OCCASION

This publication has been made available to the public on the occasion of the 50th anniversary of the United Nations Industrial Development Organisation.

DISCLAIMER

FAIR USE POLICY

CONTACT

Please contact publications@unido.org for further information concerning UNIDO publications.

For more information about UNIDO, please visit us at www.unido.org

UNITED NATIONS INDUSTRIAL DEVELOPMENT ORGANIZATION
Vienna International Centre, P.O. Box 300, 1400 Vienna, Austria
Tel: (+43-1) 26026-0 · www.unido.org · unido@unido.org

17530

RESTRICTED

October 1988

TECHNIQUES OF DIRECT COAL LIQUEFACTION PHASE II

DP/CPR/83/002

People's Republic of China

Expert Report *

Prepared for the Government of China
by the United Nations Industrial Development Organization
acting as executing agency for the United Nations Development Programme

Based on the Work of Dr. Helmut Würfel,
Consultant on Pyrosol Coal Liquefaction Technology
Blieskaste, Federal Republic of Germany

Backstopping Officer: R.O. Williams, Chemical Industries Branch

United Nations Industrial Development Organization
Vienna

* This document has been reproduced without formal editing

Since 1974 Saarbergwerke AG has been dealing with the further development of direct coal liquefaction according to the principle of IG-Farben and so does its 100 % subsidiary GfK Gesellschaft für Kohleverflüssigung mbH since 1980. In autumn 1981 GfK put into operation a pilot plant with a capacity of 6 t per day. In the beginning, the works were orientated to the further development and testing of the one-stage direct hydrogenation process. An important distinguishing feature against the IG-Farben process applied on industrial scale until 1945, was the treatment of residues. The heavy oils which had to be recirculated for pasting the coal, were no longer gained as in the past by centrifuging, but asphalt-free by a vacuum flash distillation.

In 1982, a planning study was carried out for the techno-economical evaluation of industrial coal hydrogenation, the result of which was that even if no costs would occur for the feed coal, coal hydrogenation would not be economically feasible. In 1984, a very similar result was obtained in a study performed on behalf of the BMFT (Federal Ministry of Research and Technology), where on the basis of list prices of German hard coal one ton of hard coal feed had to be subsidized by 300,-- DM.

Although it would be possible today - and not only in the Federal Republic of Germany - to build an industrial demonstration plant, for economic reasons such a decision was not made up to now. Therefore GfK performed an extensive analysis of direct coal liquefaction defining the following requirements in order to obtain an optimum process.

./2

- Selective H_2 utilization for oil production
  (suppression of hydrocarbon gas formation)
- No slurry heat exchangers and preheaters
- Auto-thermal operation
- High concentration of coal in the Slurry
- Feeding of coarser coal
- High specific coal throughput (CHSV)
- Low pressure
- Feeding of coals with high ash contents
- No sediment deposits, in particular when hydrogenating young coals.

Furthermore, the process should be simple as experience shows that the availability decreases with increasing complexity of a process.

In order to largely realize above-mentioned requirements, in 1987 GfK conceived a new hydrogenation process which was already successfully applied on bench scale. It mainly comprises the performance of reactions where the coal is conveyed to the hydrogen in a counterflow. While in the classical IG-Farben process coal slurry and hydrogen were fed from below in a co-current flow into the hydrogenation reactor after a joint preheating in a heater to a temperature exceeding 400 °C, in the new GfK counterflow reactor the coal slurry is fed from the top. From here it is conveyed towards the bottom against the rising hydrogen according to the counterflow principle. Figure 1 shows a diagrammatic sketch of the counterflow reactor. The advantages of the counterflow reactor are listed as follows:

- No slurry heat exchangers and preheaters due to the integrated direct heating
- Auto-thermal operation (at a H_2-consumption of about 3 % of coal maf)
- No secondary cracking of pasting oil and oil yield
- Less formation of hydrocarbon gas (low consumption of H_2)
- Rising H_2 partial pressure with increasing reaction way
- Higher specific coal throughput due to less inert gas and vapours in the reaction zone

- Feeding of coarser coal (dissolving instead of grinding)
- No sediment deposits
- No hot separator
- Simple up-scaling

It is especially important that the exothermic reaction is sufficient at a hydrogen consumption of 3.5 % of the coal feed in order to dispense with any heat exchanger and preheater at an inlet temperature of less then 200^{0}C for a 70 percent coal slurry. Furthermore it should be emphasized that the reactor builds less light hydrocarbons as the pasting oil from the coal slurry can evaporize at the reactor top thus avoiding a secondary cracking.

