2.2.3 Ruhrkohle AG/Veba Oel AG Coal Hydrogenation Plant A,C,C-1,D,G

The catalytic hydrogenation process is a modified version of the old German (Bergius-Pier) hydrogenation technique. The modified solids separation technique allows a solids-free distillate recycle oil for slurry preparation, thus permitting the application of moderate reaction conditions and/or a higher specific coal throughput. The plant design is based on the experimental data produced by Bergbau-Forschung.

2.2.3.1 History and Status[17,18]

In November 1977, the decision was made to construct a 200 t/d plant for catalytic coal hydrogenation. The plant was constructed in Bottrop close to a coking plant. Gas, water, steam and electricity are supplied by the coking plant and hydrogen is taken from a pipeline passing the site.

The basic engineering was carried out by Ruhrkohle AG and Veba Oel AG. Project management was also performed by both companies.

The site for the pilot plant was officially inaugurated on May 21, 1979 by the Minister of Economics of the State of North Rhine-Westphalia. The plant was completed in early 1981 and start-up operations began in July1981.

The investments for the 200 t/d pilot plant amount to $73 million. The operating costs for a three-year demonstration period are approximately $73 million. Furthermore, a six-year program of lab-scale tests at Bergbau-Forschung GmbH to characterize the product and improve the process details, budgeted at $13 million, is underway. The Bergbau-Forschung program also includes product upgrading studies.

The overall expenses of $160 million are largely sponsored by the Ministry of Economics of the State of North Rhine-Westphalia. [Although the cost of the Bottrop 200 ton/day pilot plant has been published as 300 million DM (about $170 million), current estimates place it at about 400 million DM ($230 million). Plant construction is said to have amounted to about 200 million DM ($115 million) with 200 million DM for operation of the plant.] The number of personnel required ranges between 180-250 in the first year of operation and between 170-220 in the second and the following years of operation.

The Bottrop plant has an overall process flow identical to the Bergbau-Forschung unit except that the plant will be equipped with special systems for heat recovery and for hot separator let-down processing. The solids residue will be continuously granulated for storage and not gasified on site. Large scale gasification and carbonization tests are planned to be carried out in existing gasification plant by contracts.

The Bergbau-Forschung experimental plant has been operating for almost three years. Total on-line time as of May 1979 was about 13,500 hours.

2.2.3.2 Company Experience[19,20]

The 200 t/d pilot plant design is based on the experimental unit (0.5 t/d) operated by Bergbau-Forschung, a mining research institute which cooperates closely with Ruhrkohle AG (RAG). One of their interests is in coal gasification and they are also pioneering work on a 100-atmosphere Lurgi Coal Gasification Process (the Ruhr 100 gasifier) at Dorsten. Ruhrkohle, through its subsidiary Ruhrchemie, is involved in the Texaco gasifier project at Oberhausen-Holten. It is likely that residue gasification tests will be conducted there. They also have an interest in two plants generating power from coal. Veba Oel AG is the petroleum processing part of a large German industrial group, also holding chemical and power production companies.

Since 1974 Ruhrkohle AG was a partner of Gulf in the now-discontinued SRC-II project and since 1979 a partner of Exxon in the development of the EDS process. The catalytic coal hydrogenation technology developed in Germany uses the knowledge of former years in the field of coal hydrogenation recorded partly at BASF, UK-Wessling and Veba AG.

In the area of studies, Ruhrkohle participated in the 8 million DM ($5 million) Imhausen study (see Sect. 1.1 and Appendix A for more details) which examined a Bottrop-type plant for the Australian government for the conversion of 7 million tonnes/yr (8 million tons/yr) of Australian brown coal to 3 million tonnes/yr (3.3 million tons/yr) of motor fuels. The cost of the plant would be about 7 billion DM (about $4 billion) and a Texaco gasifier would probably be used to process the vacuum bottoms. Timing on this proposal is not known.

2.2.3.3 Plant location

The 0.5 t/d experimental unit is located in Essen and the 200 t/d pilot plant is located in Bottrop, both in Ruhr district. The site of the pilot plant was officially inaugurated on May 21, 1979 by the Minister of Economics of the Land Northrhine-Westphalia.

2.2.3.4 Process flow for Bottrop Plant[17,18]

The Bottrop plant flow diagram is identical to that of the Bergbau-Forschung experimental unit (Fig. 2.10).

Design of the plant is based on the I. G. Farben process with the following modifications: (a) a solvent consisting of a distillate of middle and heavy oil is fed with coal and catalyst instead of the formerly used centrifuge overflow; (b) the residue of hydrogenation is used for hydrogen generation; and (c) the reacted solid-oil mixture is separated by distillation instead of centrifugation. Owing to these

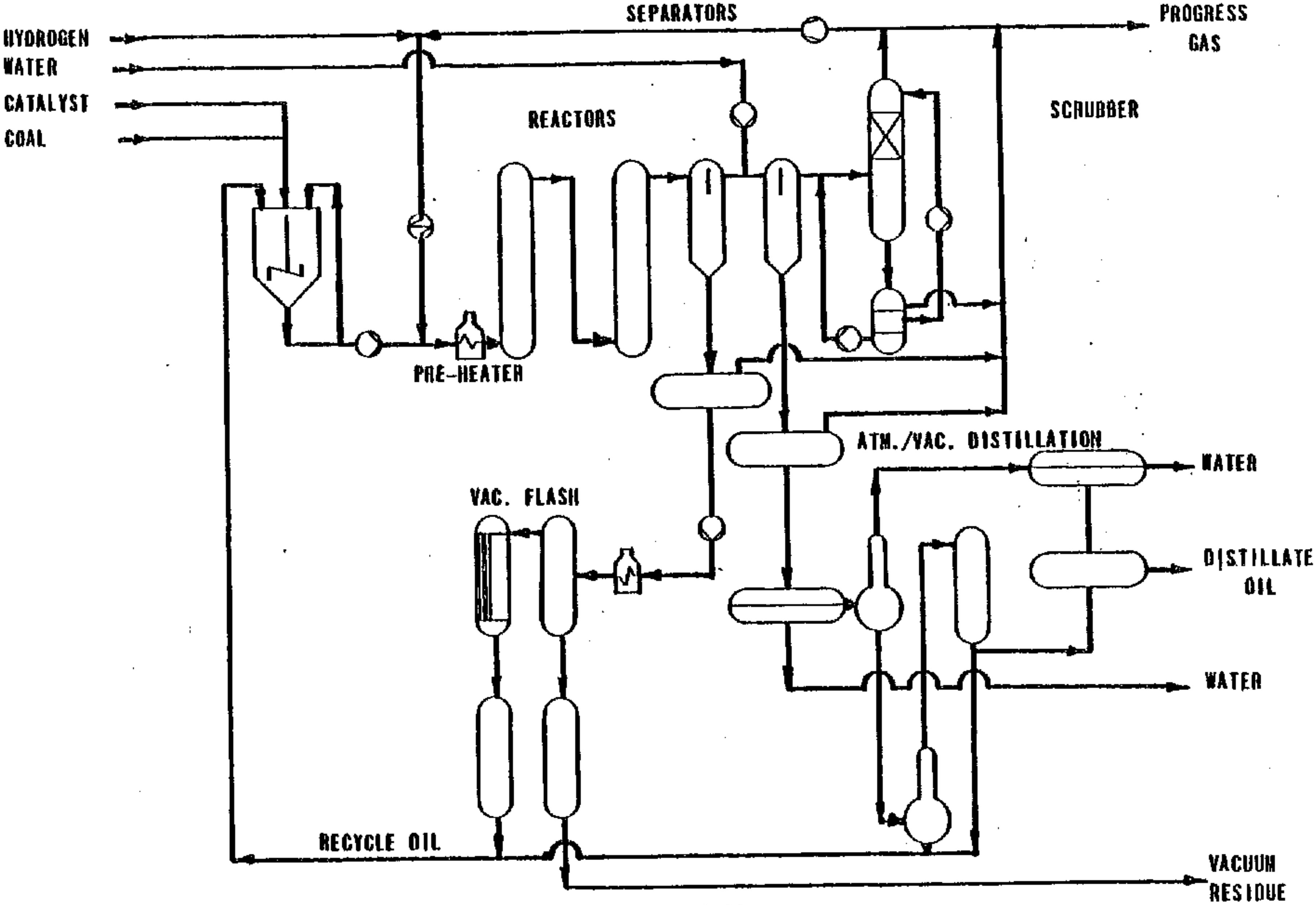


Figure 2.10

BERGBAU-FORSCHUNG EXPERIMENTAL COAL LIQUEFACTION PILOT PLANT process modifications the following improvements are expected: (a) a drop in pressure from 700 to 300 bar (10,500 to 4500 psi); (b) an increase in specific coal throughput of 50%; (c) an improvement in heat recovery; (d) an increase in the thermal efficiency of 25%; and (e) a decrease in the specific conversion costs.

