SLIDE 4

H-COAL® PROCESS FEATURES

- HIGH YIELDS OF DISTILLED, LOW SULFUR LIQUIDS FROM BITUMINOUS AND SUB-BITUMINOUS COALS AND LIGNITES -TYPICALLY 2.8 TO 3.5 BARRELS/TON OF DRY COAL

- CATALYTIC EBULLATED BED COMBINES COAL LIQUEFACTION, SOLVENT HYDROGENATION AND PRODUCT UPGRADING IN A SINGLE REACTOR

- HYDROCLONES USED TO RECOVER LOW SOLIDS RESIDUUM OIL STREAM TO SLURRY COAL FEED

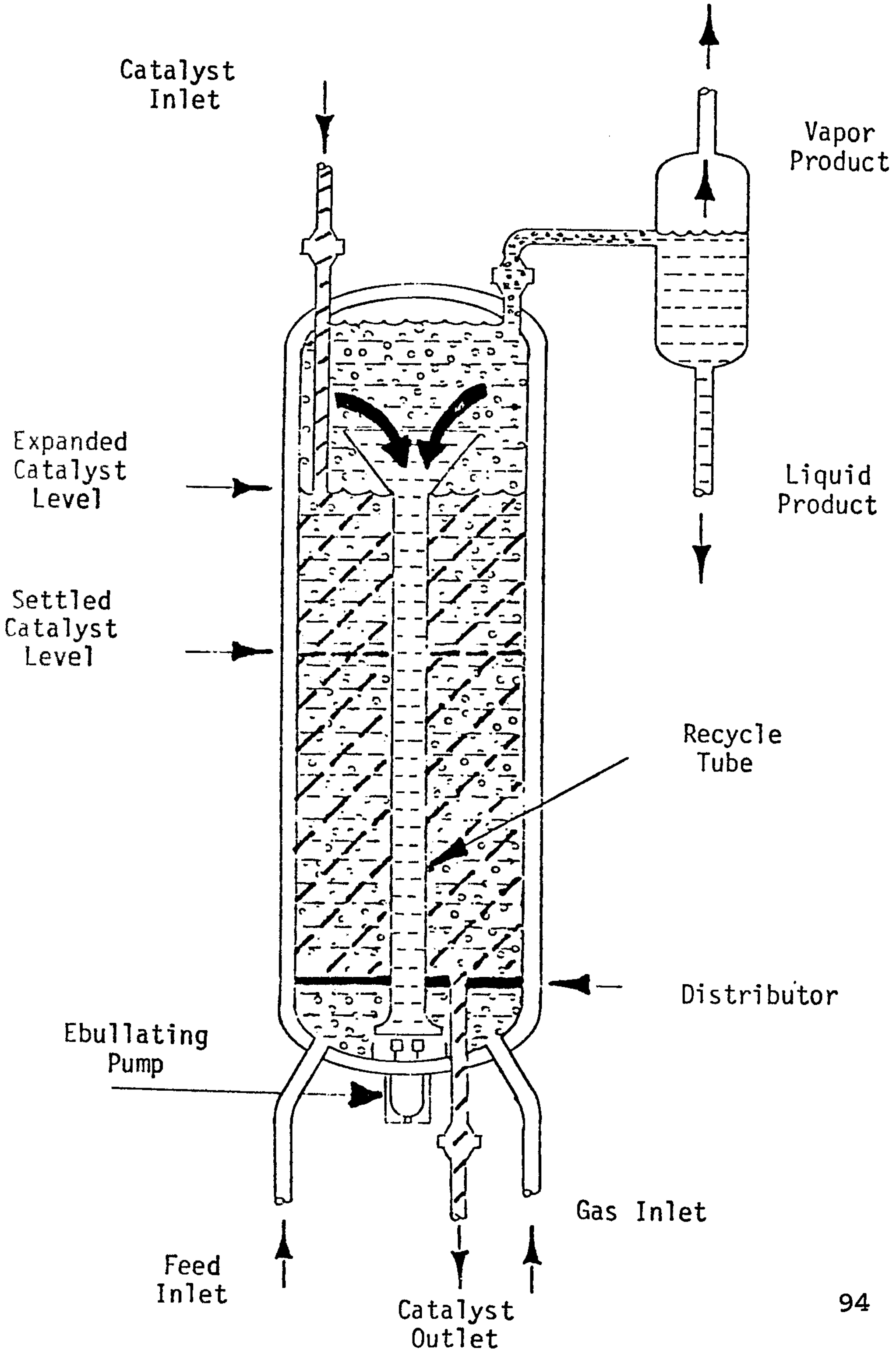
- LIQUEFACTION EFFLUENT SEPARATED BY DISTILLATION

- PROCESS HYDROGEN REQUIREMENTS CAN BE MET BY PARTIAL OXIDATION OF LIQUEFACTION BOTTOMS AND/OR STEAM REFORMING OF LIGHT HYDROCARBONS

- SOME FURTHER PRODUCT UPGRADING IS REQUIRED TO PRODUCE HIGH QUALITY TRANSPORTATION FUELS SUCH AS GASOLINE, DIESEL, OR JET FUEL.

SLIDE 5

EBULLATED BED REACTOR



SLIDE 6

COALS RUN IN H-COAL® PROCESS

EASTERN U.S.

ILLINOIS NO. 6
INDIANA NO. 5
KENTUCKY 9/14
KENTUCKY 11
PITTSBURGH SEAM (CONSOL NO. 8)

WESTERN U.S.

WYODAK
UTAH D.
BIG HORN
COLORADO
BLACK MESA

LIGNITES (U.S.)

TEXAS
N. DAKOTA

FOREIGN

AUSTRALIAN BROWN
GERMAN "STEINKOHLE"
OTHERS

SLIDE 7

TYPICAL H-COAL® YIELDS(1)

	ILLINOIS BITUMINOUS (BURNING STAR)		WYOMING SUB-BITUMINOUS (WYODAK)
YIELDS, LBS/100 LBS DRY COAL	SYNCRUDE	FUEL OIL	SYNCRUDE
H_2	(5.3)	(3.4)	(6.2)
H_2O, CO, CO_2	7.1	6.5	20.0
H_2S, NH_3	3.6	2.2	1.6
C_1-C_3	11.2	6.8	12.3
C_4-400°F NAPHTHA	18.7	13.4	25.8
400-975°F FUEL OIL	29.1	20.8	18.6
975°F+ BOTTOMS (INCL. ASH)	35.6	53.7	27.9
	100.0	100.0	100.0

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(1) BASED ON LINED OUT OPERATION IN THE 3 TON/DAY PROCESS DEVELOPMENT UNIT (PDU) AT HYDROCARBON RESEARCH, INC.'S TRENTON LABORATORY.

TYPICAL H-COAL® LIQUID PRODUCT QUALITIES

SLIDE 8

OPERATING MODE	ILLINOIS BITUMINOUS (BURNING STAR) SYNCRUDE	WYOMING SUB-BITUMINOUS (WYODAK) SYNCRUDE
NAPHTHA(IBP/350°F)		
°API	52.3	55.8
C	85.3	84.7
H	13.8	14.0
O	0.56	1.25
N	0.24	0.10
S	0.07	0.02
MID-DISTILLATE(350/600°F)		
°API	18.5	27.8
C	88.4	87.0
H	10.1	11.4
O	1.0	1.3
N	0.47	0.22
S	0.08	0.03
DISTILLATE BOILER FUEL(500/800°F)		
°API	4.9	10.9
C	89.4	88.4
H	8.6	9.4
O	1.3	1.7
N	0.63	0.46
S	0.08	0.03

PROBABLE DISPOSITION OF H-COAL® PRODUCTS

CUT	UPGRADING REQUIRED(1)	DISPOSITION(2)	COMMENTS
NAPHTHA C5/350°F	NAPHTHA HYDROTREATING, CATALYTIC REFORMING	MOTOR GASOLINE, AROMATICS PRODUCTION	SEVERE HYDROTREATING CONDITIONS, EXCELLENT REFORMER FEEDSTOCKS (88 LV% C5 PLUS, 3.4 WT% H2 YIELDS @ 103 RON)
MID-DISTILLATE 350/650°F	MILD HYDROFINISHING TO SEVERE HYDROTREATING (FOR DIESEL)	HOME HEATING OIL, DIESEL FUEL, TURBINE FUEL	NITROGEN REMOVAL FOR STABILITY, CETANE INDEX IMPROVEMENT, GRAVITY IMPROVEMENT
FUEL OIL 350°F+ OR 650°F+	NONE TO MILD HYDROTREATING	LARGE STATIONARY COMBUSTION FUEL (STEAM BOILERS)	UPGRADING REQUIREMENTS, IF ANY, DEPEND ON CUSTOMER REQUIREMENTS

---

(1) UPGRADING STUDIES HAVE BEEN CARRIED OUT ON H-COAL PRODUCTS BY HRI, UOP, MOBIL, CHEVRON, EXXON AND OTHERS. STUDIES INCLUDE HYDROTREATING, HYDROCRACKING, CATALYTIC CRACKING, COKING, CATALYTIC REFORMING, PARTIAL OXIDATION, AND STORAGE STABILITY.

(2) END USE TESTING INCLUDES DEMONSTRATION OF H-COAL PRODUCTS IN GASOLINE, HOME HEATING OIL, NO. 2 FUEL OIL AND TURBINE FUEL.

SLIDE 10

H-COAL COMMERCIAL PLANT PROJECT

LOCATION BRECKINRIDGE, KENTUCKY

COAL FEED

RUN-OF-MINE ILLINOIS NO. 6	23,000 T/D

PRELIMINARY PRODUCT SLATE

MOGAS BLEND STOCKS (C5/360°F)(1)	15,300 B/D
DISTILLATE FUEL OIL(360/950°F)	28,000 B/D
BUTANE	3,300 B/D
PROPANE	3,500 B/D
C3/950°F LIQUID PRODUCTS	50,100 B/D

BY PRODUCTS

SULFUR	570 T/D
AMMONIA	120 T/D
SNG	30 MSCF/D

---

(1) ALTERNATE PETROCHEMICAL OUTLET TO BE EVALUATED

SASOL - TONS TO BARRELS - NOW!

William A. Samuel, P.E.

SASOL - TONS TO BARRELS - NOW!

William A. Samuel, P. E.
Fluor Engineers and Constructors, Inc.
Irvine, California 92730
United States of America

Introduction

Three years ago at the Fourth COGLAC, on the panel, "Which Conversion Plants Can Be Built Now", I emphasized that Fluor was then building Sasol Two in South Africa. This paper is a report on the current status there, with emphasis on operating results. Sasol Two was constructed in two phases:

Phase One

This represents about 80 percent of the total construction effort. It includes 50 percent of all the units consisting of two or more trains, plus all off-site facilities and utilities as well as common facilities. The construction completion of the first Phase One units was achieved in late 1979, and unit completions continued through the early part of 1980. Commissioning of the Phase One units progressed in accordance with their mechanical completion, and most of the Phase One units are now in operation.

Phase Two

Construction of Phase Two is now also nearly complete, and commissioning of these units was started recently. Full production of the complex should be achieved during 1981.

Sasol Three

Sasol Three is essentially a duplicate of Sasol Two. It's construction is now moving on a schedule to bring it into start-up in late 1982, even though its authorization came as recently as early 1979. The accelerated schedule is a result of our ability to utilize the existing engineering as done for Sasol Two and to employ our world-wide purchasing capabilities to get new quotes for equipment duplicating Sasol Two for delivery on the Sasol Three program.

Overall Flow Scheme

Each plant is designed to process about 40,000 tons of coal per day. The attached simplified flow sheet entitled Sasol Synthol Process illustrates the overall process scheme for both Sasol Two and Sasol Three.[1] In the following section we discuss each of these units in some detail. Here we give a general description. Coal passes from the mine to the coal preparation units and then to the power stations and to the Lurgi gasification system. Air enters the air separation system to supply the oxygen which passes to the Lurgi apparatus along with most of the steam from the power station. In the gasifiers, the reactions occur to produce a raw gas which includes tars and some aqueous chemicals. These are separated in the quench system, with the tars going to further refining and the aqueous gas liquor going to units to recover those chemicals. The raw gas proceeds to a Lurgi Rectisol system for removal of the acid gas constituents to produce a pure gas suitable for Fischer-Tropsch reaction. This material goes to the Synthol fluid bed system along with hydrogen and carbon monoxide produced by reforming the methane produced in the gasification reactor. The reaction products from the Synthol system go to a product recovery unit from which the ethylene is separated as a salable chemical. The methane goes to the methane reforming unit, and the other products are sent to a product refinery from which are produced the LPG, gasoline, diesel, fuel oil, jet fuel, and other chemicals as noted on the flow sheet.

Unit by Unit Comments

Coal Mines.

Sasol Two and Three will be supplied by the Bosjesspruit Colliery, which is situated in the Highveld coal field. Four shaft systems are planned with combined coal production capabilities of

30 million tons per year. This will supply the design needs of Sasol Two and Sasol Three plants of 27.5 million tons per year. The mining methods are highly mechanized and include long wall and continuous mining. Mine depth varies between 390 feet and 590 feet with an average of 490 feet. The average seam height is about 10 feet. Working reserves are at least 70 years, with the time being not so much determined by the available coal as by the distance between the ultimate coal front and the plant. The current production rate amounts to between 15,000 to 20,000 tons per day. This will be increased beyond this figure as Phase Two is commissioned.

Power Stations

Up to 30 percent of the total coal will go to the power stations which produce steam for process use as well as power generation. Sasol Two imports most of its power from outside, but Sasol Three will be almost in balance as far as electric power is concerned.

Air Separation System

The biggest energy consumers are the air separation units. Sasol Two has an installed air separation capacity of 13,600 tons per day from 6 air separation units. These are the largest such units in the world.[2] The air compressors of each unit require 28.5 megawatts. The oxygen, being required at 35 atmospheres, is compressed in 6 oxygen compressors, each consuming 12.5 megawatts.

Gasifiers

The battery of 36 Lurgi Mark IV gasifiers require 8,000 tons per day of oxygen, along with 28,000 tons per day of steam and 28,000 tons per day of coal. Normally, about 30 gasifiers will be in operation, with the rest on turn-around or stand-by. These gasifiers operate at approximately 30 atmospheres pressure. The first gasifiers were started in late October 1979, and today all the Phase One gasifiers have been tested, and 16 units are more or less continuously in operation. Guaranteed capacity on the individual units has already been achieved. Gas composition is in accordance to design. The capacity of the plant is still being evaluated, but no problem is expected in achieving design production.

Raw Gas Quench

The raw gas leaves the gasifiers hot, and quenching it produces a phase of tars and oils and a phase of aqueous materials. The tar/oil streams are distilled and hydrogenated to yield gasoline and diesel in the tar-refining section, as shown on the flow diagram.

The current production of these products is in line with the present gasification load. The design load, however, was already achieved on the tar distillation plant and the oil hydrogenation plant by employing feedstock accumulated earlier.

The aqueous phase, containing ammonia and phenols as main components, is fed to a phenosolvan unit where the phenols are extracted using di-isopropyl ether. Ammonia is stripped and purified to yield anhydrous ammonia with a purity equal to, or better than, that of synthetic ammonia.

The phenosolvan with the ammonia recovery unit has completed its guarantee run earlier this year. A throughput of ten percent in excess of design was reached. The stripper liquor from phenosolvan is fed to an effluent treatment plant. For United States installation, the tar and oils could be recycled back to the gasifiers to produce more syngas. If desired, the ammonia and phenols could be incinerated in the vapor phase so that no phenosolvan unit would be required.

Raw Gas Clean-up

After cooling, the gasification raw gas is fed to a Lurgi-designed Rectisol gas purification system. Here the hydrogen sulfide and carbon dioxide are removed from the main stream, along with any residual oils and other impurities.

The resulting gas has the following composition: 86 percent hydrogen plus carbon monoxide; 13 percent methane; 1 percent nitrogen plus carbon dioxide.

The plant removes the hydrogen sulfide and organic sulfur to levels of less than 0.1 ppm, well below guarantee. The carbon dioxide removal is achieved to less than one volume percent against a guarantee of 1.5 volume percent maximum.

The carbon dioxide of the gas from the Rectisol, which includes the hydrogen sulfide, is fed through a Stretford unit where the hydrogen sulfide is converted into sulfur. The main CO_2 stream is then released to the atmosphere, at which point it contains less than 50 parts per million sulfur as H_2S. The Stretford unit, however, is still under construction, and no operating experience is available as yet.

Synthol Fischer-Tropsch Units

The purified synthesis gas from the Rectisol plant, along with the CO and H_2 from the methane reformer, next pass to the Synthol units - the real heart of a Sasol complex. The Synthol reactors employ a circulating fluid bed with a promoted iron catalyst, as shown in the accompanying simplified schematic diagram. The pressures and temperatures are moderate, and no exotic materials of construction are required. The Fischer-Tropsch reaction always produces a range of hydrocarbons, along with some oxygenated products. A typical mass distribution of such products has been reported as shown in the attached figure.[3]

By proper selection of reaction conditions, one can influence this distribution to some extent. At Sasol Two and Three, the emphasis is on transport fuels. Thus far all four Phase One Synthol loops have been in operation. No problems were experienced in achieving design feed rate, conversion, and product selectivity.

Note that the contaminant-free feedstock gas from the Rectisol system insures clean products: No sulfur, cyanides, etc.

Product Recovery

The synthesis product is first recovered by air and water cooling of the reactor tail gas. In this manner most of the synthetic oil down to the C_5/C_6 fraction is recovered.

The residual gas stream is then fed to a low temperature separation unit where first the residual C_3's and heavier hydrocarbons are recovered. Then the ethylene and ethane are recovered, and, finally, the hydrogen and methane streams are separated into hydrogen-rich and methane-rich gas streams. The hydrogen-rich stream is returned directly to the synthesis system, whereas the methane-rich stream is reformed using steam and oxygen to produce hydrogen and carbon monoxide for recycle to the synthesis complex.

The low-temperature plant has been in operation at loads commensurate with the available tail gas from the synthesis unit; that is, 50 percent of design. No specific problems have been experienced during this operation.

Construction of the methane reforming unit was recently completed, and commissioning is in progress. The intake of methane-rich gas is scheduled for July.

The Product Refinery

The product refinery utilizes conventional refinery unit processes and unit operations to produce the mixture of fuel products: LPG, gasoline, diesel, fuel oil, and jet fuel. Other units process and refine the various chemical products.

Thus far, only parts of this complex have been in operation, and it is too early at this stage to pass comment on the performance of the individual units. As already mentioned above, this area consists of conventional refinery units, so no problems are expected.

Points on the Fluor-Sasol Agreement

Fluor has the exclusive license in the United States to market and construct complete Sasol-type Fischer-Tropsch plants. The Sasol Synthol license is, however, available only to customers of arrangement between the parties. This arrangement permits Sasol to utilize Fluor's strong marketing abilities to take advantage of Sasol's proven technology in conjunction with Fluor's demonstrated engineering and construction leadership. Because of the established working relationship, Fluor and Sasol have the necessary mutual confidence to insure a successful venture.

In Summary

On most of the front end units the ability to produce at or in excess of design has already been demonstrated. On all the other units, no specific problems have been experienced which would prevent them from achieving design capacity. The complex in total suffers from the normal problems related to a grass roots plant in that the reliability of services often leaves something to be desired. This can cause unscheduled shut-downs of the complex and delays in bringing new units on line. It is expected, however, that it will still be possible to adhere to the original commissioning and production schedules.

So, in South Africa at Sasol, they are converting tons of coal to barrels of fuel - now.

References

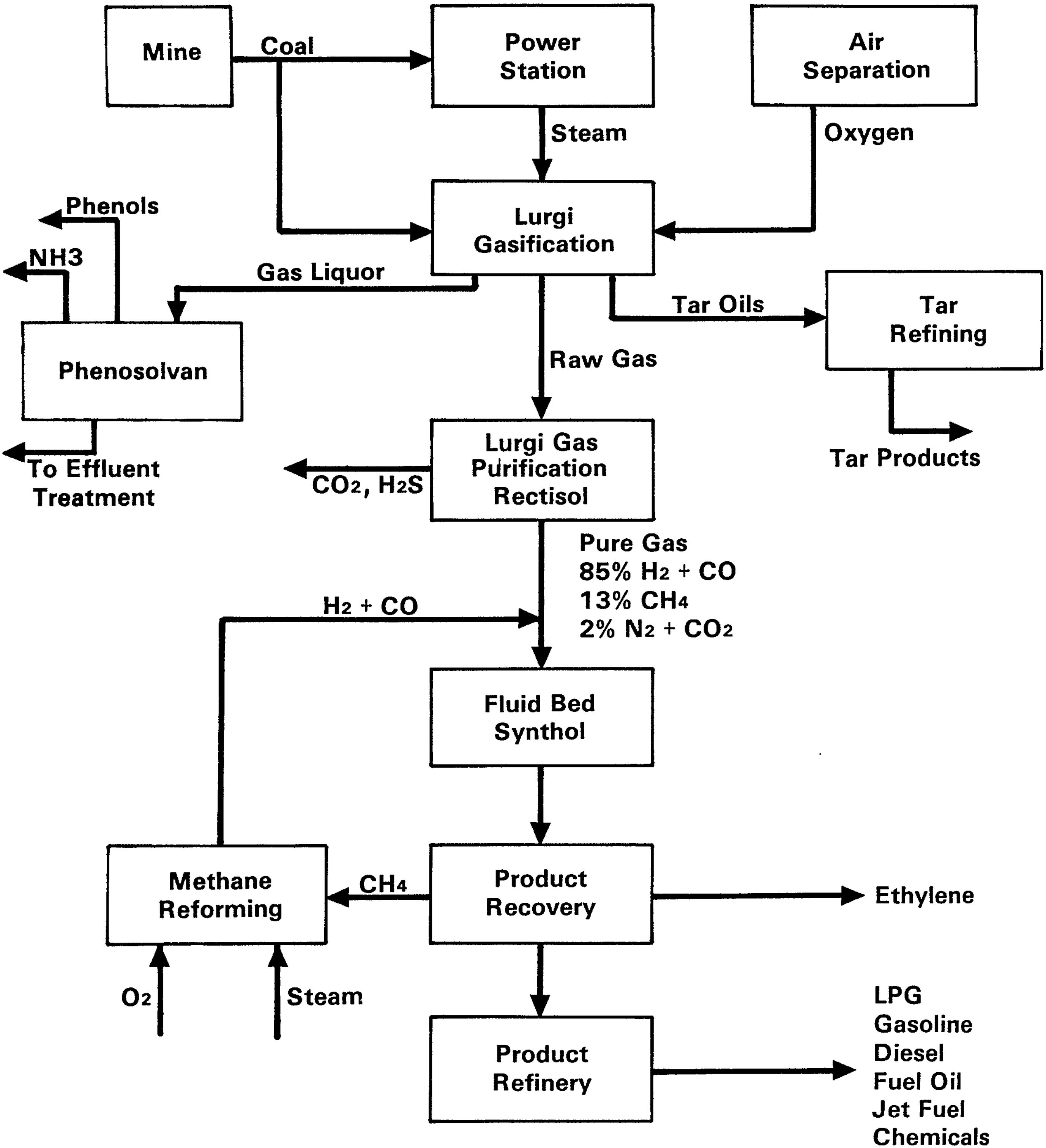
1. The Sasol Process; Sasol Public Relations, Sasolburg, South Africa. January 1980, p. 8.

