booster and saturated steam from the gasifier. The rest of the steam is led to the steam turbine for power generation.

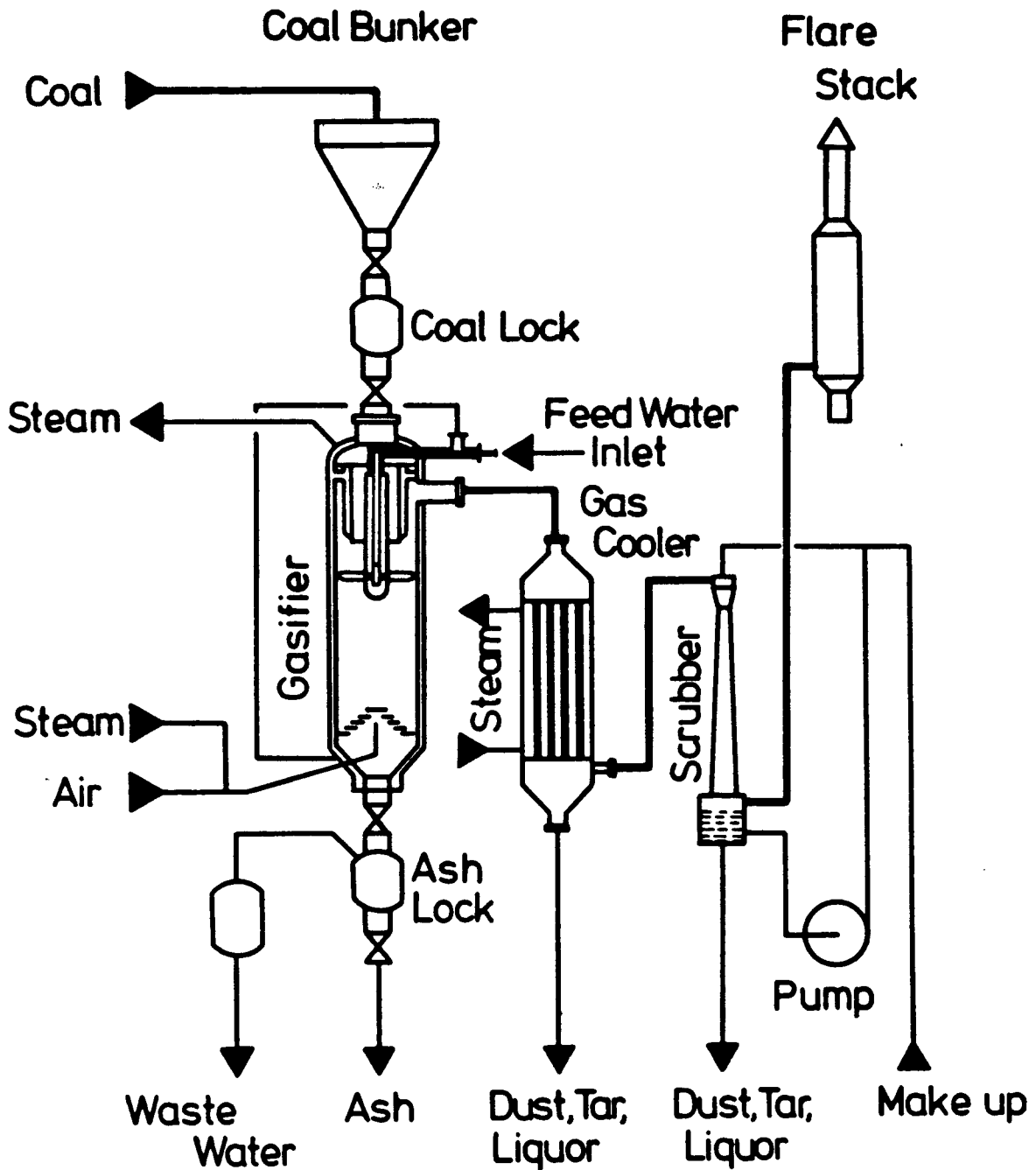


Figure 1. HIGH PRESSURE FIXED-BED GASIFICATION

The gas enters the scrubber where it is contacted by either cooling water or/and gas liquor spray. Tar and the other liquor products get condensed here and are withdrawn from the bottom of the scrubber. The liquor is recycled back to the scrubber. The final gas leaving the scrubber is free of tar and alkali vapours.

The clean gas from the gasification plant is the starting material for the gas/steam turbine combined cycle operation. This gas has a calorific value of around 4960 KJ/Nm³. This process has been tested using Singareni coal with the properties shown in Table 1. The cleaned synthesis gas which is fed to the combined cycle process has the expected properties shown in Table 2.

Process Goals

The program has the following objectives:

- To undertake technical and techno-economic evaluation of advanced power cycle concepts.
- To identify the more promising cycle configurations, define know-how gaps regarding hardware and systems, and initiate R&D Projects.
- To design, develop and install a combined cycle demonstration power plant based on coal gasification to establish engineering, product and systems designs, operating conditions and generate reliable cost data. A combined cycle demonstration plant of about 5 MW capacity is being installed at BHEL, Tiruchirapalli, for which the system design has been completed and hardware design is underway. The plant envisages a new fixed-bed pressurized gasifier, a gas turbine of about 3 MW capacity, a waste heat recovery boiler, and a conventional steam turbogenerator of 2 MW in a BHEL-patented power cycle.

Relationship to Prior Technologies

The gasifier used in this process is based on the Lurgi fixed-bed design. BHEL used the operating experience of the 1 ton/day Lurgi pilot plant gasifier at the Central Fuels Research Institute to design a modified version of the moving-bed gasifier using indigenous materials and components. Joint efforts with Lurgi of West Germany were abandoned due to high costs.

Operating Facilities

A 5-MW combined cycle facility using the high-pressure Lurgi type gasifier was to have been commissioned in 1980. This gasifier is located at

Table 1. SINGARENI COAL FEEDSTOCK CHARACTERISTICS AND MATERIAL INPUTS

Coal:	Singareni coal	6 t/h
	Particle size	6-30 mm
Ash fusion temperature:		1280°C
Net CV (KJ/kg):		17640
Proximate analysis (Wt %):		
	Moisture	4.80
	Ash	36.90
	V.M.	25.44
	F.C.	33.48
Ultimate analysis (Wt %):		
	C	42.82
	H	2.82
	S	0.62
	N	1.00
	O	5.97
	MM	40.59
	Moisture	4.18
Air:		7.2 t/h; 14 bar ($14 \times 10^5 \text{ N/m}^2$); 100°C
Steam:		2.05 t/h; 14 bar; 380°C

Table 2. SYNGAS CHARACTERISTICS OF THE BHEL GASIFIER

Net CV	4930 KJ/Nm ³
Steam	1.8 t/h; 10 bar (10^6 Nm^2) saturated
Tar	0.29 t/h; 9 bar ($9 \times 10^5 \text{ Nm}^2$); 110°C
Gas:	14.76 t/h; 9 bar ($9 \times 10^5 \text{ Nm}^2$); 110°C
Gas analysis (Vol. %):	
	CO ₂ 15.48
	CO 9.20
	H ₂ 19.48
	H ₂ O 19.42
	CH ₄ 4.58
	N ₂ 31.81
	H ₂ S 0.02

Trichy, India. BHEL is also developing a single-stage fluidized-bed gasification unit at Hyderabad and a Koller-type single-stage gasifier at Trichy. These two developments are summarized in Table 3.

Major Funding Agencies

Bharst Heavy Electrical Ltd. is funding the above mention gasification development programs.

Technical Problems

The technical problems involved with the this process include hot gas cleanup, mechanical problems associated with gasifier grid rotation, scale-up, and lock-hopper coal feeding. In addition, ball valves are being substituted for cone-type valves used by Lurgi the gasifier. These new valves have yet to be proven.

Capital Costs

Capital costs for the BHEL process combined cycle facility have not been published. However, because of the relatively dirty gas produced by the Lurgi-type gasifier it is believed that this process will be more expensive than other combined cycle projects such as the Cool Water Texaco project in California.

Table 3. OTHER BHEL GASIFIER DEVELOPMENT PROGRAMS

i. Single-Stage fluidized bed gasification unit (Hyderabad):

Coal throughput: 750 kg/h; particle size: 0-6 mm

Max. operating pressure: 10 bar

Operating temperature range: 1000-1300°C

Gasification media: Steam + air

Gas cleaning: cyclone, quenching pipe and venturi scrubber

Product gas: low-heating value gas with a net heating value of
4200 kcal/kg [17.58 MJ/kg]

Status: Conceptual design finalized; detailed engineering and design in progress.

ii. Koller-type Single-Stage Gasifier (Trichy):

Capacity: 4000 N³/h

Product gas temp.: cooled from 350-450°C to 100-110°C in a water-sprayed pre-cooler

C.V.°f product gas: 1384-1694 kcal/Nm³ at full load

operating pressure: atmospheric

Air supply: 2600 Nm³/h

Max. hot gas efficiency: 89%
cold gas efficiency: 79% at 100% full load

Steam generated by jacket boiler: 330 kg/h at 0.5 ata

Additional details: Water-cooled jacket; automatic charging and weighing equipment; rotating ash pan, at 0-2.5 rev/

Status: pilot plant under performance tests.

JAPANESE COAL LIQUEFACTION PROCESS

29. Mitsui Solvent Refined Coal ProcessProcess Description

The Mitsui Solvent Refined Coal (SRC) process was developed to produce a clean burning solid fuel from coal. This solid fuel, which has low melting characteristics, is low in ash and sulfur content. The characteristics of this solid fuel product are presented in Table 1. Testing of this process has been performed in a 5 ton/day pilot plant since 1977 by the Mitsui Coke Co.

In this facility coal is sized to less than 25 mm for storage in a coal bin. From storage the coal is sent to a dry grind pulverizer and reduced to a maximum of 100 mesh before storage in a coal weighing hopper. This hopper is capable of feeding 5 tons/day of coal to a slurry preparation tank at a constant rate. In the slurry tank a uniform slurry containing one part coal and three parts recycled process solvent is produced using an agitator and a circulation pump.

Table 1. CHARACTERISTICS OF THE MITSUI SRC PRODUCT

Characteristics of SRC

Ash	0.1 wt. % max.
Sulfur	0.3 wt. % max.
Carbon	88 — 90 wt. %
Hydrogen	5 — 6 wt. %
Low Heating Value	ca. 9,000 kcal/kg
Specific Gravity	1.25
Softening Point	approx. 150°C
Grindability (Hardgrove Index)	150

The slurry is continuously pumped from the slurry mixing tank to a slurry preheat section prior to entering the dissolver reactor. Pressurized hydrogen is added to the slurry stream at the inlet of the preheat section. The slurry entering the dissolver unit is reacted at 420° to 430°C at 1075 psi for approximately 1 hour. Most of the coal entering the dissolver is decomposed to a liquid product with the exception the coal's ash and other unreactive constituents. The coal extract is then sent to a separation system, which

consists of a high- and medium-pressure separation process. The coal extraction is separated into vapor phase products and liquid phase products, which include residue. The vapor phase products are then condensed to separate the light hydrocarbon and water from the gaseous products. The liquid condensate is stored and the recovered gases are recycled to the preheat section. Before recycling these gases are purified in a caustic soda and water wash system. A part of the washed gases are purged to maintain the hydrogen content in the gases at the dissolver. The remaining gases are compressed and mixed with make-up hydrogen before entering the dissolver preheat section.

The liquid phase products are depressurized, cooled and sent to the filter feed tank. The stored liquid products are put into a pressurized leaf filter. The operation is a repetition of precoating, filtering, rinsing, and drying, to process 6 tons of liquid products per one cycle operation. Solid free filtrate which is obtained from the filter is sent to the light end column feed tank. The stored filtrate is charged to the light end column and distilled under atmospheric pressure, then separated into naphtha, wash solvent, and SRC plus process solvent.

The naphtha and the wash solvent are transferred and stored in tanks. The SRC plus the process solvent are heated in the vacuum flash preheater and sent to the vacuum flash column where the SRC is separated from the process solvent. The SRC is sent by gravity to the solidification unit where the SRC is quenched by water and solidified. The solidified SRC is conveyed and stored as product. The process solvent is stored and used as recovered process solvent.

A process flow diagram for this process is presented in Figure 1 for the 5 ton/day pilot plant. This facility has been operated on Miike Coal (Japanese) as well as Millmerran and Victorian Australian Coals. The pilot plant test results for these coals are presented in Tables 2, 3, and 4.

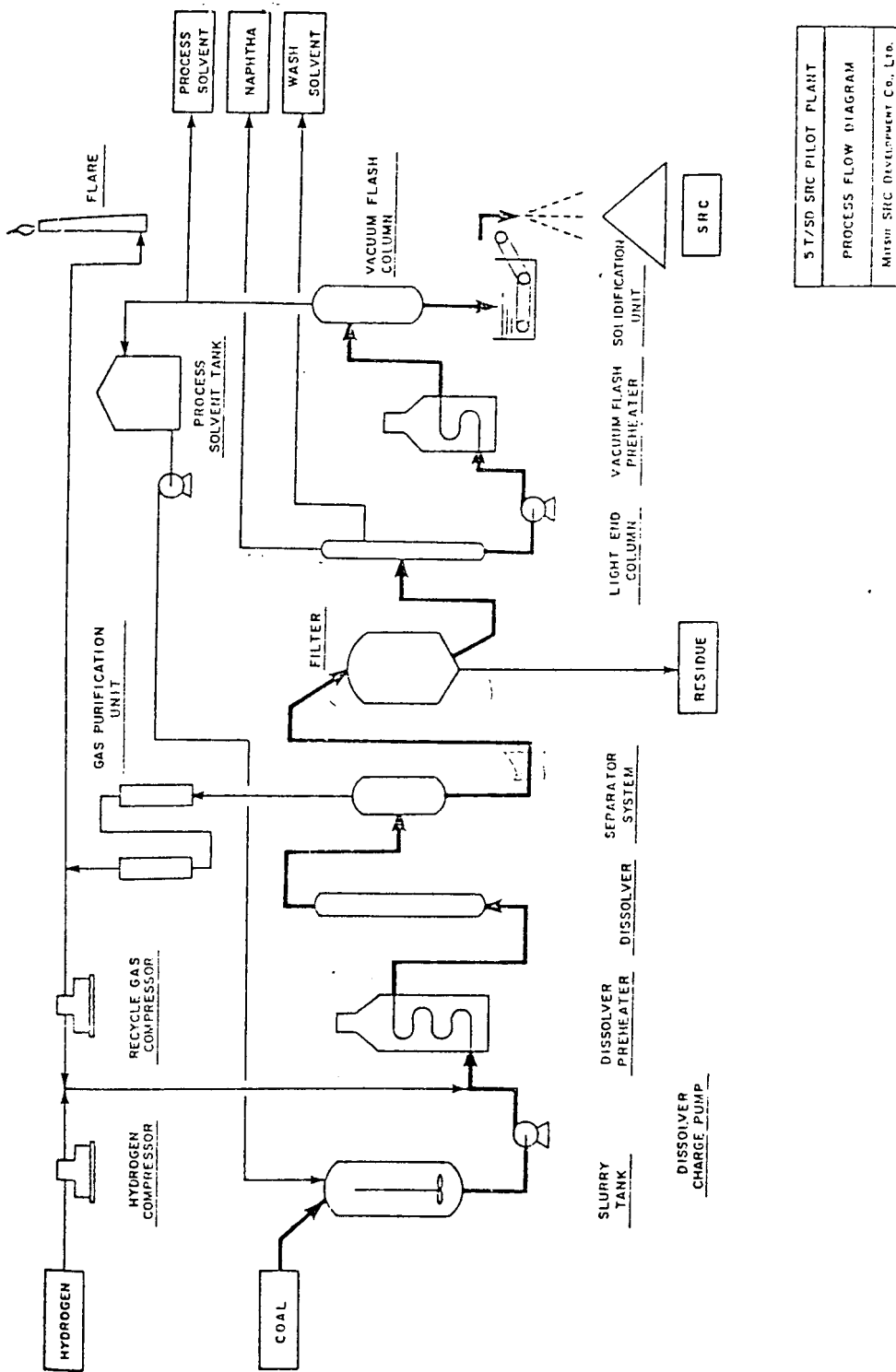


Figure 1. MITSUBI SRC PROCESS FLOW DIAGRAM

Table 2. TEST RESULTS FOR MIIKE COAL

(1) Ultimate and Ash Analysis of Coal (dry coal basis)	
Carbon	73.8 wt. %
Hydrogen	5.6
Nitrogen	1.1
Sulfur	1.7
Ash	11.6
(2) Test Result	
Solubility	97% (moisture ash free basis)
Chemical Hydrogen Consumption	2 wt. % (dry coal basis)
Distillate Yield	10 wt. % (dry coal basis)
SRC Yield	63 wt. % (dry coal basis)

Table 3. TEST RESULTS FOR MILLMERAN COAL

(1) Ultimate and Ash Analysis of Coal (dry coal basis)	
Carbon	64.0 wt. %
Hydrogen	5.5
Nitrogen	1.1
Sulfur	0.5
Ash	19.7
(2) Test Result	
Solubility	91% (moisture ash free basis)
Chemical Hydrogen Consumption	3 wt. % (dry coal basis)
Distillate Yield	10 wt. % (dry coal basis)
SRC Yield	50 wt. % (dry coal basis)

Table 4. TEST RESULTS FOR VICTORIAN BROWN COAL

(1) Ultimate and Ash Analysis of Coal* (dry coal basis)	
Carbon	67.9 wt. %
Hydrogen	4.8
Nitrogen	0.5
Sulfur	0.3
Ash	2.4

* Used as Baiquettes with 15 wt. % water.

Process Goals

The goals of the Mitsui SRC Development Co. Ltd., are to develop this process for the production of a clean fuel source as well as a source of carbon material for electrode production and as a source of coking additives.

As an energy source, even coals of high sulfur content, if turned into SRC, can be converted to a clean energy source of low ash and sulfur content. This is an important feature of SRC. In comparison with other coal liquefaction methods the SRC process is less costly because it consumes less hydrogen and in addition, SRC, if heated, can be used as a liquid fuel. By processing through hydrocracking SRC can be converted to light fuels such as gasoline.

Light and middle distillates produced from SRC process contain some chemical industry feedstocks such as benzene, toluene, xylenes, and phenols. Various raw materials for chemical industry can also be produced by hydrocracking of SRC.

As a carbon source, needle coke can be produced, with high yield, through delayed coking and calcining of SRC. This high-quality calcined coke can be raw material of ultra-high-power electrodes (UHP).

In producing blast furnace coke, the raw material blended with SRC can produce coke which has a high strength at elevated temperature. Therefore, from the viewpoint of utilizing unused coal resources (noncaking coal, brown coal) to counter the future shortage of supplies of strongly coking coal, this application of SRC has great significance.

Relationship to Prior Technology

In 1971, perceiving the promising future of coal liquefaction by a new solvent extraction method, namely, the Gulf Oil Corporations Solvent Refined Coal (SRC) process, the Mitsui Mining Company was quick to proceed with a study of the new technology. In June, 1972, the Mitsui SRC Research Consortium was organized by four enterprises, Mitsui Mining Co., Ltd., Mitsui Coke Co., Ltd., and Mitsui & Co., Ltd., to establish an SRC process research and development organization.

The consortium started liquefaction tests with more than fifty kinds of domestic and foreign coal by the autoclave method at Mitsui Coke Company's

Ohmuta Works, followed by the completion of a bench plant for continuous operation in 1973 for further study of the process. In December 1974, for the utilization of overseas leading technology, the Consortium made a contract for joint SRC development with Gulf Oil Corporation, the parent company of the Pittsburg and Midway Coal Mining Co., the originator of the SRC process.

In 1975 the consortium developed its own SRC process for the purpose of producing not only SRC as a clean coal but SRC as a substitute for strongly coking coal and coking additive for use in the production of iron-manufacturing coke. In 1973 the consortium built Japan's first coal liquefaction plant using the SRC process (coal feed 5 T/SD) at Mitsui Coke Company's Ohmuta Works with technical assistance from Gulf.

While the products of Gulf's SRC process and Mitsui process are both in the form of pitch-like solid, the raw material can be all kinds of coal, lignite, and brown coal, except anthracite. In addition, the product contains little sulfur with its heating value reaching as high as 9,000 kcal/kg, which proves its usability not only as a clean energy but also as a substitute for coking coal and high grade carbon material. Development is also under way to use SRC as a substitute for strongly coking coal and coking additive under a joint research contract made with Nippon Steel Corp. which started in April, 1979.

In the meantime, Gulf Oil Corp., the Consortium's partner, upgraded the SRC-I process to the SRC-II process capable of producing a clean liquid fuel. Gulf further experimented with the improved process at a 50 T/SD pilot plant in the suburbs of Tacoma. Plans called for the construction and operation of a 6,000 T/SD plant in West Virginia for large-scale demonstration of the process with financial support of the U.S. Department of Energy (DOE). This plan called for the involvement of the United States, West Germany, and Japan (cost borne by the governments of these countries). Gulf invited Ruhrkohle AG of West Germany and Mitsui SRC Research Consortium to organize a private joint venture corporation to carry out this plan. On the understanding that it is Japan's only private enterprise qualified to join the venture and at the request of Gulf and Ruhrkohle, the Mitsui Consortium decided to participate in the project. The private joint venture corporation, SRC International, Inc. was formally established in July, 1980. But the governments of the three

countries decided to terminate this SRC-II project because of financial problems in June, 1981. The SRC International, Inc., however, will be continued to develop further coal liquefaction studies as a private joint venture corporation.

Operating Facilities

A 5 ton/day pilot plant has been in operation since March 1978 at the Mitsui Coke Company's Ohmuta Works, Goseimachi, Ohmuta-City, Fukuoka. Construction, which started in September 1976, was completed in December 1977 at a cost of 1.8 billion Yen. Operating costs have been estimated to be 400 million Yen per year. This facility has a 5 ton/day coal input and will produce approximately 3 tons/day of SRC. The plant is operated by four groups, each of five persons, on three shifts. The operation is controlled and observed from the control room except for a part of the coal handling.

Major Funding Agencies

To cope with the domestic and overseas situation, the four consortium members established Mitsui SRC Development Co., Ltd. in February 1980 to take over the consortium's business for the further development of SRC under a substantially complete system. In addition, the seven companies which took part in building the 5 T/SD SRC plant held shares in the new company to strengthen the arrangements for further SRC commercialization. The organizational outline of this new company is presented in Table 6.

In preparation for SRC commercialization, Mitsui conducted basic tests of Victoria Brown Coal samples and developed a special dehydration technique for brown coal and a process of treating woody tissue. This development confirmed that brown coal is both technically and economically advantageous as an SRC feedstock. The State Government of Victoria, which evaluated Mitsui's technical level, has agreed to perform a feasibility study by Mitsui after June 1980 for the construction of a commercial SRC plant in the vast brown coal field area.

Capital Costs

Australia's CSR and Japan's Mitsui SRC Development Co. have agreed to a 50-50 joint venture to evaluate commercial production of solvent-refined coal and liquid fuels from lignite in Australia's Latrobe Valley in Victoria. They will spend more than \$3.5 million in studying a 6,000 ton/day SRC plant using

The Mitsui process. The study is expected to be completed by June of 1982, and a \$1.7 billion plant could be onstream as early as 1987. However, the most recent information indicated that MITSUI/CSR has postponed the construction of this facility. In addition MITSUI/CSR has expanded their development goals to include coal gasification technology through Germany's Ruhrkohle.

Technical Problems

The Mitsui SRC process is similar to the Gulf SRC development and therefore these processes should share common problems. Most of the major problems have been resolved in pilot plant research.

Table 6. ORGANIZATIONAL OUTLINE FOR THE MITSUI SRC DEVELOPMENT CO., LTD.

Founded: February 20, 1980

Head Office: Mitsui Main Building, 1-1, 2-chome, Nihonbashi-Muromachi, Chuo-du, Tokyo 103, Japan

Plant Location: 1-banchi, Goseimachi, Ohmuta-shi, Fukuoka-ken 836, Japan

Chairman: Toshikuni Yahiro
(President of Mitsui & Co., Ltd.)

President: Shingo Ariyoshi
(Chairman of Mitsui & Co., Ltd.)

Number of Employees: 72

Capital: ¥500,000,000.

Shareholders:

	<u>%</u>
Mitsui & Co., Ltd.	28.5
Mitsui Mining Co., Ltd.	23.5
Mitsui Coke Co., Ltd.	5.0
Toyo Engineering Corporation	12.0
Mitsui Engineering & Shipbuilding Co., Ltd.	8.0
The Japan Steel Works, Ltd.	8.0
Ishikawajima-Harima Heavy Industries, Co., Ltd.	6.0
Mitsui Construction Co., Ltd.	3.2
Mitsui Toatsu Chemicals, Inc.	3.0
Mitsui Miike Machinery Co., Ltd.	2.0
Yamatake-Honeywell Co., Ltd.	0.8

JAPANESE COAL CONVERSION PROCESS

30. Nippon Brown Coal "Kominic" Hydrogenation Direct Coal Liquefaction ProcessProcess Description

In 1972 the Japanese firms of Kobe Steel, Ltd., Mitsubishi Chemical Industries, Ltd., and Hissho-Iwai Co., Ltd., formed the Kominic group. The purpose of this enterprise was to exploit the vast brown coal (lignite) deposits in Victoria State, Australia, to produce a solid solvent-refined coal (SRC-I) fuel. With this in mind, Kominic group entered into a research relationship with the South African Coal, Oil and Gas Corp. (SASOL) to investigate the liquefaction characteristics of brown coal in SASOL's SRC-I liquefaction process. In 1977 Kobe Steel constructed 500 kilogram/day process development unit for further SRC-I studies on Victoria brown coals at the Iwaya Works west of Osaka.⁵ During the late '70's Japan's as well as the Kominic group's goals shifted away from solids production (for metallurgical coke) toward liquid fuels production. To meet these goals, Kobe Steel developed an additional high-pressure hydrotreating stage which when added to the original SRC-I type process produces a liquid with approximately 70% heavy oil and 30% light oil.² During this development period (1979) the Kominic group ran tests on Victoria brown coal in SASOL's SRC-II liquefaction process. After these tests, Kominic modified their SRC-I process at Osaka to an SRC-II type process using the group's new high pressure hydrotreating stage.

