Secondly, the I. G. staff have found that, when the hydrogenation process is operated in the normal way using a gradual upward temperature gradient in the reactor system, much of the asphalt make is produced in the last converter where temperature is at a maximum and hydrogen partial pressure is at its lowest. Dropping the temperature in the last converter materially reduces the asphalt make and it also helps to reduce the solids content of Abschlamm and, therefore, the hot catchpot coking danger.

At Leuna, this reduction of temperature in the last converter can be practised only in conjunction with high feed rates resulting in a high rate of heat evolution per stall. At lower throughputs, the loss of recoverable heat by cooling the last converter introduces a preheater capacity limitation.

The Leuna staff feel that still greater throughputs would be possible if it were not for two other limiting factors, (1) erosion of preheater tubes and (2) the capacity of the existing Schneckenofen. With the present high rates preheaters have to be shut down for examination every 120-150 days and this shut-down takes 6 days. Incidentally, the general maintenance shut-down of a brown coal stall takes 25 days and occurs, normally, every 15 months. Kugel ovens are considered to be superior to Schneckenofen and several were on order for installation on the Leuna plant.

(iii) Details of Plant Equipment

Paste Mills. These are simply horizontal rotating ball mills fed from each end with the appropriate quantities of dried coal and pasting oil, measurement of both being automatically controlled. The paste leaves the mill via slots cut in the side and passes through a 1 mm. screen for removal of nibs of coal, tramp iron, etc.

Paste Injectors. These are ram pumps, the power for both injection and suction strokes being hydraulically supplied. Injection rate is varied by controlling the rate of entry of water into the hydraulic cyclinder.

The gland packing for these injectors had latterly been lead alloyed with 0.1% Cadmium and this had been satisfactory. Before the war, tin packings were used.

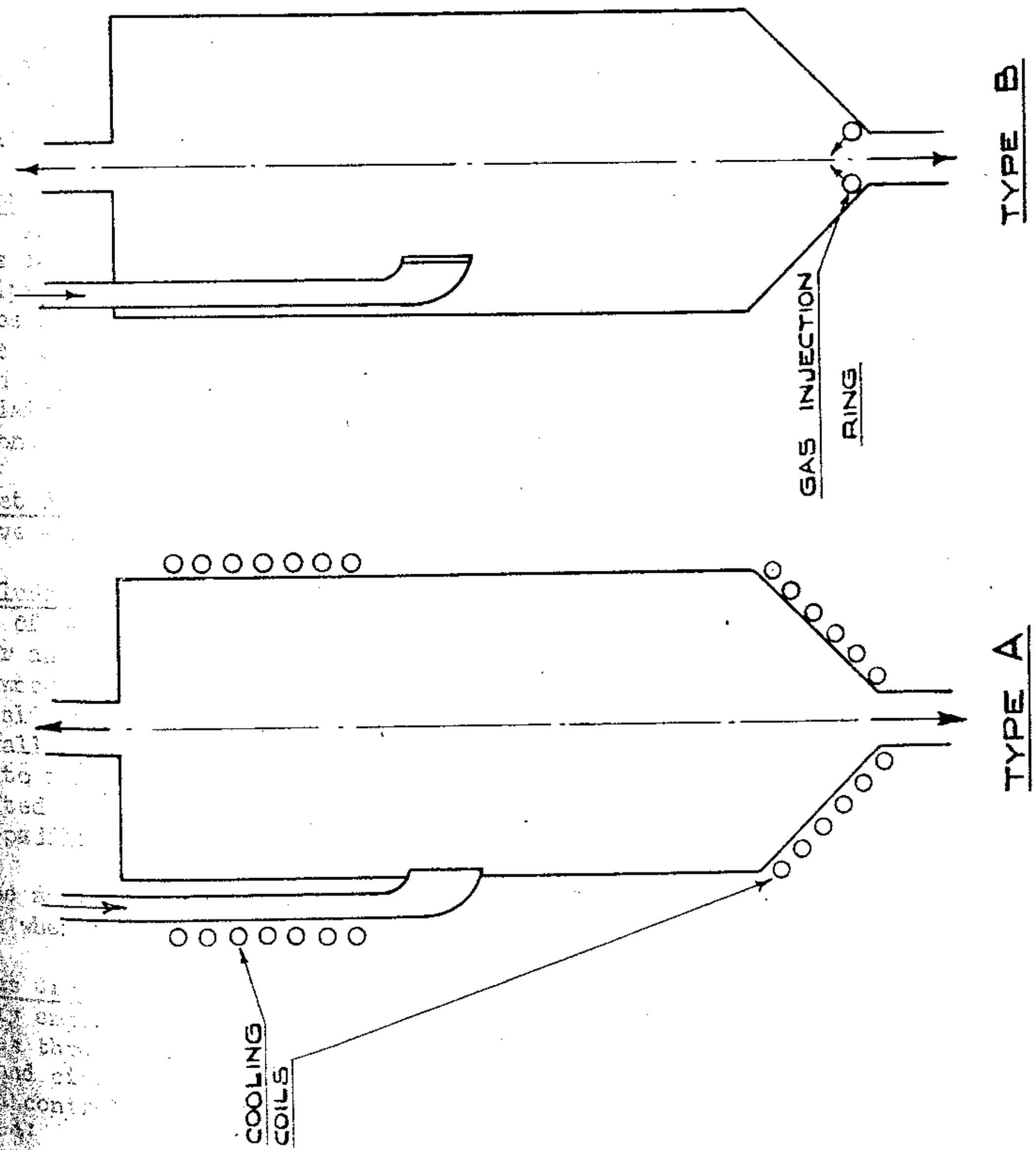
The I. G. had ceased to use nitrided rams because of the wide variation in quality which had been experienced. Ordinary case hardened rams of 400 B.H. were being used.