2. Ibid, p. 3.

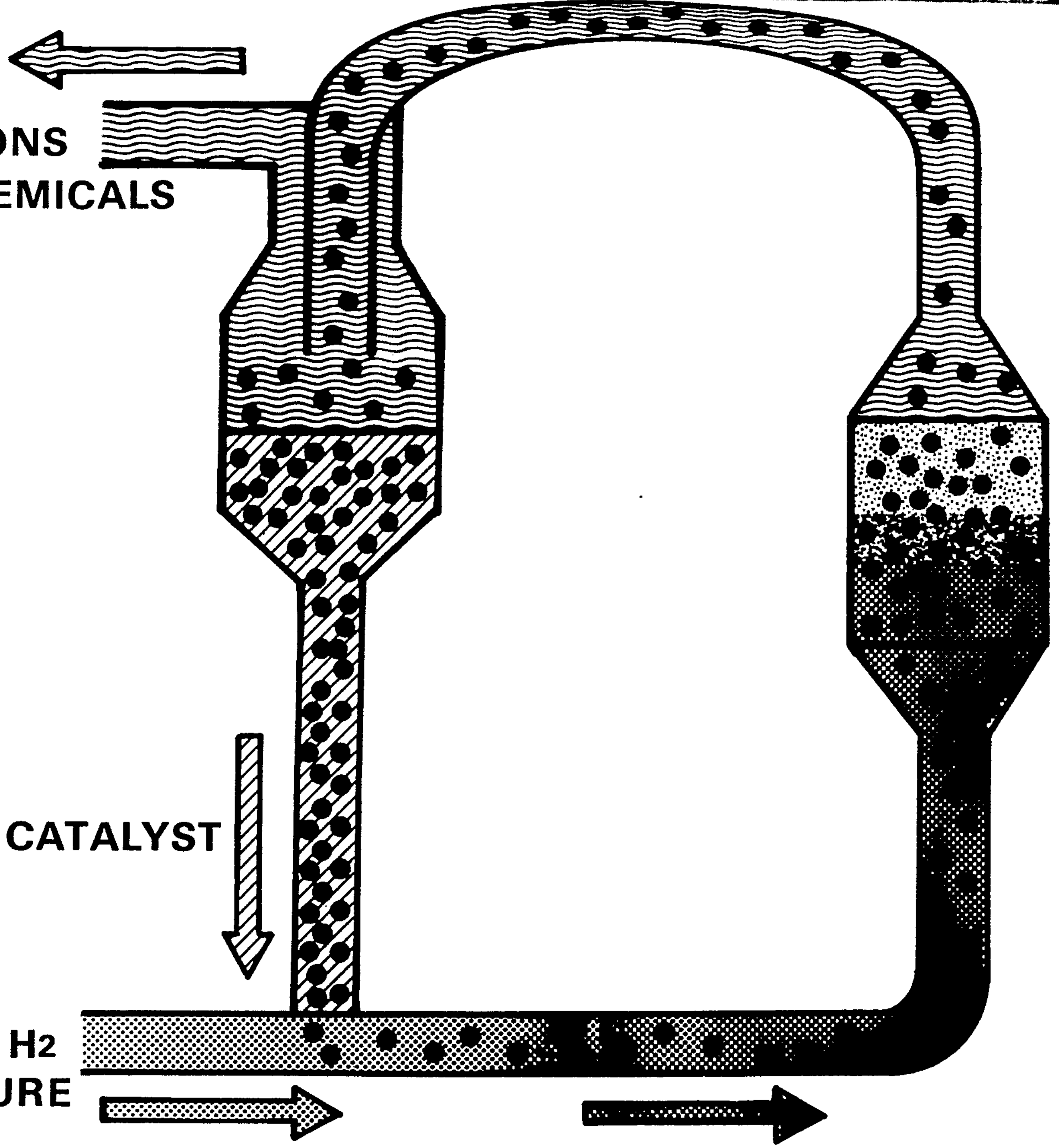
3. Hoogendoorn, J. C. and Jackson, S. B.; Sasol Projects in South Africa. Presented at "Coal Technology '79," Houston, Texas, U.S.A.

All operating data references are from personal communication from SASOL, dated June 13, 1980, which is gratefully acknowledged.

SASOL SYNTHOL PROCESS
SIMPLIFIED FLOWSHEET SASOL 2-3



FLUOR



PRODUCTS

- HYDROCARBONS
- AQUEOUS CHEMICALS
- GASES

REACTOR

CATALYST

CO + H_2
MIXTURE

SASOL SYNTHOL LOOP

FLUOR

TYPICAL SYNTHOL PRODUCT SELECTIVITY
(MASS BASIS)

	PERCENT
Methane	11.0
Ethane/Ethylene	7.5
Propane/Propylene	13.0
Butanes/Butylenes	11.0
C_5 To 375 F Fraction	37.0
375F To 750F Fraction	11.0
750F To 970F Fraction	3.0
Heavier Than 970F Fraction	0.5
Chemicals	6.0
	100.0

A COMPARISON OF DIFFERENT FISCHER-TROPSCH PROCESSES

W. -D. Deckwer

A COMPARISON OF DIFFERENT FISCHER-TROPSCH PROCESSES

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Introduction

The synthesis of hydrocarbons by the Fischer-Tropsch (FT) process presents an indirect route of coal liquefaction. This process converts synthesis gas produced by coal gasification into saturated and unsaturated hydrocarbons.

The synthesis was discovered in 1925 and within 10 years the first industrial plants went into operation. The process was based on fixed bed reactor technology (normal and middle pressure process on Co and Fe catalysts). In Germany, 9 FT plants produced about 560,000 t/a in 1944.[1,2] This was, however only about 1/6 of that amount produced by direct liquefaction. The world capacity of all FT plants amounted to 1.1 million tons. An increase in the production height was particularly hampered by difficulties with the desulfurization of the syngas and the low performance of the gasifiers.[1] In spite of the constructive advantages of the low pressure process FT transportation fuels were more expensive than those obtained from the high pressure direct liquefaction route.[2] After World War II abundant amounts of cheap oil were available. Therefore, the FT process proved no more profitable and all plants were shut down. At present, it is only SASOL in South Africa which produces hydrocarbons from FT plants using improved fixed bed and entrained fluidized bed technology.

General Aspects

Stoichiometry

From the chemical point of view, the FT synthesis is the hydrogenation of CO which, however, does not yield one definite product but a variety of compounds. The main reactions can be summarized by the subsequent stoichiometric equation.

$$nCO + 2nH_2 \rightarrow -(CH_2)_n- + nH_2O \qquad (1)$$

If the synthesis is carried out on iron catalysts reaction is most easily followed by conversion of water

$$CO + H_2O \rightarrow H_2 + CO_2 \qquad (2)$$

Thus, by combining eqs. (1) and (2) the overall reaction on Fe can be written as

$$2nCO + nH_2 \rightarrow -(CH_2)_n- + nCO_2 \qquad (3)$$

It should be noted that the conversion reaction, i.e. eq. (2), can reduce the hydrogen demand from $H_2/CO = 2$ in reaction (1) to $H_2/CO = 1/2$ in reaction (3). Thus, on Fe catalysts, the use of weak syngases, i.e. with a low hydrogen content, is possible. An important process parameter which characterizes the stoichiometry is the usage ratio. This presents the moles of H_2 needed to convert 1 mol CO. Often the usage ratio is approximately the inlet ratio.[3]

Products

The main products are unbranched paraffins and olefines. The molecular weight ranges from 16, i.e. methane, to above 20,000 depending on the catalyst, the operating conditions and the kind of process. The olefin content of the produced hydrocarbons may vary from 10 to 90%. As by-products aromates and,above all,oxygenates, i.e. alcohols, ketones, acids, and esters, are formed. Under certain conditions and using nitrided fused Fe catalysts, the oxygenates content may be as high as 49% of the C_{3+} fraction.[4] It should be pointed out that, in contrast to direct liquefaction products, FT hydrocarbons are essentially nitrogen and sulfur free.

The major interest in FT synthesis is to manufacture automotive fuels, of course. However, as the straight chain α-olefin content of the overall olefin fraction may be as high as 85% the FT products should also be considered as starting materials for further chemical processing as, for instance, the production of alcohols via oxo-synthesis and manufacturing of tensides by alkylation of aromates.

Selectivity

Various proposals on the mechanism of the hydrocarbon synthesis have appeared in the literature, however, the whole matter is still subjected to active research. It seems obvious nowadays that the chain propagation and transfer step gives a distribution of products which approximately obeys a Schulz-Flory distribution if one does not account for the C_1 and C_2 fraction.[5] Therefore, the product distribution is the broader the higher the mean degree of polymerization. Indeed, this unselectivity is an intrinsic feature of the FT process if carried out on common catalysts and at high conversion levels.

Catalysts

Iron, cobalt and nickel are the principal catalysts of the FT synthesis.[6] Mangan and ruthenium have also catalytic activity. Nowadays, only iron is of technical importance. It is believed that the action of the reactants on the metallic component leads to changes in composition and structure which present the active catalyst phase. An important role in catalytic activity play promoters which can be divided in structure influencing and energetic promoters. The most important energetic promoter of iron catalysts is K_2CO_3. Its exact dosage (usually less than 1% relative to Fe) is very important as it promotes formation of longer chains, olefins and oxygenates but simultaneously leads to coke deposition.

Several empirical working equations have been proposed for the kinetics of the FT synthesis. No general expressions are available. Summaries are found in Kölbel and Ralek [3] and Vannice [7]. Recent results on promoted Fe catalysts have been reported by Dry and co-workers.[8,9]

Heat Generation

The hydrocarbon formation is accompanied by intense heat generation. Depending on the extent of the conversion reaction (2) the heat generation amounts to (1-1,5) x 10^7 kJ per ton hydrocarbons produced. This corresponds to about 25% of the heat of combustion of the synthesis gas. It is, therefore, understood that heat removal presents one of the major problems of industrial FT processes.

FT processes

Various FT processes have been proposed and either used commercially or tested extensively.[10,11] They differ particularly on the type of reactor and the kind of heat removal. It is the reactor and its operational conditions which determine the product distribution and the upgrading scheme. At first, one can roughly distinguish between FT processes with stationary and mobile catalyst phase. Among the various fixed bed processes it is only the ARGE reactor (Arbeitsgemeinschaft Lurgi-Ruhrchemie) which meets the present standards of fixed bed reactor technology and offers high performance and production capacities. Reactors with mobile catalysts are fluidized beds (Hydrocarbon Research Inc.), entrained fluidized beds (Sasol, Kellogg), the three-phase slurry reactor (Rheinpreussen-Koppers), and the three-phase fluidized bed and fixed bed reactors with oil circulation (BASF, Bureau of Mines).

In the following the ARGE process and the entrained bed reactor which both are commercialized by Sasol will be treated briefly. Though these two processes work successfully and profitably now this must not necessarily imply that they are optimal choices under all circumstances. Other technologies may prove viable too, and,possibly, they may be better suited under certain conditions to produce synthetic fuels. In particular, three-phase reactor technology has experienced a fast development, and the aspects to carry out the FT synthesis in 3-phase reactors appear very promising. Therefore, the Rheinpreussen-Koppers slurry process and the oil circulation processes of the Bureau of Mines will be also taken into consideration and briefly characterized. Owing to abundant availability of cheap oil these processes had no chance to be commercialized.

Fixed Bed Process of Ruhrchemie/Lurgi (ARGE)

A simplified process scheme is presented in Fig. 1. The purified synthesis gas (sulfur content (H_2S) $\leqslant$ 0.3 ppm) is mixed with recycle gas, compressed to 25 bar, and passes the heat exchanger. An additional steam heater is located directly above the reactor. The reactor consists of 2052 tubes of 4.6 cm in diameter and a length of 12 m filled with promoted Fe catalyst. The overall diameter is 3 m, each reactor contains 40 m^3 (35 tons) of catalyst. The reaction temperature varies from 220 to 250° C and is slowly increased during operation in order to maintain a constant syngas conversion. The maintenance of isothermal conditions is important which is achieved by water cooling and evaporation. In addition, the gas is recycled. The recycle-to-feed gas ratio is about 2.5. One of the main products of the fixed bed process are waxes which are collected immediately after the reactor. The product gas is then cooled giving the hot condensate (heavy oil). Additional cooling condenses the reaction water and the diesel and gasoline fraction.

Typical operational conditions and characteristics of the ARGE reactor are summarized in Table 1. Under these conditions the ARGE reactor gives a maximum syngas conversion of about 50%. However, under modified conditions, conversions as high as 73% can be obtained.[10] The largest FT synthesis plant

Table 1: Characteristics of ARGE fixed bed process[10]

Operational condition	
Pressure, bar	23 - 25
Temperature, °C	220 - 250
H_2/CO Ratio in fed gas	1.3 - 2
Recycle/feed ratio	2.5
Catalyst loading (Nm^3 syngas/m^3 cat h)	500 - 700
Catalyst	
Composition	100 Fe,5 Cu,5 K_2O,25 SiO_2
Catalyst life, months	9 - 12
Reactor	
Catalyst volume, m^3	40
Number of tubes	2052
Tube diameter, cm	4.6
Tube length, m	12
Volume of catalyst per tube, l	20
Reactor performance (t C_{2+}/day)	5+

based on ARGE fixed bed reactors is operated by Sasol. The composition of the products is given in Table 2. 5 reactors each with 40 m^3 of catalyst produce 250 t of C_{2+} hydrocarbons per day. It is assumed that the present state of knowledge of fixed bed reactor technology would easily permit to enlarge reactor performance by scaling up.[10] This could be done by increasing the number of tubes and their diameter and length and by raising the operating pressure and feed flow rate, possibly simultaneously with recycle flow. An increase in reactor performance of the present 18,000 tons/year to 70,000 tons/year seems to be possible without serious difficulties.[10]

Entrained Bed Reactor (Kellogg, Sasol)

This process is the most successful approach to carry out the FT synthesis in large scale operation. It was originally developed by Kellogg and has been optimized by Sasol during more than 20 years of operation.

A scheme of the Synthol process is outlined in Fig. 2. The core presents the entrained fluidized bed reactor of about 36 m height and a diameter of 2.2 m. In the reaction zone, two

Table 2: Product distribution (%) of fixed and entrained bed processes (Sasol I)[12]

Components	Fixed bed	Synthol
CH_4	2.0	10.0
C_2H_4	0.1	4.0
C_2H_6	1.8	4.0
C_3H_6	2.7	12.0
C_3H_8	1.7	2.0
C_4H_8	2.8	9.0
C_4H_{10}	1.7	2.0
Petrol, C_5-C_{11}	18.0	40.0
Diesel, C_{12}-C_{18}	14.0	7.0
Heavy oil		
C_{19}-C_{23}	7.0	
C_{24}-C_{35}	20.0	4.0
Wax C_{35+}	25.0	
Oxygenates	3.2	6.0

cooling aggregates with recirculating oil as cooling media are installed. The reaction zone is followed by a catalyst settling hopper of 5 m in diameter. Here the tail gas leaves the reactor via cyclones which separate the catalyst fines. The reactor contains about 130-140 t of a fused Fe catalyst. The catalyst recirculation rate is 8,000 t/h. To prevent formation of higher liquid hydrocarbons which would disturb a uniform operation the synthesis is carried out at temperatures of 300 to 350° C and a pressure of 20-30 bar. The syngas enters the reactor at about 160° C. The fresh feed gas to recycle gas ratio is about 2 to 2.5 giving a syngas composition with a H_2-to-CO ratio of 6. The high ratio also suppresses formation of liquid hydrocarbon. The fresh feed flow rate amounts to 100,000 Nm^3/h giving a catalyst loading of about 700 Nm^3 syngas/m^3 cat h.

Sasol I operates 3 reactors in parallel. The mean catalyst lifetime is 42 days. On the average, 2.4 reactors are steadily under operation. Each reactor produces 60,000 t of primary products per year. The product distribution is given in Table 2 and compared with that of the fixed bed process. The Synthol process reveals a maximum in gasoline fuel while the main products of the fixed bed process are higher hydrocarbons like diesel, heavy oils and wax.

Table 3: Production figures of Sasol II [12]

Coal gasified	1030 t/h
Purified synthesis gas	1.1 x 10^6 m^3 STP/h
Products	10^3 t/a
Motor fuels	1,400
Ethylene	180
Chemicals	175
Tar products	287
Ammonia	100
Sulphur	75

The new Sasol II plant is intended to produce mainly gasoline and is therefore based entirely on Synthol technology. The Sasol II reactors have each a capacity of 2 1/2 times that of Sasol I. A simplified process scheme is shown in Fig. 3, and Table 3 summarizes the intended production figures of Sasol II.[12]

Hydrocol Process

In context with the Synthol process, the Hydrocol process developed by Hydrocarbon Research Inc. and others should be mentioned. Here, the hydrocarbon synthesis is carried out in a fluidized bed reactor. The operational conditions and the product distribution are very similar to the Synthol process, however, higher syngas conversions could be achieved. In the early Fifties, a large-scale production unit based on two fluidized bed reactors was constructed in the US (Brownsville, Texas). This plant was intended to produce 360,000 t of hydrocarbons per year and would have been the largest FT plant around the world. However, the planned production capacity was obviously never reached. After two years of operation, the plant was shut down. One can be sure that this was due only to economic reasons as cheaper hydrocarbons from oil were abundantly available during that time.

Slurry Reactor (Rheinpreussen-Koppers)

The three-phase slurry reactor was originally developed as an alternate to the fixed bed. In particular, the starting point were the following disadvantages of the fixed bed process: insufficient heat removal, nonuniform catalyst loading, use of hydrogen-rich syngases, and low conversion.[3,13] In 1951, a demonstration plant of the slurry process was erected by

Rheinpreussen and Koppers. The process scheme is given in Fig. 4. The reactor presents essentially a bubble column which is just a pressure resistant steel cylinder of 1.5 m in diameter and a length of 8.6 m. The liquid phase is molten wax. The best is to use a hydrocarbon product fraction of the synthesis itself. In the liquid phase, a precipitated and promoted Fe catalyst is suspended by the motion of the gas bubbles. The sulfurfree preheated gas is sparged at the bottom of the column. The generated heat is removed by cooling tubes located in the slurry. The reactor temperature is simply controlled by the pressure of the saturated steam in the steam collector. The tail gas leaves the reactor at the top and is precooled in a heat exchanger which partially condenses the higher boiling products. Further cooling condenses the other liquid products which are separated and upgraded in the usual manner. The higher-boiling synthesis products remain in liquid phase and are withdrawn from the suspension by filtering off.

Table 4: Performance and results of FT slurry phase demonstration plant (one-stage column without gas recycle)

Reaction volume	10 m^3
Catalyst	880 kg Fe
Pressure	12 bar
Temperature	268° C
Gas flow rate	2,700 Nm^3/h
Ratio CO/H_2	1.5
CO conversion	0.91
$(CO + H_2)$ conversion	0.89
Products per Nm^3 $CO + H_2$	
C_{1+}	178 g/Nm^3
C_1+C_2	12 g/Nm^3
C_{3+}	166 g/Nm^3
Space time yield (C_{3+} per day and m^3 reactor volume)	930 kg

In Tabel 4 characteristic operational conditions are compiled.[3,13] The slurry process gives a high single pass conversion of about 90%. The yield of C_{3+} products is 166 g per Nm^3 syngas (theoretical yield 208 g/Nm^3). The reactor of the Rheinpreussen-Koppers demonstration plant had an effective reactor volume of about 10 m^3 and produced 11.5 t of hydrocarbons per day. The process offers great flexibility with respect to the product distribution which can mainly be affected by the catalyst and such important operation variables as temperature, pressure, CO/H_2 ratio of feed gas and space velocity, something that can scarcely be done in fixed and fluidized bed processes.

Table 5: Product composition from demonstration plant (mode of operation adjusted for gasoline production)[13]

	Mass $g/Nm^3(CO+H_2)$	% wt. of total product	Olefin content %
Methane + ethane	5.7	3.2	0
Ethylene	6.3	3.6	100
C_3	40.3	22.6	75-85
C_4	9.1	5.1	70-80
40 to 180° C fraction	95.5	53.6	70
180 to 220° C "	7.1	4.0	48
220 to 320° C "	10.7	6.0	37
320° C fraction	3.3	1.9	7
Total	178.0	100	

The product distribution obtained under the operational conditions listed in Table 4 is given in Table 5. This special example leads to a maximum value of the gasoline fraction which was the main goal of the demonstration plant. Only about 4% of the total hydrocarbons produced were in the form of methane and ethane. Other examples for maximizing the fraction of lower and higher hydrocarbons, respectively, are presented by Kölbel and Ralek.[13]

The flexibility of the liquid phase process with respect to gas composition is particularly demonstrated by the fact that CO rich synthesis gases can be used which contain no hydrogen but steam (Kölbel-Engelhardt synthesis). The synthesis starts then by the formation of hydrogen from the shift reaction (2) which is followed by the hydrocarbon synthesis. The overall reaction is

$$3CO + H_2O \rightarrow -(CH_2)- + 2CO_2$$

This synthesis carried out in slurry phase on Fe or other catalysts yields practically the same products as the FT synthesis with high conversions. It was pointed out that a combination of the synthesis step with an internal water-gas shift reaction could result in substantial energy savings as well as take advantage of gasifiers which could produce low ratio H_2 to CO synthesis gas at lower cost.

Obviously, with the slurry process the chief technical problem of heat removal from the synthesis reactor was definitely solved. In addition, the slurry process is of great flexibility and gives high conversions. These advantages over the Rheinpreussen-Koppers process were confirmed by other investigations,[4,14-17] particularly by studies from the Bureau of Mines.[4,17] Schlesinger and co-workers demonstrated that by using a carbided Fe catalyst about 50% of oxygenates can be obtained from the slurry process.

One should mention that the favorable results of the Rheinpreussen-Koppers demonstration plant formed the basis for an offer made to the Indian government to erect a complete liquid phase synthesis plant with an annual production of 250,000 t of hydrocarbons in 1955. Owing to the switch from coal to petroleum, the plans for constructing such an FT plant were, however,not realized.

Oil Circulation Process (Bureau of Mines)

The oil recycle process was first investigated at BASF in Germany and later by Bureau of Mines workers.[18,20] As to modern terminology the oil recycle process was a co-current trickle bed operated in upwards flow. The heat of reaction was removed with the oil, temperature differentials of 15 to 20° C across could be attained. However, after a relatively short period of operation, the catalyst particles crumbled and matted together.[18] Due to pressure drop the bed was inoperable. Therefore, the Bureau of Mines developed a new method of operation - the moving or jiggling catalyst bed.[18-20] The linear velocity of the cooling oil was increased to such a value that the catalyst bed (particle diameter 0.4 to 2 mm, 10 to 40 mesh) expanded by about 25 to 35%. Such a three-phase fluidized bed was operated successfully for several months without any increase in the pressure drop across the catalyst bed and entirely without cementation of the bed. Fig. 5 presents a schematic picture of the synthesis reactor used in the demonstration plant of about 50-barrel-per-day capacity which was conducted by the Bureau of Mines at Louisiana, Mo.[19] Although considerable attrition of the catalyst particle occurred, this had no or little effect on the catalyst activity and catalyst carry-over from the reactor in the oil circulation lines was negligible.

