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PRODUCTION OF LIQUID FUELS

from

COAL, LIGNITE and OILSHALES

in EUROPE

OUTLOOK

for the

DEVELOPMENT in U. S. A.

by

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## INTRODUCTION

The writing of this paper intends to give a general picture of the technical methods which have been developed in Europe for the production of motor fuels from coal and oil shale. Detailed descriptions however, have been given only of those processes, which have not been described in literature yet and as far as they can be considered as methods, to which can be attributed special new features, which might give such a process a chance to contribute to the solution of the technical problems even if they will still need many years of experimenting. Methods, which may have some local use in spite of small capacity and extraordinary high labor requirements have not been considered worth while for U.S.A. scale of production and the labor conditions of this country.

Investment and production cost of several processes have been given, but it is natural, that such figures comprise mostly typical methods with which the author had to do personally. These figures should therefore be considered as an introduction and a base for the discussion of the economical questions of the fuel problems and of the relative suitability of the various typical methods with respect to the raw materials and the conditions of production and consumption available in U.S.A.

The author's general experience with operating plants of the various methods and viewpoints gathered from prospecting and developing some of the latest big plants, which have been erected in Europe, have been communicated in this paper.

It has been tried then to examine the resources of U.S.A. and the suitability of the various technical methods for utilizing those raw materials for a slowly increasing production of motor fuels in case of an insufficient supply of natural oil.

# C O N T E N T S

## HISTORICAL DEVELOPMENT IN EUROPE

	1850 - 1914 Scotland, Germany, France	1
	1914 - 1918 World War I	1
	1918 - 1933 Development of new methods	2 - 3
	1933 - 1939 Erection of big industrial plants	3 - 4
	1940 - 1944 World War II. Experience and further development of the industry	4
II	RAW MATERIALS AVAILABLE IN EUROPE	4
	A. Essential properties of solid fuels.	4
	a. Moisture content	4 - 5
	b. Ash content and fusion point of the ash	5
	c. Contents of Volatiles and tar, Degree of carbonization	5
	d. Contents of Sulphur and Chlorine	5
	e. Sintering and caking property of coals	6
	B. The Most Important Deposits of Coal and Oil Shale of Europe	6
	A. Caking and bituminous coal	6
	Great Britain, Ruhr, Northern France, Upper Silesia	7
	b. Subbituminous coal and older lignite	7
	Central Germany, Hungary, Czechoslovakia, France	7
	c. Younger Lignites	7
	Rhine Valley, Eastern Germany	7
	d. Oil shale and bituminous rock	7
	Scotland, Estonia, France, Spain, Bulgaria, Southern Germany, Sweden, Holstein, Italy, Russia	8
III	TECHNICAL METHODS OF AN INDUSTRIAL PRODUCTION OF LIQUID FUELS FROM SOLID MINERALS	8
	A. Low Temperature Carbonization	8
	a. Caking Coals	8

The Coalite retort	9
Krupp - LURGI retort	9
The B. T. retort	9-10
b. Non-caking Coals and Briquettes	10
Continuous working vertical Retorts, Otto, Didier, Koppers	10-11
Shaft Carbonizers	11
LURGI-Carbonizer	11-12
c. Lignite raw or briquetted	12
The Geissen-Furnace	12-13
LURGI Carbonizer	13-16
d. Oil Shale and Bituminous Rock	17
1. The Pumpherstone retort	17
2. The Messel retort	17
3. The Goldfield rotary kiln or Davidson retort	17-18
4. The Otto furnace	18
5. The furnace of the Grande Paroisse Company	18-19
6. TUNNEL KILNS for Carbonization of Oil Shale	19
A. The Esti-Kivioeli tunnel kiln	19-20
B. The Sillamaegi tunnel kiln	20-28
7. The Pintsch-producer	28-29
8. LURGI-Carbonizer with Gasification Zone	29
9. The LURGI-Schweitzer System	29-31
Underground Carbonization	31
Pile method	31-32
10. The LURGI Huboven For Carbonization and Gasifica- tion of High-Ash-Fuels	33-42
e. Methods of briquetting lignite and fines of coal	43-45

B. By-product - Gasification	45
a. Gas producer for industrial heating gas	45-46
b. Gas producer for water gas and city gas	46
Pintsch-Hillebrand design of this process	46-47
THE LURGI CARBONIZER WITH GASIFICATION OF RESIDUE	48-52
LURGI - Pressure Gasification process	52-57
C. Refining of Aliphatic Tars	58
a. Fractionating of tar, Extraction of paraffin wax and Creosot oils	58-60
b. Cracking of tar and distillation under several atm. pressure	60
c. Refining of tar by extraction methods	60-61
d. Hydrogenation of tar under pressure	61
D. Hydrogenation of Coal and Lignite J. G. Bergius Process	62-64
E. Synthesis of Hydrocarbons (Fischer-Tropsch) by hydrogenation of carbon monoxide	64
a. Synthesis under atmospheric pressure	64
b. Synthesis under a medium pressure (10 - 20 atm)	65
c. Medium pressure Recircling method of LURGI	65
d. Medium pressure synthesis with an Iron - catalyst	66
e. The principal types of catalyst reactors	67
F. Production of CO / H <sub>2</sub> mixtures from coal	68
a. Water gas process for lump coal	68-69
b. The Didier process	69
c. Pintsch Hillebrand method. Koppers System (Cowper system)	69
d. Wintershall Schmalfeld process	69-70
e. Winkler-gasproducer	70-71
f. Koppers-Gasification of pulverized coal	71

g.	Slagging gas-producers, Leuna works, The Thyssen-Caloxy Producer	72-73
h.	Gasification with oxygen and steam in a rotating grate gas producer	73-74
i.	Combined Carbonization-Gasification with Oxygen	74-75
j.	The LURGI-pressure gasification process	75-76
k.	Relations between the properties of solid fuels and the special features of the various processes of gasification	76-79
G.	Purification of Gas and Recovery of By-products	79
	1. Removal of dust and tar	79-80
	2. Fractionating condensation	80-81
	3. For the Recovery of benzine	81-82
	4. For the Extraction of Hydrogen Sulfide	82
	5. ALKACID process	83
	6. Potash solution	83
	7. For the recovery of sulfur Claus oven process	83-84
	8. Washing with water	84-85
	9. Activated charcoal	85-86
	10. For the removal of organic sulfur compounds	86
H.	Treatment of Waste-Water, Recovery of Phenols	86-87
	1. Tricresylphosphate process	87
	2. The Koppers process	87-88
	3. Phenosolvan process LURGI	88
	4. Biological method	88-89
	5. Filtering of waste-water through ash-piles	89

IV CHARACTER OF PRODUCTS

A. The Products of Carbonization	90-94
B. Products of Hydrogenation, Bottled gas, Gasoline Diesel Oil	94-95
C. Products from synthesis of hydrocarbons Bottled gas, Gasoline, Diesel Oil, Waxes	95

V CHARACTERISTICS OF THE VARIOUS METHODS FROM ECONOMIC AND MILITARY VIEWPOINTS

a. Dependence on the character of the coal and the desired products	96
b. Dependence from other market or industries	96
c. Investment cost per ton of liquid motor fuel	97
d. The material required for the plants	97-98
e. Laborers required for erection of plant	98
f. The cost of production	98
g. Minimum size of plants	98
h. Military Characteristics	99

VI VIEWPOINTS WITH RESPECT TO THE PROBABLE FUTURE TECHNICAL DEVELOPMENT OF MOTORS, POWER STATIONS, INDUSTRIAL AND TECHNICAL HEATING 101

VII APTITUDE OF THE VARIOUS METHODS OF PRODUCTION FOR U.S.A. CONDITIONS 103

A. U.S.A. resources of coal and oil shale and their suitability for the production of liquid fuels	104
B. Possibility of application and Special Advan- tages of the various Methods of Producing motor fuels from coal	107
a. Carbonization and By-product gasification of coal and oil shale,	107
b. Hydrogenation of tar and coal	108
c. Synthesis of hydrocarbons	108

Attempt of a prognostication of the development of  
production of motor fuels in U.S.A.

109-111

PRODUCTION OF MOTOR FUELS FROM COAL, LIGNITE AND  
OIL SHALE. Comparison of various methods on  
German conditions (1939)

111 A

111 B

Drawings attached:

- Fig. 1 Elevation of a LURGI direct-heating  
Carbonization plant Follows page 13
- Fig. 2 OTTO FURNACE Follows page 17
- Fig. 3 Pintsch-producer for oilshale Follows page 28
- Fig. 4 HUBOVEN Follows page 34
- Fig. 5 DIAGRAM of a Browncoal-Briquetting  
Plant (LURGI-KRUPP Process) Follows page 44
- Fig. 6 Hillebrand Process Follows page 46
- Fig. 7 LURGI-CARBONIZER-GASPRODUCER Follows page 49
- Fig. 8 Utilization of Carbonizer-Gasproducer Follows p. 51
- Fig. 9 Production-cost of Synthesisgas
- Fig. 10 LURGI-PRESSURE-GASIFICATION for Synthesis gas
- Fig. 11 Motor fuels from Coal and Oilshale

At end of paper



## I HISTORICAL DEVELOPMENT IN EUROPE

1850 - 1914 The distillation of solid bituminous minerals has its origin in the lack of oils and fats for illuminating purpose. The highly bituminous lignites of central Germany called pyropissis offered a chance to produce waxes and oils suitable to replace animal fats used for illuminating. An equally favorable raw material was available in some Scotch oil shales. The production was started with small retorts made of firebrick. Experimental plants with retorts of different construction in brick and cast iron followed, but finally the technical development ended about at the end of the 19th century with the Rolle oven in Germany and with the Pumphirstone Retort in Scotland. A small scale treatment of oil shale had been started also in France on the same technical line as in Scotland.

The raw materials changing slowly to lower grade in Germany as well as in Scotland, the producers were forced to make a profit from by-products. Those were found for Scotland in an ammonia fertilizer produced by steaming the retorts and separating the ammonia from the gas with the methods used in gas works.

In Central Germany the lignite char proved to be a very good fuel (grude) for special house hold stoves. A good quality of this char as well as the maximum recovery of good paraffin wax required a moderate cracking within the retort.

The capacity of both types of retorts remained low with a maximum of 5 - 10 tons of raw material per day. The production cost was high, going up still with a slowly decreasing oil content of the raw material and the decreasing price of fertilizers.

### 1914 - 1918 World War I

The lack of motor fuel forced the German government to search for new methods suitable to produce oils from coal and lignite. Several proposals were made all based on the principle of gasification of coal or lignite in a gas producer completed by an additional distilling zone. 4 industrial plants of this kind of different construction were erected in 1917 and 1918 in Central Germany to treat raw lignite. All these plants proved as a complete failure and never came to production, because the complete disintegration of the raw lignite with a moisture content of more than 50% could not be avoided as it had been expected. Only one of these plants at Regis finally could be made a success and is still working today. The gas producers have been altered for treating briquetts. Part of the char is discharged from the producer and sold for household stoves.

By-product gasification of non caking bituminous coal has been developed from 1917 in the Saar district and in Upper Silesia, but except from some small plants connected to steel-production has not reached any importance.

1918 - 1933

The research work started during the war was continued in the following period by the concerned mining and Chemical Industry together with several engineering firms.

From the Rolle oven a vertical rotating furnace with an externally heated corrugated cylinder made from alloyed steel was developed by Geisson. This furnace could treat 60 tons of lignite per day. Several industrial plants have been erected. The residue, mostly a fine powder and highly self ignitable is used for boiler-firing in power stations.

Special experience in drying and gasification of lignite helped to develop a carbonizer for lignite briquets by the DEA at Regis. This carbonizer can treat 50 tons of briquets per day.

A similar technical development carried on by the Lurgi Company, and stimulated by interests in the treatment of lignites in outer German countries finally ended with the Lurgi carbonize for all non caking coals, lignites and briquets with a capacity of 200 to 400 tons per day. Pioneer plants of this system have been erected in Greece and England, industrial plants in Canada, U.S.A., Korea, New Zealand and Sachalin.

For a big scale development of carbonizer plants in Central Germany based on lignite the quality of the char was still a serious impediment. This difficulty has been overcome in 1932 by the Lurgi-Krupp process for drying and briquetting. This process allows to produce a clean lumpy char from powdered lignite.

For coal and oil shale various other methods of carbonization have been tried. Several types of rotary kilns have been erected in the Saar district for caking bituminous coal and in Wurttemberg for oil shale. All have proved as a failure for various reasons. The steel-cylinder did not resist long enough to heat and permanently changing tension. The solid and liquid products were not of a satisfying quality.

For the lightly caking rich oil shale of Estonia two types of tunnel-kiln have been developed and successfully operated. The Kivioli tunnel kiln has a cylindric tunnel made of steel. The Silimagi tunnel kiln is rectangular and is constructed from brick. There are some other varieties concerning the arrangement of the heaters and ventilaters and the construction of the waggons which carry the shale through the tunnel. The

daily capacity of these kilns has been brought up to 500 tons per day per unit. The products and the recovery of oil are very high.

In Estonia a gas producer for carbonization of non caking oil shale has been developed by the Pintsch Company, which has been operated successfully for many years owing to the high fusion point of the residue of the Estonian oil shale.

In England the coalite retort for bituminous coal has been developed and has found a successful but limited use. In France the Grande Paroisse furnace for oil shale was developed to an industrial stage. In Wurttemberg a first Unit of the Otto- system for oil shale was successfully operated.

Based on the invention of Berguis hydrogenation of tar and coal has been developed to an industrial scale by the J.G. at Ludwigshaven and Leuna. At the same time Ruhrchemie succeeded in erecting and operating a first technical plant working with the synthetic method known as the Fischer-Tropsch process.

1933 - 1939

Any production of motor fuel from coal and oil shale in Germany and in connection with products from natural oil had proved itself dependent on a subsidy or a tax protection against these foreign products. Such protection was granted for home products in Germany. In 1933 the German government faced with the problems of non-employed and of economical and political dependence from oil producing countries placed the production of motor fuels from coal and lignite on the first 4 years plan, together with an enforced drilling program for natural oil.

The following systems were adopted and from 1933 to 1939 plants were erected or erection started:

1.	Hydrogenation of coal and lignite. factories in Rhur district Central Germany.	Capacity Metr to year 1400000 to/y
2.	Fischer tropesch Synthesis. factories in Rhur district Eastern Germany Upper Silesia	550000 to
3.	Lurgi Carbonizers for lignite and hydrogenation of tar. Central Germany Czechoslovakya	1200000
4.	Lurgi Carbonizers for lignite and coal with fractionating and refining of tar. Central Germany Upper Silesia	500000
	Total	3650000 to/yr

1.	Hydrogenation of coal. Upper Silesia Rhine valley	850000 t
2.	Lurgi Carbonizers and Continuous vertical retorts for coal and lignite with Hydrogenation or Re- fining of tar. Upper Silesia Czeckoslovakia	700000
3.	Carbonization of oil shale with Refining of tar. Estonia Holstein Wurttemberg	500000
	Total	<u>2050000</u>

In addition smaller plants of by-product gas producers and pioneer-plants for carbonization have been erected in this period.

At the end of World War II the tendency of the future development of oil production from coal and oil shale for European conditions could be clearly recognized. A long experience under various conditions allows to compare and to classify the various methods of production as is done in Section V.

## II RAW MATERIALS AVAILABLE IN EUROPE

### A. Essential properties of solid fuels.

For a classification of solid fuels from the viewpoint of producing oils, it is necessary at first to consider the principal properties of solid fuels and their influences on the industrial or domestic use of a fuel.

#### a. Moisture content.

The contents of moisture of solid fuels varies from 1 to 60% from coal to younger lignites not considering wood and peat which only exceptionally may be used for a very limited unimportant production of oils.

Often younger lignites and subbituminous coals can be mined so cheaply, that even with 60% moisture they can compete with high grade coal as for instance in the Rhine-Ruhr district. A high moisture content can be overcome by drying, but mostly the drying process means a moderate or perfect disintegration of the fuel, and makes such fuel unsuitable for most of the methods of utilization of solid fuels. With the younger lignites this difficulty can be easily overcome by briquetting without a binder. For other wet fuels the briquetting becomes

difficult and ~~costly~~ that this fuel must be utilized near the mine and by methods suitable for handling a disintegrating material.

Production of oils by carbonization, or by Hydrogenation or by a synthetical method depending on other properties of the fuel is not hindered by a high moisture content. It there is a market for solid lumpy smokeless fuel, carbonization offers a way to produce such fuel in combination with the production of oils.

b. Ash content and fusion point of the ash.

\* A high ash content of a fuel makes this fuel unsuitable for direct Hydrogenation. It also excludes carbonization with the production of a coke for an industrial or domestic market. In general such fuel must be utilized near the mine and without longer transports of the coal or the coke.

It should be utilized by complete gasification possibly with recovery of tar, the gas to be used either for production of oils by hydrogenation of tar or by synthetical methods or for the production of power or as a gas for public distribution.

The fusion point of the ash may be deciding for the process of gasification or the system of boiler-firing to be selected. If the fusion point is extremely low it may be a very costly feature of the fuel and in some cases even exclude such fuels from the production of oils.

c. Contents of Volatiles and tar, Degree of carbonization.

High contents of volatiles in the dry material are of a great value for the production of oils. Part of the oils can be recovered by the relatively cheap method of carbonization, and the high content of hydrogen available in such fuel is profitable also for the direct hydrogenation of this fuel or its gasification into gases of a high content of hydrogen.

According to the degree of carbonization the oxygen content of a solid fuel may be rather high. Mostly this results in a high oxygen content of the tar and decreases its value by increasing the cost and decreasing the efficiency of refining. If such tar is treated by Hydrogenation its oxygen content is less important.

Anthracites if cheap enough can only be used for a synthesis of hydrocarbons.

d. Contents of Sulphur and Chlorine.

Contents of sulphur are not important for the hydrogenation of coals or of tars. A high content of sulphur in the coal sometimes results in a high sulphur content of the tar but often it does not increase the sulphur content of a tar compared with normal conditions. In this case carbonization is well suitable

to utilize such a coal. In many cases an usually high sulphur content of the raw coal, which lowered its market value considerably can be reduced by carbonization to a relatively low sulphur content in the coke, and can open even the domestic market to such a fuel. The sulphur content of the coal, which mostly goes into the Hydrogen sulfide of the distillation gas can or must be utilized for the production of sulphur. A tar with a high sulphur content in general must be treated by hydrogenation in order to get a good yield of quality products of gasoline and diesel oil. Chlorine usually causes heavy corrosions on the equipment in carbonization as well as in a hydrogenation plant.

e. Sintering and caking property of coals.

Caking and highly fusing coals can only be treated in coke ovens or other outside heated retorts. A moderate sintering quality of a coal, which normally prevents gases from passing through this coal or may disturb a continuous movement of the coal in a vertical shaft, can be overcome by preheating such coal to 200 - 300° centigrade for several hours, and this effect may be increased by a certain oxygen content of the heating gas in case of a relatively small sized coal.

Such coal and non-sintering coals can be treated and heated by a direct contact with hot gases or can be gasified in a layer of fuel under condition that the proportion of fines below 1/8" is not too high. For direct hydrogenation of a coal, its sintering properties are not of any importance.

B. The Most Important Deposits of Coal and Oil Shale of Europe.

a. Caking and bituminous coal

Large deposits of coal, from anthracite to high volatile coal, are available in Great Britain. The bituminous coals are mostly caking. There are some non-caking coals in Scotland and northern England, which can be treated by direct heating. Most of the coals, however, have so much caking property that only outside heated retorts can be used. With this method, using iron retorts and a relatively low temperature of carbonization, the development is limited to a high-price market for a high volatile coke. Such a base, however, has proved too small for a production of oils of any importance.

For the hydrogenation of coal and also for the production of hydrogen gases for synthetical purposes many sorts of coal are available.

By briquetting of bituminous fines and carbonization of these briquets by direct heating methods, the base of raw material could be considerably widened.

In the Ruhr district various kinds of bituminous coals are mined, but coals which could be treated by direct heating are not available. So the situation is similar to that of England. The raw material is suitable to be used for hydrogenation of coal and for the synthesis of hydrocarbons by gasification of semi-anthracite and coke.

In Northern France and in the Saar district mostly high volatile and swelling coals are mined, which cause difficulties in any treatment. The deposits are limited so that the mine products are easily sold as a raw coal.

Large deposits of bituminous high-volatile coals are available in Upper Silesia. Mining conditions are very good. Many of these coals are suitable for carbonization by direct heating, and such an industry has made considerable progress. The low temperature coke has proved very suitable for domestic and central heating. Furthermore, carbonization of briquets made from high volatile coal seems to offer a method for replacing the production of coke in coke-ovens, the deposits of suitable caking coal becoming slowly exhausted.

Coals suitable for hydrogenation and for gasification are available on a big scale for a long period.

#### b. Subbituminous coal and older lignite.

There are rich deposits in Central Germany, cheaply mined by open-pit mining, and with a high content of tar. They are very suitable for carbonization after briqueting. They can be hydrogenated with a relatively low pressure. For these reasons, they have been preferably used for the production of liquid fuels.

Younger subbituminous coals which can be treated with the same methods, but which need no briqueting, are mined in Czechoslovakia and to a limited extent also in Hungary and southern France.

#### c. Younger Lignites.

This kind of raw material exists in large deposits in the Rhine Valley and in Eastern Germany and can be mined cheaply. Low tar contents and a high content of oxygen make these fuels preferably adapted for gasification and the synthesis of hydrocarbons. Direct hydrogenation of such lignites requires an extremely high pressure and has proved relatively expensive in this case.

#### d. Oil shale and bituminous rock.

There is an important deposit of oil shale in Estonia with a thin (2 meter) but rich seam of shale (18 to 20% of oil). Mining conditions, hitherto mostly adapted for open cut mining, are changing slowly to underground mining. The deposit widely spreads into northern Russia, the seam there going down to 50-150 m and the oil content steadily decreasing eastwards to 12%. Oil production will probably be growing

rapid in this field.

In Estonia Tunnel Kilns have been used for rich shales and a special gas-producer for lower-grade shale.

In Russia the Tunnel Kiln system has been adopted for the rich shale and a carbonizer similar to the LURGI carbonizer but of only 80 tons capacity, has been developed for the lower grade shale.

Good oil shales with 8-12% of oil are found in France, Spain, and Scotland, but the cost of mining in these fields is relatively high owing to the depth of the deposits and the hardness of the shale. The importance of these deposits is very limited therefore.

The most important deposit of oil shale of Europe is a 10 m seam of the Lias or Jurassic formation extending from southern France along the western and northern rim of the Alps to Wurtemberg and Bavaria. This 10 m seam has an average oil content of 4.8%. Open cut mining is possible to a large extent, and even in underground mining the lower 3 meters of the seam with 6.0% oil can be mined cheaply by the slope method.

Technical treatment of such a poor material with a low melting point of the ash has proved to be a hard problem by some 30 years of experimenting in Germany as well as in France. Lately carbonization and complete gasification of the residue for industrial heating and production of power in districts where other fuels are very expensive offers a chance for an extended production of oils from this Lias oil shale.

Large deposits of oil shale with 8-10% oils of very good quality have been found in Bulgaria. As the mining can be done cheaply by open cut mining, they may be of a great value when the natural oils of the southwest of Asia become exhausted.

There is an extended deposit of bituminous rock in Holstein impregnated with 12 to 18% of a good natural oil. Mining and treatment of this material, however, has proved rather difficult until now, owing to its softening in the deposit as well as in the furnace.

~~Several smaller deposits of oil shale and bituminous rock in Italy are too low grade for an economical treatment in spite of much experimenting done in these fields.~~

### III. TECHNICAL METHODS OF AN INDUSTRIAL PRODUCTION OF LIQUID FUELS FROM SOLID MINERALS

#### A. Low Temperature Carbonization

##### A. Caking Coals.

After 20 years of experimenting with various systems on a large scale, with very costly pioneer plants, only a few



designs of the same principle - that of outside heating in narrow retorts made of steel or cast iron - have proved satisfactory, and even with these retorts in general a good quality coke can only be produced if the volatile contents of the coal are not too high.

All these retort plants are operated with fines of bituminous coal, crushed, and if necessary regulated for caking property and volatiles by mixing several sorts of coal.

The three varieties of iron retorts are the following:

The Coalite Retort, developed in England, has been used for more than 15 years in several plants of medium capacity. After struggling for many years for a market in the domestic use, this carbonization industry seems to be sound economically as far as a special price for the coke can be reached in domestic or industrial use.

The coalite retort is made of cast iron 9 to 12 feet high and has a cross section 7 to 8 inches wide and 4 feet long rounded at the ends.

A number of retorts are connected to a battery similar to a coke oven. The coke is quenched for cooling. The tar is sold mostly as a fuel oil. Production of some by-products from the tar for pharmaceutical or preserving purpose seems to be essential for the economy of the plants.

In Germany the Krupp-LURGI retort system has been developed and has been operated for many years in the Ruhr and in the Saar Districts. In this system 6 rectangular cells 6 to 8 inches wide, 7 feet high and 10 feet long, made of welded steel, are connected to one unit with heating channels between the cells. For the heating of the unit, flue gas of a regulated temperature is circulated through the heating channels by means of a fan. The unit is charged by a special car, which carries the coal required for one unit and which is equipped with a device for lifting the cover of the unit and for charging and stamping the cells.

The unit is discharged into a special car suitable for the quenching of the coke underneath a quenching tower which is erected near the screening and storage place. Cells of the K. L. retorts are 20-30 mm wider at the lower end of the cell.

In the third system of steel retorts, the B.T. retort, the walls of the rectangular cells together with the welded heating channel are suspended at the higher end so that they can be swung aside for the discharging of the retorts. This enables the use of very narrow retorts of an equal width from the top to

the bottom and allows to work with very short periods of heating, as e.g. 3 hours. But the smaller weight of a charge and the increased number of charging periods nearly equalize this advantage. Stamping of the charge is made impossible by the narrow cells and the quality of the coke is lowered by its relatively reduced density.

This system has been tried a long period of experimenting. There still may arise serious difficulties in an industrial plant with the tightening of the cells between the moving walls when these walls become corroded or twisted from permanently changing temperatures and working conditions.

The steel retorts described above can also be operated with briquettes made of a slightly fusing and high volatile coal. In this case the throughput of a unit of 6 cells may be increased from 15 tons of caking coal to 25 tons of briquetted coal, owing to a better heat transmission within the charge of the retort by the fluctuating distillation gases.

Oil products are slightly cracked, which results in a relatively low degree of viscosity. Tar recovery amounts to 60% to 70% of Fischer assay.

The maximum temperature of distillation is limited to 600° Centigrade.

Another more promising method for treating caking bituminous coal, consisting of briquetting the raw material favorably after reducing the ash content and the caking quality by mixing or by a heat treatment, is still in the stage of development. It would allow to treat such coals also by the much simpler and more efficient method of a continuous shaft carbonizer working with direct heat transmission. At the same time it would allow to produce from high volatile coal a clean, dense, equally shaped coke with a regulated content of volatiles, good even for a blast furnace.

#### b. Non-caking Coals and Briquettes

Several types of Continuous Working Vertical Retorts have been developed, from experience with gas works, for the low temperature distillation of non-caking bituminous coal. Retorts made of firebricks 20 to 25 feet high, approximately 12 to 15 inches by 12 feet of cross section, and mostly two retorts connected by a common device for charging and discharging are operated by a regulated outside heating and internal steaming or recircling of distillation gas through the charge of the retort, in order to prevent the oils from cracking.

< Pioneer plants have been built by the three German firms Otto, Didler and Koppers in Upper Silesia differing to a

certain extent in the heating system and the device of charging and discharging. In these retorts screened coals or briquettes of none or a very moderate caking quality can be treated.

The temperature of distillation can be widely regulated between 600 and 1000° Centigrade according to the required amount of volatiles to be left in the coke.

The recovery of tar is 70 to 80% of Fischer assay, depending on the amount of gas recirculated into the retort. Viscosity and the contents of asphalt in the tar also depend on this same condition as well as on the character of the treated bituminous coal.

The gas available from this system of carbonization is of a high heating value from 500 to 650 BTU/cub. ft. The heating value and the quantity of gas produced depend primarily on the distillation temperature.

Shaft carbonizers have been experimented on in several mines. Industrial use has been reached only the LURGI Carbonizer in 3 plants in Upper Silesia. These plants are operated with screened bituminous coals as well as with briquettes made with sulphide liquor as a binder. The capacity of a carbonizer unit amounts to 350 to 400 tons of coal or briquettes per day. The design of such unit is nearly the same as that of the well-known LURGI carbonizer used for lignite-briquettes and described in chapter III A.c. The drier of this carbonizer, in case of treating a softening coal, can be used for reducing the fusion of the coal and for eliminating of the sticking of the charge in the shaft. It also allows to treat wet or washed coal without reducing the throughput of the carbonizer. The temperature of distillation, being easily variable between 500 and 900° Centigrade, the amount of volatiles left in the coke varies from 10 to 2% according to consumers' demand. The recovery of tar and benzine amounts to 92% of the Fischer assay. The character of the tar is slightly more asphaltic than in case of outside heated retorts, but most of the asphalt is condensed separately by a special fractionating device.

The coke, leaving the carbonizer with about 100° Centigrade continuously, is cooled sufficiently for further transport and storage by a spray of very little water, at the same time blowing air on the surface, which evaporates the water and cools the coke to about 50° Centigrade. By carbonizing briquettes made of a coal or a mixture of coals with a moderate fusing property, a very resistant low volatile, and equally shaped coke is produced, which has proved very satisfactory for the watergas producers of the Blechhammer-Hydrogenation plant.

If operated with air for combustion, the gas has a higher value of 270 and 280 BTU per cub. foot, 32 F., 30 inch. If operated with oxygen a gas with 400 to 420 BTU per foot can be produced.

By a special method of condensing the oils from the gas, 3 fractions are recovered immediately in the condensation plant consisting of 25% pitch, 65% fuel oil, and 10% benzine. The pitch with a melting point of 150°F can be used for briquetting purpose or may be hydrogenated. Fuel oil was sold to the Navy. The benzine of a high octane number is refined and used as gasoline.

c. Lignite raw or briquetted.

For nearly one century the highly bituminous lignite of Central Germany has been treated for recovery of paraffin wax by the Rolle-oven. Several plants of this type are still working, treating special briquettes now instead of raw coal. Some have been replaced by new factories equipped with LURGI carbonizers.

The Geissen-Furnace developed as a mechanical furnace on the principles of the Rolle-oven has been used for the erection of several industrial plants connected with smaller power stations. The coke produced by this method of carbonization is a fine, highly self-ignitable powder, which can only be utilized in an annexed boiler plant equipped with pulverized coal-firing systems.

The first plants which were erected used a corrugated vertical rotating cylinder 25 feet high consisting of rings made of a special chrom alloy cast iron. This cylinder is heated inside by a gas flame. The lignite is kept on the cylinder in a layer of only one inch thickness by series of wider rings fixed at the walls of the furnace around the heated rotating cylinder and leaving outlets for the distillation gas. The lignite to be treated must be small sized, with a maximum size of 10 mm, in order to prevent it from sticking in the narrows of the retort. It cannot be treated in case of a high percentage of powder below 1 mm, or if disintegrated by a sudden heating, because in these cases the movement of the material through the retort cannot be controlled. The lignite flows like water into the gas channels. In the last factory of this system erected in 1937 a straight inner cylinder made of a welded heat resistant steel was used. Part of the dried lignite had to be briquetted and crushed again to smaller grains to meet the conditions described above.

Except for a relatively high wear and tear of the machinery which possibly could be reduced by using higher grade steel, the working system of such a factory has been proved as rather complicated owing to the necessity of a special preparing and separate drying of the raw lignite and of cooling the very fine and very ignitable char. No regulation of the degree of carbonization, that is, the contents of volatiles in the char or the quality of the oils by means of an increased temperature of distillation, can be performed with this system. The oil products are unstable and of a relatively high content of oxygen. The tar recovery is high, sometimes above 100% of the Fischer assay.

— An experimental Geissen furnace of a capacity of 100 tons per day of a coal with 30% moisture has been operated also with many varieties of subbituminous and non-caking coal near Brux in Czechoslovakia, and these coals are more favorable for such a technical treatment, but the quality of the char and that of the oils is much inferior to products of other methods, working with the same coal, for the reasons described above.

For the carbonization of lignite and subbituminous coal the LURGI-Carbonizer has been developed since 1921. Originally this carbonizer was designed for treating a raw coal of 50% moisture and equipped with a correspondingly extended drying zone. Experience showed that such wet liquids are favorably dried and briquetted before carbonization. New methods of briquetting have been developed in connection with carbonization with the purpose of producing a hard lumpy coke.