It is believed that the Kominic process has been developed by the joint partner company of Kobe Steel, Ltd. In this process (Figure 1) coal is pulverized, preferably 200 to 300 mesh, and mixed with recycled pulverized catalyst (cobalt-molybdenum and/or iron, iron-sulfur) of the same consistency. The recycled catalyst is presumably recycled ash residue, depending on the coal properties. However, the catalytic effects of this ash residue are highly dependent on the coal type and coal deposit location. In addition to pulverized coal and catalyst, a hydrocarbon solvent such as anthracene oil, having a boiling point of over 150°C, is added and slurried in a slurry tank. This slurry is then pumped, at a flow velocity of from 10 to 400 m/hr to a preheater. The slurry is mixed with a high pressure, hydrogen-rich gas, and this mixture is then heated to a temperature of 420° to 440°C.

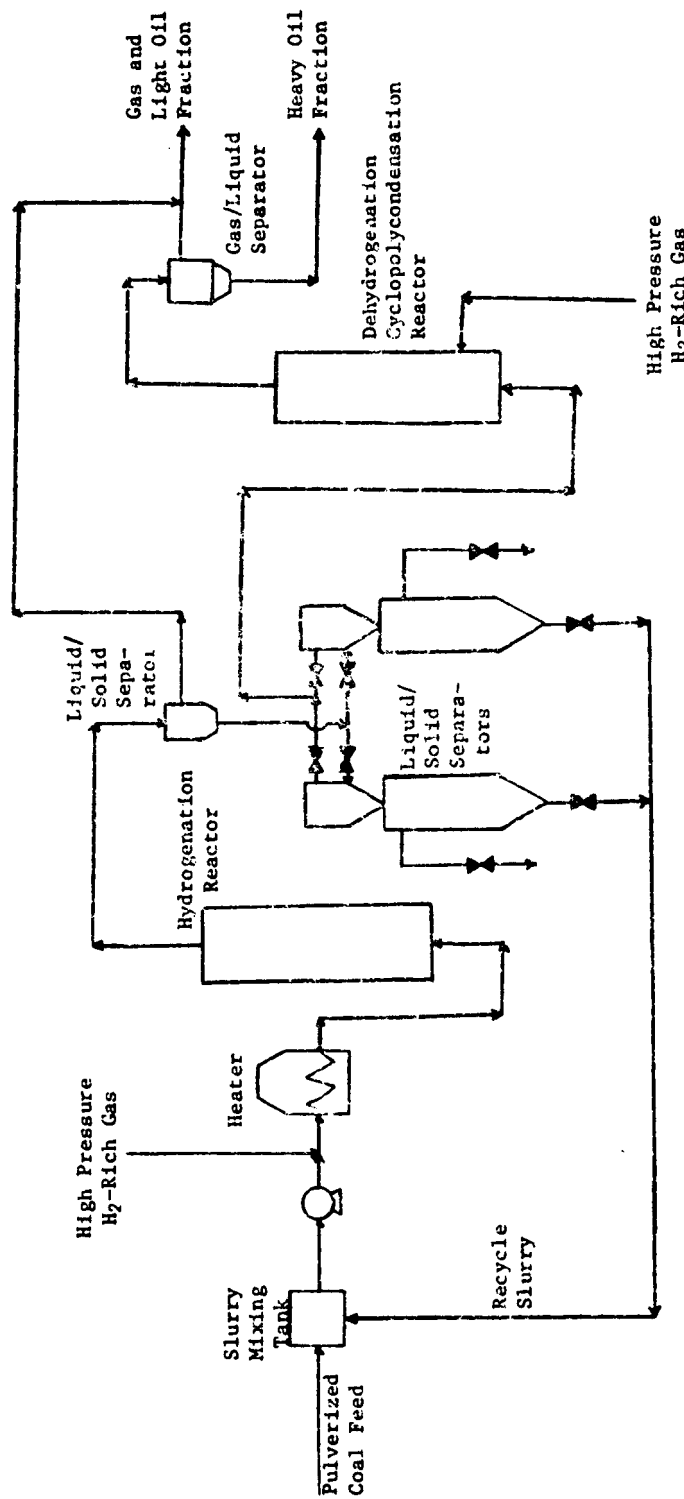


Figure 1. NIPPON BROWN COAL LIQUEFACTION PROCESS

The hot slurry mixture is then introduced into the base of the hydrogenation reactor at a pressure of 100 to 150 atm. The stream leaving the top of the reactor consists of dissolved gases in a light oil/solid residue slurry. This mixture is then sent to a solid-liquid separator.

The solid-liquid separator consists of a liquid cyclone and a solid accumulating tank. Connected to the top of the liquid cyclone is a gas-liquid outlet pipe. The solid-liquid separator is operated in a batch mode and therefore a second solid-liquid separator is utilized in an alternating pattern. The solid residue discharge from the separator is collected in a slurry tank where part of it is recycled. The liquid that is withdrawn from the separator is then sent to a dehydrogenation cyclopolycondensation reactor for further processing.

The dehydrogenation-cyclopolycondensation reactor is operated at 420° to 440°C and 100 to 150 atmospheres. In this reactor the liquid is subject to treatment under noncatalytic conditions in the presence of a small amount of hydrogen gas with a total partial pressure of 7 to 70%. The reaction time within the reactor is 5 to 90 minutes, as was the case in the hydrogenation reactor. With this treatment the higher oil liquid entering the reactor is converted from an oil having naphthenic or paraffinic-rich properties to a heavier oil having aromatic-rich properties. This oil is then sent to a gas-liquid separator for further processing. The aromatic rich oil produced in the NBCLP process can then be sent to a refinery for further processing.

To achieve the atomic ratio of hydrogen to carbon of liquid fuels, direct liquefaction processes rely on doubling or tripling the ratio from that of coal. Unlike gasification processes, which produce hydrogen internally by the water-gas shift reaction, direct liquefaction processes require the outside addition of hydrogen. About 1000 SCF of hydrogen is needed per 100 pounds of coal (MAF) feedstock. Therefore, for a 25,000 ton/day facility, about 5×10^8 CF/day of hydrogen is required. To meet this demand, hydrogen must be produced by steam reforming of methane or light oil feedstock, partial oxidation of heavy crude, or coal gasification. About 26,000 bbl/day of naphtha would be required for the process to produce a nominal 60,000 bbl/day of products. Because naphtha is not likely to be available in adequate supply in Japan, hydrogen production via coal gasification is more likely.

A U.S. patent that as recently been awarded to Kobe Steel for this process is shown as an Exhibit. A technology fact sheet for this process is presented in Table 1.

Process Goals

To further promote to commercialization of this direct liquefaction process, the Komonic group was reorganized with the addition of Idemitsu Kosan and Asia Oil on August 12, 1980.⁶ The new company, Nippon Brown Coal Liquefaction Co., is capitalized at \$2.1 million with equal 20% investments from each of the five participating companies. This new company has recently entered into negotiations for a joint Japan-Australia brown coal liquefaction project with the Victoria state government and the Australian government.

Future plans call for the construction of a 50 metric ton/day pilot plant for which construction was started in 1981 with operation expected in 1983. This plant will be constructed near the coal mines in Victoria state at an estimated cost of \$150 to \$175 million (U.S.). About 90% of this plant will be funded by the Japanese government as part of its national Sunshine program. In return for government support all technologies and know-how gained through the operation of the pilot plant will be made available to the government. The Australian government will supply the basic infrastructure, plant site, electric power, industrial water, and feedstock coal.⁷

A second phase is currently envisioned that calls for the construction startup of a 5,000 ton/day demonstration plant in 1985 at a cost of between \$833 million and about \$1 billion (U.S.). This demonstration plant would then be expanded into a commercial size facility by 1990 with the addition of five, 5,000 ton/day units. The Victoria state government has pledged to the Japanese full cooperation in supplying the feedstock brown coal for the 30,000 ton/day commercial plant. The total project cost for the first three phases is estimated at over \$4 billion (U.S.).⁶

Relationship to Prior Technology

This development is not directly related to any commercially developed process. However, the general principals of the NBCL process are similar to the SRC-II process developed in the U.S.

Table 1, Part 1. TECHNOLOGY FACT SHEET: Nippon Brown Coal "Kominic" Hydrogenation Direct Coal Liquefaction Process

NEW TECHNOLOGY (PROCESS DESCRIPTION AND GOALS): Coal is pulverized, mixed with a catalyst, slurred with a solvent, mixed with hydrogen, and heated under pressure. The mixture is then introduced to a hydrogenation reactor followed by a reactor to produce aromatic-rich products. Goals include construction of a 50 ton/day pilot plant by 1983 and subsequent large-scale facilities.

RELATIONSHIP TO PRIOR TECHNOLOGY (INCLUDING STATE OF DEVELOPMENT OF PRIOR TECHNOLOGY):

The Nippon Brown Coal Liquefaction technology is a variation on EDS and SRC-II technologies.

CHARACTERISTICS OF THE TECHNOLOGY:

CHARACTERISTICS

NEW TECHNOLOGY

PRIMARY OUTPUT (DESIGN CASE).....	Syncrude
TYPE OF PROCESS.....	Solvent extraction
FEEDSTOCK REQUIREMENTS.....	Brown coal/lignite
OVERALL THERMAL EFFICIENCY (INCLUDING BY-PRODUCTS).....	~68%
CARBON CONVERSION EFFICIENCY.....	
OPERATING TEMPERATURE.....	790°F - 825°F
OPERATING PRESSURE.....	1450 PSI - 2200 PSI
BY-PRODUCTS.....	Methane Propane/Butane Sulfur

Nippon Brown Coal "Kominic" Hydrogenation
Direct Coal Liquefaction Process

Table 1, Part 2.

TECHNOLOGY FACT SHEET:

STATUS OF DEVELOPMENT:

OPERATING FACILITIES — 0.55 metric ton/day pilot plant at the Iwaya Works of Kobe Steel west of Osaka.

MAJOR FUNDING AGENCY — Nippon Brown Coal Liquefaction Co., Japanese Government.

ANNUAL LEVEL OF FUNDING —

TECHNICAL PROBLEMS: Corrosion/erosion of components.

Feedstock coal drying technology not yet developed or purchased by

Nippon Brown Coal Liquefaction.

OTHER FACTORS AFFECTING OVERALL FEASIBILITY: Only within the last year has direct liquefaction

been pursued by Nippon Brown Coal Liquefaction. Currently, they are somewhat dependent on

technologies developed by others, including Sasol, Ltd., in South Africa, and the EDS and SRC-II

process in the United States. Direct liquefaction processes such as this have a large hydrogen

requirement which reduces the overall efficiency of production.

Operating Facilities

In 1977 Kobe Steel constructed a 500 kg/day process development unit at the Iwaya Works west of Osaka to test Victoria brown coals.

Major Funding Agencies

The NBCL liquefaction process has attained the status of a national project and as such is being partially funded by the Japanese government to a commercial level.

Technical Problems

The Nippon Brown Coal process is currently in the pilot plant stage of development. Due to the lack of available data and operating experience, critical problems for this process have yet to surface. However, due to the similarity of this process to other coal liquefaction processes, in particular the SRC process, parallel problems might be expected to occur. In the SRC process corrosion/erosion problems occur in four main process areas that are similar to those found in the Nippon Brown Coal liquefaction process. These areas include coal receiving and preparation, preheating and dissolving, filtration and mineral residue drying, and solvent recovery.

In the area of coal receiving and preparation, typical problems associated with similar facilities occurred in coal slurry centrifugal pumping equipment and high-pressure plunger-type slurry preheater charge pumps. These problems centered on packings and seal leakage, as well as check valves and plunger erosion. In the preheating and dissolving sections few materials problems developed. These minor problems involve nozzle sleeve lining corrosion and scale formation. However, in the area of high- and intermediate-pressure separator equipment, severe stress corrosion cracking of Type 304 stainless steel linings occurred. This stress corrosion cracking was attributed to polythionic acid and chloride stress corrosion cracking at weld sites. Corrosion was not experienced in any of the other materials in the separators, including carbon steel.

Corrosion/erosion material problems also occurred in the filtration and mineral residue drying areas. Most problems occurred in the heat exchanger material where severe erosion of metal tubes was encountered as well as tube bulging due to carburization in the 3/8 inch thick 304 SS dryer shell.

Severe corrosion/erosion problems have also occurred in the reboilers, air coolers, fractionation equipment, piping, and tower walls of the solvent recovery section of the SRC process. These problems have caused several major shutdowns. The most severe corrosion occurred in moving parts and materials that came in direct contact with the liquid solvent. Most of the problems required a change in material specifications to eliminate the situation. However, acceptable corrosion rates have yet to be achieved in areas such as the stainless-steel trays used in the separator towers.

Capital Costs

The investment cost of the Nippon Brown Coal Liquefaction process developed by Kobe Steel, Ltd., has not been published. However, based on a process flow sheet comparison with other coal liquefaction processes, the Nippon Brown Coal process does not differ significantly from the SRC-II process which is being developed by the Gulf Oil Corp. in the U.S. Based on this comparison, the two processes should have similar capital cost requirements. The total plant investment costs for the SRC-II process are shown in Table 2. These costs include engineering, land, plant, and G&A costs. Also included is a 20% contingency factor based on the state of the technology readiness. Make-up hydrogen production will cost about \$7.50/million Btu based on capital costs and coal requirements in addition to residues. This amounts to a product cost content for liquids of about \$3.50/million Btu. Hydrogen at \$10.00/million Btu would result in a product cost content of about \$4.50/million Btu.

Table 2. TOTAL PLANT INVESTMENT, NIPPON BROWN COAL LIQUEFACTION

	<u>1978 \$</u> <u>(Millions)</u>	<u>1980 \$</u> <u>(Millions)</u>
Major Liquefaction Equipment	675	810
Other Equipment and Materials	245	295
Labor and Erection	530	635
Hydrogen Production Plant (Coal Gasification)	825	1000
Total	2280	2740
Analysis cost basis:	\$2.7 billion (1980 \$)	
Coal Requirement:	25,000 tons/day @ 12,500 Btu/lb (including 2500 tons/day added to 7500 tons/day residues for hydrogen make-up)	

EXHIBIT. U.S. Patent to Kobe Steel for NBCL Process

B-1

B-245

United States Patent [19]

Nakako et al.

[11]

4,219,403

[45]

Aug. 26, 1980

[54] **COAL LIQUEFACTION PROCESS AND APPARATUS THEREFOR**

- [75] Inventors: **Yukio Nakako, Nishinomiya, Shizuo Yokota, Kobe, both of Japan**
- [73] Assignee: **Kobe Steel, Ltd., Kobe, Japan**
- [21] Appl. No.: **915,575**
- [22] Filed: **Jun. 14, 1978**

Related U.S. Application Data

- [63] Continuation-in-part of Ser. No. 801,920, May 31, 1977, abandoned.

[30] **Foreign Application Priority Data**

May 28, 1976 [JP]	Japan	51-62811
May 28, 1976 [JP]	Japan	51-62812
May 28, 1976 [JP]	Japan	51-62813
May 28, 1976 [JP]	Japan	51-62814
May 28, 1976 [JP]	Japan	51-62815

- [51] Int. Cl.² **C10G 1/06**
- [52] U.S. Cl. **208/10; 208/8 R**
- [58] Field of Search **208/8, 10**

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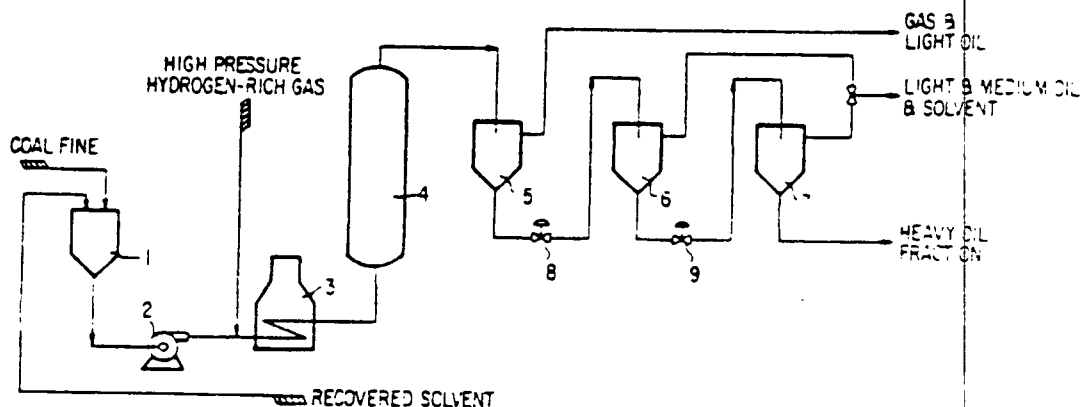
Primary Examiner—C Davis
Attorney, Agent, or Firm—Obion, Fisher, Spivak, McClelland & Maier

[57] **ABSTRACT**

A coal liquefaction apparatus which comprises a slurry mixing tank, a preheater, a hydrogenation reactor, and a gas-liquid-solid separator or separators in series and a gas-liquid separator and at least one solid-liquid separator are interposed between the hydrogenation reactor and a dehydrogenation cyclopolycondensation reactor which is positioned upstream of the final gas-liquid-solid separator.

The coal liquefaction process comprises the steps of heat treating a slurry prepared by mixing coal fines with a hydrocarbon based solvent having a boiling point greater than 150° C. in the presence of hydrogen at a temperature of 300° to 500° C. and a pressure of 50 to 700 atms. thereby forming a gas-liquid-solid mixture; separating and removing solids from said gas-liquid-solid mixture as a reaction product; separating and removing a residuum liquid fraction from said mixture; and heat treating said residuum liquid fraction in the presence of hydrogen at a low partial pressure at a temperature of 300° to 500° C. and a pressure of 50 to 700 atms.

12 Claims, 12 Drawing Figures



U.S. Patent

Aug. 26, 1980

Sheet 1 of 5

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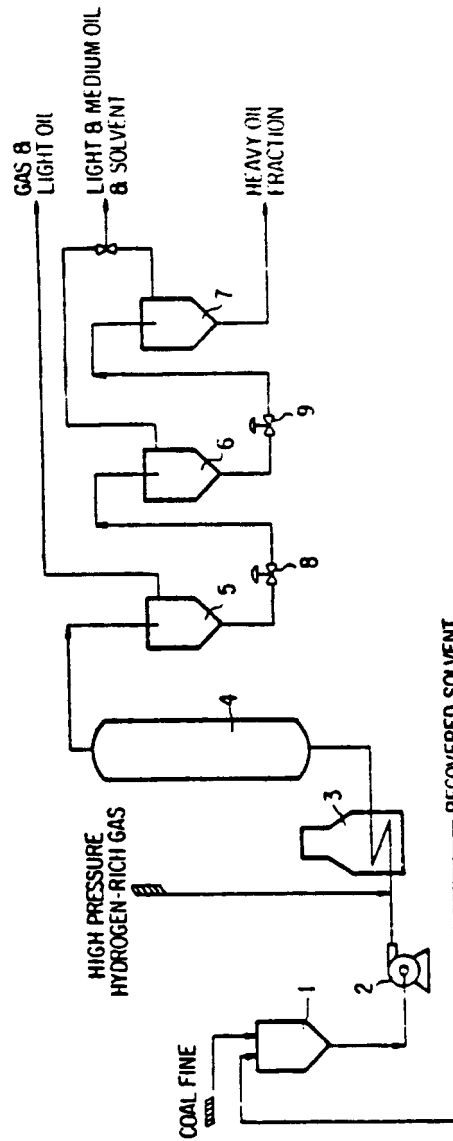


FIG. 1

U.S. Patent Aug. 26, 1980

Sheet 2 of 5

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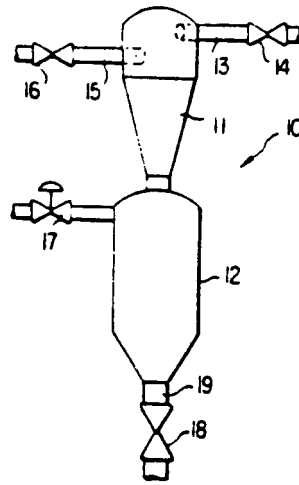


FIG. 2

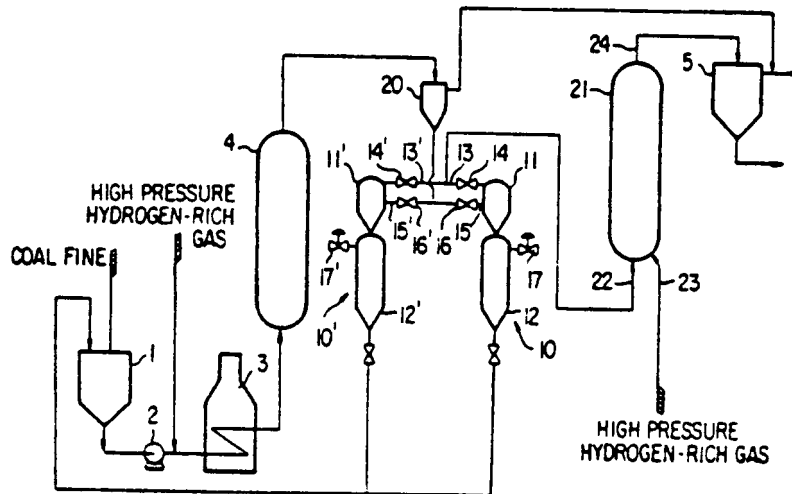


FIG. 3

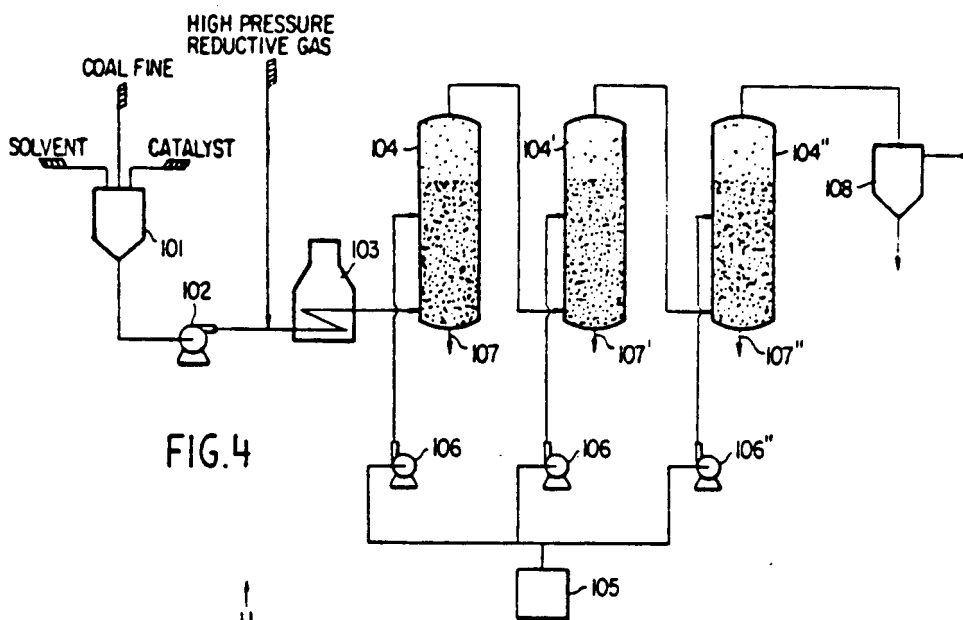


FIG. 4

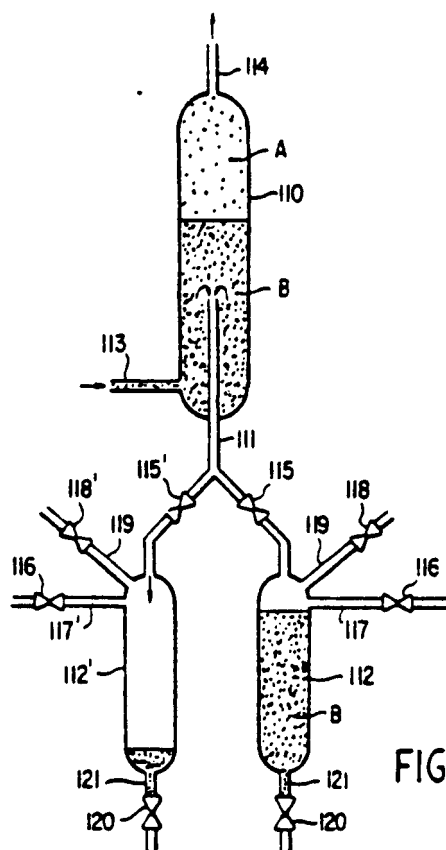


FIG. 5

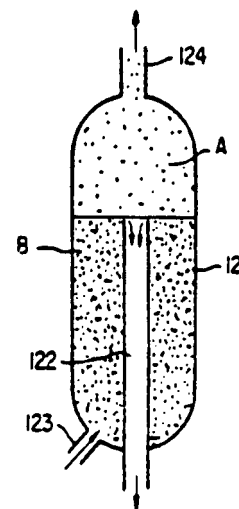
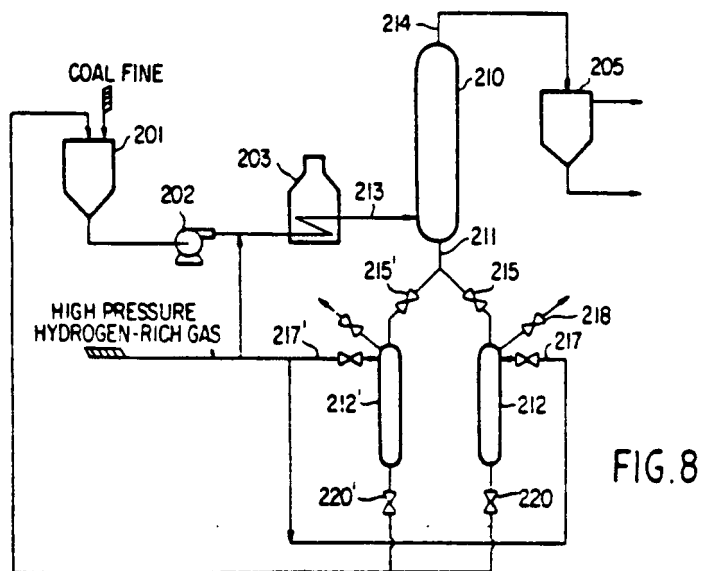
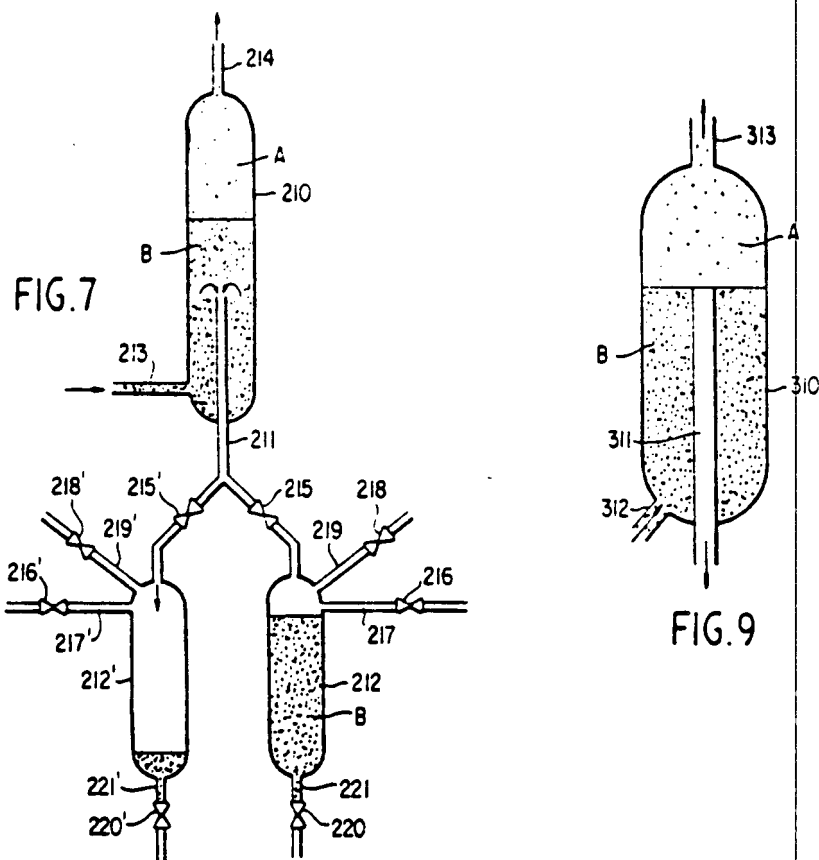