Table 6 summarizes performance of the demonstration plant operated at half capacity.[19] It can be discerned that CO rich gases can be processed at a conversion level of 86% giving 178.6 g hydrocarbons per Nm^3 syngas. The C_{3+} yield is 154 g/Nm^3. The pressure drop of the bed is only 2 bars and the temperature

Table 6: Performance of three-phase fluidized bed (Bureau of Mines demonstration plant)

Fresh feed gas flow	1750 Nm^3/h
H_2/CO ratio of feed gas	0.76
Recycle ratio	1.63
Maximum temperature	273° C
Temp. gain in reactor	8° C
Maximum pressure	23 bar
Pressure drop in reactor	2 bar
Space velocity	531 hr^{-1}
Coolant oil circulation rate	32 l/s
Conversion	85.9%
H_2/CO ratio reacted	0.71
C_1+C_2 yield, $g/Nm^3(CO+H_2)$	24.6
C_{3+} yield, $g/Nm^3(CO+H_2)$	154

gain reaches about 8° C. The liquid product distribution found in pilot plant operation of the three-phase fluidized bed reactor is given in Table 7 for two inlet ratios H_2/CO.[18] The fractions of gasoline and diesel obtainable from the oil circulation process are about the same as those from Rheinpreussen-Koppers slurry process. The gasoline fraction is more than 50%. The heavy destillate and wax cuts can readily be cracked to the diesel or gasoline fractions which results in 85% of the liquid products being obtained as transportation fuels.[18]

Table 7: Liquid products in pilot plant operation, oil circulation FT process

Product	$H_2/CO = 1.3$	$H_2/CO = 1$
Gasoline (C_{3+}-200° C	54.2	52.4
Diesel oil (200-315° C)	10.4	10.0
Heavy distillate (315-450° C)	12.2	16.0
Wax (450° C)	11.5	11.1
Oxygenates	11.7	10.5

Comparison of Various Processes

To compare the performance properties and the capacity of the various FT processes is rather difficult. In any case, it depends on special criteria which may vary considerably.

The 5 FT processes which were briefly discussed here can be distinguished in two groups. On the one hand there is the well established technology of Sasol. As a result, the fixed bed of ARGE and entrained bed reactors were steadily optimized and modernized to present-day technology. On the other hand, there are FT processes realized only in pilot or demonstration plants. These processes, i.e. the Hydrocol process and, particularly, the three-phase technology of Rheinpreussen-Koppers and of the Bureau of Mines seem to be of great potential applicability. It was the major purpose of this contribution to recall these processes which were given up more than 20 years ago, and had actually no chance to prove their effectivity over a longer period of operation and to be optimized in large-scale equipment.

It was outlined above that the fluidized bed reactor of the Hydrocol process operates at similar conditions and gives about the same product distribution as the entrained bed of Sasol. As the slurry process of Rheinpreussen-Koppers and the oil circulation process of the Bureau of Mines are very similar too with respect to operational conditions, conversion and product spectrum, we have mainly three types of FT processes, namely the fixed bed of ARGE (Ruhrchemie-Lurgi), the fluidized beds and the three-phase processes. Table 7 presents a list of some characteristic performance data which are based on Sasol I for the fixed and fluidized bed types of reactors and on the Rheinpreussen-Koppers demonstration plant in the case of the three-phase processes. From the data given in this table and the information available from the literature the advantages of the three-phase FT process can be summarized as follows:

- high single pass conversion
- high yield of C_{3+} products
- large content of transportation fuels in C_{3+} products
- high catalyst and reactor performance
- low methane formation
- high flexibility
- possibility to use synthesis gases of high CO content
- simple reactor design
- no attrition and erosion problems
- easy heat removal.

It is interesting to estimate the number of standard slurry reactors to process 1 million Nm^3 of purified synthesis gas per hour giving about 1.4 million tons of C_{3+} products per

Table 8: Comparison of various FT processes

	Fixed bed (ARGE) Sasol I	Entrained Fluidized bed Sasol I	Three-phase slurry reactor Rheinpreussen-Koppers)
Temperature, ^{0}C	220-250	300-350	260-300
Pressure, bar	23-25	20-23	12 (24)
Ratio CO/H_2 (Feed)	0.5-0.8	0.36-0.42	1.5
Recycle / feed gas ratio	2.5	2-2.4	0
Catalyst loading, Nm^3 $(CO+H_2)/m^3$ cat h	500-700	700	5,000 (10,000)
$(CO+H_2)$ conversion, %	50	77-85	90
Yield of C_{3+},$g/Nm^3(CO+H_2)$	104	110	166
Catalyst performance tC_{3+}/t cat day	1.35	1.85	5.3 (10.6)
Reactor performance tC_{3+}/m^3 reactor vol. day	1.25	2.1	0.93 (1.86)
Content in primary C_{1+} products			
Gasoline, % wt.	18	40	54
Diesel, % wt.	14	7	10

year. These values correspond approximately with Sasol II. Such a standard slurry reactor with a common precipitated and promoted Fe catalyst should favorably operate at a linear gas velcity of 10 cm/s and a pressure of 30 bar. A reactor height of about 10 m would guarantee a 90% conversion. Then 10 reactors of 5 m in diameter or 30 reactors of 3 m in diameter would suffice to process the given amount of synthesis gas. Although no thorough economical analysis is available one can assume that capital costs and operation costs would be considerably less than with two-phase reactors. One may conclude at least that application of three-phase reactor technology presents very promising and advantageous aspects which should be taken into consideration when planning large-scale production of automotive fuels by indirect coal liquefaction.

Captions

Fig. 1: ARGE fixed bed process [10]

Fig. 2: Synthol process [10]

Fig. 3: Process scheme of Sasol II [11]

Fig. 4: Rheinpreussen-Koppers demonstration plant of liquid-phase synthesis [13]

Fig. 5: Reactor of oil circulation FT process [19]

References

(1) J. Falbe (Ed.), Chemierohstoffe aus Kohle, G. Thieme Verlag, Stuttgart 1977

(2) H.-G. Franck and A. Knop, Kohleveredelung - Chemie und Technologie, Springer Verlag Berlin 1979

(3) H. Kölbel and M. Ralek, in ref. (1)

(4) M.D. Schlesinger, H.E. Benson, F.M. Murphy and H.H. Storch, Ind. Eng. Chem. 46, 1322 (1954)

(5) G. Henrici-Olivé and S. Olivé, Angew. Chemie Int. Ed. 15 136 (1976)

(6) Y.T. Shah and A.J. Perotta, Ind. Eng. Chem. Prod. Res. Dev. 15, 123 (1976)

(7) M.A. Vannice, Catal. Rev.-Sci. Eng. 14, 153 (1976)

(8) M.E. Dry, T. Shingles and L.J. Boshoff, J. Catal. 15, 190 (1969), 25, 99 (1972)

(9) M.E. Dry, Ind. Eng. Chem. Proc. Des. Dev. 15, 282 (1976)

(10) C.D. Frohning, H. Kölbel, M. Ralek, W. Rottig, F. Schnur and H. Schulz, in ref. (1)

(11) H. Schulz and J.H. Cronjé, in Ullmann's Encyclopädie der technischen Chemie, Vol. 14, 329, Verlag Chemie, Weinheim

(12) M.E. Dry, Energy spectrum, Oct. 1977, p. 298

(13) H. Kölbel and M. Ralek, Cat. Rev.-Sci. Eng. 21, 225 (1980)

(14) C.C. Hall and D. Gall, J. Hist. Petroleum, 38, 845 (1952)
(15) A.K. Mitra and A.N. Roy, Indian Chemical Engineer 1963, p. 127
(16) T. Kunugi, T. Sakai and N. Negishi, Sekiyu Gakkai Shi., 11, 636 (1968)
(17) M.D. Schlesinger, J.H. Crowell, M. Leva and H.H. Storch, Ind. Eng. Chem., 43, 1474 (1951)
(18) J.H. Crowell, H.E. Benson, J.H. Field and H.H. Storch, Ind. Eng. Chem. 42, 2376 (1950)
(19) M.L. Kastens, L.L. Hirst and R.G. Dressler, Ind. Eng. Chem. 44, 450 (1952)
(20) H.E. Benson, J.H. Field, D. Bienstock and H.H. Storch, Ind. Eng. Chem., 46, 2278 (1954)

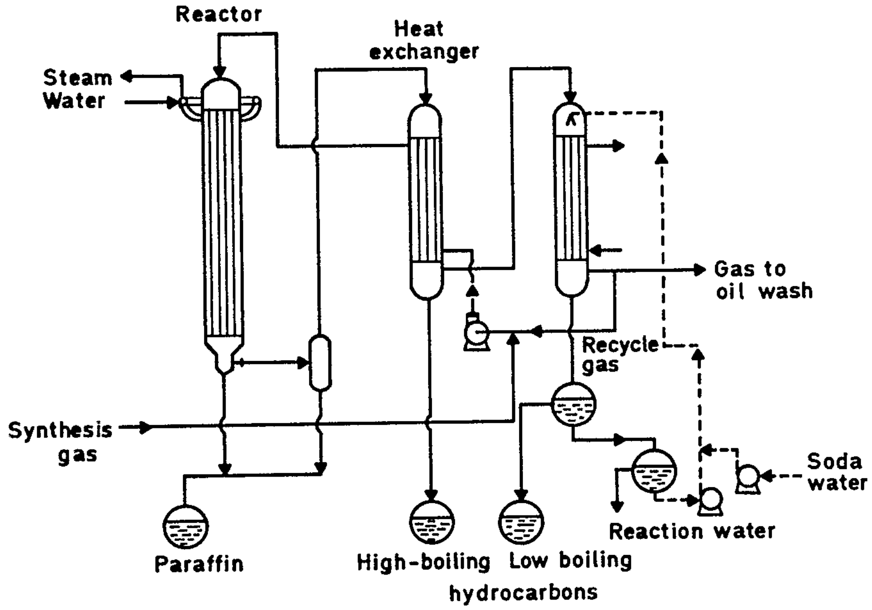

Figure 1

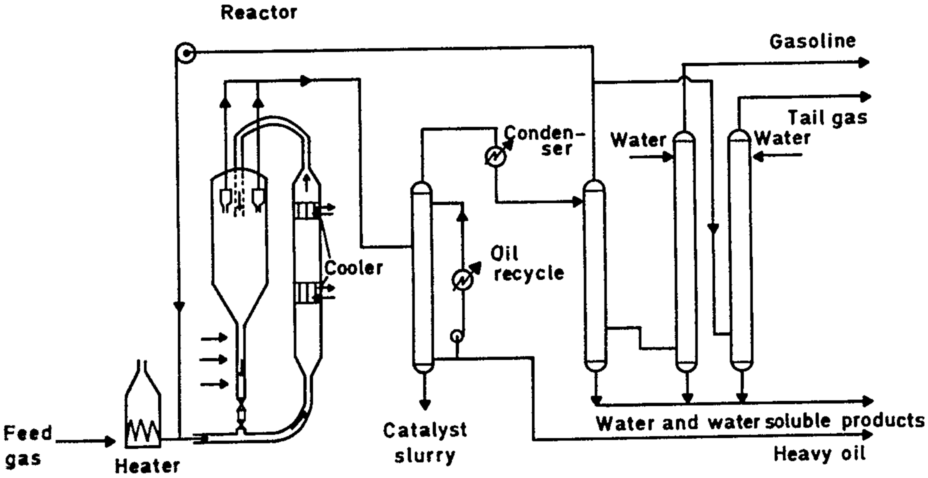

Figure 2

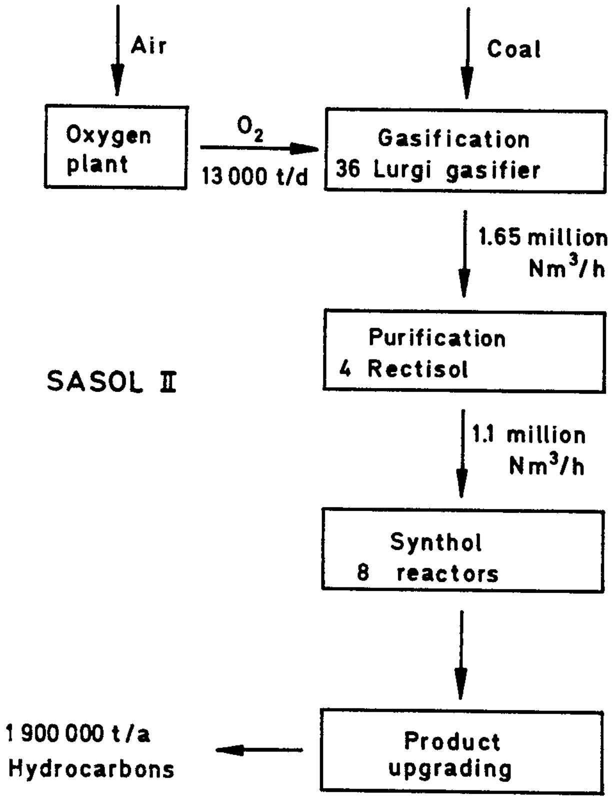

Figure 3

Rheinpreußen-Koppers Demonstration Plant

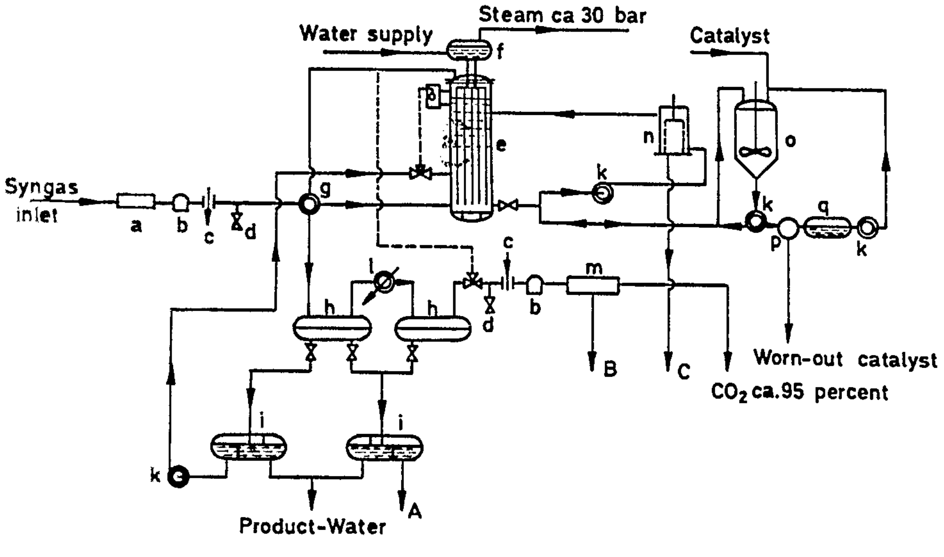

Figure 4

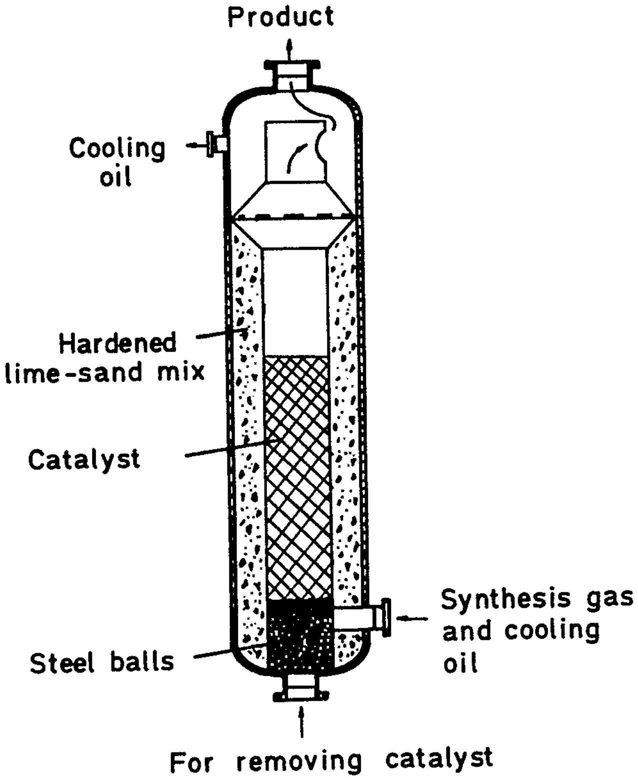
Product
Cooling oil
Hardened lime-sand mix
Catalyst
Synthesis gas and cooling oil
Steel balls
For removing catalyst

Figure 5

DEMONSTRATION AND COMMERCIALIZATION OF THE SRC-I TECHNOLOGY

John C. Tao, A. D. Bixler, and E. J. Breyne

DEMONSTRATION AND COMMERCIALIZATION OF THE SRC-I TECHNOLOGY

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Abstract

The Solvent Refined Coal (SRC-I) Process, one of the most advanced direct coal liquefaction processes available, has attracted national attention because (1) it is a partial answer to the energy crisis and (2) it is an answer that is about to be implemented. A 6000 tons per day SRC-I Demonstration Plant is now being designed and will be constructed and operated by the International Coal Refining Company (ICRC)--a partnership of Air Products and Chemicals, Inc., and Wheelabrator-Frye Inc.--under contract to the Department of Energy (DOE). The project, which is already in the detailed engineering phase, has scheduled operations at the Demonstration Plant, in Newman, KY, to begin by late 1984. Expansion to commercial production, 30,000 TPD, is planned for 1990.

This paper discusses the selection of major technologies for both the Demonstration Plant and the Commercial Plant. For the Demonstration Plant it presents the analyses that governed the selection of technologies for four processing steps:

- Deashing
- Gasification of the mineral ash residue to produce hydrogen
- Product cooling or solidification of the SRC
- Expanded-bed hydrocracking to convert the SRC to liquid fuels

For the Commercial Plant it discusses the overall design of that expanded operation, the projected product slate, product costs--comparing SRC costs with projected oil prices for the next decade when the Commercial Plant will be operating--financing assumptions, and schedule.

The objective of the Demonstration Plant--the initial 6000 TPD facility--is to demonstrate the technical feasibility, economic viability, and environmental acceptability of the SRC-I Process. In essence, the project will be using the abundant coal reserves in the United States to provide liquid and solid fuels that will be competitive with--and could displace--petroleum-derived fuels. Competitive both environmentally, because SRC products are low in sulfur and ash, and economically.

On a long-range basis, products coming from the SRC-I Plant will include:

- Gasoline
- Turbine fuels
- Heating oils
- Two-Stage Liquefaction (TSL) Solids, which are particularly low in sulfur
- Feedstocks for chemical production
- Raw materials for the aluminum and steel and other basic industries

During the first phase of the project, labeled Phase 0, ICRC prepared the conceptual design, preliminary cost estimates, marketing assessments, economic evaluation, and environmental assessments. Process options were evaluated, critical technology areas requiring additional data identified, and the economics assessed of both a "grassroots" five-module, 30,000 TPD Commercial Plant--"grassroots" in the sense that it would be built on an undeveloped site--and a Commercial Plant of the same size expanded from the Demonstration Plant. The Phase 0 work was completed in July 1979. In October 1979, the Department of Energy authorized Phase I, the detailed engineering portion of the project. Site work will start in early 1981; procurement and construction (Phase II), in 1981; and operation and evaluation of the plant (Phase III), in late 1984.

Demonstration Plant Process Description

Figure I presents a block diagram of the major processes of the Demonstration Plant.

Coal preparation means receiving, unloading, conveying, storing, reclaiming, drying, and grinding the coal as feed for the process units. Approximately 93% of the coal is ground, dried, and fed to the SRC Process Unit. The remaining 7% is ground, dried, and fed to the Gasification Unit.

In the SRC Process Unit, the coal is slurried in a process solvent, pumped to reaction pressure, mixed with a hot, hydrogen-rich gas stream, and heated in a fired heater. Within the fired heater, coal dissolution is accomplished and hydrogenation reactions begin. Additional hydrogen-rich gas is added at the exit of the fired heater and the mixture flows to the dissolver, where hydrogenation and desulfurization reactions are completed. The high-pressure, hydrogen-rich gas is separated from the product slurry, which is flashed and distilled to remove process solvent and lighter components. The remaining SRC-ash slurry is sent to the Kerr-McGee Critical Solvent Deashing Unit, where it is mixed with the deashing solvent and separated into an SRC product stream and an ash concentrate stream.

The SRC product is divided into three parts; approximately one-third goes to expanded-bed hydrocracking, one-third to coking and calcining, and one-third to product solidification. The Kerr-McGee ash concentrate stream is sent to the Coal Gasification Unit.

Gasification serves two purposes: (1) it converts the ash concentrate into an inert slag; and (2) through gasification of the carbonaceous components of the ash concentrate plus some supplemental coal, it generates the makeup hydrogen that is required for both the liquefaction and the expanded-bed hydrocracking processes. Gasification is performed at atmospheric pressure by a partial oxidation step, the oxygen being obtained from an Air Separation Unit.

The resulting gas is compressed and passed over a shift conversion catalyst to produce the raw makeup hydrogen stream. Following removal of the acid gases from this stream, the purified hydrogen stream is divided, with approximately two-thirds going to the SRC Process Unit and one-third to expanded-bed hydrocracking.

Following its use in each of these units, the high-pressure hydrogen-rich gas passes through a series of treating steps before it is recycled back to the SRC Process Unit. The combined gas stream is washed to remove chlorides and ammonia, scrubbed to remove acid gases, dried, and cryogenically processed to reject light hydrocarbons, which are used as plant fuel. Acid gases recovered from treating the combined gas stream and the makeup hydrogen stream are sent to the Sulfur Recovery Area, where molten sulfur is produced and removed in a Claus Unit and a Beavon Sulfur Removal Unit.

The expanded-bed hydrocracking process receives part of the original SRC Process Solid--sometimes called Classic SRC Solid--and converts it catalytically into naphtha, fuel oil, and low-sulfur SRC solids, called TSL Solids. The flexibility exists to produce these products in different ratios.

In the Coker/Calciner Area, the major product is anode coke, with byproducts of fuel oil and naphtha.

The single product of the Solidification Unit is SRC Process Solid. The Solidification Unit is sized to handle the entire output of the SRC Process Area so that maintenance may be performed on either or both the Expanded-Bed Hydrocracker and the Coker/Calciner Units without reducing the coal feed rate to the SRC Plant.

* * *

Technology Selection

Various trade-off studies were performed during Phase 0 and continued into Phase I to select major technologies for the Demonstration Plant. They included process design, data base, and technical uncertainty studies, as well as economic studies for both the Demonstration Plant and the full-scale 30,000 TPD Commercial Plant.

Deashing

As part of the conceptual design phase for the Demonstration Plant, an evaluation of solid-liquid separation techniques was made. The evaluation included the Kerr-McGee Critical Solvent Deashing (CSD) Process and the Lummus Antisolvent Deashing Process and was based on operating data from both the Wilsonville and the Fort Lewis Pilot Plants.

