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SC 106

"Coal Carbonization in the Laboratory"

It is of great importance to develop laboratory methods which are suitable to determine the yields of coke, gas ammonia, and benzol of a coal by carbonizing a very small quantity of coal, whereby it is necessary that the yields which are obtained by the small scale carbonization correspond with the yields of a commercial operation. Such a method is very useful for the following reasons:

Coke oven designers and constructors must very often design complete coke oven plants without being able to get detailed information about the properties of the coal which shall be carbonized in a new plant. As experience shows, the customer furnishes a few pounds of coal and expects to get information about the quantities of the products which are produced by the commercial carbonization process.

Coal mines are often interested to know the properties of the coals of various seams without being able to make full scale tests.

Such a method is also very important for coke oven operators because it is possible to supervise the proper running of the plant by a standardized laboratory test. It can be easily observed whether an increase or decrease of the yields depends on an alteration of the quality of the coal or a careless operation of the plant.

Furthermore, it is possible to investigate the influence of the carbonization temperatures on the yields.

The method which is applied in the laboratory must meet the following requirements:

It must be possible to reproduce the results within close limits and the heating equipment must be designed in such a manner that distinct temperatures can be maintained over long times.

Description of the Apparatus

The apparatus consists of:

a. The carbonization arrangement.

b. The condensing and scrubbing system.

c. The gas collecting equipment.

a. <u>The carbonization arrangement:</u>

Like all coke ovens the carbonization arrangement is composed of the carbonization chamber and the heating system which furnishes the thermal units which are required for the decomposition of the coal.

A coke oven chamber is nearly completely filled with coal. The liberated gases and vapors ascend partly through the center of the coal and partly near the oven-walls. They are collected in the upper part of the chamber, the so-called gas collecting chamber, before they leave the chamber flowing through the gas ascension pipe into the gas collecting main. The yields and the composition of the products are influenced by the temperature of the oven walls and the temperature of the walls of the gas collecting chamber which is always a little lower. Therefore, the coke oven chamber of the laboratory is subdivided into the carbonization part and the gas collecting part in order to be able to imitate the commercial carbonization process. The temperatures in both sections can be kept on different heights. The gas collecting chamber is kept completely empty in order to maintain always the same cracking surface area.

The refractory bricks of a wall of a coke oven are made of silicon compounds. The laboratory chamber is made out of quartz glass in order to obtain the same cracking properties of the walls. Quartz glass is very resistant against high temperatures, but due to a modification of the crystalline structure by the influence of temperature the quartz glass will become brittle and crumble. Approximately 100 tests can be carried out utilizing the same quartz chamber. The chamber has the form of a tube, which is closed at one end and equipped with a ground joint at the opposite end. The length of the tube is 75 cm (29.53") without the ground joint and 80 cm (31.5") including the ground part. The inside diameter is 20 mm (7.87"). The tube must have an absolute uniform width over the entire length in order to facilitate the discharge of the coke. The tube lies in two electric furnaces which are mounted on rails. One is 42 cm long (16.5") and the second 20 cm (8"). The proper regulation of the temperature is performed by heat control rheostats (slide contact type) or regulating transformers. The controlling instruments must be able to maintain any desired temperature (up to 1200°C = 2220°F) within 10°C (18°F). The temperatures are measured at the outside of the tube by unprotected thermocouples (made of Pt-Rh) whereby one of the two measures the temperature of the carbonization zone and the other that of the cracking zone. Reading resistance galvanometers show the applied temperatures. The consumption of electric current is indicated by reading ammeters.

b. <u>The condensing and scrubbing system:</u>

The condensing and scrubbing system consists of

- the tar recovery arrangement
- the ammonia absorption together with the determination of the carbonization water

the carbon dioxide and hydrogen sulfide absorption
the benzol extraction system

The tar vapors are extracted from the gas by a filtering process at a temperature which is higher than that of the dew point for water vapors. The filter consists of a short tube which is made of quartz glass. The tube is stuffed with cotton wool, which has been dried to a constant weight at a temperature of 105°C (221°F). The tube is connected with the carbonization chamber by a ground joint. The tube is 20 cm (8") long including the ground part, which has a length of 5 cm (1.97"). The width is 20 mm (0.787"). The tube is in a water-bath and is enveloped by the steam of the boiling water. The cotton wool extracts both the tar and the ammonia compounds such as ammonium chloride and ammonium sulfate. Due to the temperature of the water bath no water vapors are condensed.

The gaseous ammonia vapors are absorbed by a solution of diluted suffuric acid (16% by weight). The solution is filled into a Geissler potash absorption bulb which is connected with the tar extraction tube by means of a rubber stopper. Most of the water vapors, which are formed by the decomposition of the bitumen, are condensed in the Geissler bulb. The water vapors which remain in the gas are completely absorbed during their passage through a following U-shape drying tube with side arms and ground stoppers. The U-tube is filled with Calcium chloride.

Two Geissler bulbs, which are filled with a solution of potassium hydroxide absorb carbon dioxide and hydrogen sulfide. They are followed by two U-shape drying tubes with side arms and ground stoppers which are filled with calcium chloride. The calcium chloride retains the water vapors which escape from the U-tubes due to the vapor pressure of the potassium hydroxide solution.

The benzol vapors, which are present in the gas, are absorbed by petrolatum U.S.P. which is filled in two Geissler absorption bulbs with side arms and ground stoppers. In order to secure a complete extraction the U-tubes are packed in chopped ice. The application of a paraffin oil instead of active charcoal has the advantage that only benzol vapors but no gases like butane, propane are absorbed. The absorption bulbs are protected by a calcium chloride filled U-tube against water vapors which might come back from the gas collecting bottle.

The permanent gases are collected in a large aspirator bottle which has a capacity of approximately three gallons. The bottle is closed by a rubber stopper which is equipped with a thermometer, a water column guage and two stop-cock-tubes for the inlet and outlet of the gas or water respectively. As gas flows into the bottle the same volume of water is displaced. It is collected by a second bottle which is arranged underneath the first one.

Performance of the Test

Preparation of the coal sample:

The delivered coal is carefully mixed in order to get a representative sample which is dried in order to determine the moisture content. The average size of the coal pieces should be roughly estimated. The dried coal is ground to a size which is suitable for the performance of the analysis (particles must pass sieve #170). Two bottles are filled with the coal, one for analytical purposes and the second for reserve. The stopper of the second bottle is carefully tightened; the bottle is stored away after having been properly labeled.

Approximately 5 g of the pulverized coal are put into a weighing bottle with inside ground stopper for the determination of the hygroscopic water content. The sample is dried applying a temperature of 105°C (221°F) utilizing a drying oven. In order to get figures of the proximate analysis the content of volatiles, coke residue and ash is determined according to standardized specifications.

Exactly 15 g of the pulverized coal are weighed utilizing an aluminum scoop. The coal is filled into the quartz tube by means of a pencil brush and a funnel with a long stem. An asbestos plug is placed on top of the coal charge in order to keep the coal in its proper position.

The absorption bulbs are filled with the respective absorbing agents and weighed by means of an analytical balance. In order to prevent any losses the openings of the bulbs are tightened by means of close fitting rubber tubes and short glass rods. The U-tubes are weighed with the ground stoppers closed. The content of the U-tubes may be used for a couple of experiments. A refilled U-tube must be saturated with carbon dioxide because of the presence of alkaline compounds in the commercial calcium chloride which would absorb carbon dioxide from the gas. The carbon dioxide must be carefully replaced by dried air before the U-tubes are weighed.

The short quartz tube is stuffed with dried cotton wool and also weighed.

The gas collecting bottle is completely filled with a very diluted solution of sulfuric acid.

The absorption bulbs, the U-tubes and the gas collecting bottle are fitted together by means of rubber tubes. The stoppers of all U-tubes remain closed.

The two quartz glass tubes are connected by the ground joints which are greased by a very thin coating of pulverized graphite (graphite must be free from any grease or oil). The combined quartz tube is inserted into the tubes of the electric furnaces and the thermocouples are adjusted.

The water bath is slipped over the tar filter, which is connected with the ammonia absorbing bulb by means of a rubber stopper.

All stop-cocks are opened and all connections are carefully examined to see if they are air-tight.

Due to the difference of the height of the gas- and water-collecting bottles the apparatus is subjected to a slight vacuum pressure. Any gas bubbles which can be easily seen on their passage through the absorption bulbs indicate that the apparatus is not tight.

The two electric furnaces are in such a position, that the end of the long quartz tube which is filled with coal is not covered by the small furnace. With all the preparations and adjustments finished the carbonization can be started.

The electric furnaces and the water bath are heated to the proper temperatures.

Temperatures which are necessitated to perform the carbonization:

It was found out by trial that the temperature of the gas collecting zone must be kept at 850°C (1562°F) if the normal operation of a coke oven shall be imitated, whereas 950°C (1742°F) are necessary to meet the requirements of gas works practice.

The carbonization is performed in two stages, at first the small furnace, which is mounted on rails is moved very slowly over the coal at a temperature of 750°C (1382°F). The velocity with which the furnace slides over the coal depends on the gas volume which is liberated per unit of time. In order to secure a proper absorption of the various constituents two or three gas bubbles per second should pass through an absorption bulb. With a growing distance between the two furnaces a gas burner is kindled, the flame of which heats the quartz tube in order to prevent any condensation of oil vapors. It is also possible to wind a wire spiral around the quartz tube and to heat by electric current. As soon as the coal is completely covered by the furnace the small furnace is moved back to its outgoing position. It is now heated to higher temperatures. The temperature depends on the volatile content of the coal. A coal with a volatile content of 20% or less requires a temperature of 900°C (1652°F) whereas the temperature must be raised from 10°C (18°F) corresponding with an increase of 1% of the volatile content of the coal. A coal with a volatile content of 25% requires the application of a temperature of 950°C (1742°F). All coals which have a volatile content of 30% and more are subjected to a temperature of 1000°C (1832°F).

With the progress of the carbonization the volume of the liberated gases decreases substantially. As soon as almost no gas bubbles can be observed the carbonization is finished. The stop-cock at the entrance of the ammonia absorption bulb is closed and the connection between the bulb and the tar filter is loosened. The electric furnaces and the gas burner are shut down.

Since all absorption bulbs and U-tubes are filled with gas, the latter must be replaced by air in order to be able to determine the proper weights of the bulbs and tubes. The entrance of the ammonia absorption bulb is connected with an aspirator bottle, the stop-cock is reopened and approximately 1 liter (0.035 cft) of dried air is led through the absorption- and gas-collecting equipment.

Inlet and outlet stop-cocks of the gas collecting bottle are closed and temperature and pressure of the gas are instantly read and recorded.

All stop-cocks of the U-tubes are closed, the latter are disconnected from the absorption bulbs the inlets and outlets of which are tightened by the rubber tubes and glass rods.

The quartz tubes are disconnected and the tar filter tube is shortly dried in a drying oven whereas the coke bearing tube is weighed after cooling.

Determination of the Yields and Composition of the Gas

The content of the gas collecting bottle is mixed by shaking the bottle and two samples are drawn using gas sample tubes. The actual gas volume consisting of the carbonization gas and the air which was blown through the condensing and absorption equipment is determined by measuring the volume of water which has been displaced by the liberated gases from the gas collecting bottle.

Example:

Actual gas volume 7355 ccm
Barometric pressure: 764 mm Hg
Gas pressure read from the water column gauge 85 mm Hg.
Temperature of the gas 31°C
Reduction of the gas volume to normal conditions:
Actual pressure 764 - 85 = 661 mm Hg
Pressure of saturated water vapor at 31°C = 33.7 mm Hg
Factor for reducing the gas volume $= \frac{(661 - 33.7) \times 273}{760 \times (273 + 31)} = 0.741$
Reduced gas volume 7355 x 0.741 = 5452 ccm
Actual composition of the gas as determined by a gas analysis:

CO_2 - 1.1 % by volume
Cn Hm - 0.9 " " "
O_2 - 3.8 " " "
CO - 5.1 " " "
H_2 - 47.3 " " "
CH_4 - 21.6 " " "
N_2 - 20.2 " " "

The volume of the admixed air can be calculated from the oxygen content of the gas.

Air content $= \frac{3.8 \times 100}{20.8} = 18.3$ % by volume

Volume of the admixed air $= \frac{5452 \times 18.3}{100} = 1000$ ccm

Oxygen-volume 208 ccm
Nitrogen-volume $\underline{792}$ "
1000 ccm

Volume of the carbon dioxide: $\frac{1.1 \times 5452}{100} = 57$ ccm

Volume of the air + carbon dioxide = 1000 + 57 = 1057 ccm

Volume of the carboniz~~ation gas free~~ from CO_2, H_2S, benzol vapors:
5452 - 1057 = 4395 ccm

Volume of the carbonization gas (free from CO_2, H_2S, benzol vapors)

based on 1 ton of dry coal $= \frac{0.004395 \times 1}{0.000015} = 293$ Nm^3

Actual volume of the gaseous compounds, which are present in the gas:

CO_2 : $\frac{1.1 \times 5452}{100} = 57$ ccm

C_nH_m: $\frac{0.9 \times 5452}{100} = 48$ ccm

O_2 : $\frac{3.8 \times 5452}{100} = 208$ ccm

CO : $\frac{5.1 \times 5452}{100} = 277$ ccm

H_2 : $\frac{47.3 \times 5452}{100} = 2580$ ccm

CH_4 : $\frac{21.6 \times 5452}{100} = 1178$ ccm

N_2 : $\frac{20.2 \times 5452}{100} = 1104$ ccm

Composition of the gas free from air, CO_2, H_2S, benzol vapors

C_nH_m: 48 ccm

CO : 277 "

H_2 :2580 "

CH_4 :1178 "

N_2 :1104 - 792 = $\underline{312}$ ccm
Total 4395 ccm

$$C_nH_m = \frac{48 \times 100}{4395} = 1.1\%$$

$$CO = \frac{277 \times 100}{4395} = 6.3\%$$

$$H_2 = \frac{2580 \times 100}{4395} = 58.7\%$$

$$CH_4 = \frac{1178 \times 100}{4395} = 26.8\%$$

$$N_2 = \frac{312 \times 100}{4395} = 7.1\%$$

The following figures are utilized for the computation of the heating value of the gas:

	H.H.V.	L.H.V.
C_nH_m	16500	15500 kcal per Nm^3
CO	3020	3020 " " "
H_2	3050	2570 " " "
CH_4	9520	8550 " " "

Heating value of the gas free from CO_2, H_2S, benzol vapors

	H.H.V.	L.H.V.
C_nH_m	182	171
CO	190	190
H_2	1785	1500
CH_4	2555	2295
	4712	4156 kcal per Nm^3

The following figures are utilized for the calculation of the density of the gas:

CO_2 : 1.9769 g per ccm

H_2S : 1.539 " " "

C_nH_m : 1.2604 " " "

CO : 1.2504 " " "

H_2 : 0.0898 g per ccm

CH_4 : 0.7168 " " "

N_2 : 1.2506 " " "

Benzol vapors: 3.49" " "

Calculation of the density of the gas:

C_nH_m	11 x 1.2604 =	13.85 g
CO	63 x 1.2504 =	78.80 "
H_2	587 x 0.0898 =	52.30 "
CH_4	268 x 0.7168 =	191.50 "
N_2	71 x 1.2506 =	88.93 "
		425.38 "

Density of the gas 0.425 kg per Nm^3.

Coke yield

Quartz tube empty	115.3265 g
+ Coal	130.3265 g
Weight of the coal	15.0000 g
Quartz tube + coke	126.2786 g
Quartz tube empty	115.3265 g
Coke	10.9521 g
Percentage of coke	$\frac{10.9521 \times 100}{15.0000} = 63.02\%$

Determination of ammonia, carbonization water and hygroscopic water:

Absorption bulb filled with diluted H_2SO_4	45.3648 g
" " after experiment	46.1217 "
Absorbed and/or condensed	0.7569 g
U-tube before experiment	23.4732 g
" " after experiment	23.8483 "
Water vapors absorbed	0.3751 g

Total absorption $0.7569 + 0.3751 = 1.1320$

Small amounts of oil are condensed in the absorption bulb.

The average quantity of oil which is condensed is 0.025 g as found out by trial.

Amount of ammonia, carbonization water and hygroscopic water:

$$1.1320 - 0.0250 = 1.1070 \text{ g}$$

The content of the absorption bulb is carefully transferred to a distilling flask whereby the last traces of the solution are removed by distilled water. A solution of sodium hydroxide is admixed and the liberated ammonia vapors are distilled into a measured volume of deci-normal solution of sulfuric acid. The surplus acid is neutralized by deci-normal solution of sodium hydroxide. The quantity of ammonia can be calculated from the consumption of the deci-normal sulfuric acid solution.

applied	35.00 ml	deci-normal sulfuric acid solution
for neutralizing	10.30 "	deci-normal sodium-hydroxide solution
consumption	24.70 ml	deci-normal sulfuric acid solution

quantity of ammonia $= 24.70 \times 0.0017 = 0.0420$ g

$$= \frac{0.042 \times 100}{15} = 0.28\%$$

carbonization water + hygroscopic water $1.1070 - 0.042 = 1.0650$ g

The quantity of hygroscopic water is determined by a separate experiment.

5 g gr of pulverized coal are dried at a temperature of 105°C (221°F).

Weighing bottle with ground stopper empty	16.3587 g
" " " " " + coal	21.3587 "
Coal	5.0000 g

Weighing bottle with ground stopper and coal	21.3587 g
" " " " " after drying	21.3162 g
loss	0.0425 g

hygroscopic water $\frac{0.0425 \times 100}{5} = 0.85\%$

Quantity of the hygroscopic water of the carbonization experiment

$$\frac{15 \times 0.85}{100} = 0.1275 \text{ g}$$

Quantity of the carbonization water 1.0650 - 0.1275 = 0.9375 g

Percentage of carbonization water based on dry coal:

$$\frac{0.9375 \times 100}{15} = 6.25\%$$

Determination of the tar yield:

The cotton wool of the tar filter does not only extract tar vapors but also ammonia salts, which are floating in the gas-like unit. The dried tar filter is weighed. The increase of the weight represents the quantity of tar and ammonia salts. The cotton wool is transferred to a distilling flask, the last traces of ammonia salts are removed by a spray of distilled water, a solution of sodium hydroxide is admixed and the liberated ammonia vapors are distilled into a measured volume of deci-normal solution of sulfuric acid. The surplus acid is neutralized by deci-normal solution of sodium hydroxide. The quantity of ammonia can be calculated from the consumption of deci-normal sulfuric acid solution.

Applied	10.00 ml deci-normal sulfuric acid solution
for neutralizing	7.35 " deci-normal sodium hydroxide
consumption	2.65 ml deci-normal sulfuric acid solution
quantity of ammonia	2.65 x 0.0017 = 0.0045 g

$$= \frac{0.0045 \times 100}{15} = 0.03\%$$

tar filter before experiment	53.4573 g
" " after "	53.9318 "
tar ⌿ ammonia	0.4745 g
tar = 0.4745 - 0.0045 =	0.4700 g

total quantity of tar 0.4700 ⌿ 0.025 = 0.4950 g

$$= \frac{0.4950 \times 100}{15} = 3.30\%$$

Determination of CO_2 and H_2S

The sum of CO_2 H_2S is determined by weighing the Geissler absorption bulbs and the two following U-tubes before and after the experiment.

absorption bulb #1 before experiment	41.2354 g
" " " after "	41.4891 "
increase of weight	0.2537 g
absorption bulb #2 before experiment	42.0374 g
" " " after "	42.0900 "
increase of weight	0.0526 g
U-tube #1 before experiment	14.3216 g
" " after "	14.3672 g
increase of weight	0.0456 g
U-tube #2 before experiment	13.9546 g
" " after "	13.9882 g
increase of weight	0.0336 g

Total amount of CO_2 ⌿ H_2S:

0.2537 g
0.0526 "
0.0456 "
0.0336 "
0.3855 g

The quantity of H_2S is determined by titration using a deci-normal solution of iodine.

The alkaline content of the absorption bulbs is transferred to a vol~~umetric fla~~sk (volume 250 ml) and filled up with distilled water. Exactly 15.00 ml of a deci-normal solution of iodine are filled into an Erlenmeyer flask and diluted sulfuric acid is admixed. 50 ml of the alkaline solution are slowly pipetted into the iodine solution. The surplus iodine is determined by a deci-normal solution of sodium thiosulphate utilizing starch solution as indicated

applied 15.00 ml deci-normal iodine solution
" <u>9.35</u> " " " sodium thiosulphate solution
consumed 5.65 " deci-normal iodine solution

quantity of H_2S $5 \times 5.65 \times 0.0017 = 0.0480$ g

$$= \frac{0.0480 \times 100}{15} = 0.32\%$$

$$= \frac{22.4 \times 3.2}{34} = 2.1 \text{ cbm per ton of dry coal}$$

quantity of $CO_2 = 0.3855 - 0.0480 = 0.3375$ g

$$= \frac{0.3375 \times 100}{15} = 2.25\%$$

Carbon-dioxide which has been determined in the gas = 57 ccm

$$= \frac{1.976 \times 57}{1000} = 0.112 \text{ g}$$

$$= \frac{0.112 \times 100}{15} = 0.75\%$$

Total amount of $CO_2 = 2.25 / 0.75\% = 3.00\%$

$$\text{or } \frac{22.4 \times 30}{44} = 15.2 \text{ cbm per ton of dry coal}$$

Determination of the Benzol

The absorption bulbs are weighed before and after the experiment, the increase of the weight shows the amount of benzol vapors which have been extracted from the gas.

Absorption bulb #1 before the test:	38.9763 g
" " " after " "	<u>39.1159</u> g
Increase of the weight	0.1396 g

Absorption bulb #2 before the test: 37.6831 g

" " " after " " 37.6860 g

Increase of the weight 0.0029 g

Total amount of benzol 0.1396 + 0.0029 = 0.1425 g

$$= \frac{0.1425 \times 100}{15} = 0.95\%$$

or

$$\frac{22.4 \times 9.5}{78} = 2.7 \text{ cbm per ton of dry coal}$$

Calculations concerning the gas

Gas yield incl. CO_2 H_2S, benzol:

293 cmb per ton of coal

15.2 " " " " "

2.1 " " " " "

2.7 " " " " "

Total gas yield 313.0 " " " " "

Thermal units in form of gas per 1 kg of dry coal (without benzol)

0.293 x 4712 = 1375 kcal per kg

Since the heat of combustion of benzol (benzene) is 10,000 kcal per kg, 0.95% benzol yield represent 9.5 x 10 = 95 kcal per kg of dry coal.

Thermal units in form of gas per 1 kg of dry coal incl. benzol

= 1375 + 95 = 1470 kcal per kg of coal

Gas yield per weight = 293 x 0.425 = 12.45%

The results of the assay should be recorded applying special forms. The following pages contain all essential figures, which should be recorded. With a proper distribution of the paragraphs one sheet of normal size is large enough to represent all data of the assay.

Carbonization - Assay

Delivered from: Chariton mine, Lucas, Iowa

Physical properties: Nuts size 1"

Proximate Analysis (on dry coal)

Volatile matter	29.69%	Ash:	6.5%
fixed carbon	70.35%	Moisture:	5.6%

Carbonization Temperatures

Carbonization section 750/1000°C Cracking section 850°C

Results of the Assay on dry coal

Gas volume (free from CO_2, H_2S, benzol	Nm^3 per ton	293
Gas volume (incl. CO_2, H_2S, benzol	Nm^3 per ton	313
Higher heating value of the gas free from CO_2, H_2S, benzol	kcal per Nm^3	4712
Thermal units in form of gas (without benzol)	kcal per kg	1375
Thermal units in form of gas (included benzol)	" " "	1470
density of the gas	kg per Nm^3	0.425

A. Volatile matter on dry coal

CO_2	3.00%
H_2S	0.32%
Tar	3.30%
Benzol	0.95%
Free ammonia	0.28%
Fixed ammonia	0.03%
Carb. water	6.25%
Hygrosc. water	0.85%
Permanent gas	12.45%
Total	27.43%

B Coke	73.02%
Total A + B	100.45%

C. Composition of the Gas

C_nH_m : 1.1% by volume

CO : 6.3" " "

H_2 : 58.7" " "

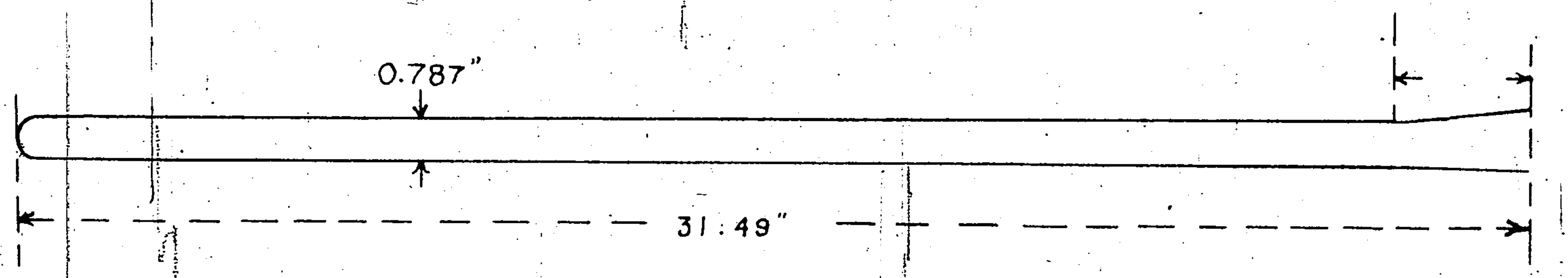
CH_4 : 26.8" " "

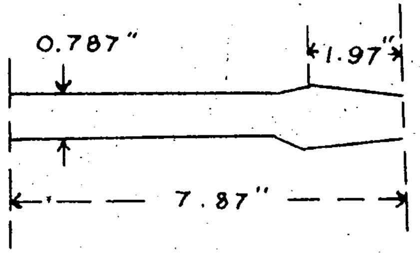
N_2 : 7.1" " "

Yield of ammonium sulphate 1.2%

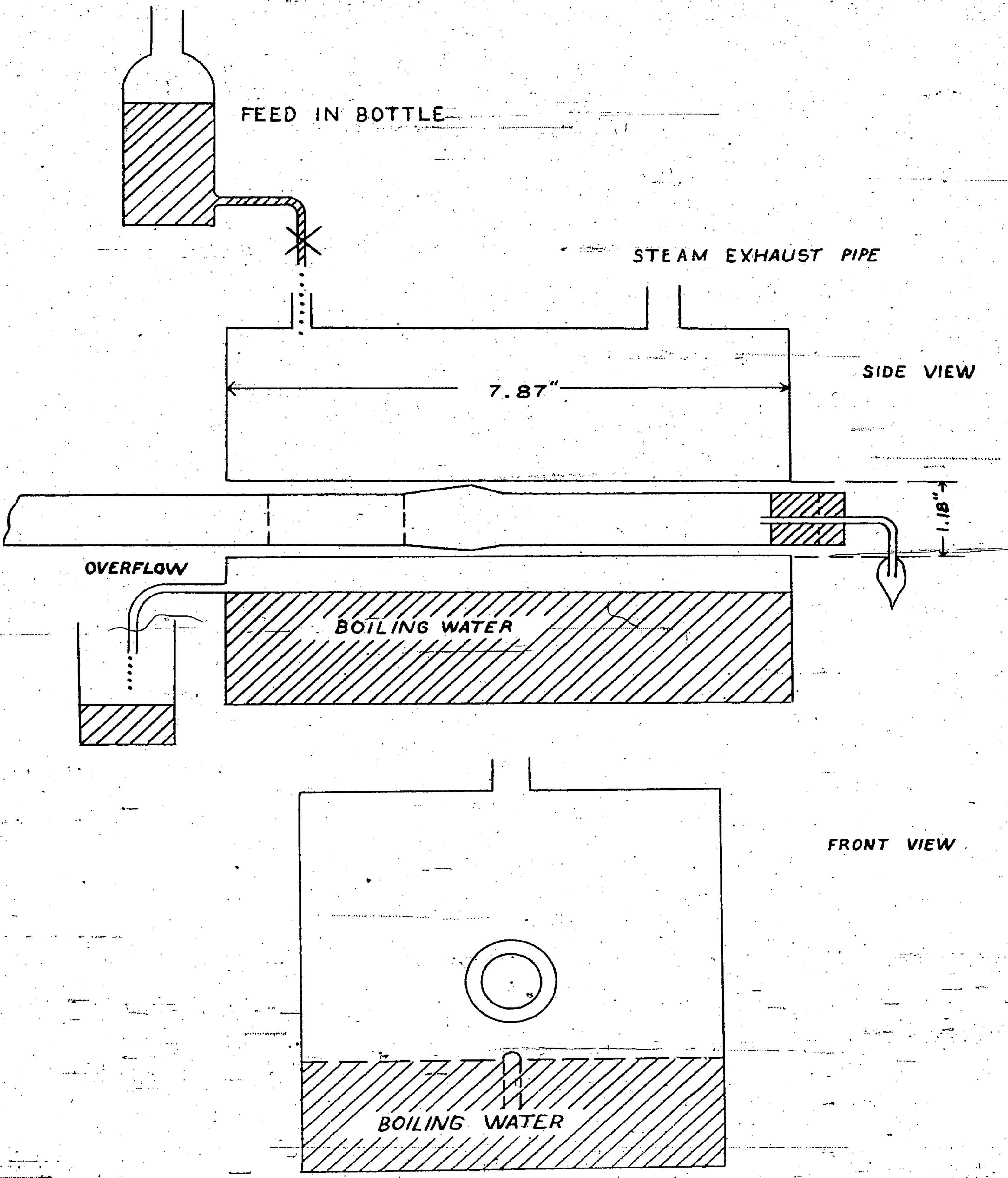
Remarks:

Swelling properties of the coal:	0.58 kg per sq cm
Caking properties: (Silesian method)	16 -
Color of the coke:	dark grey
Structure of the coke:	small pores, melted

