

Section 13  
RHEINBRAUN LIQUEFACTION OF BROWN COAL

by

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In the Federal Republic of Germany, a country which is approx. 95 per cent dependent on crude oil imports, especially the use of coal, the only raw material available in larger quantities will be a feasible solution on a long-term basis in order to serve as a substitute for crude oil.

Resources	t	55	Billion
Reserves	t	35	Billion
Output	t/a	120	Million

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**Price of Brown Coal**

Raw Coal	DM/t	20.00
Raw Coal	DM/GJ	2.40

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**Brown Coal Conversion in Liquefaction > 99%**

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**Brown Coal in the Rhenish Area**

BASE 1980

Figure 1

In using coal for making products similar in their composition to those on the basis of crude oil, brown coal, especially that occurring in the Rhenish area, will be a factor of importance, since

- contiguous brown coal deposits of sufficient size are known
- brown coal can be mined at reasonable cost thanks to the advanced mining techniques and the favourable geological conditions of the deposits
- brown coal is sufficiently reactive due to its chemical composition and can be converted by means of hydroliquefaction in a relatively easy way.

The first figure shows some details for this.

Of the well known processes for liquefying coal in order substitute products on the basis of crude oil, such as

- Fischer-Tropsch synthesis after previous gasification of coal into synthesis gas
- Mobil process starting with methanol which can be generated from coal via gasification of coal to synthesis gas and conversion of synthesis gas into methanol
- direct liquefaction of coal by adding hydrogen,

Rheinische Braunkohlenwerke AG (Rheinbraun) and its subsidiary Union Rheinische Braunkohlen Kraftstoff AG (UK) are testing direct coal liquefaction while the Mobil process is developed by UK - in cooperation with Mobil Research and Development Corporation, USA, and Uhdé Company Dortmund, FRG, - to its commercial maturity.

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

\* Raw coal with 60 wt% H<sub>2</sub>O , \*\*Coal for power and H<sub>2</sub> excluded

<b>Brown Coal Liquefaction Development Stages</b>	
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Figure 2

For the direct liquefaction of coal, Rheinbraun in cooperation with UK has operated a test plant working on a continuous basis for hydroliquefaction of brown coal "HVB" (in German: "Hydrierende Verflüssigung von Braunkohle = HVB") since 1978.

In the test plant with a coal throughput of approx. 0.25 metric tons/day the basic data for the construction of a HVB pilot plant are obtained which is planned to start operation in 1985. The pilot plant will have a coal throughput of 15 metric tons/hour. The pilot plant constitutes the final development step prior to the construction of a commercial HVB plant in the Rhenish area. The first line of this commercial HVB plant (demonstration plant) having a feed coal throughput of approx. 250 metric tons/hour of raw coal is expected to start operation in the early 90's. In this HVB demonstration plant 0,4 Mio t/a of motorfuels and/or chemical feedstocks will be produced.

In the second figure, these data are compiled.

The development on hydroliquefaction of brown coal is based on the work of Bergius and Pier in the 20's and on the know-how gained in the operation of the HVB plant on a full-industrial scale at UK-Wesseling - in 1943 this plant had an output of approx. 250,000 metric tons of motor fuel produced on the basis of Rhenish brown coal.