The engineering was carried out by Ruhrkohle AG, Veba Oel AG and other German engineering companies.

Figure 2.11 is a block flow diagram of the 200 t/d pilot plant at Bottrop. In the coal preparation unit the feed coal is dried to a water content less than 1 wt % and ground to a size smaller than 1 mm (0.04 in.). The prepared coal is stored in a bin. After slurrying with a process-derived solvent consisting of a mixture of middle and heavy oil in a ratio of 40:60 solvent: coal by weight, the coal is ground in a wet mill to a size smaller than 0.2 mm (0.008 in.) whereby catalyst is added.

In the hydrogenation unit, which consists of a high pressure pump, a preheater, three series-connected reactors, a hot separator and a separator vessel, the reaction with hydrogen takes place with makeup hydrogen and recycle hydrogen. The makeup hydrogen is supplied by a hydrogen pipeline of the Chemische Werke Huls AG passing by the site.

The coal-derived distillate oil is separated into heavy, middle and light oil in an atmospheric distillation unit. The light oil is stabilized. The bottom product of the hot separator is flashed and separated in a vacuum distillation unit into distillate oil and residue. The residue containing ash, unreacted coal and catalyst solids is further processed to a granulate. In the gas-treatment unit the gas is compressed and recycled to the process. The raw gas is delivered for further processing to the cokery.

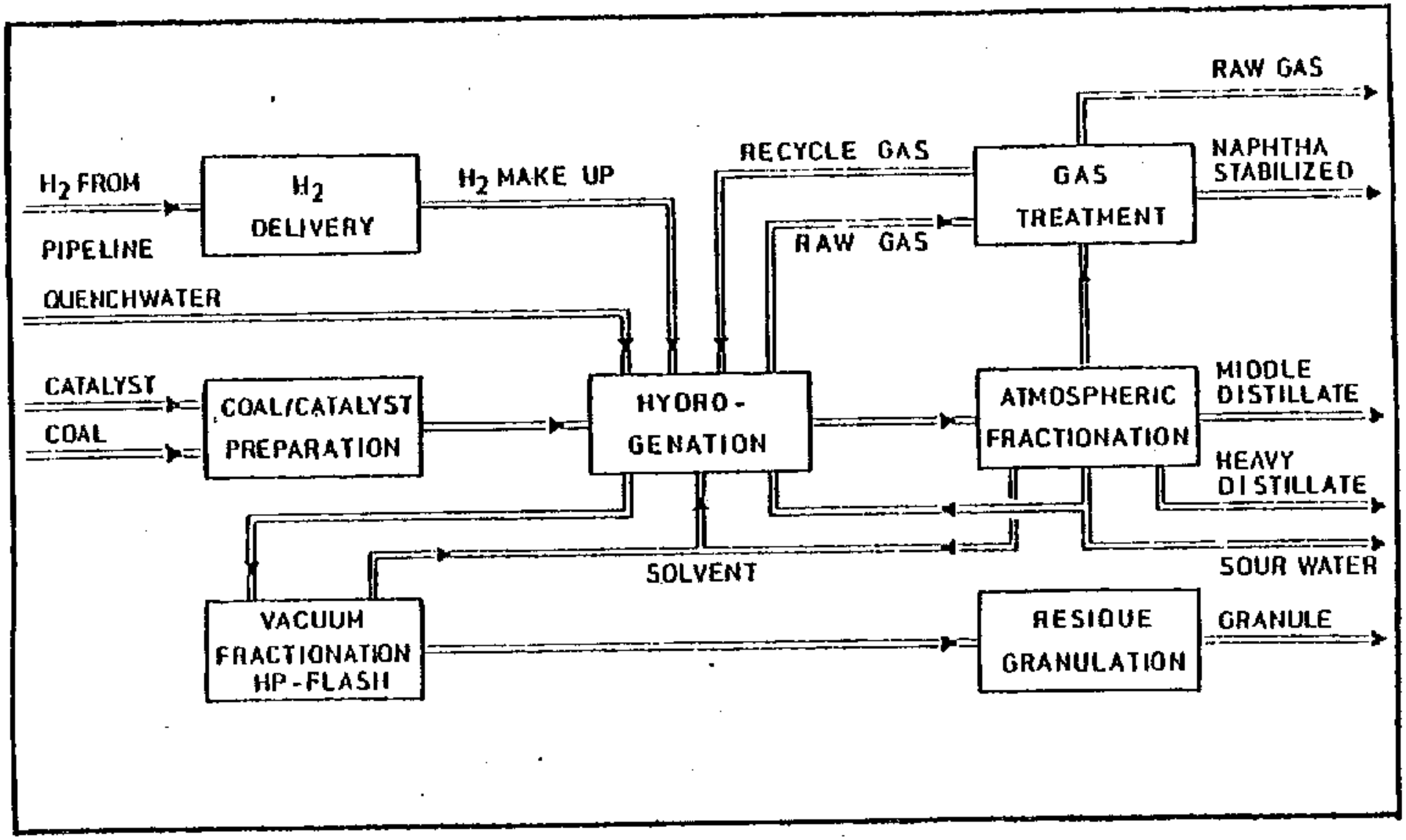


Figure 2.11. Block flow diagram of the 200 t/d Bottrop pilot plant.

Table 2.10 shows the scaleup of the fluidic conditions of the Bergbau-Forschung test reactor to the respective reactor designs. For purposes of simplification the fluidic conditions are defined as the product of the kinematic viscosity and the Reynolds number (ν x Re) of the hydrogenation feeds. The dimenions of the respective reactors are shown as diameter d and height h in the first two columns. The following three columns show the fluidic conditions of the series-connected reactors I, II, and III. The scaleup of the fluidic conditions of the pilot plant to be erected in Bottrop is identical in size with the reactor design of the 1930's, commercially proven in Scholven. $^{C-1}$

Table 2.10 Product of viscosity and Reynolds number (ν x Re) of the reactor feed from plants with different size reactors $^{C-1}$

Size of reactor	d m(ft)	h m(ft)	Reactor I ν x Re_I m^2/s (ft^2/s)	Reactor II ν x Re_{II} m^2/s (ft^2/s)	Reactor III ν x Re_{III} m^2/s (ft^2/s)	Scale-up factor
Technical plant (Scholven)	1.05 (3.4)	18.0 (59)	0.0978 (1.05)	0.1061 (1.14)	0.1116 (1.20)	175
Pilot plant (Bottrop)	0.8 (2.6)	12.0 (39)	0.0686 (0.74)	0.0950 (1.02)	0.1127 (1.21)	176
Prototype plant	2.4 (7.9)	18.0 (59)	0.2928 (3.15)	0.4058 (4.37)	0.4370 (6.70)	688
Bergbau-Forschung BmbH Laboratory	0.09 (0.3)	1.77 (5.8)			0.000639 (0.007)	1

As shown in the plot plan (Fig. 2.12), the 200 t/d pilot plant requires a total area of 58,700 square meters (632,000 sq ft = 15 acres) being 273 meters (900 ft) long and 215 meters (700 ft) wide. The main process units are located in four process areas, including the control room. They require 13,000 sq meters (140,000 sq ft) with a length of 100 meters (330 ft) and a breadth of 130 meters (425 ft). This is a quarter of the totally required area.[A]

The main process units are connected with one another by pipe racks. Roads around the process areas make delivery by heavy trucks possible.

A conveyor belt bridge connects the pilot plant with the cokery. Via this conveyor belt the feed coal is delivered from the cokery and the residue is carried back to the cokery.

The flash and blow-down system is installed within the plant borders. There are also tanks for the coal oil-fractions as well as office, change-room, warehouse and maintenance buildings.