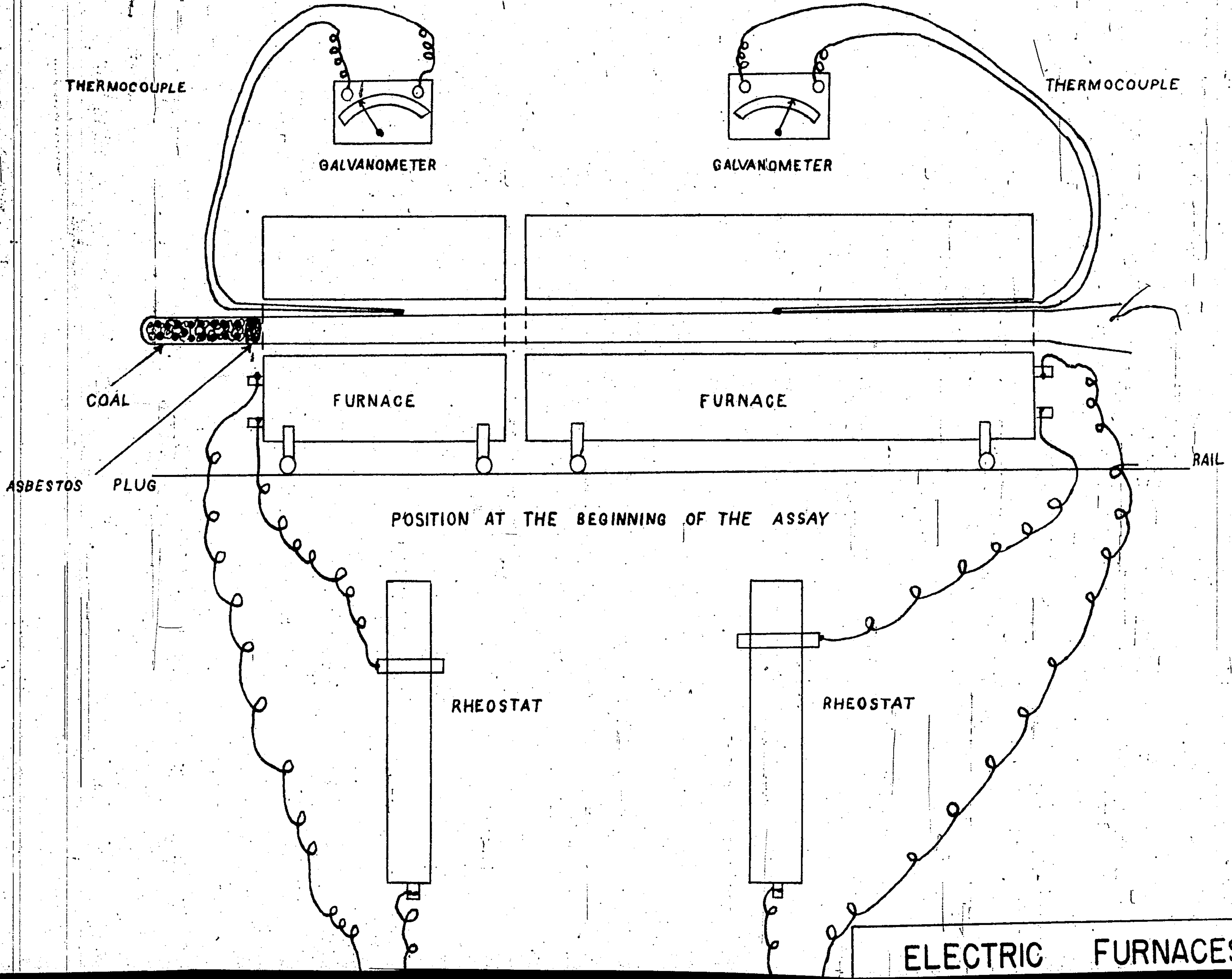




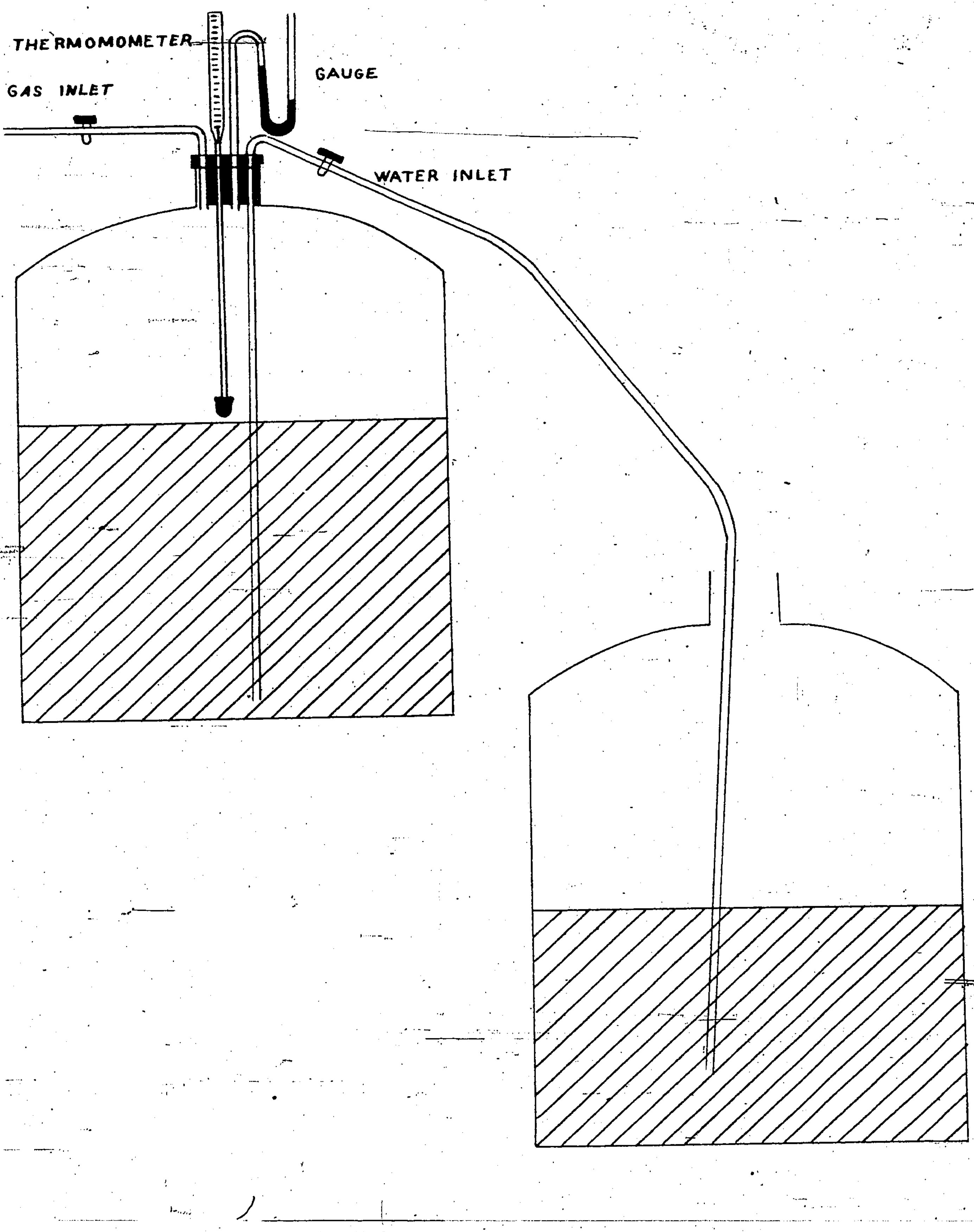
QUARTZ TUBES


FEED IN BOTTLE
STEAM EXHAUST PIPE
SIDE VIEW
7.87"
1.18"
OVERFLOW
BOILING WATER
FRONT VIEW
BOILING WATER

WATER-BATH



ELECTRIC FURNACES



GAS COLLECTING BOTTLE

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SC - 103

Low Temperature Carbonization of Slight Caking Coals.

Low temperature carbonization processes of coal intend to carbonize the coal by the application of lower temperatures than usual when the coal is carbonized in normal coke ovens. The main object of low temperatu carbonization processes consists of an increased tar yield and a coke the thermal properties of which are often superior to those of a high temperature coke. The chemical composition of the tar is substantially influenced by the application of lower temperatures which prevent a far going decomposition of the oils. One of the most remarkable properties of the low temperature tar is its high content of phenolic compounds which iften amounts to at least 20%. Experiments were carried out to use the phenolic compounds as raw material for the manufacturing of plastics whereas the oils can be refined by various methods or used as fuel oils. Depending on the content of volatiles and the composition of the bitumen coals react differently during the carbonization process. A strong caking coal begins to soften at a temperature of approximately 360 to 420°C (680 to 790°F) as a result of the decomposition of the bitumen. The coal substance becomes semifluid whereby the melted substance envelopes the solid particles and glues them together. The numerous small coal particles of the coal charge form large pieces of coke. The first step of the solidification of the semifluid substance is the formation of semi-coke or low temperature coke which still contains a certain percentage of gases. Correlating to the rising temperature the gas content of the coke is lowered. The coke shrinks and forms cracks due to a decrease of its volume. The volume of the gases which are liberated during the heating period determines the physical properties and percentage of shrinking of the coke. Only a properly shrunk coke can be easily removed from the coke oven chamber by a ram without damaging the walls of the chamber. A non-caking coal does not soften and does not become semifluid, gases are liberated and each single piece of coal preserves its initial form. Mostly intermittently working processes are applied for the carbonization of caking coals whereas continuously operated furnaces are very convenient if non-caking coals must be carbonized. Unfortunately many coals which yield large volumes of tar develop such caking properties which exclude the application of either intermittently or continuously working processes of the types usually employed. The coke, which consists of agglutinated pieces of coal is not hard enough that it can be discharged by a ram. On the other hand, the spongy coke clusters do not descend through the narrow chambers by force of gravity if continuously operating processes are employed. It is well known that comparatively large and hard pieces of coke are necessitated for the operation of blast furnaces or foundry kilns, whereas most soft and smaller pieces of coke are often suitable for gasification processes, domestic heating, operation of boilers and other purposes. Since we do not know any commercial processes which can be utilized to improve the caking properties of a coal it has often been tried to convert a slightly caking into a non-caking coal. It is possible to destroy the caking properties of the bitumen by oxidation or the influence of an elevated temperature. Oxidation is performed by oxygen which is present in flue gases. A treatment of the coal with hot flue gases results in a substantial decrease of the caking properties of a coal. Heating a coal to temperatures of approximately 20°C (35°F) below its softening point affects also its caking properties. But, depending on the size of a coal, the time which is required for an effective treatment often is so long that the economy of the process is unfavorably influenced. The processing temperatures must be carefully controlled and very narrow reactors must be employed in order to secure a uniform temperature throughout the charge. Experiments which were carried out at the Otto pilot plant showed that the Otto low temperature carbonization furnace, which was equipped with horizontal twin flues, was able to carbonize non-caking as well as slightly caking coals. But, due to the time which was required for the alteration of the caking properties of the coal, the capacity of the furnace was reduced. Since processes which alter the caking properties of the coal are not a solution of the problem to carbonize slightly caking coals, devices must be designed which are fit to handle the above mentioned coals.

An externally heated rotating drum can carbonize slightly caking coals, provided that the spongy coke clusters can be properly discharged. Due to abrasion, dust is formed when the coke is moved through the rotating drum. The dust is carried away by the liberated gases and picked up by the condensing oils, causing the formation of oil-water emulsions which are sometimes almost impossible to break.

In some cases narrow steel containers 3 to 4 inches wide and of a limited height are suitable to carbonize slightly caking coals. Due to the intermittently working process, special devices, such as rams, must be applied for the discharge of the coke. The discharge of the coke is aggravated if it is so soft that the ram cannot push out the coke but only penetrates the surface of the coke charge and comes to a standstill due to the sticking properties of the coke. It is also possible to spread the oven walls , whereby the coke is easily discharged by force of gravity. Due to the limited size of the chambers numerous units are required if large quantities of coal must be carbonized per day.

The attached sketches show the principle of a furnace which probably is suitable to carbonize slight caking coals which are fed to the furnace as nuts. The furnace consists of a cylinder into which a set of plates is placed which bear the coal charge. Revolving rakes which are shaped like plows transport the coal charge from the feed in place to an opening through which the coal is discharged to the following plate. The plates serve as containers for a hot recirculating liquid, which carries the heat necessary to carbonize the coal. The liquid is led to the bottom plate, flows from plate to plate, and is recirculated from the top plate to the heating system. The plows must be designed in such a manner that a uniform distribution of the coal is maintained and that the single piece of coal is tilted from one side to the other, so that the coal comes in close contact with the hot plate. The openings which connect the various plates must be wide enough so that coke clusters can be discharged. The following calculations have been performed in order to explain the proposed process a little more in detail, but it must be mentioned that careful considerations must be made before final designs can be determined.

Diameter of a plate .5m = 16.4 ft
Diameter of the axle .05m-= 1.64 ft
Area of a plate: 19.635 sq. meter

$$\begin{array}{r} 19.635 \text{ sq. meter} \\ \underline{-\ 0.196}\ \text{" \quad "} \\ 19.439 \text{ sq. meter} \end{array}$$

Thickness of the coal layer 0.05 m
Volume of the coal 19.439 x 0.05 = 0.97195 cbm
Bulk density of the coal 0.75
Weight of the coal per plate 0.97195 x 0.75 = 0.7289 metric tons
Number of plates per furnace 12
Weight of coal per furnace 12 x 0.7289 = 8.7468 metric tons

The carbonization time of a Krupp-Lurgi-steel furnace (width 0.06-0.07 meter) was approximately 3 hours when coal nuts were carbonized. Taking the same figure as the basis for the determination of the throughput of the furnace, the daily capacity of the furnace is

$$\frac{24 \times 8.7468}{3} = 69.97 \text{ metric tons}$$

Size of the slots for discharge of coal or coke respectively:

Width 0.25 m
Length 2.25 m
Surface area 0.56 sq. meter $= \frac{0.56 \times 100}{19.439} = 2.88\%$ of the area of a plate
Actual throughput per day 69.97 x 0.9712 = 67.95 metric tons per 24 hours

Time which is required for the coal to pass over a plate:

$$\frac{3 \times 60}{12} = \frac{180}{12} = 15 \text{ minutes}$$

Throughput per minute: $\frac{67.95}{20 \times 60} = 0.047$ metric tons per minute

$= \frac{0.047}{0.75} = 0.063$ cbm per minute

Number of rakes per plate: 16
Surface area between 2 rakes: $\frac{19.439}{16} = 1.21$ sq. meters

Weight of coal between 2 rakes: $\frac{0.7289}{16} = 0.0455$ tons

$= \frac{0.0455}{0.75} = 0.061$ cbm

Assuming that the coal is heaped up to a certain extent during its way over the plate and th t only 66% of the plate are covered with coal, the actual thickness of thecoal bed would amount to $\frac{0.05}{0.66} = 0.076$ meters

Velocity of the coal during its passage over the plate:
Circumference of the plate: 15.7 meter
Time passage: 15 minutes
Velocity at the outside of the plate: $\frac{15.7}{15} = 1.04$ meter per minute

Circumference of the axle: 1.57 meter
Velocity near the axle: $\frac{1.57}{15} - 0.104$ meter per minute

Average velocity: $\frac{1.04 + 0.104}{2} = 0.572$ meter per minute

Heat Consumption of the Process

As mentioned before, the plates are supposed to be narrow chambers through which a preheated liquid is recirculated which transfers its heat to the coal. Organic silicon compounds have been developed during recent years which have extremely high boiling points. The liquids have a very good resistance against high temperatures, and are non-corrosive. The application of hot recirculating liquids instead of flue gases has the advantage that the volumes are much smaller, because nearly the same thermal units can be transferred by 1 volume of the liquid as by 1,800 volumes of flue gases. The heating chambers become very narrow, resulting in small distances between plates. The distances between the plates are not determined by the width of the heating chambers but by the height of the rakes. Furthermore, the rate of heat transmission from a hot liquid to the steel wall of the plate is much better than that from a hot gas.

Heat consumption per 1 kg of coal:
Temperature of the discharged coke: 550°C (1022°F)

Assuming that the liberated gases are superheated if they contact the walls of the heating chambers, the average temperature of the outgoing gases is supposed to be 500°C (932°F).

Yields based on moist coal

Coke	74.9%
Tar	8.7%
Benzol	0.9%
Carbonization Water	4.7%
Gas	7.8%
Moisture	3.0%
	100.0%

0.749 x 0.28 x 550 = 115.0 kcal
0.087 x 0.5 x 500 = 21.7 "
0.087 x 85 = 7.4 "
0.009 x 0.44 x 500 = 2.0 "
0.009 x 95 = 0.9 "
0.047 x 1 x 500 = 23.5 "
0.047 x 540 = 25.5 "
0.078 x 0.25 x 500 = 9.7 "
0.03 x 1 x 500 = 15.0 "
0.03 x 540 = 16.2 "
263.9 kcal per 1 kg of coal

Assuming that 15% of the thermal units which are introduced into the furnace are lost by radiation, 236.9 + 36.5 = 273.4 kcal per kg of coal must be available at the entrance of the furnace.

Since no data were available of the physical constants of the organic silicon compounds, the following calculations must be based on various assumptions:

Spec. heat of the liquid 0.3 kcal per kg per degree C
Density " " " 0.7 kg per liter
Temperature of liquid at entrance of furnace 650°C (1202°F)
" " " " outlet " " 350°C (662°F)

Thermal units available per kilogram of liquid:
650 x 0.3 x 1 = 195 kcal
350 x 0.3 x 1 = 105 kcal
90 kcal per kg of the recirculating liquid.

Quantity of liquid which must be recirculated per kg of coal =

$$\frac{273.4}{90} = 3.04\ \text{kg.} = \frac{3.04}{0.7} = 4.34\ \text{liter} = 1.15\ \text{gallons}$$

Quantity of coal which must be carbonized:
67.95 metric tons per 24 hours
2.83 " " " hour
0.047 " " " minute

Quantity of liquid which must be recirculated:
67,950 x 4.34 = 294,903 l = 294.903 cbm = 77,900 gallons per 24 hours
2,830 x 4.34 = 12,282 l = 12.282 cbm = 3,260 gallons per hour
47 x 4.34 = 204 l = 0.204 cbm = 54 gallons per minute
3.4 l = 0.0034 cbm = 09gallons per second

In order to secure a proper distribution of the liquid which flows through the heating chamber of the coal carrying plates, it is subdivided into 8 compartments. The height of the heating chamber id supposed to be 30mm (1.18"). The velocity of the liquid during its flow through the heating chamber depends on its distance from the center of the plate.

The following table shows the velocity of the liquid, depending on its distance from the center of the plate:

Distance from the center		Velocity	
m	feet	m per sec.	feet per sec.
5	16.40	0.0578	11.384
2.5	8.20	0.116	22.83
1	3.28	0.29	57.09
0.5	1.64	0.578	113.8

The single compartments are connected by channels.
Velocity of the liquid during its flow through the channel - 1 m per sec - 60 m per minute - 196.9 feet per minute

Cross section of the channel = $\frac{0.0034\ m^3 \times sec}{1\ \ sec \times m}$ = 0.0034 sq. meter

Width of the channel if the height is 30 mm (1.18"),

$$X \times 0.03 - 0.0034;\ X = \frac{0.0034}{0.03} = 0.113\ m = 11.3\ cm = 4.45''$$

The recirculating liquid, which enters the furnace at a temperature of 650°C (1202°F) and leaves at 350°C (662°F), is led to a steel preheater which is heated by gas,

Calculation of the Heat Consumption of the Preheater

Based on 1 kg of Moist Coal.

Volume of the recirculated liquid 3.04 kg per 1 kg of coal
$3.04 \times 350 \times 0.3$ - 319 kcal
Heat losses during its flow to the preheater, 10% - 31.9 kcal
Heat content at the entrance of the preheater - 287.1 kcal

Temperature $= \frac{287.1}{3.04 \times 0.3} = 315^{o}C = 599^{o}F$

Temperature of the liquid at the inlet of the carbonization chamber 650°C (1202°F) $= 3.04 \times 0.3 \times 650 = 592$ kcal.

Heat losses during its flow from the outlet of the preheater to the inlet of the carbonization chamber, 5% $= 592 \times 0.05 = 29.6$ kcal

Heat content at the outlet of the preheater $592 + 29.6 = 621.6$ kcal

Temperature at the outlet of the preheater $\frac{621.6}{3.04 \times 0.3} = 683^{o}C$

Thermal units which must be transferred to the liquid during its flow through the preheater: $621.6 - 287.1 = 334.5$ kcal per kg of coal

Thermal efficiency of the carbonization chamber included the connecting pipes:

$$\frac{236.9}{334.5} \times 100 = 71.2\ \%$$

Gas consumption of the preheater:

The calculations are based on the utilization of a gas of the following properties:

					30" moist
higher heating value	4700	kcal per Nm^3	493	B.T.U.	per cu ft 60°F
lower " "	4170	" " "	433	"	" " " "
Theoretical consumption of air	4.27	Nm^3 per Nm^3	4.27	cu ft	per cu ft
10% less air	0.42	" " "	0.42	"	" " " "
Total air consumption	4.69	" " "	4.69	"	" " " "
Theoretical volume of flue gas	4.93	" " "	4.93	"	" " " "
Actual volume of flue gases	5.35	" " "	5.35	"	" " " "

Heat content of 1 Nm^3 of flue gas $\frac{4170}{5.35} = 780$ kcal

Heat content of 1 cu ft of flue gas $\frac{433}{5.35} = 81$ B.T.U.

The flue gases are supposed to leave the preheater at 425°C (797°F)
Heat content of 1 Nm^3 flue gas which leaves the preheater:
$1 \times 425 \times 0.38 = 162$ kcal
Thermal units available per 1 Nm^3 of flue gases:
$780 - 162 = 618$ kcal
Volume of flue gases which are required per 1 kg of shale:

$$\frac{1 \times 334.5}{618} = 0.542$$

Total heat content of the flue gas 0.542×780	= 422 kcal
Required for preheating liquid	334.5 "
Losses due to waste heat	87.5 "

Efficiency of the heating process $\frac{334.5}{442} \times 100 = 79.3\%$

Losses due to waste heat 20.7%

Heat efficiency of the total carbonization process $\frac{236.9}{422} = 56.2\%$

Volume of gas which must be burned per 1 kg of coal:

$$\frac{422 \times 1}{4,170} = 0.101\ Nm^3$$

Theoretical temperature of the flue gases at the entrance of the preheater:
2,040°C (3704°F)

Volume of the gases per hour:

Gas which must be burnod 286 Nm^3	286 Nm^3	10,850 cu ft, 60°F, 30" moist
Air required for the combustion	1,340 "	50,900 " " " " "
Flue gas	1,520 "	57,600 " " " " "

Actual volume of the flue gases:
At the entrance of the preheater: 12,900 cbm per hour
At the outlet " " " 3,890 " " "

Heat Balance of the Process

Furnished by the combustion of gas	422.0 kcal	100.00%
Losses due to temperature of waste heat	87.5 kcal	20.75%
Transferred to recirculating liquid	334.5 kcal	
Losses due to radiation of liquid		
before entering carbonization chamber	29.6 kcal	7.01%
after leaving carbonization chamber	31.9 kcal	7.56%
Transferred from liquid to carbonization chamber	273.0 kcal	
Losses due to radiation of carbonization chamber	36.1 kcal	8.55%
Transferred to coal	236.9 kcal	
Discharged by coke	115.0 kcal	27.25%
Discharged by oil vapors	29.1 kcal	6.89%
Discharged by benzol vapors	2.9 kcal	0.69%
Discharged by carbonization water	49.0 kcal	11.61%
Discharged by carbonization gas	9.7 kcal	2.30%
Discharged by moisture content of coal	31.2 kcal	7.39%

The heat losses due to radiation of the liquid can be lowered by an efficient insulation of the pipes which connect the heater with the carbonization chamber.

Sketch # 1 represents a diagram if the heat balance, and sketches # 2 and 3 show the principle of the proposed process.

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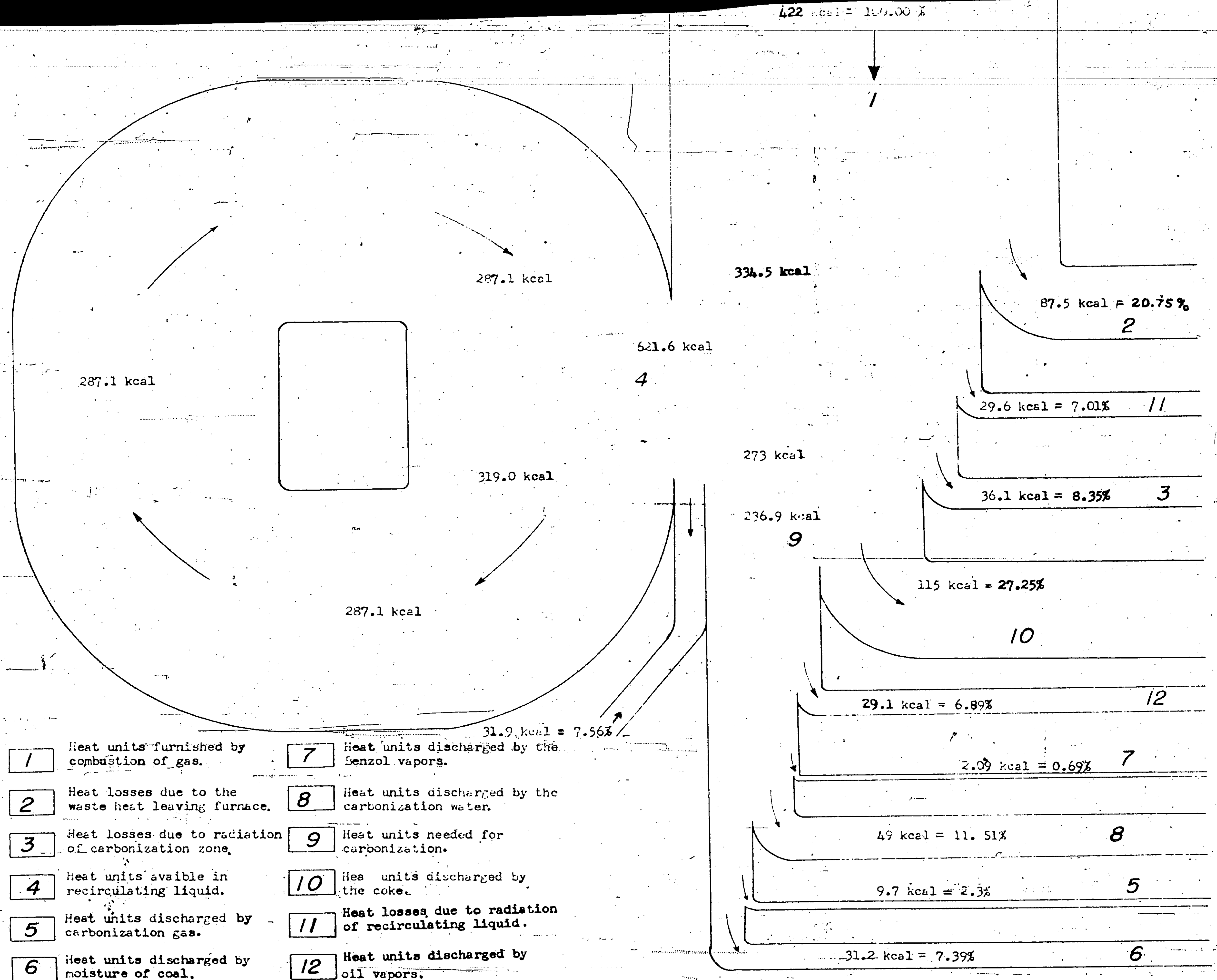
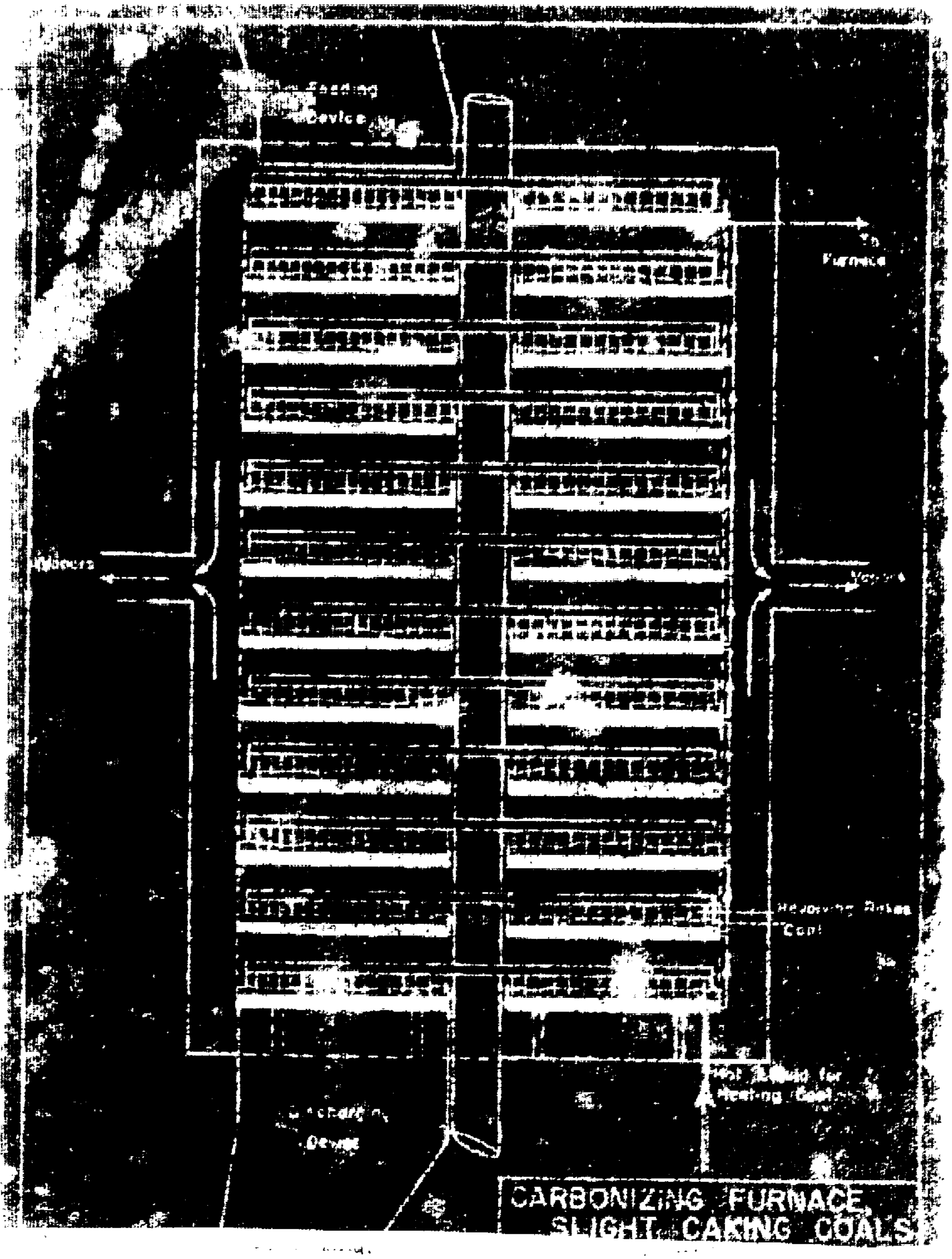
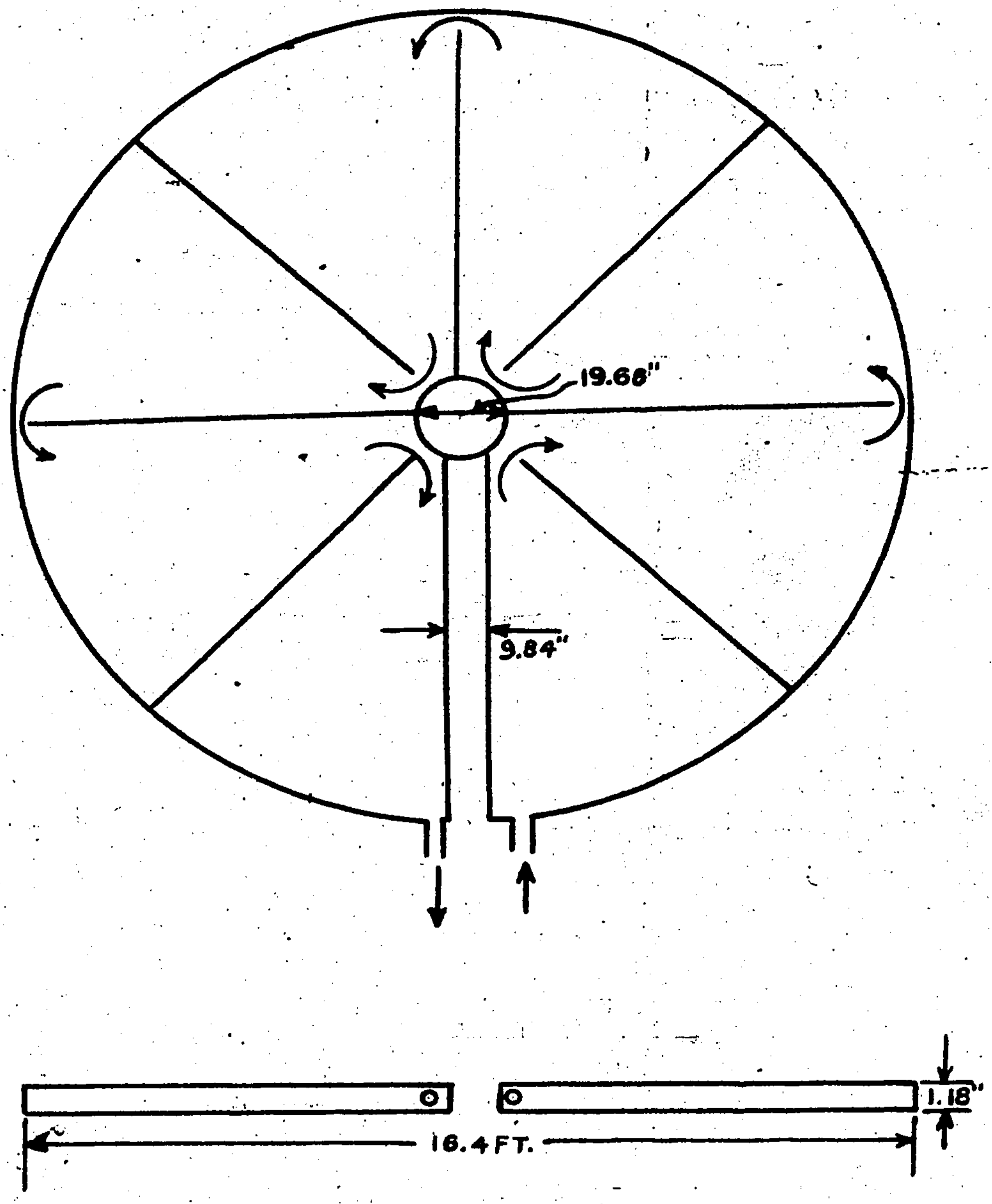


1	Heat units furnished by combustion of gas.
2	Heat losses due to the waste heat leaving furnace.
3	Heat losses due to radiation of carbonization zone.
4	Heat units avaible in recirculating liquid.
5	Heat units discharged by carbonization gas.
6	Heat units discharged by moisture of coal.
7	Heat units discharged by the Benzol vapors.
8	Heat units discharged by the carbonization water.
9	Heat units needed for carbonization.
10	Hea units discharged by the coke.
11	**Heat losses due to radiation of recirculating liquid.**
12	**Heat units discharged by oil vapors.**

HEAT BALANCE OF THE PROCESS



CARBONIZING FURNACE,
SLIGHT CAKING COALS



SECTION THROUGH HEATING CHAMBER OF THE COAL CARRYING PLATE

(20) Oppelt (20)

Determination of the commercial yields of gas, tar, benzol and ammonia of a coal utilizing a full scale coke oven.