Paste Interchangers consist of a bundle of 199 tubes in a forging 600 mm. diameter and 18 M long. Formerly 241 tubes were used but trouble was experienced with chokes. The tubes are zinc treated.

With a paste feed rate of 30 M^3/hour and a gas rate of 20,000 M^3/hour the K value is 250-200. Units for K value: Kg. cals/M^2/°C/hr. Interchangers are stripped every two years.

FIG. IV

HOT CATCHPOTS



Paste Preheaters. These are gas-fired convection type heaters of the standard I. G. rectangular design. The tube hairpins are 15.5 M long, 120 mm. bore and 171 mm. external diameter. They are fitted with fins 4 mm. thick and 10 mm. apart and which give the tubes an internal/external arearatio of 1 : 20. 6-7 hairpins are used in a normal preheater the average overall K value of which is 180-150. With high feed rates erosion of the hairpin bends is appreciable and the average life of a bend had fallen to 18 months. It was proposed to change to tubes of 160 mm. bore and 229 mm. external diameter with fins 4 mm. thick and 320 mm. square spaced 16 mm. apart. The hairpins were to have been made with a pitching of 500 mm. instead of the standard 400 mm.

Brown coal stall Converters. Leuna used converter forgings of a variety of sizes varying from 800 - 1200 mm. diam. and from 11-18 M. in length. A stall had four converters and the size was usually such that the total stall reaction volume was roughly 27 M^3.

Hot Catchpot. Several designs have been tried in an attempt to reduce coking. The type most commonly adopted is shown in Fig. IV A. It is fitted with gas cooling coils between the forging and inner catchpot wall and the inlet pipe for reactants is outside the inner vessel. The idea of the cooling tubes at the top of the catchpot is to ensure sufficient condensation that the walls never dry out. Another type (Fig. IV B) has no cooling coils but cold gas is introduced at the bottom of the vessel via a ring distributor. It is claimed that the stirring provided by this method helps to reduce coke formation.

Let down valves. These are fitted with "Widia" inserts but even so they have a life of only 200-400 hours.

Sludge carbonisation kilns. Leuna has 26 Schneckenofen arranged in sets of two. Each kiln comprises two horizontal steel tubes 2 ft. in diameter and 30 ft. long, arranged one above the other and sitting inside a brickwork gas-fired furnace. Each tube is fitted with paddles, fixed with a slight pitch on to a central rotating shaft, and these scrape the inner wall of the tube free from adhering coke. Loose iron bars are attached to the paddles in such a way that, as the paddle rotates, the bars are lifted to fall later with sufficient force to dislodge coke deposited on the paddle.

The average feed rate is 2.5-3.0 M^3/hour falling to 1.8 M^3/hour after 50 days when the unit is taken off line for cleaning.

