

Section 11
SAARBERG COAL LIQUEFACTION DEVELOPMENTS

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Saarberg-Coal-Liquefaction Developments
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Summary

The Saarberg coal liquefaction process is a further development of proven IG-Farben technology. The main difference to the IG route 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. This system could be further improved in 1979. 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 is now ready for operation.

In January 1980 Saarberg began to plan a commercial coal liquefaction plant with a coal throughput of 2 Mio t/a. This plant could start operation in the second half of the eighties.

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Saarberg an important energy company in the South West of Germany has developed multiple activities particularly in the field of coal upgrading on the basis of own hard coal resources. Beside the classical upgrading processes like low temperature carbonization, coking and electricity generation Saarberg has been engaged since 1974 in process developments for the gasification and liquefaction of coal. When we made up our mind at that time to find out a suitable technology for the production of distillate oil from coal, the decision was made to take up the direct liquefaction route since in contrast to the indirect processes higher yields and higher thermal efficiency can be attained.

In 1974 the situation in the field of coal liquefaction was characterized by the fact that in the USA there were process-developments aimed at achieving coal liquefaction at comparatively low pressures of below 200 bar. These processes still under development are not yet proven on a commercial size. In contrast the original hydroliquefaction process of IG Farben was a proven route but required a very high pressure of 700 bar.

The objective of our development work was to find out ways to modify the IG process for a much lower pressure operation while retaining the proven principal of this route. Further objectives of an optimal liquefaction process were among others improving the thermal efficiency by reducing the hydrogen consumption, increasing the coal hourly space velocity and achieving a high reliability and availability of the process.

The first figure shows how 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. Here the coal was further milled in the presence of oil to prevent oxidation. The homogenized paste was then fed to high pressure pumps for injection into the heat exchangers. Wherever the described uncorrosive iron catalyst was used the discharge pressure of the injection pumps was 700 bar. In the heat exchangers slurry and hydrogen flowed up outside the tubes countercurrently the overhead vapors from the hot separator inside the tubes. Normally coal paste and hydrogen were jointly preheated to about 430° C in a subsequent gas fired heater. The gas slurry mixture passed through 4 vertical empty reactors in which coal liquefaction took place at about 480° C. The 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 the tubes of the 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 tower. All of the 325° C + distillate was recycled as pasting oil. The main product was middle distillate 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 process.

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 severer 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 the way to solve the problem generally.

To achieve an effective reduction of the pressure the asphalt can not be recycled to the reactor. Therefore a different application of the residua had to be found: the gasification for the production of hydrogen. The separation of the asphalt from the hot separator bottom product should therefore 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 maximum 300 bar.

For that purpose Saarberg constructed a continuous bench scale unit which was put on stream in 1975 and allowed the application of the above mentioned modification of the process. In contrast to the old IG Farben process the residue preparation was operated as shown in figure 2. 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 IG Farben process the net oil yield had a final boiling point of 325° C. We have increased the final boiling point to 400° C. Thus reducing cracking in the sump phase which is only little selective in comparison to high effective fixed bed catalysts. The reaction temperature can now be about 10° C lower which leads to the suppression of undesirable by-products like light hydrocarbons having a high hydrogen content. With these changes of the IG Farben process we were very successful in the liquefaction of our hard coal at a pressure of 285 bar and a consumption of hydrogen of 5.5 % of the moisture and ash-free coal. The oil yield amounted to more than 50 %. A typical product distribution of the modified IG process is presented in the following table.

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

	wt % of maf coal
H ₂ S, NH ₃ , CO, CO ₂ , H ₂ O	11,7
C ₁ -C ₄	15,0
C ₅ - 200° C	14,6
200 - 325° C	30,2
325° C +	9,4
vacuum bottoms	19,0
unconverted coal	5,6
	<hr/>
	105,5

Thereby it could be demonstrated that the IG Farben process leads to satisfactory results even at pressures below 300 bar using cheap disposable uncorrosive iron catalysts. The conversion of the coal is satisfactory and the hydrogen consumption is considerably lower. The vacuum bottoms of the flash drum consisting of about 50 % solids and 50 % bitumen with a melting point of about 80°C 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 t coal maf/m³h to 0,65.

Beyond that we are looking for ways in which this process could be further improved. A matter of particular interest is the preheating of the coal slurry as it is shown in figure 3. The coal slurry preparation is conventional except for a high coal content in the slurry which may now amount to as much as 60 %. This thick paste is injected into the high pressure system and heated up in a unique preheating system which is described below.

