1. Introduction

Short History

Under the auspices of the Federal Ministry of Research and Technology (BMFT) of the Federal Republic of Germany, a presentation covering the production of liquid fuels from brown coal and bituminous coal was submitted to the Australian States of New South Wales, Queensland and Victoria in March 1976.

This presentation was prepared by a German consortium, headed by Professor Dr. Dr. K.-H. Imhausen, comprising the following companies :

Imhausen-Chemie GmbH, Lahr
Lurgi Kohle und Mineralöltechnik GmbH, Frankfurt/Main
Rheinische Braunkohlenwerke AG, Köln
Ruhrchemie AG, Oberhausen
Ruhrkohle AG, Essen
Salzgitter Industriebau GmbH, Salzgitter
Uhde GmbH, Dortmund.

From basic considerations, it was shown that the Fischer-Tropsch synthesis is not the optimum way of converting Australian coals into automotive fuels.

In May 1977, the German consortium proposed a technical alternative which combined coal hydrogenation, gasification of hydrogenation residue, and Fischer-Tropsch synthesis. This combined scheme offers higher overall product quality and better economics than Fischer-Tropsch synthesis alone.

On February 1st, 1978, BMFT awarded the German consortium an optimization study in order to establish the most suitable process scheme to convert coals into gasoline and automotive diesel oil (ADO). The optimization study was completed in December 1978. BMFT/Forschungsbericht T 80-048/August 1980.

Based on the results of this optimization study and on the subsequent selection of process schemes in February 1979, BMFT - and the Australian Department of National Development and Energy (DNDE) - awarded Imhausen-Chemie GmbH a contract for the preparation of a feasibility study covering the conversion of Australian brown coal and bituminous coal into liquid motor fuels. Work on the feasibility study started on April 1st, 1979. The final submission date was 31st August, 1981.

The feasibility study was financed in equal parts by BMFT and by DNDE.

2. Basic Data for the Study

2.1 Objectives

The feasibility study has the following main objectives :

a) to establish a detailed technical concept for the conversion of Australian bituminous coals into automotive fuels using coal hydrogenation, gasification and Fischer-Tropsch synthesis

b) to establish a detailed technical concept for the conversion of Australian brown coals into automotive fuels using coal hydrogenation and gasification of the hydrogenation residue

c) to establish the economics of commercial plants as per a) and b)

d) to provide summary interpretation of the main conclusions and recommendations of the final report of the study in a form suitable for submission to the Australian participants.

2.2 Technical Basis of Design

The fundamental requirements of the three plants concerning production capacity and product quality have been collected in Table 1. The proportions of gasoline, automotive diesel oil and LPG were specified by the Australian states.

Table 1 Basic Data for Plants Design

	New South Wales	Queensland	Victoria
Product capacity, motor fuels(1)	3 000 000 t/a	3 000 000 t/a	3 000 000 t/a
Ratio gasoline : ADO	1 : 1	1 : 1	2 : 1
Gasoline premium	50 %	60 %	70 %
Gasoline regular	50 %	40 %	30 %
Gasoline premium RON	97	97	97
Gasoline regular RON	92	92	92
Cetane index ADO	50	50	50
LPG production	max.15 % of liquid products	max. 15% of liquid products	no
Raw materials	bituminous coal	bituminous coal	brown coal

(1) in the feasibility study the term "motor fuels" always means gasoline and ADO.

3. Quality of the Coals from New South Wales, Queensland and Victoria (see Table 2).

The main differences of the coals from the three states are in the water and ash content.
The brown coal from Victoria has 60 wt. % water, whereas the bituminous coals from New South Wales and Queensland have only 4.2 wt.% or 7.6 wt.% water, respectively.
The ash content of the brown coal is only 1.66 wt.%, whereas the ash content of the bituminous coal from New South Wales is 17.7 wt.% and from Queensland 25.5 wt.%.

Due to the very high ash content of the Queensland coal, the coal feed for the hydrogenation step as well as for the gasification step is de-ashed.

For the New South Wales plant only the part of coal for the hydrogenation step is de-ashed.

The principles of these process steps are illustrated in the attached flowsheets (see Fig. 1 and 2).

These concepts are based on the results of tests performed with coal samples made available by Australien sources.

They consider the balance of low ash content coals for processes as well as high ash content coals for power generation purposes in a plant with full energy balance as well as the preferential supplies of low ash content coal for hydrogenation.

The plants for coal de-ashing are, however, not the subject of this study. These facilities are planned by the Australian side.