Gas Circulators. The coal and heavy oil hydrogenation system at Leuna normally employs 5 or 6 circulators each of 80-85,000 M^3/hour capacity. The machines throw a maximum pressure of 30 ats. They are operated at constant speed and circulation rate is varied by controlled bypass. Stall pressure is hand controlled and it was stated that swings of up to 20 ats. are observed:

Gas washing. Purification of the circulating gas is carried out in 9 towers each 1300 mm. diameter. Wash oil (a heavy cut of liquid phase hydrogenation middle oil) is injected partly with three throw pumps and partly by means of three machines driven by let down engines operating on the saturated wash oil. The wash oil feed rate was controlled automatically in order to maintain a constant hydrogen content of circulating gas.

Automatic Controls. Full details of the various automatic controls were obtained but these are amongst the documents (2nd Leuna visit) which have not yet reached London.

C. Brown Coal Tar Hydrogenation

The Leuna flowsheet for hydrogenation of Central German brown coal tar is shown in Fig. V. The crude tar which contains 1% water, 0.5% solids and 1-3% asphalts, is fugalled to remove slightly over 2% of its weight of sludge which is sent to carbonisation for oil recovery. The cleaned tar is mixed with "Leuchtöl", i.e. recovered oil from brown coal carbonisation gases (the crude tar + Leuchtöl mix usually contains 45% by weight of heavy oil boiling above 325°C, 35-40% distilling between 180 and 325°C and 15-20% boiling below 180°C) and with the crude product from the liquid phase hydrogenation of heavy oils. This mixture is distilled to give a 0-325°C fraction as overhead and a heavy oil bottoms which is the feed to the hydrogenation stalls.

The plant employed is substantially the same as in the case of coal hydrogenation with the exception that a special pump is installed for hot recycle of liquid from the hot catchpot to the inlet of the preheater.

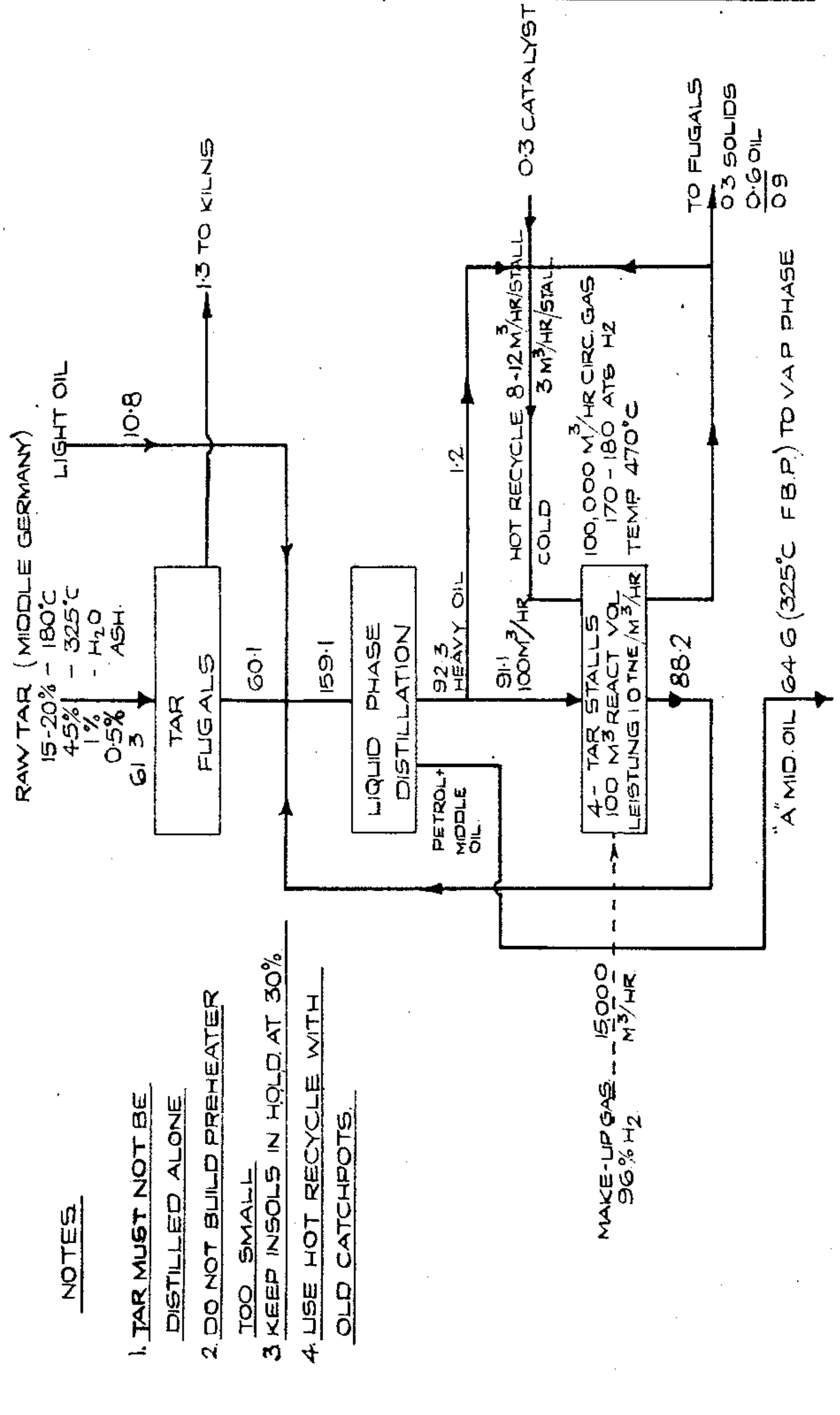
Hydrogenation conditions for heavy oil are:

