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AN APPRAISAL OF THE POSSIBILITIES FOR THE MANUFACTURE OF LIQUID FUELS FROM COAL AS VIEWED FROM PAST EXPERIENCE

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An Appraisal of the Possibilities for the Manufacture of Liquid Fuels from Coal as viewed from Past Experience.

by

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Table of contents.

Table of contents (cont.)

Introduction.

The shortage of petroleum, one of the preconditions for establishing a coal to liquid fuels facility has been delayed for some time due to the flow of oil from the arctic. It is difficult to say for how long the depletion of the oil and gas reserves, which is the conclusion of numerous investigations on energy reserves in North America, will be extended.

1) The high productivity of the American industry depends on the availability of plentiful and cheap energy. Diagrams showing the parallelity of per capita energy consumption and gross national product are as widely published as the shifting pattern of energy demand and supply toward petroleum and natural gas. The discrepancy between the discovery of new oil- and gas reserves and the increase of consumption suggests the depletion of those reserves in a few decades.

2) The technologies and even more so the economics and industrial basis of the renewable energy sources like solar-, wind-, tidal- and geothermal energies are still not developed enough. At present they require too much investment capital per unit energy, even more than coalhydrogenation. The nuclear energy industry, besides higher investment needs than conventional power plants, poses serious environmental problems, safety- and security risks, especially in the reprocessing of depleted fuel, as to replace the fossil energy industry in a few years from now.

About 25 % of the total energy in the U.S. is consumed in the form of transportation fuels. Extensive development efforts have been made in several countries to produce lightweight secondary storage batteries for automobiles. But in spite of noticeable successes the electric battery driven car can not yet compete with the much lighter car driven by the internal combustion engine, which can be refuelled in minutes. No aeroplane has flown yet with electricity as driving force, although attempts have been reported to use highly concentrated nuclear energy as heat source for airplane turbines.

3) One other aspect of an industry based entire-

ly on renewable forms of energy would be that at present and probably in the forseeable future no oganic chemicals can be produced at reasonable prices without the use of carbonaceous materials. In 1975 about 12.5 Mtons/year of aromates, solvents, plastics, eleastomers and fibres were produced from petroleum representing a total consumption of about 8.5 % of the refinery products in the U.S.

From the afore mentioned considerations it will appear that in the next decades, as far as road vehicle-transprotation is concerned, the fossil fuel industry, even if it uses coal instead of petroleum, concurrent with higher energy costs, will be indispensable, especially if organic chemicals must be produced either as byproduct or as main product.

4) The dwindling oil- and gas reserves inside the U.S. and the relative abundance of coal of every type are the driving force behind the many research activities being conducted in industry as well as in government agencies, mostly coordinated and funded extensively by ERDA. Besides direct coal combustion, coal gasification and with the viewpoint on transportation fuels, a number of competing coal liquefaction processes are investigated.

The relative costs of various forms of energy from coal can be seen from diagram 1.[*] It shows that liquid fuels are the most expensive energy besides electricity. The social impact of considerably higher priced and not so readily available energy cannot be accurately predicted at present, except that higher energy costs must and will be compensated for by savings in other sectors of the economy, for instance recreation. But this is not the place or the time to discuss the economic consequences or political issues.

[*] not there

Process Chemistry and Technology.

1) Already centuries ago hydrocarbon type liquids were produced by dry distillation of wood. This ~~technology~~ has been extended to convert peat and also coals and has led to the development of the carbonization and coking industries. The more recent of these are the Ancite- and the Lurgi-Bergbauforschung formcoke processes. As liquid hydrocarbons contain 2 to 2.5 times as much hydrogen per unit weight carbon than the average coal, the excess carbon is eliminated in these processes as char, ~~which~~ would be uneconomical if mainly liquids are to be produced.

~~An immense amount of~~ research work has been and is still ~~being done in order to change the hydrogen-carbon ratio.~~ In 1869 Berthelot reacted bituminous coal, wood and woodchar with concentrated hydro-iodic acid at about 270 to 280 °C and elevated pressure. He obtained a bituminous product with 60 % distillable oils. The investigations of Franz Fischer should be mentioned here because they led to the later development of the Fischer-Tropsch process of indirect coal-liquefaction. He replaced the hydro-iodic acid by sodium-formiate, carbonmonoxide and water and by sodium-carbonate and hydrogen. At reaction temperatures of 400 °C and about 140 atm pressure 45 % ether soluble substances were formed with 53 % boiling up to 350 °C.

2) In order to obtain higher yields of liquid hydrocarbons hydrogen must be added to the coal or similar substances to be converted. This reaction is, ~~however~~, subject to the basic laws of the chemical equilibrium and the kinetics of the high molecular substances. As hydrogen at normal conditions has a low density, it must be employed at elevated pressure to increase the number of H_2-molecules per unit coal per unit reaction space in order to shift the equilibrium in the desired

direction and to increase the speed of the reaction. Also as coals or cellulose are high molecular compounds, their reaction velocity at ambient temperature is negligible like the popular polymeric plastics. Therefore higher temperatures and pressures are required to achieve reasonable reaction rates and investment costs.

The influence of hydrogen pressure, reaction-temperature and -time on the rate of conversion is shown in tables 1a and 1b [1]. ~~These~~ the tables show that high pressure hydrogenation does not produce specification liquids at 100 % yield and even less so in one step as Bergius had originally intended. That makes a commercial operation more complicated. The non converted coal plus ash plus catalysts plus recycled solids in the paste oil must be withdrawn from the reaction space and processed to eliminate them as far as possible from the recycle oil. The reclamation of the oil in the residual sludge is, next to the hydrogen production, the most important cost factor.

With higher reaction temperatures not only the conversion increases, thus reducing the quantity of sludge to be processed, but also the hydrocarbon gas formation. The latter increases the hydrogen consumption not only by reducing the liquid yield from a given quantity of coal, but also because the hydrocarbongases have a higher hydrogen-carbon ratio. Therefore the reaction conditions must be chosen so that an economic optimum is achieved.

The yield strength of the construction materials at operating temperatures sets another limit; the increase of the reaction temperatures leads to the liberation of excessive exothermic heat of reaction, which can become eventually so large that temperature runaways cannot be halted and damage to the equipment results. W.Kroenig [2] gives the heat of reaction as 7 kcal/g H_2 or 630 kcal/Nm^3 consumed. In commercial practice it was, however, 10 % higher, probably due to the higher reaction temperatures used, or about 78 BTU/cft.

3) ~~Before the I.G.Farben~~ embarked on coal hydrogenation in the "liquid phase" they investigated fairly thoroughly the coaldust motor, which is a form of direct coal combustion

Table 2 shows some significant operating data of a complete hydrogenation processes carried out in Germany [illegible] and also the dry hydrogenation of coal. But as the results of these experiments were not encouraging, the I.G.Farben bought the patents of Bergius, who recycled the heavy portion of the product oil in order to produce a light liquid only. The invention of the I.G.Farben was the hydrogenation in vertical vessels with agitation of the liquid by upward flowing recycle hydrogen and the separation into liquid phase and vapour phase for the production of specification liquids with the aid of better catalysts. This separation and the concentrated sulhpidic catalysts produced much higher yields of specification fuels and made the construction of commercial plants possible.

The hydrogen pressure is chosen so that contrary to catalytic craking no coke is deposited on the fixed bed catalyst. The main work at Ludwigshafen was, however, the development of specific catalysts and their testing with many different coals and oils at varying conditions to provide the design-basis for optimum economics of the 12 hydrogenation plants erected in Germany from 1937 to 1943. Table 2 shows some significant operating data of a few hydrogenation processes carried out until 1944.

4) At present in the U.S. great efforts are made to improve the existing technology or to develop a new one in order to make better use of the existing natural resources of the U.S. includig the coal deposits, without incurring the disadvantages of "conventional" coal liquefaction as compared to standard petroleum refining.

The main direction of the research is centered on reducing the hydrocarbongas formation, thus reducing the hydrogen consumption by using better catalysts like molybdenum in the liquid phase. Savings in hydrogen are then traded off against higher catalyst costs. One process tries to reduce the investment costs by reducing the residence time to a few minutes only, thus saving reaction space. Others try to achieve a reduction in hydrocarbongas formation and hydrogen consumption by operating at lower reaction temperatures and pressures but at longer reaction times.[3].

The reduction or elimination of the residue-treatment plant can be achieved by reducing the ash in the feed-coal and the high molecular asphaltoid-carboid constituents from the recycle oil. The"Gelsenberg"used the baryte-process to reduce the ash of the feed-coal to about 2.8 %. The costs of reducing the ash further would have been higher than the resultant savings in the sludge processing. In connection with this the I.G.Farben investigated quite intensively the solvent extraction of coal and developed an improved "Uhde" process. Investigations have shown 4) that extraction proceeds in two steps 1)the cleavage or depolymerization of the coal-constituents - the not exactly parallel planes of the beehive-like structure - and 2)the saturation of the free valences with hydrogen. This in combination with the solvent separating the coal-fragments prevents their repolymerisation. It explains also why coal hydrogenation in a solvent yields more liquids than the"dry" hydrocarbonization. However, as this is an additional highpressure operation, extracting the coal and filtering the extract would have added more costs than could have been saved by reducing the liquid-phase operation and hydrogen consumption.

Hydrocarbonization of coal was investigated by Franz Fischer in 1914 and was resumed by the I.G.Farben in the mid 1920's on a larger scale. Pulverized coal was introduced by a pellet press into the upper part of a vertical high-pressure vessel and fell downward against the upstreaming hot hydrogen, which carried the oilvapours out of the reactionspace. The non reacted coal and ash was discharged from the bottom by pressurized water. The results and especially the heat consumption for preheating the hydrogen were, however, not too encouraging. W.C.Schroeder 5) descibes the results of hydrocarbonisation of coal at the University of Maryland, in which pulverized coal passes downward, concurrent with the hydrogen at such a speed that the residence time is only 20 to 30 seconds at 500 °C and 150 atm. With 15 % (?) stannouschloride as catalyst sprayed onto the coal 55 % liquid, 40 % gas and only 5 wt% char were obtained. For a commercial plant Schroeder estimates a yield 47.6 wt% of an aromatic, crudeoil like product of 0.92 sp. gr. Partial preheating of the hydrogen is provided. The investment- and operating costs are estimated at about one tenth of those of a "conventional" coalliquefaction plant, which produces however, finished fuels boiling in the gasoline range.

5) It is well known that coal and heavy bitumens, if heated very fast in less than five seconds or even fractions of a second in a hydrogen stream to not more than 530 °C, can be converted to a very high degree to liquids. These are very heavy and repolymerize on standing. It has been observed in commercial practice that the bottoms from the distillation of the liquid-phase hydrogenation product becomes more viscous when kept in the tank for several days. This shows that even products obtained at high hydrogen pressure are far from being completely saturated and when cooled in the absence of hydrogen repolymerize. It had been shown in Ludwigshafen that even 700 atm product oils boiling above 350 °C reduce the activity and with this the operating life of fixed bed catalysts due to carbondeposition. Heavy oils from the liquid phase hydrogenation of tars and residues can, however, be passed directly from the "hot separator" over fixed bed vapour phase catalysts. They are not repolymerized and therefore do not form as much carbon deposits, which would deactivate the catalyst prematurely. This principle is used in the "combiprocess".

6) The liquid phase operation.

The process chemistry of the hydrogenation of high molecular organic substances involves the depolymerization of coal. It begins already at about 160 °C as can be seen from the increase of the viscosity of the coal paste when heated in the heatexchangers. Only at temperatures above 260 °C the lowering of the viscosity of the oil in conjunction with newly formed oil decreases the overall viscosity of the coalpaste to normal. This forces the dilution of the coalpaste to achieve a satisfactory heattransfer in the heatexchangers.

If heated to temperatures above 400 °C the high molecular materials depolymerize further, but unfortunately into fragments of unequal molecular weight, many of them so small that they distil and eventually have less than five carbon atoms. It is a peculiarity of the coal, coal precursors or oils, that on heating, the hydrogen in them migrates to the smaller, more volatile fragments, while the nonvolatile portion due to hydrogen starvation condenses to larger aromatic structures with very little hydrogen but with all the ash, called char or coke. This type of reaction is used in the cracking of heavier petroleum fractions into ones of lower molecular weight plus coke, deposited on the catalyst.

The velocity of the hydrogenation especially of the higher molecular substances depends like every other heterogeneous chemical reaction on the extent of the accessible surface and the diffusion rate of the reactants. With oil the diffusion is the faster the lower the molecular weight and the more saturated it is. The hydrogenation of coal is therefore effected only to a very small extent directly. Most of the hydrogen is transferred via rehydrogenation of the "solvent oil" like a one step extraction plus hydrogenation. While the aim of the coal extraction at about 420 °C is a minimal reduction of the molecular weight with insufficient hydrogen partial pressure at the end of the reaction, the coal liquefaction reduces the molecular weight of the product to a boiling range below 350 or 325 °C at higher hydrogen partial pressures, and this enables and necessitates temperatures above 480 to 490 °C.

At about 480 °C the speed of the hydrogenation just keeps pace with the depolymerization and thus prevents condensation and excessive cokeformation. If the temperature is permitted to increase further the cokeformation can exceed the hydrogenation and temperature runaways occur. The temperatures were measured by 2 x 24 thermocouples, inserted into two thermowells of about 35 mm ID, which extended from the top cover to the bottom of each vessel. The electrical conduits were made pressure tight because the high solids concentration at the bottom eroded the thermowells so that some leakages occurred. The risk of coke formation is always present in the hot separator, in which hydrogen and product vapours are separated from the liquid residuum. The liquid part of this vessel must therefore be cooled. Cooling of the liquid does not affect the vapour-space above it.