This type of reactor has been operated since September 1987 in the bench scale hydrogenation facility of the GfK and, up to now it proved to be very worthwhile. Aside from high-quality German hard coals also hard coals with an ash content of up to 22 % and various brown coals have been hydrogenated.

The new GfK counterflow hydrogenation process was firstly applied at a pressure of 200 bar. Its superiority with regard to a hydrogenation reactor fed from below became abvious as the oil yield was by approx. 10 % points of coal maf higher at otherwise unchanged conditions. In figure 2 this is demonstrated clearly.

Furthermore I wish to point out that the total gas amount introduced into the reactor may be considerably lower in the counterflow reactor. As the above-mentioned results with the counterflow reactor have not been optimized yet, we assume that at 200 bar an oil yield of about 50 % (boiling end point approx. 400 ^{0}C) might be possible at an optimum process performance feeding German hard coal.

Beside the development of the counterflow process for the conversion of coals and heavy crude oils, GfK has extended the process for certain applications by an additional second stage for coking the residues from hydrogenation. This results in an increasing oil yield. Before going into this new two-stage so-called PYROSOL process, I wish to describe in principle the coal behaviour during hydrogenation with regard to the products generated depending on the hydrogen consumption (see figure 3).

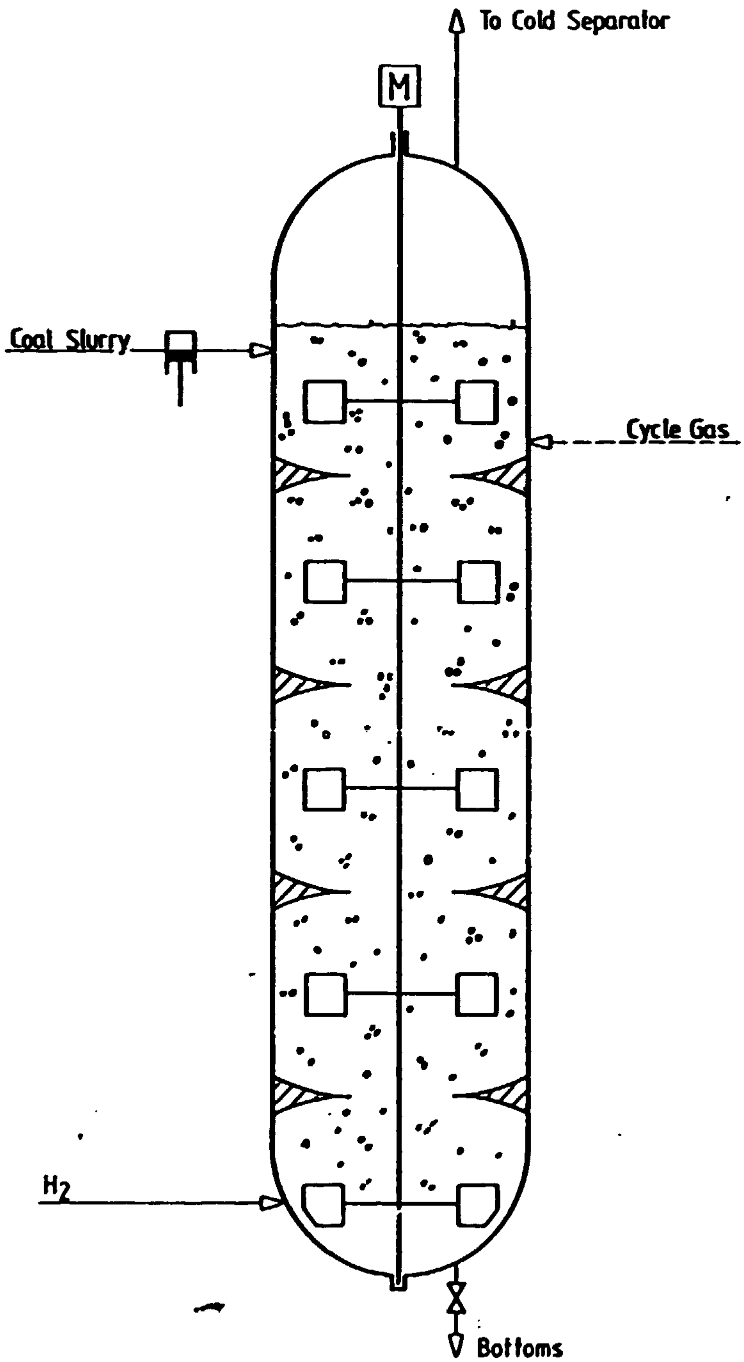
According to this the coal can be largely dissolved at a hydrogen content of about 2 %, however, the main product still is a bitumen which will only be converted into distillate oil during further reaction (hydrogen addition). Here increasingly gaseous hydrocarbons are produced. The one-stage direct liquefaction for producing a maximum oil yield can be seen in the right part of the figure. The oil yield gained here is about 50 % referred to the maf coal. In contrast, also a milder operation with reduced oil yield may be realized, which is shown in the middle of the figure. Due to the milder hydrogenation conditions of about 440 °C and 200 bar, the oil yield is limited to about 30 %, however, the amount of bitumen is correspondingly higher.