FIG 6



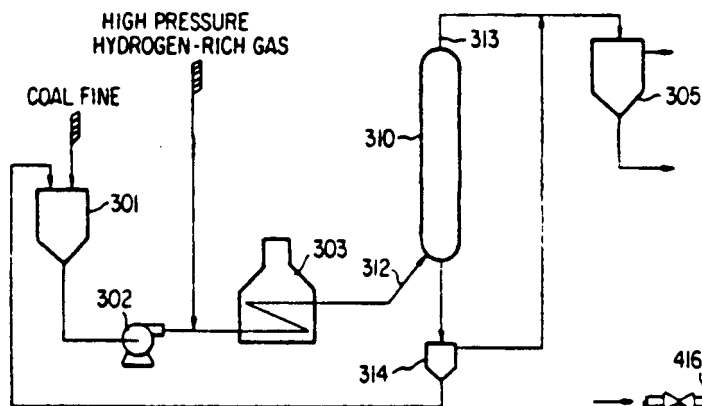


FIG. 10

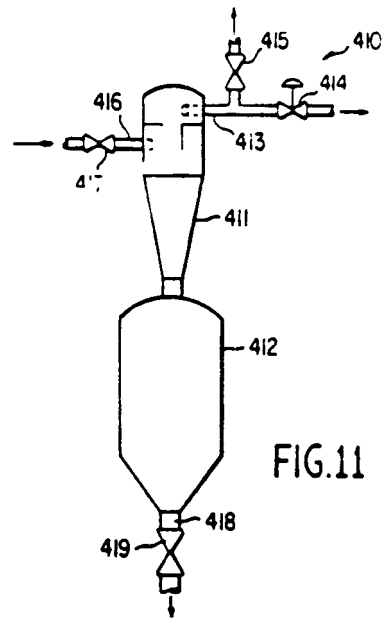


FIG. 11

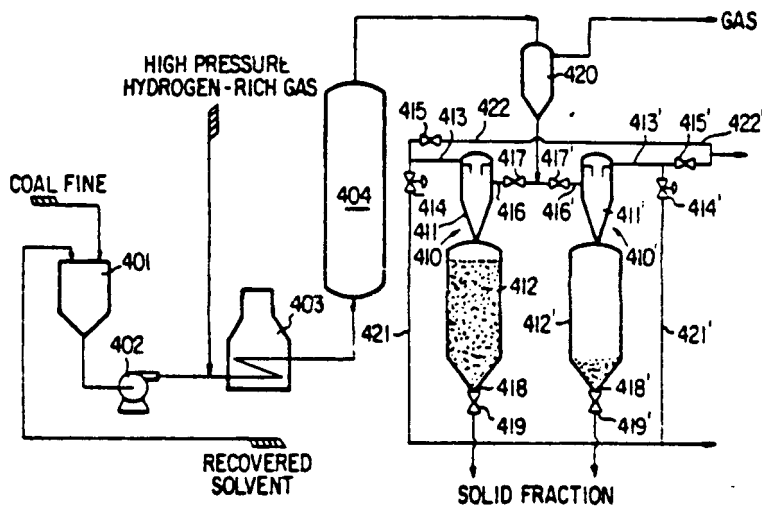


FIG. 12

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COAL LIQUEFACTION PROCESS AND APPARATUS THEREFOR

This application is a continuation-in-part application of co-pending application Ser. No. 801,920, filed May 31, 1977, and now abandoned.

BACKGROUND OF THE INVENTION

1 Field of the Invention

The present invention relates to a coal liquefaction process and an apparatus therefor, and more particularly to a coal liquefaction process which can be performed efficiently to improve the yield of reaction products, particularly, the heavy oil product which is well suited as a metallurgical carbonaceous carbon material.

2. Description of the Prior Art

A coal liquefaction process is known in which coal fines are treated in the presence of hydrogen to liquify the coal. The coal fines used in the coal liquefaction process include low grade coals such as bituminous, semi-bituminous, and sub-bituminous coals and lignite as well as similar solid carbonaceous materials such as shale. According to the conventional process of the type described, coal fines, a hydrocarbon solvent having a boiling point of over 150° C., and a suitable catalyst such as a ferro-sulfuric system catalyst, if desired, are mixed in a slurry, (The use of a catalyst may not be necessary or essential because the ash in coal functions as a catalyst), and then the slurry is preheated in a preheater. A high pressure hydrogen-rich gas is added thereto preferably prior to the preheating of the slurry. The preheated slurry and a high pressure hydrogen-rich gas are passed into a reactor where a hydrogenation reaction is conducted at a high temperature and pressure, e.g., 300° to 500° C., 50 to 700 atms. Then, a mixture of reaction products of reactor effluent is introduced into two or more separators connected through pressure-reducing valves to each other, wherein the pressure is progressively reduced and gas, liquid and solid are flash distilled.

At the present time, the objective in the liquefaction of coal is to form a heavy oil product having a high boiling point for use as a metallurgical carbonaceous material for use, for instance, in the manufacture of steel-making coke or carbon electrodes for alumina electrolysis. The liquid product or effluent, generally, includes solids such as ash, unreacted coal, catalysts, and insoluble reaction products. Accordingly, the removal of these elements would improve the quality of the heavy oil product for its intended use. In general, a metallurgical carbonaceous material should have an ash content of less than 10%.

Coal liquefaction process hitherto has been beset with many formidable problems, which will be described as follows:

Problem 1

Because of excessive hydrogenation, the yield of a heavy oil fraction in the liquid reaction product is not high enough. Moreover, solids condense along with a heavy oil fraction, in the final stage separator, where solids and heavy oil are to be separated. However, in the conventional method, a mixture having a high viscosity results at this stage, so that much time and effort must be devoted to filtering in the separation stage to separate solids from the oil. For this reason, a light oil is

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added to lower the viscosity of the mixture, and if required, the mixture is heated, followed by centrifugal separation, sedimentation separation, or separation by means of separators such as liquid cyclones. In any event, a light oil in the case should be added to the oil in a considerable amount, and this results in an unwanted increase in the amount of the mixture to be treated, which causes an increase of the number of apparatus for separating the solid and liquid and deteriorates an economic effect. In addition, upon flash distillation, a solid fraction and a heavy oil fraction both pass through pressure reducing valves, so that if the pressure is instantaneously reduced to a considerably lower level, then wear of the pressure reducing valves occurs. To avoid this, many separators and pressure reducing valves have to be used in order to gradually reduce the pressure of the system. The use of many such separators and reducing valves increases the expense of capital equipment.

Problem 2

In the coal hydrogenation reactor, a mixture of hydrogen gas or a high pressure reductive gas such as $\text{CO} + \text{H}_2\text{O}$, $\text{CO} + \text{H}_2\text{O} + \text{H}_2$, $\text{CO} + \text{H}_2$ or H_2 rich gas and the coal slurry which has to be preheated is subjected to a liquefaction reaction at a high temperature and pressure, followed by flash distillation to separate the product obtained into gas, liquid, and solid products. It is advantageous to introduce the slurry and the high pressure reductive gas into the reactor from its bottom and expel the products from the top of the reactor. In this case, the viscosity of the solvent is decreased because of the reaction at high pressure and temperature, so that a tendency arises for the settling of solids such as unreacted coal fines, catalysts and ash from the liquid. To avoid this problem, the upward rate of flow of the mixture is increased relative to the settling rate of solids during reaction. However, in order to achieve this objective, it is necessary to reduce the cross sectional area of the reactor to some extent, and the number of reactors connected in series should be increased to achieve sufficiently long residence times of the mixture for reaction in the reactors. This is uneconomical because many pieces of apparatus such as gas-liquid separators, pipe, and couplings must be used. Moreover, more maintenance problems arise because of the more extensive use of equipment. One of the attempts to solve this problem has been to reduce the number of reactors while the liquid effluent from one reactor is recycled to another, thereby extending the residence time of the slurry within the reactors. Alternatively, a reductive gas in great amounts is injected into the reactor to retard the settling of solids in the liquid reactant. However, in this technique, the concentration of unreacted coal in the reactor is equalized both at the entrance and exit of the reactor, so that the reactor itself changes in type from a piston flow reactor to a complete mixing reactor, with the result that the reaction efficiency decreases substantially relative to the reaction space or volume of the reactor.

Problem 3

A high boiling point and high viscosity reaction product is obtained from the bottom of the separator in the final stage of the multiple stage flash distillation. Accordingly, the degree of condensation of solids is not sufficiently high, thereby requiring further separation of solids from the liquid. However, because of the high

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viscosity of the reaction product, satisfactory separation of solids cannot be attained by a filtering process. For this reason, as has been described earlier, a light oil is added to the liquid product to decrease the viscosity of the mixture or heat is applied thereto, followed by centrifugal separation, sedimentation separation or separation in a liquid cyclone. Accordingly, the amount of the mixture to be treated is increased, thus failing to meet practicability requirements. It is therefore evident that no satisfactory separation process for solids has yet been found.

SUMMARY OF THE INVENTION

Accordingly, one object of the present invention is to provide a coal liquefaction process and an apparatus therefor, which improves the yield of liquified product suitable for use as a metallurgical carbonaceous material, while avoiding the wear of pressure reducing valves, and dispensing with multiple stage separators and pressure reducing valves.

Another object of the present invention is to provide a coal liquefaction process and an apparatus therefor, which provides an improved reaction efficiency relative to the space within the reactor, without using many reactors and couplings.

Still a further object of the present invention is to provide a coal liquefaction process and an apparatus therefor which improves the separating efficiency of solids from the liquid in separators after the hydrogenation reaction.

Yet a further object of the present invention is to provide a coal liquefaction process and an apparatus which eliminates the public nuisance problem caused by the disposal of catalysts.

Still a further object of the present invention is to provide a coal liquefaction process and an apparatus, in which solids may be efficiently separated from a high boiling point, high viscosity reaction product obtained from the bottom of a final stage separator, in a reasonable manner.

According to the first aspect of the present invention, solids are separated from a reaction mixture of low viscosity and at high temperature immediately after the hydrogenation reaction, and the reaction mixture from which the solids have been removed is then subjected to a dehydrogenation-cyclopolycondensation reaction under a low partial pressure of hydrogen at a high temperature under non-catalytic conditions. The dehydrogenation-cyclopolycondensation reaction is a reaction in which a light oil is dehydrogenated under non-catalytic conditions at a low hydrogen partial pressure, thereby being converted into a heavy oil, while the fraction of the reaction product which has been given a naphthenic or paraffinic-rich property because of the addition of an excessive amount of hydrogen, is dehydrogenated and cyclopolycondensed. More particularly, the reaction mixture from the hydrogenation reactor is introduced as it is or after passing through a gas-liquid separator, into a solid-liquid separation system consisting of solid-liquid separators having pressure reducing valves, with the lower portions of the separators being connected to solid accumulating tanks, and with the top portions thereof connected to gas-lined outlet pipes. The liquid fraction separated therein is subjected to a non-catalytic heat treatment in the presence of hydrogen at a low partial pressure. Suitable solid-liquid separators employable in the present invention are cyclones, sand cones, and the like

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The non-catalytic heat treatment is such that the reaction product is maintained at a given temperature for a given period of time in the presence of hydrogen at a low partial pressure. Any type apparatus may be used, as long as the above described conditions can be maintained. For instance, a device having the same construction as that of the reactor, or heating vessel which is used for preheating may be used as the non-catalytic heat treatment vessel. More specifically, the reaction mixture from a hydrogenation reactor is introduced as it is, or after passing it through gas-liquid separators into solid-liquid separators at a temperature equal to or less than the temperature at the exit of a reactor but, in any case, a temperature 100° C. no less than the latter. In the solid-liquid separators, solids accumulate in the lower solid-accumulating tank, while liquid and gas, if any, overflow and are withdrawn through overhead gas-liquid outlet pipes. The liquid fraction thus withdrawn is mixed with a hydrogen-rich gas, as required, and then introduced into a dehydrogenation-cyclopolycondensation reactor. Meanwhile, the reaction product from the hydrogenation reactor contains an excessive amount of a high pressure hydrogen-rich gas, so that hydrogen need not be added in this stage. However, when the reaction product passes through a gas-liquid separator, the addition of hydrogen is required, or a small amount of high pressure hydrogen-rich gas should preferably be introduced into the dehydrogenation reactor. In the dehydrogenation reactor, a reaction mixture devoid of solids is maintained at a high temperature in the presence of a small amount of hydrogen or at a low partial pressure under non-catalytic conditions so that the portion of the product which possesses naphthenic or paraffinic properties, is dehydrogenated and cyclopolycondensed, thereby being converted into a heavy oil fraction which imparts an aromatic-rich property to the oil which in turn yields a heavy oil well suited as a metallurgical carbonaceous material. In this respect, the presence of a small amount of hydrogen or a low partial pressure of hydrogen is mandatory for preventing an excessive amount of dehydrogenation-cyclopolycondensation. The reaction mixture subjected to the dehydrogenation reaction is withdrawn from the top of the dehydrogenation-cyclopolycondensation reactor, then passed through separators and then flash-distilled by reducing the pressure through pressure-reducing valves. However, because the reaction mixture is devoid of solids in this stage, the pressure-reducing valves are not damaged and there is no longer the need to separate solids from the liquid in the separator.

Meanwhile, in the solid-liquid separating system, when one solid accumulating tank becomes filled with solids, then the solid-liquid separating system therefor is shut off from the reaction-mixture-inlet passage, whereupon the pressure in the separator is reduced to atmospheric pressure by means of a pressure-reducing valve, and then, the accumulated solids are discharged through a bottom outlet port, as required. The solids thus discharged contain materials having a catalytic function, and thus may be used again in the coal slurry.

At least two solid-liquid separating devices in parallel are provided for one reaction system so that two solid-liquid separating devices may be used alternately, i.e., according to the so-called batch system operation. More particularly, the reaction mixture from the hydrogenation reactor is first introduced under high pressure into one solid-liquid separating device, and when the

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device is filled with solids, then the connection is switched from the filled device to the other solid-liquid separating device in order to introduce the reaction mixture into the latter, while the pressure in the first solid-liquid separating device is reduced to atmospheric pressure in order to discharge solids therefrom. This cycle of operation is repeated for an efficient continuous separation of solids from liquid.

In the second aspect of the present invention, the diameter of the reactor is increased and the number of reactors is reduced, while retaining the desired level of efficiency required for liquefaction or the hydrogenation reaction. In other words, the upward flow velocity of the reaction mixture in the reactor is adjusted in order to accelerate the settling of solids therein, and solids thus settled are discharged from the bottom of the reactor, while a fresh catalyst is supplied, as required, thereby maintaining the desired hydrogenation reaction.

More specifically, in the present invention, at least two reactors each having a solid outlet port in the bottom of the reactors are connected in series, and a preheated mixture of a coal slurry consisting of coal fines, catalyst and a high pressure reductive gas is introduced into the first reactor through its bottom port so that it passes through the reactor at such a flow velocity that solids may settle in the reactor. In this case, the reaction mixture is separated into a relatively solid-rich layer and a relatively solid-lean layer. The solids which settle are discharged from the solid outlet port provided in the bottom portion of the reactor. In this respect, one or two solid accumulators are connected to the bottom of the reactor, so that solids may be stored therein in a sufficient amount, followed by flash distillation, and then the withdrawal of the solids. At the same time the solids present in the reaction mixture cannot be completely separated in the first reactor, and hence, overflow of solids occurs along with the reaction liquid, which are separated in the succeeding reactor in the same manner.

In the second embodiment of the present invention, the catalyst substantially separates from the liquid and is removed in the first reactor, so that fresh catalyst should be supplied to the second reactor and thereafter through pipes leading to the catalyst accumulating tank to promote the hydrogenation reaction. Accordingly, the reaction is conducted in an efficient manner because of the supply of fresh catalyst. In addition, different kinds of catalysts may be used in reactors. For instance, a catalyst of the cobalt-molybdenum system, iron or iron-sulfur which possesses a high activity in the liquefaction reaction, is used in the first reactor for a highly efficient reaction, while a catalyst of a low activity is used for the second reaction and also thereafter when the reaction medium contains a relatively small amount of unreacted coal. Furthermore, no catalyst is supplied to the final reactor, so that a product possessing a naphthenic or paraffinic property, because of excessive hydrogenation is heated in the presence of a low partial pressure of hydrogen under non-catalytic conditions for the dehydrogenation-cyclopolycondensation reaction, thereby converting the liquid product into a heavy oil product having aromatic characteristics, which is well adapted for use as a metallurgical carbonaceous material.

The flow velocity of the reaction mixture of the present invention depends on the kinds and grain sizes of coal fines and catalysts used. In short, the flow velocity

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should be selected such that the solids in the reaction mixture may settle, thus leaving a solid-rich layer and a solid-lean layer therein. For instance, when an iron oxide catalyst is used, and the grain sizes of the catalyst and the coal fines are 200 mesh, then the lowest flow velocity of the slurry stream should be about 10 cm/sec to prevent settling of the solids, i.e., 360 m/hour, while the flow velocity of the reaction mixture in order to fluidize the same is about 1.5 m/hour. In an ebullated type of reactor, the flow velocity should range from about 1.2 m/hour to 360 m/hour. If the flow velocity is excessively low, then the liquefaction reaction does not proceed satisfactorily, but instead, coking occurs. Thus, the flow velocity should preferably be over 10 m/hour. On the other hand, if the flow velocity is greater than 3600 m/hour, then the undesirable excessive overflow of solids takes place. The grain sizes of the coal fines and the catalyst particles should range from 50 to 400 mesh, preferably from 200 to 300 mesh. For the grain sizes in this range, the flow velocity of the slurry may range from 1 to 3600 m/hour, preferably from 10 to 400 m/hour.

In the third aspect of the present invention, the reaction mixture is separated into a solid-rich layer and a solid-lean layer, with an interface between the two layers being maintained at a given equilibrium level. In the solid-rich layer of a given volume, ash and unreacted coal fines are present which promote the hydrogenation reaction. On the other hand, in the solid-lean layer, the dehydrogenation-cyclopolycondensation reaction occurs which results in the yield of a heavy oil product having an improved aromatic property, which is preferable from the viewpoint of a desirable metallurgical carbonaceous material. In addition, the formation of two layers permits the separation of solids of a lower ash content in an increased amount. Furthermore, the solid-rich layer thus separated may be withdrawn, as required, so that solids may be added to the slurry for reuse as a catalyst, thus saving the amount of catalyst to be used. More particularly, in the present invention, in the hydrogenation reaction of coal fines, a tube having an opening tip is inserted into the hydrogenation reactor, while the other end thereof is connected to an ash accumulator which is maintained substantially at the same pressure level as that of the hydrogenation reactor. Then, the pressure in the accumulator is adjusted so that a solid-rich layer may be introduced into the accumulator in order to maintain the interface between the two layers at a given equilibrium level, such that the volume ratio of the solid-lean layer to the solid-rich layer falls between 1/6 to 2.

In still another feature of the present invention, a tube having an open tip is inserted into the reactor through the base of the reactor, while the other end of the tube is connected to ash accumulators, which have a solid withdrawing means at the base of the reactor. The ash accumulators have gas pressure, flow rate control means and gas injection means in the tops of the accumulators. As a mixture of slurry and high pressure hydrogen-rich gas is introduced into the reactor, only the solid-lean layer is withdrawn from the top of the reactor, so that the interface between the two layers ascends. When the interface between the two layers passes over the open tip of the tube to a desired height therefrom, which depends on the reaction conditions, the size of the reactor and the like, the solid-rich layer is introduced into an ash accumulator in an amount proportional to the amount of the reaction mixture

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being fed therein. Upon the introduction of the solid-rich layer into the ash accumulator, a high pressure hydrogen-rich gas or hydrogen is charged into the ash accumulator substantially at the existing pressure level in the reactor, and then the pressure in the accumulator is adjusted to a level somewhat lower than the pressure in the reactor so as to allow the introduction of a solid-rich layer into an ash accumulator, i.e., by continuously bleeding a gas at a given rate therefrom. As a result, the interface between the solid-rich layer and the solid-lean layer may be maintained at a given equilibrium level. The solid-rich layer introduced into the ash accumulator is flash-distilled and added to the slurry for reuse. In the ash accumulator system, two ash accumulators may be used in an alternate embodiment.

According to the fourth aspect of the present invention, the interface between the solid-rich layer and the solid-lean layer is maintained in close vicinity to the open tip of a tube which is inserted in the reactor by withdrawing the solid-rich layer through the open tip of a tube, thereby providing an equilibrium between the solid-rich layer and the solid-lean layer.

The tube, as used herein, may be fixedly or movably inserted into the reactor, with the end thereof being connected via a pressure reducing valve to a slurry tank or a solid-liquid separator, such as a liquid cyclone. In this case, as well, the volume ratio of the solid-lean layer to the solid-rich layer should preferably range from 1/6 to 2.

If ash, catalyst and unreacted coal fines are separated from the solid-rich layer, then the hydrogenation reaction efficiency decreases, and unreacted coal undergoes a coking reaction, thereby adversely affecting the yield of an intended product.

Upon adjustment of the level of the interface between the solid-rich layer and the solid-lean layer to a vicinity close to the open tip of the tube in the reactor, when a mixture of slurry and a high pressure hydrogen-rich gas is continuously introduced into the reactor, the solid-lean layer alone is withdrawn from the top of the reactor, so that the interface between the two layers ascends to the open tip of the tube. In this stage, the solid-rich layer is withdrawn through the tube in order to maintain the interface between the two layers at an equilibrium level which is close to the open tip of the tube. The solid-rich layer thus withdrawn is flash-distilled as it is, and then added to the slurry for reuse as a catalyst, or otherwise separated into liquid and solids, while the liquid fraction is added to the solid-lean layer again, and the solid fraction is recovered so that it can be added to the slurry for reuse. In this case, the solid-rich layer thus withdrawn is of low viscosity, thus facilitating the separation into liquid and solid phases.