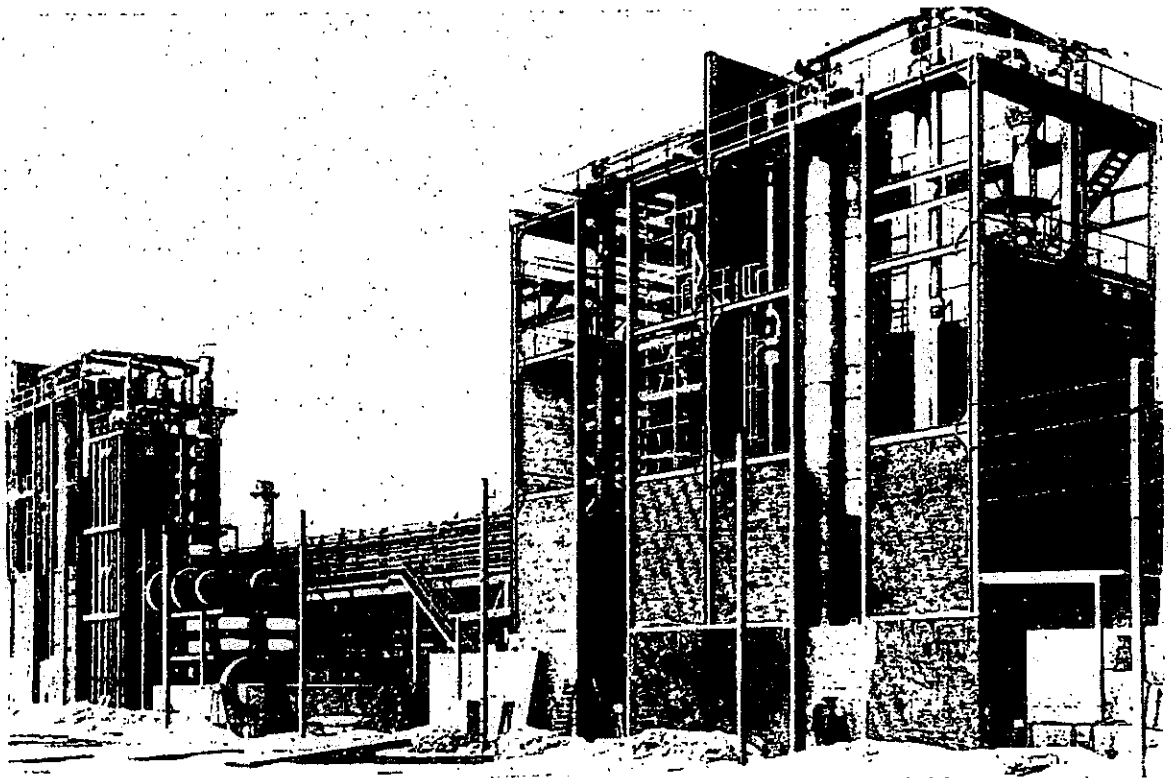


Figure 3

The third figure gives a partial view of the sump-phase high pressure section of the Wesseling plant in 1943.

In the vicinity of this "historic" place - these old reactors are still used today for high pressure reactions i. e. for methanol production - the HVB test plant was erected.