Mechanical methods for ash separation were also investigated including centrifuges, hydroclones, and various filtration techniques--such as pressure leaf filters (U.S. filter, Funda); pressure candle filters (Bergbau, Industrial); and pressure rotary drum precoat filters (Johns-Manville).

The initial analyses indicated at least two promising approaches: (1) Kerr-McGee Critical Solvent Deashing Process; and (2) filtration based on the U.S. vertical leaf filter, which was selected for the Phase 0 design. An economic comparison of the Kerr-McGee Process at an 81% SRC recovery with an array of twelve 5000 ft^2 U.S.filters (95% SRC recovery) proved to be a standoff. The higher capital cost for filtration was offset by the higher losses of SRC in the Kerr-McGee Process. But because filtration operates in a batch mode, has high maintenance requirements, and subjects plant workers to a potential exposure to heavy hydrocarbons, a continuous process was ultimately favored during Phase 1 evaluation.

The Lummus Antisolvent Deashing Process. A schematic flow diagram for the Lummus Antisolvent Process is shown in Figure II. Dissolver effluent passes to high-pressure and low-pressure separators, where light gases, including unreacted hydrogen and H_2S, are removed. The remainder goes to a fractionator to remove light liquids with a boiling point below 500°F.

The resulting slurry is sent to a feed surge drum. The feed consists of process solvent (500°-850°F), SRC, mineral residue, and unconverted coal. The feed is mixed inline with antisolvent and distributed within a gravity settler to settling chambers. Each chamber is a section of an inclined conical surface. The heavy phase flows to an underflow outlet pipe. The clear, light liquid phase flows to an overflow collection pipe, which takes it to antisolvent fractionators (including a vacuum column) for recovery of antisolvent. SRC from the vacuum column is cooled and solidified. An underflow stripper recovers antisolvent from the settler underflow--the stripper bottoms are sent to a vacuum unit for recovery of process solvent. The vacuum bottoms, containing up to 40 wt % solids, are used to generate hydrogen as gasifier feedstock.

The Kerr-McGee Critical Solvent Deashing Process. Unlike the Lummus Process, which is an agglomerative gravity sedimentation process, Kerr-McGee CSD is a solvent extraction process operated at conditions near the critical point of the deashing solvent, for which two liquid phases are formed. A schematic flow diagram of the Kerr-McGee CSD Process is shown in Figure III.

Dissolver effluent is let down to remove light gases and then fractionated to remove almost all of the process solvent. The feed slurry to the CSD unit is a vacuum bottoms product with 3-10% process solvent. The vacuum bottoms product is mixed inline with deashing solvent at temperature-pressure conditions near the critical point of the deashing solvent and fed to the First Stage Settler.

The overflow light liquid phase contains deashing solvent, soluble components of the SRC, and less than 0.1 wt % ash. The underflow heavy liquid phase contains deashing solvent, unextracted portions of SRC, ash, and unconverted carbon. The overflow is heated above the critical point of the deashing solvent to reject the SRC. This can be done in one or more stages to separate lighter SRC components (LSRC) from the heavier, less soluble components. The SRC product is cooled and solidified. The underflow from the First Stage Settler is flashed to recover deashing solvent. The remainder, including ash, unconverted carbon, and SRC, forms a finely divided powder known as ash concentrate, which is then fed to a gasifier to generate hydrogen.

Kerr-McGee Process chosen. The Lummus Process is at an earlier stage of process evolvement than the Kerr-McGee Process. Lummus has not yet demonstrated a sustained operation that produces an acceptably low ash in the overflow and a high enough (70% plus) SRC recovery.

Several mechanical methods, such as rotary drum and horizontal leaf filters and centrifugal techniques have been eliminated from consideration for high capital and operating costs, low mechanical reliability, inability to meet product ash specifications, production of a dilute underflow, inability to scale up to commercial size--or some combination of these.

Based on the above analysis, detailed engineering is proceeding with the Kerr-McGee CSD Process as the base case, but allowing sufficient plot space for a filtration system. If near-term developmental results for filters at the Wilsonville Pilot Plant are particularly encouraging--or if there are serious new concerns with the scale-up risks of the Kerr-McGee CSD Process--a parallel design effort for filtration will be initiated.

Gasification of Mineral Ash Residue for the Production of Hydrogen

As we said in describing the process, the Gasification Area has the dual function of rendering the ash from the coal feedstock into an inert residue material and, at the same time, generating the makeup hydrogen required for both the expanded-bed hydrocracking and the SRC Process. One complication regarding the selection of a gasification technology is that this step has not been piloted in any of the four coal liquefaction Pilot Plants--SRC-I at Wilsonville, AL; SRC-II at Tacoma, WA; Exxon Donor Solvent at Baytown, TX; and H-Coal at Catlettsburg, KY.

Another complication is the fact that the feed characteristics will vary, depending upon the selection of the deashing process. For example, mineral ash residue from filtration contains up to 60% ash, but Kerr-McGee ash concentrate typically contains no more than 40% ash. Although both deashing processes produce solids in very fine particles, the solids from the Kerr-McGee Process are signficantly smaller than those from filtration. The fine particle size of the feedstocks favors the selection of an entrained flow gasification process, rather than moving-bed or fluidized-bed processes.

Three gasification processes investigated. During the Phase 0 work on the SRC-I Process, evaluations were made of Texaco, Gesellschaft für Kohle Technologie (GKT)--formerly known as Koppers-Totzek--and Shell-Koppers Coal Gasification Processes for producing hydrogen for the SRC Process from a mixture of filter cake and coal. The Shell-Koppers Process had a small economic advantage over the Texaco Process; the atmospheric pressure GKT Process was the most expensive. Despite its economic advantage, the Shell-Koppers Process was considered insufficiently developed at that time--and the Texaco Process was tentatively selected in conjunction with filtration deashing for the Phase 0 study. Filter cake had been successfully gasified by Texaco in its Montebello Pilot Plant (see EPRI Report AF-777).

However, as noted above, in the initial work on the detailed design of the SRC-I Plant, the Kerr-McGee CSD Process was selected over filtration, and an evaluation of the potential slurry concentration that could be achieved with the ash concentrate was undertaken by both Texaco and ICRC. This work indicated that an economic slurry concentration of the Kerr-McGee ash concentrate in water could not be achieved with the very fine particle size of this material. Although pelletizing and regranulating the ash concentrate would be expected to result in desirable particle sizes, this procedure would be an extra, unproven processing step for the facility.

In the same period, small-scale tests were undertaken by GKT to gasify an ash concentrate blended with 10 wt % of coal and a filter cake blended with 32.5 wt % of coal. Both feeds were successfully gasified in the pilot scale runs.

GKT Process recommended. An evaluation of the processes indicated that, even with the added cost of granulating and crushing to optimize particle size and thereby slurry concentration, the Texaco Process would have a small economic advantage over the GKT Process. However, in the absence of the granulating and crushing steps, the resulting decrease in achievable slurry concentration would present such a severe penalty to the Texaco Process that the GKT Process would have a substantial advantage. For these reasons--(1) the technical risk associated with the granulating and crushing steps; (2) the lack of sufficient demonstration for the Texaco Process; and (3) the potential uncertainties in the characteristics of the ash concentrate feedstock--the GKT Process has been recommended for use in the Demonstration Plant.

Product Solidification

Three alternatives for product solidification were considered: (1) indirect cooling in a small batch on vibrating trays (Rexnord); (2) indirect cooling on a continuous moving belt (Sandvik); and (3) direct contact cooling by injection into a water bath (Mitsui-Miike).

Rexnord and Sandvik. In the two indirect cooling process options, the molten SRC is sprayed onto a metal surface for indirect cooling. The SRC is open to the atmosphere for the entire cooling process.

Cooling is accomplished by spraying cooling water on the opposite side of the moving belt in the case of the Sandvik belt--and by circulating cooling water through coils on the opposite side of the cooling trays in the case of the Rexnord system. In neither option does the cooling water come into direct contact with the SRC.

Mitsui-Miike. The Mitsui-Miike Process cools and solidifies SRC by direct contact with water. Because it has not been used in the U.S. up to now, the Mitsui-Miike Process is discussed in detail here. Figure IV represents a schematic flow diagram of the process.

The hot molten SRC (pitch) is first cooled against Dowtherm in the pitch cooler. The extent of the cooling depends on the softening point and viscosity of the pitch. The stream is divided into multiple streams which are then dropped into the solidifier through cylindrical nozzles at a height about 6 to 12 inches above the water surface.

In the solidifier, the denser pitch is cooled and solidified into rods as it falls through the water bath. The water flows upward countercurrent to the pitch. At the bottom of the solidifier vessel, the solid pitch is transferred by the screw conveyor to the bucket conveyor located outside the vessel. The bucket conveyor transfers the solid pitch to the screen conveyor where water is removed. Product pitch is then transferred to storage via belt conveyor(s).

The process water, which cools and solidifies the SRC, is supplied by the submerged circulation pump in the water pit. The process water, which contains fines, is cooled in the process water cooler against cooling water. It is then injected into the solidifier at the bottom section of the vessel through multiple nozzles. Most of the process water returns to the water pit via overflow ports at the top of the solidifier vessel. Water separated by the screen conveyor and a small fines purge stream from the bottom of the bucket conveyor is also returned to the pit. Due to vaporization which occurs in the solidifier, makeup water is required and it is introduced into the solidifier below the process water inlets.

Fines are generated in the process primarily by crushing and attrition in the conveying equipment, especially in the screw conveyor. They are removed from the system by sending a slip stream of the recycle process water to the fines separation unit. The separation can be achieved by either centrifugation or sedimentation. The clarified water is returned to the water pit for reuse. There is no water treatment required except fines separation.

Comparison of the three alternatives. The Mitsui-Miike system is compact and simple compared with both Sandvik and Rexnord. The extensive duct work and subsequent vapor treatment devices required by these two systems can be either eliminated or simplified. Fume generation is much lower for the water-bath system, due to the short distance between the feed nozzle and the water surface.

The temperature of the water bath has a strong effect on the shape and strength of the solidified product. If the water temperature is too low, the product is brittle and breaks easily during conveying and handling; if it is too high, the pitch will form bubbles at the water surface and the product is popcorn-like instead of rod-shaped. Other factors that affect product strength and fines generation are the softening point, the precooled temperature, and the thermophysical properties of the pitch.

The process design is based on the Rexnord cooler because it is the most economical and technically proven process. But the direct contact cooling option is being investigated to determine its technical feasibility. Because of the attractiveness of its simpler, more compact design and the environmental acceptability of its low fume generation, Mitsui-Miike, if proven technically feasible for the specific conditions of SRC-I, will likely be incorporated into the Demonstration Plant.

Expanded-Bed Hydrocracking

The Phase 0 final report for the SRC-I project proposed that an expanded-bed catalytic hydrocracker be incorporated into the facility to convert one-third of the Classic SRC Solid to gases and liquids boiling at less than 850°F (Two-Stage Liquefaction or TSL). The original proposal was to run two-thirds of the Classic SRC Solid through the hydrocracker at 50% conversion to minus 850°F material. The remaining 50% of the SRC Solid would be produced as low sulfur solids (TSL Solids). This has been modified. DOE accepted the overall concept of maximizing product flexibility by adding the catalytic hydrocracker--but asked that only one-third of the Classic SRC Solid be made available to the hydrocracker. ICRC then proposed that this one-third be converted at 85% severity to naphtha and fuel oils below 850°F. This is the design basis under which ICRC has been proceeding since early 1980. However, the Demonstration Plant is being designed to test both proposals.

The economic attractiveness of the 85% conversion is essentially the same as the 50% conversion case, with the higher capital and operating costs being compensated for by the higher value of the lighter product slate. The cost of the liquid fuels produced are within plus or minus 15% of the projected value of these fuels, depending on the finance method employed. Furthermore, the proposed hydrocracker, using the Lummus/Cities Service LC-Fining technology, will be capable of producing low sulfur TSL Solids--as contrasted to higher sulfur Classic SRC Solid (approximately 0.21% sulfur versus 0.80% sulfur). Our present commercial contacts are interested in using the low sulfur solid product to displace low sulfur fuel oil in boilers that had been converted from coal.

Commercialization

A great deal of flexibility exists in selecting the product slate for the Commercial Plant. An SRC-I facility would produce approximately 75 wt % of its product as Classic SRC Solid and the remaining 25 wt % as a range of liquid products. A Two-Stage Liquefaction facility based on low severity hydrocracking would produce approximately 40 wt % of the product as TSL Solids and 60 wt % as liquid products. A high severity hydrocracker would result in only 10 wt % of TSL Solids and 90 wt % of liquids.

The ultimate selection of the Commercial Plant configuration will be determined by market forecasts for the various products. The decision is still several years away, but analyses of Two-Stage Liquefaction facilities have already been performed for both low severity hydrocracking and for high severity hydrocracking.

High Severity Hydrocracking in TSL

Figure V depicts a schematic for a Commercial TSL Plant in which all the SRC is processed through a hydrocracker at high severity. The Commercial Plant is based on 30,000 TPD (33,333 tons per stream day) of raw coal feed. Of this feed, 28,754 TPSD are fed to the SRC liquefaction section to produce 13,427 TPSD of SRC, along with a substantial quantity of product oils, and--from the Kerr-McGee Deashing Area--7773 TPSD of ash concentrate. This ash concentrate, along with the remaining 4579 TPSD of coal, is fed to a Shell-Koppers Gasification Unit to produce 561 MM cfd of makeup hydrogen.* Of this quantity, 261 MM cfd are required in the SRC Liquefaction Area; the remaining 300 MM cfd are used for hydrocracking the SRC.

Total liquid products from both the SRC liquefaction and hydrotreating steps are 6082 TPSD of C_5-400°F material; 6331 TPD of 400°-650°F material; 1499 TPSD of 650°-850°F material; and 68,000 MM Btu/day (higher heating value) of Liquefied Petroleum Gas (LPG). In addition, 1466 TPSD of low sulfur TSL Solids are also produced in the hydrocracker. As noted in Figure V, approximately two-thirds of the product oils are produced in the hydrocracker with the remaining one-third being produced in the SRC Liquefaction Unit.

The combined higher heating value of the above products represents 640,000 MM Btu/day higher heating value. In addition to the 33,333 TPSD of feed coal, 548,000 Kw of electricity will be required to operate the Plant. Using 9,500 Btu/kilowatt hour, the combined energy input into the plant is 975,000 MM Btu/day, resulting in an overall plant efficiency of 65.6%--including energy losses associated with generating all of the plant power requirements. Table 1 shows the energy balance of the 30,000 TPD SRC-I Plant.

An aggressive schedule for a Commercial Plant sets up a five-year design-and-construction period beginning in 1985, with the plant going onstream about 1990. Using this time frame as a basis, both capital and operating costs have been developed for the Commercial Plant described above. For convenience, the values have been de-escalated and costs are presented in this article as if the plant were onstream in 1980.

Capital Costs Total $3570 MM. Table 2 shows a capital cost summary for the major sections of the plant: SRC Liquefaction and Deashing; Expanded-Bed Hydrocracking; Hydrogen Production and Treatment; and Utilities and Offsites, including Coal Preparation. The subtotal for plant and equipment cost is $2375 MM. With the addition of license fees, land, and an initial charge for catalysts and chemicals; allowing a contingency of 20% on the above total; adding interest during construction; plus working capital, and startup costs--the total project cost is $3570 MM.

Operating costs total $1,295 MM annually. The operating cost summary is presented in Table 3. These costs have also been de-escalated to a 1980 onstream date. After startup in 1990, the plant is assumed to have a 20-year operating life. The operating costs presented are for a typical operating year having a 90% onstream factor or 328 1/2 days per year onstream.

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*This configuration assumes the successful development of the Shell-Koppers Technology by 1985.

Major operating costs for the facility will be:

- Coal, at $1.30 MM Btu, with a total annual cost of $365 MM
- Power, at $0.034/kwh, an annual cost of $145 MM
- Catalysts and chemical consumption, $55 MM.
- Maintenance materials, $60 MM
- An operating and maintenance crew of 1645 persons, with an annual cost of $100 MM. (This includes provisions for holiday, vacation and sick leave, fringe benefits, and plant supervision and overhead.)

The subtotal for the various expenditures is $725 MM per year. As evaluated for this study, capital charges represent 16% of the project costs and a total annual cost of $1295 MM per year. These charges are based on a 65% debt/35% equity for Plant financing. The 20% contingency on capital shown in Table 2 has been included in the operating cost summary. Borrowed capital, 65% of the financing, has been allotted an interest rate of 9%; the 35% equity portion is assumed to return a profitability of 15% on a discounted cash flow basis.

Tables 2 and 3, in summarizing the capital and operating costs for the Plant, include a reasonable interest rate and return on equity. A financial analysis, required to determine whether the production from the facility has adequate value to permit this rate of return, is presented in Table 4. The total products, presented in Figure V, have a total heating value of 210,300 MMM Btu/year. To achieve the profitability assumed in Table 3, these products would be required to have an average price of $6.15/MM Btu.

As both Classic SRC Solid and TSL Solids are new products for which a market value has yet to be determined, an indirect method of deriving a price for these materials was used. The TSL Solids represent only about 7.5% of the product heating value from this Plant. Consequently, any value for this product, derived by placing values on the much larger quantities of liquid products, will be sensitive to the capital and operating costs as well as to the product values employed. To minimize this sensitivity to cost factors, the value of TSL Solids was assumed equivalent to the value of Classic SRC Solid, which was derived as follows:

> The capital and operating costs for processing 33,333 TPSD of coal to produce Classic SRC Solid without the expanded-bed hydrocracking step were generated. The liquid products, representing 28% of the total products on a Btu basis, were projected at market value. These projections, forecast by Air Products and Chemicals, Inc. for 1990, have been de-escalated to a 1980 basis.
>
> Naphtha--$7.69 MM/Btu
> Medium oil (#2 oil)--$6.63 MM/Btu
> Heavy oil (#6, low sulfur oil)--$5.57 MM/Btu

After subtracting the total value of the liquid products from the required annual cost of the facility, based on the financing assumptions presented in Tables 2 and 3, a price was derived for Classic SRC Solid that represented the remaining 72% of the heating value of the products.

For the current Plant, the TSL Solids were given a market value equivalent to the value of Classic SRC Solid derived in the above manner. Since the TSL Solids are even lower in sulfur than Classic SRC Solid, this is a conservative assumption. Deducting the assumed value for the Classic SRC Solid from the annual cost of operating the Plant, the average cost for the liquid fuels produced (representing the remaining 92.5% of the product heating value) is $6.40/MM Btu.

Based on the projected unit prices in 1980 dollars listed above and the quantities of liquid products shown in Figure V, an average market value is found for the liquid fuels produced of $6.95/MM Btu. Comparing that value to the average cost for the liquid fuels produced, $6.40/MM Btu indicates a margin of approximately 9% for exceeding the assumptions of this analysis without disturbing the 15% return on equity. These figures are encouraging as to the viability of a Commercial Plant with high severity hydrocracking.

Low Severity Hydrocracking

Figure VI depicts a schematic for a Commercial Plant based on a low severity hydrocracker. This Plant, too, is based on 30,000 TPD (33,333 TPSD) of raw coal feed. However, as less hydrogen is required, a larger fraction of the coal is sent to the SRC Unit, making that unit slightly larger than is called for in the high severity design; however, the hydrogen gasifier is substantially smaller. For the low severity design, approximately half the product oils are produced in the hydrocracker along with 7578+ TPSD of TSL Solids.

Tables 1, 2, 3, and 4 contain information on the low severity case in the same detail as for the high severity case. The low severity case achieves a thermal efficiency of 70.3% at a total projected capital cost of $3.1 MMM and an annual operating cost of $1.16 MMM.

With the value of solid fuels set by the same method (and the same value) as in the high severity case, the average value of liquid fuels, $6.75/MM Btu, exceeds the average cost (including profit) of producing those fuels by 4%. This result is also encouraging with regard to the future potential of an SRC-I coal refinery.

The authors wish to acknowledge the support of the U.S. Department of Energy under Contract DE-AC05-78-ORO-3054.

Table 1

Facility Energy Balance

	High severity hydrocracking	Low severity hydrocracking
Output, MM Btu/day HHV		
LPG	68,000	28,000
Naphtha (C_5-500°F)	240,000	165,000
Medium oil (400°-650°F)	230,000	175,000
Heavy oil (650°-850°F)	54,000	51,000
TSL Solids	48,000	246,000
	640,000	665,000
Input, MM Btu/day HHV		
Coal	850,000	850,000
Electricity	125,000	96,000
	975,000	946,000
Overall efficiency	65.6%	70.3%

HHV = high heating value

Table 2

Capital Cost Summary

(Cost in MM $, de-escalated to 1980 onstream)

	High severity hydrocracking	Low severity hydrocracking
SRC liquefaction and deashing	$ 640	$ 685
Expanded-bed hydrocracking	515	355
Hydrogen production and treatment	760	605
Utilities, offsites and coal preparation	460	445
Subtotal plant and equipment	2,375	2,090
License fees,[1] land, initial catalysts and chemicals	80	70
Contingency	490	430
Interest during construction	320	280
Startup costs	140	110
Working capital	165	145
Total project cost	$3,570	$3,125

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[1] Paid out to third parties

Table 3

Operating Cost Summary[1]

(Cost in MM $, de-escalated to 1980 onstream)

	High severity hydrocracking		Low severity hydrocracking	
Item	Quantity	Annual cost[2]	Quantity	Annual cost[2]
Coal @ $1.30/MM Btu	33,333 TPSD	365	33,333 TPSD	365
Power @ $0.034/kwh	548,000 kw	145	422,000 kw	115
Catalysts and chemicals	-	55	-	45
Maintenance materials	-	60	-	50
Operating and maintenance labor	1,645 persons	100[4]	1,455 persons	85[4]
Subtotal		725		660
Capital charges[3] 16% of project cost		570		500
Total		1,295		1,160

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1. Assumes startup in 1990 and 20-year operating life.

2. 328.5 days/yr onstream

3. Based on 65% debt, 35% equity, 20% contingency on capital, 15% discounted cash flow, 9% interest rate on debt

4. Includes manpower allowances for vacations and sick leave, fringe benefits, plant supervision, and overhead

Table 4

Financial Analysis

(Cost in \$/MM Btu, 1980 \$)

	High severity hydrocracking	Low severity hydrocracking
Total Btu produced, MMM Btu/yr	210,330	218,538
Average required price,[1] \$/MM Btu	\$6.15	\$5.30
Solid SRC cost,[2] \$/MM Btu	3.25	3.25
Average cost of liquid fuels produced,[3] \$/MM Btu	6.40	6.50
Average market value of liquid fuels produced, \$/MM Btu	6.95	6.75

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1. Derived from annual operating cost presented in Table 3 and total Btu produced.