Referring to the picture, 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 heat exchanger. By adding part of this diluent oil the content of coal slurried in the paste is reduced to about 50 %. 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. 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. 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 a quite new idea which eliminates the heat exchangers needed for preheating the slurry. In this method the coal slurry is directly mixed with the hot separator overhead vapors. 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, the slurry is preheated and diluted by the condensed oil. Also the use of coarser coal could be possible now. Since the coal slurry is being transported only in tubes there is little danger of sedimentation.

Coal practical diameters up to 1 mm 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 of CO_2 , H_2O , CH_4 etc. According to the state of the art 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 the heat exchangers. This new method is pictured in figure 4. We believe that this arrangement can help to make coal liquefaction simpler and more economic.

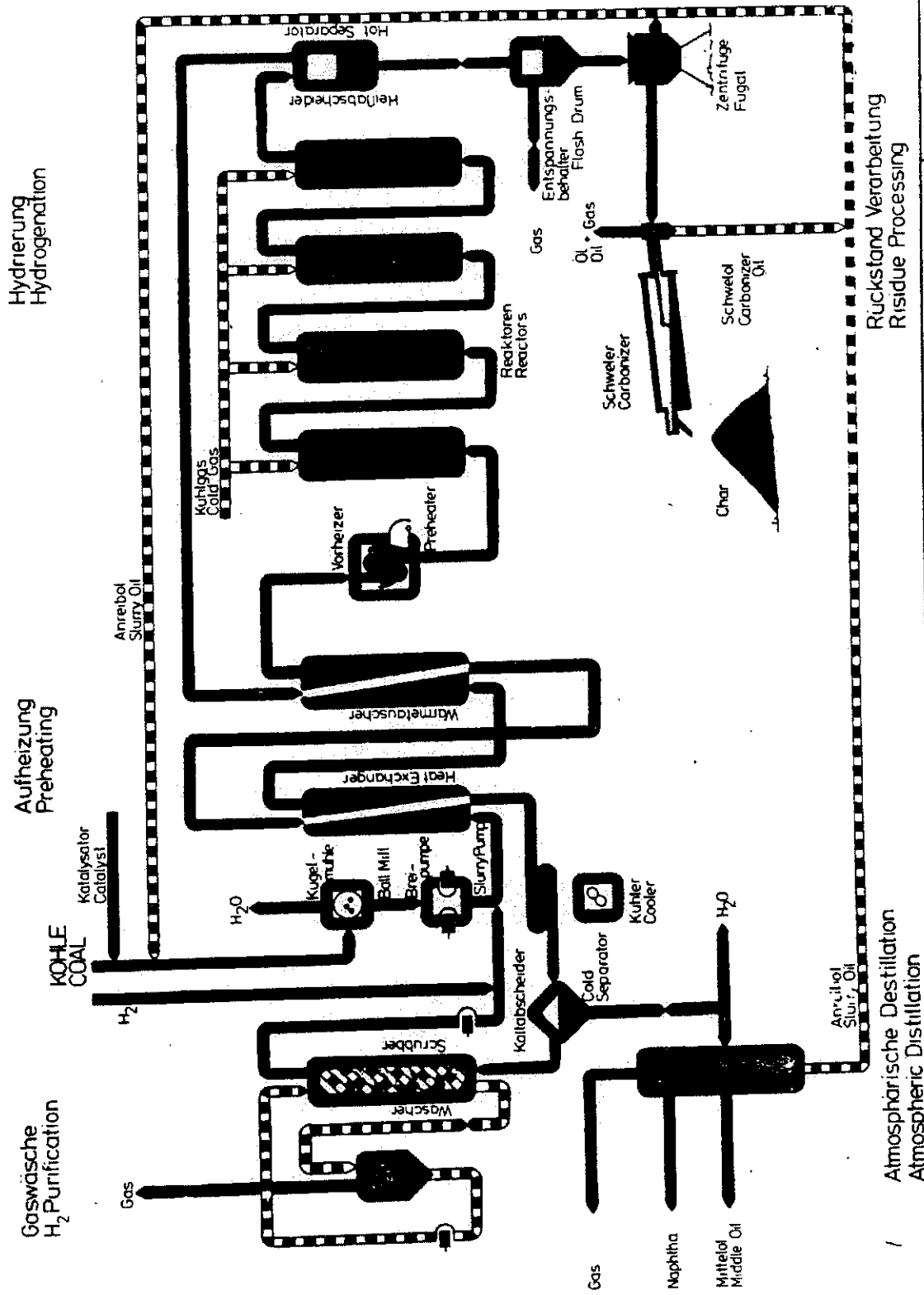
Simultaneously with the performance of the experimental program in the bench scale we have followed the upgrading of the sump phase oil to marketable products through a cooperation with the BASF. Our main objective is the production of gasoline particularly of high octane blending components. For this 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 to 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.