Quantity of the liberated by-products during the course of the carbonization

The determination of the quantities of gas, tar, benzol and ammonia which are obtained by the commercial carbonization of a coal are in most cases performed in such a manner that the recovered by-products of an entire coke-oven battery are measured over a couple of days or weeks. With the known throughput of the battery the percentage of the recovered by-products can easily be calculated. Because it is often not possible to furnish certain kinds of coal in quantities large enough that a battery can be fed over the period which is necessary to obtain the desired results, other methods must be employed. Some coke oven plants are equipped with a small condensing and scrubbing system which can be connected with one of the ovens of the battery. Such installations are comparatively expensive. It is not only interesting to determine the yields but to investigate the quantities of by-products which are developed during the single hours of the carbonization. Such investigations provide valuable information about the course of the carbonization process which is necessitated for the most efficient design of the heating system of a coke oven.

A special method has been developed in the laboratory of the Otto Company which meets the above mentioned requirements and does not need the construction of an expensive condensing and absorption system.

The principle of the method is as follows:

The gas volume which flows through the ascension pipe of a coke oven is measured by means of a Pitot-tube which is made of quartz glass. A small volume of gas is sucked off from the ascension pipe in order to determine its content of tar, ammonia, benzol and its heating value, density and composition. The volumes of gas, water and oil vapors can be calculated from the before mentioned data.

Design of the Pitot tube

The Pitot tube consists of two separate parts, which are strung together by means of asbestos-threads. The single parts are made of quartz glass in order to secure a high resistance against temperatures and corrosion. The Pitot tube is inserted into the center of an ascension pipe and the differential pressure is determined by an extremely sensitive gauge which is capable to indicate pressures of 0.01 mm of head of water.

The formula for the calculation of a gas volume if a standardized Pitot tube is utilized reads as follows:

$$V = d^2 \times \frac{\sqrt{h}}{\sqrt{\gamma}}$$

where V is the actual gas volume in cbm per hour

d is the diameter of the ascension pipe in cm

h is the differential pressure in mm head of water

γ is the actual density of the gas inside the ascension pipe in kg per cbm

Since the quartz tube does not comply with the standardized sizes of a Pitot tube a correction factor must be applied which must be determined by trial. The factor of most of the tubes was 0.75, so that the complete formula reads as follows:

$$V = d^2 \times 0.75 \times \frac{\sqrt{h}}{\sqrt{\gamma}}$$

In order to determine the temperature and the pressure of the gas during its flow through the ascension pipe a thermocouple (connected with a galvanometer) and a small pipe (connected with a sensible pressure gauge) are inserted into the ascension pipe not far away from the pressure take off end of the

Pitot tube.

The length of the Pitot tube, the thermocouple and the pressure take off pipe depend on the width of the ascension pipe.

Determination of the composition of the gas

A small tube is inserted into the ascension pipe above the Pitot tube through which a gas sample is sucked out of the ascension pipe.

The gas passes through a condenser which is connected with two bottles, which are placed into an ice bath. Tar and water vapors are condensed and collected in the bottles. A glass tube which is stuffed with cotton wool or a small electrical precipitator removes the oil mist which is present in the gas. Two bottles which are partly filled with diluted sulphuric acid extract the ammonia vapors from the gas. The gas is purified from hydrogen sulfide by iron ore which is filled into a drying tower. Benzol vapors are completely extracted by active charcoal, which is placed in small iron containers. The purified gas is measured by a gas meter whereby pressure and temperature must be recorded. A side stream of the gas is sucked into a large aspirator bottle (approximately 100 liter = 3.5 cu. ft.) for the determination of the heating value of the gas by means of a calorimeter and the spec. gravity of the gas by a Bunsen-Schilling apparatus. The gas is sucked through the condensing and absorbing system utilizing a filter pump which is operated with compressed air.

Performance of the investigation

The ascension pipe of the chamber which shall be investigated is equipped with the borings for the introduction of the Pitot tube, thermocouple and so on.

The condensing system is set up and made ready for use but not yet connected with the ascension pipe. The coal which is fed to the chamber is weighed and a representative sample is taken for the determination of the moisture content of the coal and for additional analytical investigations. As soon as the chamber is filled with coal the test tubes are inserted into the ascension pipe and connected with the respective apparatus. The filter pump is put in operation and regulated in such a manner, that approximately 500 liters (15-17 cft) of gas per hour are sucked through the condensing system. The controlling instrument which regulates the volume of water which leaves the aspirator bottle is regulated in such a manner that the bottle is nearly filled with gas after one hour. The velocity of the gas flow must be kept uniform. It is supervised by measuring the gas volume which is drawn off per minute from time to time by observing the gas meter.

The temperature, absolute pressure and differential pressure are read every 2 or 5 minutes and recorded. In the meantime a second set of the condensing system is prepared and kept ready for use.

After one hour the condensing system is disconnected from the cooler and the already prepared system is used for the next hour. The absorption bottles, the tar filter and the benzol absorption tubes are carefully labelled and put aside for later investigation in the laboratory.

A calorimeter and a Bunsen-Schilling Apparatus are installed not far away from the battery. The aspirator bottle is brought to the test room and the gas content is shortly mixed by shaking the bottle. Two gas sample tubes are filled with gas, carefully labelled and set aside for investigation in the laboratory. The calorific value of the gas and its specific gravity are determined and recorded. Afterwards the aspirator bottle is brought back to the battery and prepared for further use.

Assuming that the carbonization time is 22 hours the following items are necessary:

44 bottles for tar and water condensate

22 tar filters

5 drying towers for iron ore

5 safety bottles for the removal of the last traces of hydrogen
sulfide

44 benzol absorption tubes

1 gas meter

2 aspirator bottles

1 filter pump

1 barometer

1 thermocouple with galvanometer

1 Pitot tube with differential pressure gauge

1 Pressure take off tube with pressure gauge

1 Condenser consisting of an iron tube and a water jacket
made of glass

1 iron tube to suck the gas from the ascension pipe

Ample supply of all fragile gadgets to make sure that the investigation is not to be interrupted due to lacking gadgets. Forms for the recording of the readings should be provided because they facilitate the computation and the compilation of the investigation.

Three chemists should always be present for the supervision of the test and for the determination of the heating value and the specific gravity of the gas.

The following pages represent the forms, which must be filled out during the course of the test (forms ## 1,2, 3, 4.)

Form #1

Readings for the computation of the gas volume

Date: 15 July 1946

Hour of carbonization: 1st

Time: 10:30 – 11:30

Time	Differential pressure (h) h	√h	Absolute pressure (p) mm head of water	Temperature t °C	Barometric pressure b mm Hg
10:30	2.55	1.59	5.0	625	746
10:35	2.65	1.63	4.5	626	
10:40	2.48	1.57	5.3	628	
10:45	2.50	1.58	5.2	626	
10:50	2.45	1.56	5.1	629	746
10:55	2.53	1.60	5.5	630	
11:00	2.38	1.54	5.3	630	
11:05	2.48	1.57	4.9	630	
11:10	2.79	1.67	5.0	629	
11:15	2.45	1.57	5.2	630	746
11:20	2.45	1.56	5.0	632	
11:25	2.52	1.59	5.1	632	
Sum		19.03	61.1	7545	2238
Average		1.586	5.1	629	746

Form #2

Gas volumes sucked through the condensing system

Date: 15 July 1946
Hour of carbonization: 1st
Time: 10:30 - 11:30

Time	Gas meter liter	Pressure (p) mm head of water	Temperature t °C	Barometric pressure (b) mm Hg
10:30	3125.3	- 5	27	746
10:35		- 6	27	
10:40		- 5	27.5	
10:45		- 5	27.5	
10:50		- 6	27.0	
10:55		- 6	27.5	746
11:00		- 5	27.5	
11:05		- 6	27.6	
11:10		- 5	27.5	
11:15		- 6	27.5	
11:20		- 6	27.6	
11:25	3638.5	- 5	27.5	746
Sum		- 66	328.7	2238
Average	513.2	- 5.5	27.4	746

Reduction of gas volume to normal conditions

p mm Hg: $\frac{5.5}{13.6} = -0.4$

b - p = 746 - 0.4 = 745.6 mm Hg

Pressure of saturated water vapor at 27.4° C = 27.4 mm Hg

Pressure for reduction 745.6 - 27.4 = 718.2 mm Hg

Factor for reduction: 0.8597

log. Factor: 0.9343 - 1

log gas volume: 2.71029

" factor : 0.93430 - 1

log red. gas volume 2.64459

Reduced gas volume 441.1 liter

Test #2

Specific gravity and density of the gas sample

Bunsen - Schilling

Date: 15 July 1948
Hour of carbonization: 1st
Time: 10:30 - 11:30

	Air Sec.	Gas Sec.
1.	69.3	45.0
2.	69.2	45.2
3.	69.3	45.1
Sum	207.8	135.3
Average	69.27	45.1

Spec. gravity (air = 1

$$\frac{\text{gas}^2}{\text{air}^2} = \frac{45.1^2}{69.3^2} = \underline{0.423}$$

Reduction of the density to normal conditions

Barometric pressure (b) 746 mm Hg

Average pressure of air and gas during the test P: 160 mm head of water

$$= \frac{160}{13.6} = 11.7 \text{ mm Hg}$$

Temperature in the apparatus: 27°C

Pressure of saturated water vapor at 27°C vp = 26.7 mm Hg

$b + p - vp = 746 + 11.7 - 26.7 = 730$ mm Hg

$f = 0.8740$ log $= 0.94150 - 1$

1.293 x 0.8740
log 1.293 = 0.11160
log 0.8740 = $\underline{0.94151}$ - 1
0.05311
= 1.130 kg per cbm

actual wt. of dry air : 1.130 kg per cbm
+ moisture content of air: 0.025 " " "
actual wt. of moist air : 1.155 kg per cbm

1.155 x 0.423
log 1.155 = 0.06258
log 0.423 = $\underline{0.62634 - 1}$
0.68892 - 1
= 0.4886 kg per cbm

actual wt. of the moist gas = 0.4886 kg/cbm
- moisture content = 0.0250 " "
actual wt. of dry gas 0.4636 " "

Weight reduced to normal conditions

0.4636 : 0.874
log 0.4636 = 0.66614 -1
log 0.8740 = $\underline{0.94151 -1}$ = $\underline{0.5304 \text{ kg per } m^3}$
0.72463 -1

Form 24

Heating value of the gas sample

Calorimeter

Date: 15 July 1946
Hour of carbonization: 1st
Time: 10:30 – 11:30

	Calorimeter Reading inlet	outlet
1.	15.36	25.41 °C
2.	15.36	25.45 "
3.	15.36	25.42 "
4.	15.37	25.46 "
5.	15.37	25.45 "
6.	15.36	25.43 "
7.	15.36	25.41 "
8.	15.37	25.45 "
9.	15.37	25.44 "
10.	15.37	25.40 "
Sum	153.65	254.32
Average	15.37	25.43

Barometric pressure (b) : 746 mm Hg

Gas pressure (p) : 6 mm head of water

Gas temperature : 24 °C

Gas volume applied : 10 liter

Quantity of water utilized : 4.340 kg

Temperature rise of the water : 25.43 − 15.37 = 10.06 °C

Heating value = $\frac{\text{Quantity of water} \times \text{temperature rise} \times 1000}{\text{gas volume}} = \frac{4.340 \times 10.06 \times 1000}{10}$

= 4366 kcal per cbm

Reduction of the heating value to normal conditions

$p = \frac{6}{13.6} = 0.44$ mm Hg

b = 746 mm Hg

− vp = 22.4 mm Hg

b = 746.00 mm Hg
+ p = 0.44 " "
746.44 mm Hg
−vp 22.40 " "
724.04 mm Hg

$f = \frac{760 \times (273 + 24)}{724.04 \times 273}$

log 760 = 2.88081
log 297 = 2.47276
5.35357
− 5.29592
log f = 0.05765
f = 1.142

log 724.04 = 2.85976
log 273 = 2.43616
5.29592

Reduced heating value = 1.142 x 4366

log 1.142 = 0.05765
log 4366 = 3.64008
3.69773

Reduced heating value: 4986 kcal/ Nm³

Determination of the [illegible] [illegible] in the Laboratory

1a) Composition of the gas

The gas samples which have been collected in the gas sample tubes are analysed whereby the content of CO_2, C_nH_m, O_2, CO, H_2, CH_4 and N_2 is determined. The results are listed. The list shows the alteration of the composition of the gas during the carbonization. The heating value and the density of the gas are registered together with the gas analysis (Form [illegible])

Determination of the [illegible] [illegible] in the Laboratory

1st Composition of the gas

The gas samples which have been collected in the gas sample tubes are analysed whereby the content of CO_2, C_nH_m, O_2, CO, H_2, CH_4 and N_2 is determined. The results are listed. The list shows the alteration of the composition of the gas during the carbonization. The heating value and the density of the gas are registered together with the gas analysis (form [illegible])

[illegible]

Dates: July [illegible]

[illegible]

The Quantities of tar and water condensate

The bottles which contain the tar and water condensate are weighed before the condensate is transferred to a separatory funnel. The last traces of oil condensate are removed by [illegible] which is poured into the separatory funnel. The empty bottles are weighed again. The difference of the weights is the quantity of oil and water condensate which was present in the gas volume, which has been sucked through the condensing system. The oil water mixture is diluted with [illegible] in order to separate the water from the oil condensate. The water is drawn off and weighed. The oil which is nearly free from water is transferred to a distilling flask where the [illegible] together with the water is removed by distillation. The quantity of water which can now be calculated.

Example:

Bottle #1 incl. tar + water	[illegible] g	Bottle #2 incl. tar + water	[illegible] g
" " empty	[illegible] -	" " empty	[illegible] -
Water + tar condensate	[illegible] g		[illegible] g

Total condensate [illegible] g

Water drawn off [illegible] g

Water determined by distillation 6.0 g

Total quantity of water [illegible] g

Total quantity of tar [illegible] - [illegible] = [illegible] g

Additional tar has been extracted from the gas by the tar filter. The cotton wool of the tar filter is charged into an extraction thimble and the tar is extracted by boiling chloroform. The chloroform is evaporated and the quantity of the tar residue is determined by weighing the extraction flask.

Example:

Extraction flask with tar residues [illegible] g

" " empty [illegible] g

Tar residue [illegible] g

Total quantity of tar [illegible] + [illegible] = [illegible] g

The quantities of tar and water which have been condensed during the single hours of the test are listed, and calculated on 1 Nm³ of gas.

Form #8

Date : 15/16 July 1948

Water and Ice Contents

Hour of observation	Time	[illegible] m³	Water %	Water g per m³	Ice %	Ice g per m³
1	10:30-11:30	0.445	375	850	25	120
2	11:30-12:30					
3	12:30-13:30					
4	13:30-14:30					
5						
6						
22	7:30- 8:30					

Determination of the Ammonia

The content of the ammonia absorption bottles is poured into a volumetric flask (volume 1000 ml) the water condensate which has been collected from the tar-water-condensing bottles is added and the flask is correctly filled up with distilled water. 50 ml of the solution are pipetted into a distilling flask, a solution of sodium hydroxide is added and the vapors are distilled into a known volume of a normal solution of sulfuric acid. The surplus acid is neutralized by a normal solution of sodium hydroxide. The quantity of ammonia can now be calculated from the consumption of sulfuric acid.

Example:

applied normal solution of sulfuric acid :	25.00 ml
necessary for neutralizing normal sol. of sod. hydr.:	9.40 "
consumed normal solution of sulfuric acid	15.60 ml

Quantity of ammonia which is present in 50 ml of the solution of ammonia = 15.6 x 0.017 = 0.2652 g

Quantity of ammonia which is present in 1000 ml of the solution of ammonia = 0.2652 x 20 = 5.304 g

The quantity of 5.304 g ammonia was present in 0.441 Nm^3 gas.

Quantity of ammonia which is present in 1 Nm^3 gas

$$= \frac{5.304 \times 1}{0.441} = 12.03 \text{ g per } Nm^3$$

The figures which pertain to the single hours are listed.

Dated: 15/16 July 1945

Form #7

Content of Ammonia

Hour of carbonization	Time	Gas Nm^3	NH_3 g	NH_3 g per Nm^3
1	10:30-11:30	0.441	5.304	12.03
2	11:30-12:30			
3	12:30-13:30			
4				
5				
6				
7				
22	7:30- 8:30			

Determination of the benzol vapors

The determination of the benzol vapors is similar to the method which is applied if the benzol content of coal gas is determined. Vapors of hydrocarbons are absorbed from active charcoal and can be removed by blowing steam through the container which contains the coal. Steam and hydrocarbon vapors are condensed. The condensed vapors are collected in a burette. Since the hydrocarbons are insoluble in water and have a lower specific gravity than water they are separated from the condensed water and form a layer, which swims on top of the water. The volume of the condensed hydrocarbons is determined and transferred into weight by multiplication with the specific gravity.

Example :

Container #1 : liquid hydrocarbons 11.7 ccm

" #2 : " " 3.3 "

Total 15.0 ccm

Specific gravity of the hydrocarbons 0.863

Weight of the hydrocarbons 15.0 x 0.863 = 12.95 g

Content of hydrocarbons of 1 Nm^3 of gas : $\frac{12.95 \text{ x } 1}{0.441}$ = 29.4 g

The results of the investigation of the single hours are listed (Form #8).

Dates: 15/16 July, 1946

Form #6

Content of liquid hydrocarbons

Spec. gravity 0.863

Hour of carbonization	Time	Gas Nm^3	Liquid hydroc. ccm	Liquid hydroc. g	Liquid hydroc. g per Nm^3
1	10:30-11:30	0.441	15.0	12.95	29.4
2	11:30-12:30				
3	12:30-13:30				
4					
5					
6					
23	7:30-8:30				

Determination of the gas volume which flows through the ascension pipe.

The gas volume can be calculated using the following formula:

$v = d^2 \times f \times \frac{\sqrt{h}}{\sqrt{\gamma}}$ v= Actual volume of the gas per hour which flows through the ascension pipe.

d= diameter of the ascension pipe in cm

f= factor of the Pitot tube which must be determined by trial.

h= differential pressure indicated by the pressure gauge (in mm head of water).

γ = actual gravity of the gas (kg per cbm)

Since the actual gravity of the gas cannot be directly measured it must be calculated from various data which can be determined.

Example:

diameter of the ascension pipe 30 cm

average temperature of the gas (Form #1) 629°C

average differential pressure $\sqrt{h}$ (Form #1) 1.585

average absolute pressure (p)(Form #1) 5.1 mm head of water =0.4 mm Hg

average barometric pressure (Form #1) 746 mm Hg

weight of the gas determined

in the Bunsen-Schilling apparatus (Form #3) 0.5504 kg per Nm^3

water content of the gas (Form #6) 880 g per Nm^3 = 0.880 kg per Nm^3

tar content of the gas (Form #6) 120 g per Nm^3 = 0.120 kg per Nm^3

Method of procedure:

The water content of the gas which has been determined by weight must be transferred into volume:

18 kg of water equal 22.4 Nm^3 steam

1 " " " equals x " "

$x = \frac{22.4 \times 1}{18}$

log 22.4 = 1.35025

log 18 = 1.25527

log x = 0.09498

x = 1.224 1 kg of water equals 1.244 Nm^3 of steam

Volume of steam which has been condensed calculated on 1 Nm^3 of

gas = 0.850 x 1.244

log 0.850 = 0.92942 - 1

log 1.244 = 0.09482

log v = 0.02424

v = 1.057 Nm^3

1 Nm^3 gas + 1.057 Nm^3 steam (2.057 Nm^3) weigh 0.5304 + 0.850 = 1.3804 kg

1 Nm^3 of the mixture which flows through the ascension pipe weighs $\frac{1.3804}{2.057}$ kg

log 1.3804 = 0.14000

log 2.057 = 0.31323

0.82677 -1

= 0.6711 kg per Nm^3

Calculation of the actual density of the gas:

$$f = \frac{273 \times (746 + 0.4)}{(273 + 629) \times 760} = \frac{273 \times 746.4}{902 \times 760}$$

log 273 = 2.43616

log 746.4 = 2.87297

5.30913

- 5.83602

log 902 = 2.95521

log 760 = 2.88081

5.83602

log f 0.47311 -1 f = 0.2972

actual density = 0.6711 x 0.2972

log 0.6711 = 0.82679 - 1
log 0.2972 = 0.47311 - 1
0.29990 - 1

actual density 0.1995 kg per cbm

Volume of the gas per hour which flows through the ascension pipe:

$$v = d^2 \times 0.75 \times \frac{\sqrt{h}}{\sqrt{\gamma}} = 30^2 \times 0.75 \times \frac{1.585}{\sqrt{0.1995}}$$

$$\sqrt{0.1995} = \tfrac{1}{2} \log 0.1995 = \tfrac{1}{2} \times 9.29990 - 10 = 4.64995 - 5$$

$$= 0.64995 - 1$$

$$= 0.4466$$

log 900 = 2.95424

log 0.75 = 0.87506 − 1

log 1.585 = 0.20003

3.02933

− log 0.4466 = 0.64995 − 1

3.37938

= 2395 cbm per hour

$= 2395 \times 0.2972$ Nm^3 per hour

log 0.2395 = 3.37938

log 0.2972 = 0.47311 − 1

2.85249

= 712 Nm^3 per hour consisting of carbonization gas and water steam

$$= \frac{1 \times 712}{2.057}$$

log 712 = 2.85247

log 2.057 = 0.31323

2.53926

Gas = 346.1 Nm^3 per hour

$$\frac{1.057 \times 712}{2.057}$$

log 1.057 = 0.02424

log 712 = 2.85248

2.87673

log 2.057 0.31323

2.56350

water vapor = 366 Nm^3 per hour

$$= \frac{366}{1.244}$$ kg of water condensate

log 353 = 2.54830

− log 1.244 = 0.09482

2.45348

= 284.1 kg water condensate

The gas contains in addition to the water vapors a certain quantity of tar vapors which influence its density and volume.

The following calculation considers both the water and the tar vapors:

In order to be able to convert the weight of the tar into volume its molecular weight must be known.

Assuming a molecular weight of 200, the volume of the tar vapors is as follows: $\frac{22.4 \times 120}{200}$ = liter per Nm^3 gas

log 22.4 = 1.35025

log 120 = 2.07918

3.42943

− log 200 2.30103

log v = 1.12840

v = 13.44 liter

1 Nm^3 gas + 1.057 Nm^3 steam + 0.0134 Nm^3 tar vapors (2.0704 Nm^3) ~~weigh~~ 0.5204 + 0.860 + 0.120 = 1.5004 kg.

1 Nm^3 of the mixture which flows through the ascension pipe weighs

$\frac{1.5004}{2.0704}$ kg

log 1.5004 = 0.17621

log 2.0704 = 0.31605

0.86016 − 1

0. 7247 kg per Nm^3

actual density of the gas = 0.7247 x 0.2972

log 0.7247 = 0.86015 - 1

log 0.2972 = <u>0.47311 - 1</u>

0.33327 - 1

actual density of the gas = 0.2154 kg per cbm

Volume of the gas per hour which flows through the ascension pipes

$$v = 900 \times 0.75 \times \frac{1.585}{\sqrt{0.2154}}$$

$$\sqrt{0.2154} = \frac{1}{2} \log 0.2154 = \frac{1}{2} \times 9.33327 - 10 = 4.66663 - 5$$

$$= 0.66663 - 1 = 0.4641$$

log 900 = 2.95424

log 0.75 = 0.87506 - 1

log 1.585 = <u>0.20005</u>

3.02935

- log 0.4641 <u>0.66665 - 1</u>

3.36270

= 2305 cbm per hour

= 2305 x 0.2972 = Nm^3 per hour

log 2305 = 3.36270

log 0.2972 = <u>0.47311 - 1</u>

2.83581

685.2 Nm^3 per hour consisting of carbonization gas, water steam and tar vapors.

Gas $= \frac{1 \times 685.2}{2.0704}$ Nm^3

log 685.2 = 2.83581

log 2.0704 = $\underline{0.31605}$

2.51976

= 331 Nm^3 per hour

Steam $= \frac{1.057 \times 685.2}{2.0704}$

log 1.057 = 0.02434

log 685.2 = $\underline{2.83581}$

2.86015

log 2.0704 = $\underline{0.31605}$

2.54410

= 349.95 Nm^3 per hour

$= \frac{349.95}{1.244}$ kg water condensate

log 349.95 = 2.54410

log 1.244 = $\underline{0.09482}$

2.44928

281.2 kg per hour water condensate

Tar vapors $= \frac{0.0134 \times 685.2}{2.0704}$

log 0.0134 = 0.12840 - 2

log 685.2 = $\underline{2.83581}$

0.96421

log 2.0704 = $\underline{0.31605}$

0.64816

= 4.448 Nm^3 per hour

$= \frac{4.448 \times 200}{22.4}$ kg tar condensate

log 4.448 = 0.64816

log 200 = $\underline{2.30103}$

2.94919

log 22.4 $\underline{1.35025}$

1.59894

39.71 kg tar condensate per hour

Comparison of the two methods of calculation:

	without tar	with tar
Gas Nm^3 per hour	366.1	331
Steam " " "	366	349.95
Tar vapors " " "	–	4.446
Steam kg " "	294.1	281.2
Tar kg " "	41.5	39.7

As the table shows the tar content of the gas is essential if correct figures should be obtained. Even if the assumed molecular weight of the tar (200 g per mol.) should not be correct it is better to include the tar in the calculations.

Yield of Ammonia

The yield of ammonia is obtained by multiplication of the gas yield per hour with the ammonia content of 1 Nm^3 gas.

Example:

331 x 12.03 = 3982 g = 3.982 kg

Yield of Benzol

The yield of benzol is obtained by multiplying the gas yield per hour with the benzol content of 1 Nm^3 gas.

Example:

331 x 29.4 g = 9731 g = 9.731 kg

The results are listed, they show the formation of gas, water, tar, ammonia and benzol during the carbonization. The sum of the yields per hour represents the total yield which has been obtained by the carbonization of the coal charge of an oven (Form #9).

[illegible]

Dates: 15/16 July 1945

Yields of Byproducts per hour

Hour of carbonization	Time	Gas m^3	Water kg	Tar kg	Benzol kg	Ammonia kg
1	10:30-11:30	331	281.2	35.7	9.721	3.982
2	11:30-12:30					
3	12:30-13:30					
22	7:30-8:30	150	0.2	0.1	0.2	0.0
Total		5,710	2,910.0	600.0	170.0	55.5

In order to be able to compare the yields which are obtained by the carbonisation of various coals the yields must be calculated per ton of dry coal.