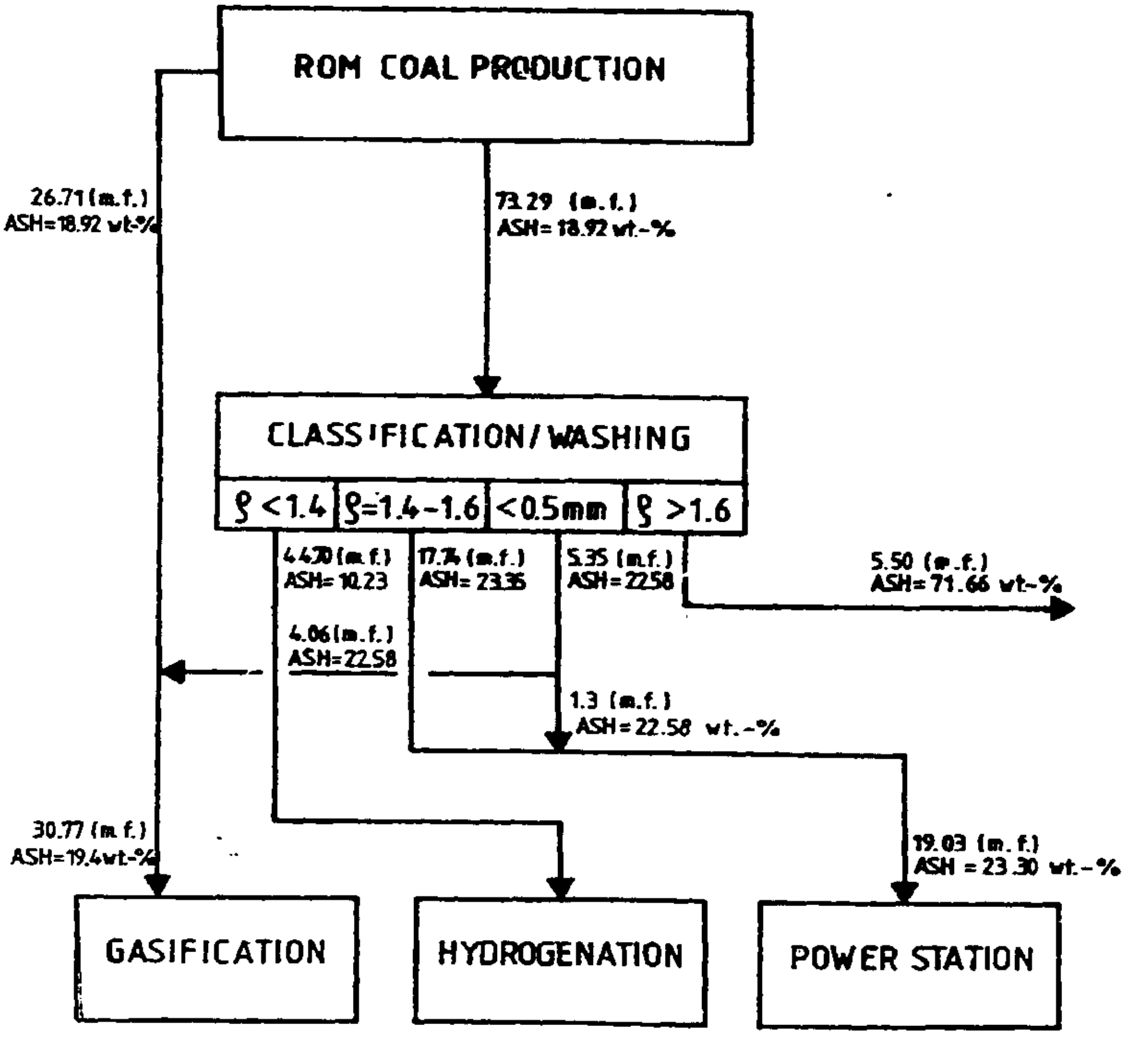
The capital related costs as well as the operational costs are contained in the coal prices for the conversion plants.

Table 2

		New South Wales		Queensland		Victoria	
		as delivered	maf	as delivered	maf	as delivered	maf
Carbon	wt.%	64.8	83.1	52.8	79.2	27.2	70.9
Oxygen		7.0	9.0	9.0	13.5	8.9	23.1
Hydrogen		4.1	5.3	4.0	6.0	1.8	4.7
Nitrogen		1.6	2.0	0.5	0.7	0.3	0.9
Sulphur		0.5	0.7	0.3	0.4	0.1	0.4
Other		0.1	0.1	0.1	0.2	-	-
Ash		17.7	-	25.5	-	1.66	-
Moisture		4.2	-	7.6	-	60.0	-
LHV	(kJ/kg)	25 832	33 352	21 219	32 134	9 002	26 901
	(kcal/kg)	6 171	7 944	5 069	7 676	2 150	6 426