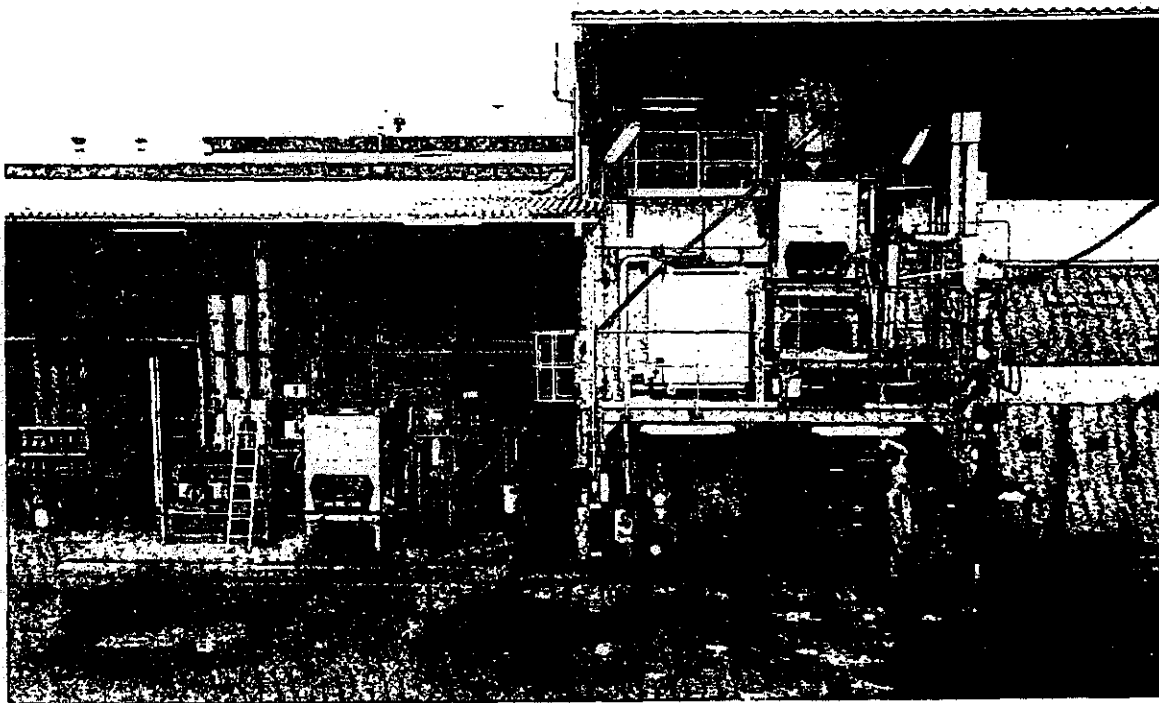
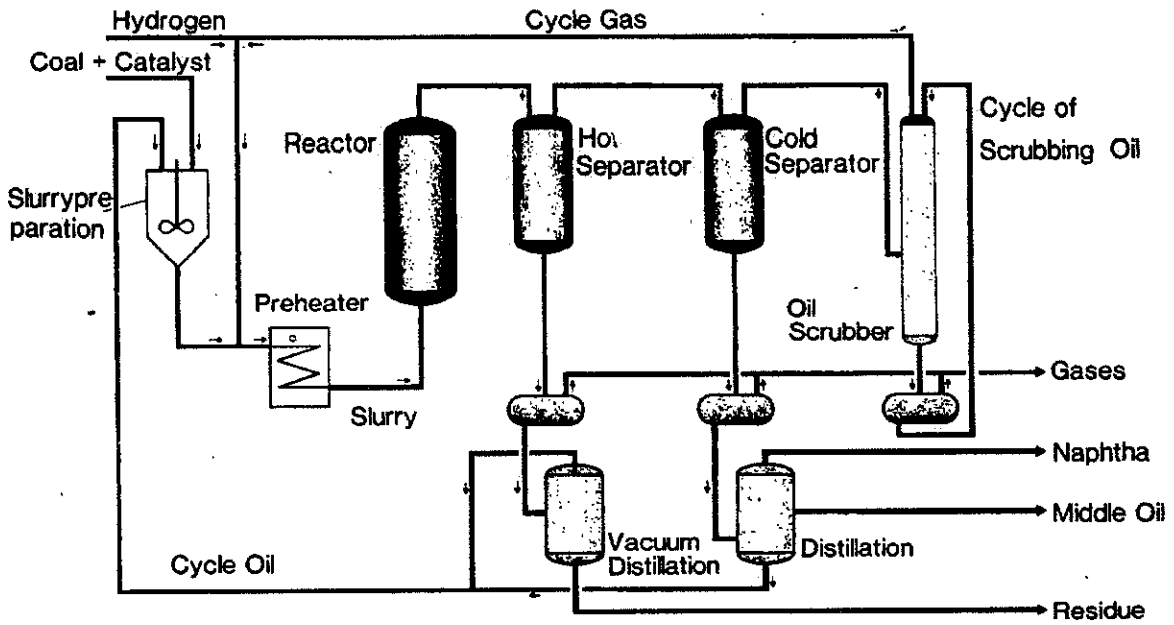


Figure 4

The fourth figure shows the outside view of the HVB test plant with continuous product distillation as well as coal preparation sections.

In the HVB process, liquefaction of brown coal is carried out in two stages. In the first stage, viz. the sump-phase hydrogenation, dry brown coal is converted into coal oil. In the second stage, viz. the gasphase hydrogenation - comprising the steps of syndrude refining, hydrocracking, naphtha reforming - the syncrude on the basis of brown coal is converted into feed materials for the chemical industry and/or into salable motor fuels.

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



Flow Sheet of Slurry Phase Hydrogenation

Figure 5



By means of a flow sheet showing the sump-phase hydrogenation of brown coal, the process is shown in detail in figure 5.