According to the fifth aspect of the present invention, the reaction mixture from the hydrogenation reactor is introduced as it is, or via a gas-liquid separator, into a solid-liquid separator having a solid accumulator connected to the bottom thereof. In this respect, the reaction mixture contains a solvent or a light oil and is of a low viscosity because the reaction mixture is preheated, thus providing ease of separation. In addition, a pressure-reducing valve is provided on the gas-liquid withdrawing pipe connected to the top of the solid-liquid separator, so that upon pressure reduction for flash distillation, solids will not pass through the pressure-reducing valve, thus avoiding erosion of the valve. This permits pressure reduction at a rapid rate. In this respect, part of the gas withdrawn from the solid-liquid

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separator may be cooled for liquefaction for further distillation in a distilling column. When the solid-liquid separator is filled with solids, then a pressure-reducing valve on a gas-liquid withdrawing pipe is opened in order to reduce the pressure to atmospheric pressure instantaneously, for flash distillation. The cycle of operation can be repeated for efficient solid-liquid separation.

BRIEF DESCRIPTION OF THE DRAWINGS

FIG. 1 is a flow sheet illustrative of a prior art liquefaction process for coal fines.

FIG. 2 is a diagrammatic view of a solid-liquid separating device within the scope of the present invention.

FIG. 3 is a flow sheet illustrating a liquefaction process according to the present invention, which employs two solid-liquid separating devices.

FIG. 4 is a flow sheet illustrative of one embodiment of the liquefaction process according to the present invention;

FIG. 5 is a view illustrative of one embodiment of a reactor of the present invention;

FIG. 6 is a view illustrative of another embodiment of the reactor of the present invention;

FIG. 7 is still another embodiment of a reactor of the present invention;

FIG. 8 is a flow sheet of the hydrogenation process of the present invention which employs the reactor of FIG. 7;

FIG. 9 is a yet another embodiment of the reactor of the present invention;

FIG. 10 is a flow sheet illustrative of one embodiment of the liquefaction process of the present invention which employs the reactor of FIG. 9;

FIG. 11 is a diagrammatic view of another embodiment of the solid-liquid separating device of the present invention; and

FIG. 12 is a flow sheet illustrative of the liquefaction process of the present invention which employs two solid-liquid separating devices.

DESCRIPTION OF THE PREFERRED EMBODIMENTS

FIG. 1 illustrates a prior art liquefaction process. Coal fines and a solvent such as a hydrocarbon having a boiling point of over about 150° C., and a catalyst, if required, are slurried in a slurry tank; and then the slurry thus prepared is delivered by a slurry pump 2 to a preheater 3. Before the slurry is passed into the preheater it is mixed with a high pressure hydrogen-rich gas. The mixture of slurry and a hydrogen-rich gas, which have been preheated to about 300° to 500° C., is introduced under pressure into a hydrogenation reactor 4 through its base for reaction at a temperature of about 300° to 500° C., and a pressure of about 50 to 700 atms. The reaction mixture from the reactor 4 is passed through separators 5, 6 and 7 which are connected in series in the indicated order, and then pressure-reducing valves 8, 9, provided on pipes which connect the separators in series, are opened so as to reduce the pressure gradually for flash distillation of the slurry into solids and liquid. The gas effluent withdrawn from the top of the first separator 5 is cooled for liquefaction, as desired, while a light oil fraction is distilled in a distilling column. A mixture of light and medium oils, and solvent withdrawn from the tops of separators 6 and 7 is distilled in a distilling column, and then the solvent thus recovered is recycled for use as a slurry forming sol-

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vent. Meanwhile, the heavy oil fraction withdrawn from the bottom of separator 7 contains a considerable amount of solids, which generally should be separated from the heavy oil. This is referred to as a de-ash operation.

In the first embodiment of the liquefaction process of the invention, as shown in FIGS. 2 and 3, a solid-liquid separating device 10 is positioned downstream of the reactor 4, so that the reaction mixture from the reactor 4 may be separated efficiently

The solid-liquid separating device 10 consisting essentially of a liquid cyclone 11 which is a type of solid-liquid separator, and a solid accumulating tank 12 connected to the bottom of the cyclone 11. Connected to the top of the liquid cyclone 11 is a gas-liquid outlet pipe 13, while a stop valve 14 is provided on pipe 13. A reaction mixture inlet pipe 15 is connected to the upper portion of liquid cyclone 11 at a position lower than the joint of the gas-liquid outlet pipe 13, while a stop valve 16 is also provided on pipe 15. In addition, a pressure reducing valve 17 is connected to the upper portion of solid accumulating tank 12, while a solid outlet pipe 19, having a stop valve 18, is connected to a bottom portion of tank 12.

In the liquefaction process of the invention, a non-catalytic heat treating device is positioned downstream of the solid-liquid separating device to reform the liquefaction products, thereby improving the yield of the heavy oil fraction which is suitable for use as a metallurgical carbonaceous material.

In the first embodiment of the invention, as shown in FIG. 3, two or more solid-liquid separating devices 10 and 10' are provided directly or through a gas-liquid separator 20 downstream of the reactor 4. In FIG. 3 the primed numerals are used to distinguish the second solid-liquid separating device and parts associated therewith for common use with those of the first device from the first device.

Gas-liquid outlet pipes 13 and 13', which are attached to solid-liquid separating devices 10 and 10', are connected to a gas-liquid inlet pipe 22, which is connected to the bottom portion of the non-catalytic heat treating device, or reactor 21. A high pressure, hydrogen rich, gas injection pipe 23 is connected to reactor 21, while an effluent outlet pipe 24 is attached to the top of reactor 21, which leads in turn to separator 5.

In the operation of the apparatus for the liquefaction process of the present invention, as shown in FIGS. 2 and 3, the reaction mixture from the reactor 4 is passed through the gas-liquid separator 20 at a temperature of about 300° to 500° C. and a pressure of about 50 to 700 atms. and then gas is withdrawn from the top of separator 20, while a solid mixture is withdrawn from the bottom thereof, which is then introduced into the first solid-liquid separating device 10. The solid-liquid mixture is subjected to a somewhat lower temperature and pressure than the reaction mixture prior to its introduction into gas-liquid separator 20. All stop valves and pressure reducing valves in the solid-liquid separating devices 10 and 10', are maintained in their closed positions at first, and then stop valve 16 on inlet pipe 15, which leads to the inlet of separating device 10, and stop valve 14 on inlet pipe 13, which leads to separating device 10 are opened to allow the introduction of the effluent from reactor 4 into device 10. The effluent is separated into a liquid-rich phase (this will be referred to simply as a liquid), and a solid-rich phase (the solid-liquid mixture will be referred to as a solid when used in

terms of liquid cyclone 11), while the liquid is withdrawn through outlet pipe 13 by overflow into reactor 21.

The solids thus separated accumulate in solid-accumulating tank 12. When solid-accumulating tank 12 is filled with solids, then stop valves 16' and 14' are opened. The stop valves 16 and 14 are closed, so that the introduction of the solid-liquid mixture is switched from the first separating device 10 to the second separating device 10', for the separation of solids and liquid as well as for the accumulation of solids. On the other hand, the pressure reducing valve 17 of the first separating device 10 is opened to reduce the pressure inside to atmospheric pressure, and stop valve 18 is opened so that the solids which accumulate therein are withdrawn through outlet pipe 19. The solids thus withdrawn are delivered to slurry tank 1 for reuse. Then, all stop valves and pressure reducing valves in separating device 10 are closed. When the second separating device 10' is filled to capacity with solids, then the introduction of the solid-liquid mixture is switched from the second separating device 10' to the first separating device 10. This cycle of operation can be repeated for a continuous operation.

The liquid to be delivered to reactor 21 is introduced into reactor 21 which is maintained substantially at the same temperature and pressure as that of reactor 4, wherein the liquid is subjected to treatment under non-catalytic conditions in the presence of a small amount of hydrogen which is fed into reactor 21 through gas inlet pipe 23. The treatment conditions depend on the size of the apparatus, the quality of the desired liquefaction product, and the like. In order to produce a heavy oil product which is well adapted for use as a metallurgical carbonaceous material, preferably the temperature ranges from 400° to 500° C., a total pressure of from 70 to 150 atms in the presence of hydrogen of a low partial pressure, the hydrogen partial pressure is preferably from 7 to 70% of the total pressure, and the reaction time should be as long as that of the hydrogenation reaction, for instance, 5 to 90 minutes. With this treatment, a further lighter oil fraction or a reaction product having naphthenic or paraffinic-rich properties, which is produced by the addition of an excessive amount of hydrogen, may be subjected to a dehydrogenation cyclopolycondensation reaction and converted into a heavy oil fraction, which has the desired aromatic-rich property at an increased yield of 1 to 30% in comparison to the amount of starting coals (MAF or medium abrasion furnace black). The liquid thus treated is withdrawn through outlet pipe 24 which is connected to the top of reactor 21 and is delivered to the separator for further processing, which is well known.

It is apparent from the above discussion concerning the liquefaction process of the invention, a reaction mixture devoid of solids is heat-treated in the presence of hydrogen, which results in an improved yield of a heavy oil fraction, while solids may be separated under low viscosity conditions at high temperature and pressure, thereby providing improved separating efficiency and minimizing the ash content of the liquid product.

The liquefaction process achieved by hydrogenation in the present process includes:

- (1) a high degree of hydrogenation of coal fines in the presence of hydrogen and catalyst of high activity such as a catalyst of the cobalt-molybdenum system, iron or iron-sulfur system at high temperature and pressure

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- (2) a relatively low degree of hydrogenation in the presence of an iron system catalyst or in the absence of a catalyst in the presence of hydrogen; and
 (3) liquefaction at a high temperature and high pressure in a hydrogen donor solvent having aromatic characteristics such as anthracene oil, without or in the presence of a small amount of hydrogen.

The term "hydrogenation reaction" is used herein in association with the above-described processes included in the present invention.

FIG. 4 illustrates the second embodiment of the liquefaction process of the present invention. Coal fines, solvent and catalyst are slurred in slurry tank 101 and then the slurry thus prepared is delivered by slurry pump 102 to preheater 103. Prior to passage of the slurry into the preheater a highly reductive gas is mixed with the slurry. The mixture of slurry and high pressure reductive gas, which has been preheated to about 300° to 500° C., is fed under pressure into the first reactor 104 through its base, wherein the mixture is passed from the bottom to the top at a flow rate (preferably 10 to 400 m/hour) such that the solids in the reaction mixture may settle against the upward flow of the mixture for reaction at a temperature of about 300° to 500° C. and a pressure of about 50 to 700 atms. The reaction mixture effluent, which overflows the top of reactor 104 is introduced into the second reactor 104' through its base, and then the reaction mixture effluent, which overflows the top of reactor 104', is introduced into the third reactor 104'' through its base. At this time, fresh catalyst from the catalyst accumulating tank 105 is slurred in a suitable solvent and then the slurry is delivered by means of pumps 106, 106' and 106'' to reactors 104, 104' and 104'', respectively. The solids which settle in the various reactors are discharged through solid outlet portions 107, 107', and 107'', positioned at the bases of the reactors. The reaction mixture effluent from the final reactor 104'' is introduced into a gas-liquid separator 108, and then part of the gas effluent from the top of the gas-liquid separator 108 is cooled for liquefaction, while the liquid residuum is further distilled in a distilling column. The liquid effluent from the base of gas-liquid separator 108 (in this case, the liquid may contain some amount of solids) is subjected to flash distillation under a reduced pressure into gas, liquid, and solids, followed by further distillation. The solids obtained from the distillation contain unreacted coal fines, catalyst and the like, and may be used repeatedly. Fresh catalyst can also be combined with the recovered catalyst for reuse.

In the reactor of the invention, the reaction mixture tends to separate into a solid-rich lower layer and a solid-lean upper layer. Accordingly, it is preferable that the flow velocity of the reaction mixture be adjusted by an appropriate means such as a tube which may be inserted into the reactor to withdraw the solid-rich layer, and that one or more solid accumulators having the same pressure as that of the reactor can be connected to the bottom of the reactor. Thus, gas is bled through the gas outlet pipes which are connected to the solid accumulators by opening the gas pressure and flow rate control valves provided on the gas outlet pipes, at a discharge rate which is commensurate with the rate a solid-rich liquid is introduced into the solid accumulators under pressure, so that an interface between the solid-rich layer and the solid-lean layer may be maintained at a given level. (In general, the volume ratio of the solid-lean layer to the solid rich layer should preferably be adjusted to 1/6 to 2.) In addition, the

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solid-rich liquid and the solid-lean liquid are withdrawn through the open tip of the tube inserted into the reactor, so that the interface between the solid-rich layer and the solid-lean layer may be maintained in the vicinity close to the open tip of the tube, thereby maintaining an equilibrium level within the reactor. The separation of the solid-rich layer and the solid-lean layer permits the desired dehydrogenation-cyclopolycondensation reaction to progress in the solid-lean layer, as has been described earlier, thereby increasing the yield of a heavy oil fraction having aromatic property characteristics.

A description in greater detail of the reactor will be presented.

Referring to FIG. 5, reactor 110 is shown whose base is provided with an inlet port 113, which is adapted to introduce a mixture of a slurry and high pressure reductive gas therein, and whose top portion is provided with an outlet port 114, which is adapted to withdraw the solid-lean layer therethrough. Reactor 110 is connected via pipe 111 and valve 115 to a solid accumulator 112. The solid accumulator 112 has its top portion connected to a gas injection pipe 117 which is provided with a gas injection valve 116, and a gas outlet pipe 119, which is provided with a gas pressure flow rate control valve 118. A solid outlet pipe 121 having a stop valve 120 thereon for withdrawing solids therethrough is connected to the base of accumulator 112. In the reactor shown in FIG. 5, pipe 111 branches into two pipes which are connected to two solid accumulators 112 and 112', which are arranged in parallel with each other. In this respect, like parts in the second solid accumulator are designated with like primed numerals which are in common use with the corresponding parts of the first solid accumulator.

In operation of the reactor shown in FIG. 5, the solid accumulators 112 and 112' are isolated from communication with the reactor by closing the valves 115 and 115', and the gas pressure flow rate control valves 118 and 118' as well as stop valves 120 and 120' are closed for the first time. Then, a high pressure reductive gas is introduced through the gas injection valves 116 and 116' substantially at the same pressure as the pressure in reactor 110, after which injection valves 116 and 116' are maintained in a closed position.

A mixture of slurry and a high pressure reductive gas which has been preheated to about 300° to 500° C. is introduced through the inlet port 113 into reactor 110 at a slurry flow rate of 1 to 3600 m/hour, preferably 10 to 400 m/hour. In this case, the reactor 110 is maintained at a temperature of about 300° to 500° C. and a pressure of about 50 to 700 atms. The mixture thus introduced under pressure is separated into a solid-lean layer A (this will be referred to as layer A) and a solid-rich layer B containing ash, catalyst, and unreacted coal fines in a uniformly or thoroughly mixed condition. (This will be referred to as a layer B.) In layer B, ash and catalyst are condensed and accumulate so that the liquefaction reaction is promoted. On the other hand, in layer A, which is heated in the presence of hydrogen at a low partial pressure or a small amount of hydrogen almost under catalyst-free conditions, a light oil fraction or a reaction product, which possesses naphthenic or paraffinic-rich properties and which results from excessive hydrogenation, is subjected to a dehydrogenation-cyclopolycondensation reaction, thereby being converted into a heavy oil fraction which has aromatic properties which is best suited as a metallurgical carbonaceous material.

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Layer A is continuously withdrawn through outlet port 114, while a mixture of a slurry and a high pressure reductive gas is fed under pressure through inlet port 113 into reactor 110, so that the interface between layer A and layer B ascends beyond the tip of the tube 111.

At this stage, valve 115 is opened to bring the first solid accumulator 112 into communication with reactor 110. Since accumulator 112 and reactor 110 are maintained substantially at the same pressure level, layer B is not introduced into the accumulator 112. Then, the gas pressure flow rate control valve 118 is opened, so that gas is discharged from the accumulator 112 at a rate proportional to the rate at which layer B is being introduced therein. (For instance, when the solids are present in the slurry in an amount of 25 to 40%, when the high-pressure-reductive-gas-feed rate is 14 to 30 Nm³/hour, when the feed rate of slurry is 50 to 100 kg/hour, when the volume of the reactor is 100 liters, when the reaction temperature is 400° to 450° C., and when the reaction pressure is 70 to 150 atms, the feed rate of layer B is 3 to 20 kg/hour.) As a result, layer B is introduced at a given flow rate into accumulator 112 so that the interface between layer A and layer B reaches an equilibrium at a given level with the result that the volume ratio of layer A to layer B may be maintained at 1/6 to 2, as shown in FIG. 5. The above ratio is well suited for hydrogenation in layer B and dehydrogenation-cyclopolycondensation in layer A.

When a sufficient amount of layer B has been introduced into the solid accumulator 112, valve 115 is opened, valve 115 is closed, and the first accumulator 112 is shut off from the reactor 110, so that layer B may be introduced into the second accumulator 112. The layer B, which accumulates in the first accumulator 112, is subjected to flash-distillation by opening valve 118, while residuum solids are discharged through valve 120, which is maintained in its open position. Subsequently, accumulator 112 is pressurized to the same pressure level as that in reactor 110. This cycle of operation is repeated by alternately using the accumulators 112 and 112'.

Referring to FIG. 6, reactor 121 has a tube 122 which is inserted therein through its base and opens into the reactor through its open tip, in addition to an inlet portion 123 adapted to introduce a mixture of slurry and a high pressure reductive gas, and an outlet port 124 adapted to withdraw a solid-lean layer therethrough.

In the operation of the reactor 121 shown in FIG. 6, a mixture of slurry and a high pressure reductive gas, which has been preheated to about 300° to 500° C., is introduced via inlet port 123 into the reactor 121, which is maintained at a temperature of about 300° to 500° C. and a pressure of about 50 to 700 atms. The mixture thus introduced is separated into layer A (a solid-lean layer) and layer B, which includes ash, catalyst, unreacted coal fines and the like in a uniformly or thoroughly mixed condition, i.e., a solid-rich layer. In layer B, ash and catalysts are condensed and accumulate, thereby promoting the hydrogenation reaction. On the other hand, in layer A, as in the case of FIG. 5, the dehydrogenation-cyclopolycondensation reaction takes place, so that the product is converted into a heavy oil fraction.

Layer A is continuously withdrawn through outlet port 123, while a mixture of slurry and a high pressure reductive gas is continuously fed through inlet port 123 under pressure so that the interface between the layer A and the layer B ascends.

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On the other hand, the open tip of tube 122 is set at a position of 6/7 to 1/3 of the height of reactor 121. When the interface reaches the open tip of the tube 122, layer B (as well as the layer A) is withdrawn through the open tip at a rate proportional to a feed rate of a mixture. (For instance, when the solid content of the slurry is 25 to 40% by weight, when the feed rate of the high pressure reductive gas is 14 to 30 Nm³/hour, when the feed rate of the slurry is 50 to 100 kg/hour, when the volume of the reactor is 100 liters, when the reaction temperature is 400° to 450° C. and when the reaction pressure is 70 to 150 atms, then the rate of layer B, which is withdrawn, is 3 to 20 kg/hour.) As a result, the interface reaches an equilibrium in the vicinity close to the open tip of tube 122, so that the volume ratio of layer A to layer B may be maintained in the range of 1/6 to 2. (See FIG. 6)

The solid-rich layer withdrawn from the bottom of tube 122 is flash-distilled into solid and liquid fractions. The solids are reused, because the solids contain unreacted coal fines, catalysts, and the like.

As is apparent from the above description, the diameter of the reactor can be increased and the number of reactors is reduced, while the flow velocity of the reaction mixture within the reactor is lowered, and the settling of the solids is promoted, so that the reactor provides the same advantages as those of a piston flow type reactor.

Attention will now be turned to the third embodiment of the present invention with reference to FIGS. 7 and 8. A hydrogenation reactor 210 is equipped with an inserted tube 211, with its open tip positioned therein. Tube 211 is connected to an ash accumulator 212 at the other end of the tube.

The reactor 210 is provided with an inlet port 213 adapted to introduce a mixture of a slurry and a high pressure hydrogen-rich gas, and an outlet port 214 adapted to withdraw a solid-lean layer at its top. The reactor 210 is connected via a pipe 212 and valve 215 to the ash accumulator 212. A gas injection pipe 217 equipped with a gas injection valve 216, and a gas discharge pipe 219 equipped with a gas pressure, flow rate control valve 218 are connected to the top of the ash accumulator 212, while a solid withdrawing pipe 221 equipped with a stop valve 220 is connected to the bottom portion of the ash accumulator 212. In the embodiment shown in FIG. 7, tube 212 is branched into two lines which are connected to two ash accumulators 212 and 212', which are arranged in parallel with each other, respectively. As in the previous embodiment, like parts in the second ash accumulator are designated by like primed reference numerals, which are used in common with those of the first ash accumulator 212.

As shown in FIG. 8, a pipe 213 leading from pre-heater 203 is connected to reactor 210 and the outlet port of reactor 210 is connected to a separator 205 via conduit 214. A high pressure hydrogen-rich gas supply pipe is connected to gas injection pipes 217 and 217' for the ash accumulators 212 and 212', while solid-withdrawing pipes 221 and 221' are connected to slurry tank 201.

In the operation of the liquefaction apparatus of the present invention as shown in FIGS. 7 and 8, the ash accumulators 212 and 212' are isolated from reactor 210 by closing valves 215 and 215' and the gas pressure flow-rate control valves 218 and 218' and stop valves 220 and 220' are closed for the first time. Then, a high pressure hydrogen-rich gas is introduced through gas

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injection valves 216 and 216' into ash accumulators 212 and 212' in order to bring the pressures therein to the level of the pressure in reactor 210, after which the injection valves 216 and 216' are maintained closed.

A mixture of slurry and a high pressure hydrogen-rich gas, which have been preheated to about 300° to 500° C. is introduced at a slurry flow rate of 1 to 3600 m/sec. preferably 10 to 400 m/sec. into reactor 210 which is maintained at a temperature of about 300° to 500° C. and a pressure of about 50 to 700 atms. The mixture thus introduced under pressure is separated into a solid-lean layer A and a solid-rich layer B containing ash, catalyst, and unreacted coal fines in a uniformly mixed condition. A hydrogenation reaction is promoted in layer B because ash and catalyst are condensed and accumulate therein. In layer A, the mixture is heated in the presence of hydrogen at a low partial pressure or a small amount of hydrogen almost under catalyst-free conditions, so that a light oil or part of a product which possesses naphthenic or paraffinic-rich properties, because of the addition of an excessive amount of hydrogen is converted into a heavy oil fraction which has aromatic properties and is therefore suitable as a metallurgical carbonaceous material, as prepared from the dehydrogenation-cyclopolycondensation reaction.

Layer A is withdrawn through outlet port 214 into separator 205, while a mixture of the slurry and a high pressure hydrogen-rich gas is continuously fed through inlet port 213 into the reactor, so that the interface between layer A and layer B ascends beyond the open tip of tube 211. In this stage, valve 215 is opened in order to bring the first ash accumulator 212 into communication with reactor 210. Accumulator 212 and reactor 210 are maintained almost at the same pressure level so that layer B is not fed into accumulator 212. Then, the gas pressure flow-rate control valve 218 is opened so that gas may be discharged from accumulator 212 at a rate proportional to the feed rate of layer B. Layer B is fed into accumulator 212 at a given feed rate so that the interface between layer A and layer B reaches a given equilibrium level above the open tip of tube 211, with the result that the volume ratio of layer A to layer B may be maintained over a range of 1/6 to 2. (FIG. 7) The above ratios are well suited for the hydrogenation reaction in layer B, and the dehydrogenation-cyclopolycondensation reaction in layer A.

Valve 215' is opened when layer B is introduced into ash accumulator 212 in a sufficient amount. Thereafter, valve 215 is closed so that the first accumulator 212 is isolated from reactor 210, thereby introducing layer B into the second accumulator 212', in the same manner as that of the first accumulator. The pressure reducing valve 218 is opened and the mixture is flash-distilled from the first accumulator 212. After the pressure in the accumulator 212 has been returned to atmospheric pressure, stop valve 220 is opened so that solids are withdrawn through the solid withdrawing or outlet pipe 221 and fed to slurry tank 201 for reuse. Subsequently, accumulator 212 is pressurized to the same pressure level as that in reactor 210. The above cycle of operation is repeated for the alternate use of accumulators 212 and 212'.

As is apparent from the above-described liquefaction process of the present invention, a mixture is separated into a solid-lean layer and a solid-rich layer for different types of reactions, so that ash and catalyst contents may be allowed to settle in order to promote the hydrogenation

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tion reaction, while the dehydrogenation-cyclopolycondensation reaction is promoted in the solid-lean layer so that the yield of the heavy oil fraction, suitable for use as a metallurgical carbonaceous material, is increased. In addition, solids may be separated in the reactor so that the ash content of the mixture may be reduced and the catalyst may be reused, which provides a considerable economic advantage as well as avoiding the public nuisance problem of the disposal of the catalyst wastes. The conditions of the operation are the same as those of the preceding embodiment, i.e. the withdrawal rate of layer B should preferably be in the range of 3 to 20 kg under the same conditions as those of the preceding embodiment.

The fourth embodiment of the liquefaction process of the present invention will be described with reference to FIGS. 9 and 10.

FIG. 9 shows a reactor 310 of the present invention. The reactor 310 is provided with tube 311 which is inserted through the base of the reactor with its open tip positioned therein. The reactor 310 further is provided with an inlet portion 312 in its base which is adapted to introduce a mixture of a slurry and a high pressure hydrogen-rich gas, as well as an outlet port 313 at the top which is used to withdraw a solid-lean layer therefrom. As shown in FIG. 10, a pipe leading from preheater 303 is connected to inlet port 312 of reactor 310, while outlet port 313 is connected to separator 305. The lower end of tube 311 is connected to solid-liquid separator 314.