Fed to the chamber : 20.0 tons of moist coal

Moisture content 10.5 %

Dry coal 20.0 x 0.895 = 17.9 tons

Gas yield per hour, per ton of dry coal: $\frac{331}{17.9}$ = 18.5 Nm^3

Water condensate per hour per ton of dry coal $\frac{281.2}{17.9}$ = 15.75 kg

Tar yield per hour, per ton of dry coal $\frac{39.7}{17.9}$ = 2.22 kg

Benzol yield per hour, per ton of dry coal $\frac{[illegible]}{17.9}$ = 0.345 kg

Ammonia yield per hour per ton of dry coal $\frac{[illegible]}{17.9}$ = 0.222 kg

The results as listed, they show the formation of gas, water, tar, ammonia and crude benzol per hour, per ton of dry coal. The sum of the figures represents the yield which has been obtained by the carbonisation of 1 ton of dry coal. (Form #10).

Dates 15/16 July 1946

Form #10

Yields of By-products per hour, per ton of dry coal.

Hour of carbonization	Time	Gas Nm^3	Water kg	Tar kg	Benzol kg	Ammonia kg
1	10:30-11:30	18.5	15.75	2.22	0.545	0.222
2	11:30-12:30					
3	12:30-13:30					
4						
5						
22	7:30-8:30	3.5	0.3	0.0	0.000	0.000
Total		320.0	163.0	33.5	9.500	3.100

Water condensate 16.3 %

Tar 3.35 %

Crude Benzol 0.95%

Ammonia 0.31%

The figures which have been compiled in the lists should be evaluated by diagrams. If the yields, the composition and the properties of the gas are plotted against the "hour of carbonization" the obtained curves show very clearly the process of the carbonization in a full scale coke oven. It is even possible to observe the formation of a single constituent of the gas (for instance the formation of methane) depending on the "hour of carbonization" or to find out connections between the formation of various constituents.

Numerous investigations have shown that there is no uniform production of by-products during the course of the carbonization process. Especially much water vapors escape at the end of the first half of the carbonization time and an accumulation of tar and benzol vapors has been observed at a time at which two thirds of the total carbonization time have been terminated. As an example sketch #5 represents the diagram of the composition, the heating value, and the density of the gas depending on the carbonization time.

Furthermore, it is possible to insert thermocouples into the coal charge and to determine the rise of the temperature inside the coal charge depending on the progress of the carbonization time.

The investigation should be completed by the performance of laboratory tests. The following tests give valuable information about the properties of a coal:

proximate analysis
ultimate analysis
determination of the softening point
Foxwell curve
determination of the yields of by-products (compare the report entitled "Coal carbonization in the Laboratory" July 19, 1946)

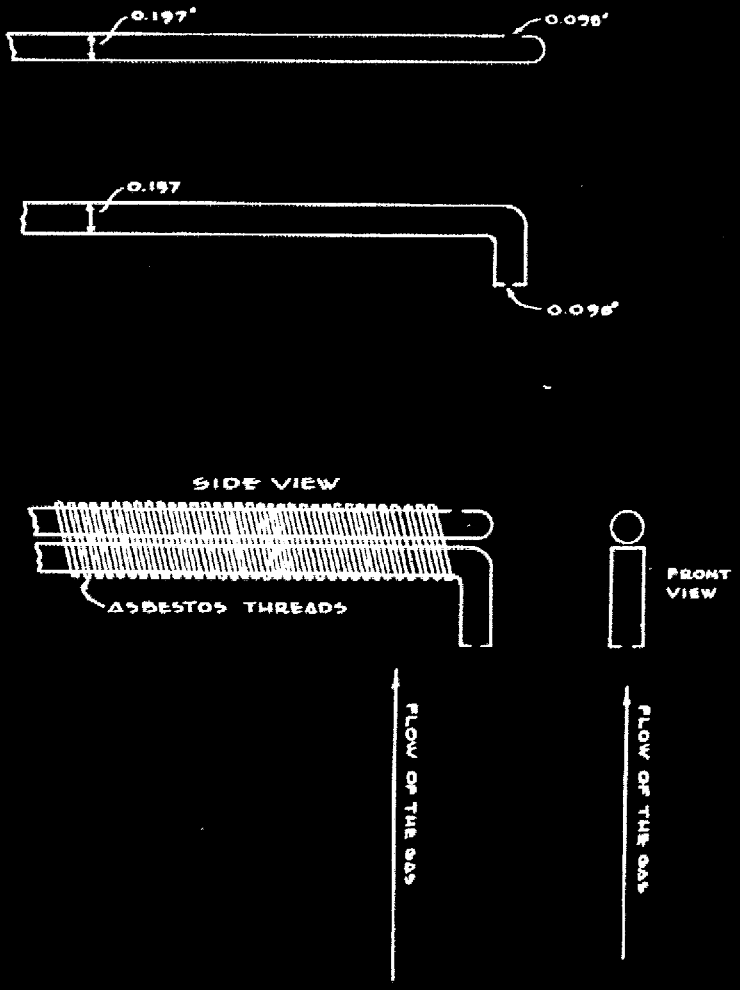
Swelling properties of the coal

Caking properties of the coal

The correct performance of the test requires skilled chemists and good teamwork.

[redacted]

[redacted]


0.197"
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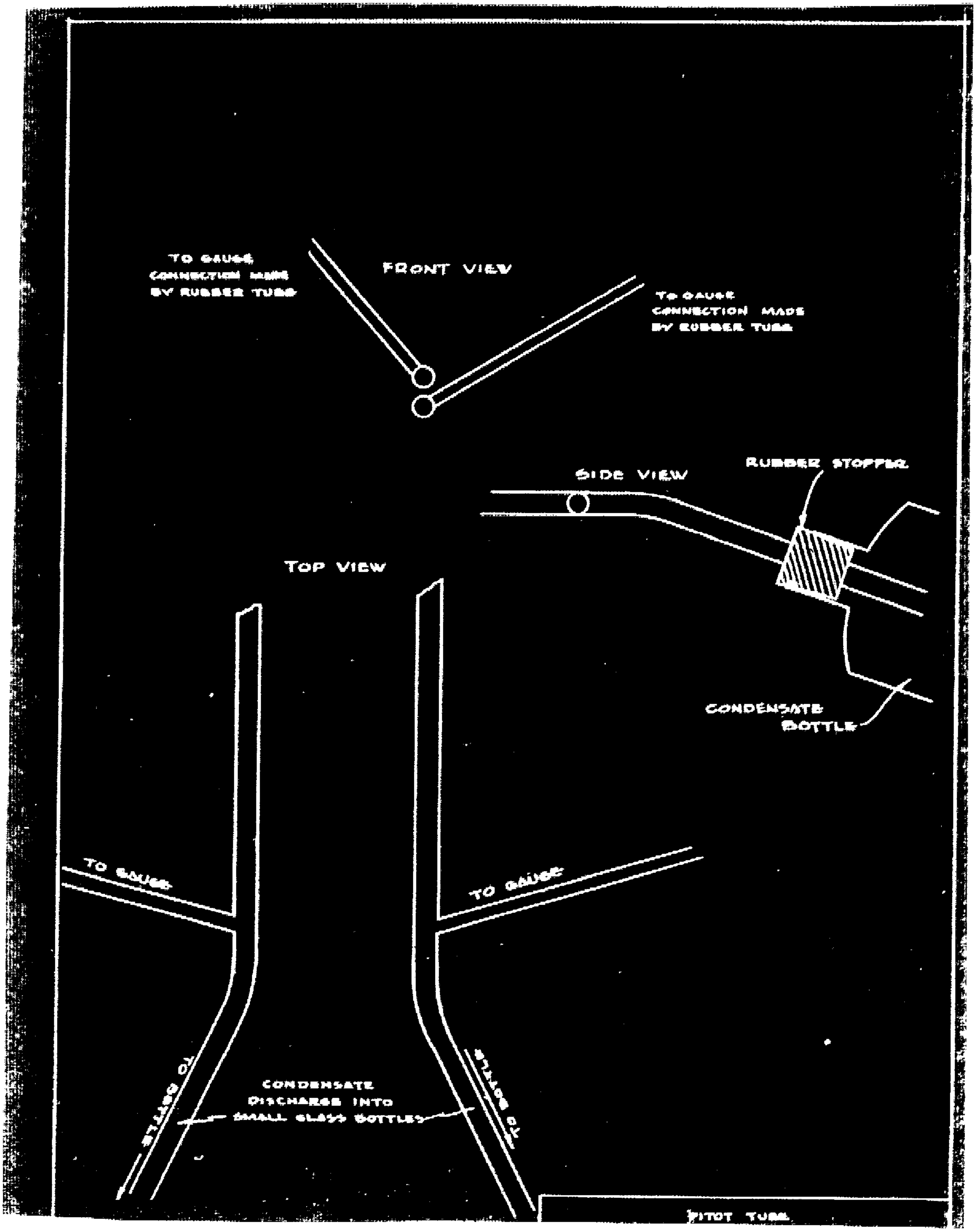
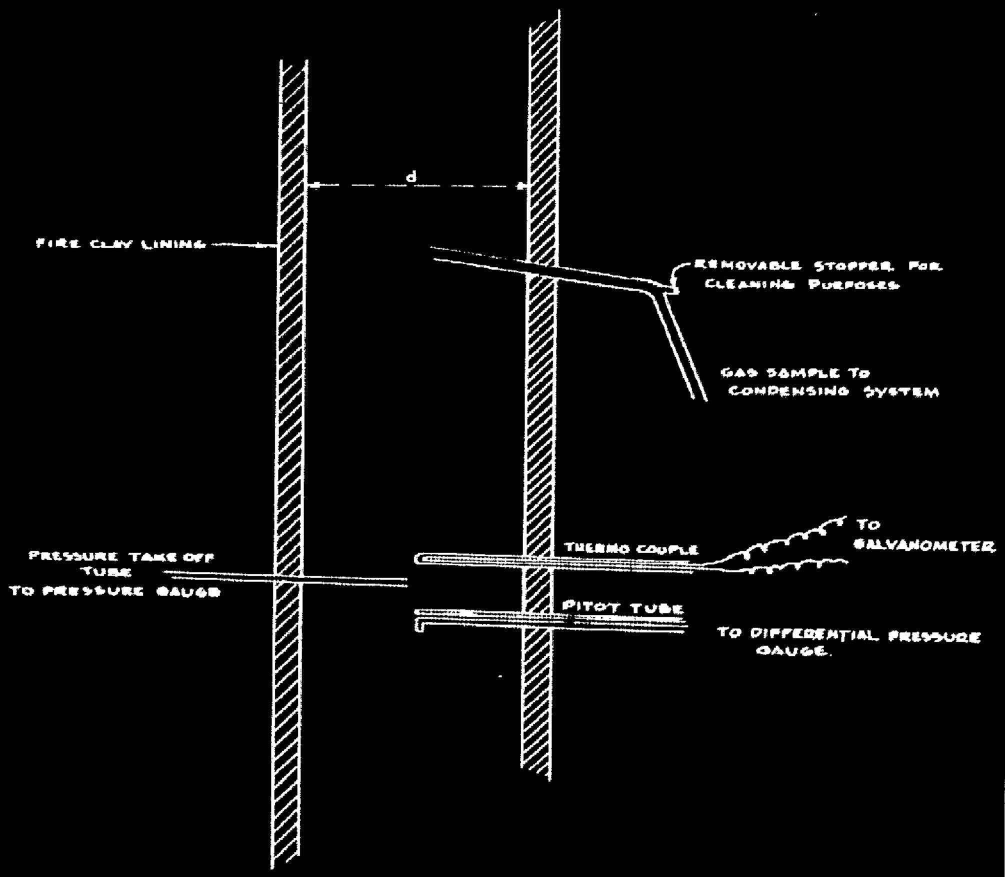
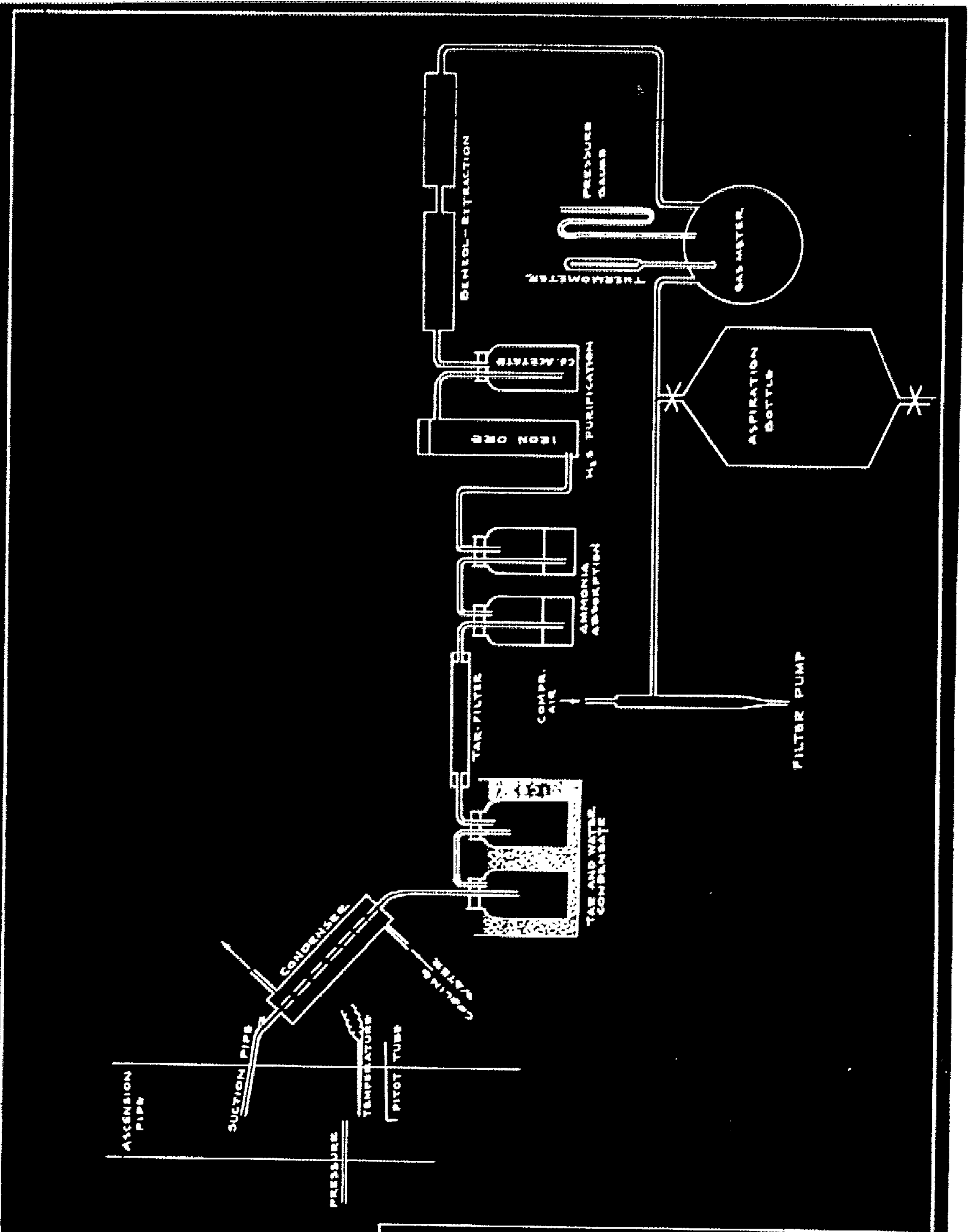
SIDE VIEW
ASBESTOS THREADS
FRONT VIEW
FLOW OF THE GAS
FLOW OF THE GAS

PITOT TUBE

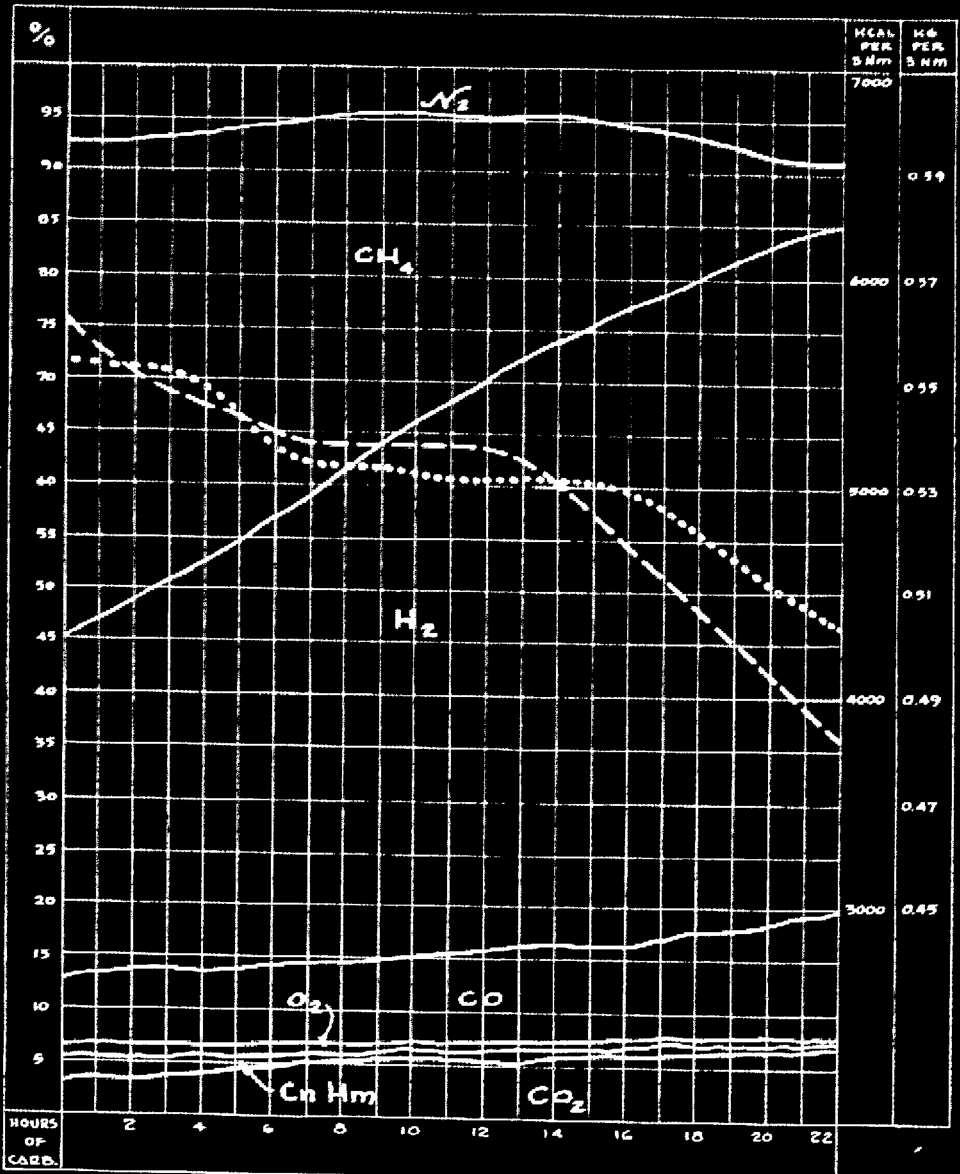

TO GAUGE CONNECTION MADE BY RUBBER TUBE
FRONT VIEW
TO GAUGE CONNECTION MADE BY RUBBER TUBE
SIDE VIEW
RUBBER STOPPER
CONDENSATE BOTTLE
TOP VIEW
TO GAUGE
TO GAUGE
TO BOTTLE
CONDENSATE DISCHARGE INTO SMALL GLASS BOTTLES
TO BOTTLE
PITOT TUBE



PREPARATION OF THE ASCENSION PIPE



CONDENSING AND ABSORBING SYSTEM



HEATING VALUE OF THE GAS — — —
DENSITY OF THE GAS ○○○○○○

COMPOSITION AND PROPERTIES OF THE GAS

21 Oppelt

21

Determination of the commercial yields of gas, tar, benzol and ammonia of a coal utilizing a full scale coke oven.

Quantity of the liberated by-products during the course of the carbonization.

Second Method (Compare the report dated 29 July 1946)

The possibilityto make exact measurements utilizing a Pitot tube depends on the velocity of the gas which flows through the ascension pipe. If the velocity is too low the differential pressure becomes so small that it is impossible to read the pressures which are indicated by the pressure gauge. A method which is often applied if the volumes of water must be measured which flow in a river bed consists of adding an exactly known quantity of a salt solution (Na Cl) during a definite period of time and the determination of the content of chlorine ions before and after the admixture of the salt solution. It is now possible to compute the quantity of water from the alteration of the content of chlorine ions. A similar method was applied for the determination of the volumes of gas which leaves the oven through the ascension pipe. Since it is not possible to spray in a solution of salts into the hot gases steam was utilized as a diluting agent. The steam content of the gas before and after the introduction of the steam can be determined fairly exact by cooling the gas and collecting the condensate. The gas volumes can be computed from the alteration of the water content of the gas before and after the introduction of the steam.

The arrangement consists of a complete condensing and extraction system before the admixture of the steam, the gadget which is necessary to introduce a constant flow of steam and another condensing system for the determination of the water content of the gas which has been diluted with steam.

The complete condensing and extraction system is the same as used by the previously described method (report dated 29 July 1946). A thermocouple is inserted close to the gas take off pipe in order to determine the temperature of the gas which flows through the ascension pipe.

The device which is used for the introduction of the steam consists of a tube which is inserted into the center of the ascension pipe, a two-way stopcock which is connected with a condenser, an orifice which controls the volume of the steam which is blown through the pipe, a sensitive pressure gauge in order to be able to maintain a constant pressure of the steam and a valve which connects the gadget with the main steam pipe. (Sketch #1). Sometimes the two-way stopcock is connected with the condenser, the steam which flows through the condenser is condensed, the condensate is collected in a graduated cylinder and measured. With the exact time known the total volume of steam which was blown into the ascension pipe can be computed. The diameter of the orifice must be determined by trial. It depends on the volume of the gas which flows through the ascension pipe per unit of time.

In a certain distance above the steam introduction another condensing system is arranged which consists of a condenser, two collecting bottles, a filter tube, a gas meter, and a filter pump which sucks the gas through the system.

Sketch #2 represents the complete arrangement.

Example:

Control of the introduced steam:

Water condensate: 250 ml = 250 g

Time: 30 seconds

Quantity per hour = 0.250 x 2 x 60 = 30.0 kg

With the exception of the determination of the gas volume the determination of the yields of tar, benzol and ammonia and the composition of the gas follows closely the method which has been described in the report dated 29 July 1946.

The determination of the oil and water content of the steam enriched gas is performed according to the directions which have been given on the pages 12 and 14 of the report dated 29 July 1946.

Calculation of the gas volume:

Water content of the gas before the introduction of the steam 850 g per Nm^3

Water content of the gas after the introduction of the steam 945 g per Nm^3

Steam which has been admixed to the gas 95 g per Nm^3

Total quantity of steam which has been admixed during 1 hour 30,000 g

Volume of gas which left the furnace per hour:

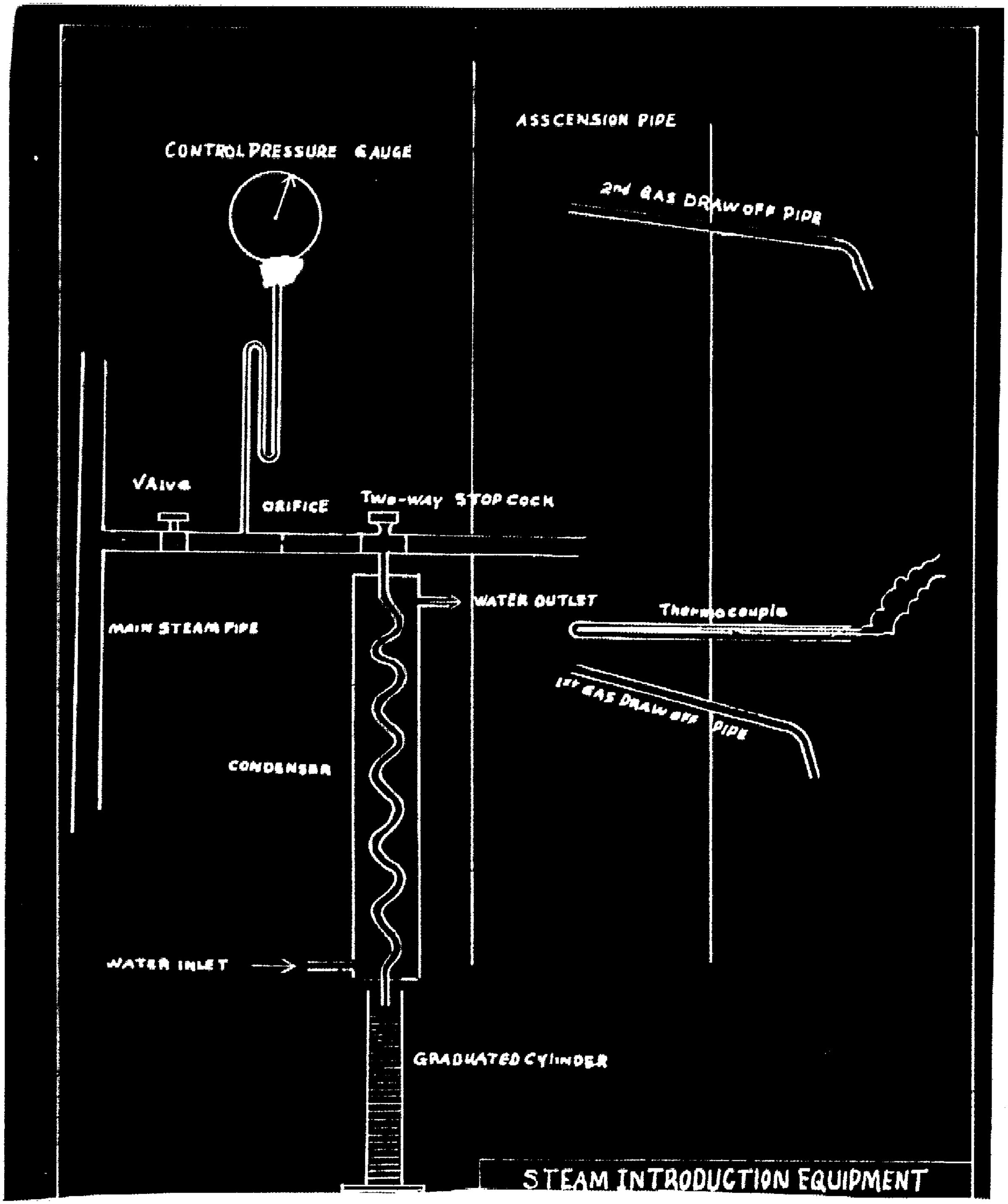
$$\frac{30{,}000}{95} = 317\ Nm^3 \text{ per hour}$$

The computation of the gas volume is easier to perform applying the above described method than to carry out the rather complicated calculations if a Pitot tube is employed.

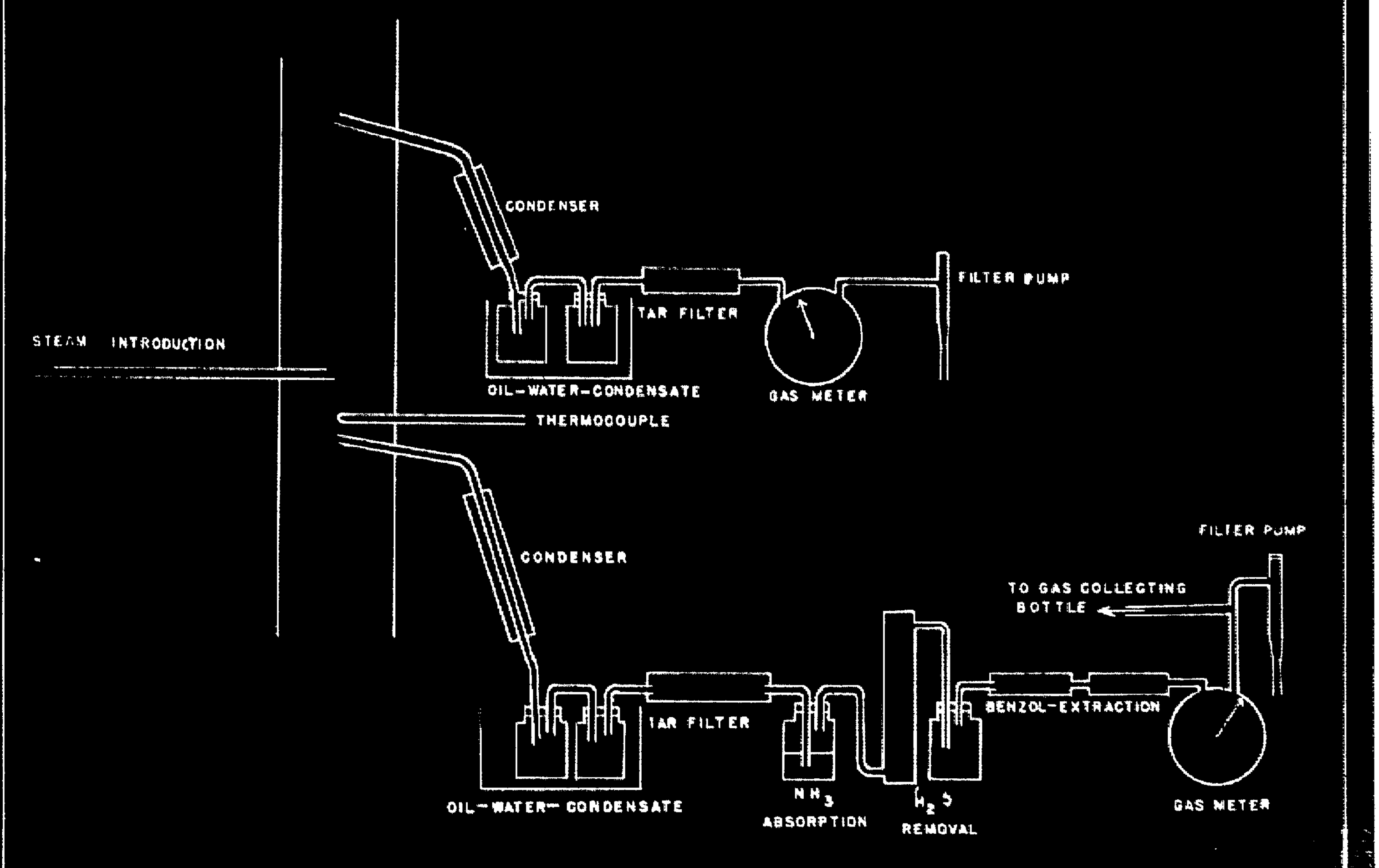
The accuracy of the method is supposedly the same as if a Pitot tube is applied because the determination of the true density of the gas requires a careful preparation and supervision of the analyses.