The LURGI Carbonizer is based exclusively on a direct heat transmission from hot gases to the coal. The working principle is shown on the Figure (1).

With the LURGI carbonizer all varieties of coals and lignites except fusing or caking bituminous coals have been treated commercially, either briquetted or raw, in many plants, among which several have a capacity of 5 to 10,000 tons per day and one has a throughput of 25,000 tons per day.

A LURGI Carbonizer plant is operated fully automatically. It needs very little labor. Repair costs are relatively low, owing to a very simple construction and any movable parts or steel being avoided, where these parts could be destroyed by high temperature or by the corrosive or abrasive action of gases and coke. The handling of the coke outside the carbonizer is simplified by the good cooling effect of the coke cooling zone and only a small amount of fines or dust are produced by the carbonization. The disintegration of the coal or of the briquettes, even with a very unfavorable raw material, otherwise highly disintegrating by a sudden rise of temperature, can be sufficiently avoided by the very slow increase of temperature which takes place during the 10 to 15 hours available for drying and carbonization only. — It has even been possible to produce a hard lumpy coke with such a method from wet coals. The content of volatiles of the coke can be easily regulated and quenching of the coke can be avoided.

The recovery of tar and benzine amounts to 90 to 95% as compared with the Fischer Assay. The tar is of a good quality, free of dust and water and its oxygen content favorably reduced. The tar products produced in this carbonizer plants are either refined or hydrogenated, mostly in a centrally located factory ~~which~~ for several carbonization plants which are located near the mines.

For the cooling and the inertization of the coke, as required for the domestic market, a special device has been developed.

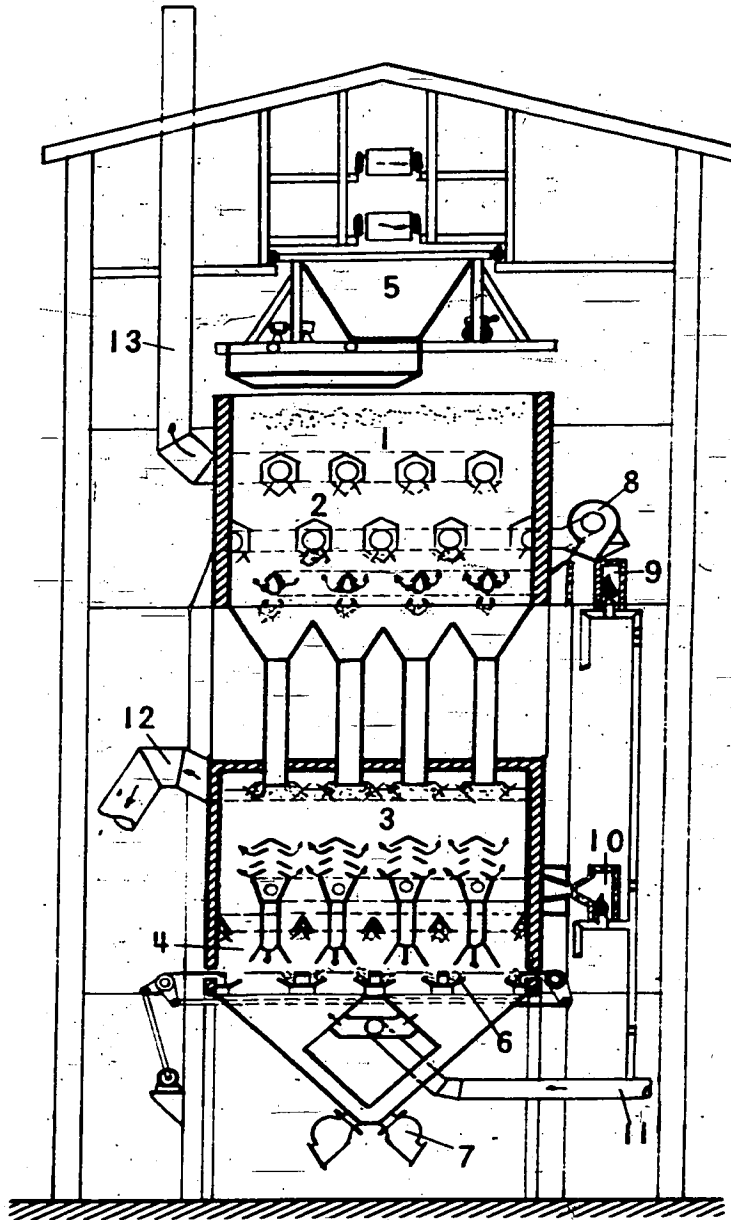


Figure 1. Elevation of a Lurgi direct-heating carbonisation plant.

- 1 Coal Hopper
- 2 Drying zone
- 3 Carbonising zone
- 4 Coke cooling zone
- 5 Coal charging car
- 6 Coke discharging mechanism
- 7 Coke discharging pockets
- 8 Circulating fan for drying zone
- 9 Combustion chamber for drying zone
- 10 Combustion chamber for carbonising zone
- 11 Admission of purified circulating gas
- 12 Offtake for mixture of circulating gas and carbonisation gas
- 13 Offtake for waste gas from drying zone

In some plants erected before the new methods of briquetting had been developed the coke is briquetted by a binder made from the tar, sometimes by oxidizing it in the tar still. These briquettes are sold for domestic purpose.

Some operating figures of a 10 unit carbonizer plant operated with a screen subbituminous coal of 12% moisture or with coal briquettes are the following

Investment Cost (without mining and briquetting)

1.	10 carbonizers incl. building and transport equipment	4,700,000 Rm
2.	Condensation plant	2,740,000
3.	Benzine-recovery plant incl. plant for distilling of scrubber oil	2,420,000
4.	Equipment for screening and cooling of coke incl. loading	800,000
5.	Storage tanks for oils	300,000
6.	Distribution system for energy steam and water	500,000
7.	Recooling system	400,000
8.	Boiler house (gas-fired) and Power station for 3000 KW, 2 x 3000 KW turbines	1,000,000
9.	Roads and tracks	200,000
10.	Laboratory, office and social buildings	400,000
		<u>13,460,000</u>
	<u>Miscellaneous</u>	<u>540,000</u>
		14,000,000
	Operating capital	<u>1,500,000</u>
		15,500,000

Operating figures for 340 workingsdays per year:

	Germany RM	U. S. A. \$
1. Total investment cost	14,000,000	8,000,000
Operating capital	1,500,000	800,000
2. Coal treated (long tons)		
$340 \times 4000 = 1,360,000$ to/year		
3. Fresh water $400 \times 8600 = 3,440,000$ m <sup>3</sup> /year		
4. Laborers and employees:		
laborers of operation, repair, social buildings, cleaning (vacation incl)	160	
Officers		
foreman	6	
engineer	3	
manager	1	
5. Production of oils (12% oil content of coal, 92% recovery)		
$1360,000 \times 0.12 \times 0.92 = 150,000$ to/year		
90% taroils	135,000	"
10% light oils	15,000	"
6. Production of coke		
50% of coal = 680,000 to/year		



Production Costs:

	Germany RM	U.S.A. \$
<u>Coal</u>		
1,360,000 to @ 12. - RM	16,320,000	
1,360,000 to @ 5. - \$		8,160,000
<u>Water</u>		
3,440,000 m <sup>3</sup> @ 0.05 RM	172,000	
3,440,000 m <sup>3</sup> @ 3¢		103,000
<u>Wages</u>		
Laborers 160 @ 3000 RM	480,000	
160 @ 2000 \$		320,000
Officers	70,000	35,000
<u>Repair and material for operation</u>		
2.5% of 14,000,000 - RM	350,000	
2.5% of 8,000,000 - \$		200,000
<u>Amortization cost</u>		
8% of 14,000,000 - RM	1,120,000	
8% of 8,000,000 - \$		640,000
<u>Interest</u>		
3% of 15,500,000 - RM	465,000	
3% of 8,800,000 - \$		264,000
<u>Taxes and Administration</u>		
	500,000	250,000
TOTAL PRODUCTION COST PER YEAR	19,477,000	9,972,000
<u>Income for coke</u>		
680,000 to @ 18. - RM	12,240,000	
680,000 to @ 9. - \$		6,130,000
TOTAL PRODUCTION COST FOR OILS	7,237,000	3,842,000
<u>Production cost per ton of oil</u>	48.2 RM	25.6 \$

## A. Oil Shale and Bituminous Rock.

1. The Pumpherstone retort developed in Scotland consists of an outside heated vertical cylinder made of fire brick. Usually 4 retorts are connected to a unit with a common heating device. Five to ten units are connected to a battery. The retorts are heated by the gas produced from the shale. The capacity of a unit of four retorts, if operated with sufficient steam and some air introduced into the retorts, amounts to 30 to 35 tons of a screened shale per day. It may be reduced to 25 tons or less if the proportion of fines below 1/4 inch is high.

The recovery of tar is from 85 to 90% of the Fischer assay. The products are of a good quality. The investment cost of a Pumpherstone plant is relatively high, but cost of operation and repair are rather low.

The Pumpherstone retort is not suitable for a softening shale or for a shale with a low fusion point of the residue.

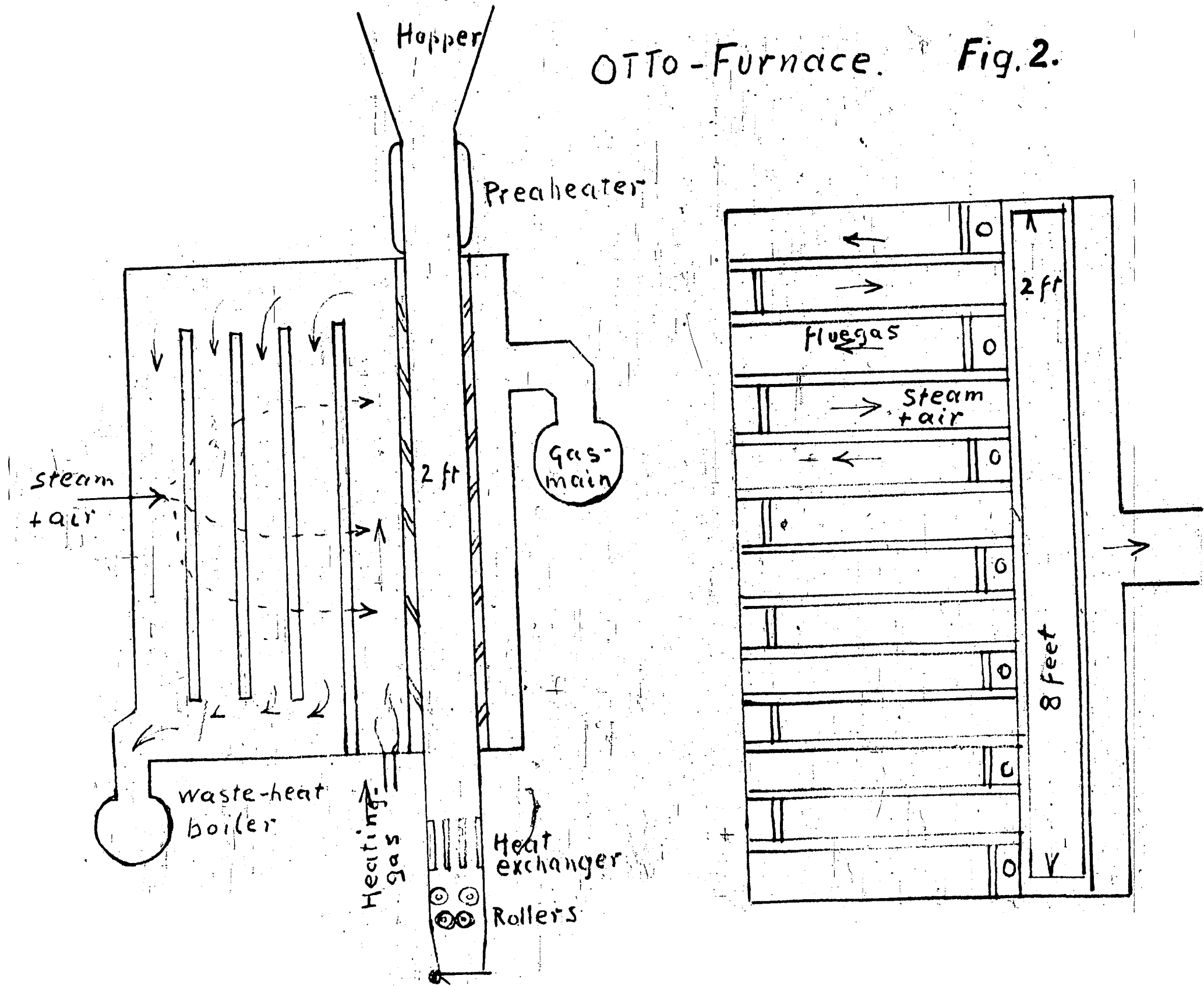
2. The Messel retort, a special design for an oilshale with a high content of carbon, combines carbonization of the shale with a complete gasification of the residue by means of steam in an outside heated retort 2 feet by 4 1/2 inches of cross section. A group of retorts is connected to one unit. The upper part of the retorts is made of steel, the lower part of one-piece fire brick tubes. Erection and operating cost of such a plant are naturally high. The dependence of the production from purchased fuel or energy is an important feature of the Messel method and allows to keep the plant working with a small profit until the very limited deposit of shale becomes exhausted. 0.5 m<sup>3</sup> of gas with a higher heating value of 3000 kcal/m<sup>3</sup> is available from a ton of shale.

3. The Goldfield rotary kiln or Davidson retort, developed in Estonia by an English group, has been working for many years in an 8 unit plant. These units, of a diameter of 4 feet and a length of 75 feet, have a throughput of only 20 tons of oil shale per day. An experimental unit of greater size provided for 75 tons capacity has been proved a failure.

The kiln is operated with high grade shale of 0 to 4 inch sizes. Scrapers are mounted inside the kiln in order to prevent the shale from sticking to the wall, which is favored by breaking a wet and fine material. The residue of carbonization discharged automatically by a double door, drops on a grate where it is burned for the heating of the kiln with the hot flue-gas. The kiln must be stopped every 6 months for a thorough cleaning. If operated with a relatively fine material this period may be reduced to 3 months.

The oil recovery amounts to 92% of the Fischer assay. The oils are of a very good quality with a high percentage of light oils (22%).

OTTO-Furnace. Fig. 2.



For oil shales with less than 20% of oil the Goldfields kiln must be considered as too expensive for the investment cost as well as for the cost of production.

4. The Otto-Furnace for oilshale has been erected in a pioneer plant in Wurtemberg. It has been operated with the low grade jurass oilshale for several years. In the Otto-furnace the shale is distilled and partly gasified in a vertical shaft of fire brick by means of superheated steam. A cross section of the furnace is shown in Figure 2. Oil shale 1/4 to 2 inch size stored in a hopper above the furnace passes through an iron pre-heater, outside heated by flue gas, into the carbonizer shaft. The brick walls of this shaft, 2 feet by 8 feet wide and 30 feet high, are perforated for the introduction of the steam and the outlet of steam and gas. The lowest part of the shaft is made of iron and is water cooled; part of the steam is produced here by cooling the residue from 600 to 300 degrees centigrade. The column of shale is supported by 4 rollers which are used to control the throughput. Annexed to the shaft is a heat-exchanger also made of fire-bricks, which superheats the steam to about 900 degrees centigrade before entering the shale. Hot combustion gas, passing along one wall of the shaft leaves the heat exchanger with 550 degrees centigrade. It is further used for the production of the steam required for the process.

Later on the process has been improved by introducing 100-150 m<sup>3</sup> of air into the chamber together with the steam. In this way the heat economy of the process has been considerably improved and more combustible gas is available from gasification of the fixed carbon of the shale.

The capacity of a unit with one shaft is 50-60 tons of shale per day. The recovery of oil is near to 100% of the Fischer assay, and the oils are of a good quality.

The investment cost is relatively high owing to the small capacity of the unit and the complicated system of heat exchangers. It might be considerably reduced by introducing air into the chamber, but with a low fusion point of the ash a considerable quantity of steam still will be required in order to avoid any fusion of the ash in the shaft. The utilization of steam for carbonization and gasification requires additional heat for the process and may raise a special problem where water for cooling purpose is not available sufficiently in summertime.

5. The furnace of the Grande Paroisse Company has been developed in France also with a low grade oil shale. This system is similar to that of the Otto furnace, the construction differing considerably, however, by subdividing the shaft into a drying, a carbonizing, and a gasifying zone and using exclusively steel for the walls of the shaft and of the heating device for the steam. After operating a pioneer plant of 100 tons capacity for several years, a unit of 300 tons per day has been erected in France. Results are not available. It seems that the system lately has been somewhat modified for a complete gasification.

The recovery of such a system should be high between 96 and 100% of the Fischer assay. The heat efficiency cannot be high and cost of plant and repair must be relatively high according to experience with similar systems.

## 6. TUNNEL KILNS for Carbonization of Oil Shale.

For the carbonization of bituminous oil shale direct heating of the shale by flush gas has been found very satisfactory for a good recovery of oil and a good quality of the Oils. The simplest equipment for a direct heating of solid material is a vertical shaft through which the material is conducted continuously. Such process, however, requires a material, which on no account sticks together or tightens the layer of material up against the flue of gas. Some rich oil shales as those of Estonia are softening under heat and may cake together to a solid rock under the pressure of a higher layer of material. A danger of this kind may exist temporarily even if the medium product of the mine does not stick together, because with a relatively thin seam of changing quality the temporary delivery of rich oil shale cannot easily be avoided.

The danger of sticking together can be reduced by not moving the material during carbonization and by reducing the thickness of the layer. Practically this can be performed in a tunnel kiln by means of flat wagons, on which the shale is placed in layers of 1 to 4 inch thickness, and which are moved through the tunnel. Volumes of hot gases are circulated through the layer of the material by means of numerous fans and gas heaters which are mounted along the tunnel. Even with the applications of this method, caking of the shale cannot be always prevented, but the operation of the plant or its efficiency cannot seriously be influenced. Though the plant and the process are rather complicated, this method has proved satisfactory for Estonian oil shale. Two types of furnaces have been developed:

A. The Esti-Kivioeli tunnel kiln

B. The Sillamaegi tunnel kiln

### A. Esti-Kivioeli Tunnel Kiln

This system is working with a horizontal cylindrical tunnel made of sheet iron. The tunnel is divided into 12 to 14 carbonization sections which are separated by a slide door from the feeding zone at one side and by another slide door from the quenching and discharging zone at the opposite side of the furnace. Two other doors close the tunnel at both ends. The heaters, one for each wagon are placed in a parallel brick-made channel. They are heated by the flue gases which originate from the combustion of oil shale. A number of fans for the circulation of the heating gas through the shale and the heater are placed parallel to the heater channel.

In the latest design of this type developed by the LURGI-Company the heaters are gas fired the heating temperature being regulated by recirculating gases, enlarged circulating fans are placed under the heaters, and each heater with its circulating fan

can be shut off for cleaning without stopping the operation of the tunnel unit.

The wagons are moved periodically. They are connected to the heater at the bottom of the tunnel by two flanges pressed together by the weight of the wagon. The positions of the wagons are adjusted automatically by a corresponding notch of the rails.

The slide gates are operated by a hydraulic gear, which serves also for the moving of the wagons. The supervision and steering device is located at a control room which may be common to several tunnel units.

The heaters consist of vertical tubes made of heat resistant steel, the heating gas passing outside the tubes, the distillation gas inside. The tubes are connected to rectangularly arranged groups. The first 2 or 3 wagons, which have entered the tunnel are operated with lower temperatures for drying only, which has proved very essential for decreased caking and a good output of oils.

#### B. Sillamaegi Tunnel Kiln

This system which was in operation at the Sillamaegi plant, uses a rectangular tunnel made of sheet iron and placed into a brick channel. This tunnel is of a larger cross section than the Kivioeli tunnel and uses larger wagons operated with a layer of shale only 1 to 2 feet thick. The wagons are closely touching each other with their frontwalls, they are tightened to the bottom of the kiln by the lower end of the sidewalls moving in grooves. Some differences of construction consist in the heating method and the drive of the wagons by an electric gear. The heaters are mounted above the tunnel. In both plants the shale is crushed and screened to 2 or 3 different sizes which can be treated in the same kiln. The thickness of the layer in the wagon in this case depends on the different sizes of the shale or the resistance of the layer against the flow of the gases.

Fines of shale cannot be treated in these tunnel kilns. The fines, which sometimes are plastic or can be made plastic by an addition of some water are formed to irregular balls by passing a rotating drum or by briquetting. They are dried after which the balls are hard enough for the carbonization in the same way as the coarse shale.

The capacity of the Kivioeli kiln has been increased to a daily capacity of 400 to 500 metric tons, whereas the Sillamaegi kiln treats 500 to 550 metric tons per day. The efficiency of the oil extraction utilizing the tunnel kiln, is 92 to 98% compared with the Fischer Assay.

The quality of the oils is very good containing a high percentage of light oil due to a slight cracking of the oil vapors in the heaters.

Operation of the tunnel kilns must be stopped and the kiln cooled down every 3 to 5 weeks for a thoroughly and costly

cleaning of heaters and pipes, so that the working days are reduced to 250 - 300 days per year. In the tunnel kiln very little permanent gas is developed from oil shale. The condensation of the oils and recovery of light oils requires a relatively very small installation. The recycled gas which enters the shale is heated to not more than 500 - 520 C. The oil vapors being slightly cracked during their circulation through the heater the tunnel process furnishes oils with a very high content of light oils.

The necessity of frequent cleaning under unfavorable working conditions and the periodical standstill of a big part of the plant makes the tunnel-system unsuitable for smaller plants and for a high labor standard. High cost of repair and labor makes its operation too expensive in most countries, and for any shale with less than 20% of oil content. For Estonian oil shale and the labor conditions of this country it has been proved as most successful for many years. Some figures of the production and operation of the Kiviõli plant are the following:

Oil shale charged (10-80 mm size)

Moisture:	7.5%	Fischer-Assay	500 C.
<u>Analysis based on dry shale:</u>		Oils	22.9%
CO <sub>2</sub>	18.8%	H <sub>2</sub> O	1.5%
Ash	46.8%	Residue	69.0%
Organic substance	34.4%	Gas	6.6%

figures are taken from the report of a thorough examination of the 4 kiln plant Kiviõli practiced in August 1938 by LURGI engineers.

The organic substance of Estonian oil shale is relatively high in oxygen and nitrogen, which determines the special characteristics of Estonian oil products.

A mixture of all the condensates made in proportion to production figures, showed the following analysis:

Oils

I. Gravity at 15°C.	0.946
H <sub>2</sub> O	1.1%
dust (unsoluble)	0.10%
red resins	8.6%
asphalts	0.5%
ash	0.02%
phenols	16.3%

Elementary analysis:

C	83.2%
H	10.7%
S	0.8%
O	5.1%
N	0.2%

II. Distillation test (Engler)

Boiling point 60° C,

to 100° C.	7%	total	7%
100-150°	5%		12%
150-185°	5.5%		17.5%
185-250°	7.8%		25.3%
250-300°	9.2%		34.5%
300-325°	4.6%		39.1%

III. Fractionating test

a. benzine - 185° C.	17.8% weight
b. gas oil 185-325° C	25.3% "
c. residue	56.2% "
Lost	0.7% "

IV. Fractions

a. Benzine	gravity 15° C.	0.743
	sulfur	0.8%
	phenols	0.8%

Distillation (Engler):

boiling point	43° C.	
-75° C.		19.5% total 19.5%
75-80° C.	4.5%	24.0%
80-100°	19.0%	43.0%



100-150°	39.0%	82.0%
150-185°	10.0%	92.0%
End point 201°		95.0%

b. Gas oil	gravity 15° C.	0.903
	sulfur	1.2%
	phenols	12.9%

Distillation (Engler);

boiling point	144° C.		
- 185° C.	6.0%	Vol. total	6.0%
185-200° C.	8.0%	" "	14.0%
200-250° C.	31.5%	" "	45.5%
250-300° C.	32.5%	" "	78.0%
300-350° C.	11.5%	" "	89.5%
End point 374° C.			95.0%

c. Residue (from 1)	
gravity 15° C.	1.051
paraffin wax	0.0%
red resins	12.0%
asphalts	1.4%

Distillation under vacuum (4 mm Hg absolute)

to 200° C.	7.0%	total	7.0% W.
200-225° C.	12.1%		19.1% W.
225-250° C.	12.1%		31.2% W.
250-275° C.	11.4%		42.6% W.
275-300° C.	11.3%		53.9% W.
300-325° C.	11.2%		65.1% W.
325-350° C.	11.8%		76.9% W.
residue (pitch)	19.6%		96.5% W.
losses	3.5%		

Analysis:

C = 82.8%

H = 9.7%

S = 0.8%

O = 6.4%

N = 0.3%

H.H.V. = 9,335 Cal/kg

L.H.V. = 8,873 Cal/kg

Distillation gas

Analysis: CO<sub>2</sub> = 23.2%

H<sub>2</sub>S + RSH = 10.0%

C<sub>n</sub>H<sub>m</sub> = 14.1%

O<sub>2</sub> = 0.0%

CO = 0.0%

H<sub>2</sub> = 97.2%

C<sub>n</sub>H<sub>2n+2</sub> = 41.0%

N<sub>2</sub> = 2.5%

H.H.V. = 7910 Cal/nm<sup>3</sup>

L.H.V. = 7156 Cal/nm<sup>3</sup>

Products on dry oil shale:

oils 22.27%

water 1.50%

gas 3.35%

coke 72.88%

Products of the plant for 1939 were delivered as follows:

gasoline to Estonian market 11,480t = 16.34%

fuel oil to German Navy 53,384t = 76.4%

various oils for local market 3,067t = 4.3%

Loss of refining 2,071t = 2.96%

70,002t

#### d. Economy of Tunnel Kiln System

For a plant of modern design, as shipped to Estonia in 1943-44, the cost of erections of a 900,000 ton per year plant ready for operation are the following:

	RM
1. Storage for 5,000 tons of raw shale with conveyers	800,000
2. Crusher, plant with screening machinery bins and building	1,000,000
3. Plant for treating fines of shale by agglomeration and drying	900,000
4. Carbonizer plant consisting of 6 kilns and transport equipment	6,100,000
5. Condensation plant with oil scrubbers and plant for distillation of Washoil	800,000
6. Top distilling of crude oil	500,000
7. Electric equipment	500,000
8. Piping for water & steam & recooling tower	200,000
9. Building for 1-8	1,400,000
10. Storage tanks for oils	300,000
11. Delivery and distributing of water, steam, energy	650,000
12. Rails and streets	500,000
13. Office building, Laboratory repair shop, social buildings	600,000
14. Power station gas heated 15,000 KW	3,000,000
15. Miscellaneous	650,000
	18,000,000

This layout provides that sufficient gas is available from another plant working with complete gasification of shale. Otherwise the power station had to be fired with oil shale, or power must be bought from outside (waterpower) and the heating of the carbonizers would require an additional gas producer plant.

The following calculation therefore shows only the influence of investment cost, labor and repair under most favorable conditions, and allows to compare this system with others.

e. Operating figures and economy of plant operated with an oil shale with 21.6% tar based on Fischer Assay and 96% recovery.

1. Capacity and products.

Oil shale treated per year	900,000 t per year
Residue with 10% moisture	734,000 t per year
Oils: Crude-oil 96%	186,000 t per year
fuel oil 78% of crude oil	145,000 t per year
benzine 22% of crude oil	41,000 t per year
gas: 22,5 cbm t (7,156 kcal per cbm) used for heating	
20,200,000 m <sup>3</sup> /year	

2. Energy, steam and cooling water

a. Electric energy

60 KWh per ton of shale 54,000,000 KWh per year

b. Fresh water

0.50 m<sup>3</sup> per ton of shale 450,000 m<sup>3</sup>/year

for power 300,000 "

c. Steam

0.4 per ton of shale 360,000 "

d. Additional heat required for kilns

320,000 cal/ton 288 Bill. Cal/year

for power and steam 180 Bill. Cal/year

3. Labor included for local repair work

480 unlearned laborers

70 learned laborers

20 for power station

40 for office, laboratory social rooms

Total 610

6 foremen

3 engineers

1 manager

4. Investment - capital

RM

18,000,000

USA \$

10,000,000

working capital

1,500,000

500,000

5. Production-cost

	Germany RM	USA \$
Oil shale, (including dump of residue)		
900,000 tons x 5.00 RM	4,500,000	
900,000 tons x 3.00\$		2,700,000
Water		
750,000 tons @ 0.05 RM	37,500	
750,000 tons @ 0.03 \$		22,500
Wages for Labor		
610 @ 3,000 RM	1,830,000	
610 @ 2,000 \$		1,220,000
Engineers	70,000	35,000
Repair and operating material		
2.5% of 18,000,000 RM	450,000	
2.5% of 10,000,000 \$		250,000
Amortization cost		
8% of 18,000,000 RM	1,440,000	
8% of 10,000,000 \$		800,000
Interest		
3% of 19,500,000 RM	585,000	
3% of 10,500,000 \$		315,000
Taxes and Administration	550,000	300,000
total cost per year	RM 9,462,500	\$5,642,500

Production cost per ton of oils:

9,462,500  
186,000

51.00 RM/tons

5,642,500  
186,000

30.4 \$ per ton

Cost of gas for heating of kiln and boilers is not included

Operating cost per ton of oil shale:

$$\frac{9,462,500 - 4,500,000}{900,000} = \underline{5.51 \text{ RM/ton}}$$

$$\frac{5,642,500 - 2,700,000}{900,000} = \underline{3.27 \text{ \$/ton}}$$

Operating cost per ton of oil;

$$\frac{900,000}{186,000} \times 5.57 = \underline{26.7 \text{ RM/ton}}$$

$$\frac{900,000}{186,000} \times 3.27 = \underline{15.8 \text{ \$/ton}}$$

The operating cost per ton of shale is extremely high owing to high investment and high labor cost.

Payment for heating gas would increase this cost accordingly.

7. The Pintsch producer system for carbonization and complete gasification of oil shale was developed in Estonia from an experience with by-product producers for lignite briquettes.

With oil shale the amount of gas produced by gasification of the fixed carbon is too low for a controlled heating of a carbonization zone, situated above the gasifying zone. In order to increase the heat transmission and the quantity of gas, which is introduced into the carbonizing zone, part of the cleaned gas is recirculated into the carbonizing zone after mixing it with the hot gas produced in the gasification zone.

The Pintsch-system is shown in Figure (3).

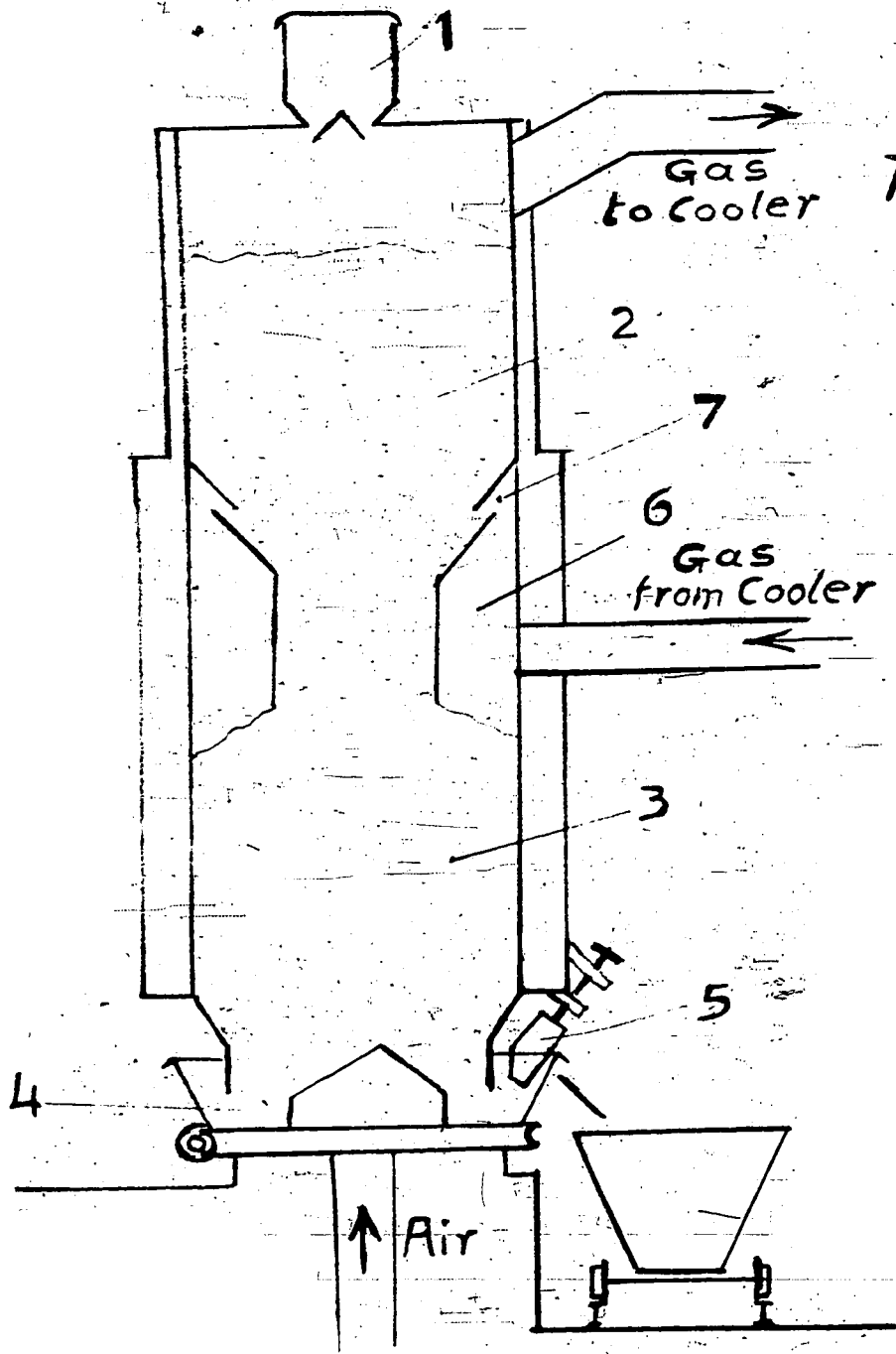
The distilling gas is introduced into the carbonizing zone with a temperature of 550 to 600 degrees centigrade. The residue of carbonization is gasified with air and relatively much steam in the lower part of the producer, which is provided with the usual device of a water-seal rotated for the extraction of the ash by means of a usual stationary shovel.