In the operation of the liquefaction apparatus of the present invention, a mixture of a slurry and a high pressure hydrogen-rich gas, which has been preheated to a temperature of about 300° to 500° C. is introduced at a slurry flow rate of 1 to 3600 m/hour, preferably 10 to 400 m/hour through inlet port 312 into reactor 310, which is maintained at a temperature of about 300° to 500° C. and a pressure of about 50 to 700 atms. The mixture thus introduced under pressure into reactor 310 is separated into a solid-lean layer A and a solid-rich layer B including ash, catalyst, and unreacted coal fines in a uniformly or thoroughly mixed condition. In layer B, since ash and the catalyst settle and accumulate, the hydrogenation reaction may be promoted in the reactor. Layer A is heated in the presence of hydrogen at a low partial pressure or a small amount of hydrogen, almost in a catalyst-free condition, and a light oil or a portion of the reaction product which has achieved a naphthenic or paraffinic-rich property by the addition of an excessive amount of hydrogen is subjected to a dehydrogenation-cyclopolycondensation reaction so that the reaction product is converted into a heavy oil of an aromatic-rich property, and is therefore well suited as a metallurgical carbonaceous material, thereby improving the yield of the heavy oil product.

Layer A is withdrawn through outlet line 313 into separator 305, while a mixture of a slurry and a high pressure hydrogen-rich gas is continuously introduced through inlet port 312 so that the interface between layer A and layer B ascends.

On the other hand, the open tip of the tube 311 is set to a height of 6/7 to 1 of the height of reactor 310. When the interface between the two layers reaches the open tip of the tube in reactor 310, layer B is withdrawn through the open tip at a rate which is commensurate with the feed rate of the mixture. As a result, the interface is maintained in the vicinity close to the open tip of

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tube 311 at all times, so that the volume ratio of layer A to layer B may be maintained from 1/6 to 2. (FIG. 9)

The layer B thus withdrawn is separated into solid and liquid fractions in solid-liquid separator 314, while solids are delivered for reuse to slurry tank 1, and the liquid fraction is fed to separator 305 for further processing by conventional prior art procedures.

The advantages and conditions of withdrawal of layer B are the same as those in the preceding embodiment.

The fifth embodiment of the liquefaction apparatus of the invention will be described with reference to FIGS. 11 and 12.

As shown in FIG. 11, a solid-liquid separating device 410 is positioned downstream of reactor 404, thereby efficiently separating solids from the reaction mixture which is introduced from reactor 404. The solid-liquid separating device 410 consists essentially of a liquid cyclone 411 which is one type of a solid-liquid separator, and a solid accumulator 412, which is connected to the bottom portion of cyclone 411. A gas-liquid withdrawing or outlet pipe 413 is connected to the top portion of liquid cyclone 411, and a pressure reducing valve 414 is provided on a branch line of pipe 413, while a stop valve 415 is provided in another branch line of pipe 413. A reaction mixture inlet pipe 416 is connected to the top portion of liquid cyclone 411 at a position lower than the point of juncture of the gas-liquid withdrawing pipe 413 with cyclone 411. A stop valve 417 is provided on pipe 416. In addition, a stop valve 419 is provided at the solid outlet port 418 at the base of solid-accumulating tank 412.

Two or more solid-liquid separating devices 410 and 410' are provided as shown in FIG. 12, directly or via a gas-liquid separator 420 downstream of the reactor 404. (Two solid-liquid separating devices 410 and 410' are provided in FIG. 12.) Like parts in the second solid-liquid separating device in FIG. 12 are designated with like primed reference numerals for common use with the corresponding parts of the first solid-liquid separating device 410. The operation of the apparatus of the present invention for separating and removing solids from a liquefied reaction product will be described with reference to FIG. 12. A mixture from the top of reactor 404, which is maintained at a temperature of about 300° to 500° C. and a pressure of about 50 to 700 atms, is passed through the gas-liquid separator 420 so that gas may be withdrawn from the top of the separator 420, while a solid-liquid mixture is introduced into the first solid-liquid separating device 410 through its base. The solid-liquid mixture introduced into the solid-liquid separating device is somewhat low in temperature and pressure in comparison to the temperature and pressure of the reaction mixture prior to introduction into a gas-liquid separator. When a solid-liquid mixture is introduced into the solid-liquid separating device 410, the stop valve 417 on the inlet pipe 416 is opened, while the stop valve 417' on the inlet pipe 416' to the second solid-liquid separator 416' is closed.

The solid-liquid mixture thus introduced is separated into a solid-lean phase and a solid-rich phase in the liquid cyclone 411. The liquid overflows through the gas-liquid withdrawing pipe 413, while stop valve 415, pressure reducing valve 414 and stop valve 419 are closed. The solids accumulate in the solid accumulating tank 412. When the solids have accumulated in the solid accumulating tank 412, the stop valve 417 is closed, while the stop valve 417' is opened in order to switch

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the introduction of the solid-liquid mixture from the first solid-liquid separating device 410 to the second solid-liquid separating device 410', for the separation of solid and liquid and the accumulation of solids. On the other hand, after the switching operation, the pressure reducing valve 414 is opened while the stop valves 417 and 415 are closed so as to isolate the aforesaid solid-liquid separating device from the other system in order that the pressure in the device may be reduced to atmospheric pressure instantaneously for flash-distillation, thereby separating the same into gas-liquid and solids. The gas and liquid are withdrawn through the gas-liquid outlet pipe 413 and line 421. The solids which condense are withdrawn through the solid outlet port 418 in the base of the solid accumulating tank by opening stop valve 419. When the first solid-liquid separating device 410 becomes empty and the second solid-liquid separating device 410' is filled with solids, the introduction of the solid-liquid mixture is switched from the second solid-liquid separating device 410' to the first solid-liquid separating device 410. Likewise, flash-distillation is conducted therein for separation of the mixture into gas, liquid and solids. In this manner, two solid-liquid separating devices are used alternately for an efficient operation by a so-called batch system operation. The gas and liquid effluents withdrawn through lines 421 and 421' pass through a condenser, as required, so that a portion of the gas may be cooled and liquefied. The liquid is further distilled in a distilling column. On the other hand, the liquid effluent withdrawn through lines 422 and 422' is further distilled in a distilling column so that the solvent which is recovered is reused as a slurry solvent. The gas product withdrawn from the top portion of the gas-liquid separator 420 is cooled and liquefied in a condenser as required.

As is apparent from the foregoing discussion, a liquefied reaction product may be separated into solids and liquid under considerably low viscosity conditions thereby dispensing with the prior art necessity of adding a light oil to the liquid to lower the viscosity thereof, thus allowing for the separation and removal of solids in an efficient manner with the accompanying improvement in quality. In addition, the size of an apparatus may be reduced to a considerable extent in comparison to the size of the prior art apparatus thus achieving the desired saving in equipment investment. Furthermore, upon flash distillation, by reduction of pressure in the system, solids do not pass through the pressure reducing valves, thus preventing valve corrosion problems. This further permits the reduction of the pressure to atmospheric pressure instantaneously, thereby avoiding the need to provide many separators. Still furthermore, in the de-ashing operation of the prior art, heat is needed to lower the viscosity of the mixture, while the apparatus according to the present invention requires no such heating, thus saving energy.

Having now fully described the invention, it will be apparent to one of ordinary skill in the art that many changes and modifications can be made thereto without departing from the spirit or scope of the invention as set forth herein.

What is claimed as new and intended to be secured by Letters Patent is:

1. A coal liquefaction process which comprises: admixing coal fines with a hydrocarbon solvent having a boiling point greater than 150° C. to form a coal slurry; admixing with said coal slurry a hydrogen-rich gas.

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hydrogenating said coal slurry by heating said hydrogen-containing admixture at a temperature of from 300° to 500° C. and a pressure of from 50 to 700 atms whereby said coal fines are liquified and a solid-liquid admixture is formed;

5 separating said liquid-solid admixture into liquid and solid fractions; and

dehydrogenating and cyclopolycondensing said liquid fraction at a temperature of from 400° to 500° C. and a total pressure of 70-150 atms in the presence of hydrogen at a low partial pressure wherein said low partial pressure of hydrogen is in the range of 7 to 70% of said total pressure,

10 to produce an aromatic-rich heavy oil product.

2. The process of claim 1, wherein said coal slurry is hydrogenated in the presence of a hydrogenation catalyst.

3. The process of claim 2, wherein said hydrogenation catalyst comprises cobalt and molybdenum.

4. The process of claim 2, wherein said hydrogenation catalyst comprises iron.

5. The process of claim 4, wherein said hydrogenation catalyst comprises iron and sulfur.

6. A coal liquefaction process which comprises:

25 admixing coal fines with a hydrocarbon solvent having a boiling point greater than 150° C. to form a coal slurry;

admixing with said coal slurry a hydrogen-rich gas;

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introducing said coal slurry-gas mixture into the lower portion of a reactor at an upward flow rate such that a solid rich layer is formed in the lower portion of said reactor and a solid lean layer is formed in the upper portion of said reactor;

5 maintaining said reactor at a temperature of from 300° to 500° C. and at a pressure of from 50 to 700 atms whereby said coal slurry is hydrogenated in said solid rich-layer and the resulting liquid reaction product is dehydrogenated and cyclopolycondensed in said solid lean layer; and

10 withdrawing a portion of said solid lean layer to thereby recover a heavy oil product.

7. The process of claim 6, wherein the volume ratio of said solid lean layer to said solid rich layer is from 1.6 to 2:1.

8. The process of claim 6, wherein said coal slurry contains a hydrogenation catalyst.

9. The method of claim 8, wherein said solid lean layer is essentially free of said hydrogenation catalyst.

10. The process of claim 6, wherein a portion of said solid rich layer is withdrawn; the solid and liquid components of said layer separated; and the solid components admixed with said coal slurry.

25 11. The process of claim 6, wherein said upward flow rate is from 1 to 3,600 m/hour.

12. The process of claim 11, wherein said upward flow rate is from 10 to 400m/hour.

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JAPANESE COAL LIQUEFACTION TECHNOLOGY

31. Mitsui Engineering and Shipbuilding Direct Coal Liquefaction ProcessProcess Description

This process, which is being developed by the Mitsui Engineering and Shipbuilding, Co., a Mitsui Group Company, is based on the direct hydrogenation process flow diagram for their newly constructed 2.4 ton/day process development unit. The key operation steps that are different from other processes include the following: (1) preheated coal slurry/H₂ gas stream containing disposable FeS catalyst is introduced into the tubular liquefaction reactor where additional quantities of H₂ gas are injected at different stages in the reactor system at about 750° to 930°F and 2840 to 4270 psia, (2) the slurry phase in the high-pressure separator is separated into solids (unreacted coal, catalyst, ash, etc.) and oil by means of a centrifuge or a hydroclone system, (3) oil thus separated is fractionated into naphtha, light oil, heavy oil, and bottoms via the atmospheric and vacuum towers, and 4) single stage operation.

Although this development is somewhat different than the original IG Farben process it closely parallels research in West Germany to modify the original IG Farben process.

Process Goals

The direct hydrogenation process for coal liquefaction is the third of the three major processes under the Japanese Sunshine project. The design of the process emphasizes improving the rates of coal liquefaction reaction to achieve high liquid hourly space velocities (LHSV) (i.e., of at least about 10). The design feature to increase both the mass transfer and chemical reaction rates consist of (a) the use of the tubular reactor to obtain turbulent flow, (b) promotion of the "bubble flow" mode, (c) maintenance of high hydrogen concentration in the process stream by use of multistage injection of fresh and recycle hydrogen streams into the reactor system, and (d) application of high reaction temperature (400-500°C or 750-930°F) and pressure (19.6-29.4 MPa or 2840-4270 psia). Another feature of the process is use of the disposable FeS catalyst.

Relationship to Prior Technology

The Mitsui process is based on the IG Farben Process and has been further refined by the government industry development laboratory, Hokkaido, in Sapporo City. The laboratory became the first Japanese institution to have completed a "continuous" test facility back in March 1979. It is still being run to seek the optimal reaction conditions for the direct liquefaction operation at the temperature range of 400 to 450°C and pressure ranging from 200 to 700 atm. The bench-scale one- and five-liter reactors are utilized in a six-week cycle: one week for test preparation, two weeks for continuous run, one week for waste disposal, and the last two weeks for analysis of coal liquids.

The Sapporo, Hokkaido, laboratory is confident that the direct liquefaction route will eventually prove the most reasonable and practical technology largely because of its simplicity. This confidence stems partly from the technical know-how accumulated over the past 16 years. The lab's basic work, aimed at utilizing the northern-most island's coal resources, started in 1965. While still providing technology to the Mitsui engineering group, the laboratory is about to launch its study to pursue the so-called coal chemistry — an avenue of research intended to make best use of coal liquids for production of chemicals.

The 0.1 tonne/day bench-scale unit is continuing its operation to conduct the support R&D for the process development unit. Main subject areas of studies include (a) evaluation of PDU operating conditions, (b) investigation of liquefaction characteristics of different coal types, and (c) determination of properties and material balance of the recycle solvent. Basic studies conducted by Hokkaido and Yamagata Universities, Asahi Chemical Industry Company, Hitachi Ltd., Japan Steel Works, and the National Chemical Laboratory for Industry are (a) catalyst development, (b) recovery systems for slurry feed and power, (c) reactor materials, and (d) coal pretreatment methods.

Operating Facilities

A 2.4 metric ton/day facility has been completed at the Nippon Kokan Kawasaki shipyard. This direct coal liquefaction PDU was completed at a cost of \$19 million.

Major Funding Agency

This process is one of four coal liquefaction efforts which is being funded by the Japanese New Energy Development Organization (NEDO). This process will compete for funding with the Sumitomo and Mitsubishi efforts for funding which will probably be used to construct one 500 metric ton/day demonstration facility in the 1985-86 timeframe. The Mitsui headed group developing this direct coal liquefaction process consists of the Mitsui Engineering and Shipbuilding Co., Nippon Kokan, Asahi Chemical Industry Co., Hitachi Ltd., and the government controlled Electric Power Development Co.

Capital Costs

Capital costs for a commercial size facility have not been published. However, this process should be similar in cost to those being developed in West Germany which are based on the IG Farben process.

Technical Problems

No major technical problems have been reported.

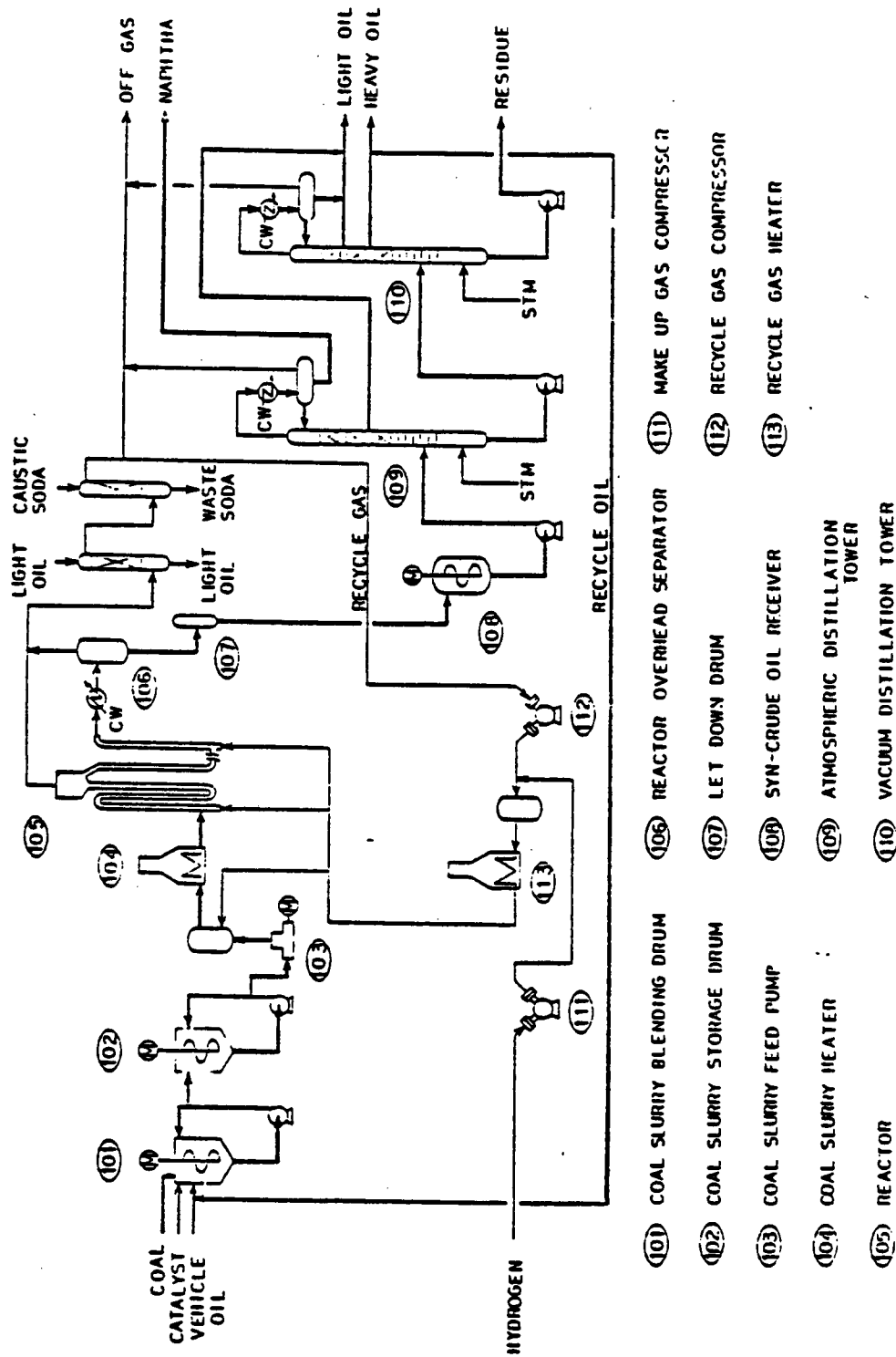


Figure 1. FLOW DIAGRAM OF THE MITSUI DIRECT COAL LIQUEFACTION PROCESS

JAPANESE COAL CONVERSION PROCESS

32. Sumitomo Solvent Extraction Coal Liquefaction ProcessProcess Description

Because of the limited amount of published information and the lack of pilot plant data, little is known of the Sumitomo coal liquefaction process. A rough process flow diagram is presented in Figure 1. As shown, coal is dehydrated and pulverized before being mixed with a hydrogen-donating solvent (tetralin) in the ratio of 2 to 5 times the coal volume to solvent.²⁴ The slurry is then mixed and heated before entering the high-pressure, high-temperature liquefaction reactor. The liquefaction reactions occur at 400 to 450°C and 100 to 150 atmospheres. The lighter products are cooled and subsequently separated into gaseous and liquid streams. The bottoms from the high pressure and high temperature liquefaction reaction are sent to a solid/liquid separator where solid solvent treated coal is produced. The liquids produced in this separator tower are cooled and sent to a distillation tower. During cooling, the heavier liquid products are collected; lighter liquids and spent solvent are separated in the distillation tower. The liquids product yield is reported to be 3.3 bbls/ton of coal.²⁶ The spent solvent is further cooled before being sent to storage. The lighter oils are also cooled and separated before being sent to storage.

The spent solvent (naphthalene) is then converted back to the hydrogen donating solvent tetralin. This is accomplished by heating and mixing hydrogen gas with the naphthalene prior to entering a series of reactor towers where the actual transformation occurs. The resulting donor solvent tetralin is then sent to storage for reuse.

The key to the process is the availability of hydrogen for the solvent reclaiming step. Although some hydrogen is reclaimed from the process, most hydrogen requirements must come from an external source. For this purpose SMI has developed a molten iron bath coal gasification process to generate the necessary hydrogen.

In the combined coal liquefaction/coal gasification mode, the hard-to-liquefy portion of the incoming coal is separated and fed to the molten iron coal gasification reactor for the production of hydrogen. The primary design feedstock choice for the liquefaction process is subbituminous coal. With

this combined process and feedstock the overall process efficiency is claimed as less than 60%.²¹ A technology fact sheet for the this process is presented in Table 1.

Process Coals

In 1970 the Sumitomo Coal Mining Co., Ltd, (SCM) a member of the Sumitomo Group, started research in the area of coal liquefaction in anticipation of an era of increased coal utilization. In 1976 SCM constructed a continuous operation 3 metric ton/day solvent refined coal liquefaction pilot plant at a coal mine near Akahera in central Hokkaido. This pilot plant produced a low ash, sulfur, and melting point solid product. In the process, a mixture of pulverized coal and hydrogen donor coal-derived solvent were heated to 400°C under high pressure. The pilot plant achieved more than 100 hrs of continuous operation in 1978.^{2,3}

In 1975 another Sumitomo Group member, Sumitomo Metal Industries, Ltd. (SMI) of Osaka, initiated research on ways of converting steam coal to binder pitch for metallurgical coke production. This investigation was prompted by a shortage of metallurgical coal in Japan at that time. Because of SCM's experience in coal conversion technology, SCM joined SMI in studying binder pitch production. This effort was soon curtailed by a decline in steel production which lessened the need for metallurgical coke. In 1978 this change in direction was illustrated by the support given by the Ministry of International Trade and Industry (MITI) to SMI and SCM for the development of the coal liquefaction technology. As part of the Sunshine Program this project has received full funding by MITI.

As an aside to coal liquefaction research, SMI has developed a ferrous bath coal gasification process. This gasification process was developed to help the steel industry toward an "oilless steel plant" capability.⁵ In addition, the ferrous bath gasification can also be used to produce hydrogen for the coal liquefaction process.

Relationship to Prior Technology

The Sumitomo liquefaction process is an extension of research conducted in the mid 70's for the production of binder pitch. This process is similar to the Exxon Donor Solvent process developed in the U.S.

Sumitomo Solvent Extraction
Coal Liquefaction Process

Table 1, Part 1. TECHNOLOGY FACT SHEET:

NEW TECHNOLOGY (PROCESS DESCRIPTION AND GOALS): The Sumitomo Coal Liquefaction Process is a direct coal liquefaction process based on providing hydrogen to liquefy a coal derived solid. Tetralin is used in the process as the donor solvent. Goals include pilot plant improvements and the design and construction of a 250 ton/day demonstration plant.

RELATIONSHIP TO PRIOR TECHNOLOGY (INCLUDING STATE OF DEVELOPMENT OF PRIOR TECHNOLOGY):

The Sumitomo technology is an extension of SRC-I technology and similar to SRC-II and EDS technology under development in the U.S. The Sumitomo separates the hard-to-liquefy portion of the feedstock for hydrogen production in a molten iron coal gasification reactor.

CHARACTERISTICS OF THE TECHNOLOGY:

CHARACTERISTICS	NEW TECHNOLOGY
PRIMARY OUTPUT (DESIGN CASE)	Syncrude
TYPE OF PROCESS	Solvent extraction
FEEDSTOCK REQUIREMENTS	Subbituminous coal
OVERALL THERMAL EFFICIENCY (INCLUDING BY-PRODUCTS)	-68% (54% excluding minor products)
CARBON CONVERSION EFFICIENCY	750°F-850°F
OPERATING TEMPERATURE	1450 PSI - 2200 PSI
OPERATING PRESSURE	Methane Propane/Butane Sulfur
BY-PRODUCTS	

Table 1, Part 2. **TECHNOLOGY FACT SHEET:** Sumitomo Solvent Extraction
Coal Liquefaction Process

STATUS OF DEVELOPMENT:

OPERATING FACILITIES - 1 metric ton/day pilot plant at Hasaki Research Center in Ibaraki Prefecture.
Tests began in March, 1981. Component testing will continue through June 1981.

MAJOR FUNDING AGENCY - MITI Sunshine Program

ANNUAL LEVEL OF FUNDING -

TECHNICAL PROBLEMS: Particulate and residue separation from the solvent in its recovery step.

Corrosion/erosion of pumps, valves and pipes.

Reactor improvement or redesign from a tubular unit to an autoclave could be required.

OTHER FACTORS AFFECTING OVERALL FEASIBILITY: Means of generating needed hydrogen (the Ferrrous Bath

Gasification Process) requires development. The overall process is contingent on a reasonably

priced reliable hydrogen supply. About 5 X 10⁸ CF of hydrogen are required per day for a nominal

60,000 bbl/day facility.

Operating Facilities

With experience gained from SMI's binder pitch research and SCM's solvent refined coal solid production investigation,³ SCM and SMI constructed a 1 metric ton/day pilot plant at the Hasaki Research Center in Ibaraki Prefecture. This pilot plant, which produces a heavy, medium and lightweight liquefied coal, started preliminary operation in February of 1980. Full operation of the pilot plant began in March of 1981.⁴

Major Funding Agencies

Funding for this process comes from the Japanese Ministry of International Trade and Industry (MITI). A discussion on the continuation of this process will be made by MITI and the New Energy Development Organization (NEDO) in mid 1982.

Technical Problems

The Sumitomo coal liquefaction process is currently in the pilot plant stage of development. Due to the lack of available data and operating experience critical problems for this process have yet to surface. But because this process is similar to other coal liquefaction processes — in particular the SRC and EDS process — parallel problems might be expected to occur. In the SRC process corrosion/erosion problems occur in four main process areas that are similar to those found in the Sumitomo liquefaction process. These areas include coal receiving and preparation, preheating and dissolving, filtration and mineral residue drying, and solvent recovery.