Applying the above described method a completely uniform distribution of the introduced steam must be secured and the gas which is sucked through the condenser must be cooled as deep as possible in order to secure its efficient removal from the gas.

[redacted]



STEAM INTRODUCTION EQUIPMENT



ARRANGEMENT FOR THE DETERMINATION OF THE GAS YIELD

(22)

(22)

ADDRESS ALL COMMUNICATIONS TO
THE DIRECTOR, U. S. BUREAU OF MINES
WASHINGTON 25, D. C.

UNITED STATES
DEPARTMENT OF THE INTERIOR
BUREAU OF MINES
WASHINGTON 25, D. C.

REC'D. ....

TIIC L.F. & L. S-C.

August 22, 1946.

MEMORANDUM for L. L. Newman.

Subject: Krupp-Lurgi Low Temperature Carbonization Equipment.

On July 26 I called at the Lurgi office at Frankfort, Germany, to obtain information requested in your letter of June 19 on the design of the circulating fan for the Krupp-Lurgi low-temperature carbonization process. I talked to Dr. F. A. Oetkin who gave me the following information.

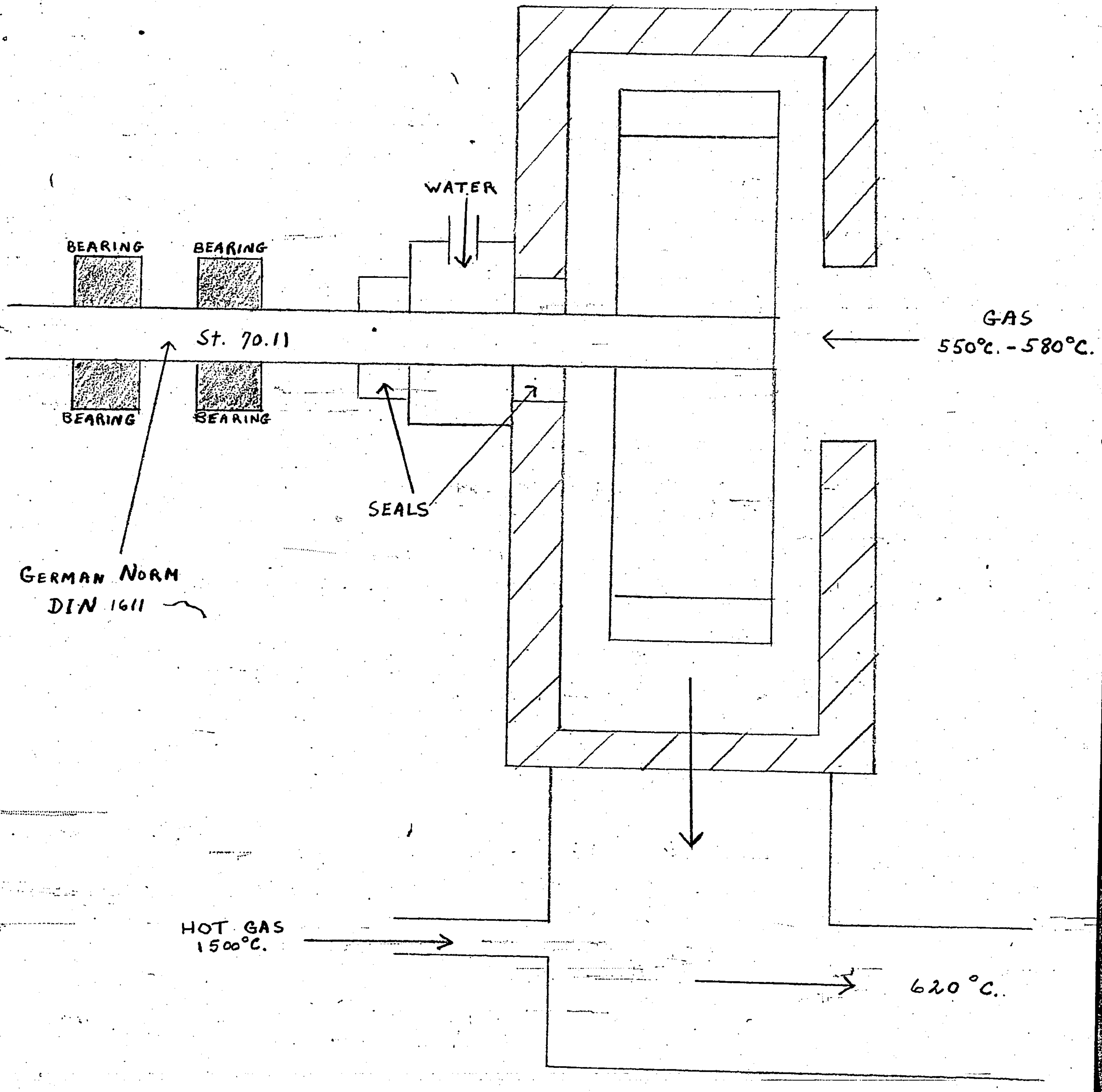
The gases enter the recirculating fan at a temperature of 550° to 580°C. and at this point never approach the 800° mentioned in your letter. After leaving the fan, the gases are mixed with combustion gases (temperature 1500°C.) which raise the temperature to about 620°C. The fan is of the centrifugal type with blades made of Krupp FKB steel. The fan housing is lined with insulating brick. The fan shaft is supported by two bearings outside of the fan housing and both on the same side of the fan housing. The end of the shaft facing the gas inlet floats. The shaft is sealed with asbestos packing at the housing and by another seal outside the housing with Burgmann packing. The space between these two seals is totally enclosed and in this space the shaft is cooled by a stream of water. A rough sketch of the recirculating fan is attached.

In discussing the process in general, Dr. Oetkin said that he would favor the Lurgi Spulgas Process for the younger coals. With the Krupp-Lurgi process using "gas flame" coal it was difficult to make good coke without the addition of coking coal.

J. D. Doherty

J. D. DOHERTY.

Enclosure 264.


WATER
BEARING
BEARING
BEARING
BEARING
St. 70.11
GAS
550°C.-580°C.
SEALS
GERMAN NORM
DIN 1611
HOT GAS
1500°C.
620°C.

[redacted]

EXHAUSTER FOR HOT GAS

KRUPP-LURGI-RETORT

The exhauster is handling hot combustion gas with A-3% oxygen. The gas enters with 500-550°C. The design of the exhauster is as normally used for flue gas of boilers. Details of construction have to make allowance only for the high temperature which weakens normal steel and even most alloyed steel so much, that is cannot be used for the wheel of the fan. This wheel is made of nickel-chrom alloyed steel, which keeps sufficient strength and elasticity even with 600°C. Specialists for manufacturing and steelproducer have to choose from available steels. The wheel should be rivitted unless the manufacturing shops owns enough experience with welding alloyed steel. Special welding wire material is necessary to avoid breaking under working conditions. Possibly the wheel must be equalized after welding, be heated to 800°C. It is advisable to compromise efficiency of the fan against strength of the construction in order to avoid heavier sheet and weak connections of same.

The shafts can be made of lower grade alloyed steel. The connection between wheel and shaft constructed for reduced heat transmission. That part of the shaft inside the housing may be protected against heat transmission from the flue gas.

Bearings should be ball or roller bearings water cooled.

For tightening of the shaft carbon rings are used.

The housing of the fan may be made of medium alloyed steel (6-8% chrom), and insulated outside or with fans of great dimensions it may be normal steel with an inner insulating layout.

23 GRAF Graf

23

Research experiments to detoxicate illuminating gases (Removal of carbon monoxide)

I. Introduction

Before 1913 in Europe carbonization gases which originated from the dry distillation of coal were almost exclusively used for domestic purposes. During the following decade the content of carbon monoxide amounted from approximately 8 per cent up to approximately 15 to 20 per cent, due to the admixture of increasing volumes of water gas thus increasing the poisonous properties of the illuminating gases. According to experiments of Flury-Zernik ("Die schaedlichen gase, Edition Springer, Berlin 1931) a CO-content of 0.2 per cent of the air which is breathed during one half to one hour is very perilous and a CO-concentration of 0.4 per cent and more has mortal effect within 5 to 10 minutes. The increasing accidents by oozing and therefore non smelling consumer gases and the numerous suicides forced the manufacturers of illuminating gas to develop and to sponsor research work in detoxicating of illuminating gases.

II. The fundamental Austrian patent # 113,333 (1927 - 1941)

Wolf F. Mueller, Ph. D., till 1927 director of the I.G. Farben factory at Leverkusen, obtained the Austrian patent # 113.333 for a process to detoxicate illuminating and consumer gases employing the following well known equilibrium reaction.

$$CO + H_2O_{steam} \rightleftharpoons CO_2 + H_2O + 10.100 \text{ Calories/kilomole}$$

The process was performed by means of catalysts which were used by I.G. Farben for manufacturing hydrogen for the ammonia - synthesis. The patent claimed that the before mentioned catalysts are suitable to convert CO into H_2 if not water gas but illuminating gases are used which contain gaseous hydrocarbons (gaseous paraffins, olefines, acetylenes, benzenes) and small amounts of organic and inorganic compounds of sulphur and nitrogen. It was further said that the conversion of the CO was commendable only to a rest content of 1 to 2 per cent by the application of 500% excess steam and at approximately 930 - 1200°F (500 - 650°C).

In 1928 W.F. Müller became professor and director of the institute of technology of inorganic substances and director of the institute of technology of fuels, where I was employed as a first assistant. During the following years I carried out experiments to detoxicate illuminating gases in cooperation with W.F. Müller and H.F. Graf. These experiments were sponsored by the management of the gas works of Vienna.

III. Laboratory Experiments at the Technical University of Vienna.

From 1928 till 1931 we made experiments in a laboratory apparatus applying 3.5 to 7oz. (100 to 200 g) of catalysts which were placed into externally heated vertical glass tubes. About 50 different catalysts were used which were composed of one, two or three components bound by various cements. The components were: potassium carbonate, copper, beryllium, magnesium, calcium, zinc, aluminum, lanthanum, active charcoal, zirconium, cerium, thorium, lead, chromium, uranium, managanese, iron, cobalt and nickel. The hand prepared catalysts were investigated under equivalent conditions of temperature, gas velocity and excess of water steam. The efficiency of the CO - conversion was measured by analysing the city gas and the obtained unpoisonous gases. One of the best catalysts, consisting of iron, magnesium and potassium carbonate was tested in an apparatus which was running during nine months 10 hours a day. The decrease of its efficiency during the test period was permissible. Due to the favorable results we agreed with the management of the gas works upon the designing and construction of a small pilot plant at one of the two gas works of Vienna, at Simmering.

IV. Small Pilot Plant at Vienna - Simmering

The pilot plant consisted of a reactor which contained 1 ton = 35cb. ft. (1m^3) of catalyst, a preheater for gas and steam, an indirect water cooler, a scrubber for the removal of the excess of carbon dioxide and the necessary control apparatus and instruments. Tests which were run during three months showed a good applicability of the catalyst and a satisfactory conversion of the CO. Carefully performed gas analysis showed that the oxygen content of 0.3 to 0.5 per cent was entirely removed and that the organic sulphur compounds were converted to hydrogen sulfide which can easily be extracted from the gas. Nitrogen compounds as HCNand NO were principally converted into ammonia. Hydrocarbons as indene, styrene and cyclobutadiene, which are as responsible for gums deposits in gas pipes as NO, were decomposed and hydrogenated. The through put was 1000 to 2000 cu. ft. p.h. (30 to 60m^3 p.h.) equal to 30 to 60 cu. ft. gas p. cu. ft. of the catalyst. Some tests showed that the throughput could have been considerably increased. It was further found out that the primarily used temperature of1100°F (600°C) was too high and that temperatures of 870 to 930°F (450 to 500°C) and even 750°F (400°C) were more favorable. An important result was the fact, that the throughput of gas increases approximately rectilinearwith the volume or weight of the catalyst. The diameter of the catalyst beads were 0.24" (0.6 cm) in the laboratory tests and 2.0" (5.0cm) in the pilot plant respectively. The volume and weight of the catalyst of the pilot plant were 10,000 times as large as in the laboratory apparatus but the surface of the catalyst was multiplied only 600 times. The pilot plant was operated with a volume of gas which was 1500 to 3000 times as large as in the laboratory apparatus. According to calculations the maximum throughput of the pilot plant would have been approximately 15000 to 20,000 cu.ft. per hour

(400 to 600m^3 p.h.). This assumption was proved true by experiments which were performed in the following years.

V. The combustion properties of the detoxicated illuminating gases

With the introduction of the admixture of water gas to dry distillation gases and the increasing use of Bunsen burners for domestic and industrial gas stoves and heating plants, the gas manufacturers payed more and more attention to the combustion properties of the consumer gases. In 1921 German gas engineers laid down guiding principles and limits for the heating value, the specific gravity, the variations of gas pressure and the maximum contents of undesirable gas components as oxygen, hydrogen sulfide, ammonia, tar and naphtalene and worthless innerts as carbon dioxide and nitrogen.

If detoxicating of illuminating gases is performed by conversion of CO into $CO_2 + H_2$ by catalysts the originated CO_2 must be extracted in order to obtain the same gas volume and the same high heating value as before. For each per cent by volume of converted CO the low heating value decreases for approximately 0.13% and the specific gravity increases for approximately 1.8% as shown by the following table 1:

	CO_2	C_nH_m	O_2	CO	H_2	CH_4	N_2
Consumer gas	2.8	1.6	0.6	12.7	46.6	20.5	15.2
Detoxicated gas	3.1	1.7	0.1	1.5	57.6	20.3	15.5

	Specific gravity (air = 1.000)	High heating Value B.T.U. p.cu.ft.	Low heating Value B.T.U. p.cu.ft.
Consumer gas	0.481	452	404
Detoxicated gas	0.385	452	398

The fundamental properties of gases which influence their combustion are 1) the heat value 2) the specific gravity and 3) the velocity of combustion. By replacing CO by H_2 the velocity of combustion of the detoxicated illuminating gas seems to increase vigoreusly because of the higher velocity of combustion of the H_2. The maximum linear velocity of combustion of the H_2 in mixture with air is approximately 520 ft. p. min. (265 cm p. sek.) and 6.6 times as large as the maximum velocity of combustion of the CO. Due to the lower specific gravity and the higher maximum velocity of combustion of the detoxicated illuminating gas, gas engineers and experts apprehented backfiring of the flames in Bunsen-burners and that changing or removing of the employed gas-jets would be necessary.

In numerous comparing experiments the injected air volume of Bunsen burners, the velocity of combustion at the actual ratio of gas and air in the burner tube, the length of the inside and outside flame cones, the minimum pressure of the gas by reducing the gas pressure until extinguishing of the flames and the content of CO in the combustion gases were measured. Furthermore tests in domestic and industrial cooking - boiling- and heating plants were carried out. A great deal of the results have been published 1934, 1936, 1937 and 1939 in "Das Gas - und Wasserfach," Berlin, by E. Graf and W.F. Müller and E. Graf. It was tested that at constant gas pressure in the jets of Bunsen burners the velocity of the detoxicated gas varies approximately with the reciprocal ratio of the root of the specific gravity according to the theoretical formula:

$$V_2 : V_1 = \sqrt{W_1} : \sqrt{W_2}$$

where

V_1, W_1 = velocity and weight respectively of the poisonous gas

V_2, W_2 = velocity and weight respectively of the detoxicated gas

Since the volume of the detoxicated gas increases at constant gas pressure, the capacity of the gas pipes and of the gas equipments are increasing in the same manner. Due to the lower specific gravity the detoxicated gas sucks less combustion air into the Bunsen burner and therefore the actual velocity of combustion becomes not very different from the actual velocity of the original poisonous gas.

The effect of the decreased air suction surmounts the increase of the combustion velocity so that the actual combustion velocity of the actual mixtures of gas and air in Bunsen burners decreases. According to this important result was the fact, that the inside flame cones become longer and not shorter as assumed. Figure 1 represents the alteration of the combustion properties of an illuminating gas depending on a gradual detoxication measured by a gas burner of a cooking plate (see the following table 2):

Table 2: Analysis of a gradually detoxicated illuminating gas

	CO	H_2	$CH_n + C_nH_m$	CO_2	$O_2 + N_2$
Original gas	17.9	51.7	19.9	3.0	7.5
Step 1	9.0	60.6	19.9	3.0	7.5
Step 2	3.0	66.5	19.9	3.0	7.6
Step 3	0.2	68.9	19.8	3.0	8.1

VI. Studies of secondary carbon-reactions

Besides the principle detoxicating reaction

1) $CO + H_2 O \rightleftarrows CO_2 + H_2 + 10.110$ Kcal P. Kmol

The following equilibrium reactions which are important at temperatures below 1100°F. (600°C) are possible:

2) $C + 2O_2$ 2 CO + 38.400 Kcal p. Kmol

3) $CH_4 + 2H_2O$ $CO_2 + 4H_2$ + 39.080 " " "

4) $CH_4 + CO_2$ $2CO + 2H_2$ + 59.300 " " "

Since we often found that the adjustment of the water gas equilibrium reaction 1) amounted up to 110% and even more, secondary carbon reactions were evident. Carefully performed gas balances at different temperatures of the catalyst and chemical analysis of the used catalysts showed that no precipitation of carbon on the surface of the catalyst took place. This result does not exclude the reaction 2) as an intermediate process with a high speed of the gasification of intermediately formed carbon. The formation of methane is possible according to the equilibrium reactions and the applied temperatures. The following table 4 shows the degree of conversion of CO (X) without and with CH_4 - composion (Y) at 1 atmosphere absolute gas pressure (= 14.2 lbs. p. sq. in.) and 100% adjustment of the equilibriums.

Table 3: Employed illuminating gas.

	CO_2	C_nH_m	O_2	CO	H_2	CH_4	N_2	H_2Osteam	Sum
Per cent by volume	2.5	2.6	0.3	12.7	43.2	22.2	16.5	31.7	131.7
Mols per m^3 gas	1.12	1.16	0.13	5.67	19.28	9.91	7.36	14.17	58.8

Table 4: Degree of CO conversion without (X) and with CH_4 - composion (Y)

°F (°C)	Conversion of X mols CO without CH_4 composion	Composion of mols CH_4 combined with CO conversion	Conversion of Y mols CO with CH_4 composion
662 (350)	5.09	5.22	5.66
752 (400)	4.64	4.39	5.60
842 (450)	4.24	3.30	5.40
932 (500)	3.77	1.84	4.78
1022 (550)	3.32	0.07	3.34

The Table 4 shows that no formation of methane is possible above 1022°F (550°C) and that the composion of CH_4 between 662 and 1022°F (350 and 550°C) favors the conversion of CO.

Catalysts with 3 and 4 components, containing Fe_2O_3 and Fe_3O_4 resp., M_gO and M_gCO_3 resp., K_2CO_3 and charcoal, were tested and the composion of methane measured. Each catalyst shows its own characteristic of adjusting equilibrium for CH_4 composion with increasing gas throughputs and optimal temperature ranges. The following table 3) contents the characteristics of a $Fe_2O_3 : M_gO : K_2CO_3 = 8 : 1 : 1$ - catalyst and shows that the CH_4 - equilibrium was completely adjusted at 1022°F (550°C) and that at lower temperatures, when the CH_4 - formation (b) is more favored, the CH_4 formation was slow, so that in practice a small volume of methane was formed. But there was no interest to develop catalysts which were able to form more CH_4 under normal pressure, because we intended to detoxicate applying higher pressure where the CH_4 - formation is more favored.

Table 5: Simultaneous adjustment of water-gas-equilibrium (a) and CH_4- equilibrium (b) at different temperatures and various throughputs (x) of cb. ft. of illuminating gas per cu. ft. of catalyst.

°F (°C)	X = 100	X = 300	X = 500	X = 700	X = 1000	X = 1400	X = 2000
662 (350)	a = 26% ——	a = 13% ——	a = 14% ——	—— ——	—— ——	—— ——	—— ——
752 (400)	a = 73% b = 3%	a = 37% b = 5%	a = 30% b = 2%	a = 25% b = 5%	—— ——	—— ——	—— ——
842 (450)	a = 99% ——	a = 87% b = 4%	a = 84% b = 8%	a = 70% b = 7%	—— ——	—— ——	—— ——
932 (500)	—— ——	a = 93% b = 16%	a = 87% b = 15%	a = 86% b = 10%	a = 79% b = 5%	—— ——	—— ——
1022(550)	a = 130% b = 100%	a = 115% b = 100%	a = 110% b = 100%	a = 110% b = 100%	a = 110% b = 100%	a = 110% b = 100%	a = 100% ——

The curve slope of the theoretical CH_4 - formation according to the results of table 4 and the practical formation of CH_4 corresponding to the results given in table 5 permits to calculate an optimum formation of CH_4 at 963°F (517°C) for iron catalysts.

Finally performed tests to determine the influence of the size of the catalyst beads showed the following decrease of the adjustment of the CO + H_2O - equilibrium with a throughput of 600 cu. ft. illuminating gas per cu. ft. of catalyst.

	over inches	(mm)	through	inches(mm)	adjustment equilibrium
Size 1	0.03	(0.75)	0.04	(1.0)	100 %
Size 2	0.08	(2.0)	0.12	(3.0)	93 %
Size 3	0.24	(6.0)	0.28	(7.0)	82 %

Some catalysts which were derived from a 3-components-iron-catalyst by weakening with charcoal gave practical the same throughputs and adjustments of equilibrium as the original catalysts. These experiments and the above shown results with different sizes of catalyst beads show that the centre of these catalyst beads takes part in reaction too.

VII. Laboratory - Experiments under elevated pressure

The water-gas-equilibrium-reaction is independing on the pressure. Whereas the composition of the CH_4-equilibriums is influenced by the applied pressure. The application of elevated pressures on the water-gas-detoxication-reaction is commendable if a higher space velocity is required.

Theoretical considerations of Langmuir, Hertz-Knudsen and others show that the adsorption speed of gases by catalysts increases at comparatively low gas pressures intensively rectilinear with the pressure. At high pressures the adsorption speed becomes practically independent on the pressure. Therefore an optimum and economical height of the gas pressure exists for each catalyst and for the used temperature. This economical pressure at the working temperature must be found out by trial.

The employed laboratory apparatus consisted of a gas compressor, a gas-storage-arrangement, a special Bosch-injection pump for distilled water, a small high pressure boiler, a pressure-and temperature proofed reactor containing about 61 cb. in. = 2.2 lbs (1L = 1kg) of catalyst, an indirect gas cooler with receiver for the condensed water, a high pressure H_2S - dry-extracter, a CO_2-scrubber, the necessary pressure reduction apparatus and instruments for controlling and measuring the volumes and temperatures.

The former used catalysts and catalysts of the American Magnesium Metal Corp. and the I.G. Farben were tried at pressures of 0, 71, 142, 214 and 284 lbs. p. sq. in. (0, 5, 10, 15 and 20 atm) and at 662, 752, 842 and 932°F (350, 400, 450 and 500°C) respectively. The adjustment of the water gas equilibrium and the CH_4-formation were measured with increasing gas throughputs.

The experiments showed that gas pressures above about 100 to 120 lbs. p. sq. in. (7 to 8.5 atm) were generally speaking not economical. The CH_4-composion varied within relative small volumes. For example the catalyst #13, containing Fe_2O_3, $MgCO_3$, H_2CO_3 and charcoal in the ratio 10 : 2.1 : 1 : 7 formed the following volumes of CH_4:

Table 6: = CH_4 - formation of the catalyst #13 in per cent by volume

	AT 752°F (400°C)				AT 842°(500°C)			AT 932°F (600°C)
In the laboratory glass tube	0.1				0.4			1.0
In the iron tube	0.2	1.5	1.5	2.8	1.0	2.0	3.6	——
At lbs. p.sq.in.	0	71	142	214	0	71	142	——

The following table 7 represents some essential results which were obtained with different catalysts at 0 and 71 lbs. p. sq. in. (0 and 5 atm.) with a constant steam excess of 400% for all experiments and a 90% adjustment of the water gas equilibrium. The composion of the used catalyst is shown in the following table 8.

Table 7: Space velocities of various catalysts at 90% adjustment of the water gas equilibrium at a = 0 and b = 71 lbs. p. sq. in.

Mark of the Catalyst	662°F(350°C)		752°F (400°C)		842°F (450°C)	
	a	b	a	b	a	b
1	—	—	250	450	—	—
4	—	—	150	320	950	1750
10	—	—	125	450	850	1650
11	—	—	250	250	—	—
12	—	—	100	300	650	1300
13	—	—	70	150	350	500
MMC	—	—	170	650	450	1200
14	—	—	200	380	850	1600
15	100	100	900	1600	1500	2800
16	—	—	550	850	850	1750
17	—	—	400	700	1100	1300
18	—	—	450	580	1050	1700
IG-Brown-Oxide	250	525	500	1200	1900	2100

Table 8: Composion of Catalysts

Mark of the Catalyst	Fe_2O_3 (burnt ochre)	Cr_2O_3	MgO	$MgCO_3$	K_2CO_3	Char-coal
1	10.0	——	1.0	——	1.0	——
4	10.0	——	——	2.1	1.0	——
10	10.0	——	1.0	——	1.0	1.0
11	10.0	——	1.0	——	1.0	2.0
12	10.0	——	——	2.1	1.0	4.0
13	10.0	——	——	2.1	1.0	7.0
MMC	——	——	2.1	——	2.1	10.0
14	10.0	0.4(nitrate)	——	2.1	1.0	——
15	10.0	1.2(nitrate)	——	2.1	1.0	——
16	10.0	1.2(sulfate)	——	2.1	1.0	——
17	10.0	1.2(alum)	——	2.1	1.0	——
18	10.0	1.2(hydroxyde from sulfate)	——	2.1	1.0	——
IG-Brown-Oxide	10.0 (nitrate)	1.0(nitrate)	Al_2O_3 0.1	SiO_2 0.1	Na_2O 0.1	

The alternation of the space velocity with elevated pressure and with a higher and lower adjustment of the water gas equilbrium is shown in the table 9 employing three catalysts, at a = 0, b = 71 and c = 142 lbs. p. sq. in. (a = 0, b =5, and c = 10 atm).

Table 9.

Mark of the Catalyst	Adjustment of the water-gas-equil.	752°F(400°C)			842°F(450°C)		
		a	b	c	a	b	c
15	100 %	500	700	700	700	800	800
	90 %	900	1600	1600	1500	2800	2500
	80 %	1200	2000	2000	1900	4300	4200
I.G-brown-oxide	100 %	300	750	1100	850	850	—
	90 %	500	1200	2100	900	2100	—
	80 %	750	1750	2800	2700	2800	—
MMC	100 %	100	200	350	200	600	900
	90 %	170	650	650	450	1200	1200
	80 %	—	—	—	—	—	—

With respect to the space velocities which were employed in the years 1928 - 1931 the obtained catalytical efficiencies were remarkable. The decreasing steam consumtion for the detoxicating process with lower temperatures is shown in the table 10, applied on the CO removal from 12 to 1% of an illuminating gas.

Table 10: Predicted steam supply

Temperature of catalyst	752°F (400°C)	842°F (450°C)	932°F (500°C)	1112°F (600°C)
Stoichiometric steam volume cu.ft.p.cu.ft.	0.11	0.11	0.11	0.11
Necessary steam excess cu.ft.p.cu.ft.	0.65	1.06	1.61	3.16
Total steam supply	0.76	1.17	1.72	3.27

VIII. Long run tests and experiments to regenerate the activity of catalysts.

2.

In order to investigate the alteration of the efficiency of a catalyst during a long run an apparatus was constructed and operated 24 hours a day at normal pressure with a simple continuous working water vaporization and admixture of the generated steam to the city gas. The arrangement was able to run over a long time without any supervision.

The used Viennese consumer gas was a mixture of coal gas, flue gas of the coke ovens, natural gas, water gas and normal producer gas. The content of undesirable components and gas impurities was mostly within the usual ranges. But due to the irregular coal supply and a not always uniform gas purification during the war years 1942 to 1944, the content of oxygen, sulfur compounds, naphtalene and even tar was beyond the ordinary limits during several days and even weeks.

The results which were obtained with 4 of the above mentioned catalysts are represented in Table 11.

In case #4 the converter contained 0.42 liters of a pre-catalyst which removed the oxygen content of the gas and two beds of the chief catalyst (0.62 + 0.62 liters) which were operated at different temperatures. This arrangement was similar to that which was applied at the detoxication plant of the Continental Gas Compant at Nordhausen, Germany.

The preheatment of the catalysts was performed by glowing through 4 to 6 hours in the illuminating gas atmosphere at 1112°F (600°C).

"Practical CO-removal" means the ratio of the removed CO in per cent by volume to the CO-content in per cent by volume of the origin gas. The ratio of the removed CO volume to the theoretically possible one (according to the water gas equilibrium) is about 10% higher than the preceding mentioned ratio.

The table shows that the activity of the catalysts was readily reduced in

Table 11: Results of long run tests.

Mark of the Catalyst	16	4	MMC	I.G. Brownoxide
Volume of the Catalyst cu. in. (liter)	50 (0.82)	49 (0.80)	50 (0.82)	25.6 + 38 + 38 (0.42 + 0.62 + 0.62)
$^{\circ}$F Temperature($^{\circ}$C)	797(425)	797(425)	797(425)	662, 807, 734 (350, 430, 390)
Employed steam Volume cu.ft.p.cu.ft.	0.90	2.0	4.0	0.97
Space velocity	600	225	110	365(500 resp.)
CO-content of illuminating gas	13.0-14.0	11.0-14.0	14.0-12.6	13.0-14.0
CO-content of detoxicated gas at test beginning	1.5	1.5	4.5	1.5
Practical CO-removal at the beginning	90%	90%	70%	90%
Practical CO-removal after 1 Month	83%	80%	55%	80%
Practical CO-removal after 2 Months	80%	62%	32%	60%
Practical CO-removal after 3 Months	*70%	68%	38%	*10%
Total test time hours	1370	2240	2190	1650
Total gas (cu.ft. through put(m^3))	28,200 (800)	13,000 (370)	6,700 (190)	36,000 (1020)

*Estimated from ~~the curve of~~ the slope. of the curve.

a relatively short time. This is partly due to the before mentioned varying content of oxygen and other gas impurities. The composition of catalyst 4 was similar to the primary catalyst #1 which was employed by the pilot plant and during the 9 month laboratory test.

During the long run test we observed, that even after short standstills, the catalytical activity enhanced.