A plant of this type was operated for many years in Estonia. The results were satisfactory with a producer of 8 feet of inner diameter, which is considerably reduced between the carbonizing and the gasifying zone for a better distribution of the gas.

Fines below 3/4 inch cannot be treated. Very rich or fusing shales cannot be treated without serious difficulties in operation. The gasification zone needs frequent stirring, which requires heavy and costly labor. The great amount of gas recovered from the shale, however, has proved an important advantage of this system where coal is not available and all the heat, steam, and energy required for the mining and the treatment of

Fig. 3.

Pintsch-producer  
for Oilshale



- 1 Feeding hopper
- 2 Carbonizing zone
- 3 Gasproducer
- 4 Ash-pan
- 5 Ash-shovel
- 6 Mixing chamber
- 7 Gas inlets

the shale and the oils must be procured from oil shale. An equal carbonization requires 1100 m<sup>3</sup> of gas per ton of shale, 700 m<sup>3</sup> coming from the gasification and 400 m<sup>3</sup> being recirculated.

With lower grade shales the recovery of benzene from a relatively high quantity of gas becomes difficult and costly with this system. Variations of design with the purpose of higher capacity and simpler construction, for example, an enlarged diameter, not reduced between carbonizing and gasifying zone, have proved a failure.

The capacity of an 8-foot producer is 35 to 40 tons of shale per day. The recovery of oil amounts to 80% of the Fischer assay.

For shales with a low fusion point of the ash and for a high labor standard this system of a gas producer becomes very expensive in operation.

8. LURGI-Carbonizer with Gasification Zone. The LURGI-Carbonizer has recently been developed for a carbonization and complete gasification of high-ash fuels. A description of this device is given in chapter III. B.

According to large scale experimenting with oil shale and the experience in carbonizing coal, this method is very efficient to treat higher grade shales with more than 10% oil and a corresponding amount of fixed carbon. With an oil recovery of 90 to 95% sufficient gas is produced to operate the carbonizers and to produce the energy and steam required for the plant.

A plant of this system with 8 Units of 2800 tons capacity is under construction in Spain.

9. The LURGI-Schweitzer system of carbonizing oil shale has been developed by the LURGI Company together with the Wurtemberg Government according to proposals of Dr. Schweitzer. It is based on the idea that low-grade shale as that of Wurtemberg can be treated economically only with a very simple method, utilizing the fixed carbon of the shale for heating. This could be realized with a combustion zone operated with air, the combustion gas entering immediately in a carbonization zone. Former experiments had proved that this process could not be realized by operating from below upwards because a condensation of steam and oil vapors could not be avoided, but it could be realized by leading the gas downwards. In this case condensate flow downwards with the gases and may be evaporated again without being cracked into gas and without disturbing the fire. The danger of heavy clinkering and sticking to the wall has been overcome by using a shaft made of a relatively thick sheet-iron which is not lined with brick nor insulated outside, and which carries the heat quickly away from the relatively low combustion zone, so that a maximum temperature of the wall of 600 Centigrade is not exceeded, the maximum point permanently moving downward. An additional cooling of the combustion zone can be performed in such an open cylinder by spraying waste water on



the surface of the shale, part of this water is evaporized and passes the combustion zone together with the air for combustion.

For the operation of this process cylinders of a diameter of 10 feet and 25 feet high are used, which are slightly wider at the top end. The shale is supported by a grate at the lower end and the cylinder, which is open on both sides during operation, is connected to the basement by a water seal. A group of cylinders are connected to a common main under the floor and to a common device for gas cleaning and benzine recovery. For filling and discharging the cylinders are carried to a filling station by means of a crane. The cylinder containing 42 tons of shale in a layer of 20 feet has a capacity of 35 to 50 tons per day according to the speed of combustion which must be lowered to 1/2 foot per hour with a poor shale of a very low fusion point of the ash, as in the case of Lias oil shale, and can be increased to one foot per hour with a higher grade shale of 3 inch maximum size. 10 to 30 cylinders are operated as a continuously working unit. The exhauster of this unit sucks the combustion air into the cylinders and carries the gas, which contains the oil vapors, through the electrical precipitator, the coolers, and the oil scrubber to a boiler plant, where the steam for the benzine recovery and for the production of energy is produced.

For the discharging, the cylinders are turned over by a special device, and filled at the same place by a special spreader. This procures an equal distribution of the shale in the cylinder without a separation of the different size and prevents an unequal streaming of the air or the gas through the shale.

By this means a treatment of the unscreened shale is made possible, if the shale is crushed to 3 inch maximum size. Only part of the fines is taken out by a rough screening before crushing, and these fines are filled on top of the layer of shale, to help a quick and equal ignition of the charge.

With the low grade Lias oil shale of only 4.8% oil content 85 to 90% of the oils have been recovered. 600 mm<sup>3</sup> of gas of a heating value of 500 to 600 kcal/mm<sup>3</sup> are produced per ton of shale. An addition of 2 or 3% of carbon has proved very favorable for better working conditions and an increased heating value of the gas.

With various oil shales of more than 10% oil content, 95 to 100% recovery of oils, and a heating value of the gas of 700 to 1100 kcal/mm<sup>3</sup> have been reached.

The necessity of a concrete building for the crane and for the pipes underneath the cylinders, which make a big proportion of this design of the process, could be avoided by a later design, in which rectangular containers 10 by 14 feet square and 10 feet high are placed on a rotating platform which carries the containers from a station, where they are filled and

when the charge is ignited, to a discharging station where they are discharged. The carbonizing takes place during the slow turning of the platform. This design needs no building and no crane. The charge can be ignited with gas and it is possible to bring part of the gas back to the containers periodically. This means avoids a loss of oils during the ignition and increases the transfer of heat from the residue to the carbonizing zone.

Recently this operating system could be simplified in the continuous working HUBOVEN described later on, in which only the layer of shale is moved on a moderately inclined grate.

Experience with the operating of the LURGI-Schweitzer experimental plant caused thorough investigation on the possibility of treating the oil shale by an Underground Carbonization in the chamber prepared in the seam and filled with shale. In this case only 40% of the shale had to be brought to the surface. The chambers could be prepared by a suitable method of shooting without any transport of 60% of the shale. More than 3 years of experimenting in horizontal chambers, for 2 years on a big scale, with a mining method proposed by Carp and operating in the 3 m seam of 65% oil content have proved, that with such a low grade material the recovery of oils cannot be brought to more than 45% of the Fischer assay, even with very careful work and with a relatively high consumption of explosives. Possibly conditions are more favorable with a higher grade shale. The closing of the chamber and the remounting of the pipelines after each run cause so much additional cost that there is very little difference per ton of shale treated, compared with the cost of one ton of shale brought to the furnace. The cost of production per ton of oil has been proved to be considerably higher than with a furnace of good design. (Production cost 300-400 Rm/ton in 1945).

The oils are of a very good quality owing to a repeated distillation of the oils within the layer of shale. Condensation and a repeated distillation of the oils could be reduced in a vertical chamber operated with a vertical draft of the gases. In this case at least 30% oil recovery can be expected with a shale of more than 12% oil content, which breaks sufficiently by shooting. The height of the chamber should not be less than 30 feet.

Another primitive method, the Pile-method, has been proposed by Dr. Sennwald. In this method the shale is placed on level ground in a pile 3 to 4 m high and 12 to 50 m square, under the pile a system of perforated conical pipes has been mounted before, which allows to suck air through the pile after the surface has been ignited by means of an additional easily ignitable fuel like peat. The gases withdrawn from the pile are treated by the usual condensing equipment where the oils are separated. After a sucking period of 18 hours the carbonization is finished. The pipes are drawn out of the pile and mounted on another place where a new pile is erected.

Experimental work in small piles has given promising

residuals and an oil recovery of 80%. In piles of greater dimensions considerable difficulties arose, which could not be overcome until now. Probably all of the ten piler plants, erected before the end of the war, meanwhile have been stopped and the process given up definitely.

1. The pile could not be operated sufficiently with a non-crushed and non-screened material. It was necessary to crush the shale to 4-inch maximum size and to cover the surface carefully with smaller sized shale.

2. For an equal ignition of the pile a relatively great quantity of peat was required.

3. Weather conditions (snow, rain, storm) often have rendered the preparing of the pile and its ignition very difficult and have prevented an equal burning.

4. The withdrawal of the pipes, which often were damaged or bent by the fire, caused considerable difficulty and considerable cost for repair and remounting.

5. The operating cost per ton of shale could not be lowered compared with a furnace plant.

6. Even with a well prepared pile and favorable weather conditions the oil recovery was not more than 45% of the Fischer assay and the oils are of a poor quality.

7. The quality of the gas was only  $300 \text{ kcal/m}^3$  (35 BTU (cb. ft.)), so it proved very difficult to burn this gas in a furnace. Combustion is absolutely necessary in a big plant because the contents of  $\text{H}_2\text{S}$  makes this gas rather poisonous.

For Carbonization and GasificationofHigh-Ash-Fuels

Oil shales and other high-ash-fuels like Cannel-coals and lower grade products of coal-mining are characterized as solid fuels of a low content of fixed carbon and a sometimes extremely high ratio of ash to carbon. An industrial utilization of such fuels naturally has to combine carbonization with gasification or combustion of the fixed carbon, because otherwise the latter could not be utilized, and the process of carbonization would require an additional fuel resulting in additional production costs. Due to the unfavorable relation of ash to carbon the combustion or the gasification of the carbon by air or oxygen generally becomes very difficult, unless the ash has an extremely high melting-point.

For this reason gasification of oil shales has been tried in so many pilot-plants in spite of the many failures, which had been experienced before. Only in one case a limited success has been reached by the normal method of introducing air continuously into a fuel bed from below. In this case the ash contains so much chalk, that its melting point is extremely high. (Estonian Oil shale).

In general heavy clinkering could not be avoided in gasifying fuels of a high ash content. The cause of these difficulties lies in the fact that a small quantity of air is extremely superheated by passing a relatively very great quantity of hot ash before it meets the fixed carbon for combustion. Usually applied agents suitable for reduction of clinkering as steaming or a very reduced combustion rate have proved too expensive. Another disadvantage in utilizing the simple method of carbonizing by a direct heating with the gases which are formed by the combustion of the fixed carbon of the fuel is that the quantity of those gases is too small to carry sufficient heat from the combustion or gasifying zone into the carbonizing zone. Cracking of hydrocarbons or their condensation and oxidation within the fuel bed is the result of this method.

In winter time, or if carbonizing a moist fuel, the condensation within the fuel bed may be so heavy that even combustion of the fixed carbon can seriously be hampered. By preheating the material and recirculating gases into the fuel bed between the combustion and the carbonizing zones, these difficulties can be reduced.

The experience made from the condensation of water and oils within the upper zones of the fuel bed have encouraged inventors to reverse the normal flow of the gases as employed by the NTU Retort and the Lurgi-Schweitzer-System. In this case condensation of liquids is reduced and a reflux of liquids into the combustion zone is prevented. But such a process can only be realized by an intermittent working process, because it is impossible to move the fuel in a shaft from the bottom to its top.

This experience shows clearly that for the treatment of bituminous high-ash material a continuous gasification process is desirable which works with a gas flow from top to bottom and in which a continuous discharge of a clinkering ash can be realized. Preheating or drying of the material, if possible, should be helpful to control carbonization and gasification. The gas should have a calorific value of more than 600 calories/ $m^3$  as its combustion cannot be avoided ( $H_2S$  content).

The Lurgi-Huboven is a device which is suitable for the above mentioned requirement. Details of the process and of its machinery have been successfully employed in many carbonization plants or in big scale experiments for the carbonization of low grade Liás-oil shale of Wurttemberg.

The process is shown on a schematic ~~drawing No. 1~~ **FIG 4** and works as follows:

The oil shale crushed to maximum size of 4 inches is carried to the bunker which in its lower part is provided with grates and roofs for circulating hot gases through the fuel. When leaving the bunker the fuel is dry and can be preheated to  $200^\circ$  centigrade. It descends through a narrow shaft into an inclined chamber which is open at the top and closed at the sides by parallel walls. An inclined grate consisting of groups of parallel iron girders such as I or rails are arranged as a support for the fuel at the bottom of the chamber. The grate can be partly moved by means of a gear. A gas collecting chamber is provided underneath the grate. The chamber is connected at its lower end with a fixed grate and is open at the end in order to discharge the ash. A separate gas collecting chamber for hot gas is arranged underneath the non-movable grate. Chambers can be provided to both sides of the bunker. A steady movement of the fuel bed is accomplished and controlled by moving part of the grate up and down for one or two inches.

Drying, carbonization and gasification of the fuel is performed by burning its fixed carbon with air and recovery of the heat which is accumulated by the hot ash. The heat transmission is carried out by circulating gases through the fuel bed. Air for combustion is sucked into the fuel by a slight vacuum which is maintained underneath the grate by exhaust

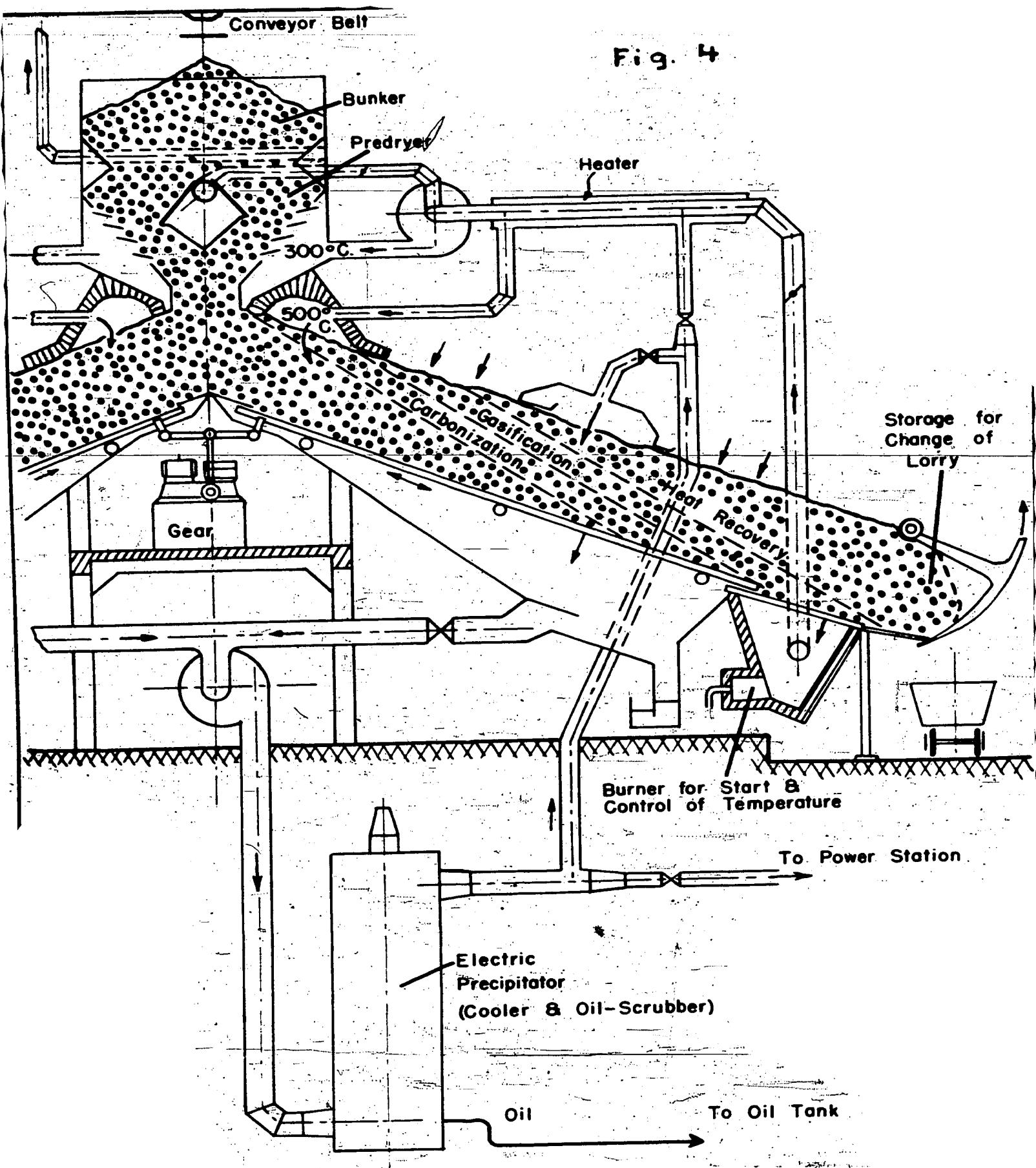


Fig. 4

**PLAN OF A HUBOVEN**  
For Gasification of Highash Fuels

fan for the carbonization gas and at the lower end by the exhaust fan of the drying zone. This fan sucks combustion gas from the lower gas chamber into the drying zone and circulates the gas with a controlled temperature through the hopper. The surplus leaves through the stack.

The oxygen content and the temperature of the gas in the lower gas chamber can be regulated by an introduction of additional air or combustible gas by a burner which is fixed to this chamber. The burner is also used for the start of the plant. In order to avoid combustion of oil at the entrance of the dried fuel into the chamber hot gas free from oxygen is used for carbonizing the upmost layer of the fuel bed. For this purpose part of the clean gas is reconducted after being heated to 500° centigrade in a heater which is heated by the hot gases flowing to the dryer. In order to increase the heat transmission from the combustion zone to the carbonizing zone if a low-grade oil shale with less than 8% of fixed carbon is carbonized oil free gas can also be recircled into the middle section of the chamber.

The residue of gasification is discharged directly into iron cars for further transport to a dump by a locomotive. The gas which is sucked from the grate by the exhaust fan is conducted to an electrical precipitator to a cooler and to a scrubber where tar oils and light oils are extracted from the gas. Part of the gas returns to the plant and the rest is available for heating purposes or for the production of electric energy.

From an economical standpoint some principles are of greatest importance for the success of an oil shale industry.

1. Cost of mechanical treatment as crushing, screening, must be reduced to a minimum.
2. A waste of fines, which cannot be used for oil production should be avoided.
3. Manpower required for the handling of material and operation of the plant must be kept at a minimum.

The grade of crushing and the necessity of fractionating of the raw material in general depends on the load or the throughput per square meter, with which the furnace shall be operated. This concerns an equal carbonization as well as the gasification with respect to clinkering and gas quality.

More than any other industry the carbonization of oil shale needs units of great capacity but with a relatively small throughput per square meter.

In this respect the Huboven seems to meet highest demands. Handling of shale and residue is very simple and needs no complicated machinery.

With one chamber on both sides of the dryer and a width of the chamber of 6 meters a working surface of the grate of 120 square meters can easily be constructed and operated in one unit. With the low gasification velocity of 0.15 m per hour, which has proved satisfactory for relatively lumpy clinkering shale, the capacity of such a unit reaches

$$120 \times 0.15 \times 24 = 432 \text{ tons per day.}$$

It can be increased considerably if the fusion point of the ash and the physical structure of the shale are favorable and if the crushing to a maximum size of 60 to 80 mm is not too expensive.

An experimental furnace with a daily capacity of 50 - 70 tons has been erected in 1944 in Württemberg (Photographs #2 and 3). It has been operated for nearly one year with Lias oil shale. The chamber of this furnace is 2.3 m wide and 6 m long, with a depth of the fuel bed of only 1.3 m. It has also been operated during one week with a moist oil shale from Brunswick. This shale was extremely crumbled by weathering and repeated handling with a shovel. Both shales have an extremely low content of organic substance and carbon and both are inclined to clinker. The experiments were hindered by the facts, that crushed material never could be delivered to the plant and the operation was frequently interrupted for several hours by air raids, lack of electric energy or cooling water. Therefore, the operating of this plant was more or less a study of details of the process and of the mechanical device. The plant was operated during 10 or 12 months and showed that the principles of the process and the design of the furnace were entirely correct:

a. A constant ignition of the shale and an undisturbed firebed could be maintained even with unscreened leafy crumbled shale of 30% moisture and 0 to 30 mm size. In both cases the fixed carbon was less than 5%, calculated on dry shale.

b. The movement of the layer of the material through the chamber was absolutely uniform and steady and without any movement within the layer in the vertical direction.

c. The movement of the material could be perfectly controlled and the discharging offered no difficulty even with a highly clinkered residue.



d. The quantity of solid material and dust passing through the grate was very low even with straight vertical outlets (15 mm wide) for the gas. This first unit had no gas recirculation and the dried and preheated shale was immediately ignited by sucking in cold air. Under such unfavorable conditions the recovery of oils and the quality of the gas could not be expected to be very good. The amount of recovered oils was only 40 to 50% as compared with the Fischer Assay. Nevertheless, the results of operation have clearly proved that with a sufficiently crushed material and recirculating of some gas a good thermal and chemical control of the process can be reached even with such a low carbon content. A good oil recovery, as well as a combustible gas for industrial use, can be expected.

Extended experimenting, which has been carried out in cylindrical furnaces of 1 to 3 m of diameter with various oil-shales and residues from coal mining, has clearly shown that with less than 4% fixed carbon the formation of such a gasification zone as it is required for a good yield of oil and gas is very difficult. Otherwise, 90 to 100% of oils and a calorific value of the gas of 800 to 1200 kcal/m<sup>3</sup> have easily been reached with oil shales of more than 10% oil content.

With an oil shale of 10% oil content and the corresponding organic matter which is more than twice as high as that of the oil-shales which were tried in the pilot plant, a recovery of 80% of the oil present in the shale and a calorific value of the gas of at least 800 kcal per Nm<sup>3</sup> (90 B.T.U. per cft) can doubtless be expected for a first commercial furnace. In the stationary cylindrical retort 90-100% recovery was normally obtained. Such a commercial unit of 400-500 tons capacity contains as a central part the bunker and preheater of 5 to 7 m cross section which are made out of concrete or steel. The gear for the movement of the grate and 2 exhaust fans for the dryer are placed on a platform underneath the dryer. Another exhaust fan for the carbonization gases is placed underneath this platform. Both the chambers for carbonization and gasification are connected with the long sides of the dryer. They are mounted on steel pillars. The grates are subdivided into 3 sections of 2 x 10 m and one non-movable grate at the lower end which is 6 x 2 m large. The lower part of the grate is cooled by water. The gas collecting chambers are made of sheet iron. They are lined with bricks in the lower part only. The thickness of the fuel bed is provided with 1.80 to 2.20 m according to the physical structure and the size of the shale.

The apparatus for the treatment of the gas is placed aside the furnace and a central gas purification plant for several units may be provided as practiced in many big carbonization plants. The standard outfit of such plants, which consists of electrical precipitators, indirect working gas coolers and oil scrubbers, can be used.

Belt conveyers are provided for the transport of the shale. Wagons or lorries, as usual in the mining industry, can be used for dumping the ash into the pit or into a natural valley.

A plant for treating 4,000 tons of shale per day consisting of 10 units comprises the following items and costs of erection (costs are based on conditions and prices of Germany 1938 to 1943).

A. Investment Costs

	RM
a. Ditch Storage for shale (grabenbunker)	450,000
b. Machinery for loading and transport	300,000
c. Crusher plant (3 units 100 tons per hour, 60 ton crusher)	400,000
d. Belt conveyers for shale	170,000
e. 10 Carbonizers	1,800,000
f. Condensation plant	2,300,000
g. Wash oil Distillation plant 150 cbm per hr	900,000
h. Storage tanks Capacity 12,000 tons	300,000
i. Distribution system for energy, steam and water incl. motors	800,000
k. Recooling system	400,000
l. Boiler house (gas fired) and Power Station 100 atmosphere 2 x 5000 kw turbines.	2,000,000
n. Roads and tracks	250,000
m. Laboratory, office and social buildings	400,000
	<u>10,470,000</u>
Miscellaneous	530,000
	<u>11,000,000</u>
Operating Capital	<u>1,000,000</u>
TOTAL	<u>12,000,000</u>

The investments for mining and dumping of residues are not included,

B. Operation figures for 340 working days per year.

1. Total investment cost	Germany	USA
	RM	\$
	11,000,000	6,000,000
Operating Capital	1,000,000	500,000

2. Oil Shale (long tons)

$$340 \times 4,000 = 1,360,000 \text{ tons/year}$$

3. Fresh water  $250 \times 8,600 = 2,150,000$  cbm per year

4. Laborers and employees

Shale storage	3 men
Conveyers	2 men
Crusher plant	3
Carbonizers	20
Condensation plant	8
Wash oil plant	2
Tanks and Loading	2
Power station	3
Electrician, Repair	3
Guards	2
	<u>48</u> per shift

3 shifts 144

1 shift

repair shop	10
outside work, cleaning	10
Laboratory	4
Office and drivers	10
Social rooms	6
	<u>184</u>

10% for vacation 18

Total Laborers 202

Officers:	4 foremen
	3 engineers
	1 manager

5. Production of oils (12% oil content of shale)

$1,350,000 \times 0.12 \times 0.8 = 130,500$  tons

90% tar oils 117,450 tons

10% light oils 13,050 tons

C. Production costs

<u>Oil shale (including dumping of ash)</u>	Germany RM	USA \$
1,360,000 tons @ 1.50 RM	2,040,000	
1,360,000 tons @ \$.75		1,020,000
<u>Water</u>		
2,150,000 cbm @ 0.05 RM	107,500	
2,150,000 cbm @ 3¢		64,500
<u>Wages</u>		
Laborers 202 @ 3,000 RM	606,000	
202 @ \$2,000		404,000
Engineers	60,000	30,000
<u>Repair and material for operation</u>		
2.5% of 11,000,000 RM	275,000	
2.5% of \$6,000,000		150,000
<u>Amortization Cost</u>		
8% of 11,000,000 RM	880,000	
8% of \$6,000,000		480,000
<u>Interest</u>		
3% of 12,000,000 RM	360,000	
3% of \$6,500,000		195,000
	4,328,500	2,343,500
<u>Taxes and Administration</u>		
	400,000	200,000
<u>Total Costs per year</u>	<u>4,728,500</u>	<u>2,543,500</u>

Production cost per ton of oil

RM

\$

$$\frac{4,728,500}{130,500}$$

36.20 RM

$$\frac{2,543,500}{130,500}$$

19.50

The heating value of the surplus gas at least 300,000 Cal. per ton of shale is available for production of power or for sale. It is not accounted for.

Operating Costs per ton of oil shale

$$\frac{4,728,500 - 2,040,000}{1,360,000} =$$

1.98 RM

or

$$\frac{2,543,500 - 1,020,000}{1,360,000} =$$

1.125

Operating costs

per ton of oils

$$\frac{1}{0.096} \times 1.98 =$$

20.60 RM

$$\frac{1}{0.096} \times 1.125 =$$

11.70

Cost of oil-shale per ton of oils

$$\frac{2,040,000}{130,500} =$$

15.6 RM

7.80

The above figures shall give but a rough estimate on the relative influence of the mining costs, the operating costs, and the loss of oil due to the necessity of the waste of fines or due to an inadequate efficiency of the process.

Figures corresponding to a 95% efficiency of oil recovery or to an increased oil content (14.3% instead of 12%) are as follows:

Operating costs per ton of oils

$$\frac{80}{95} \times 11.70 =$$

\$9.86

Cost of oil shale per ton of oils

$$\frac{80}{95} \times 7.80 = \$6.57$$

Production costs per ton of oils      \$16.43 per ton

With an oil content of 14.3% and 95% recovery, the production costs are further reduced to  $16.43 \times \frac{80}{95} = \underline{\$13.84}$

1. A waste of the "fines" which cannot be treated, influences the cost of the raw material only.

2. Higher costs of operation (capital and labor) is of a relatively higher influence on the total production costs.

3. A higher efficiency would reduce the cost of the raw material and the cost of operation.

In case of treating higher grade shales with an oil-content of 20% which probably need underground mining, the cost of the shale might be increased to \$3 per ton.

With 95% efficiency  $\frac{1}{0.95 \times 0.2} = 5.3$  tons of shale are required for the production of one ton of oil.

The production costs are as follows:

a. For shale  $5.3 \times 3.00 =$       \$15.90

For operating  $5.3 \times 1.125 =$       5.9

Total production costs per ton of oil      \$21.8 per ton

These figures show that in case of underground mining the production costs of oils are so much increased, that they become higher than those of a low grade shale which is mined by open cut mining and operated with a low efficiency of the oil extraction.

### e. Methods of briquetting lignite and fines of coal

Lignites with a high moisture content, mostly mined in open cut mining, in general are much disintegrated already after mining and transport, or they are disintegrating rapidly, when drying on the transport to consumers, or when dried in a dryer in order to increase the heating value. Such lignites can only be utilized near the mine and for boiler firing. For the utilization in a distant market or for a use, for which a solid lumpy fuel is required, it must be dried and briquetted before shipment. Briquetting is also required for the carbonization or the gasification of such lignites.

1. Lignites and most sub-bituminous coals can be briquetted without a binder only by sufficient pressure when they have been dried to a certain moisture content and grinded to a certain degree, both depending on the nature of the raw material. Very little or no grinding is necessary with the German lignites (brown coal). They easily disintegrate to a size of 0 to 4 mm when dried from 55 to 15% moisture. With this size and moisture content they can be briquetted to a solid briquette by using a pressure of 1000 to 1500 kg/cm<sup>2</sup>. For sub-bituminous coal and bituminous coal a better grinding to a maximum size of 1 mm with 50% grain under 10,000 mesh/per square mm is required and a pressure of 2500 to 3000 kg/cm<sup>2</sup>.

Ten years ago briquetting without a binder has been limited to German brown coal. Other coals, mostly anthracite and low volatile coal, have been briquetted after a moderate grinding with an addition of 5 to 7% binder and with a pressure of 2-300 kg/cm<sup>2</sup>. These briquettes are sold for domestic use.

In connection with the carbonization of coal for oil production and also for the production of a clean solid coke from non-caking coals for household and industrial purposes new methods of briquetting have been developed recently.

The process used in the lignite fields of Germany works as follows:

The raw lignite with 55 to 60% moisture is crushed to 6 mm size with a hammer mill and then dried to 15% moisture in a rotating drum drier. This drier is fitted with several hundreds of 4-inch pipes which are outside heated by steam of 2 kg/cm<sup>2</sup> pressure and through which the lignite passes slowly owing to a moderate inclination of the rotating drum. The dried lignite, which is then of the right size for briquetting, is cooled down to about 40°C and is then briquetted in a piston press (exter press) in which the lignite is pressed in an endless rope through a horizontal channel with 1200 to 1500 kg/cm<sup>2</sup> pressure.

These briquettes are also used in a big scale in LURGI Carbonizer plants for the production of oils. The coke produced by this method is relatively weak and to a great proportion smaller than 1/2 inch, so that it is mostly sold to power plants at a low price or used in a Winkler gas producer for the production of water gas.