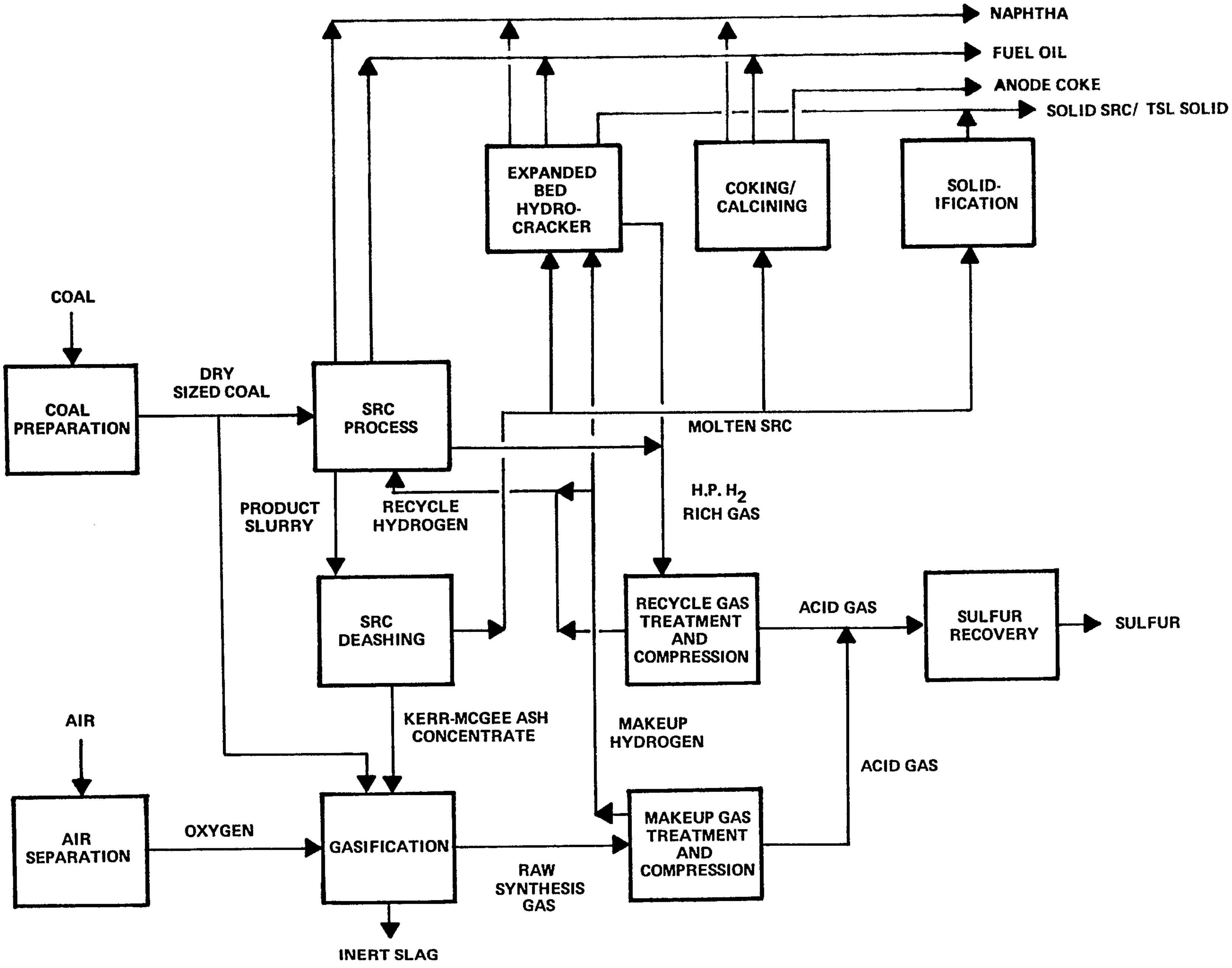
2. Derived from evaluation of investment and operating cost for a stand-alone SRC Plant constructed without an expanded-bed hydrocracker. Market value of liquid products was based on the following unit prices in 1980 \$: Naphtha--\$7.69/MM Btu, medium oil (#2 oil)--\$6.63/MM Btu, heavy oil (#6, low sulfur oil)--\$5.57/MM Btu. The above prices were forecast for 1990 by Air Products and Chemicals, Inc., and de-escalated to a 1980 basis.

   Value of SRC solids established by difference.

3. Established by difference after setting value of TSL Solids equivalent to value of SRC as described in Footnote 2.

FIGURE I

DEMONSTRATION PLANT SCHEMATIC



LUMMUS ANTI-SOLVENT PROCESS

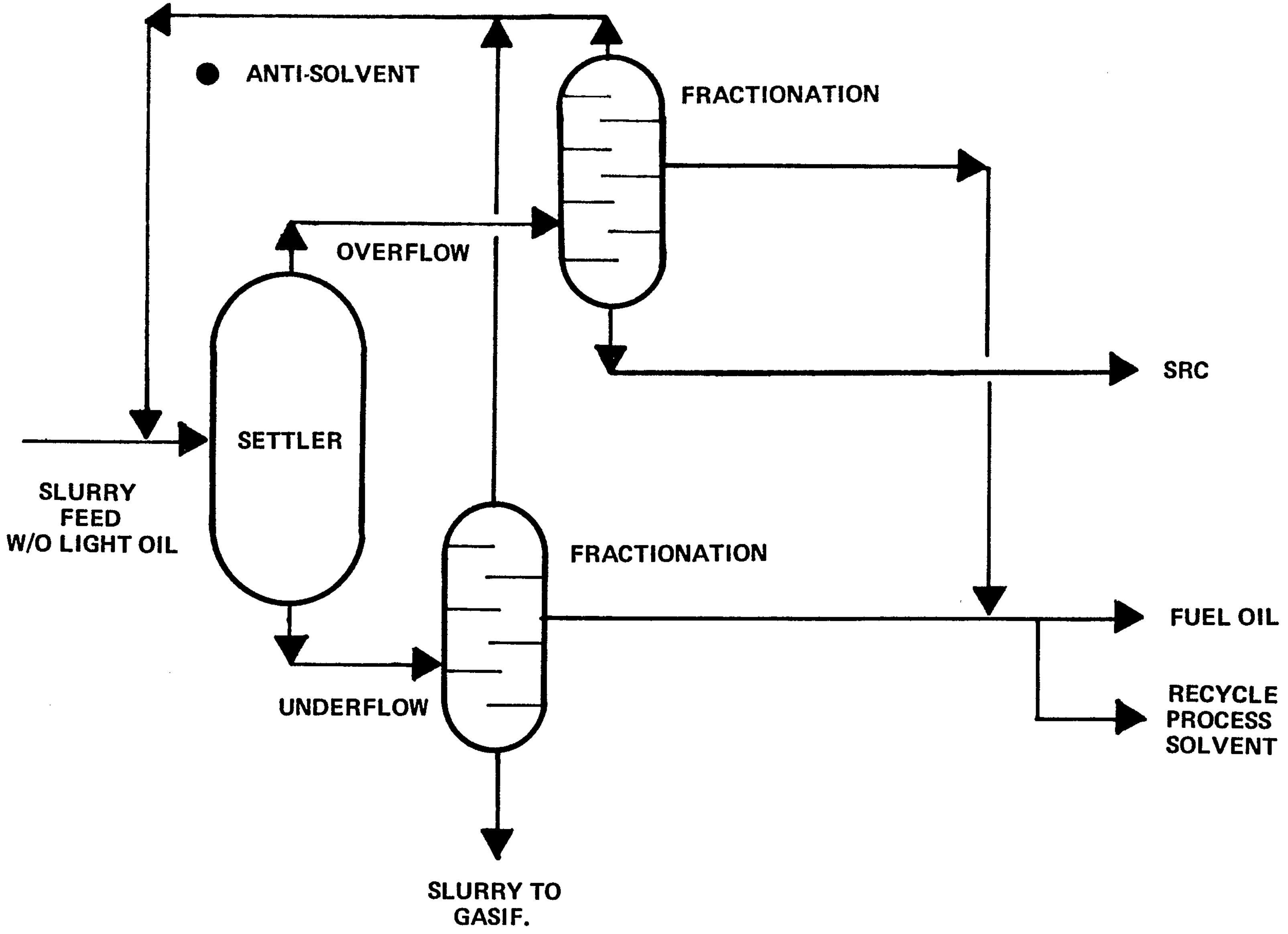


FIGURE III

SRC DEASHING SCHEMATIC FLOW DIAGRAM

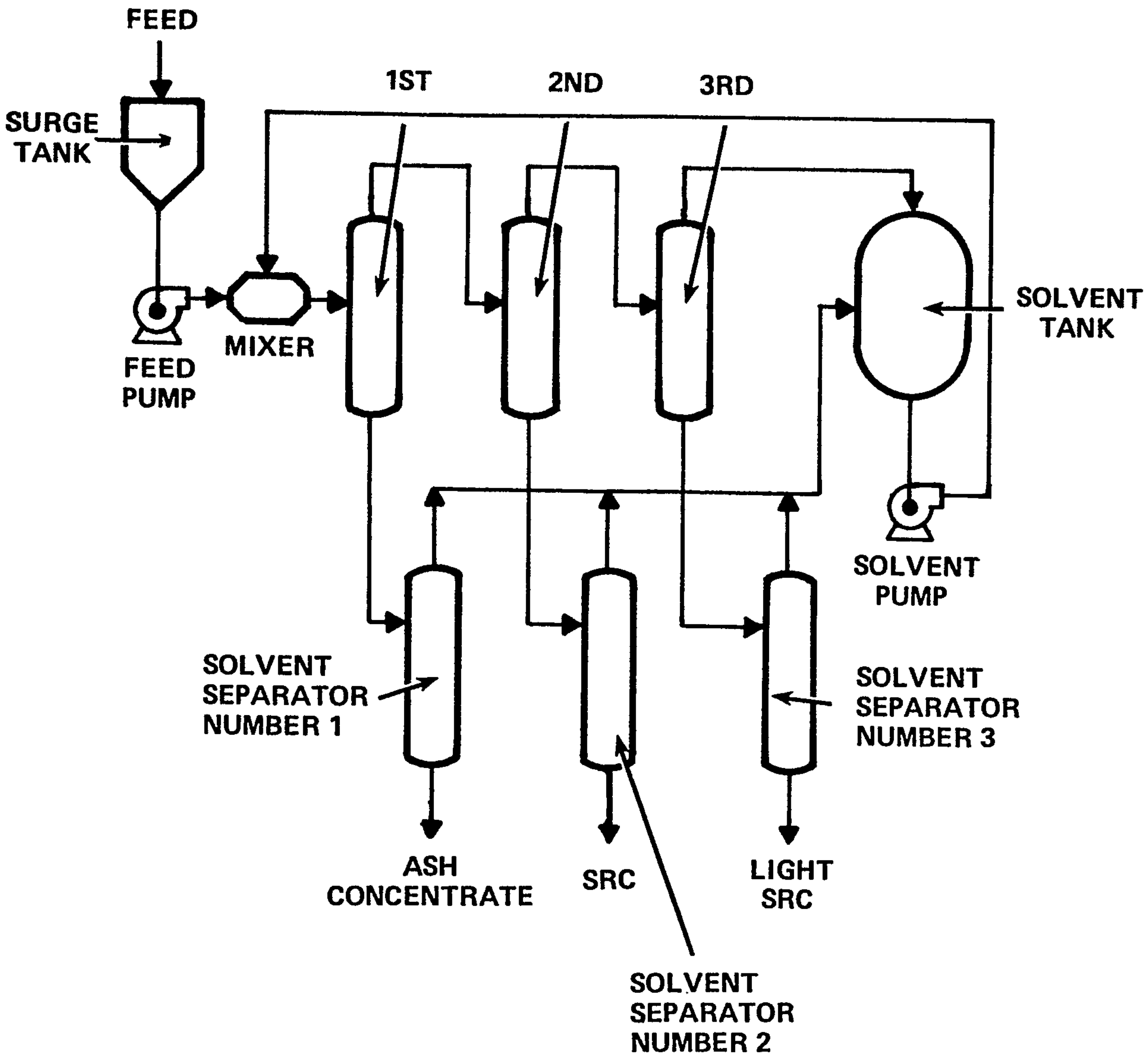


FIGURE IV

MITSUI WATER-BATH SOLIDIFICATION

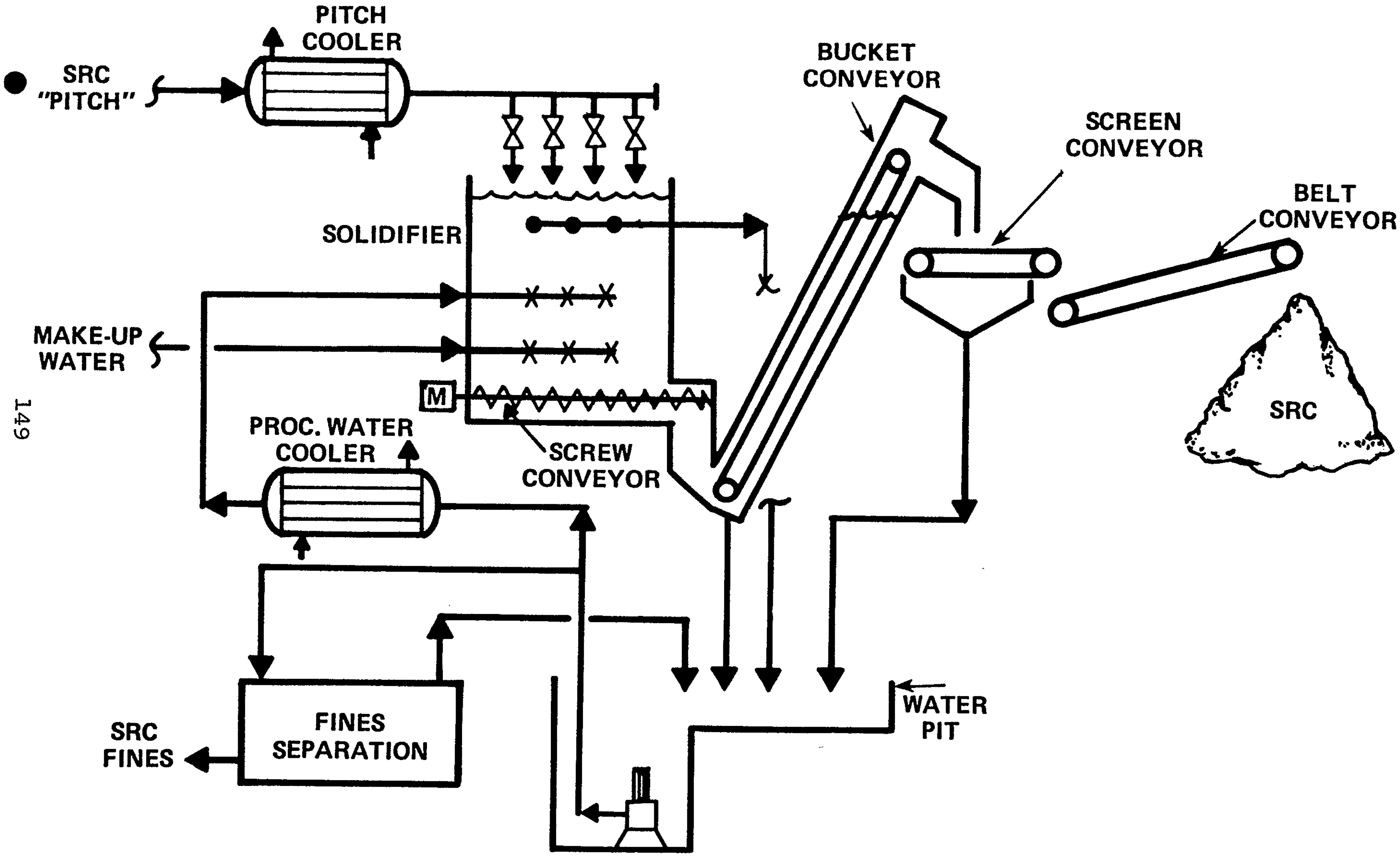
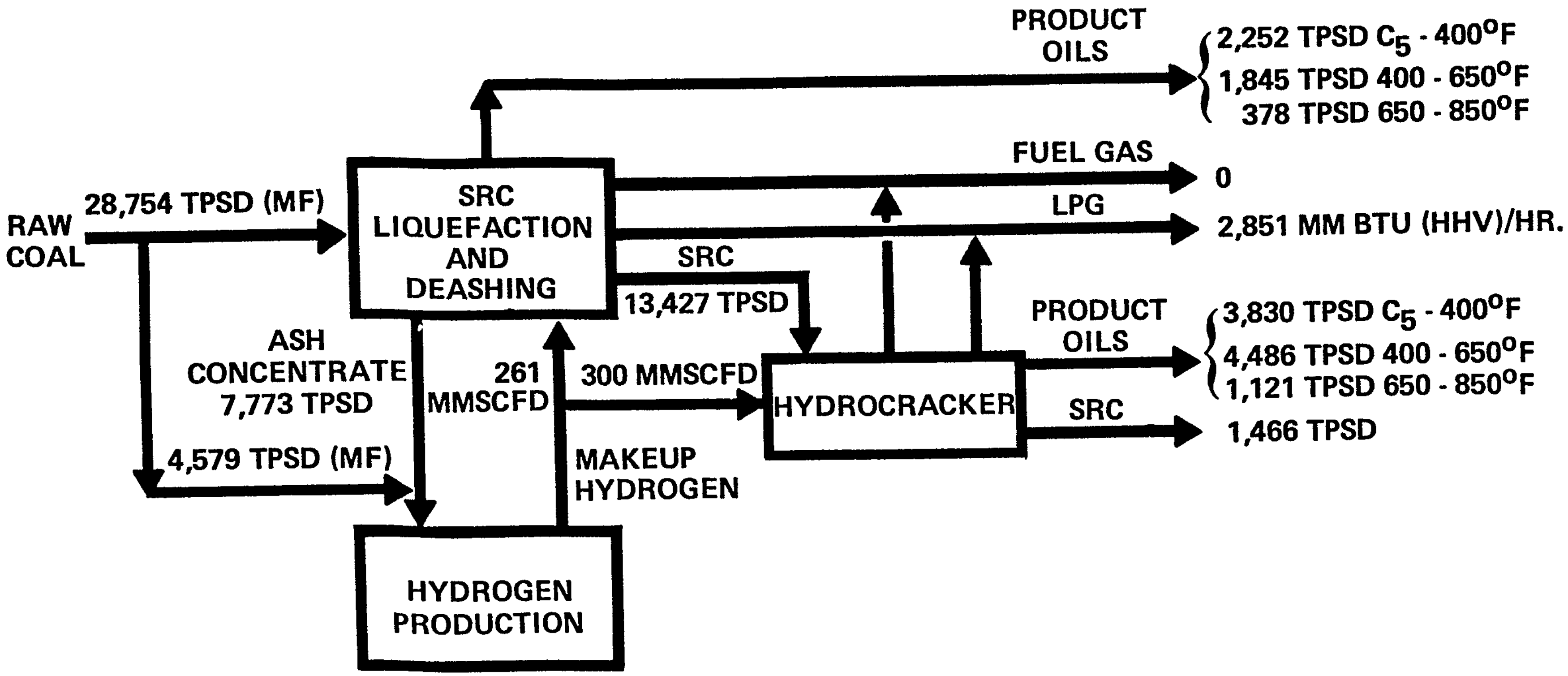
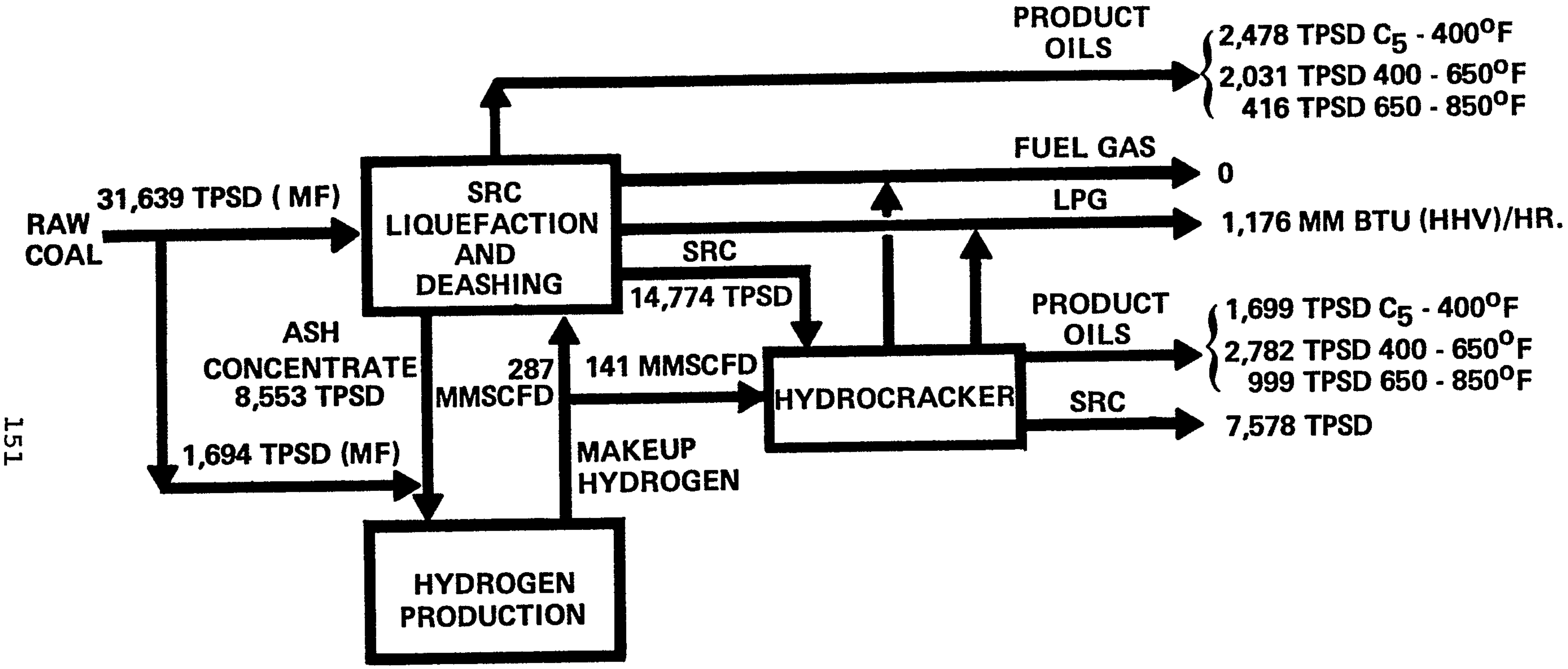


FIGURE V



SRC-I WITH HIGH SEVERITY HYDROCRACKING

FIGURE VI



SRC-1 WITH LOW SEVERITY HYDROCRACKING

References

1. SRC-I Coal Refinery, a report prepared by Southern Company Services, Inc., and Air Products/Wheelabrator-Frye Joint Venture; for the United States Department of Energy (DE-AC05-78-ORO-3054); 31 July 1979.

2. Rappe, G. C., "An Evaluation of Solids Separation Techniques in Coal Liquids," International Conference on Solids Separations in Dublin, Ireland, 17 April 1980.

3. Phillips, E. M., Kuhns, A. R., Johnson, C. A., et al., "A Comparative Study of Coal Liquefaction Performance and Economics for Solvent Refined Coal-Based Processes," 88th National AIChE Meeting, Philadelphia, PA, 10 June 1980.

4. Gasification of Coal Liquefaction Residues from the Wilsonville SRC Pilot Plant Using the Texaco Coal Gasification Process, EPRI Report AF-777, June 1978.