Circulating gas/Oil feed	1,000 M^3/Ton
Mean Reaction Temperature	480°C
Oil feed rate	0.9 - 1.0 kg/litre catalyst/hour.
Pass Conversion of heavy oil into lighter products.	35%
Total Pressure	230 ats.
Hydrogen Partial Pressure	170-180 ats.
Catalyst	5 - 6% Iron on Grude coke in suspension in the reactants.

The catalyst (10927) is made by impregnation of the coke obtained from Winkler generators with the necessary amount of ferrous sulphate and caustic soda solutions in stoichiometric proportions. The impregnated coke is dried and ground so that not more than 2% remained on a 10,000 mesh screen. There is no washing step in the preparation of this catalyst. Make up catalyst is introduced as a suspension in part of the heavy oil feed usually injected into the hot liquid recycle system. Catalyst consumption is roughtly 1% by weight on the fresh > 325°C heavy oil feed.

FIG. V FLOWSHEET FOR PETROL FROM BROWN COALTAR

ALL QUANTITIES TNE/HR EXCEPT WHERE STATED



NOTES

1. TAR MUST NOT BE DISTILLED ALONE
2. DO NOT BUILD PREHEATER TOO SMALL
3. KEEP INSOLS IN HOLD AT 30%
4. USE HOT RECYCLE WITH OLD CATCHPOTS.

The main purpose of the hot liquid recycle from the hot catchpot to the preheater is to increase the velocity of liquid in the former and so reduce the coke formation. The rate of recycle necessary depends on the hot catchpot design. With the later types, hot recycle can be eliminated and it was stated that Brux operated in this way.

In order to purge spent catalyst and non hydrogenable material in the tar, there is a small let down from the hot catchpot. Part of this product is recycled direct and the remainder treated in fugals to give a concentrate which is carbonised for oil recovery.

The yield of petrol and middle oil obtained by liquid phase hydrogenation of brown coal tar heavy oil is just over 80% by weight. Hydrogen consumption is 3.9 to 4.0% by weight. It is interesting to note that the petrol + middle oil production "leistung" per M^3 reaction volume is lower from brown coal tar heavy oil than from brown coal. The figures are 0.25-0.26 Tons/M^3/hour for tar heavy oil and 0.29-0.30 for coal.

D. Vapour Phase Hydrogenation of Middle Oils

(i) Process conditions and yields

Leuna operates the two-stage process, i.e. straight through presaturation to convert nitrogen compounds to ammonia and to hydrogenate the greater part of the oxygenated components followed by splitting hydrogenation with recycle of unconverted middle oil.

For each stage the plant consists simply of a preheater, two interchangers, three or four converters, a cooler and a cold catchpot. Reaction volume per stall is usually about 25 M^3. For the first stage, Leuna, in common with most other German hydrogenation plants, have changed from 5058 (tungsten sulphide) in the form of 10 mm. pellets, to 8376 (25% tungsten sulphide + 3% nickel sulphide on alumina) catalyst used in the form of cubes This was found to be as good as 5058 for reduction of phenols and superior to 5058 in that it gave less splitting to comparatively low quality petrol. It was not as good as 5058, however, for destruction of nitrogen compounds which is the most important function of the presaturation stage.

Leuna achieved an oil throughput of 0.6-0.8 Tons/M^3 catalyst/hour in the presaturation stage, for which an average temperature of 410°C and a hydrogen partial pressure of 210-220 ats. was employed. Circulating gas to oil ratio was 2,500 M^3/Ton. The high hydrogen partial pressure in the vapour phase stalls is achieved by introducing all the make up hydrogen for both liquid and vapour phase systems into the vapour phase circulating gas. A purge from this system is used as make up to the liquid phase.