2. A better quality coke is produced by the LURGI-KRUPP process of briquetting and carbonizing, recently developed and operated in several plants since 1935. It has been found, that with a briquette made of a fine powder, homogeneous with relation to the size and the moisture content of its particles, a dense solid coke can be produced by a very slow increase of the temperature during carbonization. The economy of such a method of carbonization is considerably improved by the much higher price of the lumpy coke as compared with former methods. A new method of drying for lignite was developed for this purpose as well as a new press able to treat a very fine powder with an increased pressure. The process works as follows (Fig 5):

The raw lignite is reduced to 4 mm size by screening and crushing. It is then dried to 8% moisture in a stream of hot combustion gas of 1000 to 1100°C. This drying takes place in less than one second and causes a moderate explosion of the greater particles by the sudden evaporation of the moisture. The waste gas of the drier, together with the coal, passes a fan in which the drying and grinding to a fine powder is completed. The gas enters this fan with 300°C and leaves it with 120°C. The fine powder of coal is then separated from the gas by an electrical precipitator. It is cooled to 45°C in a cascade-cooler by means of an inert gas circulated through this cooler and a spray cooler operated with water.

It is then briquetted in a Krupp roller-press. This press consists of a steel ring of 2 meters in diameter, within which a disk wheel of 0.8 m diameter is rotated on a horizontal shaft. The briquettes are made as an endless rope between the disk and the ring, with a pressure of 2500 kg/cm<sup>2</sup>. The ring is moving absolutely free on two roller bearings only by the friction of briquetting. The endless rope of briquettes is broken into briquettes of 4 inch length according to notches made on the rope by small teeth on the outer rim of the disk. This improved principle of briquetting needs less energy than the piston press in spite of the higher pressure. The capacity of a press amounts to 300 to 500 tons of briquettes per day.

The briquettes are very homogeneous and much harder than the normal briquettes. When heated slowly they shrink so equally without splitting that 50% loss of volatiles are equalized by 50% loss of volume and with older lignites and sub-bituminous coal the carbonized briquettes may be more solid and more resistant to abrasion than a good coal briquette.



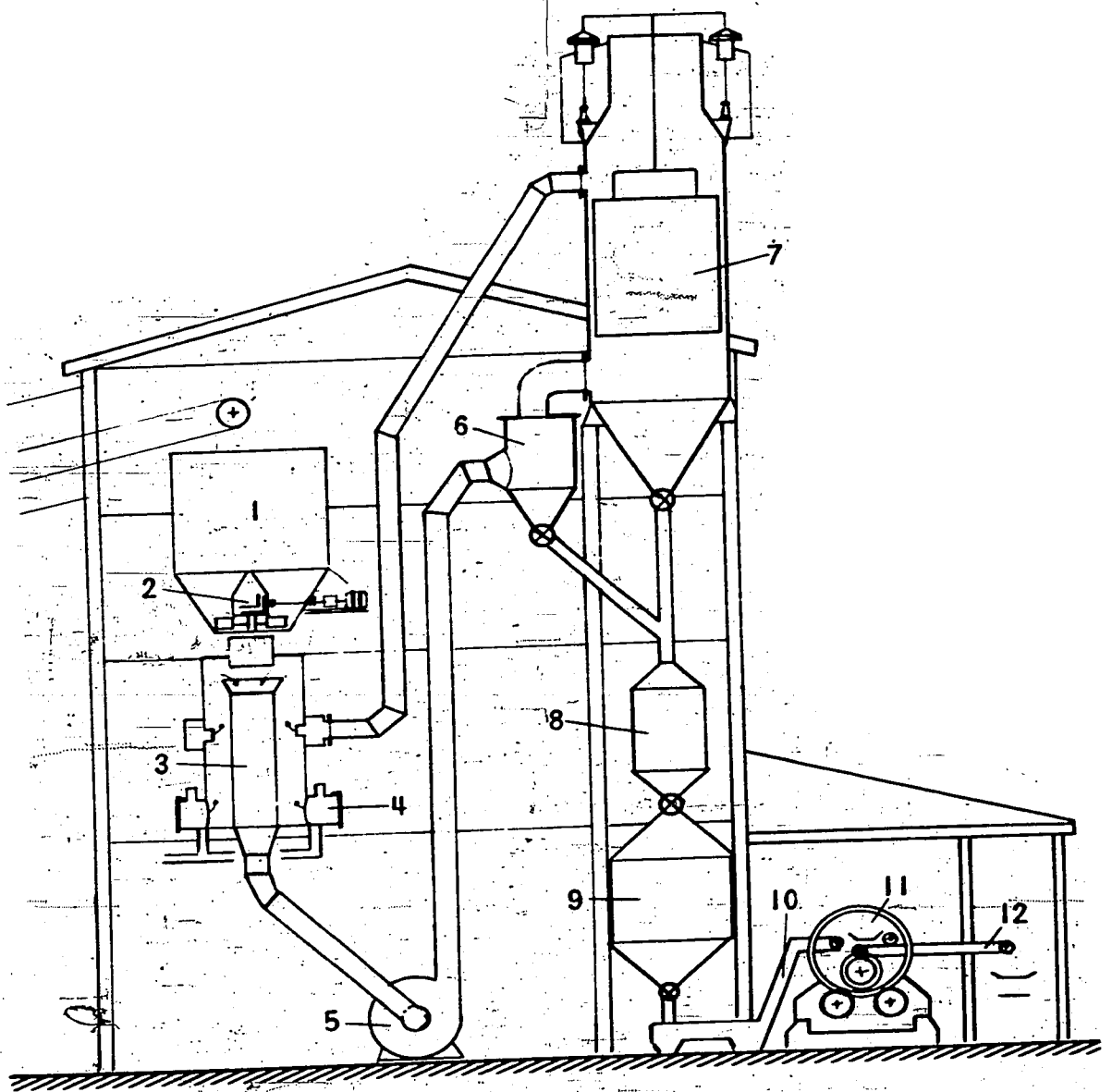


Figure 5. Diagram of a Brown Coal Briquetting Plant for Making Lump Coke. (Lurgi-Krupp Process).

- 1 raw coal bunker
- 2 coal feed hopper
- 3 coal drying oven
- 4 gas burners of drying oven
- 5 combined centrifugal fan and pulverizer
- 6 coarse pulverized coal separator
- 7 electrofilter for precipitation of pulverized coal
- 8 pulverized coal cooler
- 9 pulverized coal storage bin
- 10 pulverized coal conveyor
- 11 ring-roller press
- 12 briquette conveyor

For subbituminous and bituminous coal this method of briquetting requires a finer grinding by a pulverizer or an addition of a small amount of binder to increase the plasticity of the material. As a binder 2 to 3% of heavy oils or sulfide liquor have been used. The binder required can easily be separated from the products of carbonization.

Big scale experiments have proved that a first-class coke suitable for water gas producers and blast furnaces, can be produced in this way from bituminous and subbituminous coals which cannot be treated in a coke oven.

The fine coal being very suitable in general for a separation of ash before briquetting, it is possible to produce a high-grade coke with a reduced ash content.

3. The usual methods of briquetting with pitch or asphalt as binder have not proved satisfactory for carbonization. The roller press normally used does not allow sufficient pressure as necessary for producing a dense briquette with a reduced content of binder. Normal briquettes, made with 6 to 7% of pitch, soften in a furnace when a temperature of 70 C is passed. They cannot be treated, therefore, in a continuous vertical furnace or shaft. They could be treated without disintegration in thin layers, as for instance, in a tunnel kiln. But this method of carbonization is relatively expensive and does not allow a higher temperature than 500 C, as it is required for a good low volatile coke.

4. Briquettes made from bituminous coal with the normal equipment of mixers and roller presses but with sulfide liquor as a binder have been carbonized successfully in a 10 carbonizer plant at Blechhammer. The capacity of a LURGI carbonizer could be raised to 450 tons a day with such briquettes and the coke with 3% of volatiles was used for the production of water gas.

For briquetting, 2% of sulphite liquor (on dry basis) is used, and the briquettes are dried and hardened in a special belt drier before they are charged into the carbonizer. The recovery of oils is as normally 92% of the Fischer assay. The coke is very solid and resistant to abrasion if a moderately fusing coal is added to the mixture.

## B. By-product - Gasification

### a. Gas producer for industrial heating gas.

For the gasification of lignites, subbituminous and bituminous non-caking coals, several types of gas producers of a special design have been developed in Germany, which are equipped with a retort or a separate carbonizing zone. Usually the heating of this zone is performed exclusively by direct heating. Part of the gas produced from the coke in a lower zone by its gasification with air is sucked through the carbonization zone and conducted separately from the rest of the producer gas to a condensation plant, in which the tar vapors and sometimes also light oil

are condensed. 1 to 1.5m<sup>3</sup> of hot producer gas is required per kg coal for the carbonization zone, depending on the moisture content of the raw coal. Mostly gas producers of the rotating grate type are used, of 2.5 or 3.0 m diameter of the shaft. The gasification zone is furnished with a water jacket for a better control of the clinkering of the ash and the movement of the coal and the gas.

Such plants, up to 500 tons capacity, have been erected by the LURGI Co. and the Pintsch Co., mostly in works of the metallurgical industry.

The recovery of tar amounts to 75% of the Fischer assay and its quality is good, if a suitable coal, which does not disintegrate or fuse is used.

#### b. Gas producer for water gas and city gas

According to a proposal of Hillebrand, non-caking lumpy coal is gasified continuously with a mixture of circulating gas and superheated steam in a shaft furnace as shown in Fig (6). The gas mixture is superheated in a pair of regenerators which are intermittently heated by combustion gas. It is introduced into the fuel bed with a temperature of 1100 to 1200 centigrade and gasifies the fixed carbon. The gas producer is equipped with a separate carbonization zone, in which the oils can be recovered. The carbonization zone is heated by introducing part of the gas, which leaves the gasifying zone with 600 to 700 C, into the carbonization zone. After separating the tar oils from this part of the gas by an electrical separator, this gas goes back to the regenerator, where the hydrocarbons are cracked into hydrogen. Part of the gas leaving the gasifying zone is washed with water, which takes out dust and saturates the gas with steam. The surplus of the gas, corresponding to the actual production of gas for synthesis, is withdrawn here for further cleaning.

Another part of the circulating gas is burned in the second regenerator with air for its heating up. The output of gas for synthesis can be increased by heating the regenerators with a producer gas produced in a separated producer with air.

In the Pintsch-Hillebrand design of this process a cylindrical shaft is used for carbonization and gasification. The regenerators are placed under the gas producer, to a compact unit; the capacity of such unit is relatively small, 60 to 100 tons of coal per day.

Another design for the same principle has been developed by the Koppers Co. of Essen. In this case a rectangular shaft is used, its cross section subdivided for a better control of the flow of the gases through the coal. The regenerators of increased dimensions are placed aside the gas producers and designed similar to the cowpers of a blast furnace. The capacity of such rectangular units is considerably higher than that of the cylindrical producer. 200 to 300 tons of lignite brickettes can be gasified

Hillebrand process  
of gasification  
(Pintsch & Koppers)

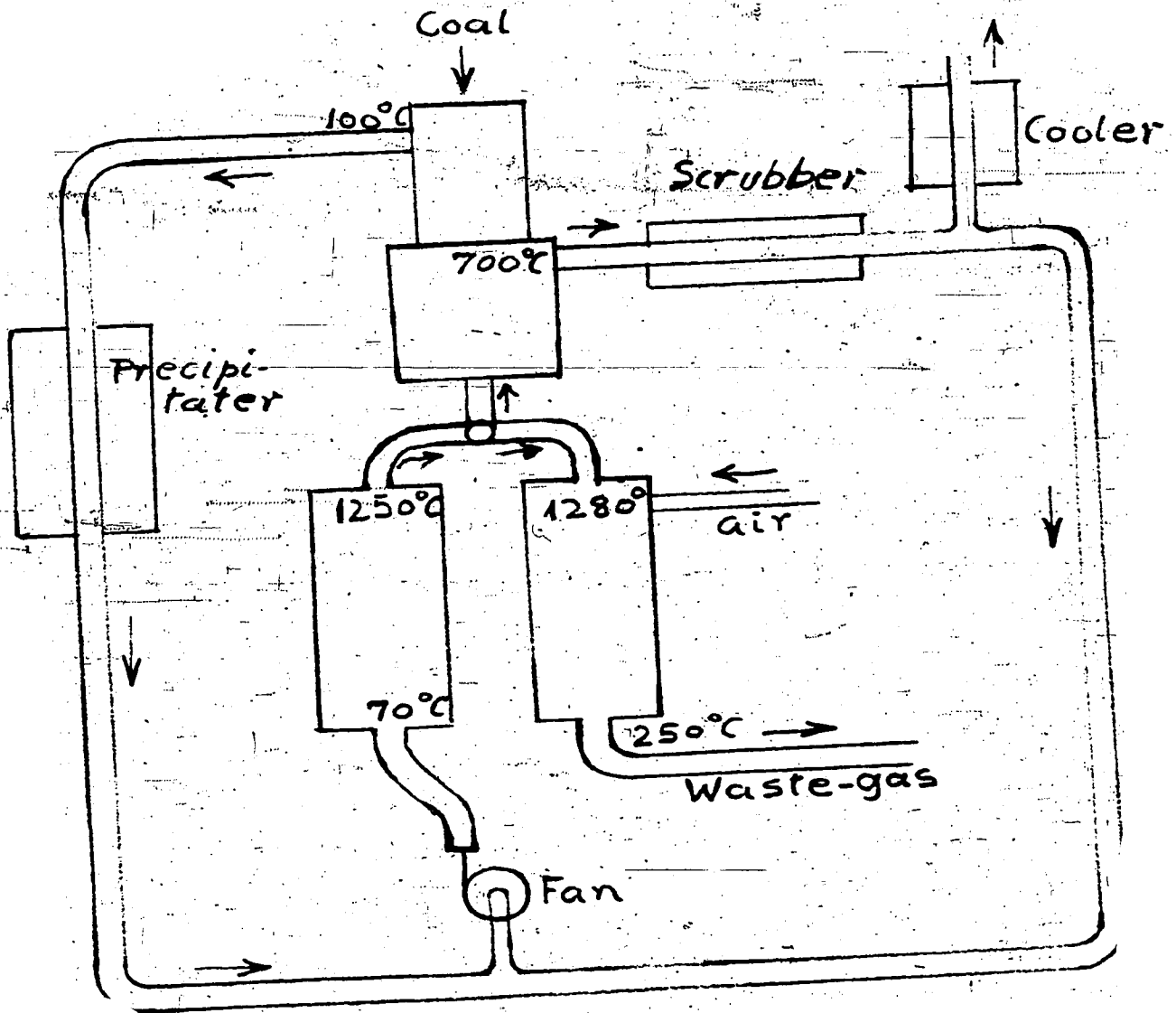


Fig. 6

in one unit per day. Part of the carbon is extracted from the producer, together with the ash, and gasified in a normal gas producer for the production of the gas, which is required for the heating of the cowpers.

A plant of the Pintsch-Hillebrand system has been erected for the production of hydrogen in the hydrogenation plant Wesselin, which is operated with lignite from the Rhine Valley.

The Koppers system has been used on a big scale for the production of water gas for the Fischer synthesis from lignite-briquettes at the Ruland plant.

For coal-briquettes of a good activity as German lignite, the Hillebrand method has proved satisfactory if gases of a high hydrogen content are required. The efficiency of this process is limited by the maximum temperature which can be employed in the cowper with respect of the fusion point of the ash of the coal. Part of this ash is carried from the gasification zone into the cowper and endangers the brickwork considerably when the fusion point of the ash is exceeded. This limit of temperature makes the process unsuitable for a fuel with a low fusion point of the ash and unsuitable, also, when a high content of carbon monoxide is required as e. g. for the synthesis of hydrocarbons with an iron catalyst.

The process is limited to a lumpy coal or a briquette which stands well against disintegration in order to avoid considerable amounts of dust in the relatively great volumes of circulating gases.

The heat economy of this method is in the same line with that of a water gas plant and amounts to 55 to 60%.

Advantages of the method are the high capacity of a unit and the possibility of cracking the hydrocarbons immediately in the gas producer or regenerator.

Gas produced from lignite briquettes in the Koppers plant is composed as follows:

12.9% CO <sub>2</sub>	56.0% H <sub>2</sub>
0.1% CnHm	1.2% CH <sub>4</sub>
0.1% O <sub>2</sub>	1.6% N <sub>2</sub>
28.1% CO	

The recovery of by-products in connection with this producer plant was low and the products not of the best quality. In both plants coals with a relatively low tar content were used. With a suitable equipment for predrying and carbonization, a recovery of 80 to 85% of the tar should be possible.

## THE LURGI CARBONIZER WITH GASIFICATION OF RESIDUE

The Lurgi Carbonizer developed for carbonization of all types of non-caking coals and lignite in order to produce tar oils and a smokeless free burning coke for domestic heating with 10-15% volatiles, or a coke for industrial purposes with 2-3% volatiles, has proved very efficient in many big industrial plants. As far as industrial utilization is concerned the coke is mostly used for the production of gas for heating or for chemical synthesis purposes. The rest is burned in power stations.

Lately a considerable interest has grown up to combine carbonization of the fuel with complete gasification of the residue. By such combination the heat economy can be considerably improved and the cost of investment and of labor can be considerably lowered. For such combination the coke-cooling zone of the Lurgi-Carbonizer had to be replaced by a gasification zone. Extended experience with gas producers of the rectangular type has been very helpful for developing the type of a rectangular carbonizer-gas producer.

Recently, interest in a combined carbonizer-gas producer has been shown by many power stations which have to work with fuels of high ash content and a relatively low melting point of the ash. In this case the efficiency of a boiler plant may be considerably reduced if fired with a pulverized fuel. With gas-fired boilers this difficulty can be eliminated and much better working conditions, reduced cost of investment, and an improved economy of power production can be expected.

The latest development in this line, however, seems to end in power production by gas-turbines, which would raise the consumption of gas considerably.

Until now, gas producers for gasification of lumpy coal have been of the rotating grate type, with a maximum diameter of 12 feet and not exceeding 50 tons per day capacity per unit for uniformly sized fuel having ash of a relatively high melting point. Recovery of oils is not feasible with such a producer.

With the principle of a rectangular shaft such as that on the Lurgi carbonizer, subdivided for an equal and controlled distribution and movement of the coal and the gases, the capacity per unit can be increased considerably. The normal carbonizer with 2 shafts of 16 m<sup>2</sup> (277 sq. ft.) each has a cross section 3.2 times larger than that of a 3.5 m (12 ft.) cylindrical producer. Furthermore, it was possible to utilize in the rectangular carbonizer the system of gasifying downwards in the direction of the flow of the coal. In this way the throughput of the small gas producers has been increased considerably. This principle of construction allowed the building of a carbonizer-gas-producer-unit for capacities of 150-200 tons of bituminous coal per day,

equal to 200-300 tons of sub-bituminous coal or lignite with 10-25% moisture content.

As in a normal carbonizer plant the charge and the discharge of the units are fully automatic. The coal is dried and preheated before entering the carbonizing zone, which allows recovery of oil and phenol with a minimum of gas and a minimum of waste water to be treated. The residue of carbonization is gasified in a separate zone. In this way the greatest part of the gas is produced nearly free of hydrocarbons and sulphur and needs only a very moderate amount of cleaning for utilization in a synthetic process. For power stations it can profitably be used without any cooling or cleaning.

By gasifying with concentrated or pure oxygen, any gas as required for the synthesis of hydrocarbons or ammonia can be produced. The gas from the carbonizing zone which contains all the hydrocarbons can be used directly as a high grade heating gas or it can easily be cracked into a mixture of hydrogen and carbon monoxide for synthetic purposes.

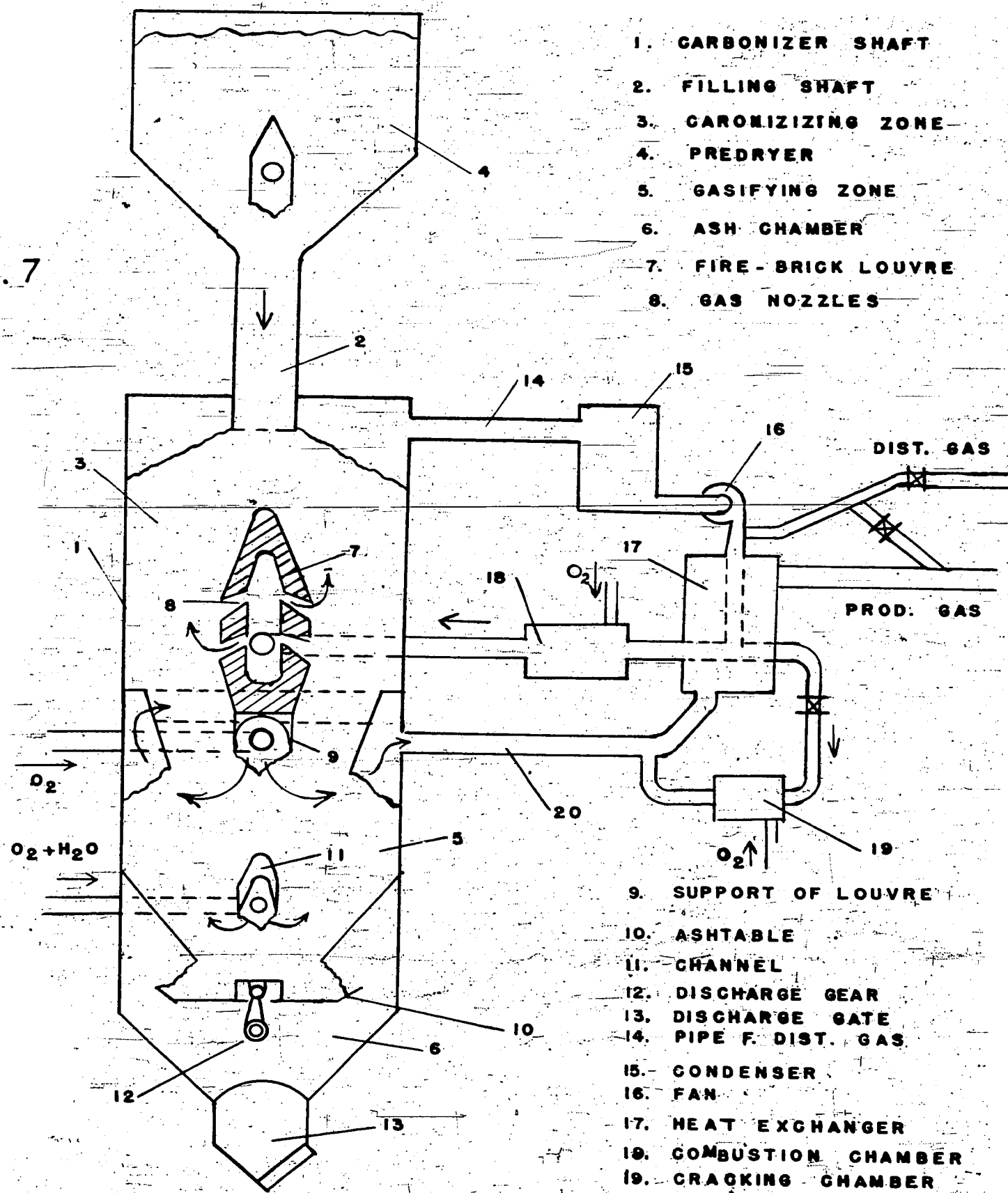
The scheme of construction and processing of the combined carbonizer-gas producer is shown on ~~Sketch 1~~. It operates as follows:

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The fuel, suitably screened (to size limits 1 to 4), is dropped into Storage bin, 4, which in its lower part contains the usual device for drying with circulating gas. The preheated material then passes through the shaft, 2, into the carbonizing zone, 3. Here it is carbonized by a circulating flow of distillation gas entering with a temperature of 600 to 900 centigrade at the nozzles, 8, and leaving with a temperature of about 200 centigrade through the pipe, 14. In the usual equipment, 15, suitable for gas cleaning and cooling, oils and benzine are extracted from the gas. After passing a fan, 16, the main part of the gas is heated in the heat-exchanger, 17, and returns to the carbonizing zone passing through the combustion chamber, 18, in which its temperature is regulated as desired by means of a partial combustion with air. Oxygen may be used if a high grade gas is required.

The carbonized fuel enters through a narrow opening into the gasification zone, 5. Here air is introduced into the fuel bed from the channel formed underneath the support, 9, which is water-cooled to prevent its burning. A second flow of oxygen, saturated with steam, enters through the channel, 11, in order to complete gasification of the rest of the carbon under conditions which prevent heavy clinkering of the ash. The gas produced in zone, 5, leaves through the pipe, 20, with a temperature of 500 to 700° centigrade, and after passing the heat exchanger, 17, it goes to consumption in a furnace or may be used for synthesis after an adequate cleaning.

Fig. 7



- 1. CARBONIZER SHAFT
- 2. FILLING SHAFT
- 3. CARONIZING ZONE
- 4. PREDRYER
- 5. GASIFYING ZONE
- 6. ASH CHAMBER
- 7. FIRE-BRICK LOUVRE
- 8. GAS NOZZLES

- 9. SUPPORT OF LOUVRE
- 10. ASHTABLE
- 11. CHANNEL
- 12. DISCHARGE GEAR
- 13. DISCHARGE GATE
- 14. PIPE F. DIST. GAS
- 15. CONDENSER
- 16. FAN
- 17. HEAT EXCHANGER
- 19. COMBUSTION CHAMBER
- 19. CRACKING CHAMBER
- 20. PIPE F. PRODUCER-GAS

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LURGI CARBONIZER—GAS PRODUCER



The fuel bed is supported at the bottom of the shafts by a table, 10. Upon this table an iron bar is mounted that is movable to both sides by a mechanical gear, 12. By altering the movement and speed of this bar, the throughput of coal and the extraction of ash is regulated. The ash is dropped from the table into the ash chamber, 6, from which it is discharged by a sluice, 13, or through a water seal.

The surplus gas from the carbonizing zone has a calorific value of 1800 to 2500 kcal/m<sup>3</sup> when air is used in combustion chamber, 18, and of 4000-4500 kcal/m<sup>3</sup> after washing with water when oxygen is used. It may be utilized separately or it may be cracked by partial combustion with oxygen in the cracking chamber, 19, after preheating it in the heat-exchanger, 17. If only heating gas is required carbonization and gasification take place with air. The heating of the carbonizing zone can be simplified in this latter case by sucking part of the hot producer gas directly upwards into the carbonizing zone.

The method of gasifying solid fuels, as described above, permits a good recovery of by-products and a separate treatment and recovery of the distillation gas and it allows, at the same time, the gasification of most of the fixed carbon in the fuel with the use of very little or no steam. In this way, a gas of a relatively high content of carbon monoxide, as required for some synthetic processes, is produced. For heating purposes, a reduced consumption of steam may decrease the production cost considerably.

In many cases coals and lignites with a high content of oil, but also a high content of ash and moisture, can be mined so cheaply that they can be considered as a very cheap fuel for industrial heating, chemical synthesis and power stations as well as for the production of oils. Combined carbonization and gasification opens a way for utilizing such fuels on a big scale.

With an industrial plant, operated with a high volatile sub-bituminous coal and with only one type of carbonizer-gas producer, practically all types of industrial gases can be produced that are required for heating, synthesis, and city gas. Even a wide variation of the quantity and the quality of the products, according to the actual market, is possible within short intervals.

The most important figures of the operation and the efficiency of the carbonizer-gas producer for a medium sub-bituminous coal are the following:-

A. Coal:	Size: 1/4 to 1"	or 1 to 3"	or briquettes
	Moisture	10%	
	Tar (Fischer Ass.)	10%	
	Ash	12%	
	High cal. value	5800 kcal/kg	
		10440 BTU/lb	

B. Thermal efficiency of the process

$$\eta = \frac{\text{high cal. val. (Tar + dist. gas + prod. gas)}}{\text{high cal. val. of coal}} = 80\%$$

$$= \frac{4640 \text{ kcal/kg}}{8350 \text{ BTU/lb}}$$

C. Recovery of tar incl. Light Oils

92% of Fischer assay

$$0.092 \text{ Kg/kg} = 0.092 \cdot 9600 = 883 \text{ kcal/kg coal}$$

$$= 3500 \text{ BTU/kg coal}$$

D. Quality of gas with variant operating conditions

	Operated with;	95% OXYGEN		
		AIR	Raw	Washed
<u>Distillation gas</u>	Nm <sup>3</sup> /kg	0.27	0.19	0.172
	CO <sub>2</sub> %	12.0	18.0	2.0
	C <sub>n</sub> H <sub>m</sub> %	0.6	1.0	1.1
	O <sub>2</sub> %	0.1	0.1	0.1
	CO%	18.3	27.9	33.4
	H <sub>2</sub> %	24.0	36.5	43.6
	CH <sub>4</sub> %	10.0	15.5	18.6
	N <sub>2</sub> %	35.0	1.0	1.2
	high cal. kcal/nm <sup>3</sup> value	2325 (243)*	3586 (377)*	4270 (448)*
	<u>Producer-gas</u>	Nm <sup>3</sup> /kg	2.62	1.2
CO%		9.0	15.0	2.0
O <sub>2</sub> %		0.1	0.1	0.1
CO%		21.8	38.2	43.9
H <sub>2</sub> %		17.0	45.0	52.1
CH <sub>4</sub> %		0.1	0.2	0.2
N <sub>2</sub> %		52.0	1.5	1.7
high cal. kcal/nm <sup>3</sup> value		1188 (125)*	2544 (267)*	2839 (297)*

\* (---) BTU/cu.ft. 60° F 30" moist

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AIR

OXYGEN

40 %

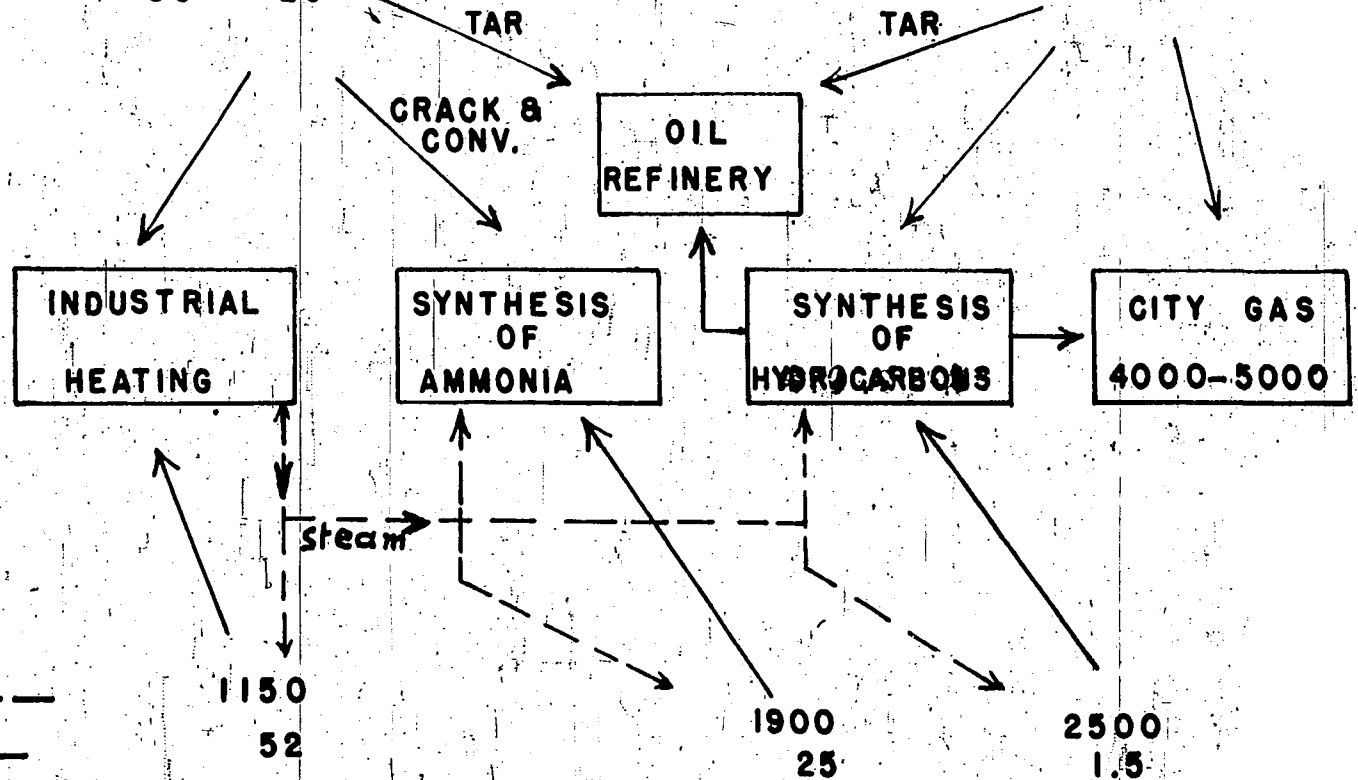
95 %

RAW GAS PRODUCED IN -- Kcal/nm<sup>3</sup> -- 2000 - 2600  
CARBONIZATION ZONE -- N<sub>2</sub> % -- 35 - 25

3500  
1.0

(SEE FIG. 3, SKETCH 1)

TYPE OF USE  
FOR GAS



RAW GAS PRODUCED -- Kcal/nm<sup>3</sup> -- 1150  
IN PRODUCER ZONE -- N<sub>2</sub> % -- 52

(SEE FIG. 5, SKETCH 1)

UTILIZATION OF CARBONIZER — GAS PRODUCER GAS  
FOR VARIOUS INDUSTRIES

A gas for the synthesis of ammonia can be produced with a 40% Oxygen-concentration.