The difference in the velocity of the hydrogenation versus cracking and condensation reactions at various temperatures enables the highpressure hydrogenation to vary the degree of conversion and the nature of the products from largely a pure refining, i.e. elimination of O_2, S and N_2 and double bonds for instance from heavy tars and residua concurrent with minimal reduction of molecular weight of the products at moderate reaction temperatures of 360 to 400 °C to an increased production of lower molecular weight compounds in the boiling range below 350 °C with increased formation of hydrocarbongases.

Table 3 shows some analytical data, which compare the quality of middleoils and heavyoils obtained by hydrogenation and by carbonization. Although the hydrogenation products contain fewer phenoles, which indicates a higher degree of refining, only those produced from lignite and shaleoil have a lower sp.gr. and a higher anilinepoint. The products obtained from bituminous coal, with the exception of the phenoles do not exhibit any visible improvement. The insufficient refining , achieved in the liquid phase, which is the result of the vapours being carried out of the reaction space in a very short time and rapid cooling, necessitates one or more further refining operations

7) The Vapour Phase Operation.

The reduction of the molecular weight of the liquid phase product is a prerequisite if transportation fuels shall be produced from them in the adjoining vapour phase hydrogenation or in a catalytic cracker. If the molecular weight of the feed is too high, carbon deposition and catalyst deactivation occurs at 300 atm pressure. However, at 700 atm residuum from the the liquid phase distillation can be processed as W. Kroenig describes it 2) over fixed bed catalyst to 92 wt% gasoline and middleoil, a yield, which is higher than obtained when recycling this oil as paste oil to the liquid phase operation. The advantage of this division of the liquid phase is the reduction of the high pressure reaction volume and other equipment.

It was the opinion of the I.G.Farben, supported by extensive investigations, which indicated a higher yield of the desired liquids, and perhaps by the lack of experience with catalytic cracking in the 1930's, that vapour phase hydrogenation at high pressure was, and from the viewpoint of energy recovery still is, preferable.

Therefore in order to produce various specification liquids a number of vapour phase processes were developed. The standard two stage 300 atm process, as shown in diagram 2, consists of a refining step with concentrated tungstensulphide, later replaced by diluted tungsten-nickel sulphide catalyst and a "splitting" stage with diluted tungsten or molybdenum sulphide catalyst. Both were arranged as a fixed bed of pellets in layers with quenchgas distribution devices in between as is well known.

8) For the production of aviation gasoline its octane rating must be increased from about 74 to 80. By using a pelleted activated char, impregnated with 15 % Cr-oxide and 5 % V-oxide, at 300 atm and 500 °C a gasoline with 50 % aromates was obtained in a good yield. This catalyst did not have a satisfactory lifetime and was abandoned. More success was achieved by processing the liquid phase middleoil at 700 atm with a catalyst of extruded terrana impregnated with 0.6 % Mo, 2 % Cr-, 5 % Zn-oxides and sulphur in one step to specification gasoline.

Lower hydrogen pressure lowers the degree of hydrogenation, which eventually leads to carbon deposition on the catalyst. If, however, a starting material is used, which has sufficient chemically bonded hydrogen, as is the case with lower boiling fractions, containing a high percentage of naphtenes, then lowering the hydrogenpressure to 60 atm or lower effects a dehydrogenation and formation of aromates. These processes like the DHD and hydroforming were employed to aromatize the fractions boiling from about 90 to 200 °C. During an operating period of 130 to 190 hours about 0.2 to 0.25 wt% of the feed was deposited as carbon on the catalyst - 10 % MoO_3 on activated Alumina. The catalyst was regenerated in about 12 hours, with

N_2-purging in 36 hours, by burning off the carbon. The purpose of this type of aromatization was only to fulfill the requirement for high octane gasoline without installing the scarce high-pressure equipment.

It has been shown that the dehydrogenation process can be used advantageously to upgrade the corresponding petroleum fractions, whereby the hydrogen liberated is used in the same reaction to saturate the olefinic bonds of petroleum-compounds. Table 4 shows some significant data of the commercial operation of some vapour phase hydrogenation- and dehydrogenation processes. The U.S.oilrefining industry has continued to perfect especially the lowpressure vapour phase processes and to adapt them to its requirements.

Chemicals from Coal by Hydrogenation

1) As seen already in table 3 the analysis of the middleoils and raw gasolines from the liquid phase hydrogenation indicate an appreciable concentration of phenoles and other taracids, which can be extracted for instance by the metasolvanprocess, an aequeous methanol. Phenoles are partially watersoluble and are therefore found in the water from the reaction or injected after the hot separator to dissolve the ammonium salts and prevent their precipitation in the cooling train, and from the addition of steam in the distillation. This water must be purified is separated from the produots in the tanks. Before discharging it, its phenol content of up to 6 g/l must be extracted for instance by the phenosolvan process, i.e.butylacetate in naphta. The combined phenoles must then be further purified. If the phenoles from all the middleoils and raw gasolines would be extracted it would make possible a phenolproduction of about 22,000 bbl/d in a 100,000 bbl/d plant and this could enhance the economics of coal hydrogenation considerably.

2) Tarbases too can be extracted from the products of the liquid phase with diluted sulfuric acid. The aequeous solution is neutralized with ammonia and distilled. According to W.Kroenig 2) the gasoline fraction contains 12 to 15 g/l omit

in one case 22 g/l, and the middleoil 42 to 56 g/l tarbases. This concentration, if all raw gasoline and middleoil is extracted, enables a production of about 5900 bbl/d in a 100,000 bbl/d plant.

3) The aromates can be extracted from the products of the aromatization processes for instance with SO_2- propane mixture. The individual aromates are then separated by precision distillation and eventual recrystallization. Higher molecular weight polynuclear aromates have not been recovered commercially from hydrogenation products, although they are present in concentrations of about 10 to 20 % pyrene, 2.4 to 4 % methylpyrene, 1.2 to 4.9 % dimethylpyrene, 0.12 to 1.12 % benzoperylene, 1.2 % coronene and 0.6 % carbazole in the sludgeoil. In Germany the demand for these compounds was met by products from coaltar.

Insertion

4) The hydrogenation offgases are those, dissolved in the products at elevated pressure. They are released and crudely separated by fractional pressure letdown. At about 25 atm a mixture, containing approximately 61 vol% H_2, 19 % CH_4 and 5.4 % C_2H_6 is released. It is used partly as fuelgas or, as was practised in 4 plants, sent to a polymer plant. Here it was dehydrogenated to acetylene and the hydrogen so liberated was returned to the hydrogenation plant. One plant converted the first offgas by steamreforming to hydrogen and produced the required fuelgas from coal.

In central Germany ethane was cryogenically separated from the first and second letdown stage -gases. It was dehydrogenated at about 300 to 400 mm Hg and 800 °C in the presence of some oxygen over porcelain pellets as catalyst. The yield was about 63.7 wt%. The ethylene was polymerized to high quality lubricantoils with a viscosityindex 105 to 115, Conradson coke 0.2 % and a sp.gr. of 0.855 to 0.860. The hydrogen from the offgases of the dehydrogenation was returned to the hydrogenation plant. When increasing the oxygenaddition to the ethane-dehydrogenation, the acetylene yield can be increased from about 1 wt% to about 33 wt% of the ethane feed.

The C_3 to C_5 offgases were liberated in the last or atmospheric letdown stage and in the stabilization of

aromates can be extracted from the [illegible]
Olefines have been produced from the HC gases during
hydrogenation

the gasoline. These gases contained only about 8 vol% H_2 besides 9 % CH_4, 15 % C_2H_6, 26 % C_3H_8, 26 % C_4H_{10} and 11 % C_5H_{12}+. The 0.7 % NH_3, 1.7 % H_2S and 0.5 % CO_2 are eliminated by waterscrubbing or eventually with organic solvents like MEA, alcacid lye etc. The organic sulphur is generally below the permissible limit of 250 mg/Nm^3 and is therefore not extracted. After a final water wash and drying operation the gas is compressed, whereby pentane and higher components are condensed and separated. The latter are added to the gasoline before its stabilization. The remaining gas was used as LPG for trucks. One plant in Pommerania separated the n-butane and iso-butane from the richgas as it was called. The iso-butane was dehydrogenated at atmospheric pressure and 550 to 620 °C in a fluidized bed of granules of 90 % alumina with 8 % Cr_2O_3 and 2 % K_2O. As carbon is deposited, the catalyst must be regenerated continuously. About 20 % of the iso-butane is converted per pass. From the converted iso-butane about 78.5 % are obtained as isobutylene. The liquid product is separated, then dimerized and hydrogenated to iso-octane. Besides diisobutylene higher polymers are formed. Isobutylene has also been alkylated with isobutane in the presence of concentrated H_2SO_4 directly to iso-octane.

The ratio of isobutane to n-butane is shown in table 5. This compilation shows that the isomerization is the more predominant the more diluted catalyst is present per unit feed material in the reaction space and especially the alumina-silica component. The influence of the alumina-silica can also be seen from the concentration of olefines as shown in table 6. Isobutylene has also been polymerized to isobutylrubber.

If middleoils are hydrogenated over fixed bed saturation catalyst at lower reaction temperatures of about 380 °C and the resultant product cooled by decompression of the propane used as diluent, the paraffin precipitates in crystals, larger than without hydrogenating the oil. The resultant paraffin can therefore be easily filtered and is relatively pure with a high content of straight chain components. If this paraffin is heated at atmospheric pressure in tubes to about 510 °C and residence times of approximately 25 seconds and then quenched, a high yield of alpha olefines, about 60 %, is obtained. These olefines have also been polymerized to excellent lubricants.

Paraffin has also been chlorinated to about 13 % Cl and the product condensed with naphtalin. The resultant product, diluted to 10 % with light lubricantoil has been used as pourpoint depressor for lubricants for a long time on a commercial scale. The oxidation of paraffines to fatty acids is universally known.

Many more compounds have been isolated in the laboratory and processes are available for commercial production, provided there would be a market for them. For instance the extract from bituminous coal, having a very low ash content, has been used for the production of electrode coke. The residuum from the vacuum distillation of the sludge has been used commercially for the improvement of the coking qualities of low value coals for the production of metallurgical coke. Polynuclear aromates could easily be extracted from the vacuum distillate. Also the coal extract, after treatment with nitrogen, has been used in the I.G.Farben coaldust motor, as its flush point could thus be depressed from 400 to 200 °C and its melting temperature sufficiently increased.

The sulphur and nitrogen in the hydrogenated coal appear in the offwater as ammoniumsulphide. In order to extract the phenole it cannot be oxidized to $(NH_4)_2SO_4$ directly. About 94 % of the sulphur are therefore produced as elemental sulphur in the Clausplant. Gasification leaves slightly more sulphur in the ash. The nitrogen of the hydrogenated coal appears almost completely in the offwater as NH_3. The N of the gasified coal appears to about onethird as NH_3 in the gasliquor. These waters are combined for phenol extraction, the NH_3 is then neutralized with H_2SO_4 and after partial evaporation the $(NH_4)_2SO_4$ is crystallized and filtered.

Operation of a Coal Hydrogenation Facility.

1) The operation of the vapour phase must be conducted at reaction temperatures, pressures and throughputs, which

enable the distillations to separate fractions of specific characteristics. By scrubbing these with soda lye or other reactants to eliminate any remaining sulphurcompounds, finished products are obtained.

The concentrated tungstensulphide catalyst, especially when fresh, exhibited the tendency of temperaturerunaways due to the relatively large exothermic heat of reaction of about 240 kcal/kg feed. This heat of reaction made it possible to operate the refining stage witout heatinput from the preheater. The splitting operation released only about 120 kcal/kg reactionheat and required a gasfired preheater.

The operation of the liquid phase in particular the coal hydrogenation is more complicated as the recycle-pasteoil quantity cannot be varied arbitrarily as with the vapour phase. The reaction temperatures and throughputs must be adjusted so that a certain level of pasteoil is kept in the storage tank. Besides this the operation of the sludge centrifuges and kilns limits the asphalt content in the diluted centrifuge feed to certain levels. Also variations of the coal fed to the hydrogenation caused a different behavior in the preheaters and the hydrogen and quenchgas requirements. A change of the coal, delivered from the mine caused always a hectic activity in the controlisle. The exothermic heat of about 578 kcal/kg coal is much greater than in the saturation stage, but the dilution with pasteoil almost 1 to 1.5 acts as heat sink and the less active catalyst of about one twentieth the concentration of the fixed bed catalyst does not so easily lead to local overheating and temperature runaways.

When the temperature must be depressed at any lower spot in a reaction vessel it was always observed that the temperature profile so created migrated gradually upward before nomalizing at the top of the vessel. Also from deviations of the solidsconcentrations at different levels it can be concluded that the content of a liquid phase vessel is not as fluid as pasteoil. When heated in a beaker and gas does not bubble upward in it like in water, but is an intermediate between a fluid and a foam.

2) In comparison to the vapour phase, which contains about 1720 kg/m^3 WS_2 or 580 kg/m^3 diluted WS_2 the average catalyst concentration in the liquid phase is about 61 kg/m^3. The upward liquid velocity in a liquid phase vessel, especially when processing tar residues or heavy oils, which are easier hydrogenated than coals, leaving a lower concentration of asphalts in the sludge, should not go below a certain minimum in order to suspend the catalyst properly. Normally due to the settling equilibrium it is more concentrated at the bottom than at the top. When catalyst becomes too cncentrated at one spot, the risk of temperature runaways and cokeformation is eminent. Also as has been experienced with the hydrogenation of lignite-tarresidua, the saturation especially at 700 atm can proceed too fast and too far so that asphalt precipitation and as a consequence cokeformation occurs. If instead of gasoline and middleoil only, heavier fueloil shall be produced besides these, then it is advantageous to recycle some middleoil. To prevent the coalpaste from becoming too viscous due to middleoil evaporation, large quantities of sludge must be recycled, partly as hot recycle directly, partly as "cold"recycle, mixed with "thick" coalpaste of up to 56 % solids in order to produce "thin" paste with only 42 to 46 % solids for preheating in heatexchangers.