There will be three slurry feed pumps installed in the pilot plant: two operating and one spare. All three pumps are driven by electric motors. These pumps have three pistons working in parallel. For test purposes two of these pumps will work with the pistons in a vertical position; the third pump will have the pistons in a horizontal position.[D]

A remote valve box is connected to the pump via a column of clean oil. Due to the pump action, some of the oil is lost in the valve box. This lost volume of oil is replaced after each stroke with clean oil. Through the design of the pump with an external valve box, an abrasion-free section of the pump piston is expected. In the valve box there are inlet and outlet valves of different design and materials in order to investigate their performance in regard to corrosion and erosion.

There will be two different preheaters installed in the pilot plant at Bottrop. Only one at a time is needed for the operation of the plant. Each heater has enough capacity to handle the design capacity of the plant. The first preheater is very similar to the ones used for hydrogenation of coal in Germany until 1945.

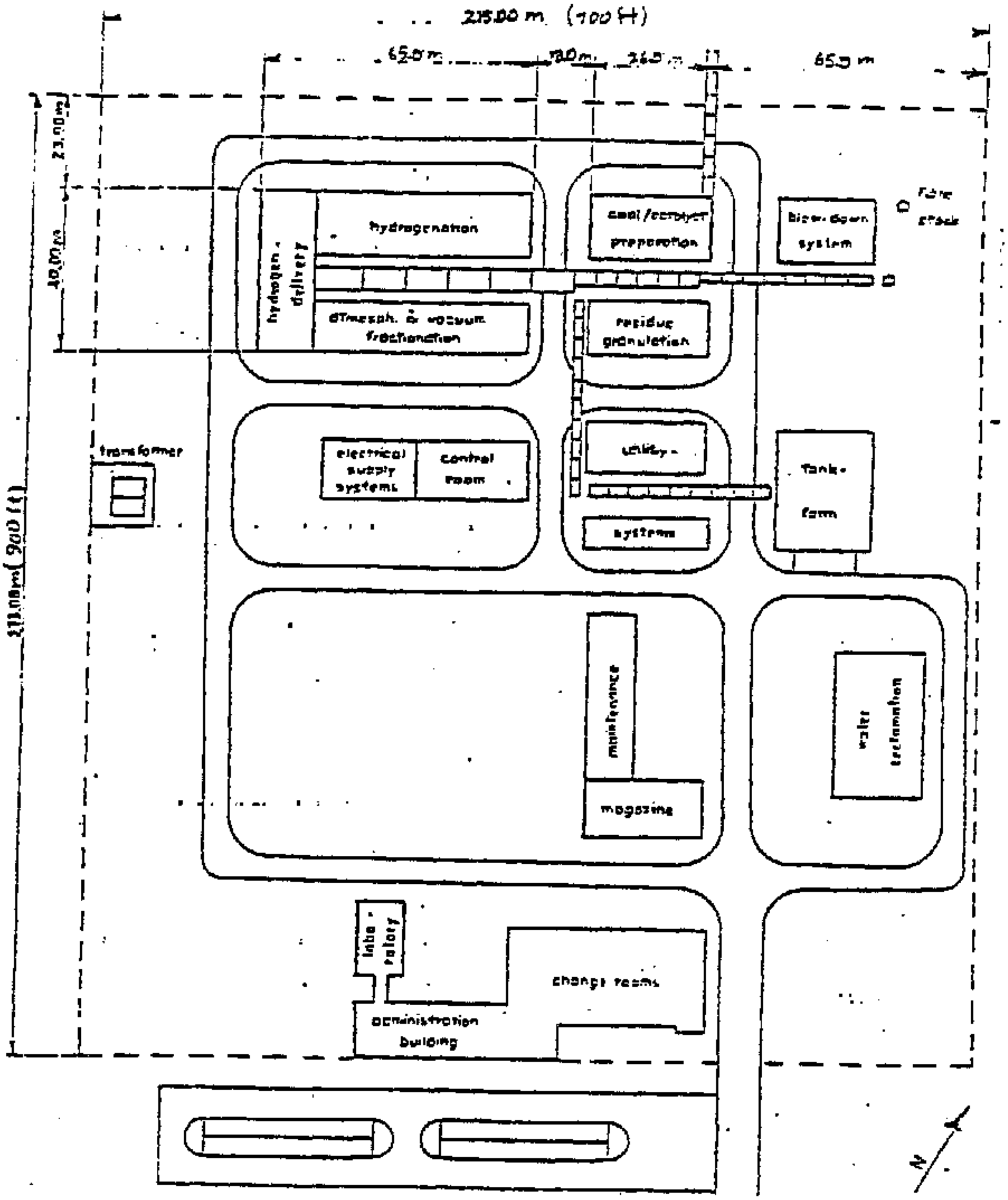


Fig. 2.12. Plot plan for the 200 t/d Bottrop pilot plant.D

In this type of preheater the heat transfer is achieved by circulating hot air thorugh the shell and the slurry through the tubes. The heat transfer in this heater is very gradual, which will prevent the slurry from coking in the tubes. The disadvantages of this heater are its size and the high cost.

The second preheater installed has a radiation furnace of a similar type frequently used in oil refineries. Because of the higher temperatures the risk of coking is also greater. Minimizing this risk while utilizing the better heat transfer characteristics of a radiation furnace, its smaller size and its lower cost is the aim of this equipment evaluation test.

Almost all reactors used for coal hydrogenation until 1945 were made from forgings. The reactors used in the pilot plant in Bottrop are of a new multilayer design. For insulation purposes a layer of refractory bricks is used. In order to protect the refractory bricks from abrasion by the slurry a stainless steel liner is installed.[D]

A new type of reactor is presently in the design stage. This type of reactor will also use the multilayer design but without refractory bricks inside. The insulation will be integrated inside the multilayer wall. The performance of this reactor will be tested in the pilot plant in Bottrop. After that the reactor will be removed from the plant and cut up for testing.

Several different options for vacuum distillation will be available during the pilot plant operation.[N]

2.2.3.5. Results[17]. Table 2.11 shows the specification of feed coals of the pilot plant for standard operation as well as the range due to the flexibility of the plant. This shows the range of utilization of different coals for hydrogenation in the pilot plant. Table 2.12 shows the feed and projected products of the pilot plant for the standard case. These data were estimated from experiments made with standard coal at Bergbau-Forschung in Essen.

The range of yields of middle oil and residue depends on the enrichment of solids in the residue as well as on the composition of the solvent.

Table 2.11 Specification of feed coal[A]

		Standard case	Flexibility
Mositure	wt % (raw)	4.30	3.8 - 14.4
Ash	wt % (m.f.)	4.52	3.0 - 17.0
Volatile	wt % (m.a.f.)	37.9	10.0 - 53.8
Carbon	wt % (m.a.f.)	84.30	75.0 - 93.0
Hydrogen	wt % (m.a.f.)	4.96	3.5 - 7.0
Oxygen	wt % (m.a.f.)	7.23	1.5 - 17.8
Nitrogen	wt % (m.a.f.)	1.68	1.0 - 1.9
Sulfur	wt % (m.a.f.)	1.67	0.5 - 5.4
Chlorine	wt % (m.a.f.)	0.16	0.0 - 0.4
Vitrinite	vol %	64	61 - 77
Exinite	vol %	17	5 - 20
Inertinite	vol %	14	1 - 14
Minerals	vol %	5	1 - 9
SiO_2	wt %	32.92	32.11 - 64.8
Fe_2O_3	wt %	32.20	3.71 - 36.00
Al_2O_3	wt %	27.63	18.48 - 34.60
Grindability	°H	52	>30
Higher heating value[a]	kJ/kg (m.a.f.)	34,600	31,500-36,800
	Btu/lb	14,787	13,540-15,820

[a] Calculated by Dulong.