COAL PREPARATION NEW SOUTH WALES

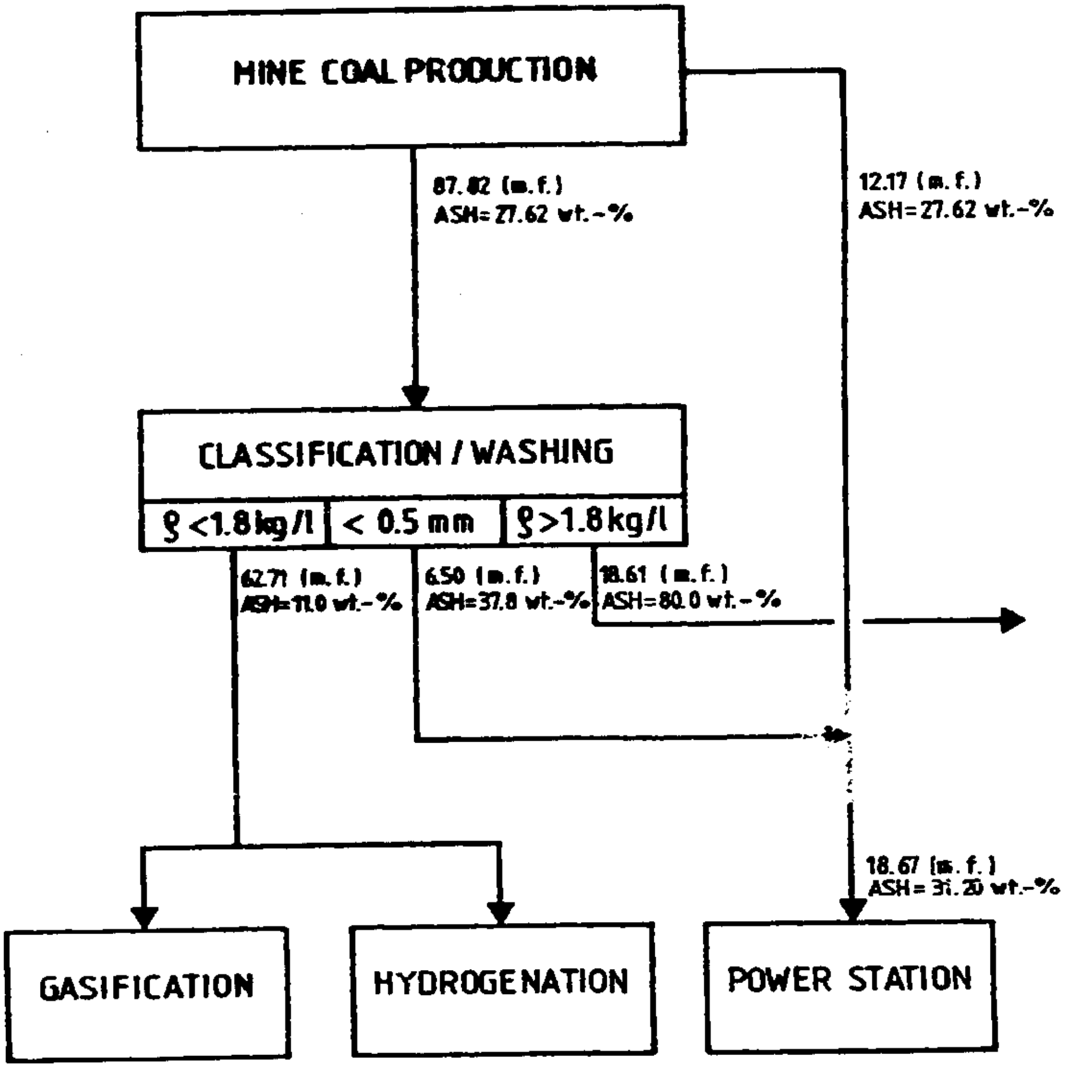
Fig. 1



CARBON LOSS : 1.9 wt.-%

COAL PREPARATION QUEENSLAND

Fig. 2



CARBON LOSS: 5.1 wt.-%

4. Hydrogenation Test

For the design of the various process components which had to be coordinated, hydrogenation tests were performed with coal samples made available from Australia. The target to be achieved, was to determine the process conditions with respect to maximize the oil generation and to optimize the design throughput capacity at the same time.

4.1 New South Wales

New South Wales coal was washed to 9 - 10 % ash content and hydrogenated in a continuous pilot plant located at Bergbauforschung GmbH in Essen. The results of the hydrogenation tests can be found in Table 3.

The oil comprises 1/3 light oil with a boiling end point of 200°C and 2/3 middle oil with an end point of 325°C. The vacuum residue contains 53 % solids and could yield 23 % of a distillate. This residue, when molten, flows easily and can be pumped without difficulty. Hydrogenation tests have shown that the New South Wales coal after ash reduction is also well suited for producing automotive fuels by hydrogenation.

4.2 Queensland Coal

Queensland bituminous coal was washed to 9 - 10 % ash and hydrogenated in a continuous pilot plant located at Bergbauforschung GmbH in Essen.

The results of the hydrogenation can be found in Table 3. The oil consists of 1/3 light oil with a boiling end point of 200°C and 2/3 middle oil with an end point of 325°C.

The vacuum residue contains 55 % solids and could yield 20 % of a distillate. This residue, when molten, flows easily and can be pumped without difficulty. Hydrogenation tests have shown that the Queensland coal after ash reduction is well suited for producing automotive fuels by hydrogenation.

4.3 Victorian Brown Coal

This coal was hydrogenated in a continuous pilot plant located at Union Rheinische Braunkohlen Kraftstoff Aktiengesellschaft, Wesseling, a subsidiary of Rheinische Braunkohlenwerke Aktiengesellschaft.
The coal samples for these tests showed important deviations from coal analyses assumed previously which were the basis of the optimization. The coal had a higher ash content. Specially the higher content of calcium oxide caused disturbances in the beginning of the hydrogenation tests. In addition, a different viscosity behaviour of the coal oil slurry required stronger drying.

The feasibility study is based on results obtained from the Victorian brown coal test sample. The problems caused by the high content of calcium oxide have been solved and are, therefore, of minor importance.

Hydrogenation tests with the Victorian brown coal have yielded favourable results and have proved that this material is well suited for producing automotive fuels. The oil comprises approx. 1/3 light oil with a boiling range from 30 to 200°C and 2/3 middle oil with a boiling end point of 325°C.
The vacuum residue contains approx. 25 wt.% ash and can be pumped without difficulty.
The most important results are to be found in Table 3.