3) The permissible heating- and cooling rates for startup and shutdown of a highpressure system, due to the relatively thick walls mainly of the outside insulated conduits, which connect the vessels, were set at 20 °C/hr up to 260 °C and from then on only 12 °C/hr. Including pressuretesting, purging with nitrogen etc. it took about 31 hours before coalpaste could be injected, starting with about 8 m^3/hr and increasing gradually to 44 m^3/hr for 40 m^3 reaction space. In the course of operation the inside of the preheater tubes became coated with a 6 mm thick sulphide crust. The outside-and with this the metaltemperatures must be raised continually until after about 11 months the safety factor, which was at the beginning about 1.2 was down to almost 1, thus forcing the shutdown for overhaul.

Auxiliary Operations.

1) The production of Hydrogen.

Several auxiliary operations are required. besides the supply of electrical power and fuelgas, the purification of the raw generator- or reformergas for the production of the required hydrogen was the most important one. The"light"-hydrocarbongases cannot provide all the hydrogen needed for the production of gasoline from coal. W.Kroenig 2) gives the approximate percentage of H_2, which can be manufactured from the methane and ethane portion of the offgases, as compiled in table 7. It can be seen that from lignite only ~30 % and even from heavy residua only 46 % of the required hydrogen can be produced. This percentage can be increased to almost 100 if propane and butane are also converted. However, due to carbon formation, the higher hydrocarbons are usually converted by partial combustion, if at all.

All these processes require their desulphurization. For the production of olefines the cryogenic separation into pure components is indispensable. The elimination of hydrogensulphur and of cause watervapour is a precondition. Today one will probably employ the Rectisol-(methanol-)scrubbing or a similar physical- instead of chemical solvent process like the alcacid-lye, used by the I.G.Farben. The physical processes permit the reduction of the sulphur to less than 1 ppmv instead of 100 ppmv. The methanol process has also a good selectivity between CO_2 and H_2S; it eliminates the entire organic sulphur and can tolerate H_2S inlet concentrations of 18 to 40 % as compared to maximal 2 % with the Stredford process. Table 8 compares some significant data of the more important sulphurremoval processes and diagram 3 depicts one Rectisol flowscheme for separating

Processes for production of H_2 are commercially available and purification of gases three different components in technically pure form from the offgas. The purified dry offgases can then be processed as discussed. If the offgases are separated cryogenically it may be economical under favourable conditions to sell the methane as naturalgas substitute and produce the hydrogen or fuelgas from coal. Table 9 gives the approximate cost of hydrogenmanufacture by some of the most commonly used established methods 6).

2) The Distillation.

The liquid products are discharged from the highpressure operation via several letdown stages into tanks. The separation of the water from these products, especially from the liquid phase is sometimes difficult, due to the high sp.gr. relative to water and due to the content of oxygen and nitrogen containing compounds. Before distilling they must eventually be heated to 200 to 250 °C at elevated pressure in order to increase the difference in density. In some cases gasoline has been recycled. The fractionation itself employs essentially the same techniques as in the petroleum refining industry including the extraction of pure compounds and manufacture of chemicals.

3) The waste Water.

The water, separated as mentioned at various places from the oil products, appears mostly in the tanks. It contains ammonium carbonates and -sulphide, taracids and eventually tarbases. Before releasing this water into the river it must be purified. In order to reduce its quantity and increase the concentration of the contaminants a significant portion of it is reinjected after the hot separator of the liquid phase.

The first mechanical separation is done in conical settlers, in which the oil floats to the top and the solids settle to the bottom. The water is withdrawn from the middle and is filtered through gravelbeds. It must be optically clear if the phenoles shall be recovered. The separated oil is then centrifuged and recycled to the liquid phase. The hydrogensulphur is driven off by passing it downward against a stream of CO_2 in a tower. The ammonium carbonate is converted to bicarbonate, which eases considerably the subsequent extraction of the phenoles, as free ammonia would saponify the butylactate solvent. The H_2S-CO_2 mixture is in most cases processed ~~treated~~ directly in the "Claus"process to elemental sulphur.

The water, mixed from all sources contains about 4 to 6 g/l taracids. It can be enriched by washing it countercurrently with a fraction boiling from 160 to 220 °C from the liquid phase and reextracting the phenoles with a part of the offwater. By this method an offwater with about 17 g/l phenoles is obtained in about 1/3 of the original quantity. Especially the lower, more valuable phenoles, which are more water soluble are preferentially reextracted, while the higher ones are left in the oil, which is combined with the bulk of the middleoil. The latter is processed in the vapour phase, where the higher phenoles are hydrogenated to aromates.

The concentrated water so prepared is extracted with phenosolvan to about 200 mg/l, which is still too high to be released. The extracted water is diluted with warm cooling water in a ratio 1 to 10 i.e.to 20 mg/l phenoles. Then by biological treatment with air at 25 to 30 °C at a pH value of 7 to 8.5 a remaining phenol concentration of about 3 mg/l is achieved. The phenol content can be further reduced to about 0.5 mg/l if this water is used to transport the ash from the boilers or gasifiers to the dump pond. Eventually after recovering the ammonium salts the water is clean enough to be reused in the plant.

4) The Sludge-Residuum Processing.

At the time when the hydrogenation plants were operated, there was no better method available to recover the highboiling oils from the residuum and to eliminate at least a part of the solids, left over from the process. In almost all plants the sludge, containing 20 to 22 % solids, was diluted with 25 to 35 % asphalt-and solidsfree heavy distillate to a solids-content of about 14 to 16 %. This mixture was separated at about 150 °C in continously operating centrifuges into about 75 % oil with 4 to 9 % solids and 25 to 30 % residue with 38 to 40% solids. The centrifuge oil was used, mostly in mixture with additional heavy distillate as paste oil, while the heavy distillate alone was used as glideoil for the heatexchangers and at various locations of a system in small quantities as purgeoil to keep the contents of important passages and valves solidsfree and fluid.

The centrifuge residue was preheated in heatexchangers to about 400 to 450 °C before entering the rotating kilns, which had a capacity of only 3 ton/hr. At about 560 °C the oil was distilled and the asphalts transformed to coke. Steelbars and -cubes ground it in the kilns to a powder, which was discharged through a waterlock. If the centrifuge residue contained middleoil it must be preheated at elevated pressure, which was letdown before entering the kiln and the evaporating middleoil was condensed. The first, hot condensate contained about 0.3 to 0.5 % dust and was added to the diluent oil for the centrifuges. The second, cooler condensate was added to the liquid phase product to be distilled.

The residua of the solvent extraction of coal can be filtered at about 5 to 8 atm and 150 to 170 °C. Eventually a two stage filtration can reduce the quantity of the residuum to be carbonized. The filterability of the extract can be significantly improved if the residuum from the highpressure hydrogenative extraction is discharged by slow pressure letdown in expansion-machines instead of through nozzles. The extraction of the coal by a solvent leaves its ash-skeleton mostly intact and the particles remaining on the filter are larger and form a more porous layer.

5) The Preparation of Heavy Oils and Coals for Hydrogenation.

The heavy oil residua must be rendered free of solid precipitates and water by addition of 1 to 3 % H_2SO_4 and centrifuging batchwise; for processing over fixed bed catalyst these feedmaterials must subsequently be filtered. In most cases these tars were distilled in combination with the liquid phase product and the heavy oil residue returned to the liquid phase.

The coal to be processed should have as little ash as economically feasible as the hydrogenation works the better the less ash and non converted coal constituents must be transported through the reaction space and eliminated in the residue processing plant. Any tramp materials are removed before crushing the coal for deashing with "setzmachines". A much better deashing of the coal, crushed to 3 to 5 mm, can be achieved with gravity fluids like the Tromp- or the Baryte process. That portion of the coal, with the ashcontent thus increased, eventually up to 35 %, is used in the powerplant or the gas~~ifiers~~producers. A large portion of the fusinite, which is almost impossible to hydrogenate and acts as ballast is also separated with these two processes from the vitrinite and clarite of the of the hydrogenation-coal fraction. Flotation processes, which would eliminate even more fusinite are generally considered to be too expensive.

Lignite cannot be deashed economically as the ash is chemically bonded to the humic acids of that coal. It is therefore taken from mines which produce the coal with the lowest ash. Lignite hydrogenates very fast; therefore the ash in in the lignite should be about proportional to the content of extractable bitumen, as the ash-skeleton plays a significant role in the conversion of the asphalts. As already seen from the isomer content of the offgases and the products of the catalytic vapour phase reactions it appears to be a retardant rather than a promotor of hydrogenation.

If the sand content of the feedcoals is too high so that settling in the reaction vessels results, then the sand must be withdrawn periodically from the bottom of the first reaction vessel. Sand has also been partially eliminated by settling of the coal paste in conical tanks. Lignite, crushed to 3 to 10 mm must always be dried, mostly by direct contact with fluegas of about 350 °C and less than 2 % oxygen.

The ironsulphate catalyst is sprayed as concentrated solution onto the coal during its transport to the drying equipment. The ironoxides are added immediately before drying. The waterfree sodiumsulphide is, however, added just before the pasting of the pulverized coal in order to avoid hydrolysis by the watervapour.

All coals, especially if they are still warm are pyrophorous and must be blanketed with inertgas. The danger of explosions is the greatest when the drying equipment is shut down, as then in the presence of coal dust air enters due to the condensation and contraction of water vapour.

Drying and grinding to about 1 mm is in most cases combined in a hammermill. The discharge is screened and the oversize returned to the mill. The undersize is fed to the "concentramills", in which the coal is ground in two stages in the presence of pasteoil to 60 % through the 100 mesh screen, corresponding to a particle diameter of 0.065 mm. The coalpaste from this mill is screened to eliminate the last traces of oversize material, mostly wood particles, and stored in tanks. Eventually a portion of it is diluted with additional pasteoil and/or cold recycle sludge and can then be"desanded". These coalpastes are pumped to the suctiontanks in the highpressure operation, from which they are constantly circulated in steamheated ring-conduits at about 125 °C and 4atm pressure, at which the pastes enter the highpressure injectors. Besides the conduits for coalpastes one ring-conduit must be provided for pasteoil for startup and shutdown and also one for purgeoil.

Economics of Coal Liquefaction.

1) A Comparison of a Standard with a more developed Hydrogenation.

No Fischer-Tropsch plant nor a highpressure plant produces gasoline from coal in Germany any more. The highpressure part of the latter is converted to the production of ammonia and alcohols, while the distillations and tanks in combination with an added catalytic cracker are operating on petroleum. Many cost estimates made by competent engineering firms, based on the results of improved versions in pilotplant runs, have been published. They all show that the present cost difference between petroleum and coal and also the costs of steel and labour do not allow to reduce the cost of liquid fuels from coal by hydrogenation to a level competitive with petroleum products, except perhaps for some chemicals. The present state of the Technology reflects only the fact that the chemistry of coal is subject to the chemical equilibrium and to the kinetics of the high molecular substances and the overall thermal efficiency. With the production and distribution facilities of the petroleum industry firmly estabished in the U.S. and with the high demand for roadpaving asphalts and electrode coke without competition from the bituminous coal tar as in Germany, the hydrogenation of refinery residua is not needed and coal hydrogenation would at best be regarded as a supplier of artificial crude for the refineries. But this viewpoint makes coalhydrogenation economically even less attractive as the higher profit margin is shifted to the finished products from the refineries.

Without subsidies or taxconcessions a highpressure hydrogenation facility could now be viable only if chemicals are the mainproduct and only the "byproduct"fuels are sold to the refineries at higher prices than crude, as they require just the finishing and distribution operation.

In mid 1976 the Austrian Iron and Steel Works quoted a price of a pressure vessel for 330 kg/cm^2 of 1.6 m ID and 26 m free length, corresponding to a reaction volume of about 40 m^3 and a weight close to 300 tons, which the railways can still transport on two cars, Can$ 537,600.- fob Montreal. The hot separator of 2.3 m ID and 16 m length would cost $ 698,300.- and one heatexchanger of 0.9 m ID and 20 m length $ 148,400.-, with bundle about $ 248,400.-. Transportation farther inland, installation, insulation, quenchgas- and other fittings installed but without connecting piping will probably increase these costs by a multiple of 3 to 4. Adding another multiple for highpressure pumps, auxiliary pumps, lowpressure equipment, structures, foundations, valves, instruments and portions for gantry- and pumphouse cranes, utility distribution system, lighting and ventilation etc. brings the cost per liquid phase system to about $ $57x10^6$ and installed to $ $124x10^6$ including the H_2-compressors.

Table 10 shows a compilation of the approximate installed cost of the equipment for one system, containing 4 vessels and other equipment to produce about 60.89 ton/hr lifuels from 77.05 ton/hr of good coal. For the production of 100,000 bbl/d, corresponding to 600 ton/hr product of an average sp.gr. 0.903, 10 such systems or stalls would be required. Table 10a presents an estimate of the probable manufacturing costs. These estimates are not meant to be accurate but are compiled for comparison only. They show that the cost of synthetic crude from coal is significantly higher than petroleum at present, that the capital costs are more than 50 % of the total manufacturing costs and that the cost of reaction space is only about 13.4 % of the total highpressure equipment and only about 3.2 % of the entire equipment.