The PYROSOL process proposed by GfK is a two-stage liquefaction process where in the first stage such a mild hydrogenation is applied followed by subsequent coking stage for the production of additional oil from hydrogenation bitumen. The advantages of this process are the low gas formation in the first stage, the low pressure as well as the possibility of subjecting the whole residues by coking to a total disproportioning in oil and coke. Another advantage of this process is the possibility of converting coals with high ash contents. As is shown by the diagram for maximum allowable ash contents in the lower part of the figure, a direct dependence is given between the liquid bitumen in the reactor and the ash content of the coal, as more bitumen in the reactor may also hold more ash in suspension, thus possibly increasing the amount of ash fed to the reactor.

Including the counterflow reactor GfK is operating a continuous bench scale facility by this process which is represented in a simplified form in figure 4. The plant can be operated either recirculating into the hydrogenation reactor the entire coker oil for pasting the coal or only the heavy coker oil. When utilizing coker oils of relatively high hydrogen content as for example occuring in brown coal hydrogenation, the entire coker oil may be added to the oil yield. Figure 5 shows a simplified flow sheet of an industrial available continuous residual coker (Lurgi-Ruhrgas type).

The two-stage PYROSOL process with subsequent residue pyrolysis represents an excellent possibility of obtaining high oil yields also from coals with high ash contents which - up to now - were not suitable for hydrogenation. By applying the pyrolysis stage the otherwise negative ballast effect of the ash is eliminated. Furtheron, the two-stage PYROSOL process can also be applied with quality coals of low ash content when coals of 80 DM/t onwards can be economically hydrogenated for the production of oil. However, if oil can only be produced economically from considerably unexpensive coals, the application of just the counterflow hydrogenation process is recommended.

It is the intention of GfK to demonstrate the new process in their 6 t per day pilot plant.