Encouraged by the good results we had obtained with the bench scale unit, in fall 1978 we began to construct a pilot plant with a coal throughput of 6 t/d. In the pilot plant not only the bench scale results will be checked

but also the described new ways of preheating the slurry will be among others intensively studied.

A photo is shown in figure 5. (Foreground: coal and slurry preparation section)

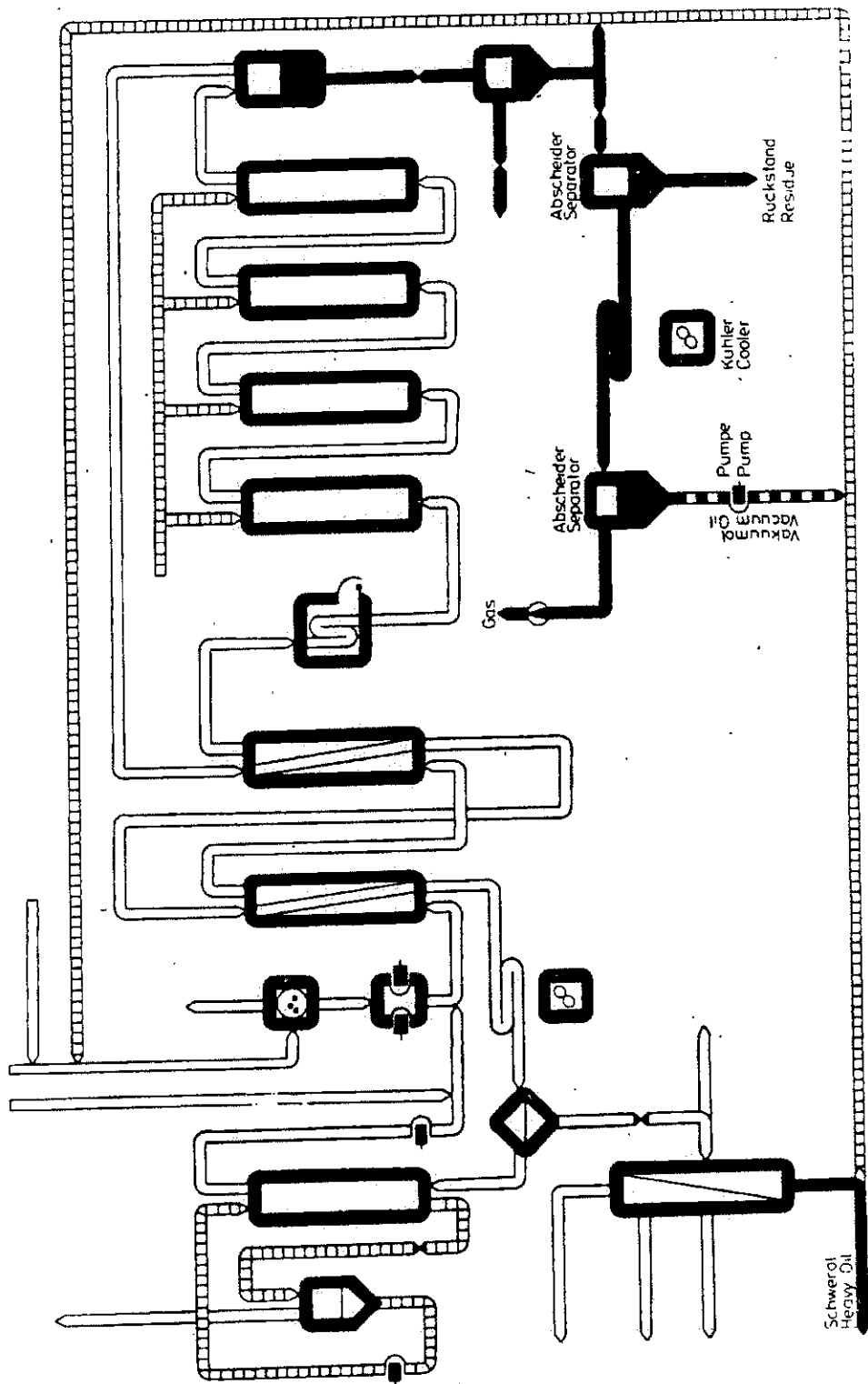
In the spring of this year Saarberg began with the performance of a study design for the erection of a commercial coal liquefaction plant in the Saarland. It is planned to convert annually 2 Mio t of coal into about 800.000 t of gasoline and chemical raw materials. The most important units of the coal liquefaction complex are the sump phase hydroliquefaction, the refining, hydrocracking and reforming of the sump phase oil and the gasification of the vacuum bottoms of the sump phase and of additional coal for the production of hydrogen. The separation of hydrogen from the coke oven gas of our coke oven plant is also being considered. The required coal quantity will be additionally produced by our mines. Scheduled date of completion could be 1986. The design study will be finished by the end of 1981. During this time many questions have to be studied in detail referring to technical problems, the site of the new plant and the economics of the process. Results and experiences from the operation of the pilot plant will also be integrated. For this study BASF has been selected as general contractor. It is the objective of this design study to provide Saarberg with a well founded basis for the decision on such a big investment.