Many attempts have been made to reduce the investment costs by increasing the speed of the reaction by using higher pressures or more active catalysts. The figures in table 10 indicate, however, that the savings so effected are negligible and are very probably overcompensated by increased costs in other departments.

Table 11 shows that the influence of different kinds of the most commonly used catalysts is negligible as could be expected from the structure of the high molecular coal and with this the size even of its depolymerization products 8).

Real savings in investment as well as operating costs could be achieved, however, if by quite different and yet cheap improved discardable or reclaimable catalystsystems the number of operating steps could be reduced, for instance as with the combi hydrogenation process. Combinations of Sn with chlorine have been used in the past at 300 atm with good success. Increasing the chlorine concentration, but without the corrosion, or eventually extracting and recycling the vanadium, which occurs in coal as well as in petroleum, from the ash could offer one of many possible solutions. Combihydrogenation of coal in one step to gasoline and middleoil, which would require only a finishing operation by standard refineries has been done on a pilot scale only at 700 atm. Only one plant in Germany processed successfully heavy oil residua in a 300 atm operation on a commercial scale to a finished product.

If combihydrogenation of coal could be perfected to work at 300 atm or lower, then such a plant would require for the same output 13 stalls, each containing 3 liquid- and 2 vapour phase vessels, i.e.65 vessels instead of 40. As everything else remains the same the investment increases only by about 6.9 %. The costs are then about as given in table 12. The figures are intended only to show the relationships of cost factors and how they can influence the economics of a hydrogenation facility. If the combihydrogenation of coal could be improved so that a sufficient lifetime of the adjoining fixed bed catalyst can be achieved or a catalyst regeneration is made possible at 300 atm, then for a cost increase of only 1.6 % a finished product could be manufacured, having a sp.gr. of about 0.863, which would have a much better chance of competing with standard refinery products from petroleum than the socalled synthetic crude, which is only a low priced refinery feed.

2) Auxiliary Operations

Besides new developments in the highpressure operation itself by reducing the number of operating steps or other improvements, the lowpressure auxiliary operations offer more chances for improvement. One of the costliest items was and is the manufacture of the hydrogen and its compression. It has been shown by several investigators 7) that carbonmonoxide has a beneficial influence on the conversion of coals, especially of lower ranks, to benzene soluble substances. Coal precursors like lignin and cellulose were found to be even more reactive than coals. Hydrogen, with exception of dehydrogenation of hydrocarbongases, cryogenic separation or by electrolusis, is always made by CO-shift conversion of generator- or reformergas, which contains 15 to 42 % CO. Using non-converted gas saves the cost of the conversion and eventually improves the hydrogenation. The H_2S-scrubbing yields an offgas less diluted with CO_2, which is beneficial for the Claus-process.

The conversion of coal to liquids by using mixtures of hydrogen with carbonmonoxide, but not with water,as Franz Fischer did, has been investigated with promising results in Ludwigshafen. At that time, however, the resultant higher asphalt level in the sludge and pasteoil would have caused an uneconomical enlargement of the residuum treatment plant and also difficulties in the kilns. Processing the sludge in a fluid coker instead of centrifuges would have eliminated this problem and would have allowed the use of purified, but not converted generator- or reformergas directly with the accompanying cost savings.

The quantity of hydrogen, which can be produced from the light offgases cannot supply all the hydrogen required and must be augmented by hydrogen from other sources, preferably from coal or better still from char, as the tar so obtained augments the liquids yield from the hydrogenation. The gasification of solid fuels can be done more economically at pressures of 30 up to about 70 atm, as the throughput is approximately proportional to the pressure. This method saves the first three or even four stages of the compression and reduces

~~the~~ its capital and energy costs by 50 or 60 %. The gasification of coal is always preceeded by its carbonization and both reactions can be combined for instance in the Lurgi-Ruhrgas gasifier, which yields more tar than corresponds to the Fischer assay.

3) The Residuum Treatment.

An other costly operation ~~which was~~ environmentally critical and operationally unpredictable, was the processing of the sludge. It was a necessity in order to recapture the oil in it for recycling. The centrifuges for unknown reasons must be shut down and disassembled sometimes after less than one hour of operation. They were closed but due to the frequent repairs the smell of the highly aromatic heavy oil could not be overlooked and forced the installation in open buildings.

The powdery solids discharge from the rotating kilns had sometimes a poor wettability and dust could not always be supressed. High asphalt contents in the centrifuge residue caused frequently ~~coke~~ caking formation in them. Also the waterlocks required attention in winter.

Without doubt the processing of the sludge could be replaced advantageously by the fluid- or flexi coker or the Lurgi-Ruhrgas gasifier, eventually in combination with a vacuumdistillation. About 66 wt% of the organics in the sludge are obtained as oil with a sp.gr. 1.06 and added to the pasteoil and 15 wt% with a sp.gr. 0.834, which is added to the product of the liquid phase. About 7.8 wt% is obtained as hydrocarbongas, separate from the watergas made by oxygen-steam blast.

By not recycling solids with the pasteoil and by replacing about 14 ton or 23.1 % of the product by tar from the gasifiers the quantity of sludge is reduced from 113 to about 49 ton/hr per system i.e.to about 44 % of the newly formed sludge. If this sludge contains as before 20 % or 9.75 ton/hr solids, consisting of 3.47 ash, 2.44 catalysts and 3.86 pyridine insolubles, then the 38.98 ton oil remaining are decomposed to 31.58 ton tars distilled, 3.03 ton hydrocarbongas and 4.37 ton additional coke. The total 8.23 ton maf coke on gasification yields 2912.5 Nm^3/hr per system clean watergas, corresponding to 2766.9 Nm^3 H_2 or 3.08 % of the requirement. The raw watergas contains 36 vol% H_2,42 % CO, 18 % CO_2 and 2 % CH_4. 0.35 ton/hr Oxygen and 3.29 ton/hr steam are used. The steam is preheated in heatexchange with the watergas discharged. Raw coal can be gasified alone or with up to 12 % sprayed on sludge as the circulation of the heatcarrier-coke is 5 to 9 times the quantity of fresh material fed to the process. One such system can process up to 100 ton/hr sludge and produce about 65 ton/hr distillate, i.e.5 such systems would be sufficient for 10 stalls. The cost per stall would be (Nov.74) about $ $6.16x10^6$.

As 3 % of the hydrogen can be produced from the sludge and about 40 % from the hydrogenation offgases, 57 % must be made from 56.961 ton/hr of high-ash coal. This quantity yields about 61,228.2 Nm^3/hr purified watergas, which together with the 46,189.7 Nm^3/hr from the offgases and sludgecoke gasification make up the total of 107,417.9 Nm^3/hr per system for 60 ton/hr product. Simultaneously from the 56.9 ton/hr gasification coal plus the 97.13 ton/hr powercoal 12.65 ton tars are produced besides 0.611 ton/hr ammonia and 1.46 ton/hr of BTX's. The tar yields on hydrogenation about 11.57 ton refined product. If the heavy portion of the tar is added to the pasteoil and so fed to the hydrogenation the hydrogen consumption is reduced from 122,322 to 107 417.9 Nm^3hr for the same quantity of product.

simultaneously the 73.45 ton/hr sludge, obtained when using solidsfree pasteoil would be reduced to 48.73 ton per hour per system, which is still too much to be sprayed onto the 154 ton/hr power + gasification coal. However, if vacuumdistilled to about 54 % solids, which is still pumpable, the 18.06 ton remaining would be only 11.7 % of the coal to be gasified. By spraying it onto the coal it might eventually be possible to dispense with the sludge carbonization entirely and with some environmental problems too.

In 1943 it was observed that when injecting 1 to 1.8 m^3 water into the liquid part of the hot separator instead of before the following heatexchanger, not only was the temperature of the liquid reduced from 425 to about 360 °C but simultaneously the centrifuges could be operated for about 6 months without disturbance and the separation was remarkably improved. When middleoil and more water of pH 6.8 was added to the sample of this sludge in a graduated cylinder and having it stand for a day it separated into 3 layers, the middle one transmitting some light and at the bottom a high solids sludge. The reaction of the sludge with the water at its critical temperature seemed to coagulate the solids around the inorganics and by some steam-cracking to reduce the molecular weight of the oil. Both reactions must, however, not be driven too far, otherwise coke is formed in the hot separator. This process could also reduce the quantity to be treated in the residuum plant considerably.

4) Preconditions for a Hydrogenating Facility.

The energy consumption of the U.S. in 1980 is estimated at about $25x10^{15}$kcal, which will be supplied to about 45 % by petroleum derivatives. If the requirement for transportation is reduced by better public transit systems and more economical cars from 25 to 20 % of the total i.e. to $5x10^{15}$kcal then this corresponds to 10.342 Mbbl/d of transportation fuel. If ~~only 10% of~~ this quantity shall be replaced by liquids from coal it would necessitate the construction of 10.3 plants of the capacity mentioned, equivalent to a requirement of about 669.5 reaction vessels already of 300 ton each. The other highpressure equipment and lowpressure parts require a multiple of this. The total steel requirement may reach eventually 1.26 Mton. As a comparison some figures of the prewar Germany shall illustrate this: the designcapacity of all 12 German plants was 4 Mton/yr of specification fuel. This required 4,000,000 ton + 8000 hr/yr + +0.055 ton/m^3 + 10 m^3/vessel = 910 vessels. These vessels were manufactured from 1937 to 1943 i.e. in 6 years, corresponding to a production of almost one vessel every other day besides all the other equipment. The U.S. requirements could eventually become about 1.85 times this quantity of steel in this particular form and this cannot be overlooked.

For the production of 100,000 bbl/d or 600 ton/hr 10 or 13 systems respectively, having a capacity of 60 or 46 ton/hr each and requiring 160 or 200 m^3 reaction space would be needed. Such a plant would consume about:

770.53	ton/hr coal	for hydrogenation
569.61	"	for hydrogen production
970.03	"	for power and fuelgas
2310.17	"	total coal consumption

The annual coal requirement is then at 8000 hr/yr 18,481,360 ton. In view of the high investment costs and from past experience with the thickwalled equipment, a lifetime of 40 years instead of the usual 25 can be anticipated for most of the pieces. The coal deposits in the vicinity of such a plant must therefore be about 700 Mtons. Deposits of this magnitude are not easy to find. The coal to be mined for 103 plants of the size mentioned would have to be per year about:

$$103 \text{ x } 2310.17 \text{ x } 8000 = 1{,}903{,}580{,}080 \text{ ton,}$$

which indicates that the present coal production must be increased by about 80 % and if the increased steel requirements are included 82 % in order to meet the demand and would probably lead to a substantial price increase for the coal, not to mention the wage demands of the unions.

Besides coal an adequate water supply is an absolute necessity. Alone the water for the production of the required hydrogen is about 261.9 m^3 chemical and 885.6 m^3 for cooling i.e. about 4210 U.S.gal/min. This water is only a portion of the approximate 157,100 m^3/hr or 691,000 gal/min cooling water, which will probably be replaced by partial air cooling.

5) Comparison with other methods of producing transportation fuels from coal and possible future developments.

From the $25.2x10^{15}$kcal/yr total U.S. energy consumption, estimated for 1980, $5.5x10^{15}$kcal/yr are from solid fuels and $17.5x10^{15}$kcal/yr are from oil and gas. To replace the latter by coal would require at 85 % efficiency $20.6x10^{15}$kcal/yr from coal. If all the coal, i.e. $26.1x10^{15}$kcal/yr would be carbonized and gasified in combination with power generation or metallurgical reduction, then the tar so obtained constitutes an average of 14 % of the heatvalue of the coal or 4.17×10^{15} kcal/yr. Converting this tar in a 150 atm combi-hydrogenation at an efficiency of 90 % to transportation fuels could provide 3.75×10^{15} kcal/yr or 74.5 % of the estimated U.S. requirement in 1980. this short calculation shows, that if all petroleum and natural gas and also shaleoil would be exhausted ~~used up~~ and all other methods to produce the needed transportation fuel from coal other than by hydrogenation would be exhausted, then about 25 % of it must be manufactured by processes other than carbonization. The high-pressure hydrogenation could well play a role in bridging this gap.

The German highpressure plants have shown an overall thermal efficiency of about 30 to 35 % including LPG's as compared to 27 to 30 % of the Fischer-Tropsch plants. The 100,000 bbl/d of finished fuels, discussed here, is based on available commercial technology and shows a thermal efficiency of 45 % and 53 % if LPG's are included. It requires an investment of about \$ 3×10^9. This compares, however, to only about \$ 450×10^6 for the production of 100,000 bbl/d of tar besides gas and char by carbonization.

The problems of deep-mining - one crippling accident per 1000 manshifts and one fatality per 3000 manshifts and only 5 ton production per manshift versus 80 to 100 ton per manshift in the stripmining industry - has induced the research in underground gasification as a means to reduce the cost of energy and to increase the safety. So far these tests have shown that the costs of recovery are not lower, especially if oxygen and water is used as gasification agent to produce synthesisgas for the Fischer-Tropsch synthesis. The underground gasification experiments in Czechoslowakia have also shown that the surface subsidance is too difficult to control and requires extensive land reclamation.

The coal extraction in a smaller plant in Germany was abandoned in favour of processing heavy residua. The difficulties in the filtration had not quite been overcome. It is, however, not entirely impossible to use this method to dissolve the coal in situ, especially those layers, which are more than 1200 m below the surface, where the temperature is higher than 45 °C and are therefore inaccessible. As the overburden has an average sp.gr. 2.0, the pressure, which could be applied in such a depth is about 240 atm, quite sufficient for hydrogenative extraction. Heating of the coal and solvent to 400°C requires about 580 kcal/kg coal or 10 % of its heatvalue.