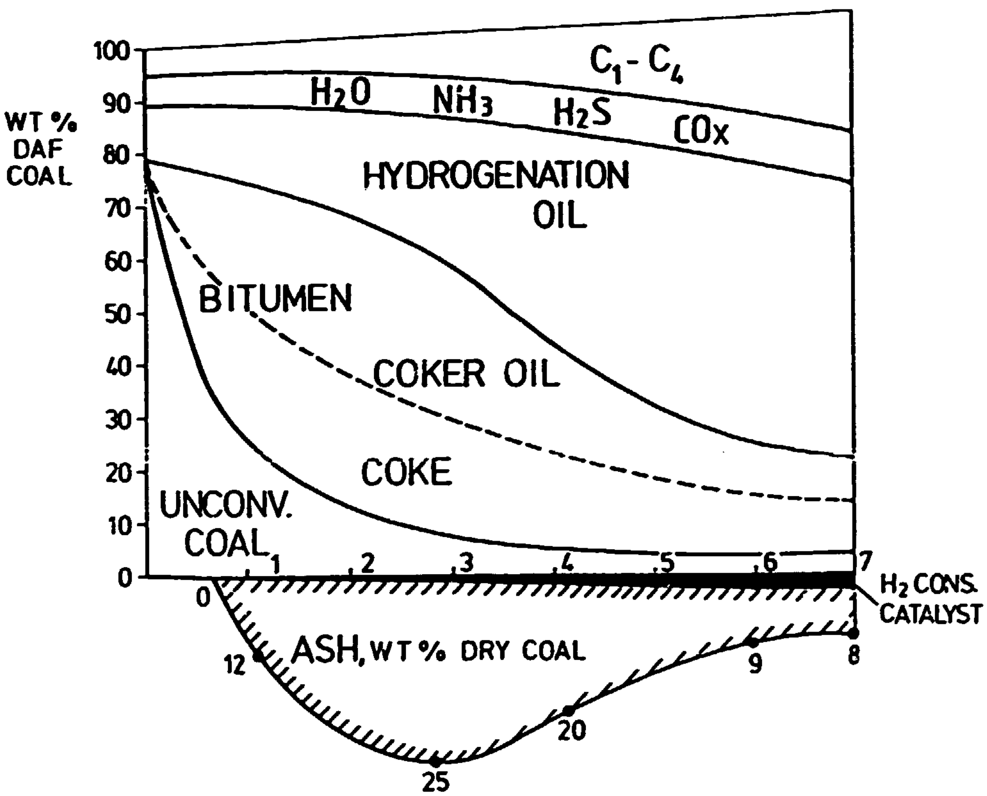
GfK	Counterflow-Reactor	1988

Figure 1

Comparison of the yields at 200 bar between counterflow reactor and co-current flow reactor

	Counterflow	Co-current flow (4)
Net Oil Yield	46	36
$C_1 - C_4$	14,0	
H_2-consumption	4,6	
Coal	Ensdorf (Saar)	(Ruhr)