Table 2.12. Feed and projected products of Bottrop pilot plant (standard case)

	Feed	
Coal, m.a.f.	t/d	200
Hydrogen H_2, makeup	m^3/d (ft^3/d)	220,000 ($7.8x10^6$)
Process water	t/d	41.2
Catalyst (Fe_2O_3)	t/d	4.0
Power	kWh/d	108,000
	Projected Products	
Gas	t/d	61.3
Naphtha (stab.) I.B.P. 200°C (390°F)	t/d	24.4
Middle oil 200-325°C (390-620°F)	t/d	69.0-74.7
Heavy oil >325°C (620°F)	t/d	-
Residue	t/d	68.6-74.3

Table 2.13 shows specifications of crude and refined coal oil fractions. The crude fractions contain high contents of sulfur, nitrogen and oxygen which have to be removed by refining in order to use the products as gasoline, diesel fuel, heating fuels and raw materials for chemicals. The following steps of upgrading are planned for the PDU: refining, hydrocracking, reforming, extraction, and steam cracking.

It is expected that motor fuel from coal can be produced with a thermal efficiency of approximately 55%. The production costs of fuel from coal are determined by the size and the lifetime of a plant. Depending on the chosen premises the costs of fuel from coal will vary. For a plant size with a fuel production of 400,000 tons per year and a service time of ten years, for example, the conversion costs (1979) were estimated to be 0.30 DM/1 ($0.65/gal). For a larger plant with 2 million tons of products per year and a service time of 20 years, the costs were estimated to be 0.16 DM/1 ($0.35/gal).

The production costs of fuel at the larger plant are at 0.63 DM/1 ($1.36/gal) based on German hard coal. On the basis of imported US coal the costs are lowered to 0.51 DM/1 ($1.10/gal).[A]

Table 2.13 Specifications of crude and refined coal-oil fractions[A]

Specifications	Light oil		Middle oil	
	Crude	Refined	Crude	Refined
Gravity at 15°C g/cm^3	0.865	0.827	0.990	0.993
Sp. g. (59°F)	0.865	0.827	0.990	0.993
Aromat. C, wt %	46	31.6	64	50.0
Naphth. C, wt %	10	27	28	31.3
Paraff. C, wt %	43	41.4	28	18.7
Carbon, wt %	85.25	87.80	87.40	88.5
Hydrogen, wt %	11.15	12.25	9.10	10.75
Sulfur, wt %	146 ppm	2 ppm	0.60	0.23
Nitrogen, wt %	0.24	2 ppm	0.60	0.23
Oxygen, wt %	3.5	0.1	3.0	0.4
Base value, mg NH_3/l	1100	3	8800	2900
g/cc	1.1x10^{-3}	3x10^{-6}	8.8x10^{-3}	2.99x10^{-3}
CFPP, °C (°F)			-26 (-15)	-47 (-53)
Flash point, °C (°F)			93	67
Viscosity, mm^2/S			3.1	2.1(50°C)(12°F)
Centipoise			3.1	2.1
Heating value, kJ/kg	41,000	43,000	38,500	40,900
Btu/lb	17,625	8,485	16,550	17,585
Fuel value, kJ/kg	43,000	46,000	40,500	43,250
Btu/lb	1,455	19,775	17,410	18,595
Phenols, wt %	15.4		16.2	0.29
Bases, wt %	0.25		6.5	2.32
Benzene, wt %		3.9		
Toluene, wt %		5.3		
Ethyl benzene, wt %		2.0		
Xylene, wt %		4.4		
ROZ (clear)		79.2		
Boiling range				
Initial boiling point, °C(°F)	76 (169)	43 (109)	212 (414)	169 (336)
10 vol %, °C(°F)	102 (216)	81 (178)	225 (437)	205 (401)
50 vol %, °C(°F)	158 (316)	149(300)	253 (487)	244 (471)
End point, °C(°F)	206 (403)	211(412)	324 (615)	303 (579)

The 200 tonne/day (210 ton/day) pilot plant at Bottrop will eventually be scaled up to a prototype-scale plant having a capacity of about 5000 tonnes (5500 tons) per day of coal.[N] The prototype will include two 2500 tonne (2750 ton) per day trains and would be used to demonstrate the feasibility of full-scale trains for a commercial plant. The time schedule for construction of the 5000 tonne/day plant was estimated as one year for preliminary engineering, three years for detailed engineering, and three years of construction after completion of the engineering. Purchase of long lead-time equipment would start at the end of the first year and site preparation at the end of the second year of engineering. This would mean that the plant would be finished approximately six years after start of design if no significant delays are encountered.

After construction of the first 5000 tonne (5500 ton) per day facility, the second, third, and fourth stages would follow until a commercial-size facility is in existence - around 1993 or 1994.

The total throughput of such a commercial plant is estimated to be about 9 to 10 million tonnes (10 to 11 million tons) per year of coal containing 5% ash and 6-8% moisture. Liquid products would include one million tons per year of naphtha (reformer feed) and two million tons per year of distillate oil which could be hydrotreated to fuel oil with a boiling range of 200 to 325°C (390 to 620°F).

It is expected that the yield of liquid fuels would be about 50% of the coal throughput, indicating that approximately 6 million tonnes (6.6 million tons) of coal would be fed annually to the liquefaction process and the remainder [3 to 4 million tonnes (3.3 to 4.4 million tons) per year] would be used for utilities and for the production of hydrogen to be utilized in the conversion plant and in the oil upgrading facilities.

It was estimated that the capital cost of this plant might be 10 to 30% higher than that of an EDS (Exxon Donor Solvent) plant of comparable capacity. However, if product upgrading facilities were added to the EDS plant it is believed that the total capital costs including upgrading (already included in the German plant) would be in the same range for the two systems.

2.2.4 Saarberg Coal Liquefaction Pilot Plant [S,AA]

The Saarberg coal liquefaction process is a further development of proven IG-Farben technology. The main differences are a lower pressure, reduced hydrogen consumption and improved preheating of the coal slurry which is achieved by mixing oil partially condensed from hot separator vapors to the slurry. The new method dispenses with any indirect heat transfer elements for preheating the slurry. Thus a better availability and thermal efficiency of a coal liquefaction plant can be achieved. These improvements will be demonstrated in a 6 t/d pilot plant which began operation in July 1981.

In the 6 t/d coal hydrogenation pilot plant ground coal is dried and added to recycled solvent along with small quantities of catalyst consisting of iron-based sulfates (1.21% on maf coal), red mud (2%) and sodium sulfide (1%). The catalyst cost was quoted at 20 Dpf/kg (about $0.05/lb). The mix is heated to 430°C (800°F) and fed to four reactors in series at a pressure of 300 bars (4500 psi). Oil or gas quench is used to maintain the reaction temperature between 470-475°C (880-890°F). The reactor effluent is fed to a separator which produces a bottoms containing oil, ash, unconverted coal and catalyst. This is letdown to a pressure of 50 bars (750 psi) and then fed to a vacuum still. The reason for the intermediate pressure letdown from 300 to 50 bars (4500 to 750 psi) is an attempt to recovery energy via a piston engine device.

Bottoms from the vacuum flash, which contain 50% solids and 50% bitumen, are granulated and gasified to provide hydrogen for the plant. Gases from the hot separator supply part of the heat to the reactor feed via a concentric tube heat exchanger. Part of the condensate oil from this cooler is used to dilute the feed mixture. In the feed preparation system, recycle oil and feed coal (0-3 mm) (0-0.12 in.) plus catalyst are ground to a maximum particle size of 0.1 mm (0.004 in) in a ball mill. This mix is then diluted to 38 wt % solids with condensate from the reactor overhead gases.

2.2.4.1 History and Status

Saarberg constructed a continuous bench-scale unit which was put on stream in 1975. The purpose of this unit was to prove the application of the modifications to the original IG-Farben process. Based on results from the bench-scale unit, construction of a pilot plant with a coal throughput of 6 tonnes (7 tons) per day was begun in 1978. Construction was completed and the plant put into operation in July 1981.[k] In January 1980 Saarberg began a design study of a commercial coal liquefaction plant with a coal throughput of 2 million tonnes (2.2 million tons) per year which could start operation in the second half of the eighties. BASF was selected as the general contractor for the study, which will be finished by the end of 1981.