The early experiments of Franz Fischer have shown that a substantial portion of the coal can be liquefied at temperatures of 400 °C at pressures of 100 to 140 atm with carbonmonoxide and alkaline water. This reaction also could eventually be used to recover the heatvalues of coal as liquids. As the solvent is a liquid it would eventually make a better control of the surface subsidance possible.

Conclusion

This short report is meant to illustrate the technology and the role, which eventually can be played by the highpressure coal liquefaction in providing a portion of the U.S. requirement for transportation fuels. At the present petroleum prices it cannot yet compete with standard refinery technology. But combination with other energy-, metallurgical- or ceramic-industries can improve the economics of a highpressure facility to a level, where it could become competitive, if the pricedifferential between petroleum and coal continues or widenes and if the environmental restrictions force the use of carbonization-gasification of the coal before burning it. With continued efforts towards improvments the hydrogenation of coal could eventually serve to bridge the shortages of liquid fuel supplies expected in the not too distant future and until the investment costs for renewable energy production are reduced to a level, more competitive with petroleum products.

The development of the coal liquefaction from the early investigations to commercial plants has been discussed. Possible improvements and eventual tradeoffs against disadvantages have been mentioned in conjunction with a discussion of the chemical fundamentals of these processes. A relative cost comparison of one improved production scheme, based on available commercial technology is presented to demonstrate the economic influence of some variables. Possibilities of the utilization of the experience aquired for future developments, for eventual combined operation with other industries and for underground energy recovery are outlined.

Contrary to coal carbonization and coking, which is always performed at ambient pressure, the other two types of coal conversion, i.e. the gasification and liquefaction are operating at elevated pressure and require gastight enclosure of all reacting materials being processed in order to avoid losses. The coal preparation was performed in completely enclosed crushers and grinding mills. In this operation no cancerous diseases were ever observed due to the absence of coaldust. Contrary to this in deepcoalmines lung diseases occur frequently. All buildingswere designed so that no gases or vapours could accumulate and stuffing boxes were vented to the outside and to special conduits. If the centrifuging and kiln operations were replaced by vacuumdistillation and the residue gasified,any health hazard would be eliminated.

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Table 1a.

Influence of Hydrogen-Pressure and Reaction-Time on the Liquifaction of Coal.

Initial Hydrogenpressure kg/cm^2	Reaction Temperature °C	Reaction Time min.	Benzene-insolubles wt% of maf.
70	400	5	91.96
"	"	35	63.12
"	"	65	37.01
140	"	5	95.59
"	"	35	37.04
"	"	65	17.63
210	"	5	85.12
"	"	35	22.44
"	"	65	7.84

Table 1b.

Influence of the Reaction -Temperature and -Time on the Liquifaction of Coal at 175 kg/cm^2 initial Hydrogenpressure.

Reaction Temperature °C.	Reaction-Time min.	Benzene Insoluble wt%
400	20	57.2
"	50	28.4
"	80	10.9
"	140	4.1
"	200	2.1
420	20	31.1
"	50	7.2
"	80	3.5
"	140	1.4
"	200	2.1
430	20	9.5
"	50	2.7
"	80	1.4
"	140	2.1
"	200	1.6
440	20	9.1
"	50	2.1
"	80	1.6
"	140	1.9
"	200	1.2

Table 2.

Quantities in the Liquifaction of Characteristic Materials to 100 ton/hr Auto-Gasoline.

Feed or Product for 100 ton/hr		Units metric	Bituminous Coal to: Middleoil only	Bituminous Coal to: Middle-+ Heavyoil	Lignite to Middleoil only	Coaltar-Residue to Middleoil	Lignite-Tarresid. to Mdloil	Asphalt-bas.Crude to Mdloil
Feedmaterial dry		ton/hr	189.0	192.7	242.4	144.2	139.5	135.7
Feedmaterial maf		ton/hr	179.2	182.7	213.6	142.8	139.5	135.7
Operatng.Pressr.		kg/cm^2	700/300	700/300	230/230	700/300	230/230	700/300
Chem. H_2 100%	Liqd.Ph	Nm^3/hr	176,200	176,490	158,700	124,900	52,050	54,920
	Vapr.Ph	Nm^3/hr	54,930	49,780	53,400	70,700	56,530	38,230
Total H_2 97.5% incld.dissolved		Nm^3/hr	289,500	287,300	243,400	233,850	126,150	112,350
React.Vol required	Liq.Ph	m^3	491.0	374,4	394.0	539.5	318.0	317.5
	Vap.Ph	m^3	280.6	236.1	282.0	338.0	286.0	155.2
Quantity Distilled	Liq.Ph	ton/h	247.6	318.8	289.3	256.4	300.6	242.0
	Vap.Ph	ton/h	210.2	193.4	180.3	237.3	213.3	136.6
Quantity	Centrifuged	ton/h	266.0	348.3	226.0	-	4.4	16.8
	Carbonised	ton/h	63.3	62.8	136.1	25.6	3.1	5.8
Hydro-Carbon Gases	CH_4	ton/hr	11.31	10.90	7.39	10.41	5.55	5.43
	C_2H_6	ton/hr	9.41	10.01	6.81	9.67	6.73	3.52
	C_3H_8	ton/hr	14.72	15.01	11.11	14.19	10.57	9.04
	C_4H_{10}	ton/hr	13.73	13.74	14.17	14.21	16.24	15.02
Knock Value of 105ton/h Gasoline(C_4+)		C.N.M.	72	72	66	74	64	67

Table 3.

Comparison of Productfractions from Hydrogenation and Carbonisation of Coals and Shale.

Fraction of	Lignite-Coal		Bituminous-Coal		Shale-Oil	
	Hydrogenation	Carbonisation	Hydrogenation	Carbonisation	Hydrogenation	Original
Rawgasoline						
sp.gr.kg/l	0.738	-	0.740	-	-	-
% Phenoles	6	-	9	-		
% Naphtenes	28	-	45	-		
% Aromates	18	-	17	-		
O.N.R.	62	-	70	-		
Middleoil:						
% Phenoles	14	24	19	36	13.8	19.0
fractn. 70-240°C						
sp.gr.kg/l	0.878	0.899	0.950	0.945	0.914	0.938
Aniline pnt °C	24	15	-15	-6	20	8
Cetane No.	-	-	-	-	41.5	39.5
fractn. 280-310°C						
sp.gr.kg/l	0.906	0.912	0.980	0.975		
Aniline pnt. °C	33	31	-14	-10		
Heavyoil fractn. + 325°C						
sp.gr.kg/l			1.070	1.040		
% Phenoles			2	7		
% Asphalts			1.5	3.5		
Viscosity °E			17	35		

Table 4

Significant Data on the Vapourphase Hydrogenation of 100 ton/hr Liquidphase Products.

Operatimg Pressure kg/cm^2	300 Refining	300 Splitting	300 Refing+Splitt.	700 Aromatisatn.	300 Refing + Splitt+60DHD
Operating Temp. °C	390 - 420	400 - 430	390 - 430	480 - 500	DHD:500-530
Catalyst	$Al_2O_3/WS_2/NiS$	Terrana/WS_2	$Al_2O_3/WS_2/NiS$ + Terrana/WS_2	HActvtd Clay +Zn/Cr/Mo/S	Clay+MoO_3
Reaction Vol. m^3	100 - 170	95 - 135	305	218.3	305 + 350
Feed Material	Liquidph.Middleoil+Gasolin	Refined+Recycl.Middleoil	Liquidph.Middleoil+Gasolin	Liquidph.Middleoil+Gasolin	Gasolinfract. 85 - 250°C
" " ton/hr	100	112.1	212.1	257.6	212.1 + 91.0
Recyclegas Nm^3/ton	3500-4000	1500-2500	5750	2500	5750+1500
Hydrogen Consumption(97.5%)Nm^3/ton	375	212	675**	409**	288 less 80
Hydrocarbongas,wt%	3.03	9.12	19.96	16.9	30.2
Avg.C of H-C gas	3.09	3.65	3.48	2.39	2.39
Product distilled C_4+ ton/hr	96.6	104.0	200.6	236.6	286.5
-C_4 gas ton/hr	0.39	1.82	2.21	3.42	6.98
Gasoline(C_5+)produced ton/hr	26.3	60.9	81.5*(aviatn)	82.5*(aviatn)	73.7(aviatn)
Raw-Gasoline O.N.R	-	74.5	74.5	79.0	-
Aromatescontentvol%	-	5	5	48.0	66.0
Middleoil produced ton/hr	70.8	41.3(recylced)	41.3	157.6	-

* includes Gasoline from Liquidphase. **includes dissolved Hydrogen.

Table 5

Ratio of Iso-Butane to Total Butane in Hydrogenation Products

Liquid Phase	10 - 20 %
Refining Vapour Phase	50 %
Splitting Operation	75 %
Aromatization (700 at)	20 %
Dehydrgenation (50 at)	35 - 40 %

Table 6

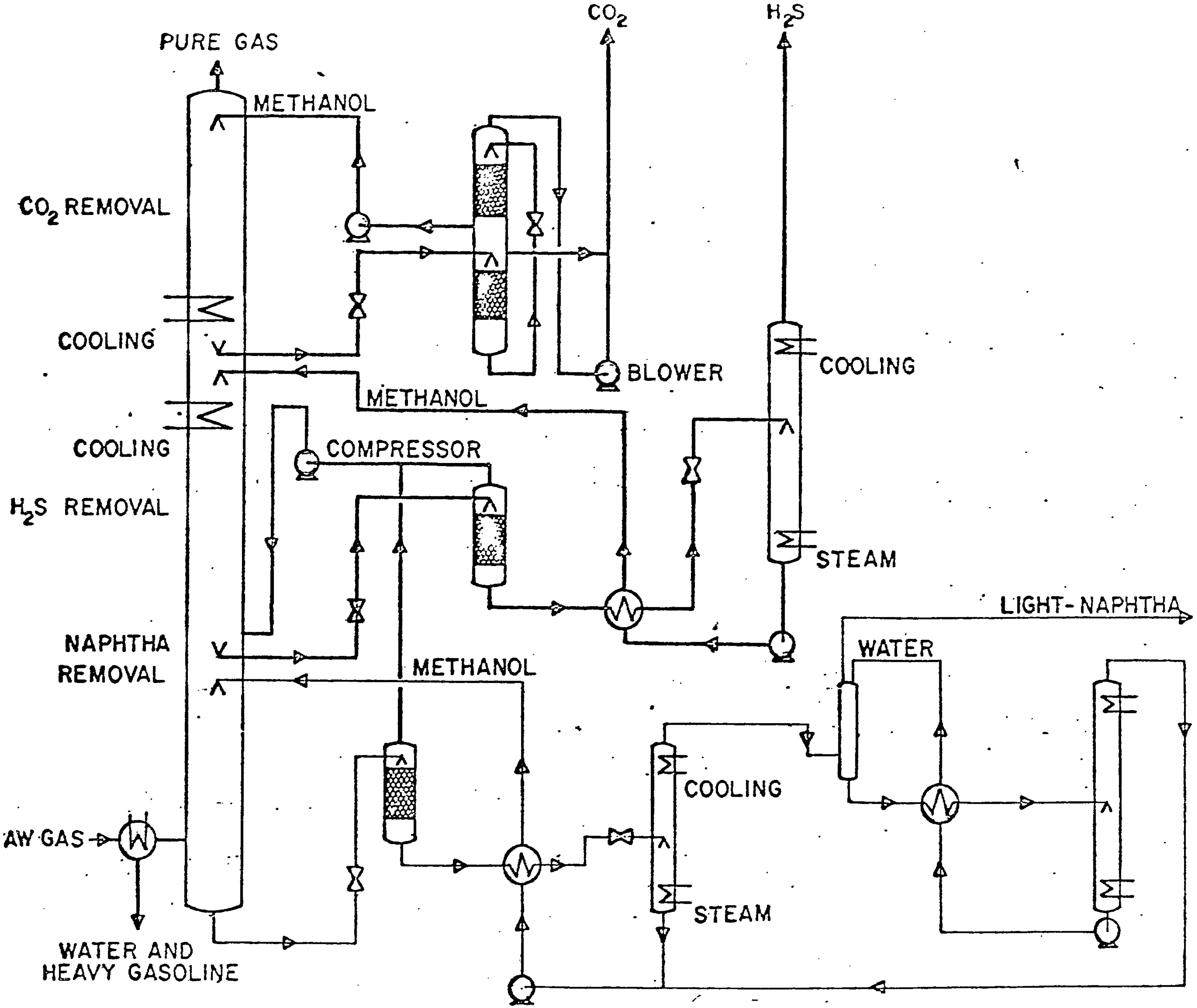
Olefines Content in Cracking Products

	C_2	C_3	C_4+
Thermal Cracking	10	33	45
Catalytic Cracking	35	45	55

Table 7

Percentage of total Hydrogen Consumption, which can be manufactured from the Methane and Ethane in the Hydrogenationoffgases from various Starting materials in % of total Hydrogen required.