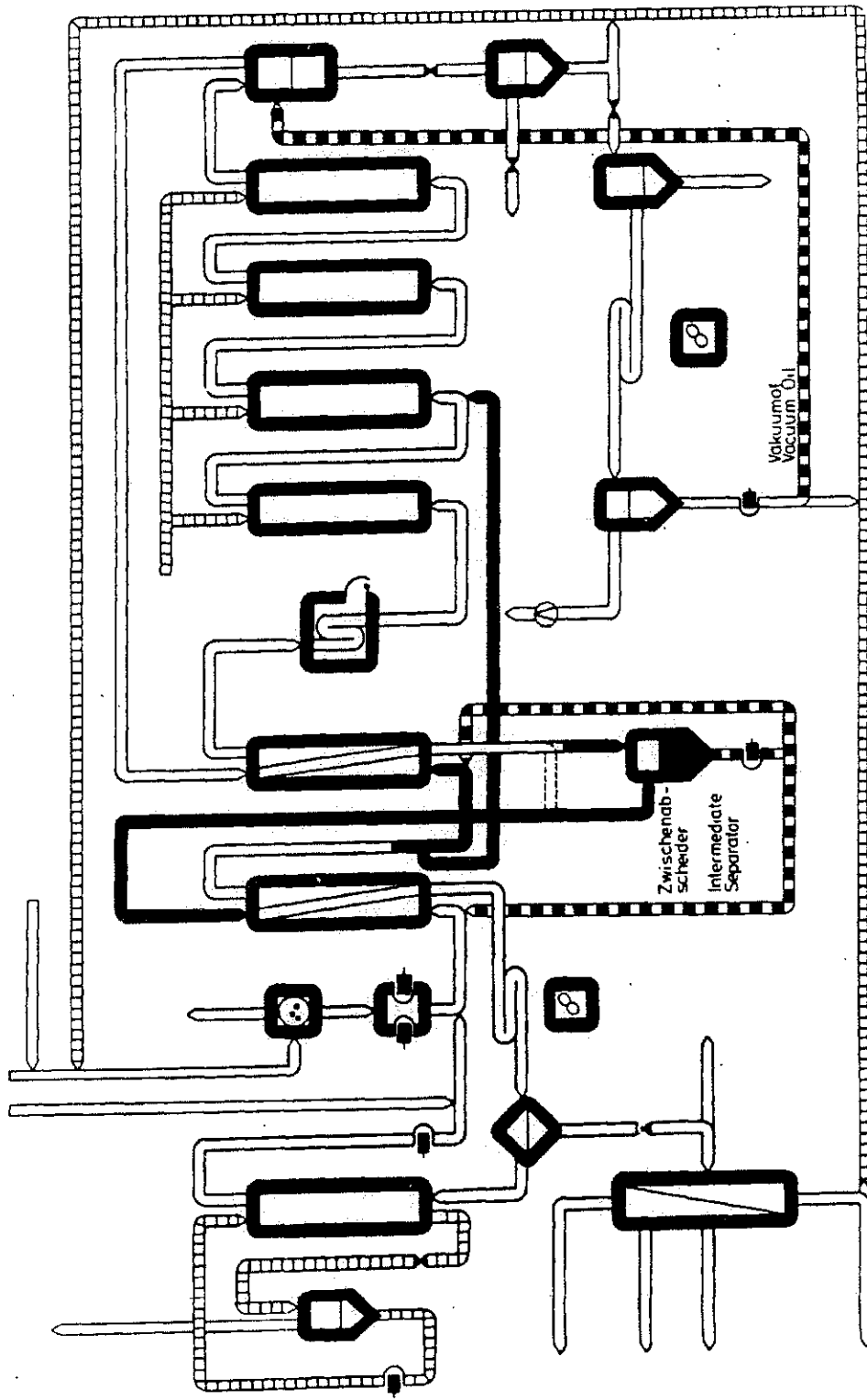
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Figure 1