2.2.4.2 Company Experience

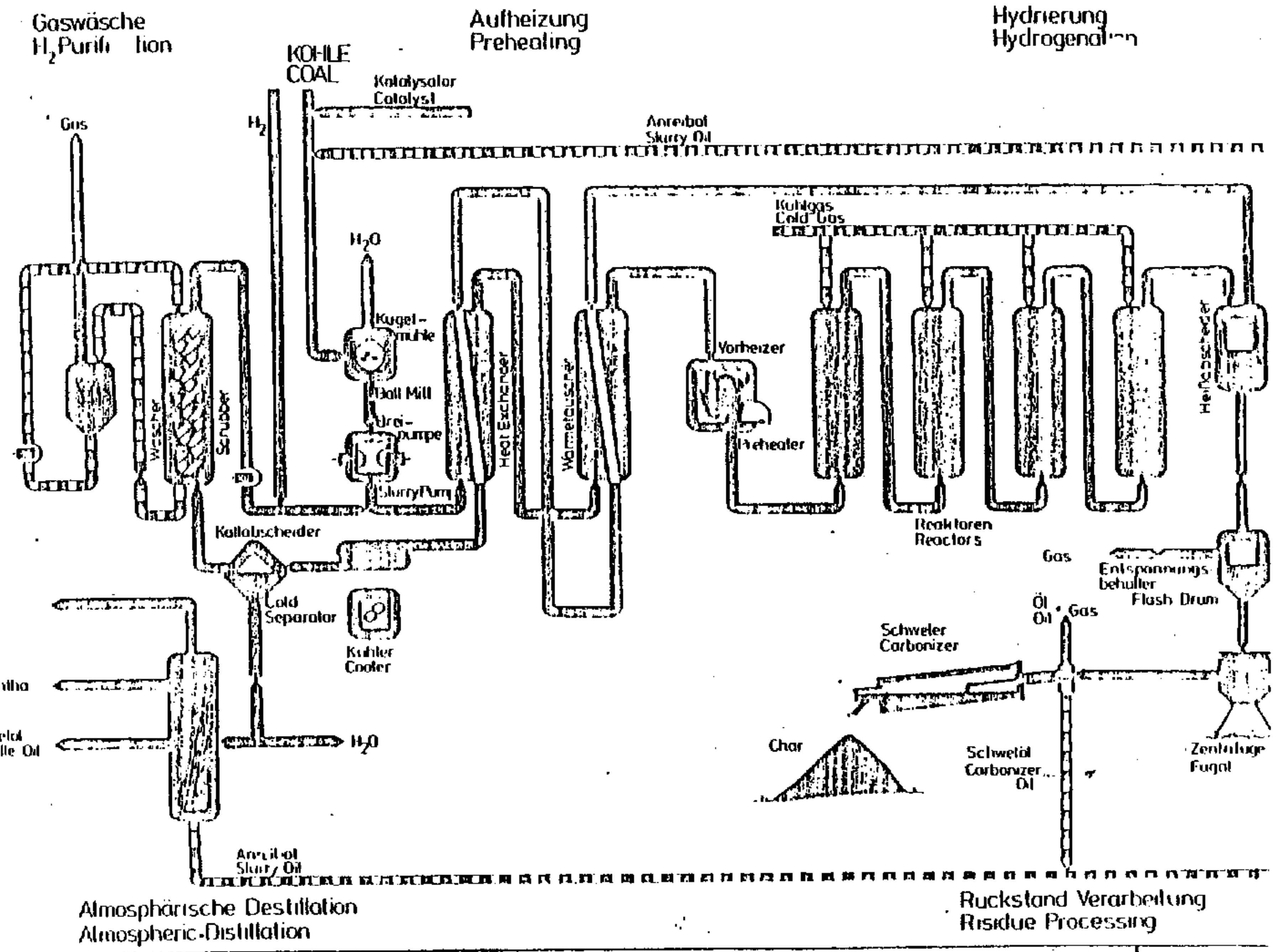
Saarbergwerke, an important energy company in the southwest of Germany, has developed multiple activities particularly in the field of coal upgrading on the basis of its own hard coal resources. Besides processes such as low temperature carbonization, coking and electricity generation, Saarberg has been engaged since 1974 in process developments for the gasification and liquefaction of coal. Saarberg decided that the most suitable technology for the production of distillate oil from coal was the direct liquefaction route since higher yields and higher thermal efficiencies could be attained than with the indirect route.

2.2.4.3 Plant Location

The 6 ton/day coal hydrogenation pilot plant is located at Volklingen/Fürstenhausen directly adjacent to the Saarberg/Otto process coal gasification plant, the Fürstenhausen coke works, the Saarland oil refinery, the Fenne power plant, and the Saarbergwerke AG central laboratories.

2.2.4.4 Process Flow for Saarberg Pilot Plant

Figure 2.13 shows how the IG Farben process was performed until 1945. Crushed and dried coal was mixed with a combination of red mud, Fe SO_4 and Na_2S and recycle oil to form a paste. This was done in rotating mills with steel balls as grinding elements in the presence of



AARBERG

Saarberg Kohlehydrierung
Coal Hydrogenation

FIGURE 2.13

1980/1

oil to prevent oxidation. The homogenized paste was then fed to high pressure pumps for injection into the slurry feed heat exchangers. The discharge pressure of the injection pumps was 700 bars (10,500 psi). In the heat exchangers, slurry and hydrogen flowed up outside the tubes with overhead vapors from the hot separator flowing countercurrently inside the tubes. Next, the coal paste and hydrogen were preheated to about 430°C (806°F) in a gas-fired heater. The gas/slurry mixture was then passed through four vertical reactors in which coal liquefaction took place at about 480°C (896°F). The exothermic reaction was controlled by the addition of cold recycle hydrogen. The effluent from the last reactor was separated into an asphaltene-free overhead product and a bottom product containing the unconverted coal, ash, catalyst, heavy distillate and residue oil. These gases and oil vapors from the top of the separator passed through the tubes of the slurry feed heat exchangers and were finally cooled. While the remaining gas was scrubbed to recycle purified hydrogen back into the process, the liquid was withdrawn from the cold catchpot and passed to an atmospheric distillation tower. All of the 325°C+ (617°F+) distillate was recycled as pasting oil. The main product was middle distillate oil which normally was converted to naphtha by hydrocracking. The hydrocracked naphtha and refined sump-phase naphtha were then reformed to high octane gasoline by the IG DHD (German acronym for pressure hydrogen dehydrogenation) process.

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

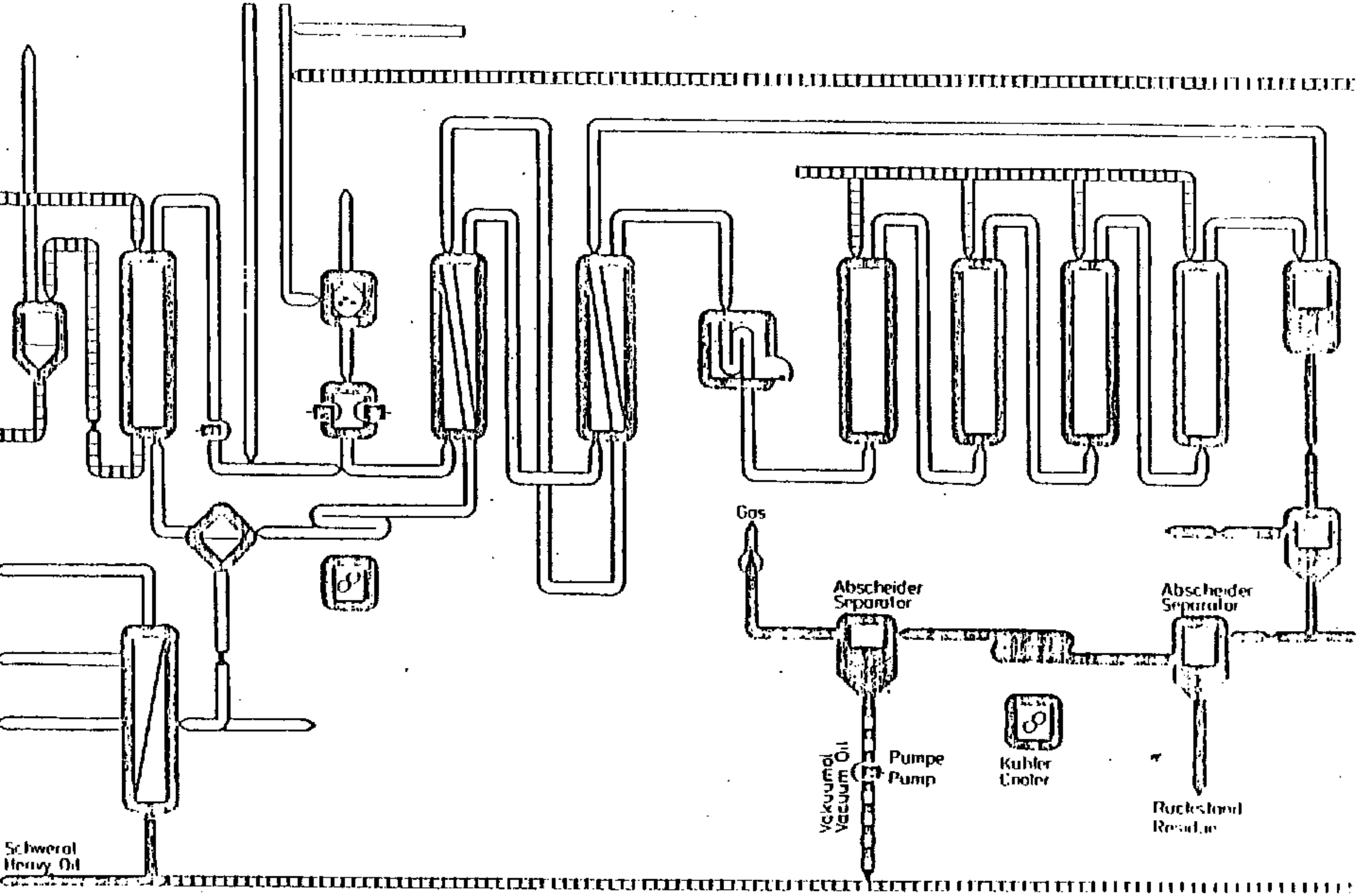
To achieve a reduction of the pressure, the asphalt cannot be recycled to the reactor. Therefore a different application of the residue had to be found: gasification for the production of hydrogen.