In the area of coal receiving and preparation typical problems associated with similar facilities occurred in coal slurry centrifugal pumping equipment and high-pressure, plunger-type slurry preheater charge pumps. These problems centered on packings and seal leakage, as well as check valves and plunger erosion. In the preheating and dissolving sections a few material problems developed. These minor problems involve nozzle sleeve lining corrosion and scale formation. But severe stress corrosion cracking of Type 304 stainless-steel lining occurred in the high- and intermediate-pressure separator equipment. This stress corrosion cracking was attributed to polythionic acid and chloride stress corrosion cracking at weld sites. Corrosion was not experienced in any of the other materials in the separators, including carbon steel.

Corrosion/erosion material problems also occurred in the filtration and mineral residue drying areas. Most problems occurred in the heat exchanger material where severe erosion of metal tubes was encountered as well as tube bulging due to carburization in the 3/8 inch thick 304 SS drying shell.

Severe corrosion/erosion problems have also occurred in the reboilers, air coolers, fractionation equipment, piping, and tower walls of the solvent recovery section of the SRC process. These problems have caused several major shutdowns. The most severe corrosion occurred in moving parts and materials that came in direct contact with liquid solvent. Most of the problems required a change in material specifications to alleviate the situation. However, acceptable corrosion rates have yet to be achieved in areas such as the stainless-steel trays used in the separator towers.

For the most part, the problems discussed above are applicable to most coal liquefaction processes and not just the SRC processes or the Sumitomo liquefaction processes. For this reason these generic problems will probably not hinder any one process toward commercial development.

Capital Costs

The investment cost of the Sumitomo coal liquefaction process developed by Sumitomo Coal Mining Co. Ltd. and Sumitomo Metals IND., Ltd. has not been published. However, based on a process flow sheet comparison with other coal liquefaction processes, the Sumitomo process does not differ significantly from the EDS (Exxon Donor Solvent) process which is being developed by the Exxon Research and Engineering Co. in the U.S. (see Figures 2 and 3.²⁹) Based on this comparison, the two processes should have similar capital cost requirements. The total plant investment costs for the EDS process (in 1975 dollars) are shown in Table 2.³⁰ These costs include engineering, land, plant, and G&A costs. Coal gasification facilities for hydrogen production have been substituted for the naptha reformers of the 1975 EDS design. Also included is a 20% contingency factor based on the state of the technology readiness. All costs are based on a facility that uses 25,000 tons/day of coal and produces 60,000 barrels/day of liquid products.

Table 2. TOTAL PLANT INVESTMENT FOR 25,000 TON/DAY
LIQUEFACTION FACILITY

	1975 Dollars (millions)	1980 Dollars (millions)
Offsites (including coal preparation, handling, product tankage, loading, utilities)	300	660
Liquefaction Facilities	300	660
Solvent Hydrogenation	100	220
Hydrogen Recovery and Gasification	450	1000
Gas and Water Treating	<u>60</u>	<u>130</u>
Total (including 20% contingency)	1210	2670

Analysis Cost Basis: \$2.7 billion (1980)

Coal Feed: 25,000 tons/day @ 12,500 Btu/lb (including
2500 tons/day for make-up hydrogen required
in addition to 7500 tons/day coal residue)

JAPANESE COAL GASIFICATION PROCESS

33. Sumitomo Molten Iron Coal Gasification ProcessProcess Description

The Sumitomo process consists of four main systems: coal preparation, coal injection, molten iron gasification, and gas cleanup. Coal is initially sized 20 to 50 mm for storage. From storage the coal is sent to a hot-gas-swept pulverizer where the coal is sized 70% to 80% through 200 mesh and simultaneously dried to 5% to 15% moisture. The dried, pulverized coal is then sent to storage before injection into the gasifier.

One of the most important elements of this process is the coal injection system. This system consists of a specially designed lance that injects coal, steam, and oxygen simultaneously into the molten iron bath. Unlike other submerged lance designs, this non submerged top-blowing lance utilizes a ultra high speed blowing technique to form a cavity in the molten slag as shown in Figure 1.

As the feed is injected into the gasifier a 2000°C flame is generated, and the coal injected at the molten iron surface is cracked in a matter of seconds by the extremely high temperatures. The molten bath assures complete conversion of the reactants while absorbing unwanted elements such as ash and sulfur. The molten ash that forms on the surface of the molten iron bath is skimmed off and withdrawn in the molten state. Part of the slag is composed of CaS, which is formed when the sulfur in the coal reacts with CaO that is added to the molten bath to promote desulfurization. From the literature it is unclear whether the CaO is injected with the pulverized coal or added in a separate manner. The molten iron bath also acts as a buffer when excess coal is dissolved in the bath as carbon. This helps to maintain a steady gas composition.

The possibility also exists to inject scrap metal or iron ore into the bath instead of steam. The steam, which acts as a heat moderator, can therefore be replaced to allow hot metal production as well as synthesis gas production. A typical synthesis gas composition for steam or scrap injection is presented in Table 1.

The pilot plant gasifier has a fire brick lining which contains the 1500° to 1600°C molten iron bath. This pilot plant gasifier has a maximum inner

Table 1. EXAMPLE OF GENERATED GAS ANALYSIS

Generated Gas	Injection of Steam	Use of Scrap
	2150~2220 Nm ³ /T — Coal	2000~2050 Nm ³ /T — Coal
CO	60 ~ 64%	64 ~ 68%
CO ₂	<3%	<3%
H ₂	29 ~ 33%	24 ~ 28%
N ₂ + Ar	<4%	<4%
Calorific value	<2600 Kcal/Nm ³	<2600 Kcal/Nm ³

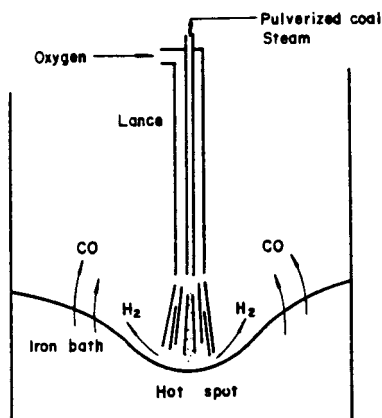


Figure 1. BLOWING METHOD

diameter of 2.3 meters, a height of 4.2 meters, an inner volume of 10 m³ and a 15 ton molten iron bath capacity. The molten iron bath is initially prepared within the gasifier, which is initially operated in the batch mode. Slag removal requires that gasifier operation stop and the vessel be tilted. This system will not be used in commercial operation, however, and current pilot plant operation incorporates a new continuous slag removal system.

From the gasifier the raw synthesis gas flows to the gas treatment system: a two-stage venturi scrubber and a wet wash system. However, a dry dust collector has been installed at the pilot plant to collect dust samples and recycle material back into the gasifier. Figure 2 is a flow diagram of the pilot plant. The material balance and energy balance for this process is shown in Table 2 and Figure 3, respectively. The material balance is representative for operation on coal with the properties shown in Table 3. Table 4 is a technology fact sheet for this process is presented in Table 4.

Process Goals

The objective of this development, which started in 1978, was to develop a molten iron gasification technique. This included:

- Determining facility performance
- Designing for safe operation
- Optimizing operation
- Generating a stable gas composition.

The work was carried out at the Sumitomo Central Research Laboratory where the fundamental concept and optimum lance structure were also investigated. Pilot plant operation, which started in May 1980, will be used to —

- Improve the yield of coal gasification
- Make high-calorie gas with stable composition
- Produce lower sulphur gas by optimizing slag desulfurization
- Minimize iron loss.

Future plans call for the construction of a demonstration facility at Sumitomos Kashima Steel Complex in Kashima, near Tokyo, by 1983-84. A feasibility study is also underway for the construction of a commercial size facility in Australia by the end of 1988.

Table 2. EXAMPLE OF MATERIAL BALANCE

	By Injection of Steam	By Use of Scrap
Coal	2.5T — Coal/H	2.5T — Coal/H
Oxygen	575Nm ³ /T — Coal	2.5T — Coal/H
Steam	>150Kg/T — Coal	--
Carrier gas (Ar)	50Nm ³ /T — Coal	50Nm ³ /T — Coal
Scrap	--	>500Kg/T — Coal
Lime	80Kg/T-Coal	80Kg/T-Coal
Flux	10Kg/T-Coal	10Kg/T-Coal

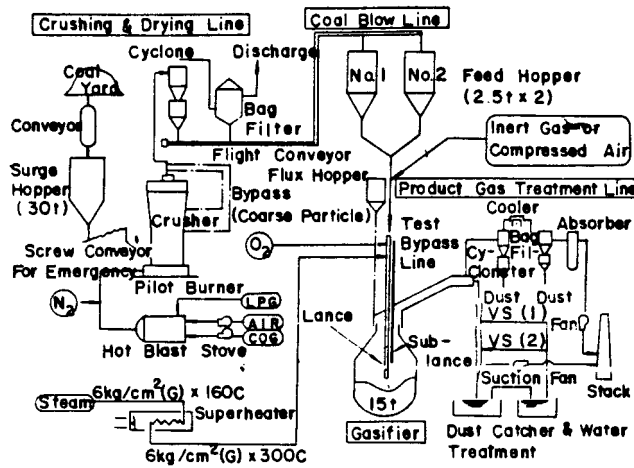


Figure 2. FLOW DIAGRAM OF COAL GASIFICATION TEST PLAN

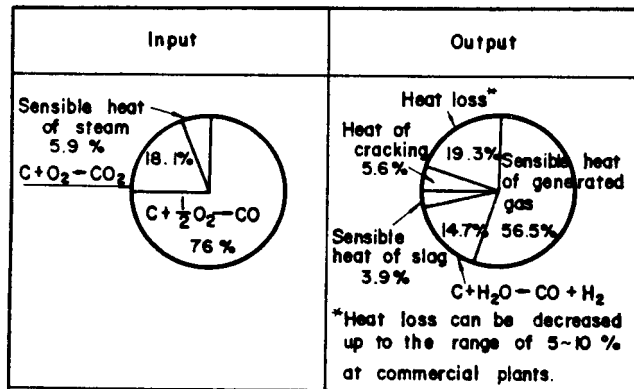


Figure 3. HEAT BALANCE

Table 3. PROPERTIES OF COAL USED FOR THE TEST

<u>Analysis</u>	<u>Coal Sorts</u>	<u>Coal A %</u>
Proximate analysis	F.C.	55.4
	Ash	8.0
	V.M.	34.4
	Mo	2.2
Ultimate analysis (d.a.f.)	C	84.3
	H	5.2
	O	7.9
	N	1.8
	S	0.8
	SiO ₂	50.4
	CaO	8.9
	Al ₂ O ₃	26.8
	P ₂ O ₅	2.3
	Ash analysis	MgO
	MnO	0.1
	Fe ₂ O ₃	3.6
	TiO ₂	1.8
	Na ₂ O	0.2
	K ₂ O	0.2

Table 4 Part 1 TECHNOLOGY FACT SHEET: Sumitomo Molten Iron Gasification Process

NEW TECHNOLOGY (PROCESS DESCRIPTION AND GOALS): This process which is being developed by Sumitomo Metals Industries Ltd., is based on the gasification of coal in a molten bath of iron. The special feature of this process is the injection of coal, oxygen, steam or iron ore into the bath through a specially designed lance. This process is especially suitable to high sulfur coals, since lime injection can be used to help desulfurize the gas within the molten bath.

RELATIONSHIP TO PRIOR TECHNOLOGY (INCLUDING STATE OF DEVELOPMENT OF PRIOR TECHNOLOGY): This process is a modification of the Applied Technology Corp. molten iron process which was developed in the early 1970's in the U.S.

CHARACTERISTICS OF THE TECHNOLOGY:

<u>CHARACTERISTICS</u>	<u>NEW TECHNOLOGY</u>	<u>PRIOR TECHNOLOGY</u>
PRIMARY OUTPUT (DESIGN CASE).....	Synthesis gas (315 Btu/SCF)	Similar
TYPE OF PROCESS.....	molten iron gasification	Similar
FEEDSTOCK REQUIREMENTS.....	all types of coal	
OVERALL THERMAL EFFICIENCY (INCLUDING BY-PRODUCTS)..	~70%	
CARBON CONVERSION EFFICIENCY.....	99.9%	
OPERATING TEMPERATURE.....	2000°	
OPERATING PRESSURE.....	1 atm	
BY-PRODUCTS.....	none	

Table 4 Part 2 **TECHNOLOGY FACT SHEET:** Sumitomo Molten Iron Coal Gasification Process

STATUS OF DEVELOPMENT:

OPERATING FACILITIES - 60 ton/day pilot plant has been in operation since May 1981 at Sumitomo's
Kashima Steel Complex in Kashima near Tokyo.

MAJOR FUNDING AGENCY

Agency at Industrial Science and Technology in MITI of the Japanese government.

ANNUAL LEVEL OF FUNDING -

Molten iron losses are due to high coal injection rates.

TECHNICAL PROBLEMS:

OTHER FACTORS AFFECTING OVERALL FEASIBILITY: Thermal efficiency of the process and economics of building

this system outside the steel industry.

Relationship to Prior Technology

A coal gasification process similar in nature was developed by Applied Technology Corp. in the early 1970's. A paper describing this process is presented as an Exhibit. In addition, the West Germans are developing the Kloeckner process (presented elsewhere in this report) which is also based on the Applied Technology design. The primary development in both the Japanese and West German processes is in coal injector systems.

Operating Facilities

Research on the basic molten iron gasification concept was started by Sumitomo Metals Industries, Ltd., in January 1978. The schedule of the technical developments to date for this process are presented in Table 5. Sumitomo is currently operating a 60 ton/day coal gasification pilot plant at then Kashima steelworks.

Major Funding Agency

A portion of the financing for pilot plant construction and operations is sponsored by the Agency of Industrial Science and Technology in the Ministry of International Trade and Industry of the Japanese government.

Technical Problems

The main problem with the previous molten iron gasification design was control and stability of the lance coal injection system. Sumitomo seems to have overcome the severe corrosion and mechanical stability problems which were a major problem in the Applied Technology Corp. research effort. No other technical problems have been published in the literature. However, Sumitomo seems to be having a problems with molten iron loss. This is probably due to the rapid injection rate. This does not look like a difficult problem to solve.

Capital Costs

Sumitomo has not published the capital cost information for this process. This process may be less capital intensive where it is possible to modify a steel-making facility for synthesis gas generation. Otherwise this process will probably be more capital intensive for applications outside the steel industry.

EXHIBIT. Fuel Gas From Molten Iron Coal Gasification

FUEL GAS FROM MOLTEN IRON COAL GASIFICATION
(190 - 940 BTU/ft³)

Paul LaRosa
Director of R&D
Ronald J. McGarvey
Project Engineer
Applied Technology Corporation
135 Delta Drive
Pittsburgh, Pennsylvania 15238

ABSTRACT

Applied Technology Corporation is developing several coal gasification processes to produce fuel gases with heating values ranging from 190 to 940 BTU/SCF. These processes are based on the molten iron gasification concept in which coal is injected with steam or air into a molten iron bath. Steam dissociation and thermal cracking of coal volatile matter generate hydrogen and carbon monoxide. The coal sulfur is captured by the iron and transferred to a lime slag from which elemental sulfur can be recovered as a by-product. The coal dissolved in the iron is removed by oxidation to carbon monoxide with oxygen or air injected near the molten iron surface.

Three processes are discussed: The Two-Stage Coal Combustion Process which uses coal/air to yield a 190 BTU/SCF offgas; the PATGAS and ATGAS Processes which use coal/steam/oxygen to yield product gases with heating values of 315 and 940 BTU/SCF, respectively.

The main advantages of molten iron coal gasification are its low pressure operation and its complete flexibility regarding coal properties. This method uses crushed (minus 1/8 inch) coal regardless of sulfur content, caking, or fines generation tendencies. All of the coal is gasified and there are no char or fines by-products to be marketed. Capital investments are 28, 85-112, and 202 \$MM for the Two-Stage, PATGAS, and ATGAS Processes, respectively based on a gas BTU output equivalent of a 250 MMSCFD SNG plant. Using the standard gas industry price estimating method, the estimated 20-year average price of gas ranged from \$0.35 (Two-Stage, 190 BTU/SCF) to \$1.10/MMBTU (ATGAS, 940 BTU/SCF) when coal is available at \$0.30/MMBTU.

INTRODUCTION

The expanding economy and rising standard of living in the United States has resulted in an increased demand for new energy sources and alternative means for utilizing available energy reserves. As coal represents much of the known recoverable energy reserves of the United States it must, of necessity, be the base for meeting future energy needs. Coupled with the increasing energy demand is an acute awareness of environmental protection. These environmental factors place restrictions upon coal utilization because of its sulfur content. These factors require that economically and environmentally viable processes be developed to produce clean fuels from sulfur bearing coal.

To expand coal's participation in total energy consumption, Applied Technology Corporation (ATC), a subsidiary of Houston based International Systems and Controls, is developing (under sponsorship of the Environmental Protection Agency and the American Gas Association) several coal gasification processes. These processes are based on ATC's Molten Iron Coal Gasification method which has the capability of gasifying coal and other carbonaceous fuels (independent of sulfur content or caking tendencies) to produce essentially sulfur-free fuel gases. The processes are: The Two-Stage Coal Combustion Process which produces a hot low BTU fuel gas (190 BTU/SCF) for combustion in power plant boilers, the PATGAS process which produces an intermediate BTU fuel gas (315 BTU/SCF) for combustion or as a synthesis gas, and the ATGAS process for substitute natural gas (SNG-940 BTU/SCF).

GASIFICATION BACKGROUND

Molten Iron Coal Gasification Concept

The strong affinity between sulfur and iron and the solubility of carbon in iron form the basis of the Molten Iron Coal Gasification method. In this new coal gasification method, coal is injected into a bath of molten iron. The coal's fixed carbon and sulfur dissolve and are retained by the iron, whereas the coal volatiles crack and appear as offgas. The solution rate of coal in molten iron is mass transfer controlled^{1,2*}. Since coal contains volatile matter it literally explodes when contacted with the molten iron (2500°F) to produce tremendous increases in surface area. Because of this surface area increase, a relatively short residence time is required for total coal solubilization.

*Numbered references are listed at the end of this paper

This carburization process is common to all three of the Molten Iron Coal Gasification Processes, i.e., Two-Stage, PATGAS, and ATGAS. The processes, however, are distinguished by the decarburization process, i.e., the oxidation of the carbon dissolved in iron to carbon monoxide.

In the Two-Stage Process, compressed air is used to inject the coal into the iron. The dissolved carbon is gasified by reaction with additional combustion air which is injected slightly below the surface of the molten iron to yield carbon monoxide to the offgas.

This hot offgas (30% CO, 15% H₂, 55% N₂) is then utilized as a low-BTU (190 BTU/SCF) fuel gas for power plant boilers. To maintain the gasifier in thermal balance, the combustion air is preheated (1000-1200°F).

In the PATGAS and ATGAS processes, steam is used as the coal carrier and the dissolved carbon is gasified by injecting oxygen (rather than air) slightly below the molten iron surface. Because of the high temperature, the steam dissociates. These dissociation products plus the injected oxygen react with the dissolved carbon to yield carbon monoxide and hydrogen to the offgas. In these oxygen based processes, steam must be used to moderate the iron temperature and keep the gasifier in thermal equilibrium. The offgas resulting from steam and oxygen injection (64.5% CO, 35% H₂, 0.5% N₂) can be utilized in several ways. In the PATGAS process, the offgas is used as an intermediate BTU fuel gas (315 BTU/SCF) or as a synthesis gas to produce other organic compounds. In the ATGAS process, the offgas is subjected to shift conversion, methanation, and compression to produce an SNG product (940 BTU/SCF).

By equalizing the carburization and decarburization rates, a steady state carbon level in the molten iron is achieved. By maintaining the molten iron carbon content at a relatively high level (3 to 4%), the injected air (or steam and oxygen) react preferentially with the dissolved carbon without sulfur oxidation. Thus, the Molten Iron Coal Gasification method produces a CO-H₂ bearing offgas that is essentially free of sulfurous compounds.

To prevent the sulfur content of the molten iron from building up, the dissolved sulfur is continuously transferred to a molten, lime-bearing slag floating on top of the molten iron. The slag layer contains lime to react with the dissolved sulfur to produce calcium sulfide and to flux the ash constituents of the coal. Lime is added to the gasifier in the form of limestone mixed with the coal.

The gasifier slag, containing 4 to 8% sulfur as calcium sulfide and all the coal ash, is continuously removed and desulfurized with steam

to yield desulfurized lime-bearing slag, and an elemental sulfur-hydrogen sulfide offgas. The desulfurization offgas is cooled to condense the elemental sulfur and the remaining H_2S gas mixture proceeds to a Claus plant for further sulfur recovery. A portion of the desulfurized slag is recycled to the gasifier to utilize its lime content. The remaining desulfurized slag is for use as road-building aggregate.

A number of carbonaceous materials, from petroleum coke to sub-bituminous coals, regardless of coking properties with a wide range of sulfur and ash contents can be gasified to clean gaseous fuels by this method; by-products consisting of desulfurized slag, elemental sulfur, and granulated iron are environmentally acceptable for stockpiling in the event they cannot be sold.

Gasifier

A conceptual drawing of the Molten Iron Coal Gasification vessel is shown in Figure 1. The gasifier is a cylindrical, refractory-lined vessel which contains molten iron and a floating slag layer. Coal and limestone are injected through carburization lances into the molten iron. In the Two-Stage Process, compressed air is used to convey the coal/limestone whereas for PATGAS and ATGAS, the coal/limestone mixture is injected using steam as the carrier. By simultaneously injecting air (Two-Stage) or oxygen (PATGAS/ATGAS) slightly below the surface of the iron, the dissolved carbon from the coal is oxidized to carbon monoxide. In PATGAS and ATGAS, the injected steam dissociates and reacts with the dissolved carbon to produce hydrogen and additional carbon monoxide to the offgas. Thus, the cracking of coal volatiles, the oxidation of dissolved carbon with air or oxygen, and the dissociation of steam (PATGAS/ATGAS) produce a hot offgas containing carbon monoxide and hydrogen.

Limestone is added to the gasifier to flux the coal ash constituents and to remove sulfur from the molten iron. The gasifier is equipped with a port for the removal of slag which is desulfurized with steam to recover elemental sulfur. As coal contains pyrites, iron is also produced; therefore, to maintain the iron level relatively constant, iron is removed from the gasifier and granulated for sale.

Experimental Result Summary

The experimental work, sponsored by the EPA has the objective of developing the Two-Stage Process so that high sulfur coals can be gasified with air for electrical power generation. Under the AGA sponsorship, experimentation was conducted to demonstrate the feasibility of steam-oxygen injection into molten iron to produce a carbon monoxide/hydrogen offgas.

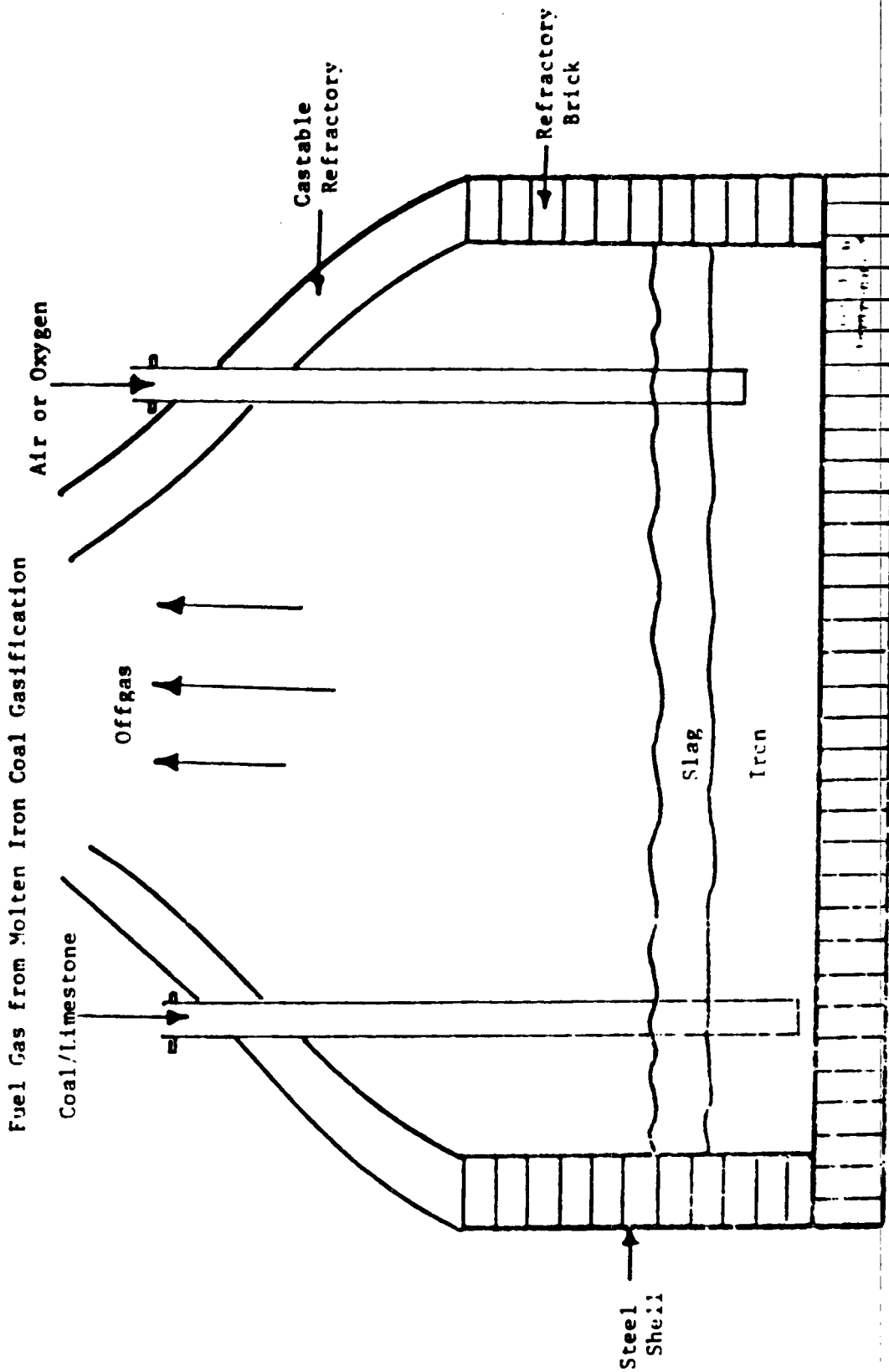


FIGURE 1-1-1 - CROSS SECTION OF MOLTEN IRON COAL GASIFIER

Under EPA sponsorship, data have been obtained using the gasifier shown schematically in Figure 2. An induction furnace (4000 lb capacity, with 25 inch I.D.) was selected to simulate the gasifier as it is a convenient device for preparing and maintaining a molten iron bath at a specified temperature. The offgas handling system was equipped with gas analyzers to permit continuous analysis for SO_2 , NO_2 , NO , H_2 , O_2 , CO_2 , and CO content. Following sampling, the offgas was flared and quenched prior to exhausting to the atmosphere. An EPA-type particulate sampling train was utilized to collect offgas particulate samples.