COAL-BASED AMMONIA AND METHANOL MANUFACTURE USING KOPPERS-TOTZEK GASIFICATION

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Introduction

A coal-based ammonia and methanol plant, using Koppers-Totzek gasification, has been operated for over 5 years by AECI Limited in Modderfontein, South Africa. Capacity of the plant is 1 000 metric tons/day of combined ammonia and methanol, and the plant forms part of a chemicals complex producing nitrogen fertilisers and explosives.

Initially reliability of the plant was poor, largely due to process and mechanical problems in the gas processing sections of the plant, as well as the steam-raising units. As these and other problems have been overcome, plant availability and output have greatly improved, and today availability is comparable to that of gas and naphtha-based plants.

This paper provides a description of the process and briefly reviews the experience we have had in operating a modern coal-based ammonia and methanol plant. The Koppers-Totzek gasification process is described in some detail since the objective of this Conference session is primarily to present operating experience with existing commercialised coal gasification technology.

A subject which is receiving considerable attention at this time in relation to coal conversion processes is environmental impacts, and this topic will be considered in relation to the Koppers-Totzek based plant at Modderfontein.

Process Description

Koppers-Totzek gasification

The Koppers-Totzek process is a high temperature, atmospheric pressure, entrained bed gasifier employing pulverised coal and was developed by Friedrich Totzek and his co-workers in Germany between 1936 and 1942. The objective was to develop a process which was not very sensitive to coal properties and would produce a clean synthesis gas consisting mainly of CO and H_2, and containing practically no hydrocarbons, phenols or other similar contaminants.

Pulverised coal is entrained into a stream of premixed oxygen and steam and the reaction mixture enters the gasifier via burner nozzles at either end. Very rapid exothermic reactions occur causing the temperature in the core of the flame to rise to over 2000°C. Subsequent endothermic reactions, and heat loss through the gasifier wall, which is employed to raise steam, cause the temperature to decrease, in this case typically to about 1600°C. Residence time in the gasifier is very short, typically about 0,5 to 1 second.

The coal feedstock for gasification on the Modderfontein ammonia and methanol plant is obtained from collieries situated about 90 km from the plant, and supplied by rail. A typical analysis of the feed coal is given in Table 1. The coal is pulverised and simultaneously dried to about 1,5% moisture in two ring and ball mills; the coal dust particle size is typically 90% less than 90 μm. The pulverised coal is supplied to a system of bunkers, and fed to the gasifiers via screw feeders.

Six two-headed Koppers-Totzek gasifiers are employed, and the coal dust from the screw feeders is entrained by a premixed oxygen/steam flow into the gasifiers. A proportion of the mineral matter in the coal is slagged in the gasifier and impinges on the walls where it accumulates and subsequently flows to a slag outlet in the base and is quenched and granulated in a water bath and removed by a continuous scraper system.

The remaining mineral matter leaves the gasifier mainly in association with unconverted carbon, as fly ash. The exit temperature from the gasifier is about 1600°C and since the mineral matter is largely molten under these conditions the gas is quenched with direct water injection to about 900°C to avoid fouling of heat transfer surfaces with slag. A waste heat boiler system raises steam at 55 bar, after which the gas is contacted with water in washing towers where most of the solids in the gas are removed. The solids are settled from the washing water, thickened to a slurry with about 15% solids and pumped to nearby dams where the major part of the water is recovered and recycled for re-use.

The gas is subjected to further dust removal before passing to raw gas compressors prior to the gas purification plant. A typical composition of raw gas is given in Table 2.

Gas purification and CO shift conversion

A block diagram of the whole process is shown in Figure 1. The dust-free raw gas from the gasification plant is compressed to 30 bar in two parallel steam turbine-driven raw gas compressors. The compressed gas enters the first stage of a two-stage Rectisol gas purification unit.

Before entering the first stage of the Rectisol unit, which is shown schematically in Figure 2, HCN is removed from the gas in a water absorber. The gas is then scrubbed in the H_2S absorber with methanol containing dissolved CO_2, which removes H_2S and COS to a combined concentration of less than 1 ppm, but does not absorb a significant amount of CO_2.

The gas is then compressed to 50 bar, and passes to the CO shift conversion unit where a conventional high temperature shift conversion catalyst is employed, and CO content of the gas is reduced to about 3%. The steam required for shift conversion is supplied by the waste heat boilers on the gasification plant.

The converted gas, after passage through a heat exchanger train, enters the second stage of the Rectisol unit where CO_2 is removed from the gas to less than 10 ppm in a two stage CO_2 absorber.

Methanol containing dissolved H_2S and COS from the H_2S absorber is stripped in two stages in the H_2S rewash column and the H_2S stripper. The overhead product from the H_2S stripper contains 60% H_2S and COS and is suitable for sulphur recovery using, for example, a Claus kiln, or for sulphuric acid manufacture.

Methanol containing dissolved CO_2 is stripped in the CO_2 stripper and a proportion of the CO_2 is recovered as a pure byproduct for use in urea manufacture.

The final gas purification stage, which is shown in Figure 3, involves removal of final traces of CO_2 in two molecular sieve adsorbers, and a liquid nitrogen wash unit to remove final traces of CO and CH_4.

Ammonia and methanol synthesis

The purified gas from the liquid nitrogen wash unit is combined with pure nitrogen from an air separation plant in the stoichiometric ratio for ammonia synthesis. A conventional ammonia synthesis loop operating at 220 bar is employed, and the high degree of purity of the synthesis gas eliminates the need for a voluntary purge to avoid inert accumulation.

An ICI low pressure methanol plant is installed which draws synthesis gas from the gas purification section.

Review of operating experience

The Modderfontein plant was commissioned near the end of 1974 but initially difficulties were encountered in attempting to achieve continuous operation at relatively high production rates. The major causes of breakdowns during the first year of operation were mechanical in nature, and a large number were related to the steam raising plant. Two large spreader-stroker boilers are employed each rated for maximum continuous output of 184 t/h of steam at 100 bar and 510°C. Tube and grate failures were very persistent problems which contributed very significantly to plant shutdowns or rate reductions. These in turn generated mechanical problems throughout the plant due to intermittent operation, startups and emergency shutdowns.

The phase of serious mechanical problems was eventually overcome and during periods of continuous operation a more detailed assessment of the process could be made. Considering the Koppers-Totzek gasifiers it was evident that the units could produce at least the rated output of gas and that the gas composition was very close to that predicted by the designers. Three main difficulties were however identified. These were refractory lining attack, low carbon conversion efficiency and excessive generation of carbonaceous flyash. All three problems were inter-related and were subsequently shown to have been the result of the differences in nature between South African bituminous coal and coals of the Northern Hemisphere.

Extensive experience had been gained in operation of Koppers-Totzek gasifiers on lignite and Northern Hemisphere bituminous coal, but at Modderfontein slag attack of the gasifier refractory linings occurred which necessitated reduction of the operating temperature to preserve the linings as an interim measure until the problem could be investigated. The original feedstock to gasification was shown to be unsuitable due to very low reactivity and poor mineralogical characteristics. This resulted in very poor carbon conversion under conditions where the refractory linings were being protected from excessively high operating temperatures. In addition the poor mineralogical characteristics lead to very little slag formation during gasification which consequently meant that the bulk of the coal ash left the gasifier as fly ash. The fly ash is disposed of in lagoons situated about 1 km from the plant and the excessive quantity of flyash could not be catered for in the lagoons originally constructed for the Modderfontein plant.

After investigation of the lining problems improvements were made in the lining and cooling system of the gasifiers. In addition, a better coal was selected and subsequently shown to have far better mineralogical and petrological properties than the original coal, for use in Koppers-Totzek gasifiers. Even so, the inherent low reactivity of South African coal still resulted in relatively low carbon conversion and a high proportion of flyash.

After resolution of the main process and mechanical problems referred to, performance of the plant has greatly improved and today availability is comparable to that achieved on gas and naphtha-based plants. This statement is significant considering the far greater complexity of the coal-based plant and the fact that a great deal of solids handling is required.

Environmental assessment

As a result of the high gasification temperature the Koppers-Totzek process produces no coal distillation products, and hence aromatics and phenols do not occur in the raw gas or water from the washer-cooler towers after gasification. The only hydrocarbons produced are methane at a level of about 100 ppm, and trace quantities of C_2H_6 and C_3H_8.

Since the raw gas containing H_2S, COS, HCN and NH_3 is contacted with water at various stages of the process, water streams containing these species are produced. All these streams are fed to a common contaminated water system which also treats the fly ash slurry from the washer-cooler towers after the gasification plant. The combined streams pass through a settler/clarifier where ash is separated and the clarified water stream is recycled to the plant. The settled ash slurry is pumped to lagoons where the major part of the water is recovered as runoff. This water represents the only liquid effluent leaving the Modderfontein plant and due to the long contact time with the flyash the dissolved H_2S and HCN are both converted to innocuous species. Cyanide content of the runoff water is typically 0.2 ppm while H_2S is not detectable.

As with liquid effluents, gaseous emissions do not present any problems on the Modderfontein plant. Besides sulphur oxides and particulates from the boiler plant and the flue gas drying circuit of the coal preparation unit, the only sources of gaseous emission are tail gases from the Rectisol and liquid nitrogen wash units. The tail gases are either flared or used as supplementary fuel for the boiler plant. The major proportion of sulphur in the gasifier coal feed appears in a concentrated stream from the H_2S stripper in the Rectisol unit. This stream was originally to be combusted in a pyrite roaster for sulphuric acid production but since the permanent shutdown of the sulphuric acid plant it is now planned to instal a Claus kiln to treat this gas stream.

From the above it can be concluded that no significant environmental problems have been experienced during five years of operation of the Modderfontein plant, and that the Koppers-Totzek process is inherently a coal gasification process characterised by low environmental impact.

Conclusion

In the light of AECI experience the Koppers-Totzek gasification process offers distinct advantages for production of synthesis gas from coal, for applications such as ammonia and methanol manufacture. Production of a "clean" raw gas with no byproducts (hydrocarbons, aromatics or phenols) and minimal environmental impact make it a suitable process for integration into a chemicals manufacturing complex such as Modderfontein.

Although initially plant reliability was poor, availability has continually improved and considering the complexity of a coal-based plant such as this when compared to gas or oil-based plants, the availability currently achieved is highly gratifying. As a fully commercialised process, Koppers-Totzek gasification must be considered as a candidate for production of synthesis gas from coal for projects under evaluation over the next 5 to 10 years.

Table 1

Typical analysis of gasification feed coal

Ultimate analysis (dry basis) % m/m

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

Table 2

Typical analysis of raw gas by volume (dry basis)

CO	58%
H_2	27%
CO_2	12%
CH_4	100 ppm
H_2S	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

FIGURE 1.

No. 4 AMMONIA PLANT - PROCESS SCHEME

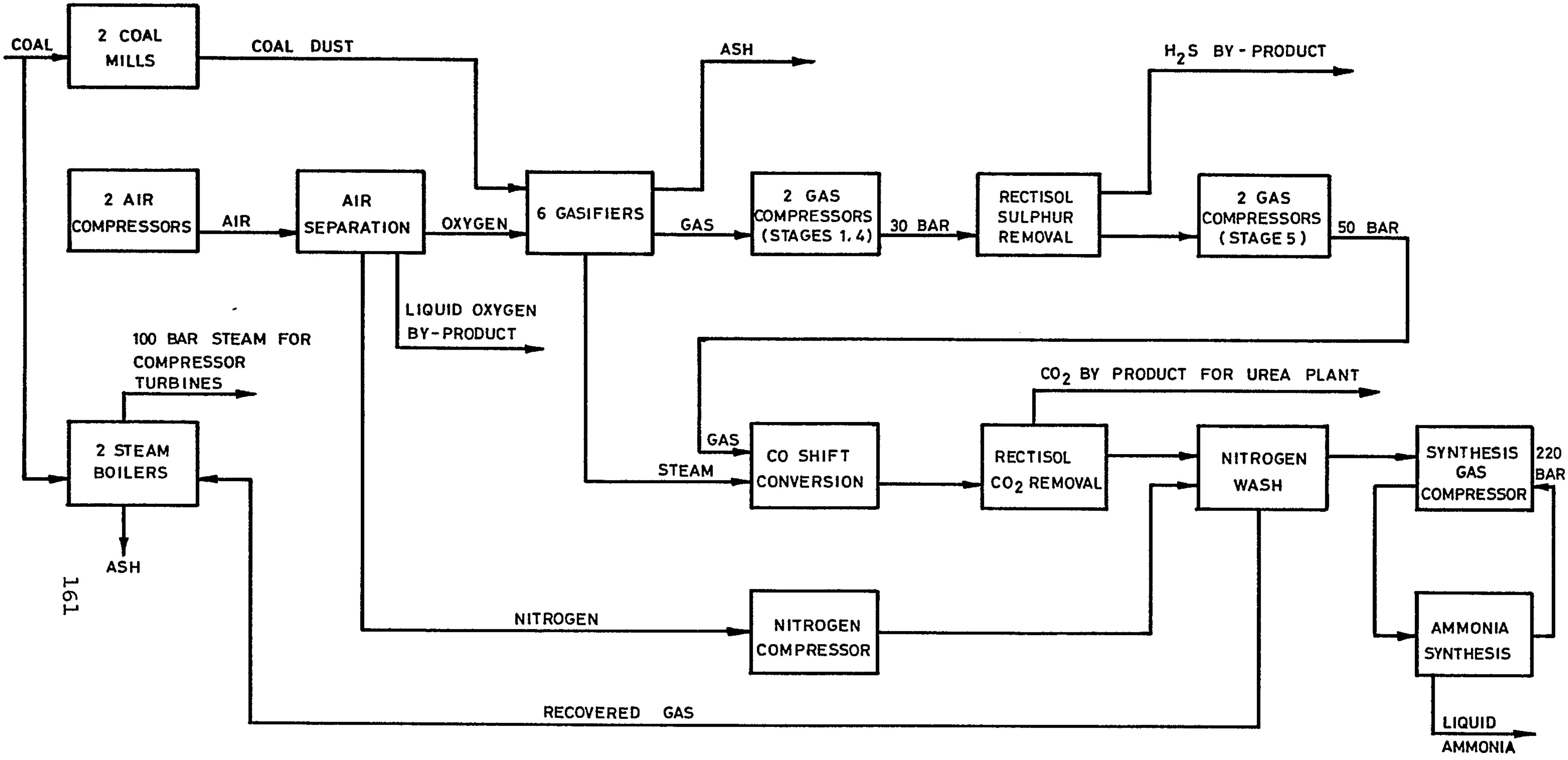


FIGURE 2.

MODDERFONTEIN No. 4 AMMONIA PLANT

SIMPLIFIED SCHEMATIC DIAGRAM OF RECTISOL PROCESS

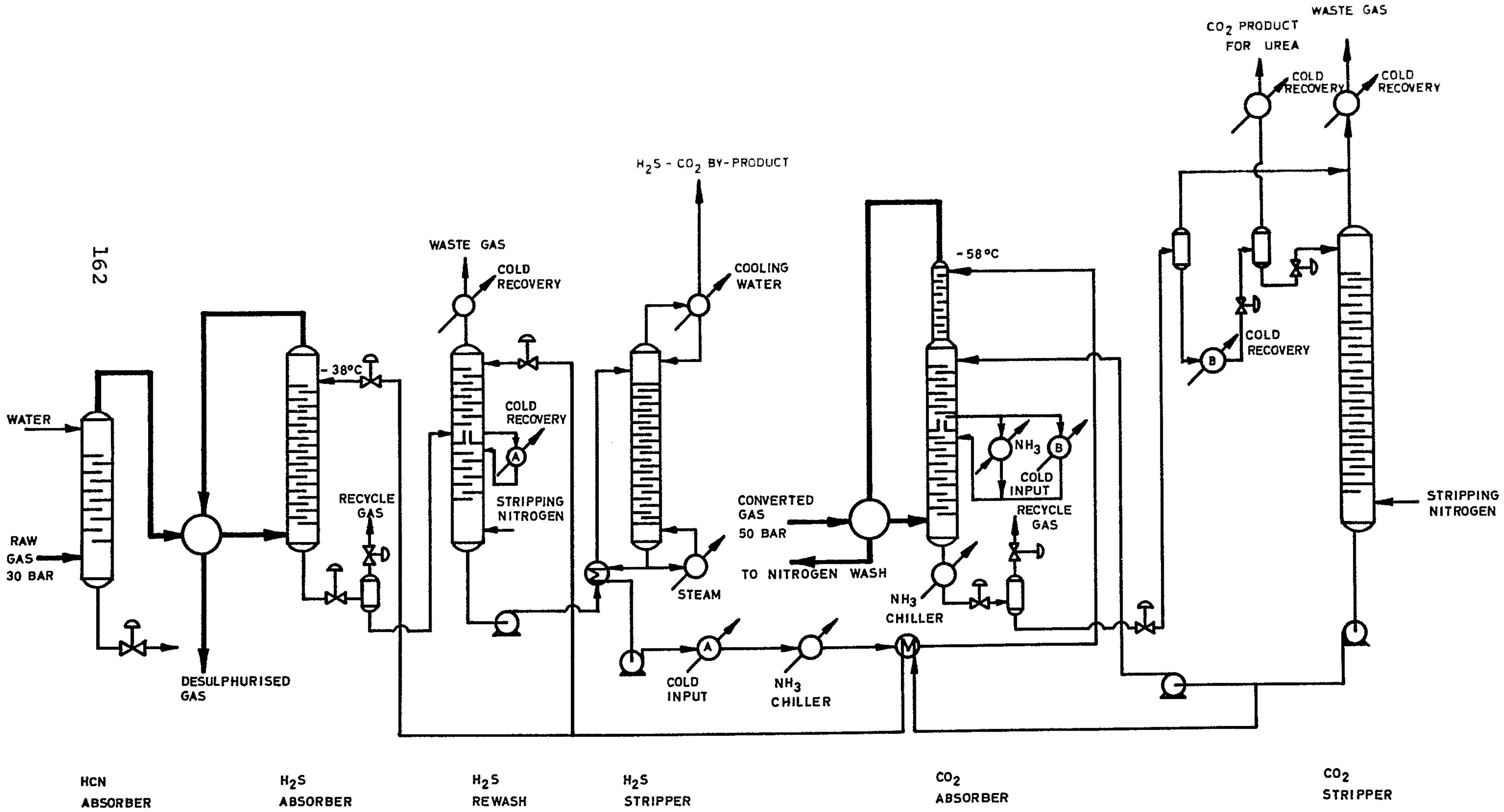
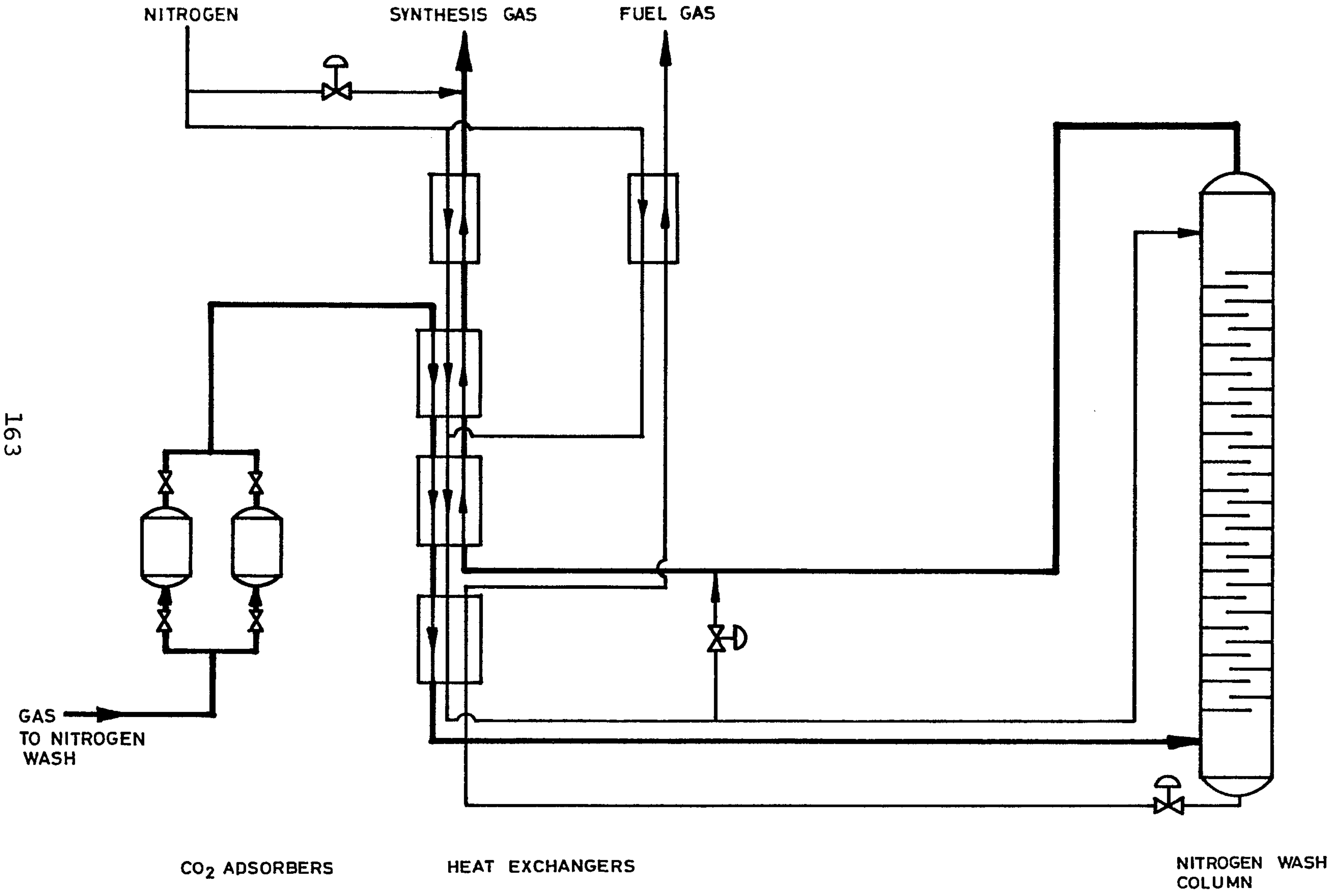


FIGURE 3

SIMPLIFIED SCHEMATIC DIAGRAM OF NITROGEN WASH PROCESS



OPERATING EXPERIENCE GAINED WITH THE "PKM/GSP" COAL FIXED BED PRESSURE GASIFICATION PROCESS

Wieland Schutter
Peter Genauck

OPERATING EXPERIENCE GAINED WITH THE "PKM/GSP" COAL FIXED BED PRESSURE GASIFICATION PROCESS

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1. General Survey

Due to the situation caused by World War II and more particularly by the lack of appreciable resources of bituminous coal and crude oil, the German Democratic Republic was faced with the task of basing the national economy to a great extent on domestic lignite resources.

To this end, experts set to work to develop opencast mining technologies and equipment enabling coal to be produced economically and reliably using a broad range of mining machinery adapted to a great variety of deposit conditions.