Annexed table shows the operating conditions in connection with various industrial utilizations of the gases. Many types of manufacturing can be combined according to the conditions of the market. Five types of gas, each being variable in its contents of Nitrogen, Hydrogen, Carbon monoxide and in its heating value according to the momentary demand of consumers, are available from the two zones of the Carbonizer Gas producer. Variations in composition can be made within a few hours. Even a temporary production of coke is possible by shutting down the gasifying zone.

In the LURGI - Pressure Gasification process gases with a high content of hydrogen are produced under a pressure of 10 to 20 atmospheres by complete gasification with oxygen and steam. This process originally was developed for the production of city gas from lignite. For this problem working under pressure promised a low velocity of the gas in a bed of small sized fuel and therefore a high throughput even with the unfavorable fuel. A favorable influence of the pressure on the formation of gaseous and liquid hydrocarbons could be expected and the high amount of  $\text{CO}_2$ , which is unavoidable for a gas of a high content of hydrogen could easily be eliminated by washing the gas with water. The problems presented by the necessity of continuously introducing solid fuel and the extraction of the ash have been solved satisfactorily in a pioneer plant. Later on even a fully automatic charger has been developed.

The process works as follows:\*

Screened coal 2 to 10 or 5 to 25 mm size is gasified in a cylindric shaft producer, which is completely covered by a water jacket. The pressure of this jacket is equalized with that of the fuel-bed by a connecting pipe. The device for the extraction of the ash is of special design with regard to the limited space available in a high pressure plant and with regard to the high concentration of oxygen.

A mixture of 15% oxygen and 85% superheated steam is introduced at the bottom of the producer into the fuel, which is gasified to a primary gas of a high content of  $\text{CO}_2$ , but also a high content of  $\text{H}_2$  and  $\text{CH}_4$  according to the equilibrium of 20 atm. pressure.

In the upper part of the fuel bed the coal is carbonized by the sensible heat of the producer gas. Oil vapors are carried off with the gas to a scrubber, immediately connected to each producer, in which the gas is washed with a circulating condensate of water and heavy oils at a temperature of 140 C. Dust and asphalt are taken out here. Any sticking of the pipelines

\* Chemistry of Coal Utilization p. 1663.

could be avoided with a gas with such a high content of tar oils. The gas is cooled in a series of surface coolers to 25 C, then washed in an oil scrubber for the extraction of benzene. Then it is washed by water for the elimination of  $\text{CO}_2$  and  $\text{H}_2\text{S}$ . After extracting the last traces of  $\text{H}_2\text{S}$  with iron oxide it is ready for use in a public distributing system. In case of a high content of hydrogen sulfide the gas can be washed profitably with a potash solution before the washing with water for the extraction and recovery of sulphur.

For the production of a gas for the synthesis of hydrocarbons, or for the production of hydrogen, the gas is cracked either by a catalytic method or by a partial combustion with oxygen.

For local industrial heating or for power production in a gas turbine, the gas is produced with air under pressure. It may be used without cooling, when the coal has only a low content of tar. The process has been used for many years in 3 plants producing city gas from lignite.

A plant for the production of gas for the synthesis of hydrocarbons with an iron catalyst under pressure has been under construction at the end of the war.

With a lignite of 28% moisture the throughput of a producer of 8 3/4 feet in diameter was as high as 180 to 200 metric tons per day. (70 to 80 metric tons of fixed carbon). Coals of a low softening point of the ash (below 1100°C) can be gasified with a reduced throughput of 40 to 60 tons of fixed carbon.

1. With this process non-caking coal can be gasified, even small sized, with a very high throughput and with less than half of the oxygen required for a gasification under atmospheric pressure.

2. Condensation of oils, cooling, and cleaning of the gas requires much less equipment under pressure.

3. The recovery of light oils is considerably improved and cheaper than under atmospheric pressure.

4. The equipment of compressors and their considerable power consumption are saved in case of utilizing the gas under pressure.

5. A gas of 4000 to 4500 kcal/Nm<sup>3</sup> (high vel.) can be produced immediately by complete gasification of solid fuels.

6. The efficiency of the process is high 83 to 85% of the heating value of the fuel is recovered in the oils and the cleaned gas.

7. 70 to 80% of the oils contained in the fuel is recovered in a very good quality. 20 to 25% of the oils are light oils with a high proportion of aromatic oils.

The disadvantages of the process are a relatively high consumption of steam and in case of low fusion point of the ash the relatively low content of carbon monoxide which is unfavorable for use in some synthetic processes.

Some results of the pressure gas producer plant at Bohlen are the following:

The plant is operated with screened dried lignite, a sort of waste delivered from several briquetting plants in two qualities:

1. Grains 2-6 mm from the dryers (unsuitable for briquetting).
2. Slack 2-10 mm of briquettes (abrasion product).

Operating figures for the month of Jan. 1941

Analysis of coal:	moisture	%	18
	volatiles	"	73
	fixed carbon	"	
	ash	"	8
Fischer assay:	moisture	%	22.8
	tar	%	14.4
	coke	%	52.8
	gas + loss	%	9.0
Cal. value (high)	kcal/kg		4600
Analysis of gas			Cleaned gas
	CO <sub>2</sub> + H <sub>2</sub> S	%	1.9
	C <sub>n</sub> H <sub>m</sub>	"	0.8
	O <sub>2</sub>	"	0.2
	CO	"	20.6
	H <sub>2</sub>	"	52.2
	CH <sub>4</sub>	"	22.0
	N <sub>2</sub>	"	2.3
Higher heating value	Kcal/nm <sup>3</sup>		4400
Recovery of gas	954 nm <sup>3</sup> /ton of coal		

Tar oils and light oils: Recovery 82% of Fischer Assay

Recovery of tar ( $>180^{\circ}\text{C}$ ) 94.4 kg/ton

Recovery of light oil ( $<180^{\circ}\text{C}$ ) 23.6 kg/ton

Consumption of oxygen:

$\text{nm}^3/\text{nm}^3$  of gas 0.146

Consumption of steam:

for gas producer 1.32 kg/ $\text{nm}^3$  of gas)

for heating 0.26 kg/ $\text{nm}^3$  of gas) 1.59 kg/ $\text{nm}^3$

Power consumption per  $\text{nm}^3$  of gas:

for oxygen 0.186 KWh

for producer plant and gas cleaning 0.019 "

for gas-washing 0.086 "

Efficiency of gasification:

Efficiency =  $\frac{\text{high heat val. of gas} + \text{high heat. val. of tar}}{\text{high heat. val. of coal}}$

.700 = 83.8%

Results of gasification of several other fuels are the following:

	<u>Lignite</u>	<u>Anthracite</u>
Moisture %	27.4	6.6
Tar content	10	-----
Volatiles / f. Carbon	67.5	88.4
Ash	5.1	5.0
Cal. val. B.T.U. p. lb	8520	13680
Size inches	0.08-0.39	0.12-0.39

Composition of purified gas

$\text{CO}_2$  3 1.0

$\text{C}_n\text{H}_m$  0.5 0.3

$\text{O}_2$  0.1 0.0

CO	22.8	27.9
H <sub>2</sub>	48.7	52.4
CH <sub>4</sub>	22.6	16.9
N <sub>2</sub>	22.3	1.5
Spec. gravity	0.448	0.410
BTU p. cft dry gas	456	437
Oxygen per 1000 cft	150	198
Steam used per 1000 cft	66	83
Tar recovery % of tar cont.	72	—

The LURGI — pressure gasification process is superior to other methods, where a non-caking small sized fuel shall be converted into a high grade gas for long distance supply, or where a gas of a high content of hydrogen is needed under pressure.

Fuels with a high tar-content and a high ash-softening point are favorable for a low production cost. Some figures of the investment and the production cost of a plant for the production of 240 mio nm<sup>3</sup> of city gas in 7500 hours per year are the following

Coal (subbituminous) 1/8 to 1 inch size

Moisture	%	15.0
Fixed Carb + volat.	%	72.2
Ash	%	7.8
Tar	%	8.7
High calor. val.	kcal/kg	4940
Ash-softening point		1150 °C

City gas	CO <sub>2</sub>	%	3.0
	C <sub>n</sub> H <sub>m</sub>	%	0.5
	O <sub>2</sub>		0.1
	CO		22.8
	H <sub>2</sub>		48.7
	CH <sub>4</sub>	%	22.6
	N <sub>2</sub>		2.3



high cal. value	4300 kcal/nm <sup>3</sup>
spec. gravity	0.448
Recovery of gas	835 nm <sup>3</sup> /t
Consumption of oxygen	0.16 nm <sup>3</sup> /nm <sup>3</sup> of gas
Consumption of steam for gas prod. and heating	1.35 kg/nm <sup>3</sup> of gas
Consumption of water (fresh water)	300 nm <sup>3</sup> /hour
Consumption of energy:	
incl. oxygen plant	0.22 Kwh/nm <sup>3</sup> of gas
tar recovery:	70% of tar content
	1/5 as benzine

Annual figures:

Gas	240 mio nm <sup>3</sup>
Coal	288,000 tons (metric)
Oxygen	38.4 mio nm <sup>3</sup>
Steam	318,000 tons
Energy	53 mio KWH
Water	4.8 mio m <sup>3</sup>
Tar	14,000 tons
Benzine	3,520 tons

Labor and wages:

140 laborers  
3 foremen  
2 engineers  
1 manager

Investment cost (1943)

12,000,000 Rm (5,500,000 - \$)

(steam and energy are delivered from outside)

If a higher calorific value than 4300 kcal/nm<sup>3</sup> is required, the gas can be treated in a simplified synthesis plant under pressure. From 1000 nm<sup>3</sup> of gas are produced:

44 kg of liquid products

of which	17.6 kg. waxes
	15.4 kg. gas oil
	11.0 kg. benzin

and 670 nm<sup>3</sup> of converted gas:

CO <sub>2</sub>	8.9%
CO	5.4%
H <sub>2</sub>	45.0%
CH <sub>4</sub>	36.0%
C <sub>n</sub> H <sub>m</sub>	1.5%
N <sub>2</sub>	3.2%

High cal. value 5,200 kcal/nm<sup>3</sup>

### C. Refining of Aliphatic Tars.

Methods for refining tar have been developed in Germany for tar from Brown-coal and in Scotland for shale oil. In both cases a crude tar with a high content of paraffin wax and with 10 to 15% of tar acids had to be treated for the production of solid paraffin wax and of oils for illuminating purpose.

For nearly a century the crude tar produced in Rolle-oven-plants has been refined by intermitten distillation in vertical stills. From 1925 an increased production of tar and the necessity of delivering to other consumers, like diesel engines forced to improve these old methods of small capacity and low efficiency

#### a. Fractionating of tar; Extraction of paraffin wax and Creosot oils.

The crude tar from the Rolle-oven may be considered as a tar, which has been mildly cracked under atmospheric pressure during the carbonization. In this way the contents of oxygen compounds and asphalt is considerably lowered as compared with a real low-temperature-tar, the contents of paraffin wax, which resists to such mild cracking, is increased relatively.

The Rolle-oven tar was refined in several smaller plants

The crude tar was intermittently distilled in vertical stills into 3 fractions light-oil, medium-oil and paraffin fraction and a pitch residue. The residue was distilled in a second still to coke for electrodes. Part of it was sold directly for manufacturing purposes in the electric industry. For briquetting of coal or as a road-tar its binding quality was insufficient, unless the residue had been treated with oxygen in the still.

Light-oil and medium-oil were washed with acid and sodium-hydroxide and then distilled for gasoline and diesel oil. The paraffin fraction was separated into paraffin-wax and oil by sweating methods. Creosots and the heavier oils were used as fuel oil. Considerable quantities were shipped for many years to Northern America for creosoting masts and sleepers.

The paraffin wax was the most important product and was exclusively used for candles.

With the development of a market for motor fuels and with the changing of the character of the crude tar, due to other methods of carbonization, these old methods of refining proved itself uneconomic. The market for paraffin wax became saturated and part of the tar had to be treated for the production of gasoline, diesel oil and fuel oil.

Extraction of tar with methanol was introduced by the Riebeck-works. The DEA developed methods of continuous distillation and of cracking of the paraffin-fraction. Cracking of crude tars, which had been tried in several experimental plants could not be considered as a success, as the low contents of hydrogen, as compared with natural oil, allowed only a low yield of light and medium oils and caused high operating cost by an exceedingly high formation of coke in the cracking device. When operated for gasoline a yield of 30% of the crude tar was not exceeded even with the highly paraffinic tars from Brown-coal.

Only in 1944 the cracking of tar was considered again as a method to replace part of the Hydrogenation plants, which had been destroyed by air-raids. The cracking plants however had not been finished before the end of the war.

Cracking of the Paraffin fraction has been operated for many years in the Rositz factory. The paraffin fraction was converted into gasoline and diesel oil. Creosot and heavier oils were treated for navy fuel oil and coke for electrodes.

Until 1935 the crude-tar was freed from ash by an acid treatment and settling in huge tanks, but this expensive method could be eliminated and the refining system could be simplified in connection with the fractionated condensation of the tar products in the newly erected carbonizer-plant Regis. From then the refinery received three separate fractions, crude-benzine, medium oil and heavy tar, all practically free from water and dust, the heavy tar after passing a scraper-centrifuge. The two lower-boiling fractions were also free from paraffin wax.

The production cost of the refinery for gasoline, diesel oil, fuel oil and electrode-coke were considerably reduced in this way.

A plant for the extraction of the medium oils of all the carbonizer plants of the Saxonia district with methanol (Merasolvan) was nearly completed at the end of the war.

b. Cracking of tar and distillation under several atm.

pressure.

While the cracking of a coal tar even in the case of high contents of paraffin wax cannot be considered as a practical method of treating such tars, the treatment of certain fractions, especially of the paraffin fraction from sub-bituminous coals or shale oil by a mild cracking or the distillation under 10 to 20 atm. pressure can be considered as a method for refining tars of a relatively high hydrogen content under condition that a sufficient market for diesel oil and fuel oil is available. When used as a method of viscosity breaking by cracking of the paraffin wax the loss by gasification and the difficulties of coke formation can be sufficiently lowered as required for an economical working. For coal tars it will be advisable to treat only the paraffin fraction by cracking methods. Shale oils with low contents of oxygen and asphalt may be distilled under pressure or even cracked with good results and operating conditions. If only a high yield of gasoline is wanted some high-grade shale oils can be strongly cracked, but all the other tars must be treated by hydrogenation.

In case of a high sulfur content of the crude-tar, hydrogenation is the only method for a sufficient reduction of the sulfur contents of the gasoline and diesel oil fractions.

Catalytic refining of light and medium oil in the vapor phase with an iron-oxide catalyst has been tried successfully in pioneer plants and with better effects of desulphurization than the acid treatment. This method possibly can be very favorably used for gasoline and diesel oil from oil shale.

c. Refining of tar by extraction methods.

A first commercial plant of this method has been erected in Saxonia (Espenhain) for treating 300,000 tons per year of brown coal-tar from LURGI-carbonizers. This plant was delivered by the Edeleanu Company. Tar and medium oils are extracted with chlorinated hydrocarbons and sulfur-dioxide for the production of diesel oil, white paraffin wax, fuel oil and coke for electrodes. The paraffin wax is separated continuously from the solvent by means of belt-filters of special construction.

The crude-benzine from the carbonizers was sent to a hydrogenation plant for refining, because such benzines with 2 to 3% sulfur can be sufficiently desulfurized only by hydrogenation.

As a primary distillation before the extraction could not be

avoided with respect to the quality of the paraffin wax (color), the method has proved as rather complicated and expensive in this case. It could be considerably simplified for the production of diesel oil and fuel oil, if the highest boiling fraction of the tar is condensed separately in the carbonizer plant and treated by hydrogenation. A similar extraction method possible could be most economically used for the shale oil of green-river shale, which is relatively low in its sulfur and asphalt content.

d. Hydrogenation of tar under pressure.

Plants of this type played a most important role in the German system of gasoline production from lignite and sub-bituminous coal. Combined with carbonizer plants it has been proved as the most economical method of utilizing such coals and the cheapest method for the production of gasoline from coal.

Compared with the direct hydrogenation of coal, the hydrogenation of tar is considerably simpler, because those difficulties which result from handling a solid material and from the inorganic substance introduced with the coal-ash, are avoided.

The consumption of hydrogen per ton of gasoline, depending on the hydrogen contents of the crude tar, is considerably lower and for this reason the consumption of steam and energy is also considerably reduced, which results in a corresponding reduction of the investment cost and the labor required for the operation of the plant.

Coal-tar is hydrogenated in a liquid phase, where the catalyst is mixed with the tar and in a two or three stage gas phase, where the catalyst is placed in layers of small briquets.

It seems that the I.G. at the end of the war had succeeded in further improvements for the hydrogenation of tar and heavy oils. The liquid phase has been operated with a fixed catalyst and the gas phase in one stage only. It has been claimed that also the octane number of the gasoline has been improved by this method. 1.3 tons of tar are required for the production of 1 ton of gasoline and 0.12 tons of liquid gas ( $C_3 + C_4$ ) which is used mostly in the bus-traffic.

For 1 m. ton of gasoline are further required:

1600 to 1700 Nm<sup>3</sup> of Hydrogen

1800 KWh

200 m<sup>3</sup> of fresh water

2 to. of steam.

For a plant of a capacity of 200000 m. tons of gasoline per year approximately

75,000,000 RM

have been invested in Germany in 1938, included the production of power but not included the carbonizer plant for the production of the tar. Part of the tar could be produced as by-products of the power station and of the production of hydrogen without an additional investment.

The hydrogenation of tar and tar oils as a refining method until now must be considered as the only method for the production of gasoline from heavy oils and from those oils, which contain a high percentage of oxygen or sulfur compounds. It must be considered as important for any future system of producing gasoline from coal and residue from natural oil.

#### D. Hydrogenation of Coal And Lignite J. G. Bergius Process

This well known method has been described so many times so that it can be referred to literature. \*

The organic matter of coal as liquid hydrocarbons reacts with hydrogen under a pressure of 300 to 800 atm. at 350 to 450 C. to build hydrocarbons with a higher contents of hydrogen as those contained in gas oil and gasoline for motors. The hydrogenation takes place in the liquid phase for coal and heavy oils as well as in the gas phase for lighter oils. In both phases catalysts are used. In the liquid phase the catalyst is mixed with the coal and the heavy oils as a fine powder and in the gas phase it is placed in the reactor as a layer of grains or small briquettes through which the hydrogen and the oil vapors are passed.

The coal is dried and pulverized and then mixed with the catalyst and heavy oils to a paste, which is pressed into the preheater and reactor by a pump. Light hydrocarbons pass directly into the reactors of the vapor phase but the bulk of the primary product is discharged and separated into medium oil - heavy oils and a residue by distillation. The residue is carbonized by means of a special design of rotating drums which are heated from outside the heavy oils are brought back to the liquid phase reactor together with fresh coal. The medium oils go into the three stage gas phase where they are converted into gasoline. Gasoline and the so called liquid gas ( $C_3 + C_4$ ) are condensed from the residual gas by cooling and scrubbing methods, the residual gas is washed with Alcaid liquor for the extraction of hydrogen sulfide. Part of the gas is converted into hydrogen and reconducted to the reaction chambers.

The efficiency of the hydrogenation process depends highly on the character of the coal, not only for the consumption of hydrogen but also for the capacity of the reaction chambers. Coals with less than 2.5% of hydrogen and residues from coke oven tar are unsuitable for hydrogenation because their molecules are too stable. Coals with a maximum of hydrogen and a relatively low contents of oxygen are readily hydrogenated with a minimum consumption of hydrogen gas. In this case (high volatile bituminous coal and brown coal) hydrogenation even can be operated

with a lower pressure (300 to 500 atm.). Caking coals and lignite: need higher pressure for a sufficient reaction speed.

As the consumption of hydrogen is the most important item in the production cost of a hydrogenation plant, the character of the coal therefore is of the highest importance.

Sometimes difficulties have been reported from other components of the coal. The character of the ash caused deposits in the preheaters of the Wesseling plant (Rhine valley lignite) and arsenic compounds caused difficulties of a similar character in the preheaters of the Brux plant. A contents of chlorine may cause very serious difficulties by corrosions.

The Leuna plant was operated with only 300 atm. with the Saxonia brown coal. The Wesseling plant had to be operated with 750 atm. and the plants operated with bituminous coal in the Ruhr and the Uppersilesia coal fields worked with pressures from 600 to 800 atm.

Difficulties with tightening the equipment for these high pressures may be considered as temporary only, they were caused by the lack of some raw materials during the war.

Medium figures for the consumption of hydrogen and energy personally communicated by Dr. Pier are the following:

Coal (net)	1.66 to per	ton of gasoline
Hydrogen	2500 nm <sup>3</sup>	per ton of gasoline
Steam	2 to per	ton of gasoline
Energy	2700 kWh	per ton of gasoline

The total consumption of coal for one ton of gasoline amounts to 5 tons of bituminous coal with 5% ash. It is increased accordingly with younger coals of a lower heating value and higher oxygen contents.

The methods available for the production of hydrogen are described in Chpt. F. In the German plants the Winkler producer has been used for the gasification of lignite char in Saxonia, the normal watergas producer operated with coke has been used in the Ruhr and the Upper Silesia districts. The Pintsch-Hillebrand process has been used for gasifying lignite briquettes. In all the plants the primary gas is cleaned from hydrogen sulfide with iron oxide, then the CO is converted into hydrogen.

The gas is then compressed and washed with water under 10 atm. pressure for the removal of CO<sub>2</sub>. In the Blechhammer plant the CO conversion was operated successfully under 8 atm. pressure. The investment cost of the latest plants erected has been reported with 800 to 1000 RM per ton of the per annum gasoline production.\*

\* Reichsamt f. Wirtschaftsausbau

The treatment of the sludge from the liquid phase by special rotating drums is a very costly process, until now it could not be replaced by a better method. This sludge material is highly caking and sticks to the walls of any retort. For these reasons the drums must be of a heavy construction and have to be provided with heavy steel bars, which scrape the inner walls during the revolution of the drum.

The rest of the hydrogenation machinery has been developed to a high technical standard.

Direct hydrogenation of coal probably will remain the most economical method for the production of gasoline from high volatile bituminous coal with a low ash content. If the coal has a high content of tar, a combination with carbonization, as far as energy steam and hydrogen are required, is favorable for a reduction of the cost of investment and production and for an improvement of the operating conditions.

#### E. Synthesis of Hydrocarbons (Fischer-Tropsch) by hydrogenation of carbon monoxide

##### a) Synthesis under atmospheric pressure.

This process also has been described in the literature  
\* The chemistry of the process has been widely discussed.

The first plants erected in Germany have been operated under atmospheric pressure with a cobalt catalyst and with a 2 : 1 ratio of  $H_2$  and CO. The economical results of these plants caused some disappointments in so far as the primary products of the plants consisted mostly of saturated hydrocarbons with a very low octane number and a considerable part of the primary products had to be cracked for gasoline, which caused an additional loss and additional cost. Plants operated with a gas produced directly from coal or with an additional splitting of coke-oven gas in the water gas plant suffered from a relatively short time of activity of the catalyst due to an insufficient extraction of resinous compounds and organic sulfur.

Experimenting under several atmospheres pressure showed that with a medium pressure the production of primary liquid hydrocarbons per  $nm^3$  of CO /  $H_2$  could be increased from 120 gr to 145 gr and the production of waxes could be considerably increased. These waxes were required for an intended future production of fats and fatty acids.

\* Chemistry of Coal Utilization II



b.) Synthesis under a medium pressure (10 - 20 atm)

A second group of works erected from 1937 to 39 was designed for a pressure of 10 atm. Though the results of these plants were improved concerning the throughput and the efficiency of conversion, the primary products were still of a low octane number or had to be cracked to a considerable extent. The increased production of waxes spoiled the catalyst still more than in the plants working under atmospheric pressure. These difficulties were increased further by the construction of the reactors in which the catalyst had to be placed in vertical tubes, a design which lowered the heat transmission as compared with the design used for atmospheric pressure.

Some operating figures of these plants are the following:

Time of activity of catalyst 6 months

Cost of catalyst 25 -RM/to of products

Liquid primary products 145 gr/nm<sup>3</sup> gas

containing 20% paraffin wax

35% oils

35% benzine (below 200° C.)

Olefins in benzine 15 - 20%

The cost of a plant with a capacity of 40000 to of liquid primary products was 25,000,000 - RM based on the gasification of coke in a water gas plant (1939).

For the production of motor fuels this result could not be considered satisfactory from the standpoint of quality of the benzine as well as the production cost.

From 1938 intensive laboratory and experimental work was going on to improve the result of the existing plants, to develop new catalysts and new reactors suitable for working under pressure. The lack of cobalt during the war was another reason for an intensive experimenting. The necessity of producing a costly gas of a very high hydrogen content if possible should be eliminated by adapting the process to normal watergas.

c.) Medium Pressure Recirculating method of IURGI:

Improvements were expected from recirculating the synthesis gas through the reactor and the condenser due to the decreased reaction time (higher speed) and the lower concentration of hydrocarbons as well as to the improved heat transmission.

A pilot plant erected in the Hoesch-Synthesis plant showed very good results and several plants of the medium pressure type accepted this operating method. The process consists in a

longer activity of the catalyst, higher output per ton of catalyst as well as per  $\text{nm}^3$  of  $\text{CO} + \text{H}_2$  and a much better quality of the primary product. In case of water gas the cost of conversion could be reduced to a certain extent.

Results of this method of operating are the following:

Time of activity of catalyst 8 - 10 month

Cost of catalyst 15 RM/to of products

Liquid primary products 150  $\text{gr}/\text{nm}^3$  -gas

containing 10% paraffin wax

30% oils

60% benzine (to 200° C.)

Olefins in benzine 50 to 60%

Cost of plant is only moderately reduced by this method (3 to 5% per ton of finished products).

Production costs for the finished products are reduced by 10% approximately.

d.) Medium Pressure synthesis with an Iron - catalyst.

In several years of experimenting in the laboratories of the concerned companies, iron catalysts have been found, which operated under 10 atmospheres pressure produce 140 to 145  $\text{gr}$  of liquid primary products per  $\text{nm}^3$  of - gas. These products can be of a paraffinic nature of more olefinic or even alcoholic, according to the composition and the method of manufacture of the catalyst. Most of these catalysts contain copper and an addition of several percent of alkaline increases the production of paraffin wax. The advantages of recircling with respect to the olefin content and the removal of paraffins from the contact exist also with the iron catalyst. The iron catalyst works with a ratio of  $\text{H}_2 : \text{Co}$  of 1 : 1 as well as with the ratio 1 : 2. In the latter case mostly  $\text{CO}_2$  is formed in the synthesis. The reaction temperature of an iron catalyst is slightly increased (220 to 235° C.).

Investment and production cost of a plant of this kind probably will not be essentially lowered as compared with a cobalt catalyst plant. The various methods as used for the production of synthesis gas or available for new plants are described in Chpt. F.

As in any other process of producing hydrocarbons from coal the production of by-products with the production of the synthesis gas is of a great influence on the production cost of the gas or of the total production of motor fuels. The value of such by-products may also be recognized in the fact that the German government (Reichsamt) did not allow to build new power plants for a

coal with more than 6% tar content on dry base, unless the tar was recovered.

A combination of by-product gasification and synthesis based on a coal or subbituminous coal with a high tar content seems to be the method of lowest cost for the production of motor fuel and a method to meet all the quality requirements of the total consumption of motor fuels. In some cases of a cheap high volatile coal the value of the by-products can cover nearly the total production cost of the synthesis gas and the production cost of the synthesis products is reduced correspondingly by nearly 50% as compared with a gasifying method, in which by-products are not recovered.

In order to give a rough idea of the influence of by-products on the cost of the synthesis gas fig. 9 shows the various items of production cost of the gas for a high volatile subbituminous coal as those of central Germany.

#### e. The principal types of catalyst reactors.

It has been proved as very essential to keep the temperature of the catalyst constant within a very short interval. A decrease of this temperature reduces the reaction speed and a high temperature results in the formation of a considerable quantity of methane. As the reaction itself is highly exothermic, the transmission of the reaction heat to a cooling medium is one of the fundamental problems of the synthesis and of the construction of reactors. This problem has been satisfactorily solved for the synthesis under atmospheric pressure. The catalyst is placed between vertical sheets of iron 1 mm thick, which are mounted on horizontal pipes with a distance of 8 to 10 mm between the sheets. Inside the pipes water is circulated. The reaction heat is used to make steam of 20 to 25 atm. pressure, which is collected in a steam collector mounted above the reactor.

This reactor has become a standard design for the synthesis under atmospheric pressure, it is 4.8 m long 1.8 m wide and 2.7 m high. It contains 10 m<sup>3</sup> of the catalyst and can treat 1000 to 150 Nm<sup>3</sup>/hour of gas in the first stage.

For the synthesis under a medium pressure as 10 atm. this design could not be used, because the straight walls cannot be built strong enough with a reasonable cost. The medium pressure plants used cylindrical vertical reactors the middle part of which is similar to a tubular cooler and the catalyst is placed in the vertical tubes. Some variations have been experimented, varying the diameter of the tubes. Also double tubes forming a ring space for the catalyst have been tried. Various types of spirals and crossed iron bars have been placed into the tubes of a relatively larger diameter, with the purpose to increase the heat transmission from the catalyst to the tubes, and to find a maximum ratio of catalyst space to the total volume of the reactor. Only the single tube of 15 to 18 mm and the double tube with a ring space of 8 to 10 mm have proved satisfactory for a good heat transmission and

equal temperature of the catalyst. All the other varieties could not satisfy, most because the removal of the spoiled catalyst caused very great difficulties.

The relatively small volume of the catalyst in this design of reactors increases the cost of the reactors compared with those of an atmospheric pressure plant considerably. This fact becomes even more important when an iron catalyst is used, because in this case the temperature of the reaction must be increased from 200 to 230 C. and the outer cylinder of the reactor has to stand against 30 to 40 atmospheres pressure.

It has been proposed to put a reactor of a similar design at the rectangular reactor for atmospheric pressure inside of a cylinder, which must resist to the gas or reaction pressure only. In this case the high pressure of the cooling fluid is limited to the cooling pipes and the reaction temperature can be increased to 270 centigrade even without an additional cost.

## F. Production of CO + H<sub>2</sub> mixtures from coal.

### a. Watergas process for lump-coke.

In this well-known method coke is gasified by means of superheated steam in a cylindrical shaft provided with a rotating grate for the extraction of the ash. The fuel bed is periodically heated up by combustion air, and then superheated steam is introduced, which is decomposed into hydrogen and carbon monoxide at the surface of the hot coke. The heat stored in the fuel bed being rapidly consumed by the endothermic reaction of gasification, the periods of introducing air and steam must be changed in short intervals. The hot glue gas of the combustion period, which also contains CO, is used to heat a regenerator and then a waste-heat boiler. The steam for gasification is preheated in this regenerator before entering into the fuel bed. This method, originally developed for producing gas for public supply in gas works, has been improved for industrial use in England and Germany and has been used to a great extent for the production of hydrogen for the synthesis of fertilizers and for the synthesis of hydrocarbons and the hydrogenation of tar and coal.

These plants have proved to be very satisfactory for a safe and nearly automatic operation in units of a relatively great capacity. The production of the gas, however, is expensive because it needs a solid lumpy coke and because the heat recovered in the gas with this method is only 60% of the coke consumed. Other fuels have been tried. One plant in the Ruhr district is operated with low temperature coke from a carbonizer plant, and a big watergas plant in Upper Silesia was operated with carbonized egg-shaped briquettes. Though this method widens the field of watergas plants for other fuels than coke from caking low volatile coal, the production cost could not be essentially lowered. It could be lower compared with gasifying a coke from coke ovens, if a very cheap slack-coal or a cheap sub-bituminous coal can be converted into

a lumpy coke by cheap briquetting and carbonizing methods.