Separation of the asphalt from the hot separator bottom product could be accomplished by flash vaporization at reduced pressure. By this means an asphaltene-free vacuum distillate oil could be fed back to prepare the coal slurry thus leading to the expected reduction of the pressure to a maximum of 300 bars (4500 psi).

Saarberg constructed a continuous bench scale unit which was put on stream in 1975 and allowed the application of the above mentioned modifications of the I. G. Farben process. In contrast to the old process the residue preparation was operated as shown in Fig. 2.14 The hot separator sludge is passed to an intermediate pressure drum for the separation of a hydrogen-rich gas and then flashed into an atmosphere of reduced pressure. While the vacuum bottom product is withdrawn from the flash drum at the bottom, the overhead vapors are condensed and this oil is used as an asphaltene-free slurry oil. Also, the cut point of the atmospheric distillation was changed. With the I. G. Farben process the net oil yield had a final boiling point of 325°C (617°F). This has been increased to 400°C (752°F), thus reducing cracking in the sump phase which is only slightly selective in comparison to highly effective fixed-bed catalysts. The reaction temperature can now be about 10°C (18°F) lower which leads to the suppression of undesirable by-products like light hydrocarbons having a high hydrogen content. These changes to the I. G. Farben process proved successful in the liquefaction of hard coal at a pressure of 285 bars (4275 psi) and a consumption rate of hydrogen of 5.5 wt% of the moisture- and ash-free coal. The oil yield amounted to more than 50%. A typical product distribution of the modified I. G. Farben process is presented in Table 2.14.

2.2.4.5 Results

It has been demonstrated that the modified I. G. Farben process leads to satisfactory results even at pressures below 300 bars (4500 psi) using cheap disposable iron catalysts. The conversion of coal is satisfactory and the hydrogen consumption is considerably lower. The



Saarberg Kohlehydrierung
Coal Hydrogenation

FIGURE 2.14

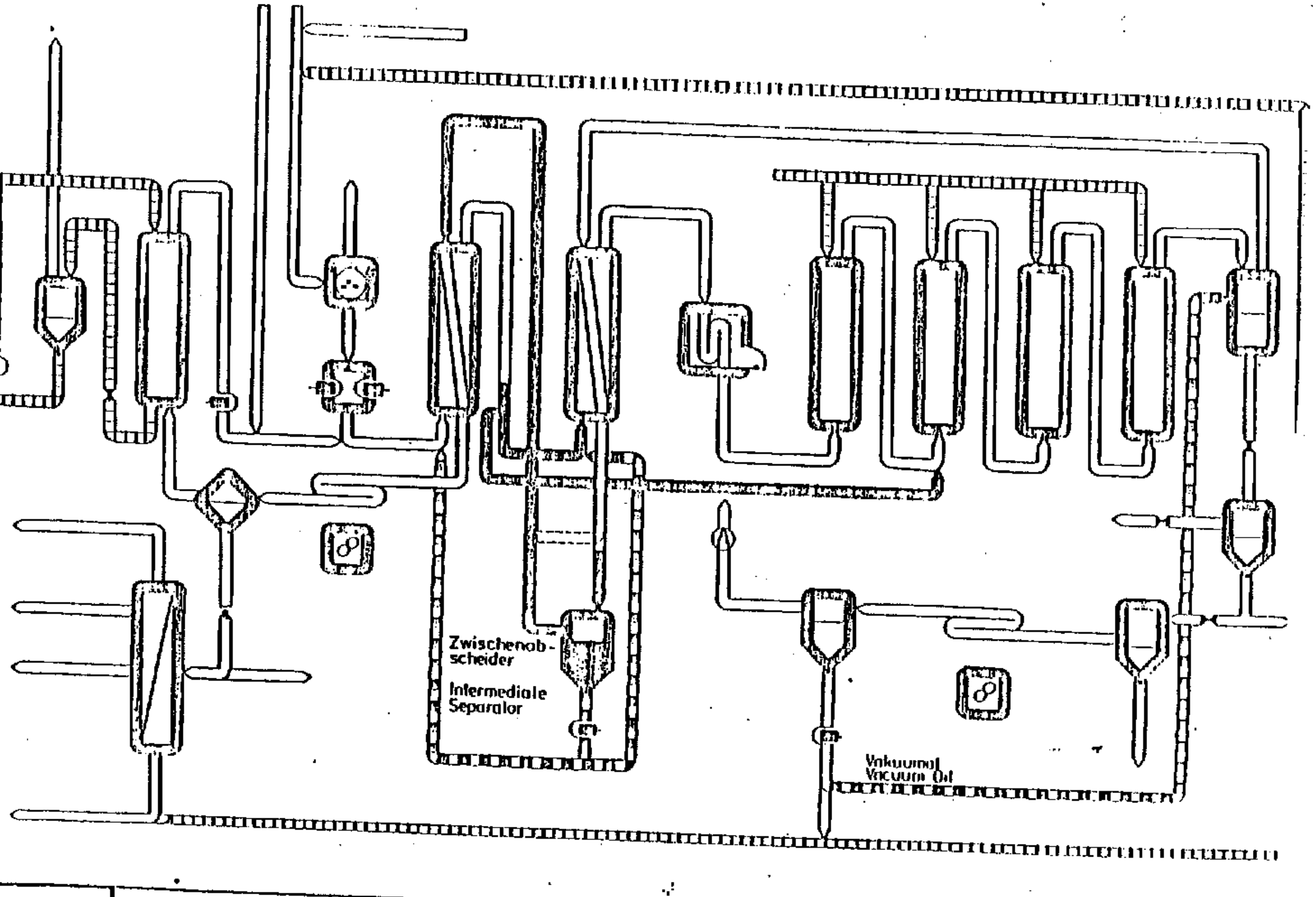
Table 2.14 Coal hydrogenation product distribution ("Ensdorf" bituminous coal)

	wt% of maf coal
H_2S, NH_3, CO, CO_2, H_2O	11.7
C_1-C_4	15.0
C_5-200°C (392°F)	14.6
200-325°C (392-617°F)	30.2
325°C + (617°F+)	9.4
vacuum bottoms	19.0
unconverted coal	5.6
Total	105.5

vacuum bottoms of the flash drum consisting of about 50% solids and 50% bitumen with a melting point of about 80°C (176°F) can be fed to a pressure gasifier in a liquid state for the production of hydrogen. Due to the fact that less asphalt is being recycled to the reactors the coal-hourly-space-velocity in the reactors can be raised from 0.365 to 0.65 tonnes of maf coal/m^3-hr (0.011 to 0.020 tons of maf coal/ft^3-hr).