Saarberg Kohlehydrierung
Coal Hydrogenation



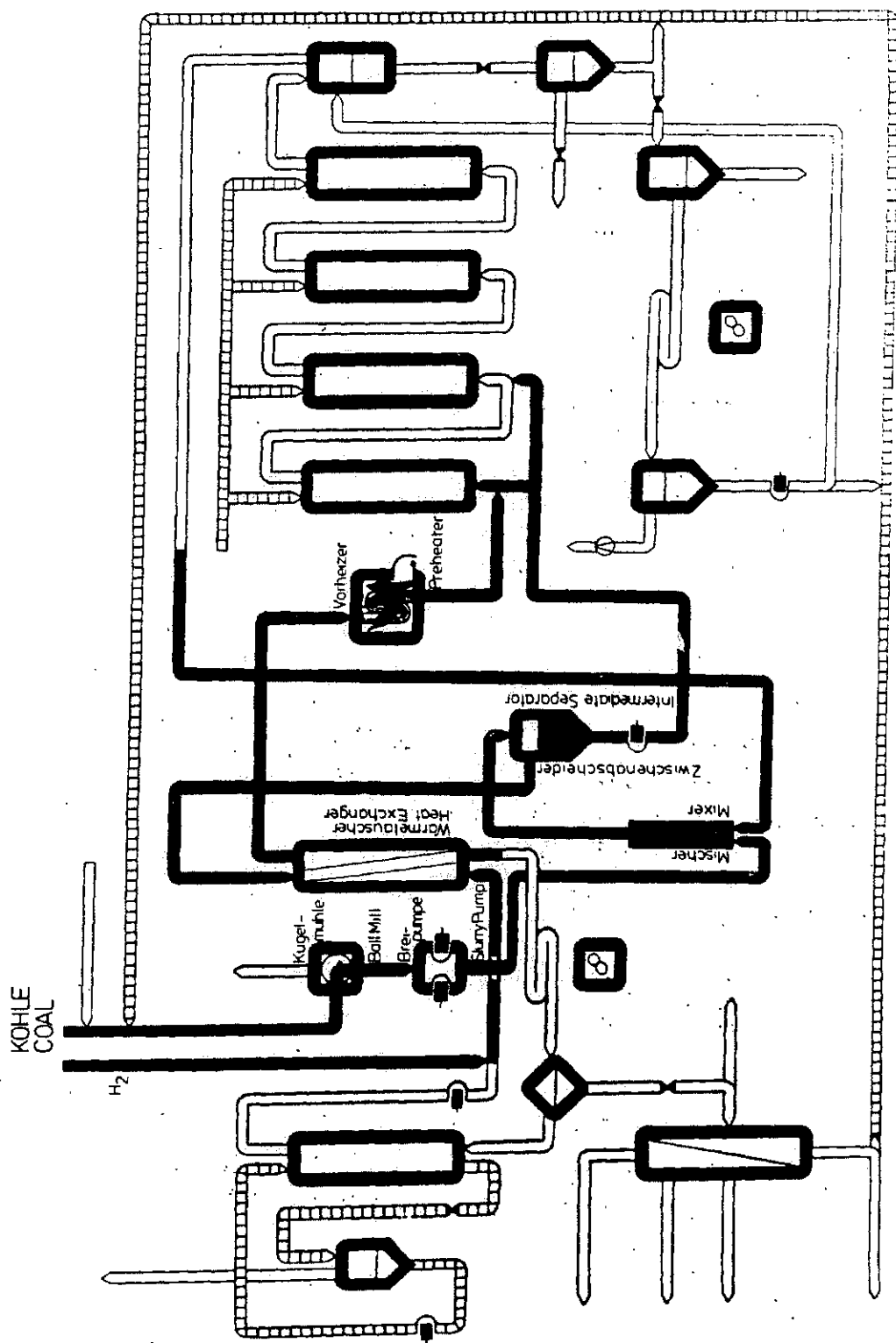
Saarberg Kohlehydrierung
Coal Hydrogenation