In the sump-phase hydrogenation, coal with a grain size of  $< 1$  mm and dried to a water content of 10 to 12 per cent by weight is used. Together with sulphur and iron salts as catalysts, the coal is slurried with a cycle oil produced in the process. The slurry is heated to a temperature of  $350^{\circ}\text{C}$ . In this process, water is evaporated from the slurry and humic acids are destroyed to such an extent that calcium carbonate does virtually no longer stick in the hydrogenation reactor - a problem specific to the "old" brown coal hydrogenation.

Following this thermal pretreatment, the slurry is pumped into the high-pressure section. Before the slurry enters the preheater, hydrogen consisting of fresh gas and cycle gas is added. The mixture enters the hydrogenation reactor at temperatures of approx.  $420^{\circ}\text{C}$ . Under a reaction pressure of some 300 bars, the major part of conversion from coal into coal oil takes place.

The average retention time of coal in the reactor amounts to some 30 minutes. The products of hydrogenation are separated in a so called hot separator at temperatures of about  $400^{\circ}\text{C}$  into a bottom product containing solids and into a top product free of solids. The top product of the hot separator is cooled down to some  $50^{\circ}\text{C}$ . This cooling process takes place by external water cooling in the presently operated test plant. In future in the follow up plants this energy is largely used for heating up the coal oil slurry. In the so called cold separator the condensed

oils are separated from the gases. The gases, viz. the top product of the cold separator, are subjected to an oil scrubbing unit in which in particular the hydrocarbons are scrubbed out of the gases. The scrubbed gas is recycled as cycle gas into the hydrogenation process.

After its depressurization, the bottom product from the hot separator which contains heavy oils, catalysts and coal ashes is distilled under vacuum.

The vacuum distillate is used as a component of the cycle oil. In future follow-up plants, the residue from vacuum distillation will be utilized for generating fuel gas or hydrogen to be used in the process. At present, appropriate research work in this field is performed.

After its depressurization, the bottom product from the cold separator is subjected to a multistage distillation.

The distillate fractions, viz. naphtha and middle oil, are the oil yield of the HVB process. The distillation residue is used as a further component of the cycle oil. The gases occurring at the different depressurization stages will be processed in the subsequent follow-up plants and will be used for the generation of liquid gas and/or for the generation of H<sub>2</sub> or as fuel gas.