The better efficiency of the catalyst #1 in the years 1918 to 1932 might be explained by this observation. But it must be clearly said that one and the same catalyst works mostly more satisfactory in a pilot or commercial plant than in a laboratory-glass apparatus. The results which are represented in table 11 offer the possibility to compare the activity of different catalysts for practical reasons, although the comparison is not quite exact because of the mentioned variations of the content of oxygen and other impurities in the used gases.

In order to investigate the alteration of their activity, the catalysts should be tried in long run tests on the influence of the gas impurities by exact chemical analyses, the alteration of the crystal structure by x-ray studies and the chemical alteration of the catalyst by analysing the compounds which were absorbed by the catalyst. Due to war time difficulties it was impossible to perform this intended investigation.

Rough qualitative comparisons showed that the catalyst #4 for example was fritted easily and contained naphthalene, whereas the catalyst #16 was non-fritted and free from naphthalene but contained more sulphur compounds and benzene hydrocarbons. The influence of the temperature, the time and the atmosphere of pretreating the catalyst is well-known. On the other hand we observed a favorable influence of standstills, ~~so~~ that we intended to prove the activity after standstills and by elevating the temperature to the applied pretreating temperature during the long run test.

The I.G. - Farben - brownoxide catalyst which was kept at normal laboratory temperature in a nitrogen atmosphere was refilled into the reactor and the test continued.

Surprisingly the activity was high at the beginning of the following second period. This appearance can be explained by the slowly performed cooling of the catalyst in the reducing illuminating gas atmosphere when the first period of testing was finished. During the following second period of the long run test, the activity deteriorated in 45 days from a 85% to a 20% CO-removal, as it is shown in figure 2.

The intended regeneration by reheating to the pretreating temperature of 1112°F (600°C) was now performed during 48 hours. The activity was elevated up 75% CO-removal and was slowly deteriorated in the following third period during 170 days to a 50% CO-removal.

The intended second reheating and the regeneration by treating with pure hydrogen, and purging with steam or nitrogen at different heating conditions could not be performed because the tests must be finished in spring 1944 due to war time restriction. The catalyst has been preserved.

IX. Experiments and investigations to detoxicate by conversion with simultaneous removal of the CO_2.

During three years I was ordered to investigate the detoxicating process of Boesher and Marischka. The patented process applied the mineral ankerite as a catalyst, containing lime, magnesium and iron in the form of carbonates. Ankerite is a worthless by-product of iron-ore cleansing in Styria, Austria. The ankerite is used as a combination of a catalytic substance plus an absorbent for the CO_2

formed by the conversion reaction. The conversion temperature was 930 to 1130°F (500 to 550°C) and the absorbent regeneration temperature 1560 to 1830°F (850 to 1000°C). The catalyst was circulated between conversion and regeneration furnace. Natural ankerite granules and beads ~~mixed and formed of~~ crushed ankerites were proved in order to study the alteration of the catalytic activity. The experiments were sponsored by Zahn & Co. Berlin and C. Marischka, director of the gas work in Vienna.

The degree of retrogression of the CO_2- absorption and the conversion was proved in long run tests, which were performed in the laboratory apparatus as similar as possible to the experiments in the pilot plant for approximately 350.000 cu. ft. gas per day at Vienna - Leopoldau. The combustion properties of the obtained gas were published by E. Graf in "Das Gas-und Wasserfach, 1939 H. 22. The ankerite catalyst was also used in the single conversion reaction without absorbing action at different temperatures and elevated pressures. Thereby this catalyst showed few activities.

X. Laboratory experiments to investigate and improve the "M.M.C." - Catalyst

During 1936 and 1937 W.I. Muller and I were asked by the Austro-American-Magnesia-Corp. at Radenthein, Austria, to investigate a catalyst which was composed of beech-charcoal, caustic magnesia and potassium carbonate for the conversion of water gas to hydrogen. The activity of MgO as a single catalyst and in mixture with charcoal or potassium carbonate respectively and the activity of various mixtures of the three components was studied in a laboratory apparatus. The important influence of the size of coal and magnesia and their intimate mixture were investigated by X-ray tests. Optimal temperatures and times to pretreat natural magnesia by glowing were found out. It was observed that absolutely pure magnesia was inferior to technical magnesia due to a content of small amounts of iron of the latter. The admixture of technical iron ochre to the tree-component-catalyst showed increasing space velocities at temperatures of 750 to 840°F (400 to 450°C) and at elevated pressures.

XI. Various studies on detoxicating illuminating gases.

A 175 cu. ft. p. h. (5m^3 p. h.) laboratory detoxicating plant was constructed with a proper heat exchanger in order to determine the heat transfer of the gases at forced convection in the iron tubes. The cold illuminating gas was mixed with water steam and the mixture preheated to 660°F (350°C) whereas the detoxicated gas was cooled from 790 to 800°F (420 to 450°C) to approximately 300°F (150°C). We obtained overall coefficients depending on the gas velocity and gas composition, which we applied to compute and design commercial plants.

Another laboratory plant was designed to investigate the heat transfer gas-iron tube-gas at gas pressures up to 570 lbs. p. sq. in. (40 atm) but it was not constructed due to war time difficulties.

The simultaneous removal of the excess carbon dioxide and the formed sulfur hydrogen by means of water at 43, 71, 100, 142 and 214 lbs. p. sq. in. (3, 5, 7, 10, and 15 atm) was proved in connection with the detoxicating process.

When the experiments under elevated pressure (see chapter VII) were performed, the H_2S was extracted in a properly designed iron ore box under the detoxicating pressure. The absorption was performed at the elevated pressure where as the regeneration by aeration of the iron oxide was carried out at normal pressure during standstill times. There was no alteration in the adsorption activity of the iron oxide to be observed whether we operated at elevated or at atmospheric pressure.

Further I was appointed an independent expert by the community of Vienna, the "Deutsche-Continental-gas-ges". Berlin, the J. Pintsch A.G. - Berlin, and the Bamag-Meguin-Berlin, in a patent interference with the "Gasent" - (Gasentgiftungs-Gesellschaft) - Berlin, which claimed to detoxicate illuminating gases to 1% CO at 750 to 930°F (400 to 500°C) using iron catalysts and maintaining the combustion properties without removal of the converted carbondioxide by means of admixing additional volumes of coal gas to the illuminating gas before the detoxication process.

The alteration of the combustion properties using the conversion without removal of the carbon dioxide was measured and compared with those of the conversion followed by removal of the carbon dioxide. The result of my investigations and those of R. Heinze, Prof., Berlin was the nullification of the Gesent-patent.

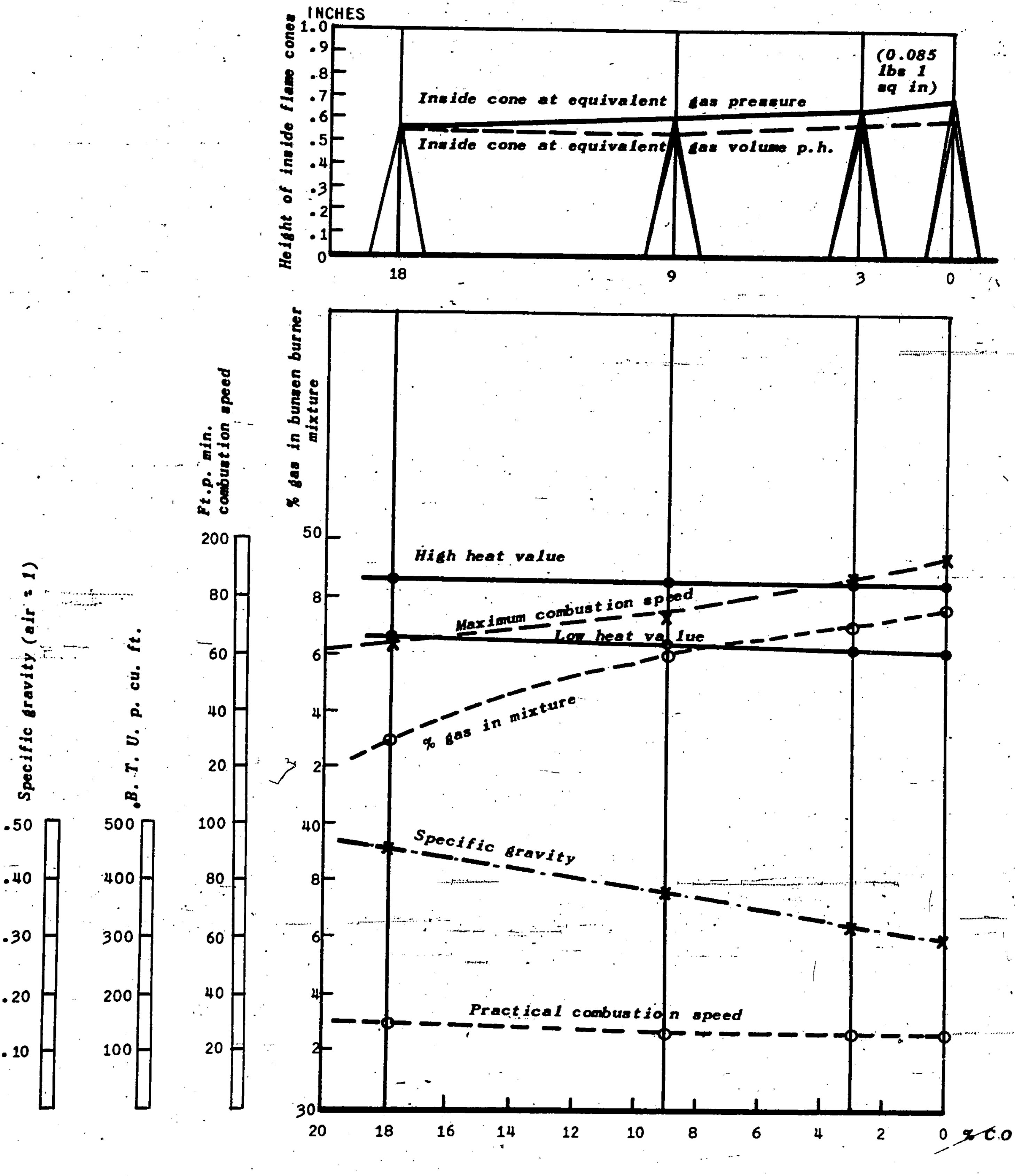
XII. Conculsion and Outlook.

The detoxicating process by conversion of carbon monoxide in illuminating gases to about 1% into hydrogen is practicable. The duration of the activity of the catalyst and therefore the fixed costs of the catalyst are still doubtfull.

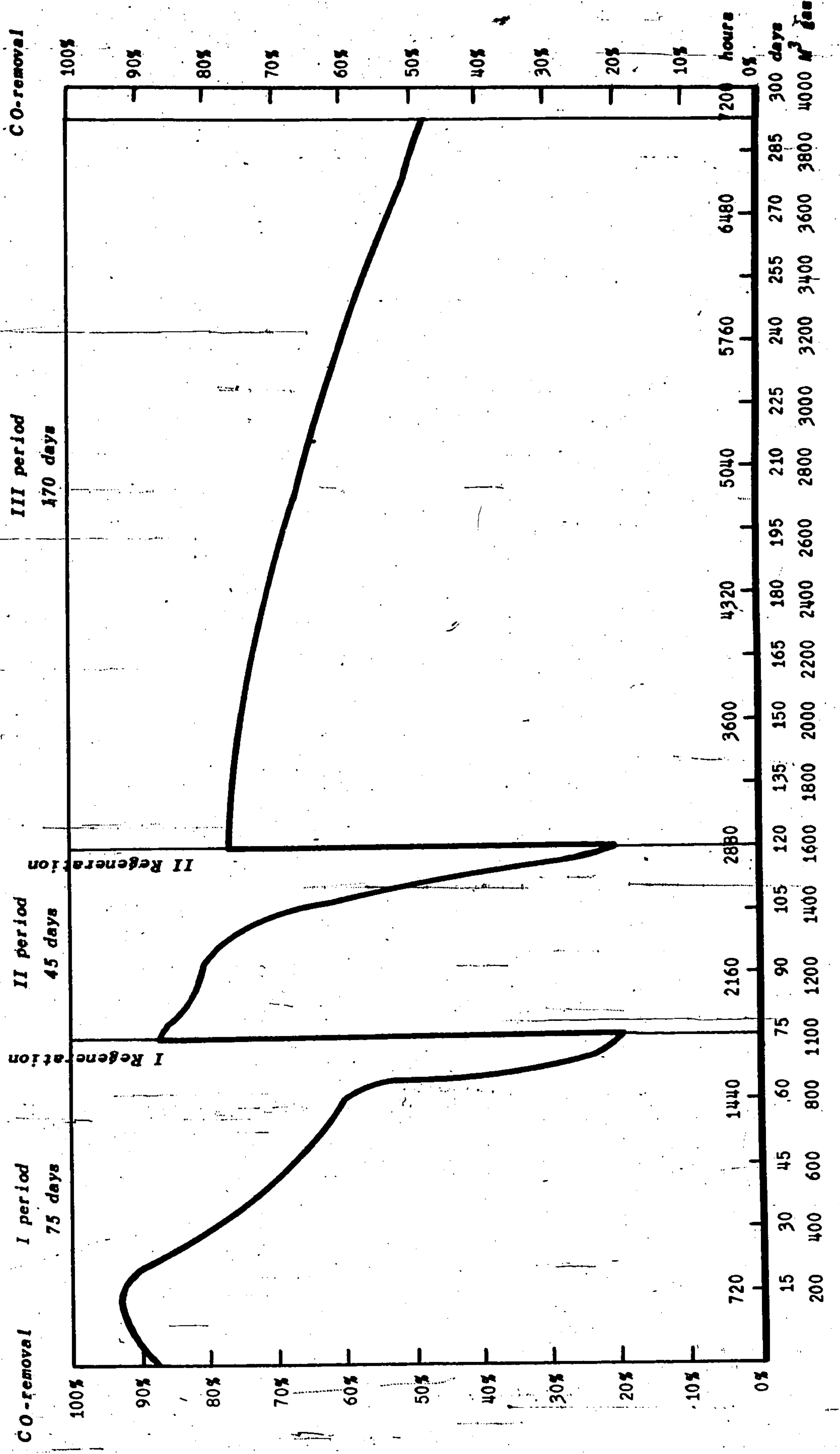
The costs of detoxicating including the removal of carbon dioxide and sulfur hydrogen have been calculated to approximately 0.5 to 0.7 Pfennig per cubic meter of gas. This costs seems to be relatively high. The detoxicating process without removal of carbondioxide by admixing additional volumes of coal gas causes alterations in coal throughput and coke production. The calculation naturally shows a cheaper process.

The detoxicating process becomes more interesting with regard to the refining of the gas which is performed simultaneously with the conversion of the carbon monoxide. The removal of oxygen, organic sulfur compounds, nitrogen compounds and of undesired hydrocarbons results less corrosion in pipes and gas using equipment, less formation of gum deposits and therefore an important saving in maintenance costs.

The favorable results of detoxication and recovery of sulfur under elevated pressure lead to considerations to perform cooling, extraction of tar, recovery of ammonia, prussic acid, naphthalene and benzene under elevated pressure. Calculations of the managers of the Viennese Gas Works, which are the most modern gas works of Europe, showed great advantages by performing the mentioned processes under approximately 70lbs. p. sq. in. (5 atm.). If this modern gas purifying plant under pressure would be financed, it is evident, the detoxicating and refining process will be applied too.



ALTERATION OF COMBUSTION PROPERTIES



ACTIVITY OF A CATALYST DURING A LONG RUN TEST

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~~CONFIDENTIAL~~

HUBMANN

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The Detoxification Plant

At Bohlen is an experimental and pilot plant to treat 500 - 1000 m 3/h of city gas for the elimination of part of its CO content.

The plant is connected to a LURGI-plant for gasification under pressure. The production of the gas and its conversion take place under 20 atmosphere pressure. The raw gas coming from the producer plant is of relatively good cleaning owing to the washing with water under pressure, which has taken out CO_2 and part of the organic sulphur.

Detoxification of gas hitherto has been developed and tried out based on the principle of converting $CO + H_2$ to CH_4. In this case the products of conversion are of no higher value than the price paid for the heating value of the gas. This former method therefore must raise the production cost per heat unit in the finished gas at any rate. In most cases this additional cost has proved so high that it cannot be accepted by the consumer, especially not if the gas is used for industrial purposes.

The LURGI method used at Bohlen is based on the principle of converting CO to a marked product with a much higher price than city gas, so that detoxification pays by itself and the price of city gas even might be lowered.

At Bohlen the gas with 4500 Cal/m^3 is sold now with 2.6 pfg/m^3 delivered into the pipe lines of the distributing company. CH_4 accordingly is sold at 5.5 pfg/m^3. At the Bohlen plant the conversion of CO into Benzine and high melting waxes is aimed at. In this case 10,000 calories are paid with 25 pfg/kg as benzine and with 50 pfg/kg as paraffin wax. Under such conditions the Bohlen plant can make even a profit from detoxification of the gas without raising the price for city gas.

Description of Process

The raw gas first passes Activated Carbon to take out the rest of light oils and all those organic compounds which might form resinoids poisonous for the catalyst. The gas then is heated to about 200°C by a surface heater. Then it enters a tower filled with alcalic iron oxide. Here the rest of organic sulphur is taken out. The gas passes to a chamber with the catalyst for conversion of CO. The catalyst is of the cobalt type, when a high yield of paraffin wax is wanted, and of the iron type, when more benzine is aimed at. Then the gas goes to a condenser with moderate cooling where the high melting waxes come out. The gas is then neutralised by washing with sodium hydroxid to protect the equipment against organic acid. In a second cooler the gas is cooled down with water. The rest of medium oils and part of the benzine are condensed there. Activated carbon takes out the rest of the benzine and if desired the so-called liquid gas (butanes, propane). A compressor is provided to bring the gas to the original pressure as required for long distance transport. Part of the gas may be recircled for better control of the reaction and improved quality of the products.

The plant has started operation in winter 1944. Exact experimenting in long periods, however, has been prevented by the many air raids on Bohlen and the industrial district of Saxonia. Some periods of several days of undisturbed working have given the results which have been expected.

One nm^3 of raw gas	CO_2	3.0%
	CO	19.5%
	H_2	50.5%
	CH_4	24.0%
	C_nH_m	0.9%
	N_2	2.0%
	O_2	0.1%

Yields

0.675 nm^3 of converted gas

CO_2	8.9%
CO	4.45%
H_2	44.90%
CH_4	37.25%
C_nH_m	1.5%
N_2	3.0%

and 44 gr of liquid products

of which

17.6 gr waxes

15.4 gr gas oil

11.1 gr benzine

The carbon monoxide remaining in the gas depends on the through-put of gas per ton of catalyst and per hour. With a through-put reduced to 50% or the mass of catalyst increased by 100%, the content of carbon monoxide is not more than 1%, the rest of the plant is unchanged in its dimensions.

The composition of the city gas after conversion can be regulated most easily by altering the composition of the raw gas, especially its content of CO_2, N_2 and CH_4. In most cases a lower grade and cheaper gas may be produced primarily, compared with primary production of a gas with 4500 cal/nm^3.

Sketch appended.

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WAHL

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Comparison of Results
of Oil Shale Carbonisation in Estonia

Altho it seems to me that in the United States under the prevailing circumstances there might be very little interest only in developing the deposits of oil shale I suppose to be entitled to lay down some characteristic data of the Estonian Shale Oil Industry. I guess they will be of any value to this country too in a not very far future, because they are the only available practical details on oil shale treatment altogether, based not on estimates but on experiences gained within two decades in the largest oil shale industry of the world.

Item	A	B	C	D	Uni
I General:					
Carbonisation Method	Kivioli Tunnel Kilns	Pintsch Vertical Rotating Generator	Groendal-Ramen Tunnel Kilns	Davidson Rotating Retort	
Company	Eesti Kivioli A.S. Estonian Shale Oil Corporation	Esimene Eesti Polevkivi Toostus First Estonian Oil Shale Industry	Estimaa Olikonsortium Oil Company of Estonia	The N.C. Gold Fields Ltd.	
Location of Works	Kivioli	Kohtla-Jarve	Sillamae	Kohtla	
Founded	1922	1918	1928	1931	
Patent Owner	Eesti Patendi A.S. Tallinn	Juli Pintsch Berlin	A.B.Industrimethoder, Stockholm	T.M.Davidson Edinborough	
Nationality	Estonian	German	Swedish	English	
Licenses sold to	German Australian South-African	Manchurian	Swedish	Scotish French	Ent pri
Total employed in 1939	2000	3200	600	350	Pe
II Mine					
Kind of mining	underground	underground open cut	open cut	underground open cut	
Organic substance of shale	34	35	37	40	%
Moisture of shale	12	11	8	11	%
Hauling in 1939	510.000	666.500	217.000	61.000	to
Shale per one miner	2.5	3.4	4.0	3.7	da
El. power consumption	4	2	3.6	3.4	to KW to

XXI Carbonisation

Item	A	B	C	D	Unit
Kind of method	indirect heating Scavenging gas	direct heating	indirect heating Scavenging gas	external heating	
Shale classified	yes	yes	yes	no	
Yield of Oil on moist material	20	17	19	21	%
Yield of Oil from Fischer Assay	96	81	92	89	%
Heating performed by means of	fines 58 oil 40 res. gas 2	Coke 97 res. gas 3	oil 95 res. gas 5	residue 98 res. gas 2	% %
Annual working time	315	350	300	345	days
Annual employed persons per one ton crude oil*	1.5	1.25	1.2	2.35	[illegible]
El. power consumption*	166	92	150	70	KWh/ Ton
Yield of gasoline	16	2.4	18	18	% of oil
Fuel Oil Analysis 1943 Spec. gravity	1.006	0.991	1.049	1.028	
Heating power	37,800	38,300	38,200	38,500	BTU/ Kgs
Viscosity at 50°C	8.1	4.82	19.4	20	°E
Pour point	-21	-25	-6	-13	°C
Flash point	88	61	100	87	°C
Water content	1.6	1.1	1.4	1.2	%
Ash content	0.03	0.17	0.02	0.70	%
Benzol insoluble	0.15	0.37	1.08	1.025	%
Phenole content	18.9	18.8	25	23	%
Distillation Test acc. Engler -200°C	0.0	6.0	0.0	0.0	%
-225°C	2.5	10.5	0.0	1.0	%
-250°C	4.5	15.5	5.0	2.5	%
-275°C	9.0	22.0	12.0	4.0	%
-300°C	16.5	29.0	22.5	7.5	%
-325°C	27.0	38.0	38.0	20.5	%
-350°C	42.0	53.0	55.0	45.0	%

Note:

* Shale Carbonisation and Condensing Plant only, but all periodical repairs and maintenance considered.

Item	A	B	C	D	Unit
IV Capacity & Output					
Crude Oil Capacity 1924	4,500	12,000	—	—	Tons/yr
" " " 1931	34,500	12,000	15,000	6,000	"
" " " 1936	80,000	28,000	15,000	12,000	"
" " " 1939	80,000	70,000	50,000	12,000	"
" " " 1943	130,000	110,000	35,000	12,000	"
" " " 1924	28,500	75,000	—	—	BBL/yr
" " " 1939	500,000	440,000	315,000	75,000	"
" " " 1943	820,000	750,000	220,000	75,000	"
Shale throughput of <u>one</u> carbonization unit	400	40	250	32	Tons/day
ditto on trial only	450	100	—	—	"
Output in 1939 of:					
Crude oil (tons)	70,000	61,000	38,500	11,500	Tons
Fuel " "	56,500	56,800	34,000	8,500	"
Diesel " "	540	1,300	1,100	800	"
Gasoline "	11,500	1,500	7,400	2,100	"
Crude Oil (BBL)	440,000	390,000	245,000	73,000	BBL
Fuel " "	356,000	360,000	215,000	54,000	"
Diesel " "	3,400	8,500	7,000	5,000	"
Gasoline "	73,000	9,500	47,000	13,500	"
Total output of crude oil since foundation	3,150,000	3,070,000	885,000	610,000	BBL
During	14½	23½	9	12	Years
Other products in 1939	—	—	—	—	—
as i.e. Bitumen	2,000	1,700	200	564	Tons
Impregnating Oil	200	117		60	"
Carbolineum	300	1,200		80	"
Roofing Tar	100	180		60	"
Acetone	70	—	—	—	"
V Specific Consumption	—	—	—	—	—
Electric power spec.:	—	—	—	—	—
per run of mine shale	4	2	3.6	3.4	KWhs/
per shale for one ton oil	22	12	44.5	16	"
per crude oil	225	112	195	90	"
per fuel oil	252	131	200	95	"
per gasoline	351	-	275	185	"
per crude oil	36	16	31	24.2	KWhs/BBL
per fuel oil	40	21	32	15	"
per gasoline	56	-	44	30	"

Item	A	B	C	D	Unit
Steam per Oil	1.9	0.5	1.7	1.8	Tons/ton
Water " "	0.0086	0.0061	0.0145	0.0079	Tons/h
Iron per annual output	75	60	75	80	Kgs/ton
Iron " lifetime "	5	4	5	8	"
Iron " annual "	26	21	26	27	lbs/BBL
Iron " lifetime "	1.75	1.4	1.75	2.7	"

<u>VI Commercial</u>

Cost price of 1 ton Crude	A	B	C	D	Unit
Oil: Run of Mine Shale	23	38	27.5	25	%
Power, Steam, Water	13	4.5	11	10	%
Wages, Salaries	15.5	13.5	18	28	%
Sundries	5	3	1.5	1	%
Interest on Capital	32	29	34	35	%
Taxes, Insurances	11.5	12	8	1	%
Total %	100.0	100.0	100.0	100.0	%
Total EKr.	90.77	84.69	83.45	90.05	EKr/Ton
minus Rebates	9.19	23.89	8.15	6.41	"
Total 1939 EKr.	81.58	60.80	75.30	83.64	"
Total 1939 $	21.80	16.40	20.00	22.20	$/ton
Total 1939 $	3.40	2.60	3.16	3.50	$/BBL
Cost price 1938	69.76	60.10	68.20	75.60	EKr/ton
" " "	2.95	2.55	2.90	3.22	$/BBL

<u>Balance sheets per December, 31st, 1939 showed:</u> -

Item	A	B	C	D	Unit
Capital Stock	380,000.-	800,000.-	133,000.-	560,000.-	$
Intangible Assets	4,000,000.-	3,350,000.-	1,860,000.-	850,000.-	$
Tangible Assets	930,000.-	560,000.-	270,000.-	200,000.-	$
Estate	17,000.-	1,240,000.-	133,000.-	570.000.-	$
Good Will (est.)	930,000.-	55,000.-	80,000.-	no	$

Parity of Stocks per August 1940, valuated by the Russians for indemnity of foreign shareholders after nationalization of this industry:

Item	A	B	C	D	Unit
	400	160	160	100	%
Net profit 1939	7,000.-	54,000.-	16,300.-	no	$

Some general data might be added:

Item	Value	Unit
Cost price for crude gasoline in 1938:	6.15	$/BBL
" " " run of mine shale " :	0.93	$/Ton
Total hauled shale since foundation up to August 1944:	17,737,000	tons
" produced crude oil " " " " 1944:	7,715,000	BBL
" " gasoline " " " " 1944:	1,080,000	BBL

[redacted]

H. A. Wann

Item	A	B	C	D	Unit
Steam per Oil	1.9	0.5	1.7	1.8	Tons/ton
Water " "	0.0086	0.0061	0.0145	0.0079	Tons/h
Iron per annual output	75	60	75	80	Kgs/ton
Iron " lifetime "	5	4	5	8	"
Iron " annual "	26	21	26	27	lbs/BBL
Iron " lifetime "	1.75	1.4	1.75	2.7	"

VI Commercial

Cost price of 1 ton Crude	A	B	C	D	Unit
Oil: Run of Mine Shale	23	38	27.5	25	%
Power, Steam, Water	13	4.5	11	10	%
Wages, Salaries	15.5	13.5	18	28	%
Sundries	5	3	1.5	1	%
Interest on Capital	32	29	34	35	%
Taxes, Insurances	11.5	12	8	1	%
Total %	100.0	100.0	100.0	100.0	%
Total Kkr.	90.77	84.69	83.45	90.05	KKr/Ton
minus Rebates	9.19	23.89	8.15	6.41	"
Total 1939 KKr.	81.58	60.80	75.30	83.64	"
Total 1939 $	21.80	16.40	20.00	22.20	$/ton
Total 1939 $	3.40	2.60	3.16	3.50	$/BBL
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Balance sheets per December, 31st, 1939 showed: —

Balance sheets	A	B	C	D	Unit
Capital Stock	380,000.-	800,000.-	133,000.-	560,000.-	$
Intangible Assets	4,000,000.-	3,350,000.-	1,860,000.-	850,000.-	$
Tangible Assets	930,000.-	560,000.-	270,000.-	200,000.-	$
Estate	17,000.-	1,240,000.-	133,000.-	570.000.-	$
Good Will (est.)	930,000.-	55,000.-	80,000.-	no	$

Parity of Stocks per August 1940, valuated by the Russians for indemnity of foreign shareholders after nationalization of this industry:

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[redacted]

H. A. Wan

(26)

H. A. Wahl (26)

The Meiler Pile Carbonization System

Summary and Practical Results
in the Industrial Stage 1944-46
and
Tests with the Cotrell-Method
as Main Condensing Device

Index **Pages**

1. Design and Development of 10 Pile Plants

The situation of German warfare in the early summer 1944 demanded the production of "oil at any price". From this judgment only the "Wueste-Plan" might be understandable and justified.

The plan provided the erection of 10 small carbonization plants of the "Meiler"-pile-carbonization method with a daily output of 60 tons of crude oil each within a building period of three months. The following points of view were decisive: the speed of erection, the saving of raw materials, the neglect of human labor and profitableness, the power and water supply, the aerial defence and camouflage, the demand for large areas for the "Meiler-field" and the ~~production of~~ one single crude oil only.

During the winter 1943/44 and in spring 1944 the first tests to elaborate a sufficient method of the pile were made at the small pilot plant in Metzingen, Wurttemberg.

Shale of German and Estonian origin was heaped on a concrete foundation of about 10 by 10 feet surface. A suction pipe in the foundation was connected to a condensing device and the fan. Later the solid foundation was abandoned and instead of one, two or three pipes were put on the bottom of the heap. The volumina of the heaps varied between 5 and 180 tons of shale. The height of 7 1/2 feet was not exceeded owing to the small condensing unit and weak fan. Very many different tests with the Meiler-pile were made and various perceptions gained. Among them were the advantage of not moving the burned material, seemingly the little demand of raw materials and the facility of pulling the suction pipes out of the pile, the trickling of the Wurttemberg shale, the way of ignition of the pile, the speed of carbonization steered by means of the intensity of the suction, aso. The apparent yield varied between 2 and 4.5% of oil. According to the shale samples the Fischer test showed 40-80% of yield. In general these tests were optimistic and it was decided to continue research on a much bigger scale at the Schomberg Pilot Plant which was completed by 60% in the meantime. The first pile of 678 tons shale was ignited in Schomberg on July 28, 1944. The height was 9 feet, 6 suction pipes were applied, the shale was not crushed. The result was disappointing; the yield of oil was bad, the handling of shale very difficult out of the lack of any machinery to heap up the shale and the pulling out of the pipes ended in a complete fiasco. The next piles were even worse. It seems to me that in the meantime the German oil situation had become very critical, because during these first days of August 1944 the resolution by the government was determined to apply the Meiler pile carbonization method to several large plants. There was some talk about 3 plants to be built in 4 months at first, afterwards it was concluded to build 10 plants within 3 months.