Based on this development work, lignite production has been increased to about 250 x 10^6 metric tonnes per annum and will continue to be increased within the near future. At present, about 70% of our country's energy demand are met by this primary source of energy which, as mined, has a calorific value of 1,800 to 2,300 kcal/kg (i.e. 3,240 to 4,140 Btu/lb), with moisture contents ranging from 55 to 60% and ash contents, from 8 to 20%.

As a result of the increase in coal production, power plants are being erected, coal upgrading plants extended, well-known upgrading process improved and new ones introduced.

This energy policy enables a successful development of the entire GDR economy to be accomplished on the basis of domestic fuel resources.

At present, about 70% of the lignite produced is directly supplied for heat and power generation. The remaining 30% is upgraded using the following processes:

- briquetting as a primary stage of thermal and chemical upgrading, or for the supply of briquetted fuel for domestic and industrial consumers;

- lignite high-temperature coking for the production of a high-strength coke for chemical and metallurgical processes, and for the supply, as a smokeless fuel, to domestic and industrial consumers;

- lignite low-temperature carbonization to produce low-temperature tar for use in the carbochemistry, and to obtain low-temperature coke as a feedstock for synthesis gas production;

- fixed-bed pressure gasification with oxygen to produce gases for public supply.

In June 1955, the Government of the GDR decided to erect a lignite upgrading complex in the area of the Lower Lusatian lignite deposits. On August 31, 1955, the first sod was turned for the project of VEB Gaskombinat Schwarze Pumpe. In 1959, the first power plant and a briquetting plant were commissioned, followed by the gasification plant in 1964, and the coking plant in 1969. The entire complex covers an area of 3 x 4 km (about 3,000 acres), and comprises:

3 power plants,

4 briquetting plants,

1 pressure gasification plant and

1 lignite high-temperature coking plant, as well as

large-scale workshops, and

effluent purification and
water treatment facilities.

Fig. 1 shows a general layout of all process lines of the upgrading complex of Schwarze Pumpe.

The labour force of the complex amounts to 13,00 employees.

The amount of lignite processed per day is about 100,000 tonnes, yielding the following main products:

about 15 x 10^6 cu.m. (530 x 10^6 cu.ft.) of medium Btu gas,
about 30,000 tonnes of briquettes, and
about 4,000 tonnes of coke.

The power plants have an installed capacity of 1,200 MW; they also provide steam for various upgrading processes.

Thanks to the possibility of co-generation and the advantages of an interconnected operating of plants the total energy efficiency of the complex is as high as 70%, and this figure is expected to increase by several percent by 1985.

2. Technological Layout of the Schwarze Pumpe Gasification Plant

The design and construction of the lignite-based oxygen fixed-bed pressure gasification plant was undertaken by VEB PKM Anlagenbau in accordance with experience available in this field in the GDR.

VEB PKM Anlagenbau is a division of the Schwarze Pumpe gas combine and is still responsible for the construction and reconstruction of pressure gasworks. This enterprise was, for instance, the contractor for the construction of a pressure gasworks for the gasification of hard brown coal in Yugoslavia on the basis of the know how of Schwarze Pumpe.

The gasification plant of Schwarze Pumpe comprises 15 plant sections which are arranged separately in terms of function as well as space. These 15 sections are operated to cover the four main processes, i.e. production and compression of oxygen, gas manufacture, gas purification, and gas liquor purification.

Fig. 2 shows a flow diagram of the oxygen pressure gasification process.

2.1. Oxygen Production Process

The oxygen plant comprises two units having a total capacity of 70,000 cu.m. of oxygen per hour(normal conditions) (2,470 x 10^3 cu. ft./hr).

An essential feature are the newly developed oxygen turbo-compressors with an output of 36,000 cu.m./hr (normal conditions) (1,270 x 10^3 cu.ft./hr) and an outlet pressure of 27 kp/sq.cm. (397 lb/sq.in.). Within the last few years, research and engineering work has been concentrated on attaining a high quality level in the maintenance and repair of these complex systems, on implementing modifications in construction, and thus on attaining a high degree of safety and reliability.

2.2. Raw Gas Manufacture

The most imposing structure of the gasification plant is the gasifier building, which is an open-air 9,000 tonnes steel structure of 180 m (590 ft) in length and 44 m (144 ft) in height.

This structure contains 24 PKM/GSP type pressure gasifiers with shaft diameters of 3.6 m (11.8ft). These large-size gasifiers are equipped with modern automation devices.

The briquettes for gasification are produced in the combine's briquetting plants. The process of briquette gasification was developed particularly for the purpose of utilizing the entire range of grain sizes (about 40% fine-grain coal) to be found with GDR lignite. Briquetting need not be employed if coals having higher hardness properties are to be used. Also, the intermediate step of briquetting is not necessary with GDR lignite, if lumps prepared by a special process are used.

The raw gas discharged through pressure reduction in the lock hoppers is passed to a gas cooling and purification system via a gasholder. Subsequently, it is recompressed and passed to the low-temperature purification section. A proportion of 8% of the total raw gas output of the gasifiers is produced in this way. This energy gain attained by the recovery of all high-calorific value gases for gasworks gas manufacture is one of the principal features of the plant design.

This process ensures that there are no waste products except ash. All necessary safety requirements are taken into account in the arrangement of the gasifier system. Every four gasifiers are grouped together and are separated from the next group by fire-resisting walls.

Every two gasifiers are connected to a condensation train. Such an arrangement is of advantage in case of breakdowns. Each condensation train comprises five cooling units and has a capacity of 38,000 cu.m. of raw gas per hour (1,340 x 10^3 cu.ft./hr). Cooling water is supplied by the cooling towers of the plant, with the addition of fresh water which is recovered in opencast mines and is treated for the combine's uses.

2.3. Downstream Plants

There have been comprehensive investigations with regard to the selection of a suitable gas purification process. We studied a number of versions and eventually decided to employ the process of low-temperature purification with methanol in view of the high gas quality requirements for underground storage. Four trains having a capacity of 140,000 cu.m./hr (4,950 x 10^3 cu.ft./hr) of raw gas each are provided to clean the gas produced in the pressure gasification and coke-oven plants. Every two trains are operated from an engine house with control room. The expansion gas from purification, having a heat of combustion of 1,000 kcal/cu.m. (112 Btu/cu.ft.), is desulphurized and the sulphur compounds obtained are subjected to the Claus process. Elemental sulphur is recovered with a purity of 99.99%. The desulphurized expansion gas is burnt in the power plant.

The purified producer gas is passed to a blending system which is fully automated and controlled by a process computer, and is capable of blending eight component gases available within the plant to obtain a gas blend with calorific values ranging from 3,800 to 4,000 kcal/cu.m. (425 to 449 Btu/cu.ft.) to be provided as town gas.

The process computer uses mathematical models ensuring an optimum economy and attaining, at the same time, a high consistency of the specified combustion properties of the town gas.

Butyl acetate is used as a solvent for reducing the phenol content of the gas liquor yielded by the process to 30 mg/l. A further decrease of phenol contents is by biological treatment to obtain the value of 0.02 mg/l specified for volatile phenols. Six trains having a throughput capacity of 110 cu.m./hr (3,890 cu.ft./hr) of water each have been commissioned. For gas liquor treatment, novel rotating disk contractors have been developed and put into operation. They are low in energy consumption and ensure a good mixing of gas liquor and butyl acetate.

Liquid products as well as solvents supplied to the system are kept in an intermediate tank storage having a total capacity of 20,000 tonnes.

3. Operating Results Achieved

Gasification tests carried out in commercial-scale units in the GDR using different types of coals have shown the feedstock properties to have a decisive impact on media consumption parameters. (Fig. 3) The results obtained were an essential factor in an optimum planning and design of the gas combine's pressure gasworks.

Over a period of more than 15 years, the results achieved have shown that the PKM/GSP process can be operated highly reliable and economical with repair expenses steadily decreasing. Some consumption and output figures of Gaskombinat Schwarze Pumpe are tabulated in Fig. 4.

Fig. 5 lists the gas composition of the raw gas from pressure gasification. Comparisons made on an international level have indicated that the quality is nearly the same as for gases from sub-bituminous and bituminous coal gasification.

4. Process Development

Evaluating 15 years of commercial-scale operation, the fixed-bed pressure gasification process can be regarded as promising and capable of development. Thus, a further development will focus upon the following main tasks:

- fundamental investigations with a view to gaining a better control of process modelling and gathering further information on feedstock properties and their impact on the process;
- process engineering research to eliminate "bottlenecks" and to improve process parameters;
- development of novel concepts for machinery and for a further reduction of down times and of expenses incurred by wear and repairs.

The objective of these tasks is to further increase the overall process efficiency and economy. In this context, emphasis is on the following projects:

- increase of unit performance;
- increase of the energy efficiency of the entire process through
  - extensive utilization of the waste heat from individual process steps;
  - elimination of process steps;
- rational allocation of manpower through
  - automation of selected process steps (use of microprocessors)
  - improvement of occupational availability of employees
  - introduction of novel maintenance techniques;

- improvement of plant and unit availability through
  - modifications in technology, equipment design and use of materials.

Essentially, the economical results achieved are dependent on construction details and on the overall operating principle of a plant. In this respect, a great number of measures to improve gasifier operation have already proved successful in recent years. In this connection, reference may be made to some concepts covered by other publications:

1. Fully automatic charging system using the concept of electro-hydraulic control and filling level indication by means of radioactive isotopes.
2. Centralized hydraulic system for operating the coal and ash lock hoppers as well as the drive elements of the rotary grates. Use of a fire-resistant hydraulic fluid.
3. A combination of process instrumentation and control equipment in a control centre ensures a reliable performance of all equipment for operation and safety monitoring, and represents a favourable starting point for the introduction of new generations of control and monitoring instruments.
4. Open-air structures are superior to structural arrangements used so far in pressure gasworks.
5. Concepts for dust removal using Venturi scrubbers incorporated in arrangement of gasifier - scurbber cooler-waste heat broiler - raw gas purification.
6. Processing of gas liquors containing dust subsequent to their expansion in large tanks.
7. Control of corrosion of the inner shell of lined gasifiers by using cladded materials.
8. Reduction of wear in piping and elements particularly susceptible to wear.
9. Improvement of the drive elements and supports of rotary grates.

5. Ecological Aspects

As for coal pressure gasification, environmental loads are mainly caused by phenol, ammonia, hydrogen sulphide, sulphur dioxide, and organic vapours, apart from the slag produced and minor dust emissions. Continuous research with a view to reducing emissions of all kinds has made it possible to minimize environmental loads even with increasing plant outputs.

5.1. Gas Liquor Treatment

Subsequent to mechanical pre-treatment the gas liquor is subjected to a pre-purification step using butyl acetate (phenol content after purification: 30 mg/l). By biological treatment the contents of phenol and fatty acids are further reduced to 0.3 to 0.7 g BOD_5/cu.m. The purified effluents are used in dust removal systems, for wet ash removal and spraying of overburden dumps in opencast mines. In these applications, the phenol content is further reduced to 35 mg BOD /l prior to passing the effluents to a receiving watercourse.

5.2. Treatment of Gaseous Emissions

By desulphurizing the gas from coal pressure gasification the environmental load caused by sulphur is considerably reduced when compared to other processes of coal utilization for energy generation (e.g. power plants). The expansion gases from the gas purification plant are desulphurized, and elemental sulphur is recovered in a Claus plant. The desulphurized expansion gases having H_2S contents of 0.13 to 0.25% are burnt in the power plant and in the coke-oven plant.

6. Economic Aspects

A percentage breakdown of the costs of the gas produced is as follows:

Depreciation	11 %
Briquettes	41 %
Steam, oxygen, power and gas purification agents	29 %
Repairs, labour cost etc.	19 %

This tabulation shows that the cost structure of the PKM/GSP pressure gasification process is similar to that of other gasification processes. This fact is supported by fundamental investigations carried out to compare the processes of fixed-bed gasification, fluid-bed gasification and pulverized coal gasification.

These investigations clearly show the high direct dependence of gas costs on feedstock cost. Thus, the economic efficiency of the process is essentially dependent on a supply of the gasification feedstock, i. e. coal, at a low cost and the achievement of high energy efficiencies by an optimum interconnection of all operations involved.

At Gaskombinat Schwarze Pumpe, the production costs of 1 cu.m. of purified gas (normal conditions) (425 Btu) amount to 0.20 Mark. A further cost reduction is likely to be attained by the research activities mentioned. On the other hand, an increase in feedstock costs will have to be taken into account.

In the GDR, natural gas is available at present at an equivalent price of 0.16 Mark per 425 Btu.

7. The PKM/GSP Process and Its Potential Application as a Primary Step of Synthesis Gas Production

The existing version of the PKM/GSP process of gas production with subsequent low-temperature gas purification may also be used as a primary stage for synthesis gas production. In Kosovo, Yugoslavia, for instance, the process described is used in ammonia production. After appropriate conditioning, the gas from pressure gasification may be used, in principle, for all $CO + H_2$ based syntheses (e. g. ammonia synthesis, menthanol production, methanation for SNG manufacture, Fischer-Tropsch synthesis etc.).

Since these synthesis processes are current state of the art, it is possible to employ the PKM/GSP process without any difficulties to obtain products from coal which have been produced so far from oil or natural gas. Thanks to a combination of process steps in terms of energy use, the overall economy and efficiency of these processes would be similar to the data of comparable processes under development, which by the way, would involve high development risks.

8 Summary

Research work concerning the oxygen pressure gasification plant at Gaskombinat Schwarze Pumpe will concentrate particularly on increasing outputs and improving efficiencies.

The upgrading of domestic lignite by means of the oxygen pressure gasification process is one of the main features of the long-term development of the GDR energy policy.

Based on the state achieved in the GDR in the field of science and technology, of planning, management and organization of large-scale pressure gasification plants, there are favourable preconditions for developing projects, in co-operation with parties interested, of further pressure gasworks of the PKM/GSP type.

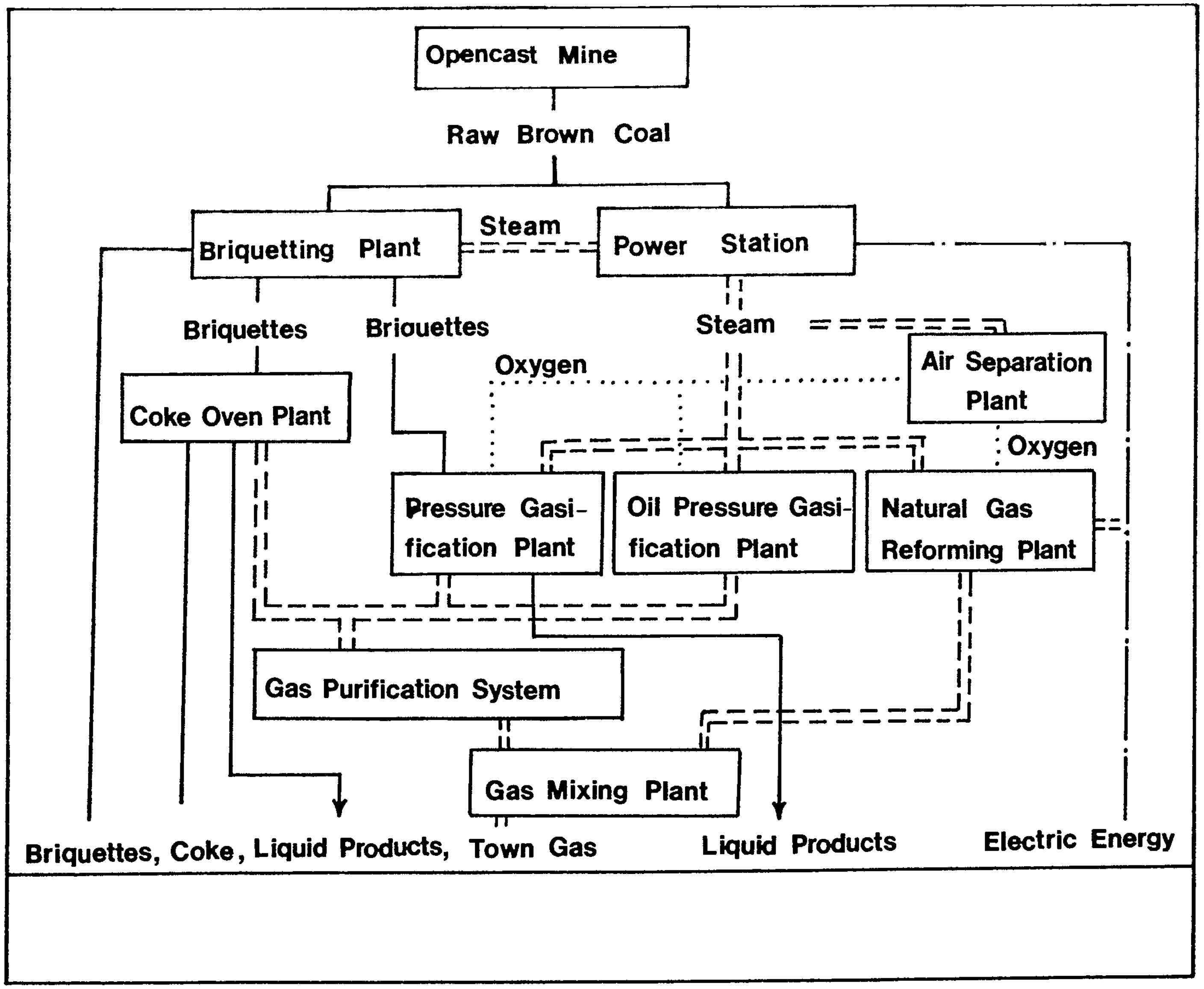

Figure 1 Simplified Flow Sheet of Coal Upgrading System at VEB Gaskombiant Schwarze Pumpe

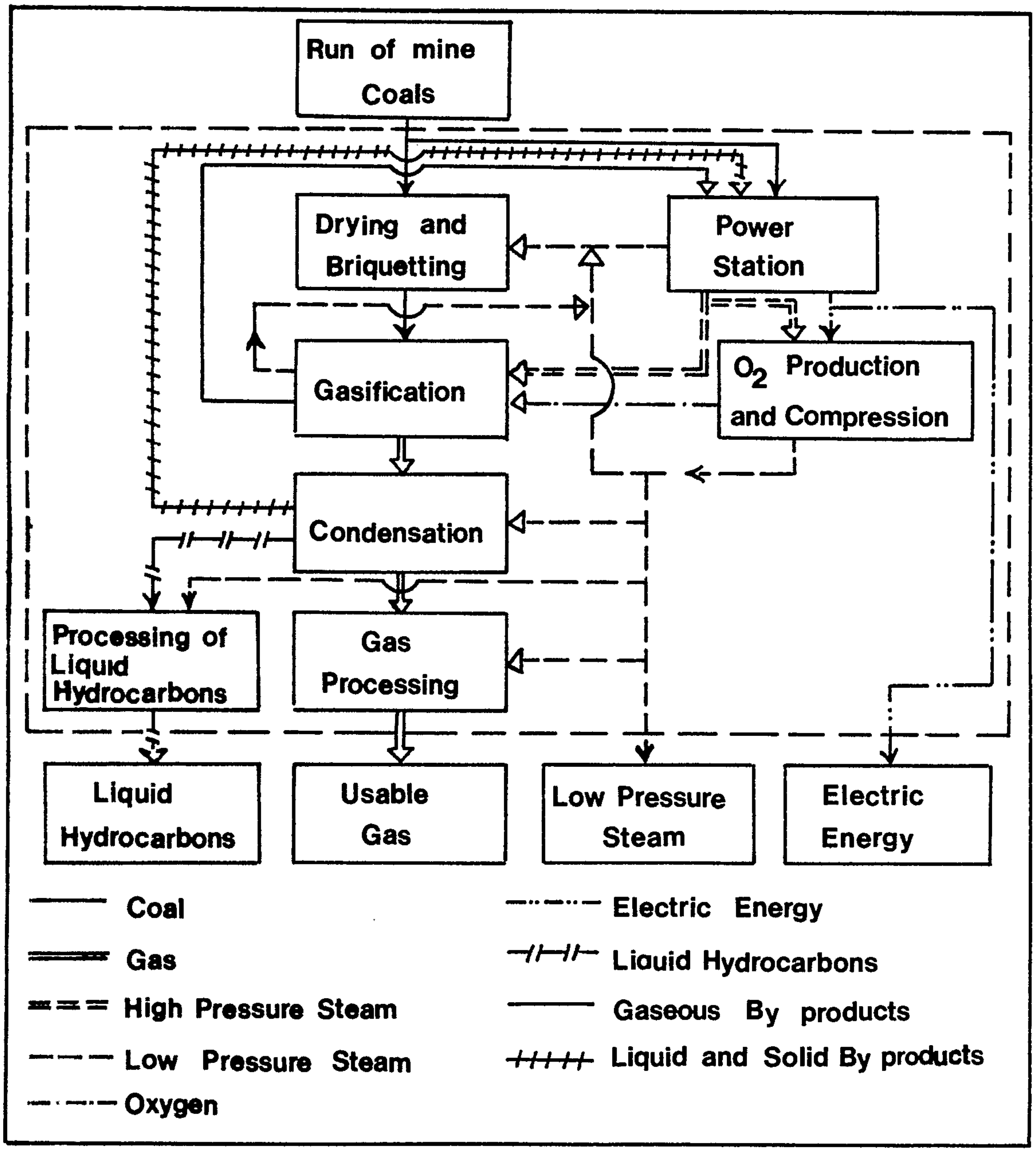

Figure 2 Flow Sheet of Fixed Bed Pressure Gasification

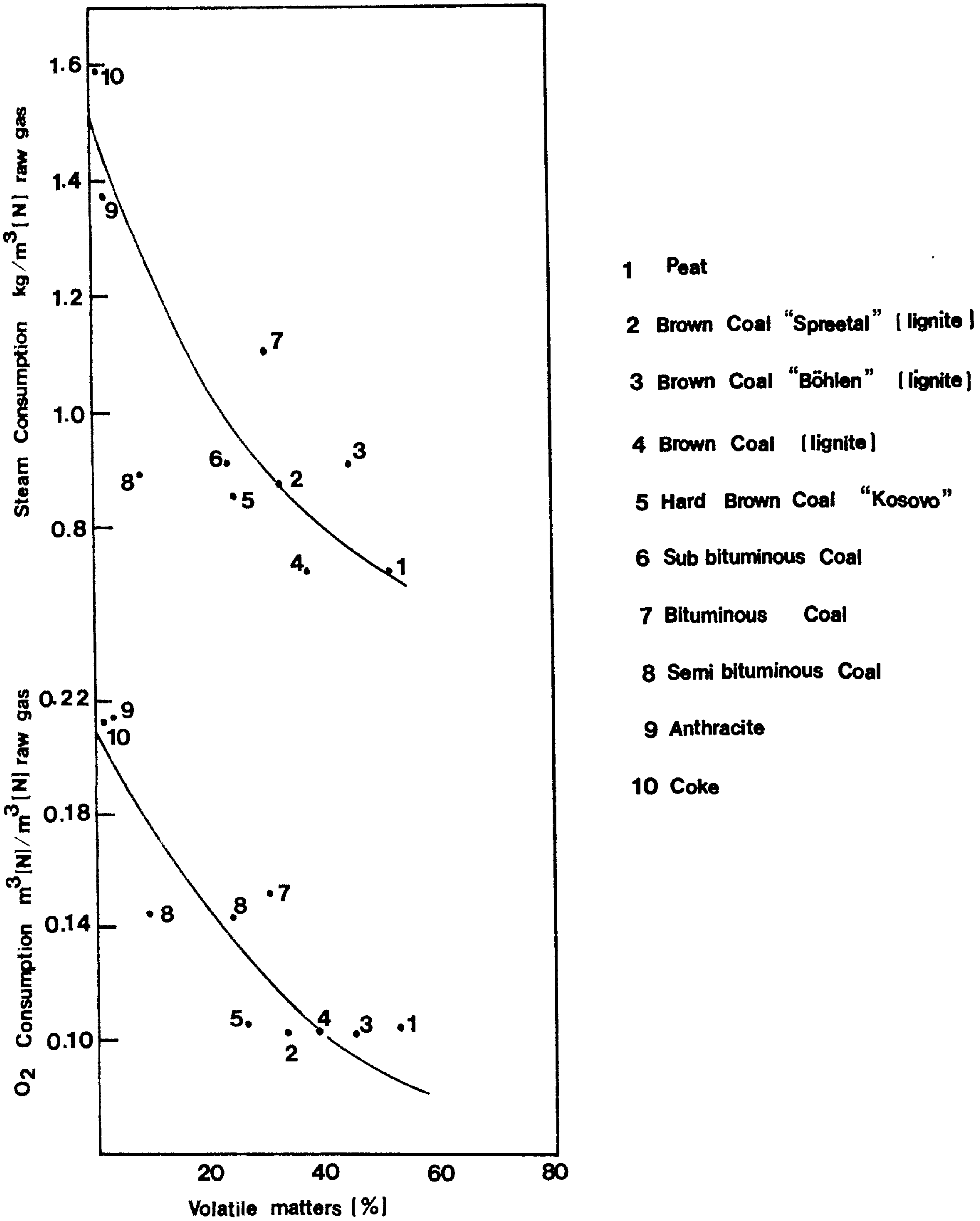

Figure 3 Steam and Oxygen Consumption in dependence of volatile matters of Coal

Oxygen Consumption	0.139	m^3/m^3 Raw Gas
Briquette Consumption	0.91	kg/m^3 Raw Gas
Consumption of Steam for Gasification	1.10	kg/m^3 Raw Gas
Steam Oxygen Ratio	8:1	kg/m^3
Feed Water Consumption	1.10	kg/m^3 Raw Gas
Electric Energy Consumption	16.0	$kWh/10^3\ m^3$ Raw Gas
Yield of Gas Liquor	1.30	kg/m^3 Raw Gas
Specific Shaft Output max	2000	m^3/m^2h
Gas Output	1490	m^3/t daf