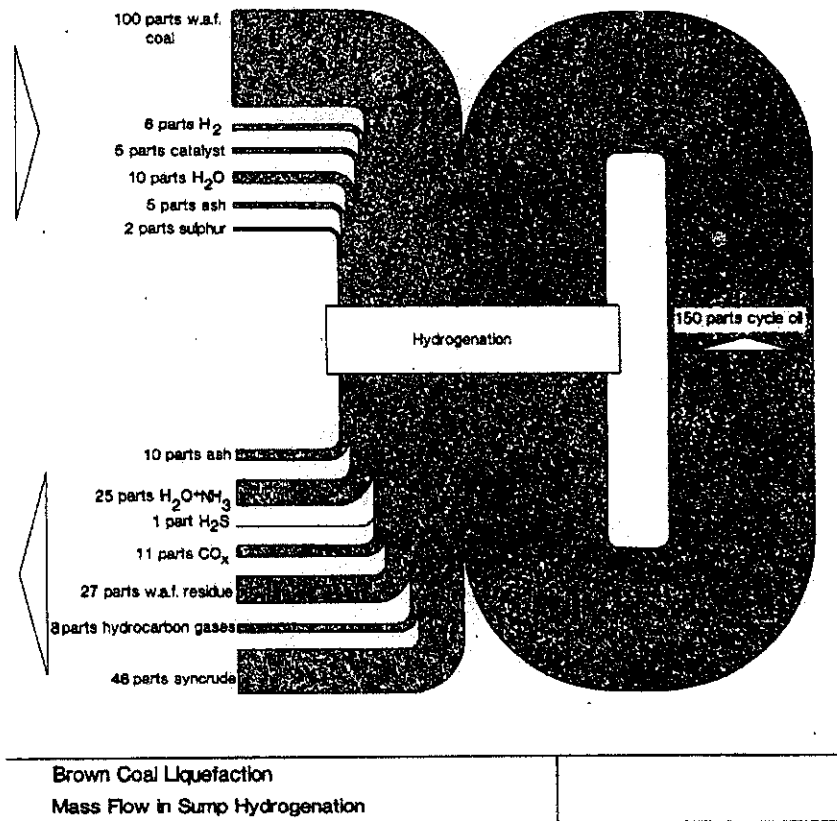


Figure 6

Figure 6 shows the net mass balance of several test runs of the sump-phase hydrogenation of brown coal. The data given are based on the use of 100 parts by weight of w.a.f. coal. Nearly 50 per cent of the organic coal material used in coal hydrogenation is converted into a syncrude from coal. This syncrude from coal is the oil yield of sump-phase hydrogenation. The syncrude from coal has a boiling range between 35 °C and 350 °C.

In order to liquefy brown coal about 6 parts by weight of hydrogen are used in the sump-phase.

The syncrude from coal still contains approx. 4 per cent by weight of oxygen and approx. 0.2 per cent by weight of nitrogen and sulphur each.

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

Brown Coal Liquefaction Operating Conditions and Results of Sump Phase Hydrogenation	
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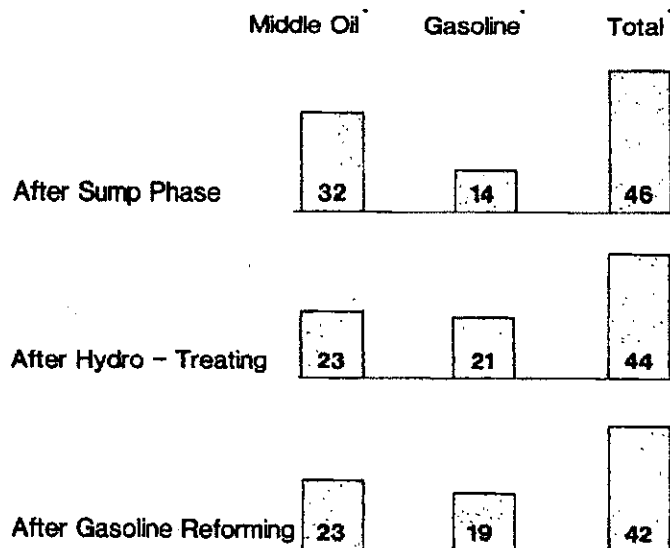
Figure 7

Figure 7 shows the improvements in the sump-phase hydrogenations of the HVB process compared with that applied on a full-industrial scale at Wesseling.

It was possible to reduce the operating pressure from 700 to 300 bars and to increase the space-time yield by some 50 per cent from less than 0.2 kg/m<sup>3</sup> reaction space and hour to nearly 0.3 kg/l.h.

The improvements made on the sump-phase hydrogenation of the HVB process compared with the sump-phase applied on a full-industrial scale at Wesseling are due to the fact that the separations of solids from coal oil made by means of centrifuges in former times are currently carried out by means of distillation processes. The solids contained in the coal oil of the sump-phase consist of non-converted coal, ash, and catalyst. By means of vacuum distillation they are removed from the sump-phase product. In contrast to former processes, this allows to produce a recycling oil for coal slurring which does not contain any asphaltenes or any portions of non-converted coal.

The solids and asphaltenes separated by distillation are gasified for generation hydrogen. In this point, too, the basic scheme Rheinbraun pursues in the developments differs from the techniques applied by UK Wesseling in former times. There, the solids were carbonized at low temperature and the carbonization residue, a coke-like substance, was disposed of. This method of treating residues using centrifuging and carbonization was technically sensitive and caused environmental problems. In contrast to this, residue gasification is an energetically efficient process which can be carried out in an environmentally acceptable way. Due to this change in process, it can be expected that it is possible to rise considerably the process efficiency compared with that in former times; this will involve that 15 metric tons of raw coal are no longer required to produce one metric ton of liquid product, but only 10 metric tons of raw coal will suffice which corresponds to a total thermal efficiency of approx. 50 per cent.