Ways in which the process could be further improved are also under investigation. A matter of particular interest is the preheating of the coal slurry as it is shown in Fig. 2.15. The coal slurry preparation is conventional except for a high coal content in the slurry which may amount to as much as 60 wt%. This thick paste is injected into the high pressure system and heated up in a unique preheating system which is described below.

Referring to Fig. 2.15 an oil stream which is condensed from the hydrocarbon vapors leaving the top of the hot separator is fed from an intermediate catchpot to the thick coal slurry proceeding to the first



Saarberg Kohlehydrierung
Coal Hydrogenation

FIGURE 2.15

1980/III

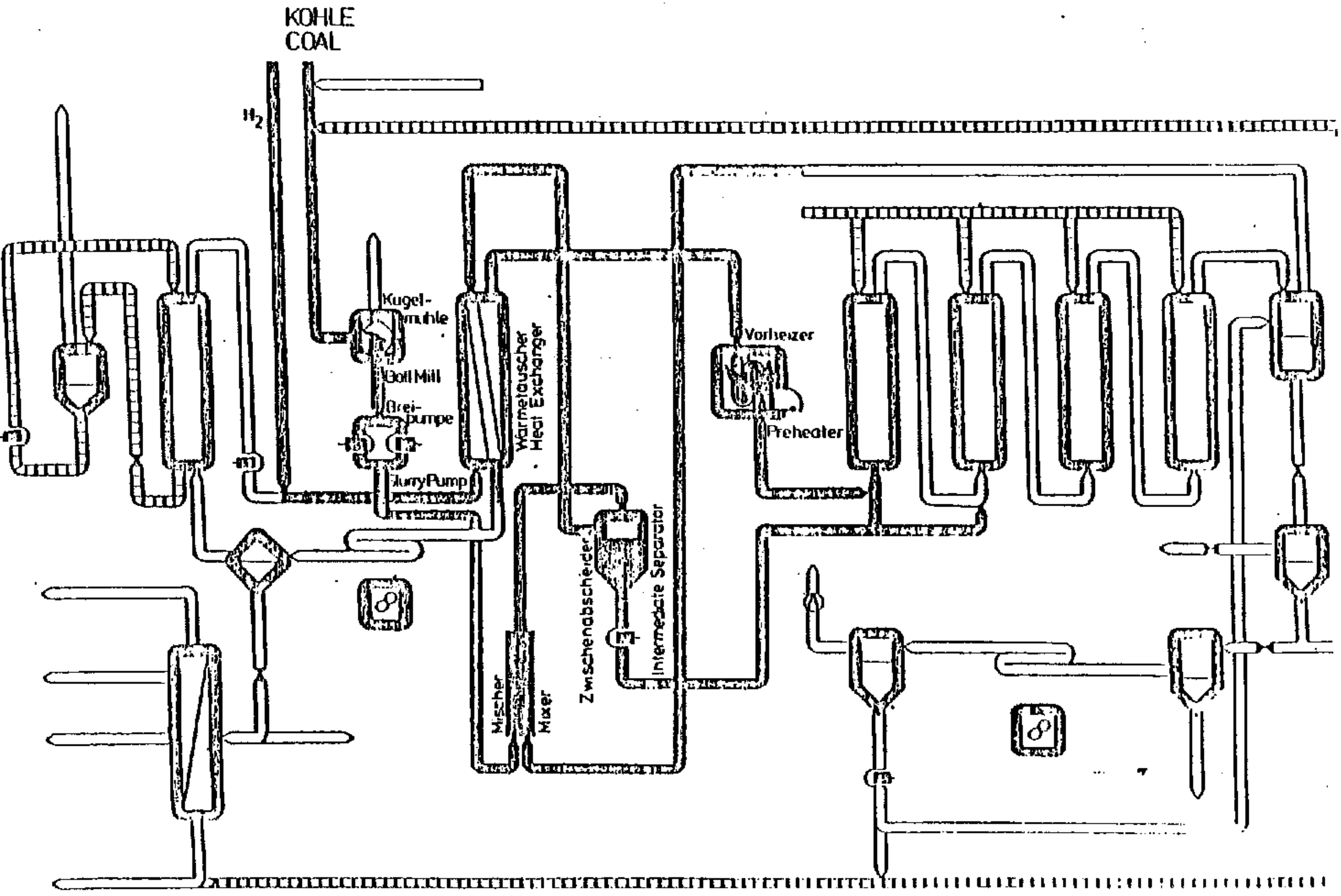
heat exchanger. By adding part of this diluent oil, the content of coal slurried in the paste is reduced to about 50 wt%. Having left the first heat exchanger, the paste is further diluted by another stream of oil from the intermediate catchpot. This advantageously results in both further dilution and heating of the slurry. The process illustrated makes it possible to recycle a considerable amount of the exothermic excess heat generated in the reactor back into the system so that the costly operation of a slurry preheater can in most cases be dispensed with.

The initial boiling point of the intermediate oil is above 200°C (392°F). The final boiling point of the remaining oil vapors which are condensed in heat exchange with the incoming fresh coal slurry is about 400°C (752°F). The described method represents an improvement particularly in energy saving. The internal hot oil recycle (which is about the same weight as the fresh coal) eliminates the need for atmospheric distillation of this slurry oil fraction. Further advantages are that this fraction remains under pressure at a high temperature. However, the use of bundle heat exchangers for preheating of the coal slurry is necessary.

Further attempts to simplify the process led to another idea which eliminates the heat exchangers needed for preheating the slurry. In this method the coal slurry is mixed directly with the overhead vapors from the hot separator. For this purpose the coal slurry is passed to a mixing zone which is also entered by the hot separator vapors. The vapors are cooled by the coal slurry, the heavy oils are condensed, and the slurry is preheated and diluted by the condensed oil. This also permits the use of coarser coal. Since the coal slurry is being transported only in tubes, there is little danger of sedimentation.

Coal particle diameters up to 1 mm (0.04 in.) are possibly applicable. The effluent of the mixing zone is separated into the hot coal slurry which is fed into the first reactor, and into the overhead vapors containing the products and the excess hydrogen. Application of this intermediate catchpot results in another advantage. The coal while being heated up normally splits off CO_2, H_2O, CH_4, etc. These compounds enter the reactor and increase the total pressure by their partial pressure. Now these compounds are eliminated from the coal slurry prior to entering the reactor. Also the physically adsorbed water is stripped off and withdrawn from the intermediate separator overhead. This preheating system has the advantage that there is a reduction of the pressure resulting from a higher concentration of the hydrogen in the reactor and a lower pressure drop for the slurry compared with the use of heat exchangers. This new method is pictured in Fig. 2.16. Saarberg believes that this arrangement can help make coal liquefaction simpler and more economic.

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



ARBERG | Saarberg Kohlehydrierung
Coal Hydrogenation | FIGURE 2.16 | 1980/IV

2.2.5 Rheinbraun Brown Coal Liquefaction Test Plant[Y]

The Rheinische Braunkohlen Werke AG (Rheinbraun) direct liquefaction process (or HVB process, from the German Hydrierende Verflussigung von Braunkohle) is based on the work of Bergius and Pier in the 1920's and operation of WW-II production facilities at Wesseling. The key feature of the HVB process is the application of this technology to brown coal (i.e., lignite).

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

These improvements resulted from the use of vacuum distillation instead of centrifugation for separating solids from the coal oil product. This new technique produces a recycle oil that does not contain any unconverted coal or asphaltenes which required the higher pressures and temperatures for decomposition to lighter material.

2.2.5.1 History and Status

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

The first line of the commercial plant, which will serve as a demonstration plant, will have a coal throughput of approximately 250 metric tons (275 tons) of raw coal per hour. Expected to start operation in the early 1990's, the demonstration plant will produce 400,000 metric tons (440,000 tons) of motor fuels and/or chemical feedstocks per year (440,000 tons/yr of motor fuel is equivalent to about 9000 barrels/day).

2.2.5.2 Company Experience

Rheinbraun currently mines 120 million tons of brown coal yearly, using modern techniques. The hydroliquefaction of Rhenish brown coal was carried out between 1941 and 1944 by Union Rheinische Braunkohlen Kraftskoff AG (URBK), a subsidiary of Rheinbraun. The yearly production was 250,000 tons (approximately 1.8 million barrels, equivalent to about 5000 barrels per day). Today URBK operates a modern refinery, producing fuels and chemical feedstocks.