Related to Normal Conditions

Figure 4 Consumption and Output Data of the Oxygen Pressure Gasification Plant of Gaskombinat Schwarze Pumpe

COMPONENTS	RAW GAS FROM COAL PRESSURE GASIFICATION	PURIFIED GAS AFTER LOW TEMPERATURE PURIFICATION
CO_2	36.7	2.0
C_nH_m	0.5	0.1
CO	8.6	18.0
O_2	0.2	0.1
CH_4	12.2	18.2
H_2	40.3	55.5
N_2	1.7	6.1

Figure 5 RAW GASES SUBJECTED TO LOW TEMPERATURE PURIFICATION

DEVELOPMENT OF THE SHELL-KOPPERS
COAL GASIFICATION PROCESS

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DEVELOPMENT OF THE SHELL-KOPPERS COAL GASIFICATION PROCESS

7th Annual International Conference
Coal Gasification, Liquefaction & Conversion to
Electricity
Pittsburgh, August 5-7, 1980, United States of America

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SUMMARY

The Shell-Koppers process for the gasification of coal under pressure, based on the principles of entrained bed technology, is characterized by:

- practically complete gasification of virtually all solid fuels;
- production of a clean gas without byproducts;
- high throughput;
- high thermal efficiency and efficient heat recovery;
- environmental acceptability.

There are numerous possible future applications for this process. The gas produced (93-98% vol. hydrogen and carbon monoxide) is suitable for the manufacture of hydrogen or reducing gas and, with further processing substitute natural gas (SNG). Moreover, the gas can be used for the synthesis of ammonia, methanol and liquid hydrocarbons.

Another possible application of this process is as an integral part of a combined-cycle power station featuring both gas and steam turbines. The integration of a Shell-Koppers coal gasifier with a combined-cycle power station will allow of electricity generation at 42-45% efficiency for a wide range of feed coals.

The development programme includes the operation of a 150 t/d gasifier at Deutsche Shell's Harburg refinery since November 1978 (Fig. 1) and of a 6 t/d pilot plant at Royal Dutch Shell's Amsterdam laboratories from December 1976 onwards. Both facilities run very successfully. With hard coal a conversion of 99% is reached whilst producing a gas with only 1% vol. CO_2.

The next step will be the construction and operation of one or two 1000 t/d prototype plants which are scheduled for commissioning in 1984/85. Towards the end of the eighties large commercial units with a capacity of 2500 t/d are contemplated. The economy, especially of these large-size units, is very competitive.

DEVELOPMENT STRATEGY OF SHELL-KOPPERS PROCESS

Shell (SIPM) have been working since 1973 on the development of a high-pressure slagging coal gasification process using their experience with and know-how on the high-pressure oil gasification process.

The firm of Koppers (now Krupp-Koppers GmbH) contributed to the development with know-how from their atmospheric Koppers-Totzek process, and by acting as the main engineering and construction contractor for the 150 tonnes per day plant built in Harburg.

The overall strategy for developing the Shell-Koppers process comprises four phases (Fig. 2):

Phase 1	: Construction/Operation of Pilot Plant in Amsterdam Capacity : 6 t/d coal intake
Phase 2	: Construction/Operation of second Pilot Plant in Harburg Capacity : 150 t/d
Phase 3	: Construction/Operation of Prototype Plants Capacity : 1000 t/d
Phase 4	: Construction/Operation of Commercial Plants Capacity : 2000 - 3000 t/d

The development to commercial application is a long term activity requiring considerable financial and manpower effort. To date, more than $ 100 million has been invested (including the construction of the two Pilot Plants).

SHELL-KOPPERS PROCESS DESCRIPTION

The Shell-Koppers process is based on the principle of entrained bed gasification at elevated pressures under slagging conditions. A general flow scheme is given in Fig. 3.

The coal feed is ground to a size of less than 100 microns, and is normally dried to a water content of about 2%. The dry coal is then pressurized in a lock-hopper system and introduced into the gasification reactor together with oxygen and steam.

The reactor is basically an empty vessel, providing a residence time of a few seconds at a pressure of about 30 bar. Flame temperatures can be up to $2000^{o}C$, but the reactor outlet temperature is normally of the order of $1500^{o}C$.

Under these operating conditions the coal is virtually completely gasified without the formation of any tars, phenols or other condensate hydrocarbons. Overall carbon conversion is about 99%.

The high reactor temperatures also cause most of the ash to melt and to flow down the reactor wall into a water-filled compartment. The remainder of the ash leaves the reactor with the product gas flow. In order to solidify the entrained ash droplets before entering the waste heat boiler, the gas is cooled to about 900°C. Depending on the specific product gas application, this is done either by recycling cold gas or by quenching with water. In the waste heat boiler high-quality steam is raised, which is normally used for driving the oxygen plant compressors.

Following the waste heat boiler, the solids are removed from the gas via an integrated system including cyclone and scrubbers. This allows of the removal of solids in a dry form. If so required, these solids (ash plus some unconverted carbon) may be recycled to the reactor.

Leaving the solids removal system, the product gas has the following typical composition for a hard coal feedstock:

H_2O	2%
H_2	28.5%
CO	65.5%
CO_2	1.5%
CH_4	0.1%
H_2S	1.4%
N_2, A	1.0%

In addition, it contains traces of COS, HCN and NH_3. Total gas production is about 2000 Nm3 per tonne of coal feed, and the gas has a lower heating value of about 2700 tcal/t. The cold gas efficiency is about 80%. Depending on the ultimate use of this gas (fuel gas, synthesis gas) further gas treating will be required, for which various, commercially available, processes can be applied.

Some characteristic features of the Shell-Koppers process are:

(a) Complete Conversion of any Coal

The process is suitable for the complete gasification of a wide variety of solid fuels, such as all types of coal and petroleum coke. Fuels with a high ash content (up to 40% wt) and high sulphur content (up to 8% wt) can be used in the Shell-Koppers gasification plant without any trouble. Even a high water content in the coal does not pose a technical problem. However, on economic grounds, as will be shown in the next section, it is advantageous to dry the coal to a moisture content of 1-6% wt.

Since the process in any case requires a solid fuel to be in dust form for gasification, the entire output of a mine, including fines, is acceptable as feed. Unlike fixed-bed or fluidized-bed processes, in the Shell-Koppers process there is practically no limitation on the coal as to ash fusion behaviour or caking properties.

(b) Clean Gas Production without Formation of Byproducts

The operation at very high temperatures ensures the formation of a high quality synthesis gas essentially consisting of hydrogen and carbon monoxide (93-98% vol. for oxygen gasification). Tars, phenols and other byproducts are absent; as a rule methane concentrations in the gas do not exceed 0.2% vol. The low CO_2 concentration (typically 1-2% vol.) facilitates gas treating.

(c) Large Unit Capacity

Both the high temperatures of above 1400°C and the high pressures are responsible for the high capacities attainable. Short-term targets are 50-100 t/h of coal per reactor, corresponding to 2.4-4.8 million Nm3/day of raw gas.

Depending on the application, the optimum pressure level can be selected. Apart from the beneficial effect of the elevated pressure on reactor capacity,there are spin-offs in terms of increased heat transfer rates in the waste heat boiler, easier gas treating and a reduction in gas compression costs.

(d) High Thermal Efficiency and Efficient Heat Recovery

The chemically bound heat in the gas produced with oxygen gasification is equivalent to about 79 to 82% of the chemically bound heat contained in the coal feed. The recovery of the sensible heat from the hot gases leaving the reactor accounts for another 12 to 15% of the heat content of the coal feed. The surplus steam produced by this cooling is generally sufficient to drive the compressors of the oxygen plant.

(e) Environmental acceptability

A negligible environmental impact can be expected from the Shell-Koppers process. This is the consequence of:

- the clean raw gas produced
- the high thermal efficiency
- the low waste water production
- the production of non-leachable and inert slag

SHELL-KOPPERS DRY FEED SYSTEM

One of the characteristic features of the Shell-Koppers process is its dry feed system. The coal is ground and dried to a moisture content of 2%, pressurized in a lock-hopper system and pneumatically fed to the reactor. The lock-hopper system is cyclic, and requires the frequent opening and closing of valves in a dust environment.
An alternative would be a "wet" system. A coal/water mixture could be made pumpable, and be compressed and transported directly into the reactor.

A comparison between a "dry" and a "wet feed system for the Shell-Koppers system is presented in Tables 1, 2 and 3. The calculations have been made with the SIPM-developed mathematical model for entrained bed gasification. This model has proved to be a valuable tool in various design studies. Its reliability for this type of exercise has been verified by comparing the mathematical model results with actual plant measurements in Harburg.

All calculations are based on the standard Shell-Koppers process scheme, with as the only difference the moisture/water content in the feed stream to the gasifier:

Case 1	: Standard Shell-Koppers "dry" feed system
Case 2	: As Case 1, but coal is not dried (moisture content of "as received" coal is 16.5%)
Case 3, 4 and 5	: Water is added to "as received" coal: 20%, 40%, 80% respectively.

Table 1 gives the main results. It shows for increasing water content:

- a lower syngas production (H_2 + CO)
- a higher steam production
- a higher specific oxygen consumption.

In the standard Shell-Koppers scheme, a high-pressure steam is generated which is mainly used to supply power to the various consumers (the main one is the oxygen plant). If there is not enough high-pressure steam available, a coal-fired auxiliary boiler is assumed; if there is a steam surplus, this is considered as an export product. Heat for coal drying (Case 1 only) is provided by an extra amount of import coal.

Table 2 presents the consequences for the overall efficiency. It shows that for Case 1 (dried coal) 4.6% extra coal is required, partly for coal drying and partly to provide extra power. For Case 5 (80% water added), 4.5% of the incoming energy is exported in the form of high-pressure steam. Table 2 clearly illustrates the reduction of net efficiency for increasing water content.

In the standard Shell-Koppers process, the raw gas is cooled to its dewpoint in the solids removal system. Subsequent cooling (if so required) is by direct air cooling. For a "wet" feed system, with its relatively high raw gas dewpoint temperature, it might be advantageous, dependent on the process application, to use the low level heat for process preheating or for raising low-pressure steam. This would increase capital costs but improve overall efficiency. If such heat recovery schemes were applied, the net efficiency differences between "dry" and "wet" feed systems would be reduced to about half of those listed in Table 2.

Table 3 is indicative of the consequences on plant investment costs. It gives the relative throughput of the main plant systems for plants producing the same quantity of synthesis gas.

Particularly significant is the increase in the required oxygen plant capacity. It reflects the combined effect of:

(1) the higher coal consumption for the same quantity of synthesis gas, and

(2) the higher oxygen consumption for the same quantity of coal feed.

A third aspect of a "dry" versus a "wet" system is the product gas composition (Table 1). For "wet" systems the CO_2 content rises sharply. Even if the CO_2 itself does not have to be removed (as in most fuelgas applicatons), it is a disadvantage, as it significantly increases the costs of H_2S removal by requiring an extra enrichment stage before the sulphur recovery unit.

For synthesis gas applications, requiring a CO-shift, a somewhat higher H_2/CO is beneficial. In addition, in the "wet" feed case, the raw gas already contains part of the steam required for the shift. If a sulphur-resistant shift is applied, this has a favourable effect on the shift economics. On the other hand, the high CO_2 content remains a disadvantage and, more generally, introducing water into the gasifier is a very expensive way of raising steam for the shift reaction.

Taking the above three aspects together (net efficiency, plant investment, product quality) it is evident that a "dry" feed system offers remarkable advantages.

The dry feed system with lockhoppers has performed completely satisfactorily in both the Amsterdam and Harburg plant.

In an effort to develop further the concept of dry pressurizing and dry feeding, SIPM are seriously studying the possibilities of a continuous dry pressurizing system as an alternative to the cyclic lockhopper system.

CURRENT DEVELOPMENT STATUS

The Pilot Plant (built under Phase 1 of the programme) has been in operation at Shell's Amsterdam Laboratory since December, 1976. To date the plant has successfully operated for over 4000 hours on a number of feedstocks, including various hard coals, German brown coal and coke ex Athabasca tarsands.

The present function of the pilot plant is to widen the range of feedstocks tested, to continue collecting fundamental process data and to test new equipment.

Besides the operation of the pilot plant, extensive research activities, directly related to the gasification project, continue at the Amsterdam Laboratories. These include:

- the operation of component test facilities, e.g. for burners, feeder systems and valves
- the development of advanced measurement and control techniques
- reactor model studies (both mathematical and physical).

The Pilot Plant of 150 t/d capacity (built under Phase 2 of the programme) started operation by the end of 1978 at Deutsche Shell's Harburg refinery. The plant has now completed 750 running hours, with a longest uninterrupted run of 240 hours.

A summary of the operational experience is given in Table 4.

The main objectives of the plant are:

- to confirm the operational results of the 6 t/d Pilot Plant on a larger scale
- to develop scale rules for scale-up to Prototype and Commercial plants
- to implement new technical developments and to collect component reliability data
- to collect environmental impact data.

Surveying the total operating experience in the two plants in Amsterdam and Harburg, it may be concluded that the basic concept of the Shell-Koppers process has been demonstrated. Important in this respect are the following:

- a conversion of 99% is obtained
- CO_2 content in the product gas can be maintained between 1 and 2%
- reactor conditions (temperature) can be controlled accurately.

On this basis, Phase 3 of the development programme was initiated early in 1980. This phase calls for the construction and operation of Prototype Plants of a capacity of about 1000 t/d of coal each, to be completed in 1984/1985. The first of these will be erected at Moerdijk (The Netherlands). and will produce fuel gas for a combined cycle power station of a local utility company. A second plant, the location of which has not been decided yet, will probably produce synthesis gas for methanol production.

Basically, the Prototype Plants will be scaled-up versions of the Harburg Pilot Plant, but the design will be such that technical innovations, currently being investigated in the two existing plants, can be accommodated.

PROSPECTS OF THE SHELL-KOPPERS PROCESS

It is now envisaged that, following the successful operation of the two Prototype Plants, the first Commercial Plant will come on line in the second half of the eighties. Apart from the direct use as fuel gas producer, there is a whole range of further applications for the Shell-Koppers process. Three applications are listed below:

(a) In combination with combined cycle power generation

This may become the first type of commercial application and for that reason has been taken as the basis for the first 1000 t/d Prototype Plant. A block scheme is given in Fig.4. In this scheme the gasification and combined cycle station are kept separate. Full integration, in which the high-pressure steam systems of the gasifier and the combined cycle station are combined, is also possible. Overall efficiency of such a system, using gas turbines with inlet temperatures of about 1200°C, is in the range of 42-45%.

(b) Production of hydrogen-rich synthesis gas for methanol and Fischer-Tropsch type syntheses

This type of application has been chosen as the basis for the second Prototype plant. Two possible flow schemes are presented in Fig. 5 and Fig. 6. The optimum route is dependent on many parameters, including the composition of the feedstock, the availability and cost of utilities, etc. Detailed design optimization studies are at present being carried out in this field.

(c) Production of hydrogen in coal liquefaction schemes

This application follows the same scheme as under (b), but includes a deeper CO shift plus a methanation step to remove the remaining CO. In direct liquefaction processes, this scheme may be used to convert the bottom products into hydrogen which in turn is used for liquid upgrading.

A similar feedstock is the fluid coke from the Canadian Athabasca tarsands. This feedstock has already been successfully gasified in Amsterdam.

ECONOMICS

The investment for a 36 x 10^6 Nm3/day synthesis gas plant, based on US location mid-1980, is estimated at $ 1 billion. This figure applies to the processing of a 10% ash, 10% moisture hard coal and includes the investment of coal handling and storage, coal mill and dryer, oxygen plant, water treatment, ash disposal facilities and off-sites.

Excluded is the investment in plants for the further processing of the dry, particulate and sulphur-free synthesis gas.

The corresponding cost of the synthesis gas before and after conversion into methyl fuel and subsequently into gasoline on a heating value basis is shown as a function of the coal feed unit cost in Fig. 7.

For a low-cost coal feed ($ 1/MMBTU), as would be available from open-cast mining of large coal reserves, the synthesis gas cost would amount to some $ 3/MMBTU. For deep-mined coal ($ 2.5/MMBTU), as available in Europe, the cost could be as high as $ 5/MMBTU.

The corresponding figures for methyl fuel are $ 6.5 and $ 9.5/MMBTU and for gasoline $ 6-8 and $ 9-11/MMBTU respectively. The economics of Shell-Koppers coal gasification are only marginally affected by variations in coal rank, ash content or moisture content. Only the cost of the coal mill and dryer and of the ash disposal facilities are significantly influenced, but these have a minor impact on the overall economics.

TABLE 1

COMPARISON OF "DRY" AND "WET" FEED SYSTEM

MAIN GASIFICATION DATA

		Coal dried to 2%	Coal as received	water added to coal		
				20%	40%	80%
Coal (A.R.) to process	(kg)	1000	1000	1000	1000	1000
to auxiliaries	(kg)	45	15	0	0	0
Moisture removed	(kg)	145	-	-	-	-
Water added	(kg)	-	-	200	400	800
Oxygen to Gasifier	(kg)	700	750	810	880	1025
Steam to Gasifier	(kg)	15	-	-	-	-
O_2/MAF Coal Ratio	(-)	0.92	0.99	1.07	1.16	1.35
H.P. Steam produced	(kg)	1155	1320	1510	1735	2180
Raw Gas produced	(Nm3)	1610	1775	2020	2270	2770
(H_2 + CO) produced	(Nm3)	1520	1450	1365	1270	1070
Raw Gas Composition	(% vol)					
H_2O		1.7	9.0	18.6	27.3	41.4
H_2		28.7	27.8	26.0	23.3	18.1
CO		65.6	54.0	41.7	32.5	20.5
CO_2		1.6	7.0	11.8	15.1	18.5
H_2S		1.5	1.4	1.2	1.1	0.9
N_2 + A		0.9	0.8	0.7	0.7	0.6

1) Coal: Illinois # 6; 16.5% moisture, 7.6% ash (A.R.)

TABLE 2

COMPARISON OF "DRY" AND "WET" FEED SYSTEM

NET PLANT EFFICIENCY

	Coal dried to 2%	Coal as received	water added to coal		
			20%	40%	80%
Coal (A.R.) to process	100	100	100	100	100
Raw product gas	81.2	77.3	72.5	67.1	56.6
H.P. steam produced	16.0	18.3	20.9	24.1	30.2
Own energy requirement					
Steam to Gasifier	0.2	-	-	-	-
Electric Power	18.5	19.6	20.9	22.5	25.7
Heat for drying	1.9	-	-	-	-
Total	20.6	19.6	20.9	22.5	25.7
Total coal in	104.6	101.3	100	100	100
Total raw gas out	81.2	77.3	72.5	67.1	56.6
Total H.P. steam out	0	0	0	1.6	4.5
Net Efficiency	77.6	76.3	72.5	68.7	61.1

1) Heat content of coal (A.R.) to process is set at 100

2) Heat content is based on net heat of combustion

3) Raw gas at dewpoint temperature; latent heat not included

4) Heat content of "power" is based on heat content of equivalent amount of coal required to raise power

5) Heat content of "steam" is based on heat content of equivalent amount of coal required to raise steam.

TABLE 3

COMPARISON OF "DRY" AND "WET" FEED SYSTEM

INDICATION OF MAIN PLANT SYSTEM CAPACITIES

	Coal dried to 2%	Coal as received	water added to coal		
			20%	40%	80%
Syngas Production (Nm3 H_2 + CO)	100	100	100	100	100
Total Coal (A.R.) (incl. Coal to Auxiliaries)	100	102	105	115	135
Feed to Gasifier (inc. transport medium)	100	120	150	185	285
Oxygen	100	110	130	150	210
H.P. Steam	100	120	145	180	270
Raw Product Gas	100	115	140	170	245

1) Individual system throughput for Case 1 (Dried Coal) is set at 100.

TABLE 4

SHELL-KOPPERS 150 t/d PILOT PLANT

SUMMARY OF OPERATING EXPERIENCE

Operating Hours	750 Hours
Longest Uninterrupted Run	240 Hours
Pressure	19 Bar
Temperature	1500 °C
Coal Feed Rate	4.5 t/h
Oxygen/Coal Ratio	1.05 Kg/kg MAF
Steam/Coal Ratio	0
Raw Gas/Coal Ratio	2.1 Nm3/kg
Carbon Conversion	99%
Gas Composition:	
H_2	25.6 % vol.
CO	65.1
CO_2	0.8
H_2S	0.4
N_2 + A	8.1