Gas made in the presaturation step is the equivalent of 3% of the carbon in the feed. Roughly half this gas (by weight) is butane, a third propane and the remainder ethane and methane in approximately equal proportions. The yield of butane-free product depends of course on the oxygen, nitrogen and sulphur content of the feed. In the case of total 0-325°C product from brown coal tar it is 95% and hydrogen consumption is 3.9%

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by weight. Using petrol + middle oil from brown coal hydrogenation as feed, the yield of saturated product is 97% and hydrogen consumption 3.7-3.8%.

The saturated product from both brown coal and brown coal tar contains an appreciable amount of material boiling in the petrol range. This fraction has a comparatively poor knock rating and it is desirable to keep it separate from the better quality product obtained by splitting hydrogenation. It is, therefore, usual to distil the saturation product, taking overhead a 0-150 or 160°C cut if motor gasoline is the desired end product or a 0-120°C fraction if aviation base petrol is being made. It was stated that 0-120°C saturation stage petrol ex brown coal hydrogenation has a motor method octane number of 70-71 while the rating of the corresponding product from brown coal tar was only 68-70. Presaturation stage petrol is frequently used as feed for the D.H.D. process.

The middle oil from the presaturation stage contains 2-3 mgs. nitrogen per litre. It can be used direct as a Diesel fuel and has a Cetane number of 40-42 and a pour point of -15°C. The pour point can be lowered to -40°C by cutting 15% from the higher boiling end but this reduces the Cetane number to 38.

Splitting hydrogenation of saturated middle oil is carried out over a tungsten sulphide on Terrana earth catalyst (6434). Other process conditions are:

	For Motor Gasoline	For Aviation Base Gasoline
Oil throughput Kgs/litre catalyst/hour	0.8	0.8
Conversion/pass	60%	55%
Circulating gas/oil feed ratio. M^3/Ton	1,500	1,700
Hydrogen Partial Pressure	210 - 220 ats.	210 - 220 ats.
Temperature	400°C	410°C

Circulating gas is water washed before entering the stall to reduce nitrogen to less than 0.1 mgrm/litre oil feed. Motor gasoline is cut to an end point of 190°C and has a volatility of about 30-35% at 100°C. Aviation base gasoline distils 65-70% at 100°C and has an end point of 150°C.

When making motor gasoline, 15% of the carbon in the 150-325°C saturated middle oil feed is converted into hydrocarbon gas consisting of 83% by weight of butane, 15% propane, 1% ethane and 1% methane. It is possible, however, to include 40-50% of the butane in the final petrol so that the net weight yield of motor gasoline is 91-92% on the feed.

20% of the carbon in the 120-325°C feed is converted into hydrocarbon gas of the same composition as given above when making aviation base petrol. It is not possible to include butane in the liquid product and the net yield is thus only 81% by weight.