For the reason that in Central Germany cheap fuels are available which cannot be gasified in a watergas producer, new methods had to be developed which could gasify lignite-briquettes or dried lignite of a small size or even pulverized lignite and lignite-coal.

Several plants using lignite briquettes were erected at the Ruland Synthetic Oil factory.

b. The Didier-process of gasification uses a continuously operated vertical retort for the gasification with steam. In the upper part of the retort the coal is carbonized, the gases of this zone are taken off downwards in the middle of the retort, so that oil vapors and hydrocarbons are cracked in the retort. Part of the carbon is discharged from the retort with the ash and gasified in a separate gas producer, which makes the low grade gas required for the heating of the retort. Nearly 50% of the fixed carbon is required for the heating.

An industrial plant of this system has been erected for the production of gas for the Fischer synthesis at Ruland. It was operated with lignite briquettes for many years. The quality of the gas meets the requirements of the Fischer process, but the process has proved relatively expensive for investment and for production cost compared with other methods.

c. The two variations of the Hillebrand method of gasifying with a circulating stream of a superheated mixture of gas and steam, the Pintsch-Hillebrand plant and the Koppers System have been described in Chapter B.

Due to the high velocity of the gas within the fuel bed, which is necessary if a high gasification rate shall be reached, these methods also require a high grade briquette or a lumpy non-disintegrating, non-caking coal. The coal must be relatively reactive with a relatively high softening point of its ash. The heat-efficiency of this process is only 55 to 60%, so its use has been limited to good quality lignite briquettes.

d. On a similar base of chemical reaction the Wintershall-Schmalfeld process has been developed. In this case briquetting of the lignite is avoided. The dried lignite, which is disintegrated by the drying process to a size practically between 0 and 2 mm, is gasified in suspension in the stream of heated gas and steam.

This method equally uses a pair of regenerators to heat the reacting gas by the waste heat of the reaction gas, but naturally the quantity of ash carried away with the gas into the regenerator is still greater in this case, compared with the preceding method. Gasification in suspension requires a relatively high temperature to complete the reaction, so the danger of fused ash, which destroys the brickwork of the regenerators and of the reaction chamber, is still greater in this case and the uncompleted reaction results in

a loss of carbon and in a relatively high content of  $\text{CO}_2$ . This original method of the plant has proved a failure. Finally, it could be considerably improved by the use of oxygen in the reaction chamber. By this means it is possible to keep the temperature below the softening point of the ash and still to gasify with a temperature, which is high enough for a complete combustion of the carbon and a good reduction of carbon dioxide. No figures are available from this improved plant, but from other experiments and from the theoretical equilibrium it can be expected that a raw gas of 12-15%  $\text{CO}_2$ , 40-45%  $\text{CO}$  and 45 to 50%  $\text{H}_2$  can be produced with less than 3% of  $\text{CH}_4$  +  $\text{N}$ . This gas is of a high quality for the synthesis of hydrocarbons, if the  $\text{CO}_2$  contents are eliminated, but it requires compression and washing with water, even when it shall be used under atmospheric pressure.

A recovery of tar oils is not possible in combination with this process. It can be expected that any slack coal, even in case of high caking properties and a low softening point of the ash, can be gasified with the improved method of introducing oxygen into the reaction chamber. A moderate pulverizing of the coal will be necessary in general.

e. The Winkler-gasproducer, specially designed for the gasification of German brown coal and the fine char produced by carbonization of this fuel, gasifies the coal in a relatively low layer of fuel (about 40 inches), which is kept in a turbulent movement by the high velocity of the steam and oxygen introduced from below. Smaller particles are carried away with the gas and are further gasified in the upper part of the producer. In the latest design of this process a cylindrical producer of 15 ft. of inner diameter and 75 ft. high has been used. On the bottom of the cylinder a scraper is rotated mechanically, which keeps the layer of coal moving and brings the ash to a screw conveyer which is connected to the ash receiver. Only a small amount of the ash comes out at the bottom; most of it is carried off with the gas. Oxygen and steam are introduced through the wall in two rows of jets, the lower several inches above the scraper, the higher immediately above the fuel bed. A considerable amount of the fine coal being carried off with the gas, it is necessary to screen the fuel to 2 to 10 mm size. This probably is also required for a better control of the turbulent movement of the fuel bed. The gas, which leaves the producer with about 1000° C. is conducted through a waste-heat boiler to a multiclone, a cooler and a scrubber. It is then treated by iron oxide, compressed, and washed with water for the elimination of  $\text{CO}_2$ .

When gasifying a lignite char (Grudekoks) of 3 to 5 mm size with 30% ash from a LURGI Carbonizer plant 300 m. tons of char could be gasified per unit and per day. 42 to 45% of the fuel is carried on with the gas and separated to some 85% in a dry state from the gas, the rest by washing with water. The dust contains roughly 50% of carbon and may be used in a pulverized fired boiler without a further grinding only 10% of the introduced ash is extracted at the bottom, this ash containing 25% of carbon.

From the high amount of sensible heat carried off by the raw gas and from the high proportion of carbon separated in the cleaning device, it can easily be recognized that the efficiency of gasification of this process is very low and that a good reactivity of the fuel is essential for this method. The amount of  $\text{CO}_2$  must be high in the gas, so that a use for chemical reactions is bound to compression and washing with water.

A typical analysis of the gas as normally produced from char for hydrogenation plants is the following:

$\text{CO}_2$	1.0%
$\text{CO}$	38.0%
$\text{H}_2$	59.2%
$\text{CH}_4$	0.8%

The oxygen consumption also is relatively high with this process and amounts to 300 to 320 nm<sup>3</sup> per 1000 nm<sup>3</sup> of the above gas. The steam consumption practically can be covered by waste heat. Only 65 to 70% of the carbon used is gasified.

If a cheap low volatile coal or a cheap reactive charcoal is available, the Winkler producer may be most suitable for the production of a high hydrogen gas, favored by the high capacity of a unit and low labor cost for its operation.

f. Recently, gasification of pulverized coal by oxygen and steam has been developed and tried in an experimental plant by the Koppers Co. at Essen. A vertical cylindrical reaction chamber is used, which is subdivided by several eccentric narrows, producing in this way a turbulent rotating movement of the gas and the pulverized fuel. Oxygen, superheated steam, and fuel can be introduced in several levels of the reaction chamber. A secondary reaction chamber is provided to complete the reaction. A hot coke filter protects the plant against explosions from oxygen entering through the reaction chamber in case of a lack of fuel. The heat of the raw gas is utilized in a waste-heat boiler. Then the gas is washed in a scrubber. Steam and oxygen are superheated in a pair of regenerators, which are gasfired so that ash in these generators is avoided.

The temperature in the reaction chamber is kept below the fusion point of the ash.

It is claimed by the Koppers Co. from calculations that in a unit of 900,000 cu. ft. capacity per hour, when gasifying a bituminous coal of 8.8% ash and 20.5% volatiles on dry basis, a gas with the following analysis and consumption of material is produced: \*

\*According to L. L. Newman.

CO <sub>2</sub>	%	11.0
CO	"	54.0
H <sub>2</sub>	"	34.0
N <sub>2</sub>	"	1.0
Coal dust used		27.6 lbs p. 1000 cu. ft.
Oxygen used		270.0 cu. ft. p. 1000 cu. ft.
Steam at 2 atm		11.1 lbs p. 1000 cu. ft.

Even if these figures may be considered optimistic with regard to the contents of CO<sub>2</sub> and the rate of gasification of the carbon and possibly also for the oxygen consumption, the Koppers process may prove more efficient than similar methods of gasifying in suspension, like the Winkler and the Wintershall Schmalfeld-process, and more economical than the primary Koppers equipment for the Hillebrand method, because any pulverized cheap fuel can be gasified with a relatively high temperature. The heat efficiency of this process probably will be lowered to a certain degree if a higher content of H<sub>2</sub> is required, as for hydrogenation purpose.

As in all the processes of the group, it requires compression and washing of the gas for any further use, except combustion, in order to eliminate part of the CO<sub>2</sub> content. By-products cannot be recovered with this method.

g. Slagging gas producers have been operated with oxygen and with a lumpy coke in the Leuna works on an industrial scale and also in the Ruhr district in large scale experiments with the Thyssen-Galocsy producer.

In the Leuna works a shaft producer of a relatively low height was blown by one set of nozzles only, the temperature or ductility of the ash was controlled by an addition of limestone or by recirculating part of the slag.

The Leuna gas producer was operated with several kinds of fuels, even with an ash content of 50% and with steam as well as with CO<sub>2</sub>. When operated with steam a gas containing

6% CO<sub>2</sub>, 62% CO  
31% H<sub>2</sub>            1% N<sub>2</sub>

was produced, with an oxygen consumption of 270 cu. ft. per 1000 cu. ft. of CO + H<sub>2</sub> gas, \* and a steam consumption of 18 lbs. capacity per unit 500,00 cu ft per hour.

Operated with CO<sub>2</sub> for the Production of CO Gas, a gas of the following composition was produced:



3.0 % CO<sub>2</sub>

92.5 % CO

3.0 % H<sub>2</sub>

0.0 % CH<sub>4</sub>

1.5 % N<sub>2</sub>

O<sub>2</sub> consumption 310 cu ft per 1000 cu ft of CO + H<sub>2</sub>.

CO<sub>2</sub> consumption 258 cu ft per 1000 cu ft of CO + H<sub>2</sub>.

The Thyssen-Galocsy Producer was operated with a very high fuel bed according to the theory that by reintroducing gas into the lower part of the fuel bed together with the oxygen for its combustion, the reaction zone could be enlarged and a better reduction of CO<sub>2</sub> could be reached. Results of this plant do not differ essentially from the results of the Leuna producer.

The slagging producer of Leuna Works may be considered as a good solution for the problem of producing a gas rich in CO or even a nearly pure CO gas, if CO<sub>2</sub> is available from another process. For a gas as required for the synthesis of hydrocarbons, the H<sub>2</sub> contents might be adjusted accordingly by a conversion of part of the gas into H<sub>2</sub>. If the same grade of coke is used as suitable for a normal watergas plant, the latter seems to be superior with regard to production cost, due to the relatively high consumption of oxygen in the slagging-method.

Probably for such reasons the Leuna slagging producer has not been adopted for other works of the I. G. Concern, as e.g., for the Auschwitz Works erected 1942-44. In this works gasification with oxygen and steam in a cylindric producer equipped with a Brassert A. B. C. grate was adapted for the production of a gas for the synthesis of methyl alcohol.

h. Gasification with oxygen and steam in a rotating grate gas producer has also been used for the production of hydrogen for a fertilizer plant at Meran (Tirol). The producers were delivered by the Viag Co. of Vienna.

Operating figures are not available from these plants. In the Auschwitz plant coke from a non-caking bituminous coal with 3% volatiles produced in a Lurgi carbonizer plant was used in the size of 1/2 to 1 1/2 inch and with a throughput of 60 to 80 tons of coke per day and per gas producer.

The quality of the gas depends on the proportion of oxygen to steam, which is limited by the softening point of the ash. In case of a high softening point of the ash and using coke, the raw gas (calculated from other experience) should be composed as follows

CO <sub>2</sub>	%	14.8	24.8
CO	%	42.0	24.0
H <sub>2</sub>	%	42.0	50.0
CH <sub>4</sub>	%	0.2	0.2
N <sub>2</sub>	%	1.0	1.0

Approximate consumption per 1000 cu ft of (H<sub>2</sub> + CO):

O <sub>2</sub> cu ft	250	230
Superheated Steam	lbs 40-45	60-70

When using a non-caking bituminous coal, the raw gas contains 3 to 5% hydrocarbons (mostly CH<sub>4</sub>) and the consumption of oxygen and steam per 1000 cu ft may be 5 to 10% lower.

This method of producing mixtures of hydrogen and carbon monoxide may be most economically used for the gasification of a small-sized coke, if a gas with a medium or a high hydrogen content is needed for a synthesis under several atmospheres pressure. In this case the elimination of CO<sub>2</sub>, which is unavoidable, makes little additional cost and may be equalized by the high thermal efficiency and a relatively cheap investment and operating cost.

1. Combined Carbonization-Gasification with Oxygen as developed from the LURGI Carbonizer has been described in Chapter. Moist and high ash coals can be gasified with this method to a gas of a low content of CH<sub>4</sub> and with a high yield of by-products.

For the production of gases of a high content of H<sub>2</sub> + CO the carbonization zone and the gasification zone are operated with oxygen. The surplus gas of the carbonization zone which contains nearly all the sulfur is cracked whereby the CH<sub>4</sub> is converted into CO and H<sub>2</sub>, the resins and organic sulfur are destroyed or converted into H<sub>2</sub>S. In this way most of the sulfur is concentrated in a small amount of gas, and can be recovered with a relatively small cost.

The gas produced in the gasification zone, which amounts to 80% of the total gas and contains only traces of sulfur is purified only by a waterwash and the alkalic iron oxide placed before the catalysts.

In case of hydrogenation an extraction of sulfur is not required for the gas from the gasification zone.

The ratio of CO to H<sub>2</sub> can be altered in a wide range from 2:1 to 1:2, if necessary by recycling CO<sub>2</sub> from the waterwash into the gasification zone.

In some cases the carbonization zone can be operated with air and the gas be used for the production of steam and energy. This method is very economical for coals of a high tar content. It allows recovery of a maximum of tar and a very reduced investment cost for the production of steam and energy.

When gasifying a subbituminous coal with 21% moisture, 4% ash 7.2% tar and with a heating value of 5220 Kcal/kg. The following gas is produced from 1 kg of coal:

	Carbonizer gas			Producer gas	
	Raw	Split	Washed	Raw	Washed
Nm <sup>3</sup> /kg	0.18	0.21	0.18	1.2	1.07
CO <sub>2</sub>	18.0	14.0	2.0	12.8	2
C <sub>n</sub> H <sub>m</sub>	0.9	-	-	-	-
CO	27.9	37.9	43.2	56.2	62.8
H <sub>2</sub>	36.5	44.8	51.2	28.5	31.8
CH <sub>4</sub>	15.5	2.0	2.2	0.8	0.9
N <sub>2</sub>	1.0	1.3	1.4	1.7	1.6
Oxygen Nm <sup>3</sup> /kg	0.03	0.05	0.05	0.3	0.3
Steam kg/kg required	-	-	-	0.5	0.5

The capacity of a unit depends on the size of the coal and the fusion point of the ash normally 250 - 300 tons of subbituminous coal 1/2 to 2" size or 200 to 250 tons 1/8 to 1/2" can be gasified per unit and day.

j. The LURGI-pressure gasification process, as developed for the production of city-gas also was described in Chapter B. The most important advantages of this method consist in the possibility of gasifying a small-sized raw coal with a good yield of by-products with a low consumption of oxygen and without the necessity of compressing the gas for the elimination of CO<sub>2</sub> and for utilization under pressure.

This process has been adapted to the requirements of the production of a gas mixture of H<sub>2</sub> and CO. When carbon or coke is gasified under pressure of 15 atm with oxygen and considerable amounts of steam formation of 4 to 5% of CH<sub>4</sub> cannot be avoided practically. By the volatiles of a raw coal this figure may be increased to 8 to 10 per cent in the CO<sub>2</sub> free gas. For synthetic purpose and also for the hydrogenation of coal and tar such a high content of CH<sub>4</sub> not only reduces the output of active gas for the synthesis but it lowers also the absolute pressure of CO + H<sub>2</sub>

and causes an additional loss of these gases because the reaction practically cannot be continued below a certain concentration (30% of  $\text{CO} + \text{H}_2$ ). The contents of  $\text{CH}_4$ , which is increased in the residual gas to 50%, could be utilized by a splitting of the  $\text{CH}_4$  into  $\text{CO} + \text{H}_2$ .

The  $\text{CH}_4$  content could also be reduced by splitting  $\text{CH}_4$  in the raw gas under pressure. After a thorough investigation by experiments and calculations, it has been found that the production of a raw gas with a high  $\text{CH}_4$  content and splitting of the  $\text{CH}_4$  under pressure is the more economical method. It allows the production of a gas of a very high concentration of  $\text{CO} + \text{H}_2$  and the  $\text{CO}_2$  content of the raw gas facilitates splitting and increases the  $\text{CO}$  content which is limited in the raw producer gas by the softening point of the coal-ash. This process may be considered as a gasification with oxygen in two stages, the first comprising carbonization-gasification of the solid fuel to a maximum of  $\text{CH}_4$  and with a minimum of  $\text{O}_2$  and the second consists in splitting the hydrocarbons into  $\text{CO}$  and  $\text{H}_2$  by a further addition of oxygen. In this second stage resins and organic sulfur are perfectly destroyed. The  $\text{CO}$  content of the gas can be regulated by operating part of the gas producers with  $\text{CO}_2$  and oxygen or by introducing  $\text{CO}_2$  from the water wash into the splitting chamber.

The process as developed for the production of  $\text{CO} + \text{H}_2$  mixture is shown in fig. 10. It shows also the principal figures of operation with a subbituminous coal of a medium tar content. A production of 4.0 to/h of primary products of the synthesis is combined with a production 3.0 to/h of high-grade tar oils and high octane benzine without an additional cost. Synthesis gas is of a very high concentration of  $\text{CO} + \text{H}_2$ . The high cost of oxygen is equalized by the savings for a compression of the raw gas, which would be required in case of producing the gas under atmospheric pressure.

The cooling and cleaning equipment and the pipelines required for an operating pressure of 20 atm. are considerably cheaper than those required for the same quantity of gas treated at atm. pressure.

#### k. Relations between the properties of solid fuels and the special features of the various processes of gasification.

Except from the gasification of a hard lumpy coke in an intermittently working water-gas plant, which still has considerable advantages if a solid coke is available at a low price all the other proposals and pioneer plants finally ended in adopting pure oxygen for the reactions at least as a means to overcome difficulties caused by the fusing of ash. Nearly all the methods used in the latest plants built in Europe, were based on an exclusive use of oxygen as gasification agent in a continuously working gas producer.

The characteristic features of the various methods can be seen from the following table:

Process	Oxygen	Fuel	Quality of Gas			By-Products	Remarks
			CO <sub>2</sub>	CO	H <sub>2</sub>		
			%	%	%		
Watergas-Producer	No	Lumpy coke	Low 5	40	48	No	Expensive fuel. Low efficiency. No oxygen.
A.B.C. Contin. Producer	Yes	Small-size coke or anthracite	High 15-20 2 (washed)	40-25 46-33	40-50 46-66	No	By product in a separate carbonizer
Slagging Producer (Leuna)	Yes	Lumpy coke	Low 6 3	62 92.5	31 3	No	With steam With CO <sub>2</sub> High ash fuels
Carbonizer-Gas producer (LURGI)	Yes	Lumps and smalls All non-caking coals	High 13-25 W.2	50-25 60-33	30-50 35-66	Yes	Also moist high ash fuels
Winkler Producer	Yes	Smalls of coke Low vol. coal	High 24 W.2	29 37	44 56	No	Low efficiency. High oxygen cont. By-prod. in separ. carbonizer
Pulverized Coal-process (Koppers) (W.S.)	Yes	Any pulverized coal incl. caking	Medium 11-15 W.2	54-40 60-46	34-45 37-52	No	
Pressure Gasification (LURGI)	Yes	Screened slack smalls of non-caking coals and lignite	High 30-32 W.2	45-30	52-66	Yes	Gas production under pressure. Splitting of CH <sub>4</sub>

The table shows that each of the gas producing methods has special features with regard to the fuel quality required, the possibility of recovering tar oils from a bituminous coal, the possibility of producing high CO contents or high H<sub>2</sub> contents and the necessity of eliminating CO<sub>2</sub>.

Production of gas with a high content of H<sub>2</sub> necessarily requires elimination of CO<sub>2</sub>.

A primary gas with a high CO content and a low CO<sub>2</sub> content can only be produced by the water-gas method and with the slagging producer, but in both cases an expensive coke is required.

A gas with a high CO content also can be produced with the pulverized coal methods, but the CO<sub>2</sub> content of the gas is so high that an elimination of CO<sub>2</sub> possibly cannot be avoided. Any coal slack including caking coal can be used, but no by-products can be recovered.

Carbonization-Gasification with oxygen is the method suitable for gasifying coals with a high tar content, even fuels which have a high moisture content (35%) and a high content of ash.

Gasification under pressure allows gasifying even small-sized coal with a high throughput and with a good oil recovery. It seems most economical when a gas with a high hydrogen content is required. It allows producing the gas under a pressure of 20 atm. It is very economical in connection with gas-turbines for the production of power.

In the next table the character of the coal is taken as a basis for classifying the gasification methods:

TABLE  
C O A L

Low Ash Content			High Ash Content	
Pulverized C. Proc. Pressure-Gas A.B.C. Producer	Slack Smalls Lumps	ANTHRACITE	Slack Smalls Lumps	Pulverized C. Proc. ? Carboniz.-Gasif. Slagging Producer
Pulverized C. Proc. Watergas Proc. A.B.C. Producer Slagg. Producer	Slack Coke	CAKING BITUM. COAL	Slack Lumps	Pulverized C. Proc.
Pulverized C. Prod. Pressure Gas. Carboniz-Gasific. A.B.C. Producer	Slack Smalls Lumps Coke	NON-CAKING BITUM. COAL	Slack Smalls Lumps	Pulverized C. Proc. ? Carboniz.-Gasif. Slagging-Prod. ?
Pulverized C. Prod. Pressure-Gas Carboniz.-Gasific. Winkler-Producer	Slack Smalls Lumps Coke	SUBBIT.-COAL and LIGNITE	Slack Smalls Lumps	Pulverized C. Proc. ? Carboniz.-Gasification

For several of the methods the application is limited to special fuels, mostly coke. Gasification of pulverized coal covers the whole range of solid fuels, if the ash content is not too high. For non-caking coals with a high tar content the recovery of by-products may be deciding in favor of pressure gasification or combined gasification carbonization. The latter is very suitable also for high moisture and high ash fuels.

#### G. Purification of Gas and Recovery of By-products

Except for use of producer gas in metallurgical factories where the gas is often burned hot in a furnace connected directly with the producer plant, gases from coal must be cleaned for their further use. Dust, tar, and water vapor have to be removed to avoid difficulties with pipelines, valves, instruments and burners. Some gaseous compounds have to be removed because they would disturb the chemical reactions for which the gas shall be used. In general, a maximum concentration of  $H_2 + CO$  is required. Sulfur compounds and such compounds which might form a solid residue are dangerous for the constructing material or spoil the catalysts used for the reactions.

##### 1. Removal of dust and tar.

For the removal of dust, Cyclones or Electrostatic Precipitators must be used when the dust shall be obtained dry or the gas shall not be cooled. Cyclones only take heavier grains; electrical precipitators can be built for a very high purification rate. If the solid particles must be removed completely washing with water in a scrubber or with a disintegrator usually is sufficient to protect the following stage of the process, mostly the oxide boxes, against dust. If the raw gas contains only very little dust, washing with water alone is sufficient.

A gas which contains tar and dust at the same time may be very difficult to handle unless either the quantity of dust per cu. ft. or the quantity of tar is very low. As a matter of fact, the problem of handling a gas with a high content of tar and a high content of dust has not been solved. In this case the dust must be taken out above  $400^{\circ}C$  by cyclones, but mostly the temperature of the gases when leaving the carbonizer or the gas producer is not high enough. Sometimes the dust contained in the gas is so fine that it does not separate in a cyclone, as e. g. with rotary kilns. Many systems of carbonization or gasification had to be given up because of difficulties with treating the gases. If the temperature of the gas goes below  $350^{\circ}C$  the heavy tar is condensed at the walls of the pipes and coolers and forms solid layers together with the dust, which block the pipes within a short time. Mechanically such a gas could be handled by a disintegrator, but the tar oils recovered in this way are considerably spoiled or even worthless because of a considerable content of dust and the polymerization which takes place during the pumping and heating of the tar. If operated below the dew-point of the gas the tar is still more spoiled by emulsion

of tar, dust and water. It is impossible to utilize such a tar by refining methods.

Gasification and carbonization of fines or of a pulverized coal in a suspension, which has often been proposed and sometimes experimented on a big scale, may finally be proved impossible, unless the tar oils are completely split into permanent gases.

Normally the gases from carbonization or by-product gasification contain high amounts of tar, from 50 to 300 grams/m<sup>3</sup> of gas 5 to 30 grams of coal dust, the dust being relatively heavy. The temperature of the gas is between 100 and 400°C. In order to separate as much of the dust as possible before condensing the tar, the hot gas is guided through a wide gas chamber, in which the direction of the gas is turned at the bottom of the chamber with a low speed, from downwards to upwards. Such chambers settle most of the dust without condensing much of the tar. They often are provided with a water-seal, through which the dust can be extracted periodically. Then the gas enters an electrostatic precipitator, after its temperature has been brought near to the dew point of the gas by spraying water in it. Normally this temperature is kept between 120 and 80°C in order to condense all the asphaltic compounds and the paraffin wax completely in this fraction without condensing any water. Then the gas is cooled in surface coolers to 25°C or less (practically 3 to 5°C above the water temperature). It is then washed with oil in an oil scrubber for the recovery of light oils.

This system of condensation has proved very satisfactory for the considerable improvement with regard to the quality of the tar oils and the facilities in refining reached by this roughly fractionating system. The heavy oils are free from water, which would cause difficulties in the distillation. The medium oils condensed in the cooler easily separate from water. They are free from asphalt, paraffin wax and insoluble residues and can easily be refined for gas oil. Operating cost of this system also has proved very low as compared with disintegrators or only cooling of the gas, because the consumption of energy of the electrostatic precipitator is very low and the coolers are kept absolutely clean and can be calculated with a much higher heat transmission.

2. For the LURGI carbonizer plants erected for bituminous coal in Upper Silesia another method of a fractionating condensation has been developed, with the purpose of immediately producing a pitch with 65 to 70°C melting point and a fuel oil for the Navy with a low viscosity and a pour point below 0°C. In this case the effect of an electrical precipitator is reduced by the high viscosity of pitch and possibly separating of dust and asphalt on the surface of the precipitator. The pitch fraction is separated at a temperature of 130 to 150°C by forcing the gas through a number of nozzles with a speed of 20 to 30 m/sec., the direction of the gas being turned at the point of highest speed. The nozzles are permanently washed by the pitch. The fuel oil is then condensed in the surface cooler and it may be regulated in



its flash point, if necessary, by scrubbing the hot gas with this oil after the pitch condenser.

3. For the recovery of benzine, which in general is economical with carbonizer gas, the old method of scrubbing with wash oil is used. New types of scrubbers have been developed in order to replace the very voluminous towers used for coke ovens. Compared with coke oven gas, the gas of directly heated shaft carbonizers or gas producers has a lower concentration of light oils and needs less oil per cbm of gas. An equal distribution of the oil over the whole cross-section of large scrubbers is difficult under this condition.

Three types of oil scrubbers are installed in the large carbonizer plants built in Germany.

The Walther Feld spray washer consists of a cylindrical tower of 10 to 13 feet diameter and 33 feet high, which is equipped with a vertical rotating shaft in its center. Conical rings are mounted on this shaft. They are provided with holes or openings which spray the oil in 6 or 8 stages into the gas, which slowly moves upwards in the tower. This equipment is relatively cheap but it needs much electrical energy if a good recovery of the light oils is wanted.

A washer proposed by Weindel consists of a tower 65 to 80 feet high equipped with 10 to 12 layers of Raschig rings, each layer 5 feet thick. Between the layers of rings flat basins are mounted on a central rotating shaft. This device procured an equal distribution of the wash oil through all the layers of the tower. This equipment is more expensive but it needs very little power and supervision and has a good washing effect.

A third type of a washer called Stufenwascher consists of a large tower 28 to 33 feet high and 33 feet of diameter, the cross section divided into 8 to 10 vertical towers, each with 3 to 4 layers of Raschig rings or equipped with layers of wood. The gas flows through each of the vertical sections successively and is washed by the oil, which passes the sections by means of a corresponding number of circulating pumps in counter current to the gas. In the middle sections the washing is intensified by pumping 5 to 6 times the quantity of oil over these sections as compared with the fresh wash oil delivered to the first section of the washer. This system has also proved very efficient.

Normally an improved washer of the Weindel system, with a distributing device somewhat simplified, seems to be the best equipment.

For gases of a higher concentration of the light oils, as for outside heated carbonizers and for pressure gasification, a single tower of the coke oven plant type has been used with good results.

For the reactivation of the wash oil by distillation, steam heated equipment as well as gas fired heaters have been used,

The wash oil is taken from the medium oils produced in the same carbonizer plant. In general these oils have the best washing effect for the benzines produced the same coal. They are fractionated to a boiling range of 220 to 320 °C in a small separate column.

The wash oil is saturated with 2 to 3% of benzine in the scrubber depending on the primary concentration in the gas. In the pressure gasification process 5 to 6% saturation is usual. In the distillation plant the oil is topped to 0.1% of benzine below 180 °C (Englertest). The equipment of heat exchangers, coolers and of the column is the usual of oil distilling plants

In plants where the hydrogen sulfide has been taken out by iron oxide before the benzine, activated charcoal can be used with very good results. This method is of a great advantage in the case of using the gas for a Fischer synthesis, because the activated charcoal also extracts the resinous and part of the organic sulfur compounds of the gas.

With the wash oil plant normally 2 to 3 grams of benzine boiling above 50 °C are left in one cbm of gas. With an activated charcoal plant the recovery is complete.

4. For the Extraction of Hydrogen Sulfide various methods have been used.

In general an intense purification as required for the synthesis of hydrocarbons needs iron oxide at least for the last traces of H<sub>2</sub>S, but liquid methods of purification have often been used as a primary stage for the recovery of sulfur from a gas with a relatively high sulfur content. In case of utilizing the gas for boiler firing such liquid methods are sufficient and more economical. For a hydrogenation plant they may be sufficient if in the CO conversion plant a catalyst is used, which allows traces of hydrogen sulfide as e. g. 20 to 30 gramm of sulfur per 100 cbm, which seems to be the practical maximum of purification for a liquid method.

Iron oxide, mostly used in the form of LUXmasse (from aluminum factories) and improved in its physical structure by an addition of some natural ore, was used in smaller plants in the box system; in plants of a higher capacity the tower system was usual, which provides mechanical facilities for the changing of the containers which carry the iron oxide in the tower.

For the purification of gases under several atmospheres pressure, special towers have been developed which allow a quick removal and a good tightening of the cover of the tower.

In case of part of the hydrogen-sulfide being removed by a liquid method or by a water wash under pressure, the iron-oxide equipment may be reduced to a 3 or 2 stage passing only.

5. The liquid method, known as the ALKACID process has been used on a big scale for the recovery of sulfur from gases of the LURGI-carbonizer plants erected in Saxony. It has also been used in hydrogenation plants for the purification of the residual gas of hydrogenation.

In the alkacid-process a special solvent called alkacidlauge is used, which was delivered by the Leuna works. The cost of this solvent being relatively high, it was very essential to avoid consumption of the solvent by certain compounds of the gas as Cyan compounds,  $\text{CO}_2$ ,  $\text{O}_2$ , and  $\text{SO}_2$ . Difficulties from corrosion and too high consumption of solvents have been overcome by a primary wash of the gas with a potash solution. Finally the Alkacid process was considered as the most economical method for removing 80 to 90% of the sulfur from gases with a high content of hydrogen sulfide.

6. A plant for washing with a Potash solution has been delivered by the Koppers Co. for a LURGI carbonizer-plant but here also difficulties arose with corrosion and unexpected high consumption of the solution, so that this method was not considered as more economical than the alkacid method working under the same conditions.

In Pressure gasification plants the  $\text{H}_2\text{S}$  is removed in the most simple way together with part of the  $\text{CO}_2$  by a waterwash and is burned for superheating the steam used for gasification.

7. For the recovery of sulfur in connection with the Alkacid and with the Potash solution method, the gases from the regeneration of the solutions which contain from 50 to 80%  $\text{H}_2\text{S}$  are burned in a Claus-oven with a reduced quantity of air. Elementary sulfur is recovered in blocs. The waste gas of the Claus-oven, which contains much sulfuric acid, is diluted by air before leaving a relatively high stag, or it may be treated by the sulfidin-process for the recovery of sulphuric acid.

The sulfur produced in the Claus-oven is clear yellow. It can be produced at a cost of 100 to 120 Rm from the highly (2%  $\text{H}_2\text{S}$ ) concentrated carbonization gases of German lignite. In this case the complete operating cost of the liquid solution plant and the Claus-oven are charged to the sulphur. In Hydrogenating or Synthesis plants a considerable part of the purification cost can be charged to the produced motor fuels.