Experimentation consisted of simultaneous coal and air injection into the molten iron bath using medium volatile bituminous coals with 0.6 to 3.5% sulfur. Expendable non-cooled ceramic lances were used for injection with a pneumatic transport system for coal injection. Slags consisting of CaO , CaS , SiO_2 , and Al_2O_3 with low basicity (0.1 to 0.6) were used to cover the molten iron bath.

The primary objective of simultaneous coal-air injection experimentation was to determine the offgas composition during steady-state operation using high sulfur coal and high sulfur-low basicity slag. In addition to the primary study of SO_2 and NO_x content in the combustor offgas, the concentrations of other gaseous compounds (H_2 , CO_2 , O_2 , and CO) were determined. Data were also obtained during this work on the combustor offgas dust loading, the sulfur retention of low basicity slags and refractory life.

The results of this experimentation show that a boiler feed gas essentially free of SO_2 (less than 50 ppm), can be generated using high sulfur coal (3.5% sulfur) and low basicity (0.1-0.6) slags.

Preliminary dust loading data show that the combustor offgas particulate concentrations can be lowered by a 95 to 97% efficient electrostatic precipitator to meet EPA Standards of Performance. The dust loading data obtained are felt to be conservative due to size limitations and operational difficulties of the experimental combustor. With a larger diameter gasifier, an effective freeboard zone (area above the iron/slag bath) can be incorporated to significantly reduce particle entrainment and offgas particulate concentrations.

A slag study indicated that sulfur does transfer from the iron to the slag. Slag sulfur contents in excess of 14% were achieved; however, as expected from previous bench-scale studies, these high sulfur slags (about 10% sulfur) exhibit an increase in viscosity. Although high sulfur retention by the slag is desirable, the associated increase in viscosity could present some problems in slag removal from the gasifier. Consequently, for best flow characteristics, the slag sulfur content should be maintained at the design specification, 4 to 8%.

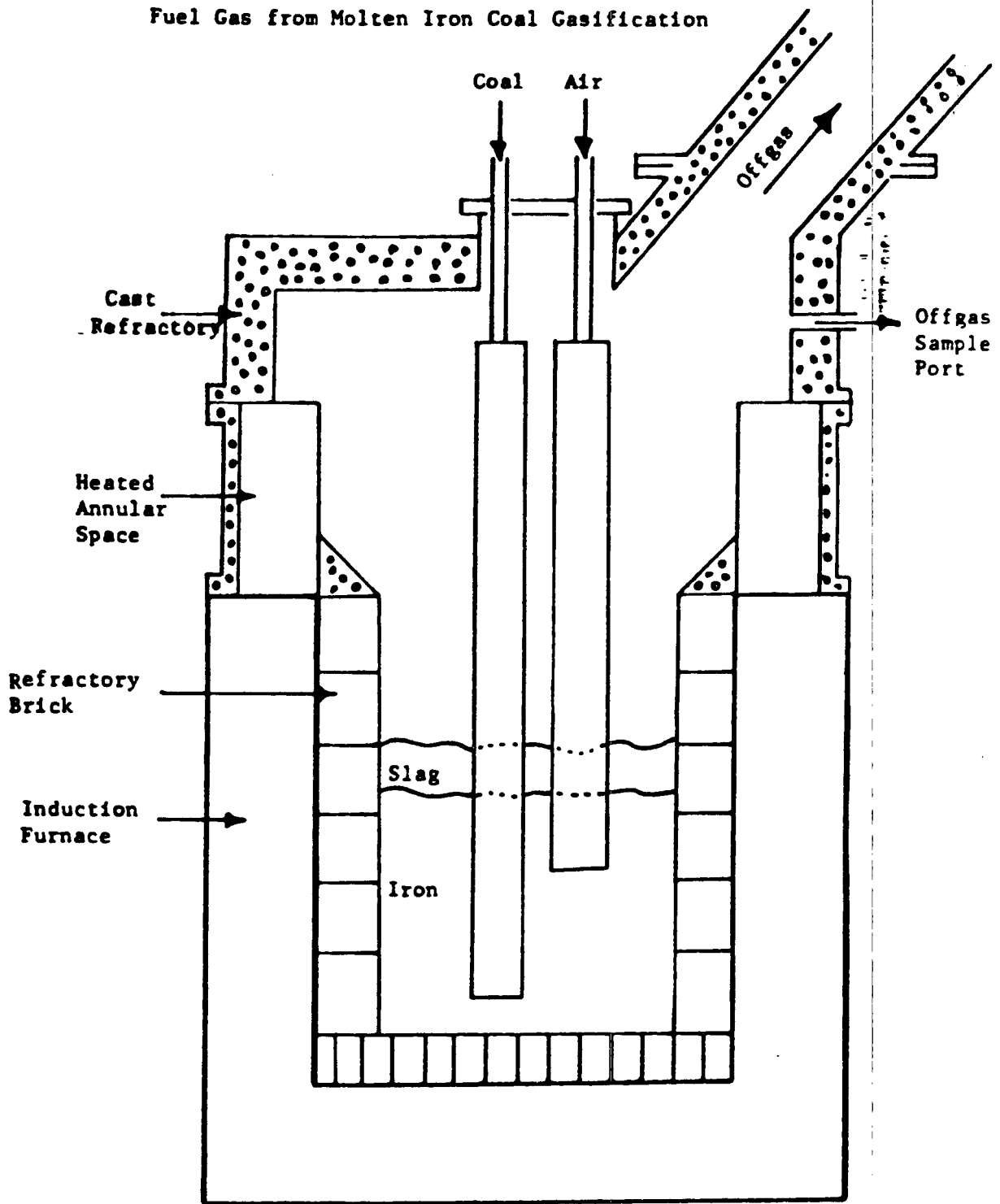


FIGURE 2--EXPERIMENTAL GASIFIER

Under a current EPA contract (Contract No. 68-02-1368 July, 1973-75) data will be collected to determine the refractory life of the various commercial bricks used in the gasifier over prolonged periods of time. Present data based on two and four week experimental tests suggest that a number of commercial bricks may provide a satisfactory combustor service life of about one year.

A study of coal trace element accumulation in the molten iron and/or slag will be the subject of a study under the current EPA contract. Results to date are encouraging and indicate that nickel and vanadium are essentially totally captured by the iron and slag. By retaining such trace elements in the slag/iron, they will not be present in the offgas to pollute the atmosphere and foul catalysts when the offgas is subjected to further processing.

Bench-scale experimentation on the affect of weather conditions on sulfur release from sulfur-bearing slag and the use of oil-cooled lances for coal/air injection have been completed. The results show that desulfurized slags containing as high as 3% sulfur can be stockpiled without adverse environmental effects. Experimentation with oil-cooled metal lances submerged in molten iron has shown that lances are a technically feasible means of injecting coal and air into molten iron.

Under AGA sponsorship, the feasibility of steam and oxygen injection into molten iron to produce a carbon monoxide/hydrogen offgas was demonstrated. These tests (using the EPA experimental gasifier) demonstrated the feasibility of the Molten Iron Coal Gasification Concept and the PATGAS and ATGAS processes. Specifically, the results showed that:

1. Steam and oxygen react with carbon dissolved in molten iron to form a gas mixture rich in hydrogen and carbon monoxide.
2. The offgas produced by the molten iron coal gasification method is essentially free of sulfur compounds and analysis of the offgas showed that neither hydrogen sulfide nor carbonyl sulfides were present.

PROCESS DESCRIPTION

A brief summary of the Molten Iron Coal Gasification Process is shown in Table I. In the following sections, a brief description of each process is presented with schematic flow sheets and major process streams for each process.

TABLE I
Process Description

Process *	Raw Materials	Product Gas		Pressure, PSIG	Heating Value, BTU/SCF
		Composition	Temperature, °F		
Two-Stage	Coal	30% CO	2040	2-5	190
	Air	15% H ₂			
	Limestone	55% N ₂			
PATGAS	Coal	63.5% CO	100	2/1000	315
	Oxygen	36% H ₂			
	Steam	0.5% N ₂			
	Limestone				
ATGAS	Coal	93% CH ₄	100	1000	940
	Oxygen	4% H ₂			
	Steam	3% Inerts			
	Limestone				

* All processes yield as by-products granulated iron, elemental sulfur and desulfurized slag.

Two-Stage Coal Combustion Process

The Two-Stage Process is shown in Figure 3 with the important process streams numbered. A description and the rates of these streams for a 1240 mw power facility are given in Table II. For economic comparison of the various Molten Iron Coal Gasification Processes, each has been sized to yield approximately the same BTU/day output -- equivalent to a 250 MMSCFD SNG plant producing a 940 BTU/SCF product gas.

TABLE II
1240 MMSCFD Two-Stage Process Streams

<u>Stream No.</u>	<u>Description</u>	<u>Rate, TPH</u>
1	Coal, 4% Sulfur 13,100 BTU/lb	386
2	Combustion Air	1377
3	Limestone	24
4	Transport Air	27
5	Boiler Feed Gas (30% CO, 15% H ₂ , 55% N ₂)	52 MMSCFH
6	Iron	6
7	Elemental Sulfur	14
8	Desulfurized Slag (1% Sulfur)	47

As received coal is crushed (minus 1/8 inch), dried, combined with limestone, and injected pneumatically using compressed air into the molten iron bath. Combustion air to gasify the dissolved carbon-in-iron to carbon monoxide is compressed and preheated via heat exchange with the hot gasifier offgas prior to injection into the molten iron bath. Sulfur-bearing slag is continuously removed and enters a desulfurization unit where it is contacted with steam to generate an elemental sulfur H₂S-bearing offgas. After condensation of the elemental sulfur, the H₂S-bearing gas proceeds to a Claus plant for additional sulfur recovery. A portion of the resulting desulfurized slag is recycled to the gasifier to utilize its lime content and the remainder is available for sale as road building aggregate. The hot gasifier offgas proceeds to a heat exchanger to preheat combustion air and then to the power plant boiler for steam generation.

Fuel Gas from Molten Iron Coal Gasification

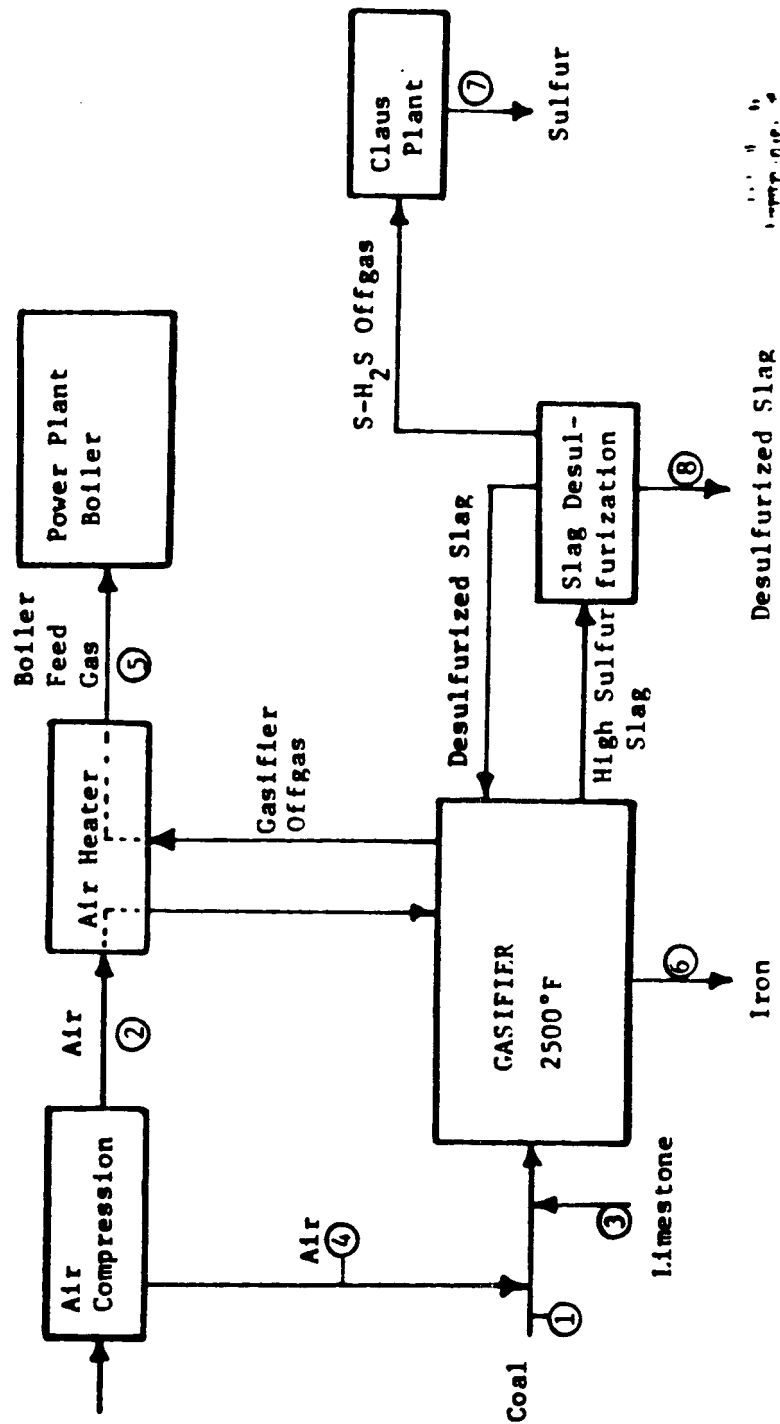


FIGURE 3--TWO-STAGE COAL COMBUSTION PROCESS

PATGAS Process

The PATGAS Process is shown in Figure 4 with the important process streams numbered. A description of the streams is presented in Table III.

TABLE III
750 MMSCFD PATGAS Process Streams

<u>Stream No.</u>	<u>Description</u>	<u>Rate, TPH</u>
1	Coal, 4% Sulfur 13,100 BTU/lb	467
2	Oxygen (50 psig)	276
3	Limestone	31
4	Steam for Coal/ Limestone Injection (50 psig)	52
5	PATGAS 100°F, (63.5% CO, 36% H ₂ , 0.5% N ₂)	31 MMSCFH
6	Iron	15
7	Elemental Sulfur	18
8	Desulfurized Slag (1% S)	47

As received coal is crushed, dried, combined with limestone and injected pneumatically using steam into the gasifier molten iron bath. Oxygen from an onsite oxygen plant is injected into the gasifier to gasify the dissolved carbon-in-iron to carbon monoxide. The coal/limestone transport stream also dissociates to give additional carbon monoxide and hydrogen to the gasifier offgas. Sulfur-bearing slag is processed as in the Two-Stage Process with steam to produce elemental sulfur. Part of the desulfurized slag is recycled to the gasifier and the remainder is utilized as road building aggregate.

The high temperature offgas enters a waste heat boiler to produce high-pressure process steam. This steam is utilized in the oxygen plant to drive compressors and for slag desulfurization. The cooled offgas is scrubbed and further cooled to remove excess moisture. The resulting PATGAS can then be utilized onsite or compressed to 1000 psig. PATGAS has a heating value of 315 BTU/SCF and can be burned as a fuel gas used as a synthesis gas.

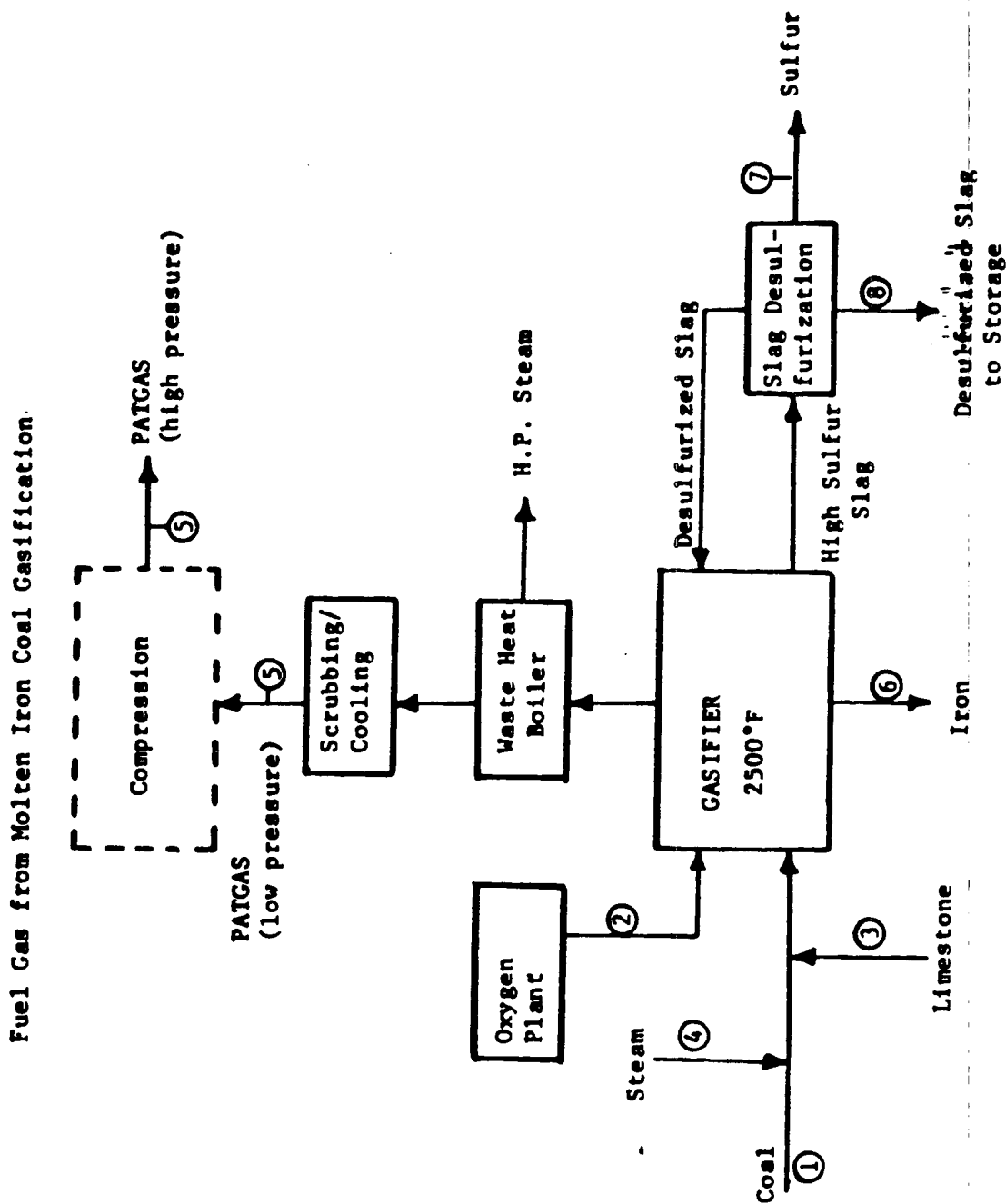


FIGURE 4--PATGAS PROCESS

ATGAS Process

The ATGAS Process is shown in Figure 5 with the important streams numbered. These streams are described in Table IV for a 250 MMSCFD plant.

As seen in Figure 5, the operations of ATGAS are the same as PATGAS to the point of producing a low pressure PATGAS. The low pressure PATGAS is then compressed to about 600 psig and fed to a shift converter where a portion of the carbon monoxide is reacted with steam to yield a hydrogen to carbon monoxide ratio of 3 : 1 in the gas. The carbon dioxide produced is removed in a hot carbonate scrubbing purification unit resulting in less than 2% carbon dioxide in the gas. The gas then enters a methanation unit where the carbon monoxide and hydrogen react to methane. The methane rich gas is cooled to remove excess water and then compressed to yield ATGAS (SNG 940 BTU/SCF) at 1000 psig. Steam produced in the waste heat boiler is used for gas compression, oxygen plant compression needs, slag desulfurization, and shift conversion.

TABLE IV
250 MMSCFD ATGAS Process Streams

<u>Stream No.</u>	<u>Description</u>	<u>Rate, TPH</u>
1	Coal, 4% Sulfur 13,100 BTU/lb	641
2	Oxygen (50 psig)	379
3	Limestone	43
4	Steam for Coal- Limestone Injec- tion (50 psig)	71
5	Gasifier Offgas (2500°F, 65% CO, 35% H ₂)	43 MMSCFH
6	Iron	20
7	Sulfur	25
8	Desulfurized Slag (1% S)	64
9	ATGAS (100°F 1000 psig, 93% CH ₄ , 4% H ₂ , 3% Inerts) ⁴	10.5 MMSCFH

Fuel Gas from Molten Iron Coal Gasification

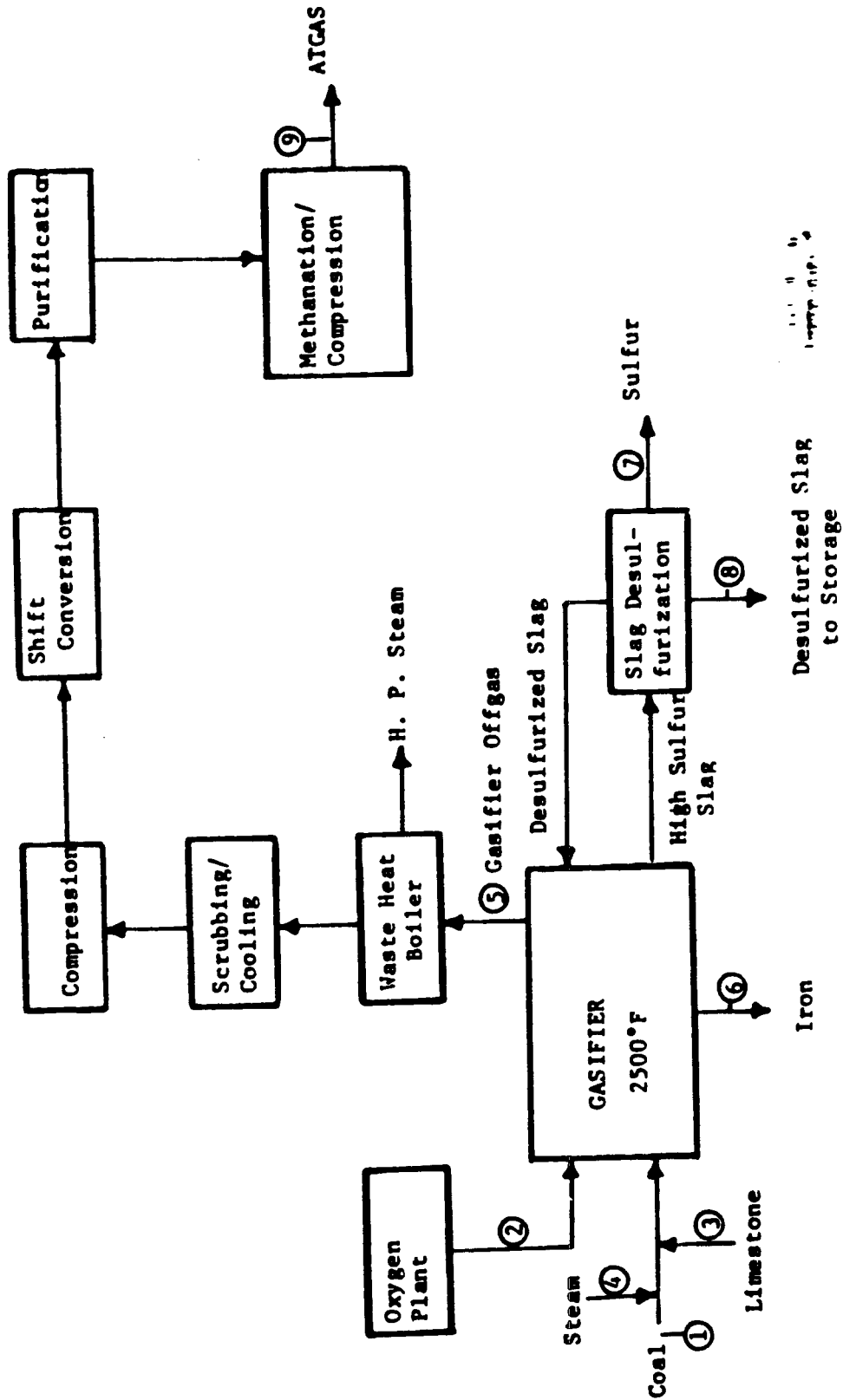


FIGURE 5--ATGAS PROCESS

Advantages

The Molten Iron Coal Gasification Processes have several inherent advantages over gas-solid contact gasification in either fixed-bed or fluidized-bed reactors. Their advantages are summarized as follows:

1. Since gasification is conducted at a low pressure, the mechanical problem of feeding coal into high pressure gasifiers is eliminated.
2. Coking properties, ash-fusion temperatures, and fines generations of coal are not a problem.
3. Coal sulfur content creates no problems as the sulfur is retained in the system and recovered as elemental sulfur from the slag.
4. The extreme flexibility with regard to the physical properties of the coal used and its sulfur content permits economical use of coals that cannot be readily marketed elsewhere.
5. Relatively coarse crushed coal having a wide size consist can be utilized without any special pretreatment.
6. Char is not produced and the high gasification temperature eliminates tar formation.
7. The product gas is essentially free of sulfur compounds.
8. Shutdown and startup procedures are greatly simplified as compared to fixed or fluidized-bed reactors (molten iron can be contained in the vessel over long periods without use -- for almost immediate startup).
9. Gasifier response time is short and turndown ratios can be varied between 0 and 100% without adversely affecting the unit.