In vain we warned. It was a big blunder to transfer the sufficient results of a tiny research station like Metzingen to the industrial stage. There was no time to await the perceptions of the large scale tests at the pilot plant. Two big companies in Berlin who did not have the slightest idea about the difficulties were commissioned with the design and the erection. Technical particulars were not regarded at all, as for instance, the pulling of pipes, handling and crushing of shale, the soft and partly muddy surface of the areas provided for the meiler fields, the climate of the region situated between 1500 and 2400 feet a.s.l. with a severe winter, snow and long lasting periods of rain. In order to use the main advantage of the pile, i.e. not to remove the residue, it was concluded to apply the so-called "Wander-Meiler" (wandering piles) for the 10 plants. This postulated the handling of about 2000 tons of shale and earth within 24 hours on very limited areas, the movement of rails and sleepers on partly burning piles. Dry storage of fine shale, peat and wooden chips for ignition of the piles was another problem. In particular, it was entirely connived that the pile, according to perceptions of the small scale tests, was to be made up with great care which was connected with a lot of skilled handiwork. There were no experienced men and the workers had to be drilled by the same few scientists who ran the laboratory tests.

Therefore I give in the following a comparison of the description of the Meiler-Pile Carbonization System written by me on July 25, 1944, after conclusion of the Metzingen tests with the real handling of the Meiler-Pile as it was practically applied in the industrial scale up to two years later, i.e. in July 1946. I lay stress on the fact I have emphasized in my report of July 25, 1944, that the perceptions of the Metzingen tests are not adequate for the industrial stage and are not transferable to the latter.

A. The Oil Shale of Wurttemberg.

1944: The total depth of the shale averages about 27 feet. The oil contents vary between 4.5 and 5.9%. The specific gravity of the shale is 2.7, of the included limestone 2.8. The color of the shale is dark gray, the limestone gray to plain gray. Therefore the separation of limestone from shale after mining is very difficult. Out of this reason the separation of the three mighty limestone banks has to take place on the spot. The pile requests a shale as free of lime as possible to avoid explosions, especially during the ignition. Otherwise large parts of the surface were not ignited. The average moisture of the run of mine shale is 5%. The moisture of the medium to be carbonized has to be less than 8%, the moisture of the ignition shale not more than 4%. By rain spraying the moisture of the run of mine shale does not increase, on the other hand, the rain is very disadvantageous to the middle and fine shale, because of the cementing of the small particles. By this, large areas of fine shale become air-tight and the ignition is very bad.

1946: The oil contents of the shale we had to use varied between 4 and 9% according to samples taken from every 10th lorry. The sampling was insufficient owing to the circumstances that we did not have any real crushers and screening devices. The man who took the sample took it from the average shale on the lorry and did not consider that on the same lorry there were some large pieces of not crushed limestone, nor did he regard the amounts of dirt in the lorry which were inevitable owing to the crude operation of the big excavators. In 80% of the lorries, lime and dirt in varying amounts were present. Thus the real oil contents might be 3 - 8%. The picking out of limestone failed completely because the working speed of the excavators could not be reached by the handiwork of picking out the hardly distinguished limestone pieces. The average moisture of the run of mine shale did not pass over 6% in spite of heavy rains; the average moisture of a complete pile reached sometimes the enormous percentage of about 15%: the pile bound the heavy rains like a sponge. Contrary to the first assumptions this was not disadvantageous to the yield of oil, in opposite: the tests with artificial spraying <u>before</u> the ignition showed that the water vapors worked like scavenging gases, produced primary condensation centers for the oil gases and increased the total oil yield considerably. On the other hand, they overloaded the condensing devices, especially the Cotrell precipitation apparatus. But these disadvantages are of secondary issue. Artificial spraying <u>after</u> the ignition did not give any results: the water produced in the burning zone of the pile a remarkable increase of watergas effecting explosions in the Cotrell. The increase of the moisture percentage of the fine shale remained in the industrial process of very bad influence. The less the moisture the less the amount of necessary ignition material. Nevertheless we failed to undercut the moisture of 3.5%, sometimes even being higher owing to the climate and the outdoor handling of the shale, a main disadvantage of the meiler-pile.

B. The Granulation and Crushing of Shale.

<u>1944</u>: The run of mine shale must be classified according to the granulation:

rough shale: 30 - 200 mm size (1¼-7 inches)
middle " : 10 - 30 mm " (½ - 1¼ inches)
fine " : up to 10 mm " (½ inch)

The necessary amounts for a 1000 ton pile are:

rough shale: 94.5% over 35 mm
middle shale: 5.0% 10 - 35 mm
fine shale: 0.5% up to 10 mm

To make one meiler tight to its neighbor an additional amount of fine shale is required. This results in the contact-surface of both the meilers by 1/5 inch thickness. It is impossible to classify the shale by special system of blowing. The most pieces break in sizes of about 6 x 50 x 60 cm, about 10% in sizes of even 25 x 60 x 90 cm. The amount of fine shale depends on the number of manipulations of the shale, as for instance, loading, unloading, crushing, etc., but does not reach by itself the necessary quantities. Therefore crushing is necessary.

1946: Owing to the circumstance that for one plant only a crusher lately could be obtained, we had to work without granulated shale, only the necessary amount of fine shale was produced by primitive crushers. For the "standard meiler of 1700 tons" one took 1700 tons of not granulated run of mine shale (97%) and 60 tons (3%) of fine shale. The air tight packing against the other meilers was made of dirt and clay after abandoning the much better fine shale owing to the inability of producing and additional 100 tons of fines per pile. The packing of dirt had to be made 3 feet thick. It gave an extra load of 800 tons of earth moving per one pile. Nevertheless, this dirt dried and became pervious to air, such minimizing the oil yield. The meiler consisted of completely unclassified shale, containing big rocks of limestone and much dirt. A very little classification took place by the heaping. The larger pieces fell out of the lorries more to the bottom. By comparison with the few meilers granulated by hand on the pilot plant we came to the conclusion that the influence of granulation is at the meiler-method relatively little in comparison to its other disadvantages and to the more elaborated carbonization systems. Huge shale rocks were not carbonized and burned later on out. These losses are little at this crude method and were not surmountable owing to the lack of technical apparatus.

C. The Measures of the Meiler-Pile.

1944: Up to now the maximal height has been 2.4 m (7½ feet). By increasing height a reciprocal action takes place between the suction of the draft fan and the buoyancy of the hot gases in the center of the pile. The higher the pile the greater the "chimney-effect" of the rising gases. This must be regulated by the depression of the suction, otherwise a cone-shaped zone of non-carbonized shale arises. Other limits for the height of the pile are resistance of shale and the output of the fan. It is likely that the "chimney-effect" decreases at long piles, for there are less gases arising lateral. That would mean that the suction and the "overheaping" (see Diagram No. 1) might be decreased. To safeguard a uniform burning of the pile the "overheaping" must be observed strictly. The "overheaping" is the distance from the outer end of the grates to the beginning slope of the piles surface. The "overheaping" is up to the height of the pile of 2m 4/5h, it increases at rising heights and can be at 2.4 m height 1/1 h already. The angle between the slopes and the soil has to be 36°. This corresponds to the natural pouring angle of dry shale. It is very advisable to round off all angles of the pile. Regarding the basis we have thus a radius of 4/3 h ≠ 4/5 h. For the horizontal surface of the pile we take the radius by 4/5 h. By doing so we get out of a truncated pyramid a truncated cone only in the longitude expanded for the length of the grates. After shaping the pile according to the above measures the edges of the truncated cone also are to be rounded off. This grants a uniform ignition and advance of the burning zone or carbonization zone respectively. If this is not regarded, the suction of the fan will be less at the outer edges of the cone than the buoyancy of the arising gases. The sharp edges burn upwards with flames and the shale beneath them is lost for the carbonization. If several suction pipes are put side by side we get as the ideal shape of the pile the shape of a flat loaf of bread. Then the volume of the pile is calculated and multiplied by the pouring gravity of the shale. The latter is 1.25 at piles heaped by hand and 1.22 at piles heaped by excavator. Thus we get the total weight of a pile.

The grate consists of pipes. Pipe surface has to be as great as possible. The velocity of gas must be 4 m/sec. The pipes are provided with as many holes as possible. Stability of the grates only has to be observed. To avoid the so-called "false air" it is very essential to control the O_2 contents of the gases continuously. "False air" means the excessive presence of oxygen in the carbonization gases. This is produced by the incomplete ignition or by the "wall-effect", i.e. the suction of air into the grate along the pipes or by intake of air from the neighbor pile. Therefore the pipes must be covered with fine shale and the surfaces of other piles have to be tightened.

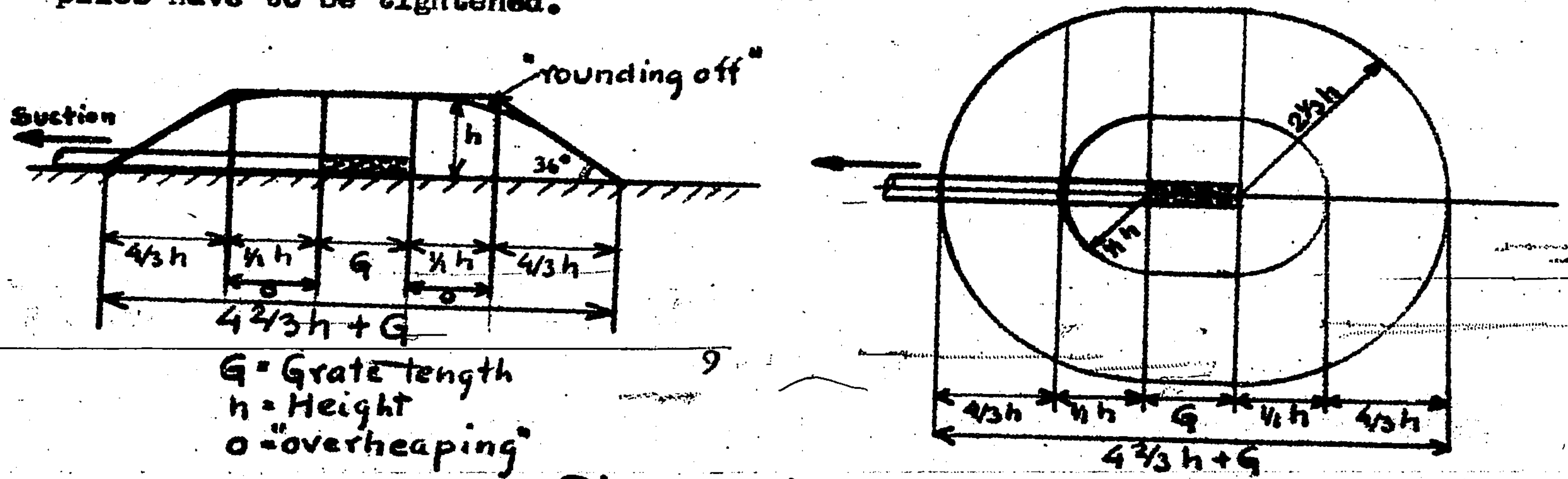
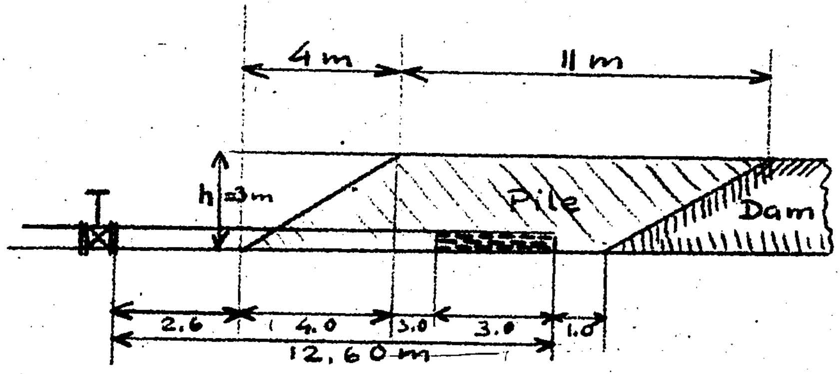


Diagram No. 1.

1946: The principal measurements were confirmed by tests made at the Schomberg Pilot Plant with meilers of 3.5 and 5 m height (11 & 16 feet). But on the other hand, it was impossible to adopt the ideal round shaped meilers in the method of wandering piles which leaned against each other side by side. The finesses of the upper rounding off have to be abandoned too. By doing so the shape of the standard 1700 tons pile originated. (See Diagram No. 2) The height is 3 m (10 feet), the length 40 m (130 feet) and the depth 11 m (35 feet). After having fixed these data once it was impossible to change the measures, for the meiler-fields, pipe lines, railroad tracks, aso., were calculated for the above measures. The piles were not surveyed singly any more to save the time-wasting handiwork. The single rows of piles were marked and the measures observed only. The total weights of the piles were not checked and as a result the pouring weight of the shale differed widely, as we made sure in the Pilot Plant by weighing every tenth lorry. Owing to size of pieces, the moisture of shale, the filling by the excavator, and other facts, the load of one lorry varied on a wide scale. When handling almost 2000 tons of shale and about 1000 tons of dirt daily it was impossible to observe the accurate prescriptions with an insufficient technical apparatus and differing weather conditions. Instead of using fine shale to prevent the intake of "false air" dirt was used on the suction pipes. This had to be moved with hand barrows on wooden planks over the rows of suction pipes while at the same time these pipes were fixed to the huge receiving pipe of 5½ feet diameter in pouring rain or in a snow gale.

Similar insufficiencies, which were not observed while planning, were discovered later. The packing against the neighboring piles had to be made with dirt, too, and did not satisfy at all. Further soil was needed for covering the old once-burned piles in order to prevent the burning of sleepers, boots and electric cables.



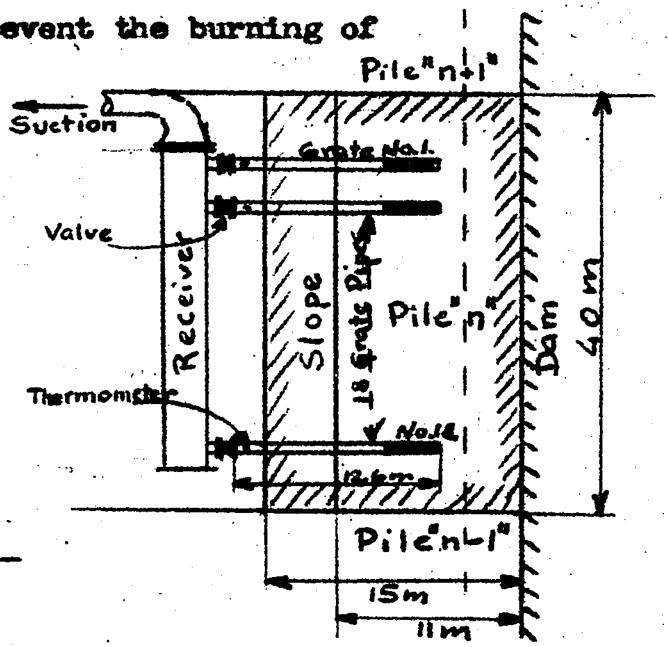


Diagram No. 2 (not in scale)

D. The Ignition.

1944: The ready pile is covered with a layer of fine shale at least 2 inches thick, i.e. about 70 kg/m^2 surface (17 lbs/1 sq foot). Then the whole pile is covered with hacked sods of peat equally (about 1/3 of the sods size, not smaller, because the peat would be gone with the wind). The necessary amount is 2 1/2 lbs/1 sq foot at dryweather; at rain the demand rises to 4 lbs/1 sq foot. Then the upper third of the pile is covered with wooden chips (7/8 lb/1 sq foot), in order to give to the upper part of the meiler a preceding against the lower parts. The ignition is very dependable on the weather. The ignition has to take place in the area protected from wind. Use slow-matches. The flames transplant equally over the whole surface towards the wind, like a peat moor burns towards the wind. The ignition lasts under normal conditions 2 hours until the beginning of the carbonization. While igniting the exhaustors are driven with highest under pressure. Suction before the ignition in order to dry the shale of the pile is not necessary. Did the ignition fail one recognizes that by the high O_2 contents of the gases and by the dark and often upwards smoking spots on the piles surface. If this is remarked early there the "post-ignition" in order to save the meiler is possible yet. This is done by heavy roughening of the not ignited spots by means of a long rake, covering with peat and chips again and ignition at bad cases by aid of spread oil. Then the fan is started at full speed again. The total peat in the cases of "post-ignition" amounts up to 5 and 6 lbs/1 sq foot.

1946: In the course of two years we had to learn that the ignition is the most essential and the most difficult proceeding of the whole meiler method. Because of the dependence of the ignition on the weather the method as such is entirely dependable on the climate. By this the sentence upon the meiler is passed for Wurttemberg at least.

By traveling over the meiler with heavy trucks, lorries and engines, the shale is pressed firmly. It has to be roughened. Big plates of shale are to be gathered from the surface because they would hinder the ignition of larger areas. Then followed the covering with 60 tons of fine shale, 9 tons of hacked peat and roughly 1 ton of wooden chips. At rain and snow the quantity of peat rose to 7 lbs/sq foot and of chips to 1 1/2 lbs/sq foot. Nevertheless the ignition and hence the yield were insufficient. All that was connected with a lot of handiwork. On the long run, that was unbearable for an industrial stage of this system. We set out to save manpower and the variety of ignition means. At last we achieved the reduction of manpower from 60 man-hours to 20 man-hours at the standard 1700-ton pile, to abandon the peat at all, to reduce the fine shale to 30 tons in the average and to relinquish the partial ignition. The fine shale had to be very dry (3.5%), instead of peat was used lignite dust (obtainable and no preparation, about 4 tons) and the chips averaged about 0.8 tons. The exhaustors were started at the maximum underpressure of 1200 mm W.C. and operated with 55,000 m^3/h for at least 6 hours. Nevertheless the ignition remained dependable on the weather and the skill of a trained crew. Big rocks of limestone hidden some feet under the surface were very disturbing because of their explosions and blowing up large areas of the surface. "Post-ignition" was seldom adopted because of its great demand for men compared with the little improvement in the yield. Very disturbing regarding the ignition and the yield was the transfer of the burning of the neighbor-pile on the pile just under treatment. To avoid that, one row of meilers (8-10 piles) had to be carbonized in close succession.

E. The Carbonization.

1944: The velocity of carbonization is the advance of the carbonization zone in perpendicular direction towards the center of the meiler. In general, one can say the more uniform the granulation the quicker the carbonization. During the ignition the velocity (in this case the burning velocity) is rather high and amounts to 15-20 cm/h. After sufficient ignition the velocity is decreased to 5-7 cm/h. This throttling shall produce a settlement of the carbonization zone. Then the velocity is increased up to 7-10 cm/h. Desirable is the increase of the velocity until the end of the process. The velocity is decreased again shortly before the end of the carbonization when the gas temperature rises to 200-250°C. The temperatures were measured at the pipes near the meiler. Before igniting the temperature corresponds to the temperature of the outer air or to the material to be carbonized, respectively. After ignition the temperature rises to about 70°C owing to the condensation of the moisture at the lower shale and decreases again at beginning evaporation of the moisture at 60°C. Then the temperature settles at 65°C, corresponding to the dew-point, and remains equal during the whole process. The increase to 250-300°C starts shortly before the carbonization is finished. The burning zone has almost reached the grate pipes and the suction is finished. It is no use to operate any further, because no oil vapors will be obtained anymore and the stability of the grate pipes is only endangered. The final rise of the temperature is accompanied by an increase of the O_2-contents in the gases. The contents of oxygen correspond before the beginning of the carbonization to the oxygen of the air, i.e. 20.9%. After ignition the O_2-contents must decrease on 0.7 to 1.4% within the first 4 hours. At good ignitions the latter averages in this range during the whole process and rises towards the end only with increasing temperature on 2-4%. At high O_2-contents the oil in the pile is roasted. The temporal course of exhausted quantity of gas corresponds to the velocity of carbonization. The depression rises at the beginning of the carbonization owing to the condensation of the moisture on the lower, still cold shale on about 350 mm, decreases then proportionally quickly on 70 mm and rises shortly before finishing the process owing to the remarkable resistances of the trickling taking full effect now only. The evaporation of the water is in general very high especially at the beginning. The water contents of very moist gases must be condensed by means of a water cooler. It was noted that the water cooler effects the condensation of the water vapours only and by decrease of the dew-point the moisture of the gases is diminished, but no oil is condensed. Very essential for the yield is the distance between the carbonization and the burning zone. The latter is a function of the moisture and of the uniformity of granulation of the shale of the piles. The more uniform the granulation and the drier the shale, the smaller is the distance and the greater the yield. Tests proved that the distance is at any rate less than 10 cm.

1946: The experiences of the operation showed that the fundamental conditions only could be observed in the practical stage of meiler-carbonizations. The O_2-contents of the gases could be kept between 4 and 10% at the more elaborated piles in the pilot plant, at the 1700 tons standard meilers of the various plants the oxygen contents varied between 7 and 12% owing to the incapability of making the piles tight against their neighbors, owing to the burning from the neighboring piles and to the insufficient ignition, and last but not least to some leakages of the almost mile-long pipe lines. In order to reduce the oxygen, the meilers were carbonised very fast, especially in the beginning of the process. The carbonisation velocity was for the first 6-10 hours 17-20 cm/h. The perception, that fast carbonised piles gave a better yield, affected to carbonize as quickly as possible. The first 1700 tons meilers were treated in 60-80 hours, the last ones in 40 hours on the average, such giving a better yield. This can be explained by the following assumptions. The difference in the granulation results in a very unequal carbonization zone. The burning zone is often nearing the carbonization area and the not yet carbonized oil is roasted. By fast carbonization the distance between both the zones is widened. Further we recognized that within the pile where a strong condensation by shock of oil vapours took place, this oil dropped on the bottom and was lost. We have measured these losses in the pilot plant and got - depending on the height of the pile - 8 to 12% of the total oil yield. On the standard plants it was impossible to catch this oil, because of the incapability to make a foundation on the bottom and to drain the oil into catchers. By faster carbonization, however, the oil vapours did not have any time to condense within the pile and were forced into the pipe line where they were caught. Especially after "invention" of the wet meilers the advantage of the scavenging gases was effective at fast carbonization. The average data of a standard 1700-ton meiler may be given as follows: total duration: 40 hrs.

Hours		1	6	11	16	21	26	31	35	39	
Velocity		18	18	15	12	9	8	8	8	8	cm/h
Quantity	of	55000	55000	43000	40000	32000	32000	32000	32000	32000	Nm^3/h
Temperature	gas	75	65	65	65	65	65	65	70	180	°C
O_2-contents		20	17	12	10	8	7	7	8	8	%

The real yield of oil according to the Fischer test could not be achieved owing to the above mentioned difficulties. It is not impossible that the yield of the average meiler reached 40-50% Fischer, for the consistency of the shale was very bad.

F. The Trickling and the Removal of Pipes

1944: Trickling arises in the midst of the pile above the grate owing to an accumulation of heat. It was not avoidable up to now. The trickling is a function of the velocity of carbonisation. The faster we carry out the low temperature distillation the greater is the accumulation of heat and the more extensive is the area of trickling. Troubles by trickling of shale onto the suction pipes were not observed up to now. It is advisable to start the removal of the suction pipes as soon as the carbonisation is finished in order to avoid the settling of the not yet dead material on the pipes. The burning zone will not be permitted to reach the grates as long as the carbonization has to be finished at 300°C temperature of the gases. It is essential to pull the pipes strictly in the horizontal. It seems to be advisable to pull the pipes by means of a caterpillar because one can expect very heavy friction.

1946: By cutting ready carbonised piles at the pilot plant we could observe that the trickling took place in 3/4 of the pile and at high or fast carbonized piles the trickling was so vigorously molten down that removal of the trickled material by aid of mechanical devices was impossible. Then rocks had to be blown up by dynamite. The grates were not affected. Nevertheless, the removal of the grate pipes was one of the heaviest tasks, and perhaps the only task, solved in an adequate manner in these two years. The first experiments to pull the pipes with a winch failed, so did the tests with a tractor, a caterpillar, and an engine. The only mechanism which succeeded was the 60 ton heavy excavator. Of course, it was impossible to operate heavy excavators on the muddy "meiler-fields" where different pipes hindered the motion. The measured friction amounted to 20-25 tons at the 10 m pipes on the Pilot Plant. The 12.6 m pipes of the standard outfit were heavier to pull. Therefore we constructed two similar devices, both operating on the corkscrew-principle. The one consisted of two pipes, the other of a lattice-mast. Both were placed against the front wall of the pile and by means of a motor and a set of pulleys removed the suction pipes. Both systems were movable by aid of motors. They were at last so developed that 4 men removed the 18 pipes of one pile within 8 hours. Nevertheless the handling of the pipes, the removal of the huge receiving pipe, the pipe fitting and the wear and tear of the pipes was a very unpleasant job. It was a factory in a permanent fitting state. The fitting had to be done outdoors all the year round. The demand and wearout of fitters was enormous.

G. The Lowering and Cooling of the Pile

1944: After finishing of the carbonization a diminution of the volume of the pile occurs. The lowering of the surface amounts on the average to 20% of the height. The lowering is a function of the way of heaping the pile, of the success of the carbonization and of the grade of trickling of the shale. Piles loosely heaped by excavators shrink more than hand-or-lorry-heaped piles. The better the low temperature distillation and the faster the pile was carbonized, the greater is the lowering. The cooling time, i.e., the time after which working on the meiler surface was possible again, is 2 days at a 7-foot high meiler. Probably at a 10-foot meiler it will amount to 3 days.

1946: The standard pile showed on the average a lowering of 50 cm (1 3/4 feet) or 17% owing to the bad carbonization of the contents. There were often large hollows and craters owing to strong trickling and explosions of the included limestone rocks. This lowering of the surface was very disagreeable, for the ready treated meilers did not perform a plane surface. In order to heap the "second floor" of meilers it is necessary to level the "first floor". Further, every following row of piles having the standard height of 10 feet had to be adjusted to that height and the preceding row had to be raised aloft by means of additional dirt to get the heaping level for the lorries. This effected a lot of labor not considered while making the theoretical design of the plants. Single standing meilers cooled fast. The 1700-ton meilers situated in a row, made of bad shale including different not carbonizable material, insufficiently carbonized, contained a huge amount of heat. This heat was sucked downwards during the process, thus enabling to work on the pile on the 3rd day. Then the heat started to rise again after finishing the suction and reached the surface during the 4th and 5th day, in which working on the surface was impossible. The cooling lasted then from 3-4 days depending on the success of the carbonization. It means in the average that the pile was free for working on the 3rd and from the 8th day on, calculated from the beginning of the respective carbonization. This fact was essential and not considered during planning too. It fixed the division of time and the various working orders.

H. The Velocity and Duration of Carbonization.

1944: One ton of Wurttembergian shale produces 500 m^3 of gas. A diagram is made showing the dependence of the carbonized tons of shale at different oxygen-contents from the amount of cubic metres of gas, i.e. $\frac{w(tons)}{t} = f\left(\frac{V(m^3)}{t}\right)$. Further another diagram is made of the volume of the pile as a dependence of the height of the pile, i.e. $V(m^3) = f(h(cm))$.

Instead of volume take the weight, i.e. W(tons) = f (h(cm)). By aid of the measured O_2-contents and the measured output of the exhaustor one takes from the first diagram the amount of carbonized shale. This amount corresponds on the second diagram to a certain decrease of the height. Regarding one hour this gives us the velocity of the carbonization. The latter will not exceed 10 cm/h (4 inches/h), otherwise the suction has to be throttled. The duration of a carbonization may be obtained thus by keeping a certain velocity of carbonization at an assumed or expected content of oxygen.

1946: These diagrams were made for all meilers. Nevertheless, it was impossible to fix the duration of the carbonisation of a pile owing to the circumstances that neither the O_2-contents nor the total weights of the standard meilers were exactly determinable. There were so many inadequate factors that in the average the duration of the process was determined by $\leqq$ 20% only. This resulted in a big waste of time regarding the workers in the industrial stage and is in my opinion one of the main obstacles to this method to an ample application in industry: there is no uniformity as basically presumed for an industrial process.

J. Piles with perpendicular suction pipes

In the Pilot Plant 6 greater tests were made with standing suction pipes. These piles gave a comparatively good yield of oil, for the condensation of vapours within the pile was less. On the other hand, it was more difficult to fix the pipes, to heap the shale by means of lorries and to pull the pipes. Owing to these facts and the rather limited volumes this method seemed not to be applicable on a larger scale either.

K. Fixed Meiler Piles

After having worked for two years with the wandering meilers of various sizes on the Pilot Plant and of the standard 1700-ton type on the "Wueste" plants, it was decided by the French government of the company to start with fixed meilers. The main advantage of the standard type — not to remove the residue — was abandoned. We started in June 1946 with the first piles of this type containing 1100 tons of shale. The main motives were: to get rid of the whole pipe and railroad struggle, to save iron and labor, to catch the oil lost by condensation and oozing into the bottom and a step forward to a better controlled and - may be - scavenging system. Really it means the capitulation of the "meiler" and a return to some system like the Schweitzer. A final judgment after the first test is not advisable. A foundation for 8 piles was built, the grate pipes put into the latter, the preparation with shale by lorries fixed on one side and the removal of the residue carried out by excavating on the other side of the row of piles. The first piles showed heavy trickling and an extremely dirty operation of the excavators at the removal of residue.

2. Technical Devices of a Standard Pile-Plant.

A. Mine

Open cut mines only were applied in this industry. The overburden, varying between 2 and 5 feet, is removed by excavators and used as tightening on the meiler field. The shale is worked by electric drilling, blasting and loading to lorries by means of excavators. The shale used in the plant where I worked was so soft that the excavator itself dug and loaded it.

Three excavators were employed in a mine for 2000 tons of shale per day.

B. Crusher and screening plant.

These were provided but nowhere performed owing to the happenings of the war.