Figure 2



1988	COAL LIQUEFACTION PRODUCT DISTRIBUTION	GfK

Figure 3

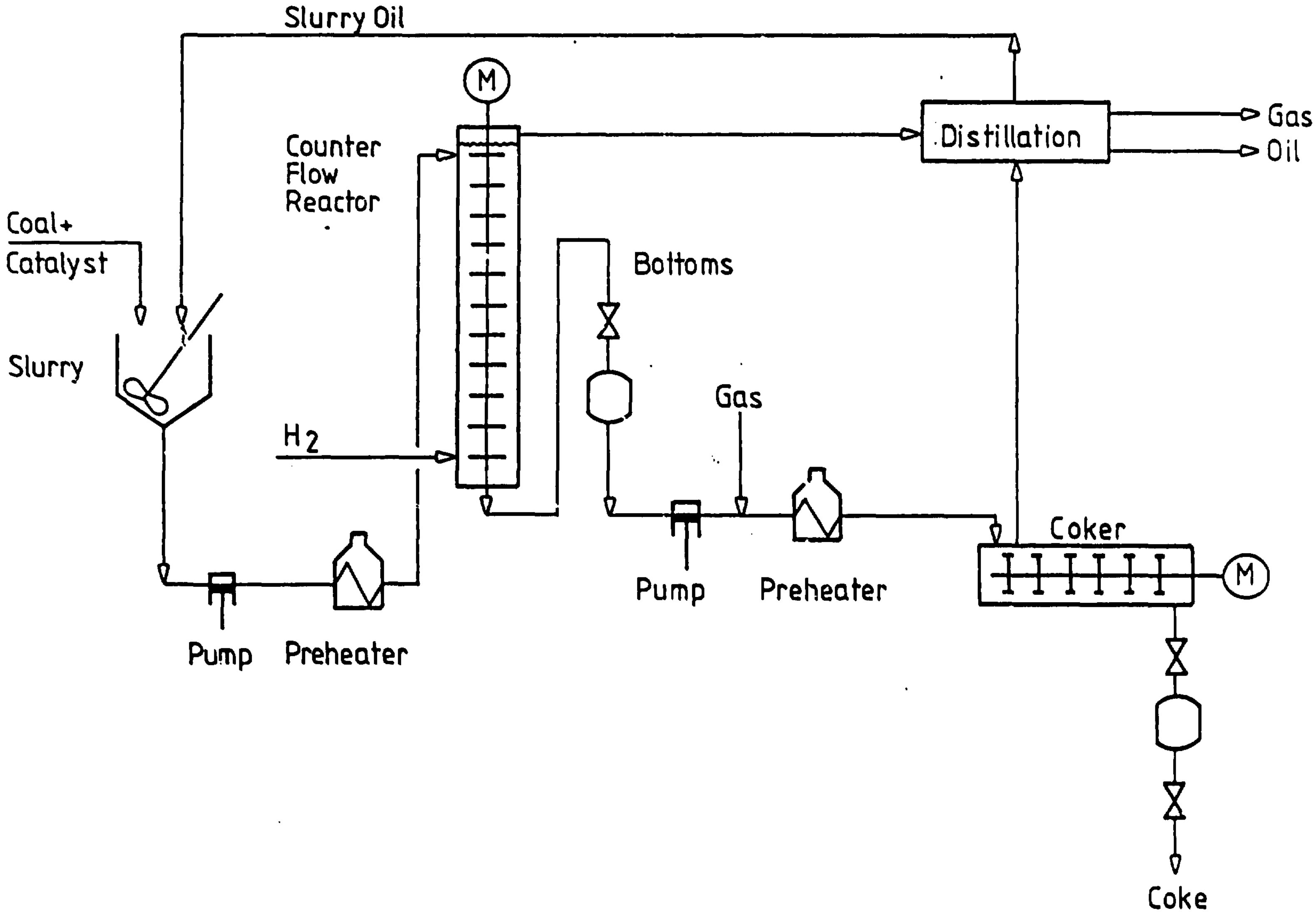


Figure 4

GfK	PYROSOL Bench-Scale Unit	1988

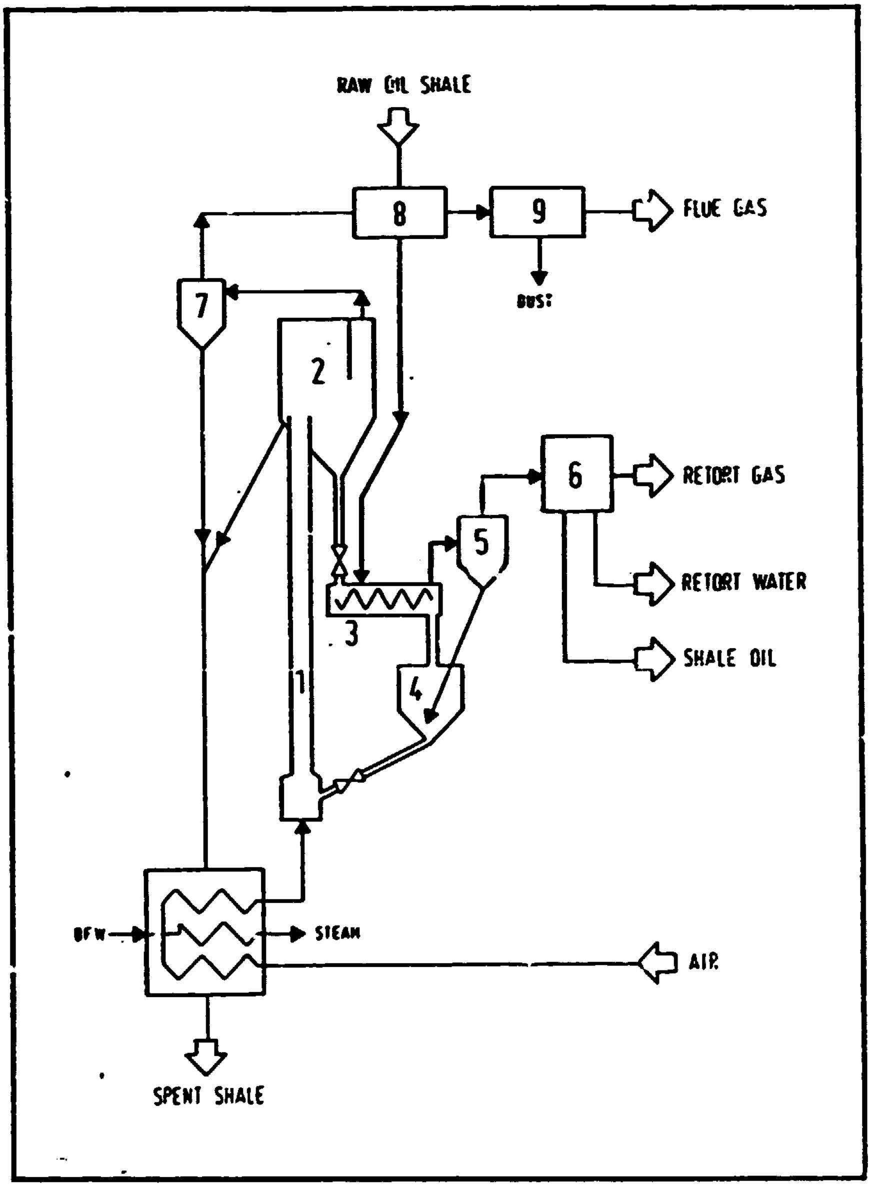


Figure 5 Simplified LR-Process Scheme

Results of coal liquefaction (PYROSOL process with counterflow reactor)

Coal	Ensdorf (Saar)	Brown coal DDR	
Ash content mf	up to 22 %	9,3 %	
Oil yield C_5^+	52)	57,2)	wt % of coal maf
C_1 - C_4	13,7)	9,6)	
H_2-consumption	4)	3,9)	
Pressure	200 bar	200 bar	
Temperature	440 °C	440 °C	

Figure 6