The extraction of  $\text{CO}_2$  from gases is unavoidable in the production of hydrogen because any process of producing hydrogen by water-gas reactions is bound to a formation of considerable amounts of  $\text{CO}_2$  in the primary stage of gasification as well as in the conversion of the CO.

Elimination of  $\text{CO}_2$  is also required for city gas produced by gasification of coal, especially in the pressure-gasification method.

For the synthesis of hydrocarbons under atmospheric pressure

elimination of  $\text{CO}_2$  has been avoided in general by using such methods of gas production which produce a primary gas of a relatively low  $\text{CO}_2$  content not exceeding 12%, or the elimination of  $\text{CO}_2$  is limited to that part of the gas which had to be converted with a catalyst.

For a synthesis under pressure it will be in most cases economical to eliminate  $\text{CO}_2$  from the synthesis gas, because the additional cost for washing is relatively low when the gas is under pressure already.

8. Washing with water under a pressure between 10 and 20 atmospheres is exclusively used for the elimination of  $\text{CO}_2$ . The water used for washing absorbs  $\text{H}_2\text{S}$  and  $\text{CO}_2$  in quantities which are proportionate to the pressure of water and  $\text{CO}_2$  and also depend on the temperature of the water. The absorbed gases are released from the water by its expanding to atmospheric pressure, but a certain amount of the gases remains in the water unless this is intensely aerated after expansion. An intense aeration of the water is possible only when  $\text{H}_2\text{S}$  has been removed before the waterwash, because most of the  $\text{H}_2\text{S}$  would be liberated with the air. For this reason normally a removal of  $\text{H}_2\text{S}$  precedes the waterwash. In connection with Lurgi-pressure gasification difficulties with releasing  $\text{H}_2\text{S}$  into the air have been avoided by using limited quantities of air, which are conducted to the combustion chambers of the boiler house. The expansion gas with a heating value of 40 Btu/cu ft is burned, together with a higher grade gas, for superheating steam.

For the absorption of  $\text{CO}_2$  towers 50 to 65 high and of 7 feet diameter are used, which are equipped with several layers of Raschig-rings. The water flows over these rings and the gas passes upwards through the water. For an equal distribution of water and gas and for their perfect contact the quantities of water and gas are so regulated that the waterflow is retarded by the ascending gas.

In order to recover part of the energy used in the water pumps the water which comes from the tower is expanded through a pelton turbine into a receiver for water and expanded gas. The gas is conducted to a burner, the water is pumped with a low pressure pump on top of the aerating tower, from which it flows to the pond for regenerated water.

This aerating tower is a closed cylindric vessel 32 to 35 feet high equipped with layers of wood in case of a  $\text{H}_2\text{S}$  containing gas. It can be constructed as an open tower of wood in case of washing a gas previously freed from  $\text{H}_2\text{S}$ . A fan is required in both cases to move the air through the tower.

Part of the power required for the pumps is recovered in the expansion turbine, which mostly works immediately on the shaft of the high pressure pump.

30 to 35% of the energy is recovered, the higher figure counted for washing with 20 atmospheres.

The power consumption for the CO<sub>2</sub> removal amounts to

0.05 to 0.07 KWh/Nm<sup>3</sup> of purified gas

or 1.4 to 1.95 KWh/1000 cu ft

the lower figures relating to a removal from 12 to 3% of CO<sub>2</sub>,  
the higher figures to a reduction from 30 to 2%

The investment cost depending on the same conditions varies  
from 1.4 to 1.6 Million RM/1 Million cu ft/hour

The labor cost is unimportant.

2 men are required per shift for 1 million cu ft/hour

9. Activated charcoal is used except from the recovery of light oils and gas oil for the elimination of resin forming compounds from the gas used for the synthesis of hydrocarbons. These resins if not removed spoil the catalysts and can reduce their activity considerably. Compounds of this kind preferably are contained in a gas produced from bituminous coal or lignite if these gases have not been heated to very high temperatures (above 1000 C) in the gas producing process. Mostly in this case the gas contains also light oils and gasol (C<sub>3</sub>, C<sub>4</sub>) which can be recovered at the same time with the activated charcoal.

Part of the organic sulfur is also removed by the activated charcoal. For this reason the catalyst for the removal of organic sulfur (Feinreinigung) is saved and lasts for a longer period.

A cleaning with activated charcoal is considered as necessary for all those gases which have been produced from coal. Even for a watergas plant, in which coke-oven gas is split in the firebed of the gas producers, the activated charcoal treatment of the gas has been proved an advantage. Clear watergas and possibly gases produced by gasifying pulverized coal or lumpy coke with oxygen need no activated charcoal, or probably can be produced free from gum forming compounds by an adequate control of the gasifying temperature.

If the Synthesis plant is operated under atmospheric pressure, an activated charcoal plant is usually provided for the recovery of the synthetic light oils and the gasoil. In a pressure synthesis plant these products can also be recovered by washing methods. The activated charcoal plant for the synthetic products may be used for the cleaning of the synthesis gas in some cases where this gas has been purified carefully from light oils before entering the charcoal. This is necessary in order to keep sulfur containing light oils away from the synthetic oils. The synthesis gas is used for cooling the activated charcoal, which previously had been heated for the evaporation of the synthetic oils.

An activated charcoal plant consists of one or several groups

of three containers each, which are filled with charcoal of approximately pea-size. The containers are equipped with a system of coils, which can be heated with steam or cooled with water. The three containers are working together so that one is operated with the oil containing gas, the next is heated with steam for the evaporation of the oil or the regeneration of the charcoal, and the third is cooled by water and by sending cold gas through the charcoal. For the periodical change of this run the plant is equipped with a number of valves and controlling instruments.

A Synthesis plant can comprise two independent charcoal plants, or the cleaning of the synthesis gas from gum-forming compound may be combined with the recovery of the synthetic oils in some exceptional cases.

10. For the removal of organic sulfur compounds, which have passed the preceding stages of purification, alkaline iron oxide is used exclusively. A purification on less than 0.2 gr of sulfur per 100 m<sup>3</sup> of gas is considered as a sufficient protection of the synthesis catalyst. This degree of purification can be obtained when the gas has been freed from hydrogen sulfide completely by a tower or box system of wet iron oxide (Lux-mass), and in case of gasifying coal, part of the organic sulfur has been converted into hydrogen sulfide by high temperature or removed by activated charcoal.

The catalyst for the conversion of organic sulfur into hydrogen sulfide, which at the same time absorbs the sulfur from the hydrogen sulfide, consists of Luxmass and NaHCO<sub>3</sub>. It is operated with 300 to 350°C continuously, the entering gas being preheated to that temperature in a preheater, heated by gas. The catalyst used in Germany was 7-15 mm size. Its production cost amounts to 100 RM/1000 kg. It was manufactured by the Ruhrchemie and delivered to the other works. The used material was thrown away.

#### H. Treatment of Waste-Water, Recovery of Phenols

In connection with the Carbonization and Hydrogenation plants which have been erected in Central Europe numerous new methods for the extraction of phenols from waste-water have been developed. Extraction with benzole which worked successfully is coke-oven-plants was not found satisfactory for carbonization plants, because benzole is not produced in these plants. Experimental plants for the extraction of phenols by means of a fraction of tar oils corresponding to the solvent naphtha from natural oil have been abandoned finally in favor of other methods which could be operated industrially with better results. Some typical analytical figures of waste water from carbonizer plants are contained in the following table:

	Waste water per ton of coal m <sup>3</sup> /ton	Contents of phenol g/ltr	Contents of NH <sub>3</sub> g/ltr	Phenol Kg/ton of coal	NH <sub>3</sub> Kg/ton of coal
Carbonizer pl. Bohlen (Brown coal)	0.136	8	5	1.09	0.68
Pressure-Gas Plant Bohlen	0.800	5	4	4.09	3.20
Carbonizer pl. Hirschfelde (lignite)	0.178	15	4	2.67	0.71
Carbonizer pl. Blechhammer (bit. coal)	0.77	6.3	5	0.7	0.55

### 1. The Tricresylphosphate-process

This process has been developed by the Leuna works and has been used in several works as Bohlen, Magdeburg, Zeitz.

The solvent has a very good solution power and the purification of the water is rather complete. Serious difficulties have been met with this process if other higher boiling carbonization products than phenol are contained in the water, such as fatty acids. In this case the acids are not evaporated with the phenol. They remain in the solvent and spoil it within a relatively short period, causing difficulties by emulsions and high expense for the consumption of solvent material.

The Tricresylphosphate-process recently has been replaced by the Phenosolvan-process.

2. The Koppers process of purifying waste water from phenols has been developed together with the Saxonia-State-Works at Bohlen. In this process the phenol is evaporated from the water by means of heating and by directly introduced steam. The vapors move upwards in a high cylindrical tower, in which they are washed with a solution of sodium hydroxide. The phenols are absorbed as sodium-phenolate. The steam is recirculated to the wash tower by means of a fan. The absorption is rather complete if the phenolate is withdrawn with 50 to 60% concentration. In order to produce a saturated sodium-phenolate, which is required for the recovery of phenols, the partly saturated solution is used to extract phenol from the naphtha fraction of carbonization. It is saturated to 90% and then sold to chemical works for the production of artificial resins.

In order to eliminate CO<sub>2</sub> and part of the fatty acids, which cause difficulties in the process and decrease the quality of the phenolate, the crude water is treated first in a column in which 5% of the water is evaporated. This vapor, which contains all the CO<sub>2</sub>, the low boiling fatty

acids and 10% of the phenol are conducted to the powerhouse to be burned under the boilers. 0.5 to 0.8 gr phenol per ltr are left in the purified water.

The crude phenol produced with this method contains 50-55% carbolic acid and 40 to 35% cresol as an average of several carbonizer plants in Central Germany.

3. Recently the Phenosolvan-process has been developed by the LURGI Company, together with the I. G. Farben works of Leverkusen. This method has been used in the factories of Brux and Blechhammer for the purification of waste water from carbonization of coal and lignite and from the hydrogenation of coal and lignite tar.

In this process Iso-butylacetate is used as a solvent. This solvent having a lower boiling point than Phenol, all the difficulties, which were found in using Tricresil phosphate, have been eliminated. The solvent remains absolutely pure. Impurities are withdrawn permanently with the crude phenol.

The process works as follows:

Waste water is extracted in one or two towers in counter-current to the solvent. The phenol solvent solution is distilled under atmospheric pressure and using small amounts of direct steam. The solvent is evaporated by means of a column and recovered with a content of 1.0% phenol. The phenol withdrawn from the bottom of the column is distilled under vacuum in order to recover the rest of the solvent. Less than 0.1% are left in the crude phenol.

The purified water contains from 100-200 mg/ltr of phenol.

Capacity of the Brux-plant:

2800 m<sup>3</sup>/day waste water

with 15 gr/ltr of phenol

Capacity of the Blechhammer-plant:

2100 m<sup>3</sup>/day waste water

with 10.2 gr/ltr of phenol

Though this process is very efficient and in most cases cheap enough to cover the total production cost from the price of the phenol, the purification of the water sometimes may not be sufficient. This is the case in densely populated districts with a limited supply of stream water like Saxonia.

4. In this case a complete purification is possible by the biological method developed by Nolte. In this method the phenols are consumed by bacteria which need for their life and good growing a good oxygen supply and a temperature between 15 and 35°C. A plant of this type has been operated for many years with crude waste water of a carbonizer plant at Golzau. The water contains 4 gr/ltr of phenol and is purified to 50 mg of phenol per ltr. It is claimed and proved by large-scale experiments that a purification to less than 10 mg/ltr can be obtained with a water primarily purified to less than 300 mgs



by other methods as is the case of the Phenosolvan process.

In the Golzau plant difficulties arose from the crude water by foaming, which is considerably reduced with a water from which phenols have been extracted.

In the Golzau plant 30 m<sup>3</sup>/h of waste water have been treated after dilution with 150% fresh water in a pond of 1300 m<sup>3</sup> water volume. 3000 m<sup>3</sup> of air per hour have been blown through the water by means of porous pottery plates fixed at the bottom of the pond. 20 kg phosphoric acid have been added to the water as an additional food for the bacteria.

As a summary of the experience with the various systems of purifying waste water, the Phenosolvan method is considered as the most reliable and the most economical method. It has been proved as the only method in which the price of products covers all the expense.

#### 5. Filtering of waste-water through ash-piles

For a long time this method has been considered and used as a means to reduce the phenol content of waste water. This method cannot be considered as sufficient in connection with an industrial plant for the carbonization or the hydrogenation of coal and tar. In this case the phenols, which temporarily have been retained in the ash pile, are carried off again into the rivers by heavy rains. But conditions may be entirely different if the water has been extracted for the recovery of phenol by a modern method. In this case the filtering of the water by spraying it on piles of the ash of boilers and gas producers or of the residue of oil shale may prove a means of reducing the phenol content of the water sufficiently and to avoid another more costly equipment like that of the biological method. A dry climate and the relatively high ash content of oil shale and other low-grade fuels are conditions which favor the use of ash piles for this purpose.

#### IV. Character Products

##### A. The Products of Carbonization

The quality of the coke produced by carbonization depends highly on the property of the coal. The ash content of the coal is increased by carbonization to 150% with bituminous coal and may even go to 200% in the coke with a high volatile subbituminous coal. A low ash coke can be produced only from a very low ash coal.

When carbonizing briquetted coals the ash content may be considerably lowered before its briquetting by the usual methods by reducing the ash content of the coal.

The contents of volatiles of the coke can be regulated between 12 and 2% with direct heating methods as well as with outside heated fire-brick retorts. With iron retorts it cannot be lowered to less than 8%. The most important features of the coke are equal size, density and resistance against abrasion. They can be achieved with non caking bituminous lump coal. Other coals can be carbonized to a very solid, dense, and equally shaped coke by using special methods for the briquetting of the coal. Lignite and subbituminous coal can be briquetted without a binder. Bituminous coal requires 2 to 3% of binder.

High ash fuels with a high tar content should be utilized by a combined carbonization-gasification process.

The character of the liquid products of carbonization depends on the coal, which was treated, as well as on the process.

Products from bituminous coal and woody lignite show high contents of pitch and oxygen compounds. The neutral oils may be as low as 50%.

The tars from lignites and subbituminous coals often have a low content of pitch and oxygen compounds from 15 to 25%. Sometimes the contents of paraffin wax of such tar may be as high as 10 to 15%.

The light oils (below 180 C) obtainable by carbonization run from 8 to 12% of the tar content. 20% can be obtained by working under several atmospheres pressure as in the pressure gasification process.

Shale oil in general is of a neutral character, sometimes it has a high content of sulphur.

With several methods of carbonization, e. g., the LURGI carbonizer system, it is possible to separate certain fractions of the oils in the condensation plant. The lower boiling oils, 30%, can be separated free of paraffin wax, so that they could be refined very easily and cheaply in a separate plant. The heavy oil (60% of the total production) preferably could be hydrogenated in a central plant connected to a series of carbonizer plants, or in case of a very high paraffin content it could be refined for paraffin wax or cracked to a maximum production of gasoline and diesel oil.

In general the hydrogen content of products from bituminous coals is low, that of products from younger coals and lignite is higher. Oils

from oil shale may be as high in hydrogen as many natural oils. Such shale oils sometimes are below 0.9 in specific gravity.

Refining of carbonization products for gasoline and gas oil can be efficient only with products of a high hydrogen content and a low content of sulphur. In this case good quality gasoline with 90 to 95 of octane number can be produced from crude benzine by the usual method of refining for benzol.

Gas oil of a moderate cetane number, paraffin wax and fuel oil can be produced from lighter tars by extraction methods. By a previous distillation under 10 to 20 atmospheres pressure the paraffin wax can be cracked, resulting in an increased output of gas oil and simplified refining.

Heavy tars from coal and tars of a high content of oxygen or sulphur should be treated by hydrogenation, because only with this method a good yield of gasoline and gas oil of good quality can be obtained.

When treating bituminous coal a direct recovery of pitch, fuel oil and crude benzine in the condensing plant can be very economical, especially when the pitch can be used for briquetting slack coal.

Raw benzines from various plants:

Plant and coal	Ruhr bit. coal	Up. Silesia bit. coal	Up. Silesia bit. coal	Bohlen brown coal	Bohlen brown coal
Process	Krupp-L. retort	Krupp L. retort	LURGI- Carbonizer	LURGI- Carbonizer	LURGI- Pressure gasif.
Recovery in % of tar cont. (Fischer-Assay)	9	9	9	10	18
Spec. Gravity Kg/l 20 C	0.765	0.763	0.8	0.828	0.840
Boiling pt. °C	57	62	60	54	56
to 100°C %	51	52	25	18	33
150°C %	90.5	90	82	74	67
180°C %	96.0	94.5	95	95	85
Phenols	---	---	12	6	8
Clefins / Aromatic %	---	---	---	78	85
Octane Number (Res-M)	---	81.4	---	90	97
Sulfur %	---	0.13	0.2	2.1	1.8

Tar-oils from Coals (mixed condensates):  
benzine not incl.

Plant and Coal	Ruhr bit. coal	Up. Silesia bit. coal	Up. Silesia	Bohlen brown coal	Bohlen brown coal
Process	Krupp-Lurgi retort	Lurgi- Carboni- zer	Continuous Vert. Ret.	LURGI- Carboni- zer	LURGI- pressur. Gasif.
Recovery % of tar cont. (Fischer Assay)	60	55 *	70	80	55 **
Spec. gravity Kg/l. 20 C	1.06	1.02	1.04	0.948	0.944
Non soluble (dust) %	---	0.01	0.01	0.2	0.05
Phenols + creasote %	18	40	30	15	10
Paraffin wax %	---	---	---	14.2	5.7
Ash %	0.12	---	---	0.008	0.01
Boiling to 180°C	4	2	5	2.3	2.0
250°C	---	35	30	15.8	25.0
350°C	---	75	---	56.3	66.5
Melting point°C	- 20	+ 5	+ 5	+32	+34
Viscosity (20°C) 59°F Engler-meth.	20	15	20	---	---

\*Further 25% are separately condensed as a pitch binder with a melting point of 65°C.

\*\*20% are recovered as benzine separately.

TAR-OILS FROM OILSHALE

Oilshale	Autun	Autun	Bulgaria	Spain	Puertollano	Estonia	
Process	Pumpherstone	LURGI-Schweitzer	LURGI-Schw.	LURGI-Carb.	LURGI-Schw.	LURGI-Schw.	tunnel-kiln*
Recovery % of tar cont. (Fisch-A)	% 80	80	85	85	80	85	95
Spec. gravity kg/l 20°C.	0.90	0.925	0.905	0.892	0.876	0.99	0.946
Melting pt.	--	--	32	--	--	--	--
Phenols / creosote	5-6	2.2	7.5	10	14.2	60	16.3
Boiling to 180°C	--	0	--	--	--	--	17.0
250°C	--	7.5	10.0	--	--	--	25.3
350°C	--	40.5	55.0	--	--	--	45.0
Sulfur %	0.5	0.6	0.7	0.6	0.87	0.86	0.9

\* Incl. benzene

0.3

## B. Products of Hydrogenation

1. Hydrogenation of coal for economic reasons is used for producing gasoline because other products than gasoline, which could not be produced at a much lower cost than gasoline, do not pay for a direct hydrogenation of coal.

Gasoline from coal hydrogenation is of a high octane number as compared with gasoline from natural oil

Gasoline from Ruhr coal: (Gelsenb. Scholven)	boiling	100°C	35-40%
	"	180°C	98%
	Gravity	20	0.745 Kg/lit.
	Oct. No. (mot. meth.)		74-78

2. Hydrogenation of tar may be operated to a hundred per cent gasoline production or to moderate hydrogenation with a yield of 50% to 60% gasoline and 50 to 40% gasoil. In a country with a high production of crude oil, gasoil is available sufficiently and cheaper from crude-oil refining. A hydrogenation plant for tar in general will be operated also to produce only gasoline.

In general the gasoline produced from tar is slightly lower in its octane number than a gasoline from a coal-hydrogenation plant. Products from a heavy coal-tar are better than those from light lignite tar.

Gasoline from Hydrogenation of Heavy Coal-Tar or Pitch Rhuroil-plant:

	Octane No. Motor Method	82-84
Gasoline from Lignite tar		
Brabag-plants:	Boiling 100°C	35-40%
	Octane No. Motor Method	70

With an addition of 0.11% of Tel (5% Tetraethyl lead) the octane number could be increased by 8 to 15.

In Germany the hydrogenation plants also produced liquid gas (C<sub>3</sub> & C<sub>4</sub> Hydrocarbons) for motors. This gas was used instead of gasoline for trucks and busses, and probably will be important also in the future wherever hydrogenation plants are located near densely populated districts.

The liquid gas from hydrogenation plants mostly consists of Propane and Butane. It amounts to roughly 10% of the gasoline production.

Recently a new improved system of hydrogenating tar oils was developed at Leuna, working with a fixed catalyst and 600 atm pressure. An increased throughput, a higher efficiency, and a higher octane number are claimed for this new method, which might

be important for treating tar oils from coal and oilshale.

### C. Products from synthesis of hydrocarbons

The primary products of the synthesis at atmospheric pressure with cobalt catalyst are mostly saturated hydrocarbons. The light oils are of a low octane number, medium oils are of a high cetane number, and the higher boiling products are paraffin waxes, which may amount to 10 to 30% of the primary products. For the production of gasoline the medium oils and waxes must be cracked, which can be done with a good efficiency but still a low octane number as between 58 and 62.

A better quality gasoline can be produced working under a medium pressure of 10 to 20 atm and recycling part of the gas. In this case 50% of the primary products are benzene containing 50 to 60% olefins. The octane number of this benzene is increased to 65 instead of 50 with the straight method.

When working with an iron catalyst under pressure most of the primary products are of an unsaturated character. The higher fraction can be converted into good quality gasoline by cracking or polymerization with a high yield.

When a subbituminous coal is used for the production of the gas the benzene from the gas producer plant with an octane number of 90 to 95 may often be sufficient to produce by mixing with the synthetic product a gasoline with 70 octane number.

Tests with mixtures of synthetic-benzene with a gas benzene from a pressure gas producer showed octane numbers as follows:

	Octane No. (Res.m.)		Octane No.		
Primary benzene	100%	44	Crackbenzene	100%	63
	90 / 10%	48.5		90 / 10%	66.5
	50 / 50%	75		50 / 50%	81.0
Gas benzene	100%	97	Gas benzene	100%	97.0

The unsaturated low boiling  $C_3$  and  $C_4$  compounds can be converted into high octane number gasoline by polymerization methods.

As a whole the main product of synthesis for motors is a gasoline with 60 to 70 octane number. The gas oil fraction of synthesis, which has a high cetane number, may be very valuable to increase the cetane number of carbonization products by mixing.

The waxes which can be produced by the Fischer synthesis are of a high melting point as compared with other waxes. Except from a limited use as industrial waxes, they must be cracked for gasoline.

V. Characteristics of the various methods from economical and military view points.

Except for those cases, where tar oils are produced as by-products, the most important economical figure for any production of motor fuels is the price of the coal or shale, or more accurately of its content of combustible matter. The quality of the raw material is less important and decides only for the method of its treatment. (fig. 11)

A high content of tar or volatiles is the second important feature of a fuel with respect to producing motor fuels. This concerns carbonization, hydrogenation and the synthesis of hydrocarbons, because even for the complete conversion of a coal into gas the volatiles contribute substantially to the quantity of gas per ton of fuel, without an adequate cost. The cost of gas production is nearly proportionate to the relation of the fixed carbon to the gasified dry coal.

Coals with a high tar content can be hydrogenated with a lower pressure and a lower consumption of hydrogen.

a. Dependence on the character of the coal and the desired products.

The efficiency of the technical methods, which are available for the production of motor fuels from coal more or less depends on certain properties of the coal. Carbonization is bound to a high tar content; hydrogenation of coal wants a low ash content and a high content of hydrogen.

Synthesis of hydrocarbons may be used with any coal which is suitable for a complete gasification. Even coals with 30% ash (on dry base) can be gasified efficiently with special methods as adapted to the properties of the coal.

The general relation between a coal of a certain content of ash and volatiles (tar) and the adaptability of the various methods of production for treating this coal is shown on Fig. 11. This Fig. also shows the principal products which can be produced in each case.

If only gasoline is required hydrogenation or synthetic methods cannot be completely avoided unless a light tar with a low oxygen and sulfur content suitable for cracking can be produced in a primary stage of carbonization.

Diesel oils and fuel oils can be produced from younger coals and oil shale by the simpler and less expensive methods of carbonization and refining. Heavy tars from bituminous coals or tars with a high sulfur content should be treated by hydrogenation.

b. Dependence from other market or industries.

The production of motor fuels depends on no other consumer if the coal is completely used for the production of motor fuels. This is the case with direct hydrogenation of coal and with the synthesis of hydrocarbons. It is also the case, if high ash fuels are carbonized or gasified with an additional production of power and steam, consumed



mostly in the mine and in the production of the motor fuels.

If coals of a high content of fixed carbon are treated by carbonization or gasification in order to recover tar oils, part of the coal or of the produced gas must be sold to other consumers. In case of low content of ash and sulfur and good properties of the coal for the production of a hard and clean coke (sometimes by briquetting the coal), the quantity of motor fuels which can be produced in this combination may be relatively high.

The production of energy or of city gas for public supply may be other combinations for a cheap production of motor fuels in case of treating high volatile coals or lignites. Delivery of gas for public supply may also be favorably combined with a synthesis of hydrocarbons as a means to increase the heating value of the gas.

In many cases such combinations have been used in Germany and have been found more economical than direct hydrogenation and synthesis of hydrocarbons.

#### C. Investment cost per ton of liquid motor fuel.

Based on the cost of manufacture in Germany in 1939, the various methods of production are compared on a table. Figures contain the total cost of a plant including power station and storage tanks. The capital required for mining is not included.

This table shows that direct hydrogenation of coal requires very high investment cost. This cost can be reduced considerably if hydrogenation can be combined with carbonization.

A Fisher synthesis plant if not combined with the production of by-products also requires a very high investment.

The investment cost of a Synthesis plant can be considerably reduced, when by-products are recovered in the gasification process. At the same time such combination allows an important reduction of the coal consumption per ton of motor fuels and the motor fuels are considerably improved reciprocally by mixing synthetic hydrocarbons with by-products of the gasification.

Low costs of investment are required if carbonization is connected with a refinery plant. The products of such method, however, are mostly gas oil and heavy oils. At least the heavy oils should be hydrogenated if a sufficient market for heavier oils does not exist.

Lowest costs of investment are required when a rich oil shale, as the Green River oil shale, is carbonized and gasified, and the oils, which are of a low content of sulfur and oxygen, are treated by refining methods.

d. The material required for the plants consist mostly of manufactured steel. The quantity of steel necessary per ton of oils per annum may roughly be considered as proportionate to the investment cost. Special alloys and special manufacturing methods are required to a

considerable proportion for hydrogenation plants and synthetic plants (high-pressure, high-temperature, corrosion-resistant-equipment). Also special instruments are required to a considerable extent. A considerable part of the equipment of these plants consist in the power plant, especially when coal is directly hydrogenated. Carbonization plants require relatively more transport means but very little high-grade manufacturing. Refinery plants require the usual equipment for distilling piping and tanks.

e. The amount of laborers required for the erection of plant may also be considered as proportionate to the investment cost. The relative amount of special work and skilled laborers is higher with hydrogenation plants and lower with carbonizer plants.

The labor required for the operation of the plants, which is important also for the investment possibly required for housing and transportation, is higher in case of hydrogenation of coal, lower with a synthesis plant and hydrogenation of tar, and lowest with carbonization and refining of tar oils. Exact figures may differ considerably, however, in each individual case.

f. The cost of production for the oils is high in case of hydrogenation of coal and pure synthesis of hydrocarbons, 0.25 to 0.30 RM per kg of gasoline or 0.70 to 0.80 RM per gallon were considered as usual in German plants of this kind. They are at the same level when tar is hydrogenated, which was bought at a price of 100 to 120 RM per metric ton. The production cost may be reduced accordingly if the tar costs less, as for instance, in carbonization of a rich oil shale, or of a cheap subbituminous coal with a high tar content. In this case the minimum cost may drop to 0.50 RM per gallon of gasoline, if hydrogenation of the heavier oils is used in the production. 0.50 to 0.60 RM per gallon may be reached in a Synthesis plant in the most favorable case of a cheap subbituminous coal, which is used for the production of gas and gasified with a good tar yield.

The production cost of one gallon of gas oil in a hydrogenation plant or a synthesis plant may be lowered compared with gasoline by not more than 5 to 10%. It can be considerably lowered if tar oils from oil shale and subbituminous coal are treated by refining methods. In this case a production cost of 0.12 to 0.15 RM/per kg or 0.42 to 0.55 RM/gallon was usual for plants in Central Germany, under condition that heavier oils could be sold as fuel oil at a price of 80. - to 100.-RM per metric ton.

g. Minimum size of plants

For the hydrogenation of coal and tar oils a capacity of 150,000 tons of gasoline per annum was considered as a minimum. For economic reasons (emergency units, investment cost, cost of supervising personnel) this minimum capacity cannot be lowered without rapidly increasing the production cost. The capacity of such plants in Germany was between 250,000 and 1,000,000 tons of gasoline per day. The largest plants, Brux and Blechhammer, with a capacity of 900,000 tons of gasoline and with investment costs of 500 Mio RM and 900 Mio RM respectively,

covered a surface of 8 to 10 km<sup>2</sup>. They probably were the largest chemical works in Europe after the Leuna Works.

For a synthesis of hydrocarbons a capacity of 30 to 50,000 tons of gasoline may be considered as the minimum size for the same reasons.

In case of treating tar by hydrogenation or refining, the minimum size of this plant may be lowered to 100,000 to 150,000 tons per year, and the carbonizer plants, which are the deliverers of tar can be spread on a group of mines. The capacity of these delivering plants of carbonizers or gas-producers is determined by the volume of their market for coke or gas.

Similar conditions are to be expected for a production based on oilshale, except that the capacity of a carbonizer plant in this case is not limited by the market for other products. The shale-oils can be treated in a refinery or a hydrogenation plant, which is favorably located with respect to deliverers and consumers.

#### h. Military Characteristics

From this standpoint the degree of danger from air raids, which is connected to each system of production, is of primary importance. But in case of war the requirements of a certain system concerning skilled labor for the erection, the operation and the repair work of a plant can also be an important feature of the system. Even the amount of the material (steel and special alloys), as well as the amount of coal or miners which are bound by the system, can be important in time of war. Works like the Brux Works in addition had required military formations to a considerable extent for its protection.

The danger of air raids had proved very high in case of the widely extended plants like Brux and Blechhammer. This danger is increased relatively by the fact that a number of different sections of the entire system of production, which all separately are essential for the operation of the work, are concentrated at one place and connected by a complicated and extended system of pipelines erected on steel structures or underground and by hundreds of underground cables. The damage done by bombs on these pipelines and cables, especially on the underground pipes for water supply and drainage, may be considered as the most serious trouble experienced in the German works.

Possibly a protection of extended works against bombing may become still more difficult in a future war, due to further progress in long distance guided missiles and robot planes.

The possibility of dividing a certain capacity into a number of smaller works, either as complete independent works of the same type (small synthesis plants) or by subdividing the total process of production into a number of separately erected works which may be interchanged as consumers and deliverers in case of damages (groups of carbonizer plants together with several plants for the treatment of tar) allows to decrease the danger of an interruption of the production considerably. In this case the production is protected for the following reasons:

1. Smaller plants can be camouflaged.
2. Smaller plants cannot be damaged as easily as big ones.
3. Damages in small plants mostly can be repaired in a short time.
4. A damage in one of a number of small plants influences the total production very little. The same damage in a large plant cuts off a relatively great proportion of the total production.
5. In case of an interchange of products between a group of works (carbonization and refining) the stoppage of one factory can be equalized for several months by an overload of the rest of the works of the same type or even by the contents of the normal tank space.

With regard to the erection of new works in time of war or of the material and labor required for repair in existing works, plants with a low investment cost per ton of motor fuel and with little special material and work invested are more favorable. Carbonization plants and refineries are superior to hydrogenation plants and plants of synthesis, but refineries cannot satisfy sufficiently a high gasoline demand, especially when based on coal. A limited hydrogenation capacity for heavy tar oils would meet this situation without the high demands for capital and labor. In time of emergency direct hydrogenation of coal or synthesis of hydrocarbons hardly can be considered adequate for the erection of new plants because in this case the available skilled labor, special material, and manufacturing capacity is urgently needed for the direct supply of the army.

VI. Viewpoints with respect to the probable future technical development of motors, power stations, industrial and technical heating

As the preceding chapters have shown, the production of gasoline as the main product from coal and oil shale is complicated and expensive for the requirement of capital as well as with regard to the production cost. The production of these plants is relatively highly endangered in time of war.