Table 3 Main Results of Hydrogenation Tests

	New South Wales	Queensland	Victoria
Raw material	bituminous coal	bituminous coal	brown coal
Input			
coal mf	111.2	110.7	104.3
hydrogen	5.7	5.3	6.0
catalyst	3.8	3.7	3.3
sulphur	1.0	1.3	2.2
Na_2S	0.2	0.2	-
Output			
oil	44.0	49.9	48.0
residue	49.0	38.2	31.3
reaction water	3.0	8.2	15.0
hydrogen gases	22.0	19.2	9.1
other gases	3.9	5.7	12.4
Main catalyst component	Gladstone red mud	Gladstone red mud	Hamersley iron ore

all figures in t

5. Technical Basis of Design

Based on the hydrogenation tests of the coal samples the different process steps were designed and are illustrated in the attached overall block flow diagrams.

5.1 New South Wales and Queensland Plants

The plants for New South Wales and Queensland are based in contrast to the Victorian plant on the combination of hydrogenation and Fischer-Tropsch synthesis in order to achieve the required specifications for gasoline and automotive diesel oil.

The drawing No. 1-3-00/01, page 1, illustrates the process blocks with the major product streams for the following units :

- Milling, drying, slurry preparation
- Coal hydrogenation including gas conditioning
- Hydrogen production and LPG recovery
- Coal oil upgrading comprising several steps

This drawing also covers the pressure gasification of bituminous coal and hydrognation residue for the manufacture of Fischer-Tropsch synthesis gas and the gasification of bituminous coal for the production of fuel gas including fuel gas purification by desulphurisation.

Milling, Drying and Slurry Preparation

The washed and de-ashed coal is supplied from the coal storage to the coal preparation section where the coal is ground and its moisture content reduced to 0.5 wt.%. The coal is slurried in recycled oil from hydrogenation and catalyst, sulphur and sodium sulfide are added.

Coal Hydrogenation and Vacuum Distillation

The hydrogenation section comprises 5 trains with 3 reactors each. After separation of products in the hot separators, the reacted slurry is charged to the vacuum distillation. The residue from vacuum distillation is supplied to Texaco gasification.

The overhead product from the vacuum distillation is a part of slurrying oil.

The gas phase from the hot separator, after passing through heat-exchangers, is cooled further. The condensed coal oil, after separation from aqueous phase is charged to atmospheric distillation.

Atmospheric Distillation / Naphtha Distillation

In this process step the coal oil is separated into the following products :

Petroleum ether
Naphtha
Middle oil
Solvent, as the more important part of slurrying oil and various gas streams.

Coal Oil Upgrading

The upgrading of the coal oil fractions requires the following process units :

- Naphtha Hydrotreating in 2 stages
- Middle Oil Hydrotreating in 2 stages
- Middle Oil Stabilisation
- Hydrocracking in 2 stages and Distillation
- Catalytic Reforming and Stabilisation

The outputs of products from these process units are :

Gasoline blending component
Diesel oil blending component
and various gas streams.

Process Gas Selection and Production of Hydrogen

Hydrogenation yields rich and lean gases which are treated separately. Rich gases are the gases with a high content of hydrocarbons. Lean gases have less hydrocarbons. The rich gases are freed from C_3 and C_4 hydrocarbons and then used with the lean gases as feed for the hydrogen production.

The hydrogen production section comprises the following units :

- Hydrogen sulfide removal (ADIP process)
- Final desulphurisation (zinc oxide)
- Steam reforming
- CO shift conversion
- CO_2 removal (Benfield process)
- Sulphur recovery (Claus process)

The total amount of hydrogen including hydrogen rich gases from the upgrading units and a small quantity from the Fischer-Tropsch synthesis is compressed to the following pressure levels depending on the requirements of the downstream process units :

70 bar
115 bar
240 bar
340 bar

LPG Recovery

The LPG recovery plant processes four gas streams. The LPG ist separated by absorption and distillation and is charged to the butane isomerisation unit.

Production of Fischer-Tropsch Synthesis Gas by Texaco Gasification of Coal and Residue

This section comprises 2 process units :

- Gasification of bituminous coal
- Gasification of vacuum residue

The raw gases are conditioned in the following process steps :

- CO shift conversion
- COS hydrolysis
- Carbon dioxide and hydrogen sulphur removal by Selexol wash
- Final purification (zinc oxide)
- Sulphur recovery (Stretford process)

The output of purified synthesis gas is sent to Fischer-Tropsch synthesis.

Fuel Gas Production

A part of bituminous coal is gasified to produce fuel gas. Texaco gasifiers are installed for this duty. The fuel gas containing 0.11 Vol% H_2S is purified in a desulphurisation plant consisting of a Selexol wash unit and a sulphur recovery unit operating by the Claus process. Fuel gases from the Fischer-Tropsch synthesis are added.

The total amount of fuel gas has 0.0041 Vol% COS and H_2S and has a pressure of 25 bar.

Drawing No. 1-3-00/01 , page 2

Fischer-Tropsch Synthesis

The Fischer-Tropsch plant consists of 2 stages and has a conversion efficiency of 87 %. The plant is equipped with 13 tubular reactors. The products, condensates and waste waters are treated in the following plants.

Primary Product Distillation

This plant separates the liquid products from Fischer-Tropsch synthesis. The process comprises atmospheric distillation, stabilisation and vacuum distillation. The products include fuel gas, LPG, naphtha, primary diesel oil and hydrocarbon fraction $> C_{18}$.