The companies are working together on the improvement and optimization of the brown coal hydroliquefaction process. A continuously operated test plant has been run by Rheinbraun and URBK since 1978.

2.2.5.3 Plant Location

The 0.25 tonne (0.3 ton) per day test plant is located in Wesseling, West Germany.

2.2.5.4 Process Flow for the HVB Process

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

In the sump-phase hydrogenation (see Fig. 2.17), coal with a grain size of <1 mm (0.25 in.) and a water content of 10 to 12% by weight is used. The coal is slurried with sulfur and iron salts (as catalysts) in a recycle oil produced in the process. The slurry is heated to 350°C (662°F). In this process, water is evaporated from the slurry and humic acids are destroyed to such an extent that calcium carbonate does not stick in the hydrogenation reactor – a problem that occurred with the old technology.

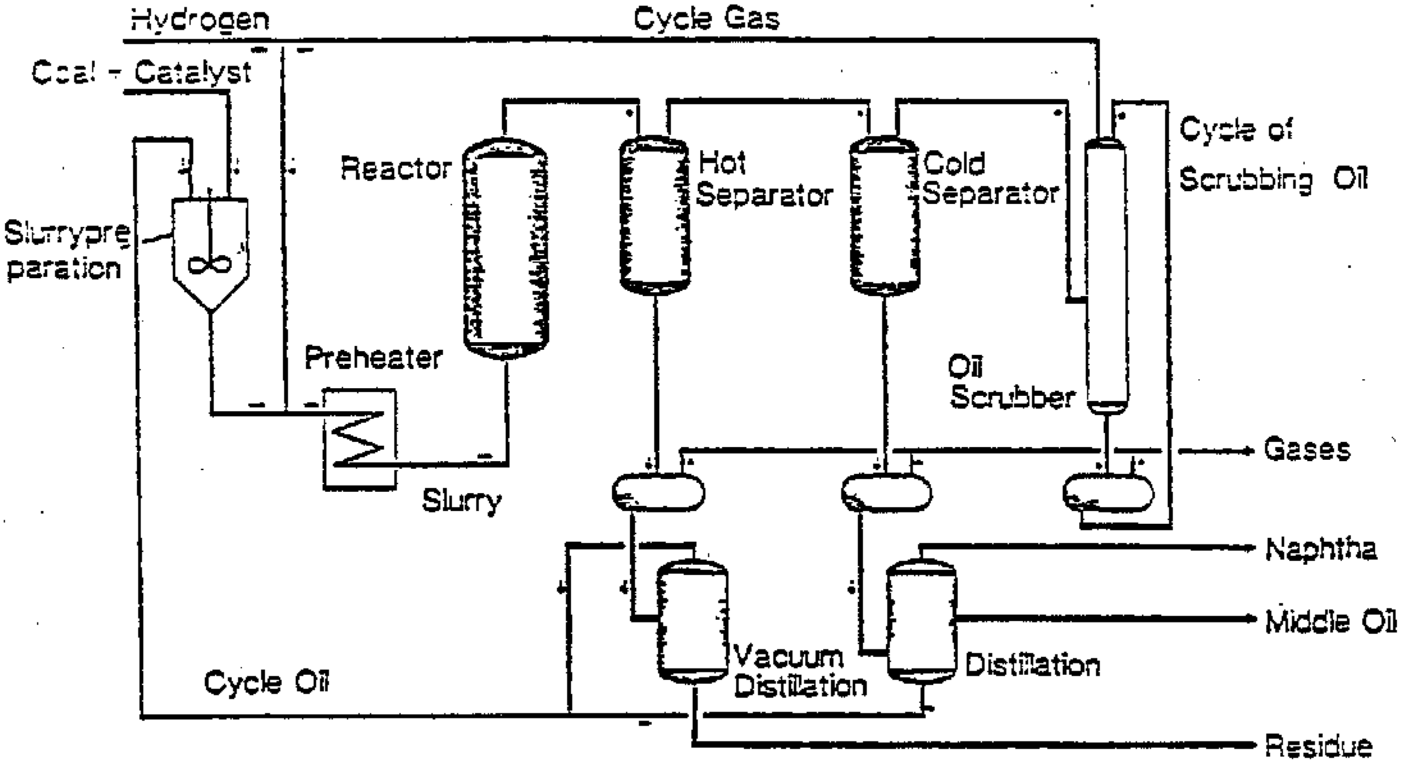


Fig. 2.17. Sump-phase hydrogenation flowsheet.

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

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

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

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

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

2.2.5.5 Results

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

About 5 parts by weight of hydrogen per 100 parts of maf coal are added in the sump-phase hydrogenation stage (see Fig. 2.18).

Vacuum distillation is used to separate the liquid product from the solids which consist of ash, catalyst, and unconverted coal. The solids are gasified to generate hydrogen. Part of the distillate is recycled for slurrying the coal feed. Vacuum overheads are subjected to further multistage distillation. The resulting distillate fractions, i.e., naphtha and middle oil, constitute the oil yield of the HVB process.

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

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

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

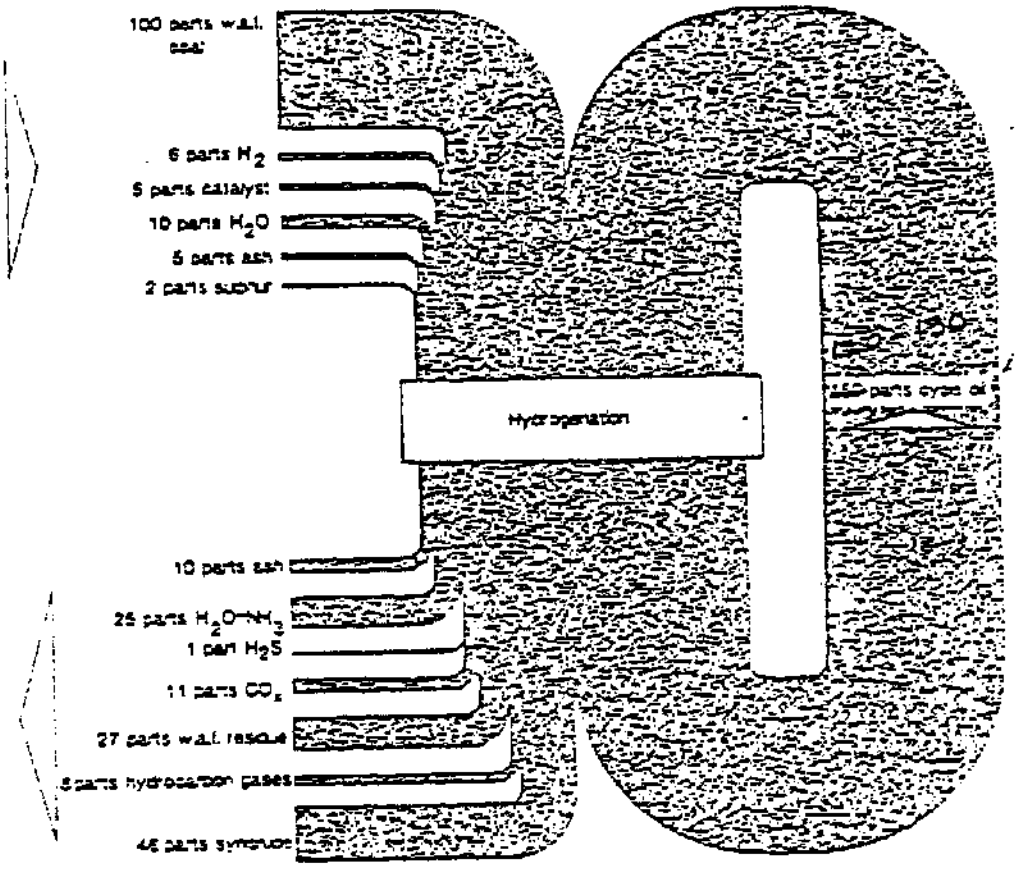


Fig. 2.18. Material balance for sump-phase hydrogenation.

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

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

The cost of producing gasoline by means of the HVB process is estimated to be approximately 50% higher than from crude oil.