The production of heavier motor fuels, as diesel oil and fuel oil, from coal and oil shale is much simpler and less expensive. It is also more efficient with regard to the raw material required per ton of production. Certain quantities of motor fuel can be produced as by-products of other industries and are very cheaply produced in this case. Enormous quantities of heavy liquid hydrocarbons could be saved also in the refining of natural oil, if the consumption of gasoline could be reduced, at least temporarily, during a war. For this reason, it seems very essential to watch the probable future development of fuel technique and fuel consumption and possibly it might be more important to adapt this development, at least as far as army equipment is concerned, to the special conditions given by a reduced production of natural oil and the necessity of producing liquid fuels from coal.

In several European countries like France and Germany, even in peacetime, reasons of national economy and the balance of trade forced the government to encourage the use of heavy oils and even substitutes like charcoal for motors. The fact that the German army for their own craft requirements had not adapted this principle proved a most important failure made by its army staff.

The development of diesel engines for all the heavier vehicles required would be the most efficient step to meet a deficiency of natural oil. The development of combustion turbines (gas turbines) for air craft and water transport, which has been started in U.S.A. and in Europe, even would allow utilization of fractions heavier than gas oil for motors. It would also allow considerable reduction of loss in the refining of tar oils.

There is no doubt that the market slowly will become more favorable to products from coal and that this development should be part of a national and international policy with respect to a saving of natural oil.

Experience in Germany proved that considerable quantities of oils can be produced cheaply in connection with industrial use of coal. Production of power and gas for industrial heating and chemical synthesis can easily be combined with the production of oils when a high volatile coal is used. In case of a lack of natural oils this combination becomes automatically attractive by an increased price for the by-products. It has been stimulated by many European governments. An increasing demand of energy and gas for heating purpose instead of coal caused by the progress of manufacturing methods and social requirements will favor more and more such plants in the future.

In Germany in several power stations pulverized firing of the boilers with coke from carbonization plants has been adopted. The development of a gas turbine of a high efficiency, which is expected within a few years, would encourage considerably the production of tar oils in combination with power stations. For the combustion in a gas turbine the gas could be produced under a pressure of 20 atm.

The factory which produces gas and oils could be placed outside the town, the gas being conducted by a pipeline to the power house and burned and expanded without further compression. A very high efficiency of power production has been expected from this new method in Germany. Even a transport of gas in pipelines for power production instead of transporting coal and electric energy might be an economical solution for a number of private and governmental, local and national problems in that case, where cheap non-caking coals of a high tar content can be used which could not be transported to the district of consumers.

The actual consumption of gas in small manufacturing industries as well as for domestic heating also might increase constantly due to economical advantages and technical and social progress. The consumption can be combined with a considerable production of oils and gasoline as by-products. In addition, a considerable amount of carbureting oil could be saved by adapting such a method of production instead of producing carburetted water gas.

## VII.. Aptitude of the Various Methods of Production for U.S.A. Conditions.

It is of great interest to analyze briefly the actual production and consumption of motor fuels in the U.S.A.

At present natural oil is the nearly exclusive resource for motor fuel. A small amount of gasoline only is produced by catalytic methods from natural gas and by scrubbing of coke-oven gas. The most important method of treating crude-oil is the cracking by heat under pressure. This method is dictated by the enormous consumption of gasoline for street vehicles. The efficiency of the cracking method is essential for the relation between the consumption of motor fuel and the quantity of crude oil which has been required for its production.

In connection with considering the actual or future necessity of replacing motor fuels from natural-oil by products from other resources, the fundamental problem which must be raised is: how can the ratio between motor fuel and crude oil required be improved?

An improvement of 10% as an example for the U.S.A. alone would save more crude-oil than that quantity which has been replaced by coal products with an enormous investment of capital and material in all the countries of the world together. The recent development of the catalytic polymerization of olefins has been of a similar effect.

Another way to this aim of saving crude oil is to replace part of the gasoline consuming motors by such motors which work with heavier oils or other hydrocarbons available from natural gas and oil refining. It is true that at present there is no economic reason for a private owner of a motor to change to another fuel than gasoline, but in the moment when certain quantities of motor fuels must be produced from coal to satisfy the market, the price of gasoline has to be increased considerably and a reduced consumption of gasoline becomes an important factor of private and national economy.

Substitutes for gasoline, whether of that kind which can be produced with a higher output from crude oil or such as can be cheaply produced from other raw-materials, have therefore been an important item in the fuel policy of other countries.

Diesel engines for tractors, heavy trucks, and for the long distance merchandise transport, the use of liquid gas ( $C_3$ / $C_4$ ) for local transport including buses, have proved very satisfactory in Europe.

The use of producer-gas for vehicles, which has been so much subsidized by the European Governments and thoroughly investigated with various fuels and equipment, cannot be considered as a solution of the problem, except for medium size water vehicles, where the weight and the space required for the equipment are not so important as on a street vehicle. For the American living standard and

labor cost not the least chance of a success can be given to this kind of motoring.

Another method to save crude oil or at a later date the enormous capital required for synthesis plants is to reduce the consumption of gas oil and fuel oil, which are used today for heating purpose in traffic, industry and household including the production of city gas. Most of these consumers could easily change to coal or gas and electrical energy produced exclusively from coal. Enormous quantities of raw material can be saved in this way, which can be converted into motor fuels even by hydrogenation much easier and cheaper than coal. Interests of national economy must lead in this direction sooner or later and the unavoidable increase of price of motor fuels will soon procure cooperation of the consumers, when the oil resources will become scarce.

By-products of gasification (tar) must be considered in the same line as far as gasification of coal for industrial heating and the production of electric energy and city gas proceeds with the lack of liquid fuel and the progress of technical methods (gas turbine).

Only when this means of saving natural oil are exhausted it becomes unavoidable to synthesize gasoline from coal, which if required for a big proportion of the actual gasoline-consumption would influence the living standard not only by an increased cost of motoring but also by absorbing a great deal of laborers in coal mining and chemical industry.

It may be taken as rather safe that then the principal consumers of liquid fuels will be the following groups;

1. High octane gasoline for air planes.
2. Gasoline for private cars and light trucks,
3. Diesel oil and liquid gas ( $C_3/C_4$ ) for buses, trucks, tractors, and medium water vehicles,
4. Heavy oils (bunker oil) for oversea and long distance water transport.

It should be remarked that technical progress of motor construction (gas turbine, jet motor) can possibly help to reduce the future consumption of light high quality gasoline in favor of a higher demand of medium and heavy oils.

#### A. U. S. A. resources of coal and oil-shale and their suitability for the production of liquid fuels.

There can be no doubt that the U.S.A., and still more the North American continent, own resources of oilshale and coal, which contain oil for thousand years to come and that the carbon content of the available coal resources, if converted into oil, will last for a similar period.



There are enormous deposits of high grade coal in the eastern states of the U.S.A. All kinds of coal are available, from anthracite to low volatile and high volatile bituminous coal, and as can be seen from the former chapters, for each kind of these coals a suitable method is available for the production of liquid fuels.

There are still greater deposits of subbituminous coal and lignite in the central and Rocky Mountain region of the U.S.A. and Canada.

From the technical standpoint all these coals are equally suitable for the production of motor fuels by one or several of the three principal methods of processing:

1. Carbonization (by prod.-gasif.).
2. Hydrogenation of tar and coal.
3. Synthesis of Hydrocarbons.

Water content, ash-content and its fusion point, sulfur content, volatiles and caking properties are practically no restrictions for the production of motor fuels from a coal. They are important only in deciding for the one or the other method of processing and for the special equipment and process to be used for the carbonization or gasification of this variety of coal.

Low mining cost per ton must be considered by far as the most important feature of any project, and a high content of hydrogen has proved as the second important point, especially when a considerable part of the fixed carbon can be utilized for the production of coke or gas for industrial and domestic consumption.

But even for an exclusive production of liquid fuels by hydrogenation or synthesis, the greatest part of the coal consumed is used for the production of gas and energy and offers a means of producing a considerable amount of the total production of liquid fuels as cheap by-products in case of a high volatile raw material.

Except from carbonization plants, which are bound to a densely populated industrial region, as a consumer of coke or gas or energy, the erection of plants for hydrogenation or synthesis is practically unrestricted in their location. Both methods requiring from 5 to 8 tons of coal per ton of gasoline production (according to the grade and the oil content of the coal) it is obvious that the cost of the coal is most important for the production cost of gasoline. It is also easily understood that such plants should be erected within coal fields which allow cheap open-pit mining. Presumed that one kg of gasoline requires 35,000 kcal of coal, the influence of the mining cost may be seen from the following table:

Coal	Gal. Value of coal	Cost of Coal per metr ton \$	Cost of Coal per ton of gasoline \$	Cost of Coal per gallon Cts
Coal (undergr)	7000	7.00	35.00	10.6
Coal (open pit)	6500	3.00	16.2	4.9
Subbit. coal (open pit)	5200	1.50	10.01	3.05
		1.00	6.67	2.02
Lignite (open pit)	3900	1.50	13.5	4.09
		1.00	9.0	2.73

As compared with a total production cost of approximately 30 to 40 cts per gallon of gasoline from coal by both the Hydrogenation and the Synthesis method, the cost of coal is very essential.

Cheap open pit mining is possible in some eastern coal fields. It is possible on an unlimited scale under very favorable conditions in the fields of subbituminous coal of Wyoming and Montana and in the lignite fields of North Dakota and Texas.

Carbonization and by-product gasification is important in connection with hydrogenation and synthesis plants in these same fields. All these subbituminous coals and lignites are very suitable for carbonization and by-product gasification by cheap methods. For the only purpose of producing coke or gas for a long distance supply, under present conditions these cheap coals are located too far from the consumers. Some non-caking coals from the eastern fields could be used in this way in order to replace carbureting oil and natural gas, probably in combination with a synthesis of hydrocarbons as far as a regulation of the heating value of the gas is required.

The production of energy, which has so successfully been combined with the production of gasoline in Germany in the lignite fields, is based in the U.S.A. nearly exclusively on bituminous coal. In this case the technical conditions are less favorable to a recovery of by-products. With the development of a gas turbine for power stations possibly the power stations could produce tar in a big scale as a by-product of gasification.

The deposits of oilshale in Colorado and Wyoming, with an oil content of several hundred billion tons as compared with the 20 billion tons of known oil resources of the world, must be considered as the most important resource of motor fuels for the future.

The oil content, the mining conditions, and the physical and chemical properties of this shale are very favorable to a cheap processing and a low production cost. The laborers required for the production of oils are relatively very low as compared with other methods operated with coal. For these reasons the production of oils from oilshale for a long time probably will be the most economical way to produce motor fuels from carbonaceous minerals.

A high grade oil can be produced to a practically unlimited extent at a price of 20 to 30 dollars per metric ton (price-level, 1940 to 1945). Calculations as well as the experience in Europe show clearly that hydrogenation of coal and synthesis of hydrocarbons cannot compete on this base even if part of the shale oil is refined by hydrogenation, and even a synthesis based on natural gas probably cannot be operated with a lower production cost.

If the crude oil is refined to gasoline, gasoil and bunker oil, only by simple methods of distillation and extraction which seems adequate to a big extent, the production cost in the oil shale field is considerably lower than with any other method starting from coal. It must be reminded, however, that under normal conditions the oil shale field is too far from the coast, and that therefore sooner or later the heavy fractions must be converted to gasoil and gasoline probably by hydrogenation methods.

B. Possibility of application and Special advantages of the various Methods of producing motor fuels from coal.

a. Carbonization and By-product-gasification of coal and oil shale.

This method is the only method available for the utilization of the oil shale. Its application seems to be unlimited as to the resources of raw material and the quality and market of oil shale products. The production cost of this process is relatively low as compared with other methods. No doubt an industry based on this method in the green-river field will produce an important part of the motor fuels in a later period.

In connection with coal, conditions in USA are different from those in Germany, where low-grade but high volatile non-caking bituminous coals and lignites are located near densely populated centers of consumption. Most of these coals need some processing like drying and briqueting to suit the market. They can be gasified and carbonized with cheap methods and coke and gas find a ready market, replacing the low grade raw coal.

In USA only high-grade bituminous coals, which are mostly caking are located near the centers of populations and industry. These coals are less favorable to gasification and carbonization for various reasons:

- 1.) The relation of by-products to fixed carbon or heating value is less favorable than with the younger coals.
- 2.) The technical methods of treating caking coals are less efficient and more expensive than those used for non-caking coals.

So carbonization and by-product gasification of coal probably will not become an important factor in the eastern states. They may have some chance at a later date, when natural gas and carburetted water gas should require new resources, or the production of electric energy should change to gas-turbines. Carbonization may also have a

field of application for the production of a high grade coke from high volatile coal by briquetting coal slagg and carbonization of these briquetts in a shaft carbonizer, in districts in which low volatile caking coal is not available.

Carbonization and by-product-gasification can be used for high volatile non-caking coals according to the market for gas and coke in many central and western fields.

It will be important in connection with Synthesis and hydrogenation plants. Presuming that only 10 million tons per year of motor fuels are produced at a later date by these methods and that for economical reasons (low mining cost) the plants are located in the lignite and sub-bituminous coal regions there will be required from 80 to 100 million ton of coal in these plants which can deliver another 5 to 8 million tons of by-products at a very low cost of production.

b. Hydrogenation of tar and coal.

Hydrogenation as a method of refining of residues from natural oil and shale oil and of tar seems to be very important as long as the consumption of gasoline prevails in motoring and the demand cannot be satisfied with the available crude and the refining methods used at present.

Plants for the hydrogenation of heavy oils probably will be used at a considerable extent for products from oil shale and by-products from coal.

Hydrogenation of coal, the method of highest investment cost and also a very high production cost, at a later date may not be dispensed with in case of a high demand of gasoline but in USA more than in other countries the high labor cost for the erection and the operation of such plants probably will show other methods more economical, than direct hydrogenation of coal. Hydrogenation of coal is preferably suitable for the treatment of the high volatile caking coals, which are difficult to be treated with other methods.

c. Synthesis of hydrocarbons.

This method requires enormous quantities of hydrogen-carbon monoxide gas. Its economy therefore depends on low production cost of such gas. No doubt it has an enormous field of application for the conversion of natural gas into gasoline and there can be no doubt that in this case the gasoline can be produced cheaper than with the same or any other method starting from coal. Synthesis plants based on natural gas could produce a considerable quantity of gasoline and diesel oil. At a later date even certain quantities of natural gas, which are used for heating purpose now, should be replaced by gas from coal because natural gas will be more valuable as a resource for gasoline.

Synthesis of hydrocarbons from coal, must be more expensive than in case of natural gas. From a technical standpoint it can be

used for any coal, even for the many cheap high ash and low volatile coals, which are unsuitable for hydrogenation. The production cost of a synthesis plant contrary to the hydrogenation process highly varies with the quality, the tar content, and the price of the coal, so that in many cases, especially with some sub-bituminous coals the synthesis process is considerably more economical than hydrogenation and the synthesis process probably will be used at a later date to a considerable extent for the production of gasoline and diesel oil from coal.

#### Attempt of a prognostication of the development of production of motor fuels in USA.

Though many of the statements taken in this paper require thorough investigation of the basic USA conditions, certain facts of the experience of European countries will be equally practicable for the USA, at least, when the supply of natural oil becomes insufficient. A prognostic picture of the possible development, even if a source of much critics and restrictions should help to understand the statements of this paper and facilitate their criticizing examination.

The necessity of saving natural oil or utilizing coal and oil shale for the production of motor fuels not only depends on the exhausting of the actual oil resources but also on the possibilities that some of the foreign resources might not prove reliable enough for political and economical reasons and that an increasing consumption in other countries might decrease the shipments from abroad.

The fact that the adaptation of the motors and the production of motor fuels from other resources than natural oil requires an enormous investment and consequently a great proportion of the manufacturing capacity for many years, if enforced by a lack of natural oil, should be another reason to reduce the consumption of natural oil at an earlier date and to direct the concerned industries according to guide principles taken from a correct prognose.

The end point of the development that is a nearby exclusive production from coal and oil shale seems rather clear, if motoring does not change to another energy than liquid fuels. This end point has been practiced in Europe. It facilitates considerably the following prognose of the development in USA.

		Addit. Production	Saving
I. Period	a) Production of more gasoline from natural gas (Synthesis)	gasoline	
	b) Hydrogenation of crude-residues and of tar	gasoline	
	c) Diesel motor for heavy vehicles (trucks, buses)		gasoline
	d) Carbonization-gasific. of oil shale	gasoline gas oil bunker oil	
	e) More coal, coke and gas from coal for heating (by-products)	gasoline gas oil	fuel oil carburett. oil
	f) Gas from coal instead of nat. gas (by-products) (synthesis of gasoline)	gasoline gas oil	fuel oil
II. Period	a) Carbonization-gasif. of oil shale	gasoline gas oil bunker oil	
	b) Synthesis of Hydrocarbons. (by-products gasific)	gasoline gas oil	
	c) Hydrogenation of tar and coal (by-product gasific)	gasoline	

Some medium investment figures as an indicator for the requirements of material, labor and erection time may illustrate the above table:

Motor fuel	Production method	Investment cost per met. ton/year
		\$
Gasoline	Synthesis from nat. gas	200-250
	Hydrogen. of oil-residues & tar	200-250
	Synthesis from coal	350-400*
	Hydrogen. of coal	450-500*
Diesel oil	Carboniz. of oil shale and refining	180-220*
	By-products from coal and refining	130-160
	Synthesis. from coal	320-360*
Bunker oil	Carboniz. of oil shale and refining	150-180*
	By-products and refining	100-130

\*based on price-level 1940-45 and including opening of the mine.

The first period may be characterized as the period of changing part of the consumption from gasoline and oils suitable for the production of gasoline to lower grade heavier oils or to coal and of increasing the recovery of gasoline from natural oil and natural gas. The second period is that of gradually replacing gasoline, gas oil and bunker oil from natural oil by products from the carbonization of oil shale, the hydrogenation of coal and the synthesis of hydrocarbons from coal.

PRODUCTION OF MOTOR FUELS

FROM COAL, LIGNITE AND OIL SHALE  
Comparison of various methods on German conditions (1939)

RAW MATERIAL	a. Coal (lignite) b. Coal (lignite) c. Oil Shale	10% tar) 14% tar) Fischer 16% tar) Assay	10% moisture 10% " 3% "	8% ash 8% "	Type & Minimum Size of Factory products metr. to/year	Investment	Investment Cost	Coal Consumption	Coke for
						cost line not incl. Mill. RM	per ton oil/year RM/to/year	per ton oil incl. energy & steam to/te	market. Consumpt of factory deducted to/year
1.	HYDROGENATION OF COAL J. G. - Bergius								
a.	150,000 Gasoline	125 (120) /	850 (810)	5.9 (5.7)					
b.	150,000 "	120 (115)	800 (770)	5.8 (5.6)					
	/ ( - - ) = 50% of products are <u>diesel oils</u>								
2.	CARBONIZATION OF COAL AND HYDROGENATION OF TAR								
a.	150,000 Gasoline	75 (72)	500 (480)	14.8 (14.6)			1,100,000		
b.	150,000 Gasoline	70 (68)	470 (450)	10.6 (10.4)			640,000		
c.	150,000 Gasoline	65 (62)	440 (420)	7.7 / 1.4 to coal- (1.2)					
3.	FISCHER-SYNTHESIS (Atm. pressure) no by-products.								
	30,000 Gasoline	21	700	6.1					



Type & Minimum Size of Factory products metr. to/year	Investment cost Mine not incl. Mill. RM	Investment Cost per ton oil/year RM/to/year	Coal Consumption per ton oil incl. energy & steam to/to	Coke for market Consumpt of factory deducted to/year
4. FISCHER-SMNTHEISIS (Medium pressure Lurgi - system ) By products				
a. 50,000 (39,000 Gasoline ( 6,500 Diesel Oil (4,500 Fuel Oil	27	540	5.0	—
b. 56,000 (41,000 Gasoline ( 9,000 Diesel Oil (6,000 Fuel Oil	29	520	4.6	—
5. CARBONIXATION OF COAL (lurgi-Carb.). REFINERY PLANT				
a. 100,000 (	42	420	12.5	680,000
b. 100,000 (10,000 Gasoline (40,000 Diesel Oil (50,000 Fuel Oil	35	350	8.4	420,000
c. 100,000 (	322	320	7.3	—
6. CARBONIZATION-GASIFICATION OF OIL SHALE (Huboven) REFINERY PLANT				
c. 100,000 (10,000 Gasoline (40,000 Diesel Oil (50,000 Fuel Oil	28	280	7.8	—

Base of Calculations:

1. Hydrogenation of coal

Coal for Hydrogenation

1.66 kg net coal = 2.0 kg Raw Coal /kg gasoline

H<sub>2</sub> Consumption

2 500 Nm<sup>3</sup>/to gasoline

H<sub>2</sub> Recovery

1 050 Nm<sup>3</sup>/ to raw coal (Winkler prod.)

2. Carbonization of coal and Hydrogenation of tar

Recovery of tar

90% of Fischer Assay

Tar consumption

1.33 Kg tar/kg gasoline

H<sub>2</sub> Consumption

1 600 Nm<sup>3</sup>/kg gasoline

H<sub>2</sub> Recovery

1 000 Nm<sup>3</sup> kg of coke breeze (Winkler)

3. Fischer Synthesis (Atm. pressure)

Recovery of primary prod.

130 gr/nm<sup>3</sup> of (N<sub>2</sub> / CO)

Finished products

90% of 130 gr = 117 gr/nm<sup>3</sup> of (H<sub>2</sub> / CO) includ. Propan

(H<sub>2</sub> / CO) Recovery

1 500 nm<sup>3</sup>/ to coal (70% eff. of producer)

4. Fischer Synthesis (Medium pressure) By-products (Lurgi-system)

Recovery of primary prod.

155 gr/nm<sup>3</sup> of (H<sub>2</sub> / CO)

Finished prod.

90% of 155 gr = 139.5 gr/nm<sup>3</sup> of (H<sub>2</sub> / CO)

Recovery of tar and benzine

80% of Fischer assay (with 20% benzine)

Efficiency of refining

90% of tar (60% Diesel Oil / 40% fuel oil)

5. Refining of tar and benzine with solvents.

Recovery of  
finished prod.

95% of tar

(10% gasoline / 40% Diesel Oil / 50% Fuel Oil)

6. Carbonization-Gasification of Oil Shale (Hüboven)  
with Refining of tar

Recovery of tar

85% of Fischer assay .

Efficiency of refining

95% (10% gasoline / 40% Diesel / 50% Fuel Oil)

Fig. 9.

Cost of Synthesis-Gas

Gasification under  
20 atm. pressure

Fuel: subbit. coal  
20% moisture  
12% tar  
h. cal. val. 5000 kcal  
per kg

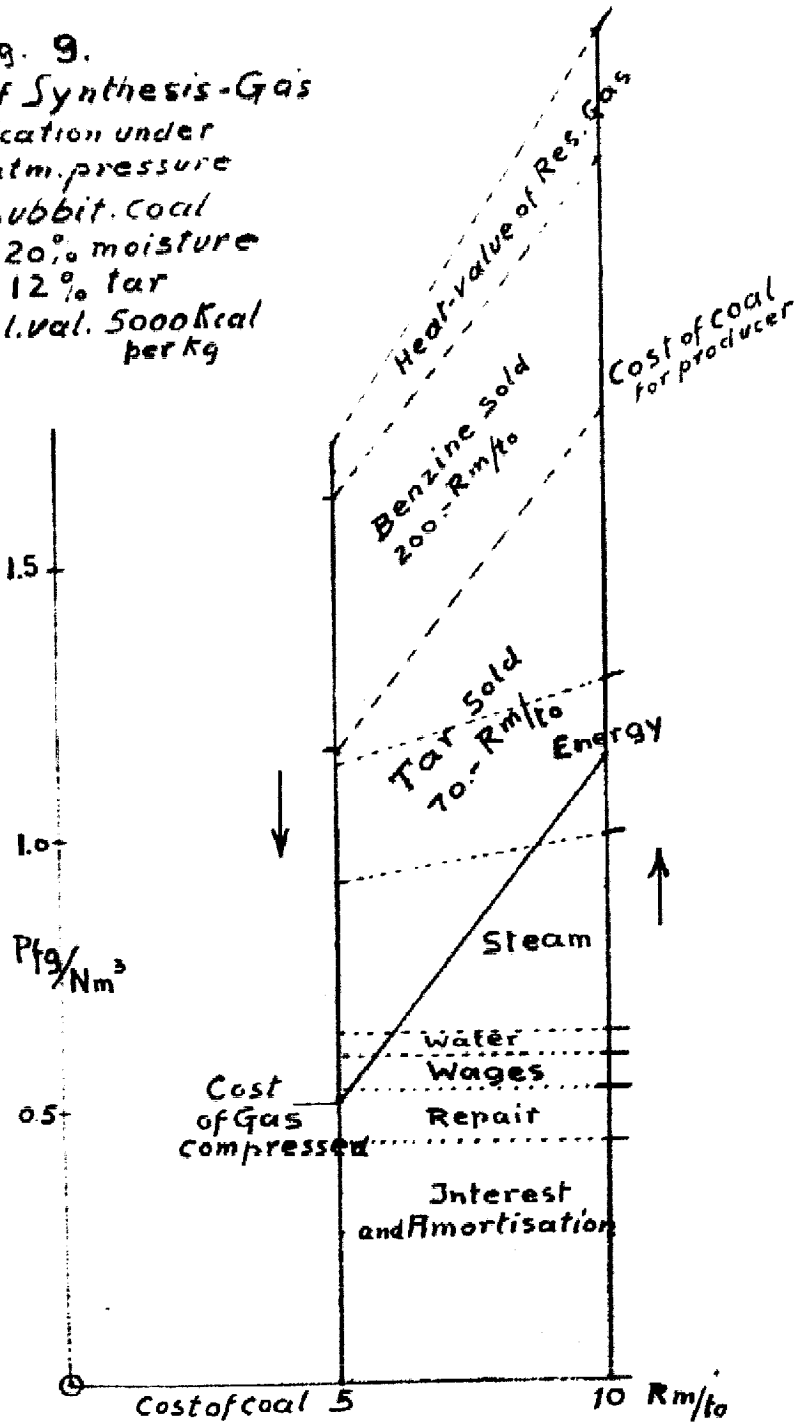


Fig. 10.

# LURGI-Pressure-Gasification for Synthesis-Gas

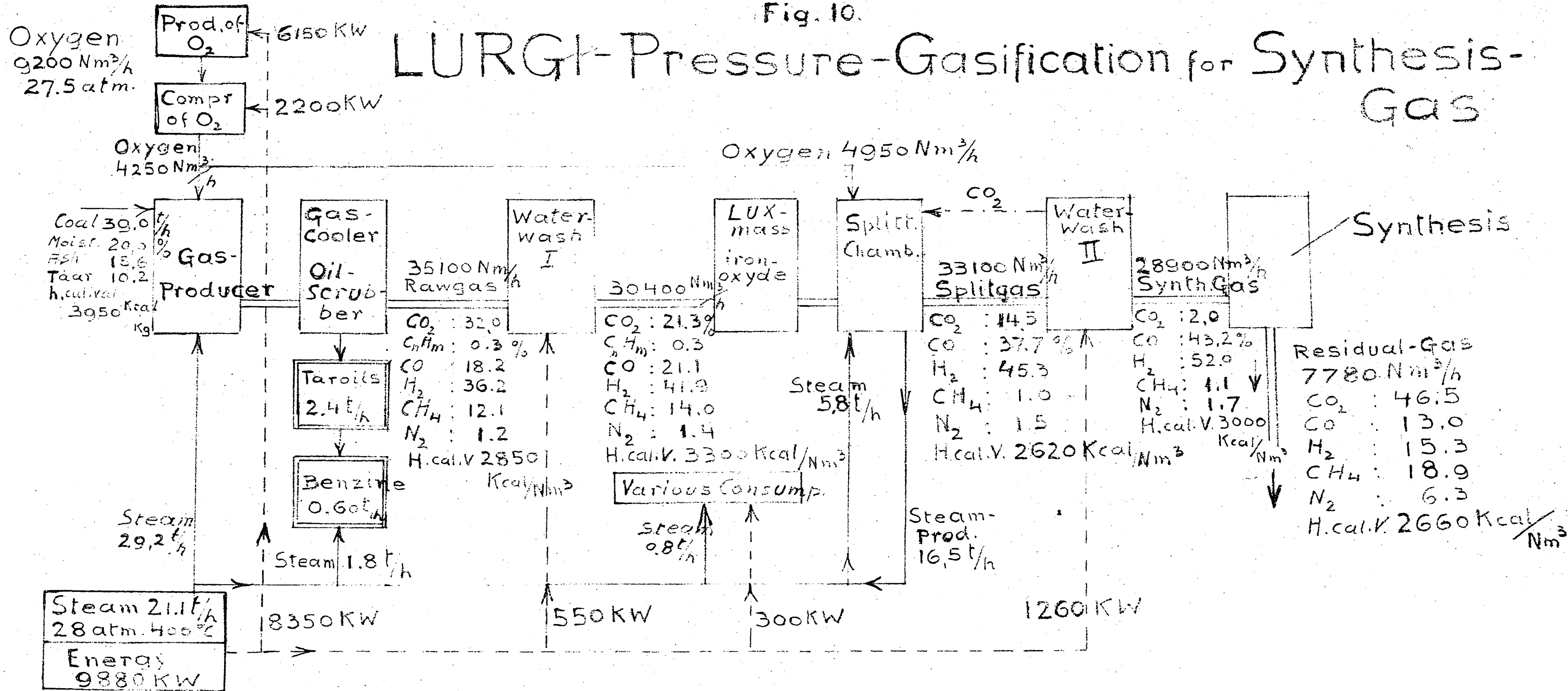


Fig 11.

MOTOR FUELS FROM COAL & OIL SHALE

RELATION BETWEEN RAW MATERIAL AND PROCESS

PRODUCTS	PROCESS	Ash low		PROCESS	PRODUCTS
GASOLINE (Dieseloil)	FISCHERSYNTHESIS	GASIFICATION with O <sub>2</sub> Lump or Pulverised	ANTHRACITE & COKE BREEZE	GASIFICATION w.O <sub>2</sub> Lump	FISCHERSYNTHESIS  GASOLINE (Dieseloil)
GASOLINE (Dieseloil)	FISCHERSYNTHESIS	COKEOVEN WATERGAS	CAKING	POWER STATION Pulverized F.	
GASOLINE (Dieseloil)	DIR. HYDROGENATION	-----	CAKING		
GASOLINE (Dieseloil) FUELOIL	FISCHERSYNTHESIS	CARBONISATION WATERGAS	HIGH VOLATILE	POWER STATION Pulverized F.	
GASOLINE (Dieseloil)	HYDROGENAT. OF TAR	GASIFICATION w.O <sub>2</sub> Pulverised	NON CAKING	CARBONIZATION GASIFICATION	HYDROGENATION of TAR GASOLINE (Dieseloil)
GASOLINE (Dieseloil)	DIR. HYDROGENATION	-----	BITUMINOUS	GASIFICATION W.O <sub>2</sub> Byproducts	HYDROGENATION of TAR GASOLINE (Dieseloil)
GASOLINE (Dieseloil) (Fueloil)	FISCHERSYNTHESIS	GASIFICATION W.O <sub>2</sub> Byproducts		GASIFICATION w.O <sub>2</sub> Byproducts	FISCHERSYNTHESIS GASOLINE (Dieseloil) (Fueloil)
GASOLINE (Dieseloil)	FISCHERSYNTHESIS	GASIFICATION w.O <sub>2</sub> Lump or Pulverised	SUBBITUMINOUS & LIGHT LOW TAR CONTENT	GASIFICATION w.O <sub>2</sub>	FISCHERSYNTHESIS GASOLINE (Dieseloil)
GASOLINE (Dieseloil)	DIR. HYDROGENATION	-----	SUBBITUMINOUS & LIGHT	CARBONISATION GASIFICATION	HYDROGENATION of TAR GASOLINE (Dieseloil)
GASOLINE (Dieseloil)	HYDROGENAT. OF TAR	CARBONIZATION	HIGH TAR CONTENT	GASIFICATION w.O <sub>2</sub> Byproducts	FISCHERSYNTHESIS GASOLINE DIESEL OIL (Fueloil)
GASOLINE (Dieseloil)	FISCHERSYNTHESIS	GASIFICATION w.O <sub>2</sub> Byproducts		CARBONISATION GASIFICATION	REFINERY (Hydrogen. of residue) GASOLINE DIESEL OIL FUELOIL (Gasoline)
			OIL SHALE		