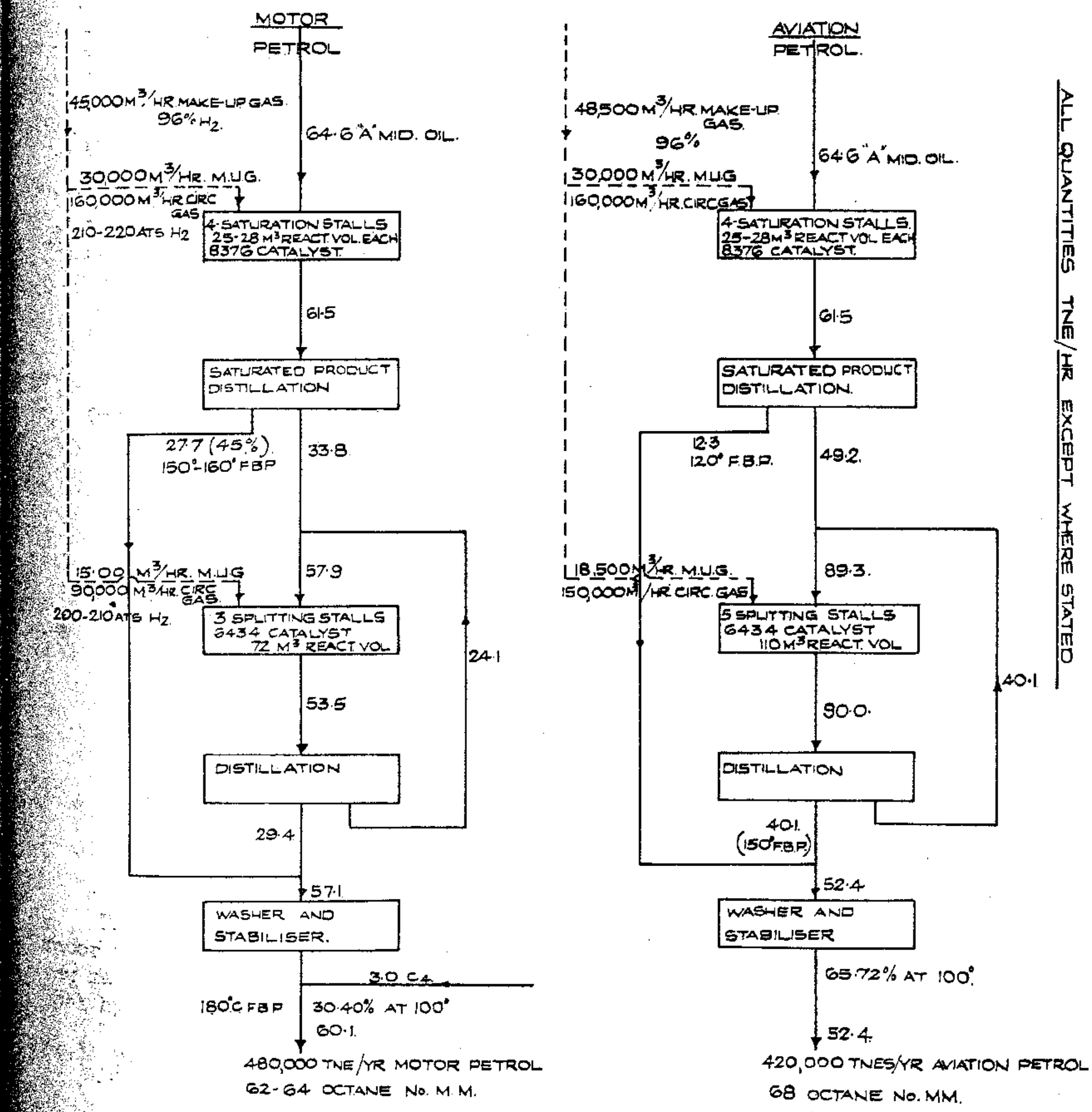

FIG VI FLOWSHEET FOR PRODUCTION OF MOTOR OR AVIATION PETROL FROM BROWN COAL MIDDLE OIL

ALL QUANTITIES TNE/HR EXCEPT WHERE STATED

Figs. VI and VIII show diagramatically the conversion to two grades of petrol + middle oil from liquid phase hydrogenation of brown coal tar (including Leuchtöl) and brown coal respectively. They show that the following overall yields are obtained:

	Motor Gasoline	Aviation Base Petrol
Brown coal tar		
(a) wt. % on crude tar including Leuchtöl	83	72.5
(b) wt. % on liquid phase petrol + middle oil	93	81
Brown coal		
(a) wt. % on ash and moisture free coal	46	40
(b) wt. % on liquid phase petrol + middle oil.	95	83

Overall hydrogen consumption figures are:

(1) Brown coal tar (including Leuchtöl) to Motor gasoline -

6.9 - 7.0% by weight on tar or .083-.084 Tons Hydrogen per Ton Motor gasoline.

(2) Same feed to Aviation petrol -
7.3% on tar or .1 Tons H_2/Ton petrol.

(3) Brown coal to Motor gasoline -
9.4% wt. on ash and moisture free coal or .203 Tons H_2/Ton motor gasoline.

(4) Brown coal to Aviation base -

9.6 - 9.7% wt. on a.m.f. coal or .24 Tons H_2/Ton aviation base.

The final blend of motor gasoline from brown coal tar has a C. F. R. motor method octane number of 62-64, while aviation base from the same raw material has a rating of 68. Corresponding ratings for petrols ex brown coal hydrogenation are about two octane numbers higher.

(ii) Details of Plant.

Injectors. Normal three throw pumps are used with electric drives. S. E. A. rings for gland packing had not been very satisfactory and the I. G. had used ACACIA wood packing with considerable success. These new packings had a life of about 1,800 hours.