Naphtha Distillation and C_5/C_6 Isomerisation

The feedstocks originating from primary product distillation and consisting of C_5/C_6 hydrocarbon fractions are separated into 3 fractions :

C_9/C_{12}	fraction to be added to the diesel pool
C_7/C_8	fraction to be added to the gasoline pool
C_5/C_6	fraction to isomerisation.

The purpose of the isomerisation unit is to increase the octane number of the gasoline product.

Tail Gas Separation

This plant separates and purifies the tail gas derived from Fischer-Tropsch synthesis.

The plant comprises 5 process steps :

- CO_2 removal
- Gas drying
- CO recovery in the COSORB unit
- H_2 recovery in the pressure swing absorption unit
- C_3/C_4 absorption and distillation

The following products are separated and recovered :

- C_3/C_4 fraction
- Hydrogen for the wax hydrocracker

- CO-containing gas for Fischer-Tropsch synthesis
- Sour gas (carbon dioxide)
- C_5 and higher hydrocarbon fractions for isomerisation
- Fuel gas, mainly consisting of methane and ethane.

Non-Acid Recovery and Mobil Unit

The aqueous phase of the Fischer-Tropsch synthesis consists of 95 % water and 5 % aliphatic alcohols, aldehydes, ketones and acids. After neutralisation and distillation the overhead product is sent to the Mobil unit. The bottom product from distillation goes to the biological effluent water treatment unit.

In the Mobil unit the non-acid chemicals (NAC) are converted to aromatic and olefinic hydrocarbons in the presence of a zeolite catalyst. The output of hydrocarbons is used as gasoline blending components.

Paraffin Wax Hydrocracker

The C_{19}+ bottom product from Fischer-Tropsch primary product distillation is passed to a hydrocracking unit to recover the low-boiling hydrocarbons such as diesel oil and naphtha. This unit comprises three process steps:

- Hydrogenation
- Hydrocracking
- Fractionation

In this unit are produced :

- Gasoline blending component
- Diesel oil blending component
- Fuel gas

Butane Isomerisation and Alkylation

In this plant the C_3/C_4 hydrocarbon streams from hydrogenation and Fischer-Tropsch synthesis are suitably treated. The C_3/C_4 hydrocarbon mixture from hydrogenation is distilled and the C_3/C_4 hydrocarbons separated to obtain the quantity of normal butane required for isomerisation and alkylation. The normal butane is converted to isobutane in the isomerisation unit and the isobutane is converted with olefinic hydrocarbons such as propylene and butylene from the Fischer-Tropsch synthesis plant to branched C_7/C_8 hydrocarbon fractions in the alkylation unit.

Output of products:

alkylates as gasoline blending component
butane as gasoline blending component
LPG, and
propane

Phenosolvan Plant

This plant separates phenols and ammonia from the process effluents water prior to biological treatment. The recovered crude phenol is returned to high pressure hydrogenation. The crude ammonia is charged to ammonia recovery plant.

Biological Process Water Treatment

The effluent water is to be treated. Hot water has to be cooled to 30°C before it is fed to the bio-treatment basins. After this biodegradation the water is filtered. The sludge is burnt.

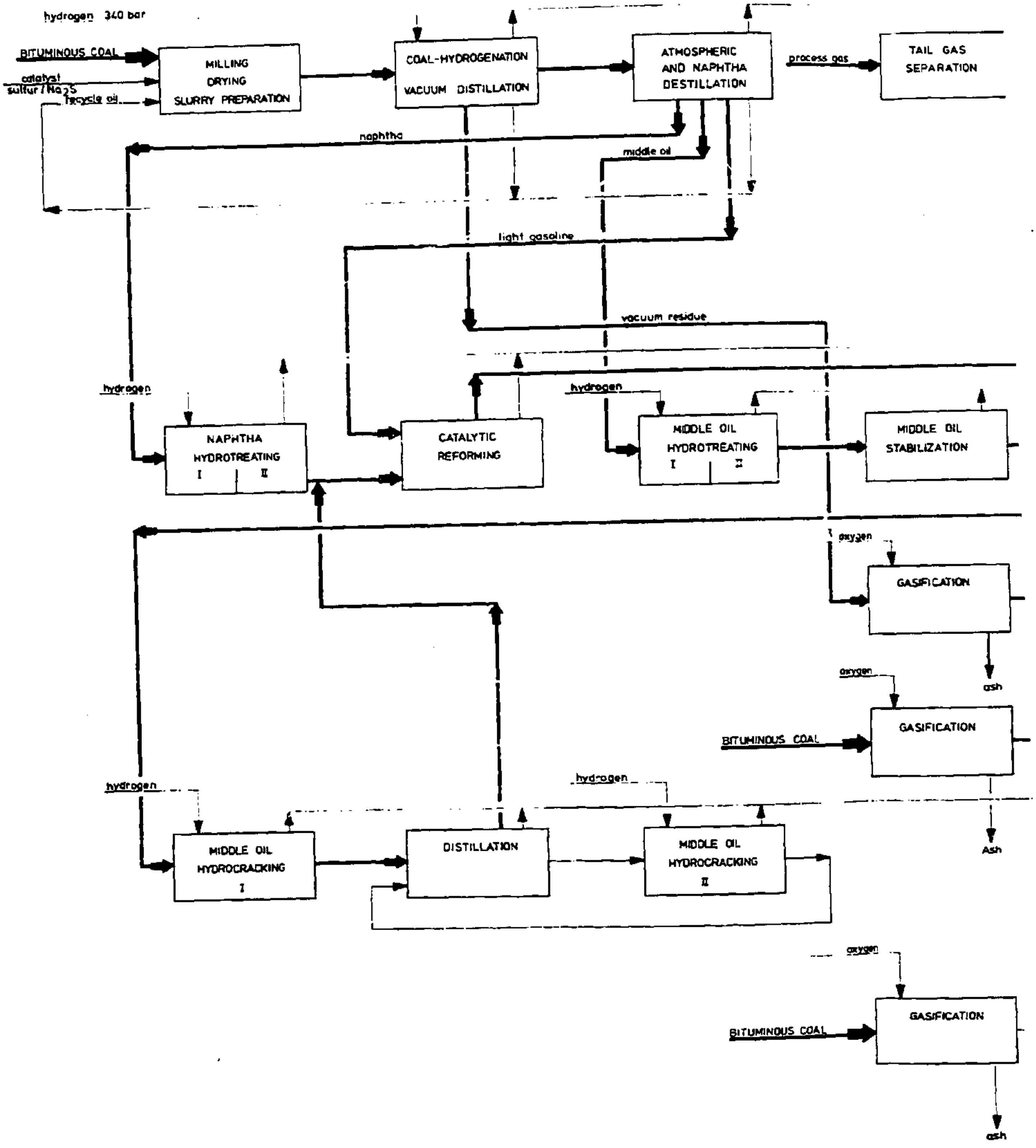
The treated water is expected to have :
a COD of 100 - 120 ppm and
a BOD_5 of 10 ppm.