Yield of C<sub>5</sub> + Hydrocarbons in Percent by Weight  
related to Feed Coal ( w.a.f. ) in  
the Sump - Phase Hydrogenation

<b>Brown Coal Liquefaction Distribution of Products</b>	
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Figure 8

The objective of the HVB process is the production of chemical raw materials and/or motor fuels. Due to their high content of hetero-atoms the products from the sump-phase hydrogenation of brown coal have to be post-treated by hydrogenation before being used for other purposes. Figure 8 shows the ratios for the motor fuel production. The results shown are an average of several test runs. Syncrude from coal of the sump-phase hydrogenation contains approx. 70 per cent of middle oil and approx. 30 per cent of naphtha. By means of a catalytic middle oil refinement, the phenolic fractions in the middle oil are converted into hydrocarbons the major part of which will boil in the naphtha-range. Thus, the ratio of naphtha to middle oil shifts from approx. 2 : 1 to approx 1 : 1 after the refinement of the middle oil and the naphtha fractions from the sump-phase hydrogenation.

	Middle Oil	Gasoline
<b>After Hydro - Treating</b>		
- Cetane Number	50	-
- Research Octane Number	-	66
<b>After Gasoline Reforming</b>		
- Research Octane Number ( unleaded )	-	97
- Research Octane Number ( leaded )	-	99
<b>Brown Coal Liquefaction Product Quality</b>		

Figure 9

While the middle oil from the refinement stage meets the requirements for diesel fuel with a cetane number of 50 without any further treatment being required it is necessary for naphtha to be further processed by catalytic reforming so that it will be in keeping with this specification.

Figure 9 shows the qualities of diesel fuel and/or gasoline after their different post-treatments in which they are characterized by their cetane numbers and/or research octane numbers.

After catalytic reforming of heavy naphtha (80 - 180 °C) the combined total gasoline fraction has a research octane number of 97 and is considered to be premium gasoline even without leading.

Thus, liquefaction of brown coal according to the HVB process allows to produce motor fuel in keeping with the present market requirements.

Due to the properties of the HVB process products, viz. high contents of paraffins and naphthenes in the refined gasoline and middle oil, high content of aromatic substances in the reformed heavy naphtha, it is possible to produce the essential basic chemicals of organic chemistry, viz. olefines and aromatic BTX substances, on the basis of brown coal.

Tests on steam cracking of refined brown coal naphtha and middle oil have shown that compared to corresponding fractions from petroleum there is no difference to be expected in the yield of olefines.

Thus, the products from hydroliquefaction of brown coal can be used as a substitute for petroleum in both the motor fuel sector and petrochemical sector without any adaptation of the present demand structures in both sectors becoming necessary.