PROCESS ECONOMICS

To establish the economic feasibility of the Molten Iron Coal Gasification Processes, the selling price of gas was estimated for each of the process plants described in the previous section. As noted previously, these process plant sizes are comparable in terms of the total heating value of the product gas produced per day. The resulting capital cost and gas selling price estimates are shown in Table V. The method used to estimate the price of gas was developed by the American Gas Association and the Institute of Gas Technology for the Office of Coal Research³.

TABLE V
Process Costs

<u>Process</u>	<u>Plant Capacity</u>	<u>Capital Investment \$MM</u>	<u>20 Yr Average Gas Price \$/MMBTU</u>	<u>Process Efficiency, %</u>
Two-Stage	1240 MMSCFD of 190 BTU/SCF Gas	28	0.35	98
PATGAS	750 MMSCFD of 315 BTU/SCF Gas	85 (Low Pressure) 112 (High Pressure)	0.51 0.58	72
ATGAS	250 MMSCFD of 940 BTU/SCF Gas	202	1.10	57

Cost estimates were based on the following: 20 year straight line depreciation, a gross return of 9% of the total capital investment, 7.5% interest cost, debt financing of 65% of the capital and a 48% tax rate.

For estimating purposes, a cost of \$0.30/MMBTU for coal and \$4/ton for limestone was used. By-product credits for sulfur (\$20/ton), iron (\$20/ton), and road aggregate slag (\$1/ton) were assumed. The capital cost for PATGAS and ATGAS includes the cost of an oxygen plant. In Table V PATGAS costs are shown for both the low and high pressure alternatives.

As seen, the selling price of the Two-Stage boiler feed gas is 0.35, PATGAS is 0.51 to 0.58, and ATGAS is 1.10/\$ MMBTU. Capital cost estimates ranged from \$28 MM for the Two-Stage Process to \$202 MM for ATGAS. Also shown in Table V is the process efficiency defined as,

$$\text{Process Efficiency} = \frac{\text{BTU's in Product Gas}}{\text{BTU's in Coal}} \times 100$$

for the various processes. The Two-Stage Process is the highest at 98% because the gas is utilized hot without compression. PATGAS at 72% has a higher efficiency than ATGAS at 57% mainly because shift conversion (which removes CO as CO₂ from the system) is not used.

REFERENCES

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2. Olsson, R. G., Koump, V., and Penzak, T., "Rate of Solution of Carbon in Molten Iron Carbon Alloys", Annual AIME Meeting of February, 1965.
3. Tentative Standard for Cost Estimating of Investor Owned Plants for Providing Pipeline Gas from Coal, Dept. of Interior, Office of Coal Research, Washington, D. C., June 4, 1965.

JAPANESE COAL LIQUEFACTION PROCESS

34. Mitsubishi Heavy Industries Solvolysis Coal Liquefaction ProcessProcess Description

The solvolysis process for coal liquefaction is being developed under the Japanese Sunshine Project supported by joint public-private funds. Among the main process features are a) the use of petroleum heavy oil as solvent instead of process-derived heavy oil and b) considerably less severe liquefaction process conditions (350° to 450°C and 0.1 to 2 MPa or about 1 to 20 atm total pressure). The liquefaction step yields solvolysis pitch (presumably a preasphaltene-asphaltene mixture in solvent), gas (mainly methane), and cracked oil (mostly n-paraffins). The liquid products undergo catalytic hydroprocessing to yield desulfurized oil.

Figure 1 shows the schematic flow diagram of the 1 tonne/d solvolysis PDU. No detailed operating procedure for the PDU is available. The procedure, however, appears to be straightforward according to the flow diagram.

Feed coal preparation (size reduction, drying, etc.) is assumed to be based on the conventional method. Crushed coal (30 to 100 mesh; particle size distribution unclear) is blended with petroleum-derived heavy oil (asphalt) in the slurry preparation tank. The coal slurry (about 33 wt% coal) is fed to the reactor after going through the preheater where it is heated to about 360°C. The major portion of the "solvolysis reaction," including solubilization of coal and conversion to preasphaltene and asphaltene, takes place in the reactor at about 380° to 450°C and about 0.1 to 2 MPa (about 1 atm-20 kg/cm²), to produce gas (methane-rich), cracked oil (mainly n-paraffins), and "solvolysis pitch."

The liquid product is brought into contact with a "suitable" organic solvent to separate solvolysis residue containing ash and insoluble matter, and refined solvolysis pitch is recovered from the extract. The ash-free pitch (a mixture of solubilized coal and petroleum asphalt) is transferred to the distillation tower where it is separated into different fractions of oil.

Since the development of the original solvolysis process, several modifications have been made to the process as well as the equipment. One such modification is the change in solvent for coal slurry from asphalt (petroleum-derived heavy oil) to process-derived, hydrotreated heavy oil. A technology fact sheet for this process is presented in Table 1.

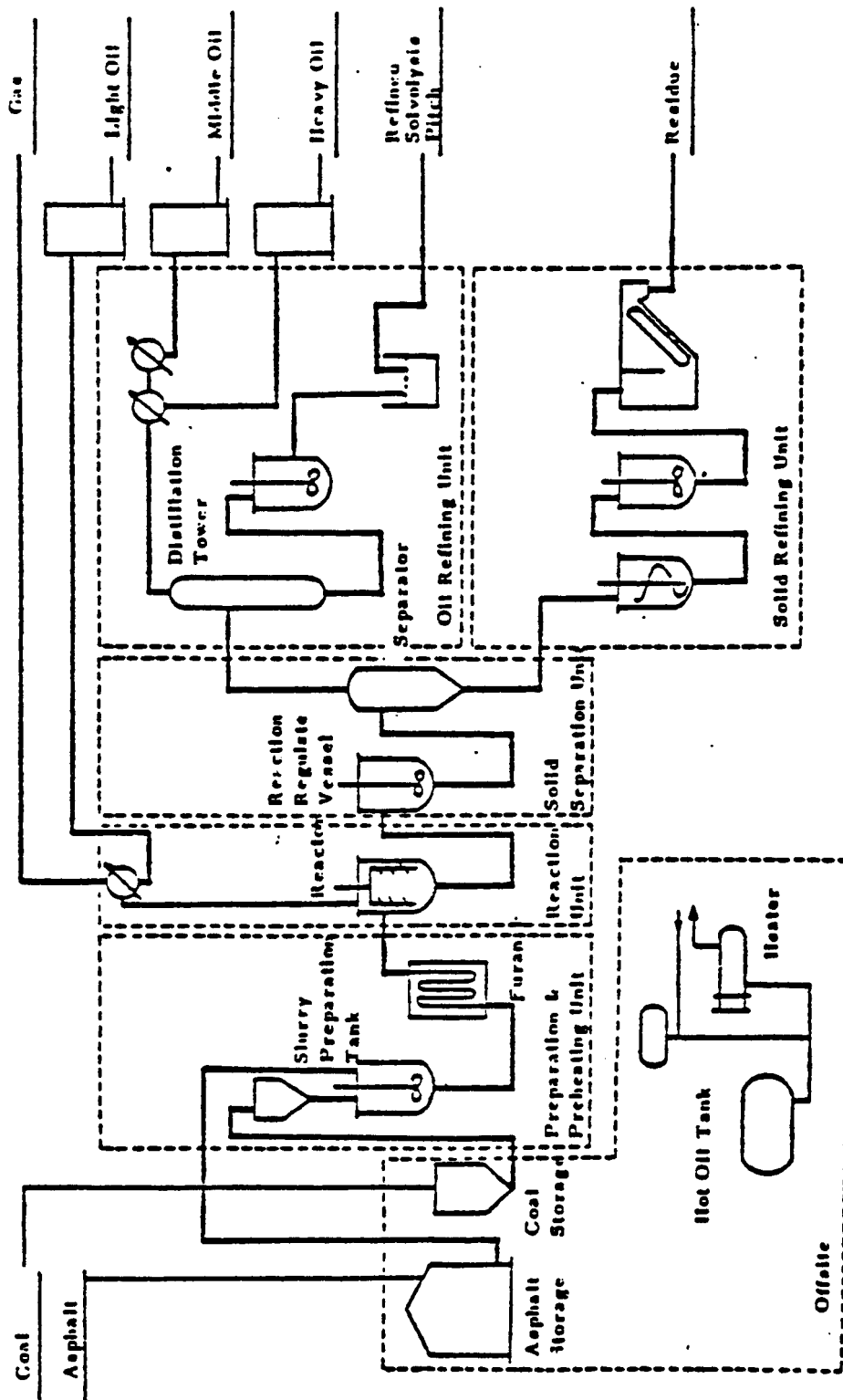


Figure 1. SOLVOLYSIS COAL LIQUEFACTION PROCESS DEMONSTRATION UNIT
(1 tonne/d)

Table 1 Part 1 TECHNOLOGY FACT SHEET: Mitsubishi Heavy Industries Solvolysis Coal Liquefaction Process

NEW TECHNOLOGY (PROCESS DESCRIPTION AND GOALS): This solvolysis process is aimed at the production of a clean liquid fuel from any type of coal. The Mitsubishi Heavy Industries, Ltd. (MHI) design uses hydrogenation of refined solvolysis pitch which is produced by the solvolysis reaction of coal, asphalt, and recycled pitch to produce a solid fuel and light oil. This process is particularly useful in upgrading low-quality coals to a premium liquid and solid fuel.

RELATIONSHIP TO PRIOR TECHNOLOGY (INCLUDING STATE OF DEVELOPMENT OF PRIOR TECHNOLOGY): None

CHARACTERISTICS OF THE TECHNOLOGY:

<u>CHARACTERISTICS</u>	<u>NEW TECHNOLOGY</u>	<u>PRIOR TECHNOLOGY</u>
PRIMARY OUTPUT (DESIGN CASE).....	Coal liquids, solid fuels	
TYPE OF PROCESS.....	solvolysis	
FEEDSTOCK REQUIREMENTS.....	all types of coal (30-100 mesh)	
OVERALL THERMAL EFFICIENCY (INCLUDING BY-PRODUCTS)..		
CARBON CONVERSION EFFICIENCY.....		
OPERATING TEMPERATURE.....	380-450°C	
OPERATING PRESSURE.....	1 atm	
BY-PRODUCTS.....	solid fuel	

Table 1 Part 2 TECHNOLOGY FACT SHEET: Mitsubishi Heavy Industries Solvolysis Coal Liquefaction Process

STATUS OF DEVELOPMENT:

OPERATING FACILITIES - MHI operates a 1 tonne/day PDU in Nagasaki since 1978.

MAJOR FUNDING AGENCY MITI

ANNUAL LEVEL OF FUNDING -

TECHNICAL PROBLEMS: Solid deposits, foaming, pumping, of slurry problems, plugging of transfer lines, and unsatisfactory solid/refined pitch separation.

OTHER FACTORS AFFECTING OVERALL FEASIBILITY: Development of hydroprocessing technology to upgrade oil production.

Process Goals

The major R&D work on the solvolysis coal liquefaction process is being conducted by three Japanese organizations: Industrial Research Institute of Kyushu, Kyushu University, and Mitsubishi Heavy Industries (MHI). The former two organizations have been concerned with fundamental studies pertaining to solvolysis. MHI has been involved extensively in different areas of coal liquefaction process development, including those under joint projects with the United States and Australia. In the solvolysis program, MHI is responsible for design, construction, and operation of the PDU and pilot plants.

Mitsubishi's original process converted coal to oil matter by using heavy oil as a solvent and heating the slurry under normal pressure at 380° to 420°C. Ash and other impurities are separated to produce solvolysis pitch, which is then separated into coke for gasification and oil/pitch for hydrogenation. But now Mitsubishi has revealed plans to develop a new type of solvolysis that derives both solvent and hydrogen from coal. While regular solvolysis works only on bituminous coal, the new method reportedly can liquefy brown and subbituminous coal. MHI has brought in three other members of the Mitsubishi group to help advance the new method.

Relationship to Prior Technology

The solvolysis coal liquefaction process is one of the three major processes (hydrogenation) being developed under the Japanese Sunshine Project, and intensive R&D activities have been in progress since the start of the project in April 1974. The process has been undergoing modification in process conditions as well as the feed and product composition over the years.

Operating Facilities

A 1 tonne/day PDU is being operated by Mitsubishi in Nagasaki, Japan. The location of a proposed 40 tonne/day pilot plant has not been published.

Major Funding Agency

This research is being partially funded under the Sunshine Project of Japan through MITI.

Technical Problems

A 1-tonne/day process demonstration unit (PDU) has been in operation since the end of FY 1977 and has experienced several equipment and operational

problems: a) solids deposit in the slurry preparation tank, b) foaming in the reactor, c) irregular pumping rates of the two plunger pumps (presumably for transporting coal feed slurry to the preheater), d) occasional plugging of transfer lines and valves, especially after a brief pause in operation, and e) unsatisfactory performance of the solid/refined pitch separator (antisolvent type). Some of these problems have been alleviated by design modification of the equipment and piping. For example, problem (a) was overcome by providing a centrifugal recirculation pump. The foaming problem was lessened dramatically by installing a recirculation loop around the preheater and by adopting a lateral, biaxial multi-disk mixer as the reactor. The performance of the plunger pump improved after the diameter of the check-valve ball for the pump was changed (increased?) and erosion-resistant material was used for the ball.

Development of hydroprocessing technologies for solvolysis pitch had been rather limited. It appears that no PDU studies on hydroprocessing have been carried out thus far. The only published study on this subject uses a laboratory-scale fixed-bed cocurrent upflow reactor for desulfurization and denitrogenation of the solvolysis pitch. Scale-up of the 1 tonne/day PDU to a 40 tonne/day pilot plant is being studied.

Capital Costs

The capital cost estimates for this process have not been published.

JAPANESE COAL GASIFICATION PROCESS

35. Mitsui M-Gas Coal Gasification Process

The Mitsui M-Gas process is understood to handle a wide range of feedstocks down to vacuum residuum. It consists of two fluidized beds that operate continuously without the need of oxygen. The fluidized beds consist of particles of a special catalyst containing alkaline earth metal. The feedstock is catalytically gasified with steam in one fluidized bed, and the catalyst is regenerated by combustion of deposited coke and added fuel in the other. The heat to support gasification is provided by the circulation of catalyst between the two beds. A pilot plant was constructed at the Kitakyusha (Mitsui Kogan Coking Co., Ltd.) site by Mitsui Engineering and Shipbuilding Co. under a joint development program. Provision for on-line withdrawal and addition of catalyst was made to maintain activity in the gasifier and compensate for attrition. The pilot plant operation is not fully integrated but is aimed at providing data to confirm and modify certain aspects of the process or for use in designing a development plant. The plant is designed for 1 atmosphere pressure but a 5 bars pressure is envisaged for a commercial design. It is possible that the product gas would be capable of upgrading to SNG either by a methanation route or to a hydrogenation gas for an oil hydrogenation unit.

No additional information has been published on this process.

JAPANESE COAL GASIFICATION PROCESS

36. Coal Mining Research Center Combined Cycle Coal Gasification Process

Coal Mining Research Center, a Tokyo-based nonprofit organization, will soon start a three-year low-calorie coal gasification program to test combined cycle power generation at a recently completed pilot plant at Yubari, Hokkaido. The \$10-million plant, completed by Mitsubishi Heavy Industries (Tokyo), is designed to gasify 40 metric tons/day of low-calorie (6200 to 6300 kilocalories/kilogram) coal produced in Kushiro, Hokkaido. Estimated to be one of the world's largest such facilities, the pressurized two-section fluidized-bed plant is designed to produce syngas with 1,200 kilocalories/cubic meter value. The process features reaction of coal's carbon with air (preheated to 300 to 350°C) and steam to convert carbon and water into hydrogen and carbon monoxide. The plant's upper section is for gasification at about 900°C. The char (coke) drops to the lower section for reaction again with air and steam at about 1000°C.

The center's plant construction and operation are financed by the Ministry of International Trade and Industry for the government's "sunshine" project. It is aimed at developing knowhow for coal gasification/liquefaction (along with hydrogen, solar energy and geothermal energy development). Once the 40 metric tons/day plant generates promising results, the center will try design of a larger unit capable of gasifying about 1000 metric tons/day of Kushiro coals. The center originally considered but abandoned the dry distillation (carbonization) technique, involving lower temperatures up to 500°C. It judged that it should be easier to handle nitrogen than to treat gas containing tar. The plant cost includes related facilities, such as desulfurization and coal ash treatment facilities.

No other information has been published on this process.

JAPANESE COAL GASIFICATION PROCESS

37. Hitachi Coal/Oil Mixture Gasification ProcessProcess Description

In this process coal is crushed to 0.1 to 1.0 mm and mixed with fuel oil to form a slurry. In the pilot plant 4 metric tons of coal are mixed with 8 metric tons of fuel oil. Dubbed a hybrid gasification process, this design produces a high calorie gas (9,000 kilocalories/m³) from steam, coal, and fuel oil. This slurry is then heated to 200°C and injected into the gasifier at 29 atm. Oxygen and steam are also fed to the gasifier as gasification agents.

The 11,300 m³/day synthesis gas output pilot plant gasifier is of a entrained-bed design, which consists of two sections in one reactor shell. The coal slurry enters at the midpoint of the gasifier shell, between the two sections, where it strikes a rotating dispersion grate. The slurry is quickly vaporized at this point, and the coal is thermally decomposed at about 700°C to form gas and char. The gases flow upward into the top section where they undergo further thermal decomposition. The char formed at the mid-section drops down to the bottom section where it is gasified with the oxygen and steam. The hot gases generated in the bottom section pass upward through the injection zone and top section. Temperatures in the bottom gasification zone are about 950°C. Ash is removed from the bottom of the gasifier in a water cooled quench zone. Tars formed in the process that exit the top zone are removed and recycled to the slurry feed section. Overall process efficiency is estimated to be 74%.

Process Goals

Electric Power Development Co. (EPDC), a government-controlled electricity wholesale company, completed construction of a \$23.6-million pilot coal gasification plant at Iwaki City, Northern Honshu, in early 1982. EPDC will test the method for three years, starting in April, to confirm the process feasibility and economics of utilizing the process for supplying town gas and industrial fuel. EPDC will try to raise the ratio of coal feedstock — a key to improving the process economics. Future plans call for the construction of a 40 ton/day demonstration plant.

Relationship to Prior Technology

This development work started as an offshot of the U.S. Synthane process but has evolved into a unique Japanese coal gasification process.

Operating Facility

A 4 metric ton/day pilot plant was completed in March 1982 at Iwaki City, Northern Honshu.

Major Funding Agency

The gasification research started in fiscal 1974, when the Ministry of International Trade and Industry (MITI), which controls EPDC, awarded Hitachi a contract to investigate the elements of such a method. EPDC joined Hitachi in fiscal 1977 for the development efforts, which are being supervised by the New Energy Development Organization (NEDO) for MITI. This is one of the Sunshine projects.

Technical Problems

Reducing the oil-to-coal slurry ratio may be technically difficult. This will probably be required to make the process more economical.

Capital Costs

The pilot plant cost \$23.6 million to construct; no cost estimates have been published for a larger facility.

SOVIET COAL CONVERSION TECHNOLOGY

38. Power Engineering High-Speed Pyrolysis ProcessProcess Description

The process being developed by the Soviet Ministry of Coal and the Krzhizhanovskiy Power Engineering Institute (ENIN) of the Ministry of Power and Electrification is based on the high speed pyrolysis of brown coal to produce a semicoke solid. This process is being developed to produce a semicoke solid which can then be transported by rail or pipeline from the central Siberian coal fields of the Kansk-Achinsk basin to the western Soviet Union. Gaseous and liquid fuels are also obtained from this process.

The ENIN has tested the high-speed pyrolysis process in both a fixed-bed and fluidized bed reactor. The fixed bed process was studied for a number of years in pilot projects at a power plant in Kalinin and at the Elektrostal plant in Krasnoyarsk. The ETKh-175 demonstration facility currently under construction at TETs-2 in Krasnoyarsk will have a capacity of 1.2 million tons (coal)/yr. In 1976, the Soviets indicated a desire to increase the capacity to 24 million tons/yr gradually through several stages. Problems in equipment and construction priorities have stalled the ETKh-175 project temporarily. Construction of this facility was to be completed in 1981 however, it is still not completed. Reportedly, the 25th Congress of the Capital Committee, CPSU proposed to build three energy-technology combines at the K-A Fuel-Energy Complex, each able to process 24 to 50 million tons (coal)/yr by high-speed pyrolysis. Construction of these combines cannot be started, however, until the results of the ETKh-175 test program become available. Only the coking facility is being considered at this time.

A fluidized-bed process for the high-speed pyrolysis of K-A coal to produce semicoke was pilot tested successfully by ENIN at Sverdlovsk by 1977. A proposed design for a demonstration-scale facility to process 2.1 million tons (coal)/yr by this method has been drawn up. There is no evidence that construction has begun.

Process Goals

The vast quantity and location of the Kansk-Achinsk brown coal deposits make them an attractive source of energy. However, their high moisture content (up to 35%), low heating value, (3,300 kilocalories/kilogram),

variable physical and chemical characteristics, and tendencies toward spontaneous combustion make it a difficult coal to handle, transport, and burn directly. For this reason the Soviets have been developing technologies to make this coal more suitable for direct combustion applications and for transportation.

Relationship to Prior Technologies

None.

Operating Facility

A 1.2 million ton/yr fixed-bed facility will be tested in Krasnoyarsk. Testing of the fluidized-bed high-speed pyrolysis process variant was completed in 1977 at Sverdlovsk.

Major Funding Agencies

The Soviet government.

Technical Problems

None found in the literature.

Capital Costs

Based on published information the high speed pyrolysis process is approximately 10% more expensive than the Soviet developed "thermocoal" process which simply reduces the coal's moisture. Estimated technical and economic data for the "thermocoal" production are presented in Table 1.

Table 1. ECONOMIC AND TECHNICAL DATA FOR "THERMOCOAL" PROCESS

Annual raw coal production capacity	7,500,000 tons
Consumption of raw coal per ton of final product	1.96 tons
Total capital investment (1 Rbl = U.S. \$1.30)	26,003,000 Rbl.

SOVIET COAL CONVERSION PROCESS

39. Institute of Fossil Fuels Hydrogenation Coal Liquefaction ProcessProcess Description

The Institute of Fossil Fuels in Moscow has developed a direct hydrogenation liquefaction process based on the Bergius-Piers process. In the Soviet process coal is crushed to a fine powder, dried in a heated vacuum and mixed with heavy oil and a catalyst. This slurry is then blended with hydrogen gas prior to entering the reaction vessel. The main reaction occurs at 100 atmospheres and at 400°C. The molybdenum and iron catalyzed reaction produces a mix of products consisting chiefly of three ranges of distillates and a residual. Vacuum distillation is used to produce distillates with a boiling range of <200°, 200 - 399°C, and 300 - 500°C. Part of the distillate production is recycled to the slurry preparation section where it is used as a hydrogen donor. The slurry is blended to a 30 wt % consistency with the recycled oil.

Preliminary results indicate that 90 to 93 percent of the coal is liquefied in this process. Approximately 50.4 wt % of the liquid product is heavy residual, 35.2 wt % is distillate in the 300 to 500°C boiling range and the remainder has a boiling point of <500°C. The final products after further processing include low-sulfur boiler fuel, high-octane gasoline, light diesel fuel, and other chemical products.

Process Goals

The goal of this research, which has been conducted since 1968, is to develop a direct coal liquefaction process based on the Bergius-Piers concept which operates at milder reaction conditions, thus reducing process costs. Major goals have included the successful liquefaction of Kansk-Achinsk lignite coals and the development of novel catalysts.

Relationship to Prior Technologies

The Soviet development effort is based on the Bergius-Piers process which was developed prior to World War II in Germany. The Institute of Fossil Fuels research closely parallels similar efforts currently underway in West Germany to improve the old Bergius-Piers process.

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Operating Facilities

Bench-scale experiments have been conducted in Moscow since 1968 which converted four kilograms of coal to one kilogram of oil. Larger scale testing is being planned for a 10 tonne/day pilot plant which is located at the Belkovskaya mine in the Moscow coal basin. Construction of this plant began in late 1980. Completion of this plant is scheduled for 1983. Plans simultaneously call for the construction of a 75 tonne/day liquefaction pilot plant at an open pit mine in Siberia's Kansk-Achinsk coal basin.

Major Funding Agencies

The Soviet government.

Technical Problems

The only cost numbers published for this process indicate that the reduced hypothetical cost of electric power generation using the liquefied coal products would cost 2.48 to 3.20 kop/kWh. No detailed costs have been published on actual plant costs. Costs are projected to be greater than the HS Pyrolysis process.

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