Fischer-Tropsch Catalyst Preparation

The capacity per year is 2,500 m^3 of reduced catalyst. For the inertisation of the catalyst nitrogen with an oxygen content of max. 10 ppm will be supplied from the air separation plant.

Ammonia Recovery

Ammonia is recovered from a stream coming from the phenosolvan plant. Liquid ammonia with a purity of 99.9 % is produced. The water leaving the system has less than 100 ppm ammonia.


hydrogen 340 bar
BITUMINOUS COAL
catalyst
sulfur / Na2S
recycle oil
MILLING
DRYING
SLURRY PREPARATION
COAL-HYDROGENATION
VACUUM DISTILLATION
ATMOSPHERIC
AND NAPHTHA
DESTILLATION
process gas
TAIL GAS
SEPARATION
naphtha
middle oil
light gasoline
vacuum residue
hydrogen
NAPHTHA
HYDROTREATING
I
II
CATALYTIC
REFORMING
hydrogen
MIDDLE OIL
HYDROTREATING
I
II
MIDDLE OIL
STABILIZATION
oxygen
GASIFICATION
ash
oxygen
BITUMINOUS COAL
GASIFICATION
Ash
hydrogen
MIDDLE OIL
HYDROCRACKING
I
DISTILLATION
hydrogen
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oxygen
BITUMINOUS COAL
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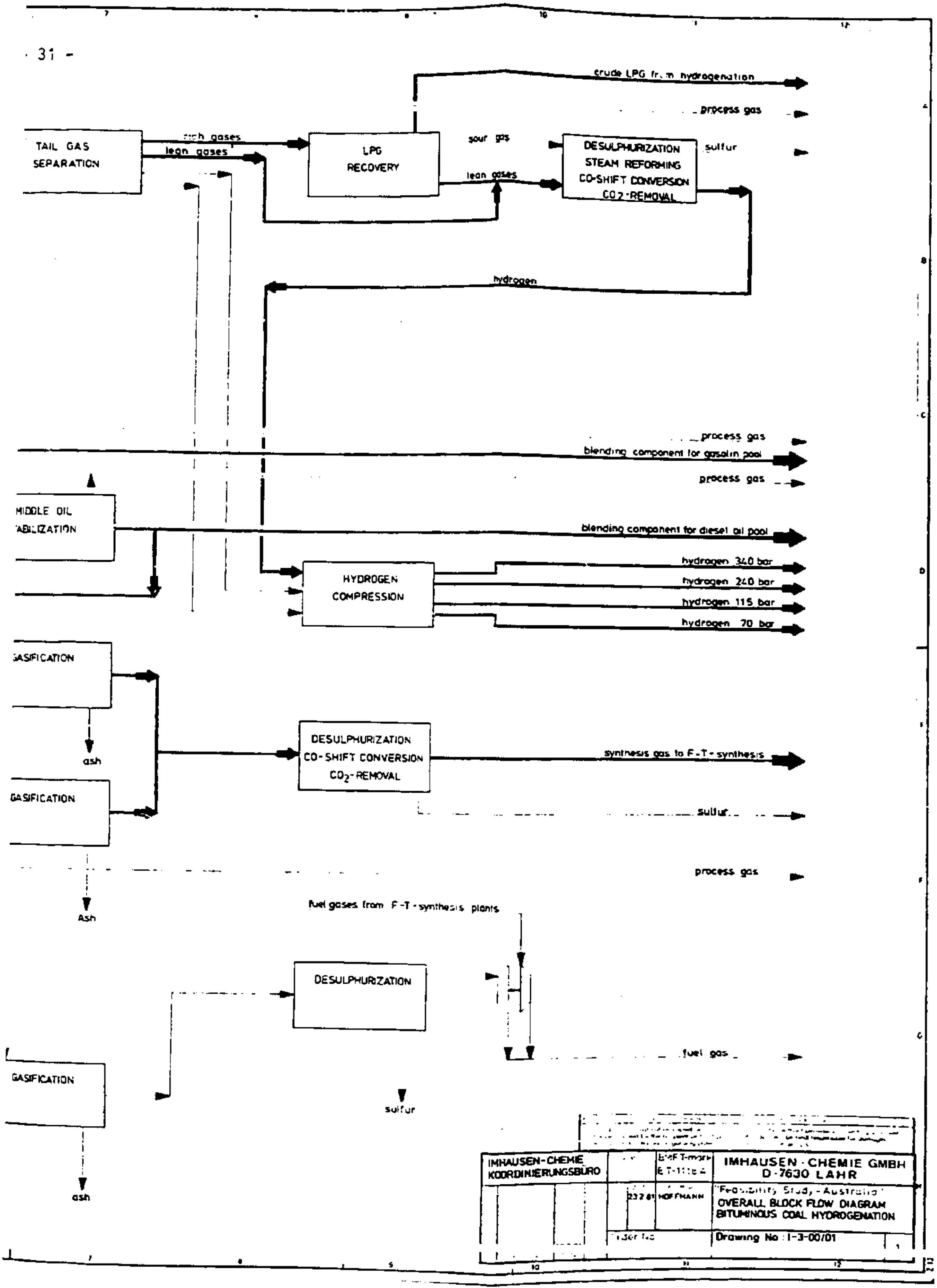