Bituminous Coal	Lignite	Residua from:		
		Bituminous Coaltar	Lignite-Carbonisation Tar	Asphalt-basic Crude
40	30	43	46	42



METHANOL PROCESS

Comparison of Various Desulphurization Processes

Per 1,000 Nm^3/Gas	Pressurized Water	Rectisol (Methanol)	Purisol (N-methyl-pyrrolidone)	Shell Adipic Acid Process	BASF-Alcazid $C_2H_6NCH_2CO_2H$	Ethanolamine CH_5O-NH_2	Hot Potash Solution	Ciammarco Vetrocoke Na_3ASO_3	Stretford, 2, 7 Anthraquinone disulfonic Acid
Circulated Solvent m^3	50 to 100	0.5 to 1.0	2.5 to 3.2	3.5	0.7 to 2.0	4	0.05 to 0.1	10 to 15	12 to 20
Electric Power kWh	25 to 50	20 (including refrigeration)	4.5 to 6.0	3.5	2	3 to 5	0.04 to 0.07	12	5 to 20
Cooling Water m^3	--	14	1.5 to 2.0	4.5	$25m^3$/ton $	corresponding to steam consumption	very little	little	some
Steam kg	--	corresponding to cooling water	30 kg	170 kg	1.5-3.0kg/kg CO_2 4-20 kg/kg S 200 kg/$1000m^3$	200 to 400 kg	1.6 to 2.2 kg	30 kg	20 to 50 kg
H_2S/CO_2 Inlet Vol %	--	18/40 vol %	--	10 vol %	10/30 vol %	--	--	Max. 2.5 vol %	Max. 2 vol %
H_2S/CO_2 Outlet ppmv	--	1 ppmv	4 ppmv H_2S 2 vol % CO_2	1 ppmv H_2S 10 ppm org.S	100 ppmv (with carbon down to 1 ppmv H_2S)	6 ppmv	100 ppmv	1 ppmv	1 ppmv
Operating Temp °C	ambient	-65°C	40 to 70	--	125°C to 150°C	--	80 to 120°C	150°C	80°C
Regeneration Pressure kg/cm^2	ambient	ambient	10 atm, 140 °C	--	Full pressure*	--	Ambient Pressure	Full Pressure*	70 kg/cm^2
Sulphur Recovery	90%	92%	92%	92%	95%	92%	85%	97%	98%
N-methylpyrrolidone Adipic Acid, Alcazid kg	--	--	0.1	negligible	negligible	negligible	--	--	--
Methanol, Soda kg	--	0.45	--	--	--	--	--	70 kg	35 kg
Hydroquinone, Anthraquinone-sulfonic-acid, A_sO_3	--	--	--	--	--	--	--	2 kg	2 kg
$ Investment/1000 Nm^3 Hr Basis: 10^6 Nm^3/hr Power Factor 0,653	--	31,252.86	--	--	34,836.95	--	--	13,220.00	13,219.89
Additional $ Investment per ton/hr sulphur recovery Basis: 6 ton/hr S Power Factor 0,54	696,000 (Claus + tailgas)	696,000 (Claus + tailgas)	696,000 (Claus + tailgas)	696,000 (Claus + tailgas)	696,000 (Claus + tailgas)	696,000 (Claus + tailgas)	696,000 (Claus + tailgas)	290,000.00	$\frac{11,500}{1,000\ Nm^3/hr\ gas} + \frac{374,500}{ton\ of\ sulfur}$
$ Operating Costs/Ton S	--	98,57*	--	--	118.08	--	--	--	34.39

*With use of waste heat for refrigeration.

*1 to 25 atmospheres

Table 8

Comparison of Various Desulphurization Processes

r 1,000 Nm^3/Gas	Pressurized Water	Rectisol (Methanol)	Purisol (N-methyl-pyrrolidone)	Shell Adipic Acid Process	BASF-Alcazid $C2H_6NCH_2CO_2H$	Ethanolamine $CH5O-NH_2$	Hot Potash Solution	Ciammarco Vetrocoke Na_3ASO_3	Stretford, 2, Anthraquinone disulfonic Aci
rculated Solvent m^3	50 to 100	0.5 to 1.0	2.5 to 3.2	3.5	0.7 to 2.0	4	0.05 to 0.1	10 to 15	12 to 20
ectric Power kWh	25 to 50	20 (including refrigeration)	4.5 to 6.0	3.5	2	3 to 5	0.04 to 0.07	12	5 to 20
oling Water m^3	--	14	1.5 to 2.0	4.5	$25m^3$/ton $	corresponding to steam consumption	very little	little	some
eam kg	--	corresponding to cooling water	30 kg	170 kg	1.5-3.0kg/kg CO_2 4-20 kg/kg S 200 kg/$1000m^3$	200 to 400 kg	1.6 to 2.2 kg	30 kg	20 to 50 kg
S/CO_2 Inlet Vol %	--	18/40 vol %	--	10 vol %	10/30 vol %	--	--	Max. 2.5 vol %	Max. 2 vol %
S/CO_2 Outlet ppmv	--	1 ppmv	4 ppmv H_2S 2 vol % CO_2	1 ppmv H_2S 10 ppm org.S	100 ppmv (with carbon down to 1 ppmv H_2S)	6 ppmv	100 ppmv	1 ppmv	1 ppmv
erating Temp °C	ambient	-65°C	40 to 70	--	125°C to 150°C	--	80 to 120°C	150°C	80°C
generation Pressure /cm^2	ambient	ambient	10 atm, 140 °C	--	Full pressure*	--	Ambient Pressure	Full Pressure*	70 kg/cm^2
lphur Recovery	90%	92%	92%	92%	95%	92%	85%	97%	98%
methylpyrrolidone ipic Acid, Alcazid kg	--	--	0.1	negligible	negligible	negligible	--	--	--
thanol, Soda kg	--	0.45	--	--	--	--	--	70 kg	35 kg
droquinone, Anthraquine-sulfonic-acid, A_sO_3	--	--	--	--	--	--	--	2 kg	2 kg
Investment/1000 Nm^3 Hr sis: 10^5 Nm^3/hr ver Factor 0,6[illegible]3	--	31,252.86	--	--	34,836.95	--	--	13,220.00	13,219.89
litional $ Investment ton/hr sulphur recovery sis: 6 ton/hr ver Factor 0,5	696,000 (Claus + tailgas)	696,000 (Claus + tailgas)	696,000 (Claus + tailgas)	696,000 (Claus + tailgas)	696,000 (Claus + tailgas)	696,000 (Claus + tailgas)	696,000 (Claus + tailgas)	290,000.00	$\frac{11,500}{1,000\ Nm^3/hr\ ga}$ $+\frac{374,500}{ton\ of\ sulfur}$
perating Cost/Ton S	--	98,57*	--	--	118.08	--	--	--	34.39

th use of waste heat for refrigeration.

*1 to 25 atmospheres

Table 9

Cost of Hydrogen produced at a Rate of 53,000 Nm^3/hr by different Processes.

Cost Item in % of those of Naphta-Steam Reforming	Steam-Reforming of Naphta	Pressure-Gasification of Coal	Gasification of Coaldust	Steam-Iron Process (Medium-Pressure)	Steam-Iron Process (High-Pressure)
Erected Plant Cost	100	176	202	275	233
Working Capital	100	129	142	194	167
Raw Material Cost ***	100	49	39	58	43
Direct Operating Costs	100	292	331	123	160
Indirect Operatingcosts	100	173	198	269	228
Cost of Production of 1000 Nm^3 purified H_2	100	128	136	-	-
Cost of Compression of 1000 Nm^3 H_2 from Purification Exit to 25 at	100	33(?)	33	-	-
Cost of Production of Compressed H_2 (96%)	100	123(?)	130	102*	95**

* 99.8% pure Hydrogen at 32 kg/cm^2 ** 99.8% pure Hydrogen at 79 kg/cm^2

*** Naphta to Coal-Price = 5.2

Table 10

Cost of High Pressure Equipment for Conversion of 116 ton/hr Bituminous Coal to 60 ton/hr Liquid Products in 160 m^3 Reaction Volume.

4 Reactors, 26m lg, 1.6m ID, @ 537,600.-	2,150,400.-
3 Heatexchangers, 20m lg, 0.9m ID, @ 248,000.-	744,000.-
1 Hot Separator 16m lg, 2.3m ID, @698,300.-	698,300.-
1 Paste Preheater + 2 Blowers 1600 kW	2,661,819.-
Total hot Highpressure Equipmt.	6,254,519.-
100% +310% for Installation etc.	25,643,528.-
2 Cold Separators 8m lg, 1.6m ID, @163,500.-	327,000.-
1 Scrubber Tower 20m lg, 0.9m ID @ 248,000.-	248,000.-
2 Letdown Vessels 12m lg 2.4m ID @ 163,500.-	327,000.-
Total Cold Highpressure Equipm.	902,000.-
100% + 235% for Installation etc.	3,021,700.-
1.5 Gasreciculators 2800 kW installed	9,736,688.-
2.5 Hydrogencompressors, 25 to 325 at, @16,000 kW	32,836,560.-
4.0 Pasteinjectors 20 m^3/hr	4,603,984.-
2.0 Oil-Injectors 10 m^3/hr	690,597.-
2.0 Hydraulic Drivers + Turbine @ 1200 kW	270,400.-
1.5 Lowpressure Transferpumps @ 320 kW	8,950.-
1 Expansionmashine + 2 Auxiliary Pumps @ 1300 kW	496,000.-
42 Electrical Valves @ about $ 4030.- installed	169,260.-
408 Hand operated Highpressure Valves @ about $2125.-	650,760.-
Instrumentation + Automatic Conrolsabout	288,000.-
Portion of Gantry Crane, Pumphouse Crane + Power-Supply, about	115,000.-
Portion of Pressure Testing Basin, Emergency, Discharge Tower and Suction Tanks, about	84,000.-
Total Pumps and Machinery	50,220,599.-
Total Highpressure Equipment	78,885,827.-
Engineering 4%, Structures 12%, Piping 20%, Utilities 10%, Insulation 4%, Cleaning and Painting % Testing 2%; Total 57%	44,964,921.-
Total Installed Highpressure Equipment	123,850,748.-
<u>Portion of Lowpressure Equipment 10% of Total:</u>	
Gasification including Purification and Oxygenplant, Tarcondensation, Gasliquortreatment and Ash removalsystem as well as Coal Storage etc.	48,423,200.-
Powerplant 1[illegible] MW Combined Cycle	18,166,609.-
Power Distribution System	9,590,087.-
Sludge Carbonisation	6,157,675.-
Distillation	7,185,890.-
Vacuum Distillation for Sludge	2,553,473.-
Storage Tanks	3,053,526.-
Shipping Facilities	241,014.-
Site, Yardimprovements, Fireprotection etc.	5,706,114.-
Total Lowpressure Equipment installed	101,077,588.-
100% + 65% for Engineering, Structures, piping etc	171,831,900.-

Table 10 cont.

Cost of complete Highpressure Equipment	123,850,748.-
Cost of complete Lowpressure Equipment	171,831,900.-
Total Costs for one System for 60 ton/hr Prod.	295,682,648.-

Manufacturingcosts at 60 ton/hr or 480,000 ton/yr: Capital Charges: 10% interest+profit, 25% depriciation, 1.7% Taxes+Insurances, 3.8% Maintenance = 18% in $ per ton	110.881
Energies + Utilities (no coal)	15.539
Repairs, Replacements, Catalysts: 25% of Capital-charges	25.861
Labour: 0.4 Manhours @ $8.50 + 15%	3.910
Shipping Charges 15 of total	1.535
General Charges 0.5% of total	0.768
Total Manufacturing Costs	158.494
Cost of 3.7944 ton coal/ton product at $ 11.0 per ton	41.739
Total Manufacturing Costs	200.233
Manufacturing Costs per bbl about:	28.705

Table 11

Catalytic Activity of some Metal Compounds At an initial Hydrogenpressure of 100 kg/cm^2 and a Residence Time of 3 hours at 400 °C.

Catalyst	Catalystconcentration per 100 g Coal*		Conversion % of maf Coal	Asphalt g/g Coal convert.
Sn + NH_4Cl	1.5 + 0.375	(1.18)	92.2	0.476
$SnCl_2$	0.8	(0.5)	90.1	0.341
$(NH_4)_6Mo_7O_{24}\cdot 4H_2O$	1.0	(0.5)	89.4	0.818
"	1.0	(0.5)	73.5	0.597
"	1.0	(0.5)	76.3	0.586
$FeCl_3 \cdot 6H_2O$	5.0	(1.03	62.0	0.526
$FeCl_3 \cdot 6H_2O$ + $SnCl_2$	5.0 + 0.16	(1.03 + 0.1)	65.7	0.570
$FeCl_3 \cdot 6H_2O$ + $SnCl_2$	5.0 + 0.8	(1.03 + 0.5)	86.7	0.449
ZnS + NH_4Cl	2.0 + 0.5	(1.34)	51.0	0.720
$FeSO_4$ reduced with SO_2 (powder)	4.25	(1.5)	72.8	0.522
$FeSO_4 \cdot 7H_2O$ impregnated	9.15	(1.84)	82.2	0.652
No Catalyst**	-	-	67.7	-

---

* Figures in parantheses indicate metal concentration g per 100 g Coal

** Reaction was carried out in glass liner

Table 12.

Additional Costs for Expansion to Combusystem:

2/3 x 4 Reactors	1,433,600.-
100% + 310% for Installation	4,444,160.-
1.5 x 13/10 Gasrecirculators installed	2,921,006.-
2.5 Hydrogencompressors + 14% installed	4,957,118.-
Total Highpressure Equipment more	11,962,284.-
Gasification, Purification, Oxygenplant etc. 14%	6,779,248.-
Powerplant 7% more	1,271,663.-
Powerdistributionsystem 7% more	671,306.-
Total more for Lowpressure Equipment	8,722,217.-
Total more for Combi Equipment	20,684,501.-
Total Costs for one System for 60 ton/hr Prod.	316,367,149.-

Manufacturing Costs per ton:

Capital Charges 18% / 480,000 ton/yr	118.638
Energies + Utilities, 7% more	16.627
Repairs, Replacements, Catalysts: 25% of Capital Charges	27.637
Labour 7% more	4.184
Shipping- + General Charges as before 1.5%	2.461
Total Operating Costs	169.547
Cost of Coal to Process 3.8507 ton/ton at $11.-	42.358
Total Manufacturing Costs	211.905
Manufacturing Costs per bbl about	29.032

Estimation of the Coal Requirement for the Generation of Electric Power or Fuel-Gas for the Production of 10,000 bbl/D liquid fuels from Coal by Hydrogenation.