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Coal Hydrogenation

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Figure 3



Saarberg Kohlehydrierung
Coal Hydrogenation

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Figure 4

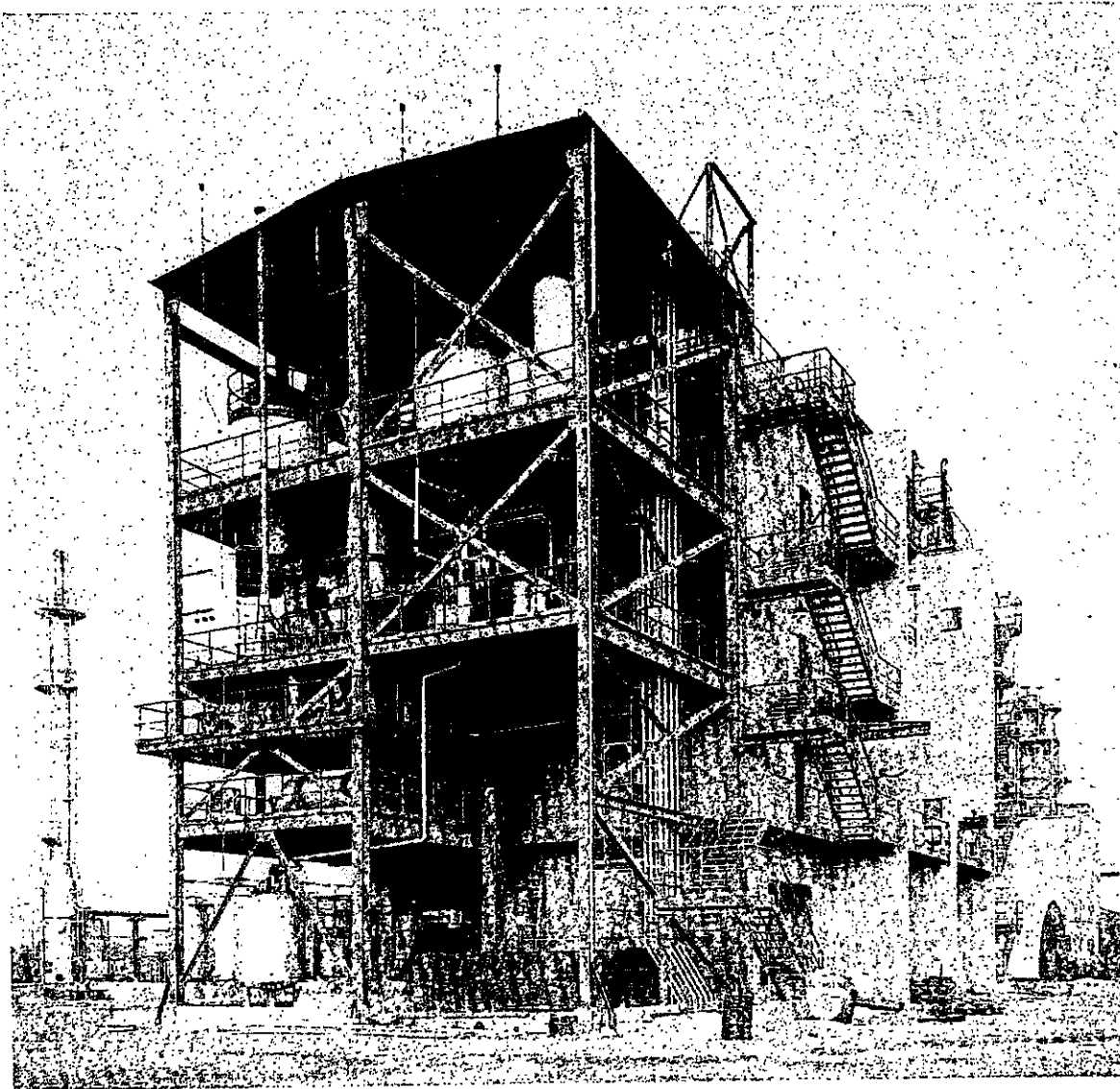


Figure 5 - Coal liquefaction pilot plant