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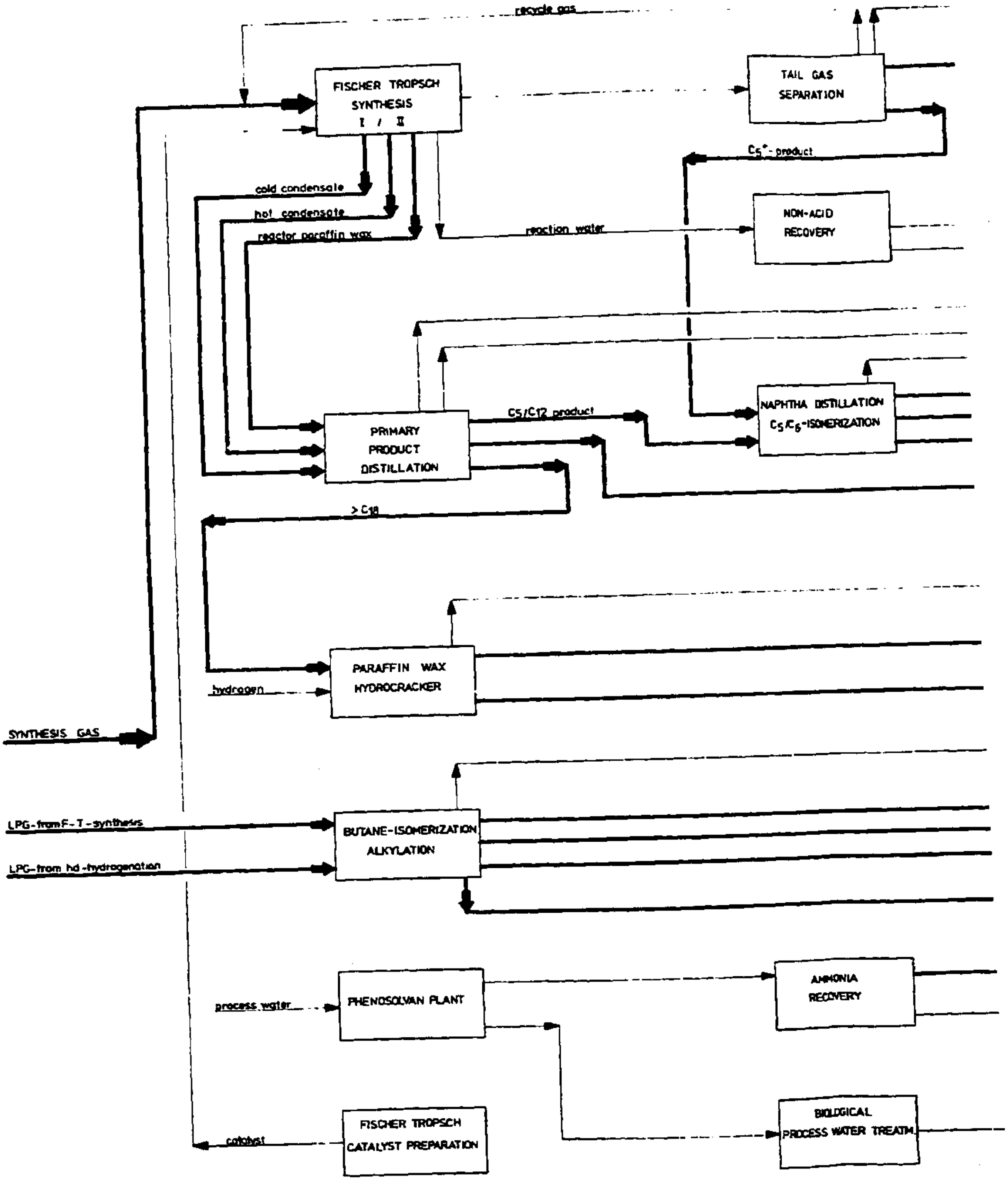
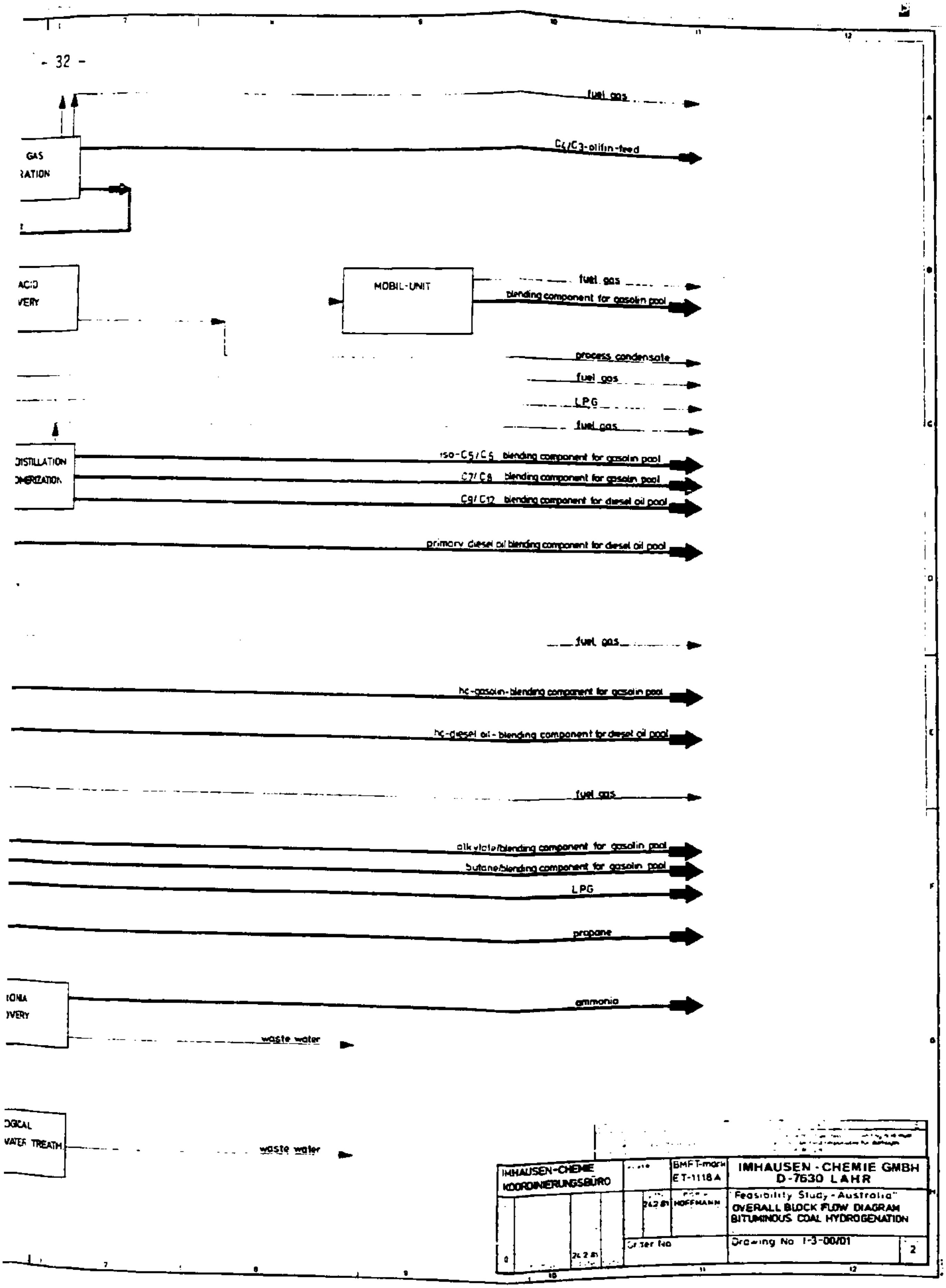
"Feasibility Study - Australia"
OVERALL BLOCK FLOW DIAGRAM
BITUMINOUS COAL HYDROGENATION

Order No.

Drawing No.: 1-3-00/01


recycle gas
FISCHER TROPSCH SYNTHESIS I / II
TAIL GAS SEPARATION
C5+- product
cold condensate
hot condensate
reactor paraffin wax
reaction water
NON-ACID RECOVERY
PRIMARY PRODUCT DISTILLATION
C5/C12 product
NAPHTHA DISTILLATION C5/C6-ISOMERIZATION
> C18
PARAFFIN WAX HYDROCRACKER
hydrogen
SYNTHESIS GAS
LPG-from F-T-synthesis
LPG-from hd-hydrogenation
BUTANE-ISOMERIZATION ALKYLATION
process water
PHENOSOLVAN PLANT
AMMONIA RECOVERY
BIOLOGICAL PROCESS WATER TREATM.
catalyst
FISCHER TROPSCH CATALYST PREPARATION



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OVERALL BLOCK FLOW DIAGRAM
BITUMINOUS COAL HYDROGENATION

Order No

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