Gelsenberg: 200 MW for 103 ton/hr coal to 55.8 ton/hr Gasoline

12+2 H_2-Compressors, 6800kW	81.6	MW
14+2 Hydraulic drivers 800kW	11.2	"
10=2 Booster-Compressors 600 kW	6.0	"
14+4 Gasrecirculators 550kW	7.7	"
10+4 Injectionpumps 550kW	5.5	"
2+1 Scrubberinjectors 550kW	1.1	"
12+2 Hotgas Blowers 450kW	5.4	"
Transferpumps, Instruments, Cranes, Lighting etc.	6.5	"
For Hydrogenation only ca.	125.0	"
For other Plant Sections	75.0	"

For one Liquid-Phase System for 60 ton/hr Product as described:

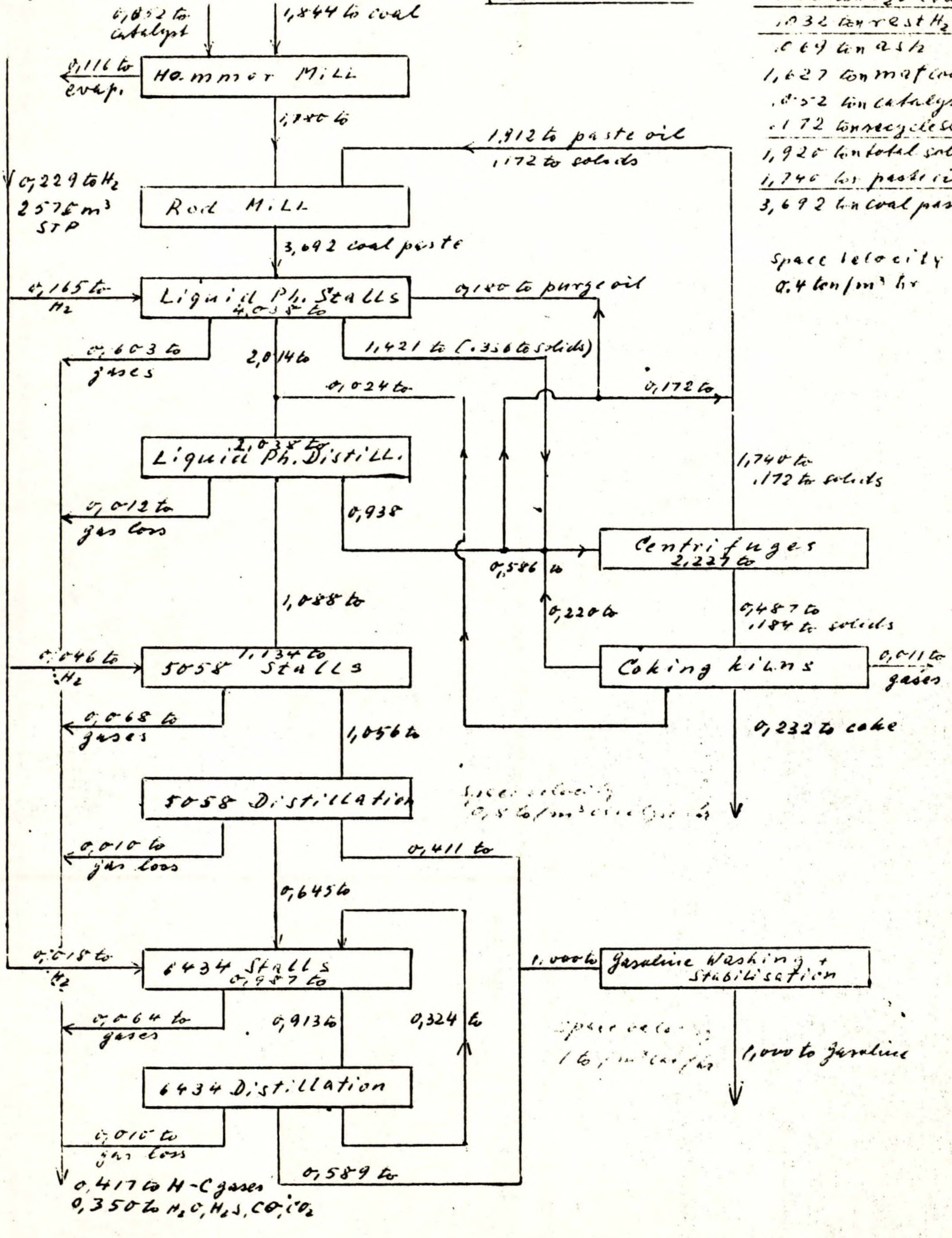
H_2-Compression from 25 to 325 kg/cm^2 for 108,000 Nm^3/hr	25.8	MW
Hydraulic drivers	4.8	"
Gasrecirculators	4.5	"
Productinjectors	3.5	"
Scrubberpumps	1.0	"
Hotgasblowers	3.9	"
Transferpumps, Instruments, etc.	2.4	"
Coalgrinding, Pumps in Distillation etc.	4.1	"
Estimated Total ca.	50.0	"

50+75=125 MW x 860 + 0.32 + 3462.202Kcal/ton = 97.030 ton coal for Powergeneration or Fuelgas.

Fig. 1.

Hydrogenation of low-ash bituminous coal (ton/ton Mot. fuel)

1,844 ton green coal
.116 ton H_2O evap.
.032 ton rest H_2O
.069 ton ash
1,627 ton maf coal
.052 ton catalyst
.172 ton recycle solids
1,920 ton total solids
1,740 ton paste oil
3,692 ton coal paste

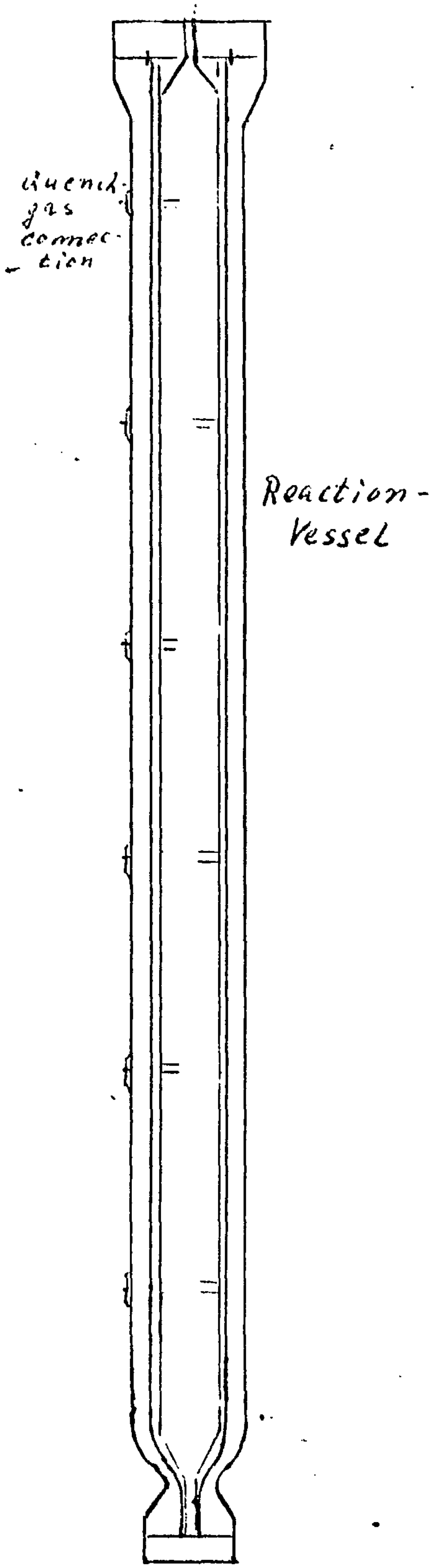


Space velocity
0.4 ton/m³ hr

Space velocity
0,8 to/m³ [illegible]

Space velocity
1 to/m³ [illegible]

Sketch B.
High-Pressure Vessels
for Coal-Hydrogenation



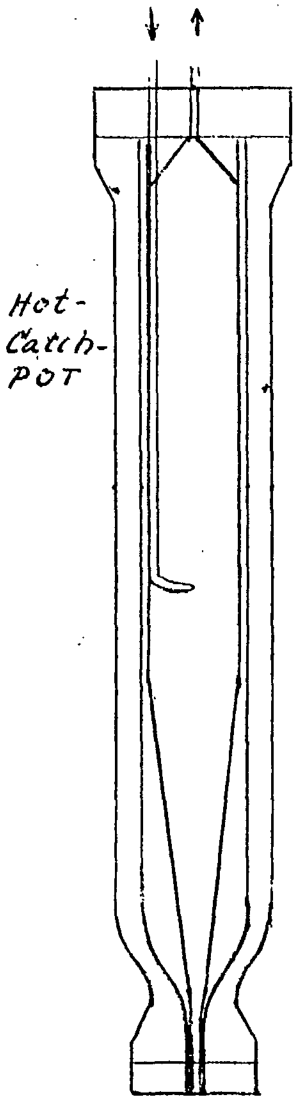


Diagram 3.

Flow Scheme for Hydrogenation of 231.04 ton Coal to 63.99 ton Liquid Fuels + LPG's (Quantities in ton/hr or Nm^3/hr).