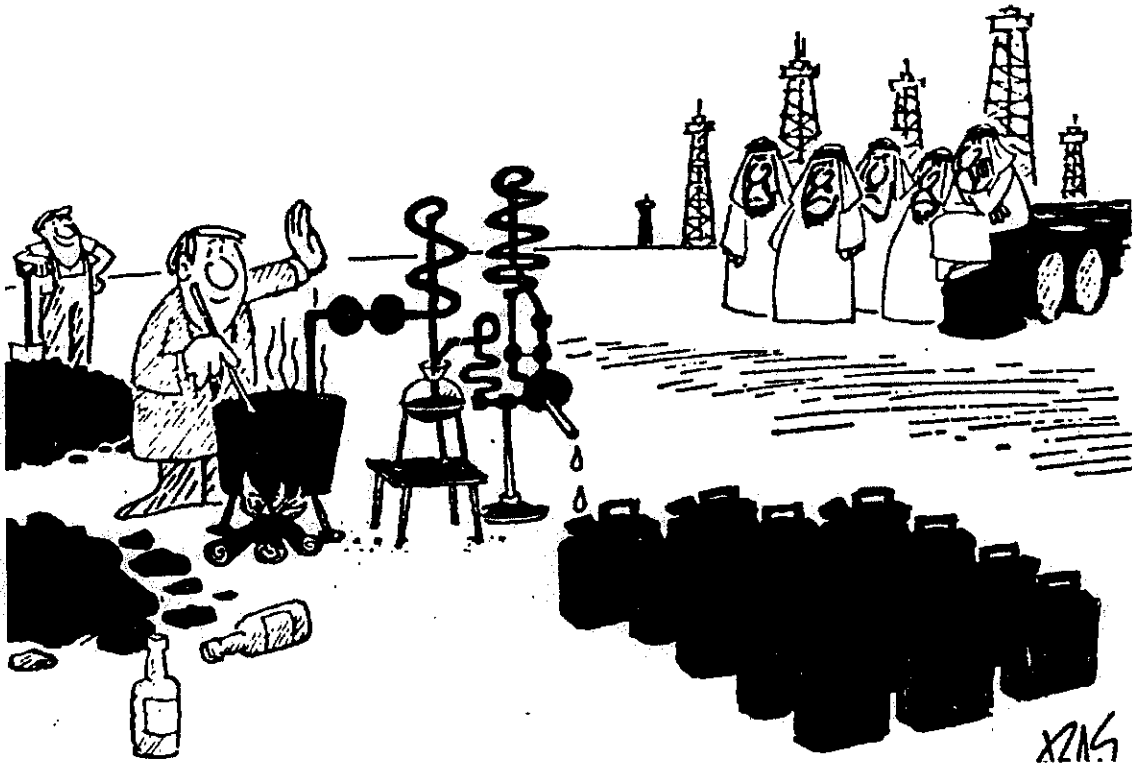


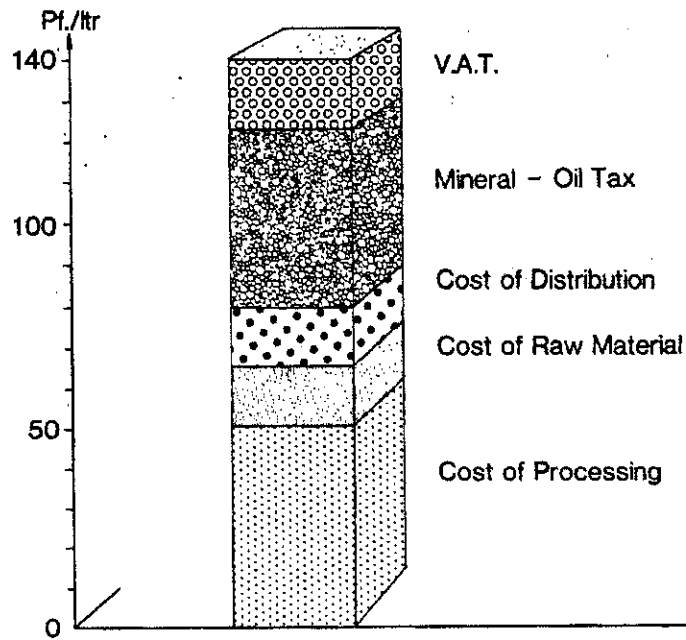
Figure 10

Does HVB make it possible to reach the dreamed-of goal pursued by the coal chemists which is shown in figure 10?

From the chemical point of view this is true. From the technical point of view, still some questions have to be settled which concern critical development stages.

Besides coal hydrogenation itself and vacuum distillation of the products from sump-phase hydrogenation, gasification of the vacuum residue from vacuum distillation, too, is part of the critical development stages for HVB in view of its use on a full-industrial scale.

What are important factors to HVB in addition to the chemical and technical ones, are the product costs and the availability of coal in FRG.



**Brown Coal Liquefaction  
Price at Gasoline Stations**

FASS 1980

Figure 11

According to the present state of knowledge the costs for the production of e.g. gasoline on the basis of Rhenish brown coal is at present still some 50 per cent higher than the cost for gasoline on the basis of crude oil ex refinery. Figure 11 shows the cost structure of gasoline on the basis of brown coal. After 1990 when the project of the commercial HVB plant is expected to be realized, it might be possible to produce brown coal gasoline in an economical way. The extent to which brown coal liquefaction can take a considerable share in reducing the Federal Republic's dependency on crude oil depends not only on the technological successes in developing coal liquefaction and on the developments in petroleum prices but in particular on the availability of coal.

From the present point of view, german brown coal and german bituminous coal are only available in approximately sufficient quantities for coal liquefaction in the Federal Republic of Germany, if in future nuclear energy substitutes part of the coal fired in today's power plants.