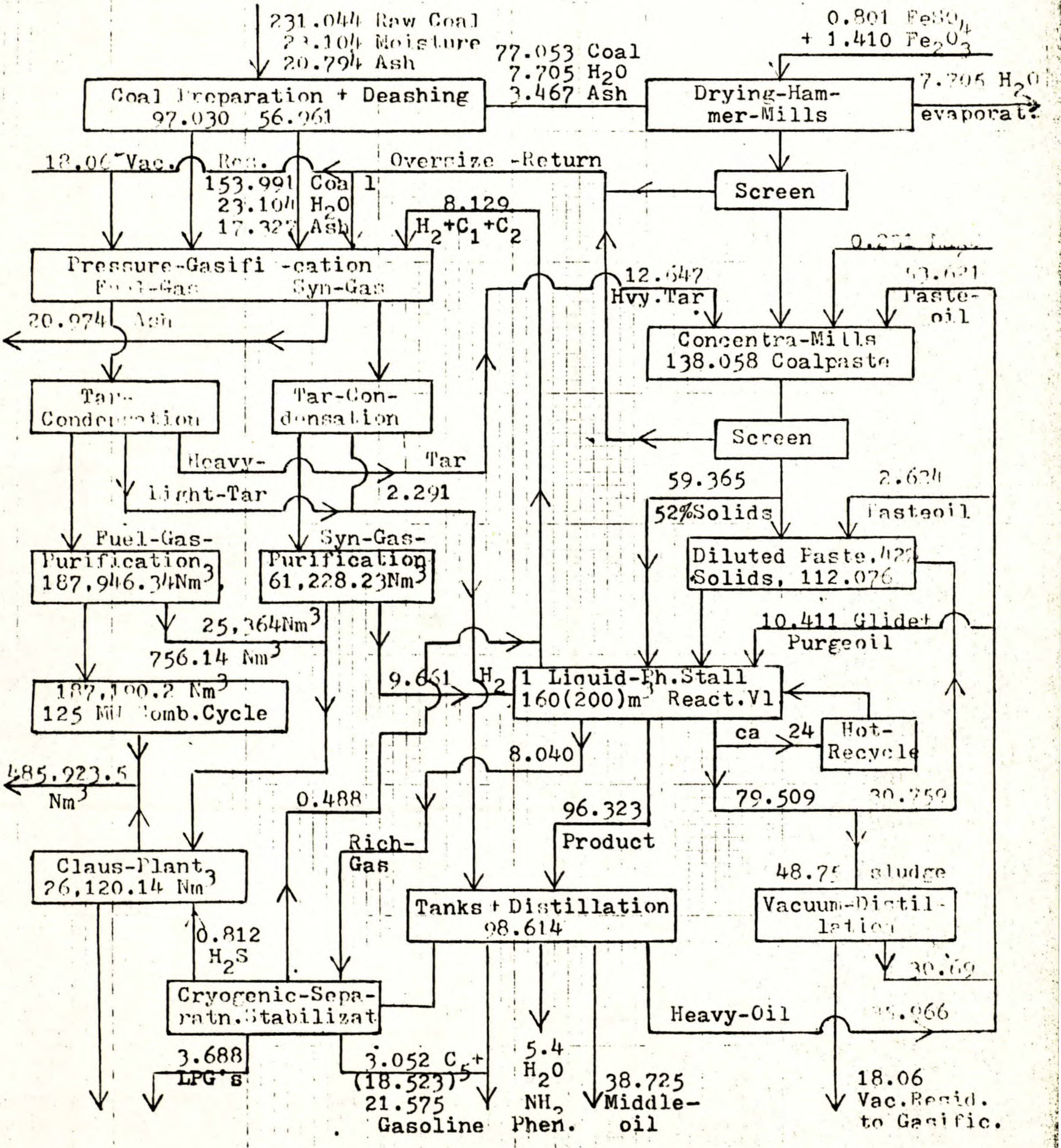


TABLE 6

COMPARISON OF THE PRODUCTION OF REDUCING GAS FROM COAL WITH VARIOUS TYPES OF GASIFIERS

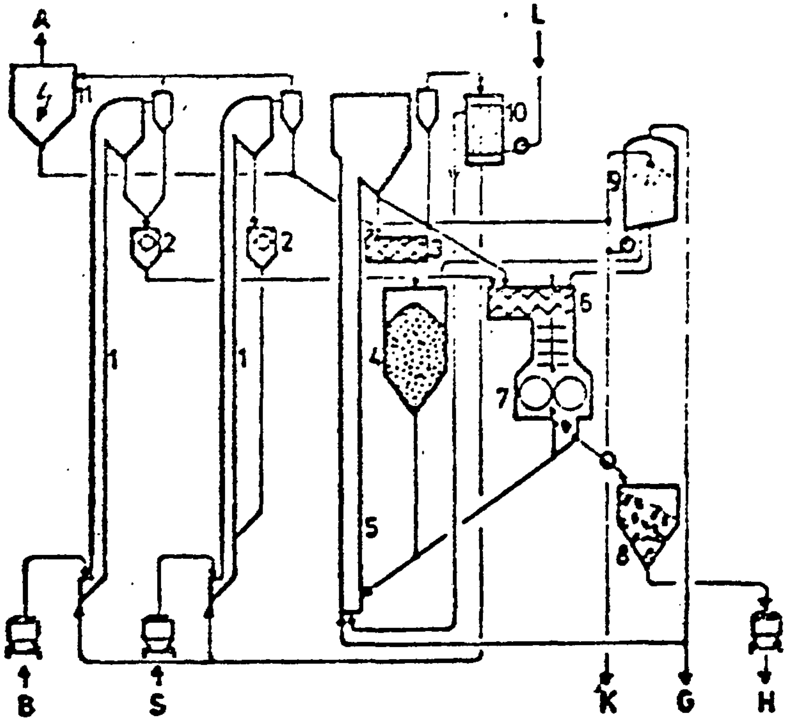
Gasification Process Licensor	Gravitating Bed Lurgi	Fluidized Bed Bamag Winkler	Entrained Bed Koppers Totzek	IGT*** Pilot Plant
Commercial Operating pressure atm	30.6	1.5	1.3	~75
Raw coal per gasifier, ton/hr	56.596*	36.561	35.167	66.842
Screen Size inch or Screen No.	<3 inch	90%< No.7 USS	90%< No.170 USS	80%< No.12USS
Maximum permissible ash content, wt/%	45	35	30	(9)?
Maximum permissible moisture, wt/%	20	8	4	6
Dry coal per gasifier ton/hr	45.249	33.636	34.500	62.831
Raw gas per gasifier Nm^3/hr	80,000	80,000	~50,000	80,170
Raw gas per ton dry coal Nm^3/ton	1,988.99	2,190.-	~1,500.-	1,199.4
Temp in gasification zone, °C	850	950-1000	1580	927
Raw gas exit temperature, °C	350-400	900-950	1440-1490	553
Raw gas analysis (depending on coal):				
Vol. % CO_2	28.44	18.9	10.83	19.09
CO	20.14	34.5	57.11	5.61
CH_4	10.30	2.9	0.1	51.99
H_2	39.27	42.8	30.73	21.20
N_2Ar	0.65	0.9	0.77	1.09
NH_3, H_2S, COS, CS_2	1.15	0.98	0.46	1.02
H_2 + CO in raw gas, vol. %	59.41	77.3	87.94	26.81
Net heat value of raw gas, kcal/Nm^3	2,481.6	2,268.89	2,524.22	3,811.94
NHV in raw gas as % of NHV of coal	91.46	75.53	66.57	65.63
Per Nm^3 raw gas required (approx):		(Leuna)		
Raw Coal kg	0.595	0.659 (0.625)	0.967	0.834
Oxygen Nm^3	0.156	0.653 (0.255)	0.404	0.269
Steam kg	0.472	1.521 (0.385)	0.321	0.449
Steam Generated kg	0.295	1.862 -	1.481	-
Steam Difference kg	-0.177	+0.341 -	+1.160	-
Heat loss on conversion of CO to H_2 to obtain a ratio of $3H_2$ to 1CO in kcal/Nm^3 raw gas	23,367	81,185	154,242	-
Reaction volume per gasifier, m^3	197.114	about 422	62.297	268.135
Raw gas per m^3 reaction vol. Nm^3/hr	405.856	236.967	722.346	298.989
Surface per kg feed coal, dm^2/kg	5.9336	41.5354	2,595.63	207.677
Coal in reaction space, kg	86,888.60	70,698	41.0282	50,071.44
Surface of Coal in Producer, m^2	26,155.67	29,326.79	1,065.076	103,986.9
Raw gas per m^2 coal surface, Nm^3/hr	3.441	2.7279	42.251	0.77096
Thermal efficiency	91.8	75.34	72.3	71.3
Installed cost per gasifier, $**	6,058,800.-	14,831,800.-	8,055,500.-	13,896,500.-
Installed cost per 1000 Nm^3/hr raw gas, $	75,747.-	185,398.-	161,110.-	173,339.-
Gross Heating Value of Coal, kcal/kg	4,444	7,192	3,396	7,333

*5.0 m ID

**Installed Cost per Gasifier + Gas Cleanup to 500 ppmv S + O_2 - Plant + Coal Grinding but excluded Steam plant and Coal storage according to an estimate by Dravo Co. in 1973.

***From Data in Report of Tzaros, Knabel et Sheridan, Oct. 1965, Hydrogasifier only corrected for inflation up to 1973.

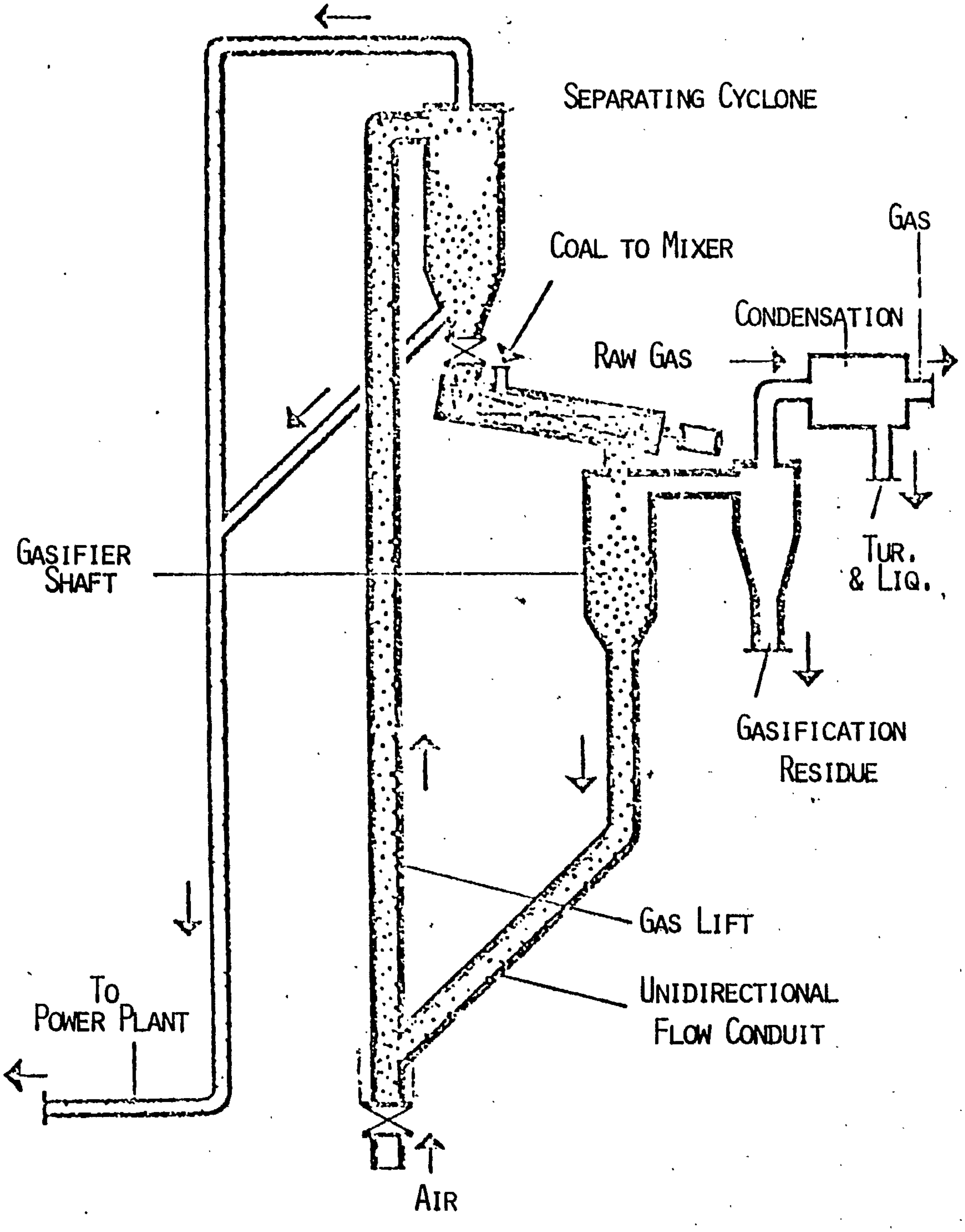
FIG. 2



FLOWSHEET OF THE BERGBAU-FORSCHUNG LURGI FORM COKE PROCESS

B	binder coal	
S	coal for charmaking	
K	oil	
G	coal gas	
H	hot briquettes	
A	flue gas	
L	air	
1	coal flash dryer	
2	crusher	
3	mixer	Lurgi-Ruhrgas-system
4	hopper	
5	air lift	
6	mixer	
7	press	
8	cooler	
9	condensation	
10	air preheater	
11	electrostatic precipitator	

FIG. 6



LURGI RUHR GAS PROCESS

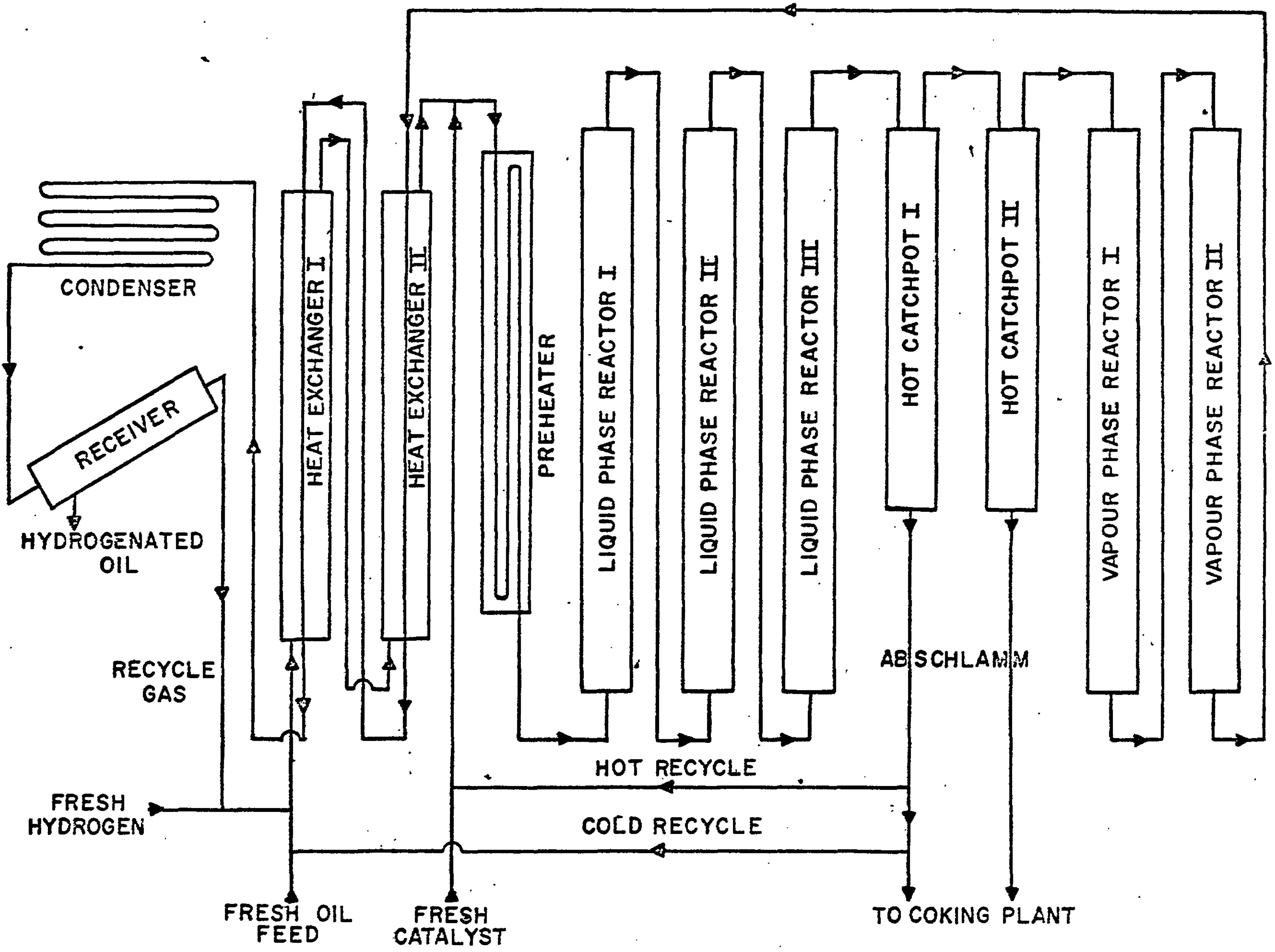


FIGURE 1 - SCHEMATIC DIAGRAM OF A COMBI HYDROGENATION STALL