This plant should consist of a delivery-bunker, a conveyor for picking out the limestone, a crusher for shale of maximum size of 600 x 900 mm and an output of 220 to 250 tons of shale up to 130 mm per hour, a conveyor to the screening plant. The latter consists of five vibration screens and the additional requirements. The screening is performed in 3 sizes of shale: fines up to 10 mm, ignition shale 10-35 mm and rough shale over 35-130 mm. These kinds of shale are taken into bunkers and loaded to the lorries by means of conveyors. The output of the latter is: fine shale 75 m^3/h, ignition and rough shale 130 m^3/h each.

We had to use on the Pilot plant as well as on the standard plant temporary crushers with inadequate screening devices. The amount of crushed material sufficed the Pilot Plant at the worst, but was much too little for the standard process. Therefore, we were forced to use uncrushed shale and crushed and screened the ignition shale only.

C. "Meiler-Fields"

The above distinctly described pile was used as the standard carbonization unit of 1700 tons shale. Given data were: the measures of the meiler, the duration of carbonisation (assumed to be 72 hours whilst planning), the duration of the cooling of the pile (assumed to be 72 hrs. too) and the available area. By these conditions one choose meiler fields of several rows of meilers, each row 8 meilers. After completion of the first floor the meilers had to be made in the second, third, aso. floor, thus filling large valleys. The first row of piles is started to heap from a dam. 2 piles are heaped at once, progressing the railroad has to be pushed on. In order to heap the last row of a - let us say 6 x 8 meiler field - the slope for the railroad must have about 60 m width. All these disadvantages are valid as long as it is impossible to heap the piles by means of loading-bridges. Each pile has 18 suction pipes of 12.6 m (40 feet) length and 350 mm (14 inches) dia. These are connected to the receiving pipe of 40 m (130 feet) length and 1600 mm (63 inches) dia. The receiver had to be pushed forward in the whole for one meiler width, i.e. for 11 m, after having carbonized the respective pile. Therefore these receivers had small wheels and were pushed on rails. By this standard outfit the alteration of the meilers measures, fixed once, was impossible. The receiver is connected to the main pipe line by means of the so-called connection pipes. These pipes as well as the main line have a standard length of 11 m (35 feet) corresponding to the width of the meiler and a diameter of 1200 mm (47 inches). After the progress of each meiler the respective connection line had to be shortened for one pipe length. One meiler-field for 6 x 8 meilers consisted of:

144 suction pipes (+ 18 spare) 40 feet by 14 inches dia.
8 receiving pipes (+ 1 spare) 130 feet by 63 inches dia.
40 connection " 35 " " 47 " "
26 main " 35 " " 47 " "

additional the T-pieces, valves, steel ring packings, bolts, and the length of the main pipe line to the condensing plant which varied between 200 and 700 m (640 and 2250 feet) corresponding to the local situation. This outfit was ordered 10 times suitable for 10 plants. The total iron weight of this outfit for the plant No. 8 amounted for instance on 530 tons. The suction pipes have compensators for thermical expansion and nozzles for adapting the thermometers. The receiver had 18 sockets for connecting the suction-pipes, each provided with a throttle-valve to regulate the suction of the respective grate. Each connection line can be closed by means of a valve. The main line has a heating pipe inside and a cooling line with sprinkler jets on the top outside. The first was used in winter to avoid the stiffening of the condensate supposed to have a high pour-point, the latter was used in summer as a condensing stage for the gases with great success. The meiler field is plane with a slight slope towards the main. On the deepest point of the main a water-valve and oil catcher were provided.

D. Condensing Plant.

The last mentioned item belonged already to the condensing plant. The long main pipe served as the first condensing stage and gave, depending upon the weather and the temperature of the cooling water used in the sprinklers, various amounts of various condensate which was pumped to one general collecting tank for crude product. The main line worked mainly as a gas drying device, but worked as an oil vapour condenser too, giving remarkable quantities of oil owing to its length at plant No. 8. Then the Cotrell-Tar-Precipitation followed. This consisted of 5 filter-boxes, oil catchers and the respective rectifier and high-tension set. The Cotrell was designed as the main condensing stage. The results achieved I shall mention later on. The obtained condensate was pumped to the general collecting tank. The gas is sucked to the exhaustor-plant now. The latter contains three exhaustors of 20,000 m^3/h each, pressure 1400 mm column of water, gas temperature of 65°C and 3000 r.p.m. Each fan is driven by a high-tension motor of 200 kw and 1500 r.p.m. A gear is switched between motor and exhaustor. For condensed oil there are collecting pipes and sealing pots of which the condensate is led to the collecting tank. From the exhaustor plant the gas is led to the burning-oven, where oil catchers are provided for the remainder of the condensing oil. This is led to the collecting tank too. The burning-oven was designed to burn the residue-gas with a low heating-power in order to annihilate the hydrogen-sulfides and the phenolic-water. This oven did not come into being at all fortunately. The oven was to be ignited by means of oil burners, the temperature of the residue gases has to be kept uniform and the oven was very sensitive to gas fluctuations as well as on interruptions of the operation as we made sure on the Pilot Plant. The very unstable operation of a standard meiler plant, as it resulted in the practice compulsory later, would have damaged the sensitive wall-up of the furnace and prevented the operation of the oven, if not the events of war had hindered the completion of these ovens in all plants. By means of a by-pass-pipe-line the gases were led into the chimney.

The general collecting tank was a pit made of concrete with a pump-sump. The condensate from main pipeline, Cotrell, exhaustor-station and chimney were collected here. Heating of the contents was provided.

The crude product was pumped from here to the separation plant and the water already separated to the phenolic-water container. The separation plant consists of two pre-heaters from which the oil is led to two wood-wool filters and then to a mixing container. The latter performs the mixing of filtered oil with "Dismulgan" in order to break the oil-water emulsion. Dismulgan is added continuously in a special solution by means of a pump. The result was very good. In summer 1945 the delivery of dismulgan by the I. G. Farben was stopped. In order to separate the oil from water and dust we installed a huge continuous centrifuge by which the heavy emulsion by means of adjustable scrapers was eliminated. Then the oil passed another small centrifuge and was pumped to the main separating tanks, where the final separation took place. The small centrifuges were overloaded as far as the crude oil was not pre-separated in the scraper centrifuge. From the separation tank the clean oil was pumped to the storage tanks and loaded in the tank-trucks. The separated oil-water emulsion was again led to centrifuges to repeat the process.

All phenolic-waters were gathered in a basin and - in abundance of the burning oven - pumped on the piles already burned for annihilating. Heating of the different containers was performed by steam. The whole condensing and separation plant including the exhaustor station was built in outdoor construction. This resulted in many troubles during the first winter. In order to smooth the operation we built houses on the exhaustor and separation plant apparatus. Further the following belonged to the standard outfit of a "Wueste"-Plant: The boiler-house with 2 boilers of 200 m^2 heating surface and a permuthit-water-softening device, the transformer-and switching-station for the electric power supply, the laboratory and different storage sheds for peat, coal and wooden chips.

The standard number of workers and employees amounted to 270 persons.

Provided was the carbonization of 30 meilers a month with an assumed output of 1800 tons of shale oil per month.

Further proper data of the technical apparatus of a standard plant as designed to come into being may be obtained from the papers pg. 2581-2608 included in the oil shale files here.

In the following a short sketch of the technical outfit of the "Pilot-Plant" at Schomberg which was destroyed by the events of war in May 1945 may be given. The Pilot Plant was erected in order to make various tests on methods of shale carbonization in general and to give the necessary information for the 10 standard shale carbonization plants in special.

The plant had two open cut mines, a temporary crushing and screening plant (the right one was never completed), two "meiler-fields" such giving allowance to different types and sizes of piles, a condensing plant consisting of air-cooler, water-cooler of 920 m^2 cooling surface, a Cotrell-tar-precipitation plant of 2 separate units with 3 + 1 filters of 32,000 m^3h gas capacity, an exhaustor-station consisting of two exhaustors of 16,000 m^3/h each and one for 20,000 m^3/h, different other condensing means, as for instance, baffle separator, nozzle separator, which were switched on the gas circuit or prepared for testing. Further, a light-spirit recovery plant based on the active-coal principle was installed. The gas-burning-oven worked for a few days only, as above mentioned. The separation plant consisted of wood-wool-separator, centrifuge, dismulgen-mixer, the necessary containers for separating the crude product and respective pumps. Storage tanks, boiler house, two laboratories, two workshops, transformer station, offices, aso. Besides this there was a special plant foreseen for trial carbonizations, where at first the Hubmann furnace only was practically tested. There was a condensing plant, a Cotrell device, laboratory, aso. The carbonization capacity was limited by these different apparatus, up to 400 tons of shale daily by the "meiler"-method, not taking into consideration the other methods.

The "Pilot Plant" was never completed and owing to a wrong design was started much too late to build. Thus the results gained from it for the 10 standard plants were poor and late. Owing to the great building capacity the number of workers and employees amounted to 700.

3. Practical Results and Profitableness of the Pile Carbonization Method

The erection of the 10 standard plants was started on October 1 1944. Within the first days of January 1945 it was expected to obtain the first oil from each of the plants. The daily output of a single plant was planned for 60 tons, i.e. per month's totaling on 15,000 tons of oil to be obtained from 10 factories. The full production together with the Pilot Plant and the enlargement of 3 existing oil works of other companies for March 1945 was scheduled to amount to 18,000 tons of oil. These estimates were never reached owing to basic miscalculations - as I tried to explain in the former chapter — owing to the lack of understanding of human and practical obstacles on the part of the extremely theoretical planning authorities, owing to the delay in building caused by extremely bad weather during October, November and December 1944 and by the lack of trust in such a late and gigantic remedy "to win the war", finally owing to war influences which paralyzed the transportation of raw materials, food and housing equipment for nearly 40,000 workers employed at once in this program.

The first standard plants were "ready" during February. At that time the first local air raids began to make some trouble. Compared with the fixed quantity of 18,000 tons the real output of oil of the whole industry was up to now in approximate data the following:

November 1944	630 tons	October 1945	210 tons
December 1944	650 "	November	650 "
January 1945	500 "	December	680 "
February	480 "	January 1946	500 "
March	1165 "	February	700 "
April	860 "	March	1100 "
May	—	April	1000 "
June	—	May	950 "
July	—	June	850 "
August	60 "	July estimated	950 "
September	170 "		

It means the real output was at the best a fifteenth of the rated capacity. Of the 10 standard plants 5 never were completed (Nos. 3,5,6,7,10), one and the pilot plant were destroyed by the happenings of the war, two are in full working condition but closed and two are producing oil all the time. The utmost one can expect of these four remaining meiler plants is a monthly output of 1800 tons of oil in the total. After completion of the plant at Frommern and enlargement of the underground plant of Schorzingen - both of them working by another method - I estimate the maximal possible oil output of this area may amount to 3500 tons per month under the presuppositions that the two closed works start again, that man labor is available, that tools, lubricants, etc., are provided sufficiently and an adequate nutrition is granted to the laborers. All these conditions were not fulfilled either under the German or under the French government. It is, of course, quite another question to what amount the oil production may rise in this district by means of other methods and new plants, if at any time in the future a stable German or Russian government should put great stress on these oil deposits.

Up to now the best output of a standard meiler plant has been 350 tons from 12 meilers within one month. This is a bare fifth of the estimate. Fifteen piles with roughly 450 tons per month are the best from my expectations.

Nevertheless, the French seem to be earnestly interested in that industry. After an interval of four months after the occupation of that area by the French First Army, the French showed an increasing interest in the production as well as in large scale testing. There may be different reasons: to ease the French imports of fuel for agricultural purposes into their zones of occupation, a means of balancing a little their occupation expenses and to make large scale tests for the benefit of their own oil shale industry by aid of the existing plant plus captured manpower and money. The profitableness in this case again does not count: the money was taken over from two German "companies" and is spent for research purposes. It is of secondary issue if the price of one ton oil by the meiler method is about RMk. 950.- and the fixed market price RMk 330.- per ton. One can expect that two works producing by another method and a third after completion may decrease this cost price, but that's of no importance. The monthly oil output of the pile-plants alone was in round figures:

November 1944	80	tons	(1)	October 1945	110	tons	(1)
December	100	"	(1)	November	390	"	(3)
January 1945	52	"	(1)	December	485	"	(3)
February	80	"	(1)	January 1946	—		
March	665	"	(5)	February	200	"	(1)
April	660	"	(5)	March	600	"	(2)
May	—			April	500	"	(2)
June	—			May	450	"	(2)
July	—			June	350	"	(2)
August	60	tons	(1)	July estimated	450	"	(2)
September	20	"	(1)				

The figure in brackets gives the number of working plants. This return shows a) a complete failure of the meiler-method in its industrial stage under the prevailing local circumstances. On the average 175 tons were produced monthly instead of 1500, i.e. 11% of the rated capacity. b) the dependency of the meiler on the climate is evident. During the severe winter of Wurttemberg the production equalled practically nil.

The results of single meilers in the pilot plant were:

Meiler	Shale	Oil	Yield	
No.	tons	tons	absolute %	acc. Fisher %
1	678	15.5	2.29	47.6
2	600	8.5	1.40	abt. 25
3	630	10.5	1.76	abt. 30
4	625	7.05	1.10	26.2
5/6	1560	19.3	1.24	28.0
7/8	1660	20.0	1.20	40.5
9/10	1390	24.3	1.75	54.0

Meiler	Shale	Oil	Yield	
11/12	1700	40.0	2.35	27.2
13/14	1670	27.0	1.65	44.0
15/16	2500	55.0	2.2	25.0
17/18	1600	26.9	1.7	25.0
19/20	1700	15.0	0.9	20.5
(S1) 21	935	10.0	1.1	23.0
(S2) 22	745	10.8	1.45	31.6
23	610	15.2	2.5	59.4
24	430	9.4	2.2	41.8
25	765	15.2	2.0	55.2
26	720	15.5	2.2	41.5
(S3) 27	392	10.5	2.7	51.5
29-33	2700	53.1	2.0	44.5
(S4) 34	300	7.4	2.5	54.0
35	450	10.0	2.2	abt. 46.0
36	600	11.5	1.9	" 40.0
(S5) 37	480	8.0	1.7	" 36.0
(S6) 38	390	6.7	1.72	" 40.0

The results of the meilers of Plant No. 8 were:

Meiler No.	Oil Gain ton	Oil Yield absolute %	Duration of Carbonization hours	Spec.power consumption KWh/to	
1	25.1	1.48	(	(	All
2	30.2	1.80	(	(	meilers
3	35.0	2.05	(abt.	(	had
4	33.0	1.95	(80	(850	a

No.	Oil	Yield	Duration	Power Consumption
5	34.0	2.0	(hrs.	(Kwhs standard
6	36.0	2.15	(in	(in weight
7	25.2	1.50	) the	) the of
8	26.1	1.55	) average	) average 1700 t
9	20.2	1.20	)	) up
10	22.0	1.30	)	) to
11	18.0	1.05	)	) No. 49
12	16.3	0.95	)	)
13	20.4	1.20	)	)
14	15.5	0.9		
15	25.0	1.5		
16	25.0	1.5		
17	27.7	1.65	(abt.	
18	20.0	1.2	(80	
19	25.1	1.5	(hrs.	
20	22.4	1.3	) in	
21	26.0	1.5	) the	
22	29.0	1.7	) avge.	
23	30.4	1.8	)	
24	28.5	1.7	)	
25	27.3	1.6	53	820
26	29.5	1.75	55	650
27	33.2	1.95	85	620
28	33.2	1.95	61	610

No.	Oil	Yield	Duration	Power Consumption
29	37.6	2.2	47	470
30	32.2	1.9	44	610
31	26.0	1.55	38	640
32	27.2	1.6	36	600
33	31.2	1.85	48	520
34	29.5	1.75	51	510
35	28.7	1.7	70	745
36	30.4	1.8	54	735
37	25.5	1.5	63	910
38	30.0	1.8	61	555
39 shale weight unknown	3.0	-	24	1630
40	31.0	1.8	53	540
41	29.6	1.75	(	
42	33.4	2.0	(	
43	31.2	1.85	(abt.	
44	32.0	1.9	) 40	
45	27.6	1.65	) hrs	
46	28.4	1.7	) in the	shale weight
47	31.2	1.85	) average	1700 tons
48	33.0	1.95	)	"
49	24.0	2.2	(abt.	
50	22.0	2.0	(30 hrs	shale weight
51	19.0	1.7	(in the	1100 tons
52	20.0	1.8	) average	"
53	21.5	1.95	)	"

The open-cut mine had a normal consumption of coal and fuel for excavators, electric power for drilling and lighting, aso.

The specific consumption of different items for the meiler-pile-plant consisting of the meiler-field, the condensation plant, workshop, laboratory, office, etc., were in the average per one ton of produced oil:

El. power	700 Kwh/to
Water	17 to/to
Steam	2 to/to
Coal	0.5 to/to
Fuel and Lubricants	0.003 to/to
Man-power	170 hrs/to
Peat	0.9 to/to
Wooden chips	0.08 to/to

These figures include consumption for general purposes, as for instance, transportation, office heating, repair and maintenance of buildings, etc. These are, therefore, no proper data for comparison. Nevertheless, one can state that the above data, especially electric power and laborers, lie highly over the normal averages of low temperature carbonisation demands experienced in Europe. Without taking into regard the other, partly major disadvantages of the meiler system, out of these consumptions the meiler method was not able to compete with regular methods of carbonisation. Even the demand of raw-materials which had to be supposedly lower than at a factory with an average mechanical outfit was much higher. For instance, the plant (without mine and rolling stock) consumed in its pipe-and-machinery-installation around 1700 tons of iron, i.e., at an expected annual output of oil of

18,000 tons and a duration of life of say five years, a relation of 0.019 tons of iron per one ton of produced oil. The really performable production of 450 tons monthly or 5,400 tons annually or 27,000 tons within five years gives a relation of 0.063 tons of iron per one ton of oil, i.e., 22 lbs of iron per one USA barrel of oil. Compared with the low temperature distillation plants built by us in Estonia up to 1940 and by the German administration until 1944 the demand on iron totalled in Estonia on 6 kgs per one ton of oil, i.e., 2.1 lbs of iron per one USA barrel of oil, or taking into consideration the oil contents and yields of both the methods, the Estonian method required 10 1/2 lbs/barrel. The relation is 2:1 in disfavor of the meiler-pile method. The apparent saving of iron ought to be one of the most advantageous items of the meiler system.

Under the circumstances of the Wuerttembergian climate, way of building, aso, then the profitableness of the meiler method is completely out of question: the cost price of 6.3 barrels varied between 800 and 950 RMK, i.e., $80.00 and $95.00 at the present exchange rate.

4. The Cotrell-Method in Connection with the Pile Carbonisation.

Besides the main gas pipe line with a water sprinkling device, the main and only condensing means for the gases was the Siemens-Lurgi-Cotrell-tar-precipitation plant.

The long pipeline worked more or less as a natural condensation, that is, in strict dependence from the respective weather, and did not enable the adjustment of condensation. Such gases and vapors of the meiler reaching the Cotrell-plant could not be influenced regarding their temperature, dust content and concentration.

The working with the tar-precipitator proved to be very unpleasant under these conditions, altho the electrostatic condensation of the Cotrell-

Lurgi system is well appreciated for dust and tar removal in the industry. Therefore, tests were started to fix and to eliminate, if possible, the obstacles of the condensation of meiler-gases by means of the Cotrell. Because I have performed these observations I shall describe them here in a more detailed way, altho I could not finish them owing to my sudden departure.

The Cotrell precipitator of a standard meiler-pile plant consisted of the following items:

5 filter boxes in one row. The gas inlet is on the side below and the outlet on the opposite side above. The conducts to the single boxes are not equal regarding their resistance to the suction. The boxes are 6 m high. The lead of the high tension voltage is performed on the top thru a cable sealing box and a porcelain wall insulator. An iron pipe leads the voltage to 144 steel wires which form the emit-electrode. These wires hang exactly in the midst between sheet-iron plates, which form the precipitating-electrode. The distance between opposite plates is exactly 240 mm. Each filter has on the bottom an oil tub which is connected over a sealing pot by the suction pipe to the crude oil tank. The voltage of 380 volts is transformed to 525 volts and can be regulated within the range of 400 to 650 volts by means of a tap-changing transformer. This voltage is conducted over several switches to a high tension transformer 525/55,000 volts. The high tension passes to a rotating disk rectifier. The high direct voltage is led over switches, ammeter for control of the ma-load of the single filters, automatic circuit breakers and cable sealing ends thru 5 single high tension cables to the wall entrances on the top of the filter boxes. To avoid live boxes the grounding is performed very properly. Two high tension and rectifier sets are installed. Each

filter can be switched likewise to one of high tension sets. Further are provided electric heating of the high tension switch room, ventilation and the electric heating of the wall entrance insulators on the top of the boxes. This heating can be regulated in 4 ranges and is independent for each filter insulator. The outfit of the plant was up-to-date and built as well as installed by the Lurgi Company in a very proper way. The high tension could be regulated in the range between 60,000 and 80,000 Volts D.C. and was checked on the under-voltage side. Control and supervision took place on the switchboard panel. Each of the rectifier sets has a maximal load of 290 mA at the rated voltage, which means all five filters can be fed in case of emergency by one rectifier allowing about 55 mA per filter. The rated capacity of each filter was 8000 Nm^3 of gas. At this capacity the filters precipitate nominally 95% of dust according to the Lurgi's guarantee. The overload of every 10% produces a decrease of precipitation of 4% of the rated capacity regarding dust and dry material; regarding oil gases of different composition there are no data experienced up to now.

Operation experiences.

The operation of the Cotrell plant in collaboration with the meiler-carbonization method exhibits a condenser with a variable dielectric. The variables are the following:

a. The oxygen content varies between 3 and 20% and is mainly dependable on the success of the ignition of the meiler pile. We have confirmed that the working of the precipitator at a higher oxygen content than 10-12% is technically not profitable. This is valid not regarding the danger of explosion, the limit of which we have fixed on 12% O_2. In contrary of this limit there occurred at different oil works explosions of filters at mere 11% of oxygen. This showed that not the oxygen alone but another gas, the quantity of which varies proportionally with the O_2-content, releases the explosions. We assume that watergas is due to these fulminations. We have observed that heavy rain after the ignition of piles of dry shale made the operation of the Cotrell very turbulent, favored fulminations and produced even heavy explosions by which some filters were destroyed. The same occurred at meilers moistened artificially after the ignition. The hydrogen content varied at these tests between 4 and 7%. The opposite was observed at meilers of very moist shale or at artificial moistening before the ignition. The water evaporizes, the steam works as scavenging gas and produces condensation-cores for the oil gases. No explosions were observed, the Cotrell works very quietly, the amperage rises but the oil yield by means of the Cotrell is rather small because of the overload of the Cotrell by precipitating water.

Various measurements showed curves according to which the "feasible load" is inversely proportional to the oxygen-content. Altho here were some declinations this rule can be considered as general. The so-called "feasible load" is the discharging current in mA between the emit-and-precipitation-electrodes which can be kept without leading to short circuits between the electrodes. The "feasible load" is proportional to the maximal voltage and to the dielectric of the condenser. At increase the spark-over-voltage is reached. The latter is automatically cut off. The "feasible load" is proportional to the efficiency of the filter, i.e. the precipitation of oil or other matter which determines the dielectric. The number of carriers is decisive. At pure oil vapors the "feasible load" would be strictly proportional to the gain of oil. This is never the case with the meiler. Therefore every admixture of moisture, dust, etc., produces an overload of the filter or participates in the silent discharge of the condenser, in both cases minimizing the yield of oil. At small oxygen-content (about 3-7% O_2) the "feasible load" can be kept from 50 to 70 mA per filter. The oxygen-content decreases after a good ignition of the pile after 8 to 12 hours below the limit of 12% O_2. Concerning our experiences thus the first quarter of the duration of the carbonisation does not enable the use of the Cotrell-precipitator. If the pile gets "false-air" from his neighbors, the whole carbonisation is spoiled, at least regarding the use or efficiency of the Cotrell-plant. The latter must be switched off towards the end of the carbonization because of increasing oxygen content and the formation of watergas. Based on our experiences one can state that the possible duration of use of the Cotrell amounts to 65% of the total duration of the pile carbonisation.

b. The amount of charge of the Cotrell is of outstanding influence regarding its efficiency. The above mentioned is valid as long as the filters are not overloaded. The rated capacity of 5 filters is 40,000 m^3/h at a temperature of 65°C and a density of 1.37 Kgs/m^3 of the gas. The meilers were run usually with 2, at necessary higher volocity of carbonization with 3 exhaustors. During the ignition they were run always with 3 exhaustors for at least 3 to 15 hours. The quantity of gases is then 44,000 resp. 54,000 m^3h. The standard Cotrell plant of 5 filters was hence overloaded by 10 resp. 36%. Therefore the efficiency of normal dust precipitators has to be decreased on 91 resp. 79% of the rated precipitation. This at normal oxygen-content. Regarding the efficiency of oil gases there were no data available and our own observations could not be completed. Nevertheless, it is sure that the precipitation owing to overload decreased on more than 90 and 75% of the rated output. Taking in consideration the demand to carbonize two piles at once "overlapped" it is evident that at least 6 filters should be applied and an evasion pipeline provided to meet the peak load. The price of an additional filter is one tenth only of the complete Cotrell plant.

c. The temperature of gas. The two variables were increased by the uncertainty of the temperature of the gases passing thru the Cotrell plant. The temperature varies between 45 and 80°C. As mentioned already, it was impossible by means of the existent apparatus to keep the gases at the Cotrell plant on a stable temperature. The latter depended on the outer air temperature. The lower the temperature, the more misty the gas, the greater handling-surfaces the single particles of the gas offer to the electrostatic field, the better is the gain of oil. The Cotrell worked during winter, therefore, much better than during summer. At hot times again better during the night than at daytime. The use of the sprinkling device on the main gas pipe was of some advantage, but comparatively little only. An effective water cooler must be used before the Cotrell, even though by this the dew-point is lowered only and the gases are dried.

d. The moisture, as mentioned already, is a further variable which can't be controlled at the meiler system. The drier the gas, the less the proportionate load of the Cotrell by precipitating water-vapors to water-condensate. This is a reason more to put a water-cooler, baffle-cooler, or shock-absorber between the meiler field and the Cotrell.

e. The concentration of the gas. Like the oxygen content, the specific content of oil, of hydrogen, of dust, etc., is vastly varying in the meiler gas owing to their origin in a not controllable carbonisation method.

f. Finally, there are as variables of the dielectric unknown admixtures, content of dust and - as we have observed at several occasions - certain "clouds" of gases of quite another composition which like large bubbles pass through the electric field of the Cotrell altering for a short period all conditions of working. The high tension was not altered, the gas temperature and the suction are constant, but nevertheless I have noticed fluctuations of the discharge current of 20 to 60% for durations varying between 1/2 and 2 minutes. The normal conditions were reached after such a cloud had passed. These fluctuations arose at all filters at the same instant and are due neither to a momentary variation of the oxygen-content nor to some electrical reason. Because of the lack of automatic gas-recorders, no proper explanation can be given. It is supposed that the clouds arise by huge explosions in the meiler. The dust-content varies, too, but never in short intervals. This depends on the kind of the heaped shale in the meiler. More fine and dry shale, especially with a remarkable percentage of dirt, as we had to carbonize often, results in more dust. Wet piles of large shale pieces gave less dust. This was made sure on several occasions by opening of the filter and checking the grade of getting dirty of the porcelain wall entrance insulators after having run different piles exactly at the same heating of the insulators. The dust content of oil showed this too.

Owing to the diversity of these disturbing components, especially if they can't be eliminated as it was the case at the standard meiler plants, the predetermination of the operation of the tar-precipitator was impossible. On the other hand, the Cotrell as a sensitive and highly developed device does not sustain these irregularities of a crude method. It cannot grant as the only condensation means in collaboration with the pile carbonization a sufficient, not to speak of a constant, oil precipitation.

Further, the following points of view at the collaboration of the Cotrell with the meiler are to be considered:

g. The equal distribution of gas on the single filters. We made sure that the velocity of gas in the different filters was by no means equal owing to the arrangement of the filters in one row and to the main pipe entrance at one end and the suction pipe outlet at the opposite end of the row. Thus the resistances to the gas suction were the greatest at the entrance (first) filter and the smallest at the (last) filter nearest to the suction outlet. A radial arrangement would have been much better. Accordingly, the velocity of gas was the highest at the last filter and decreased by steps up to the first filter. The quantity of gas was regulated at each single filter by means of valves. The "feasible load" in mA was the greatest at the (first) filter with the smallest velocity of gas and decreased correspondingly up to the (last) filter with the smallest resistance to the suction. By means of the valves this was regulated; the velocity of the gas was made equal in all filters. By doing so the total capacity of the plant was minimized naturally. The precipitation or "feasible load" is proportional to the voltage (E) and inversely proportional to the gas velocity (V) as the parallelograms of forces shows. On the other hand, it is of no use to enforce a higher "feasible load" by increasing the voltage (E) for this leads very soon to the limit voltage and to spark-overs. Further the precipitation is proportional to the density of the gas up to a certain range, possesses there a reversal point and decreases with rising density because of the fact that not all gas particles can be seized by the electrostatic field (E). If, for instance, one filter is closed by the valves entirely, the "feasible load" may rise up to 150 mA because there is no component V and all particles serve to the discharge of the condenser. Of course, this does not give any gain of oil; this filter is running idle. As is well known, this is the way of testing the insulating resistance of the filters with air and no suction. The insulating resistance was tested after repair and upkeep and showed about 160 mA at 450 V under-voltage. Besides the velocity of gas there the formation of wreaths of the gas due to extreme velocity and unsymmetrical streaming is of outstanding influence on the decrease of precipitation of oil. The vectors E and V of the electrostatic field were disturbed by these wreaths.

h. Heating of the wall entrance insulators.

The insulators are built for 110,000 Volts A.C. They are fixed between asbestos packings on the top of the filter, two for each. The inner parts of the insulator are surrounded by a sheet iron protection cylinder to perform a better distribution of the electric field. For the same reason all sharp edges in the surrounding of the insulator have to be abandoned as well as welding ridges. Otherwise, needle-effect may disturb the operation when voltages up to 80,000 Volts are used. Several insulators were destroyed by punctures at some plants. These were due to short circuits after appearance of needle-effects and due to strong heating, high voltage and soiling of the surface of the insulators. There are often strong burning traces on the surface produced by sparking-oven voltages. The surface of the insulators becomes leading after long lasting use of the Cotrell during a carbonisation process when high tension is not applied. The sediment on the isolators consists of an emulsion of tar, dust and moisture. It is very likely that the water content of this emulsion does not evaporate entirely, although the isolators were heated up to 120°C for drying purposes. The dew-point of this emulsion is seemingly above 120°C. On the other hand, the isolator will not be heated above 120°C whilst under high tension, in order to avoid a decrease of the dielectric strength of the porcelain at higher temperatures. As long as the voltage is not applied comparatively more particles of oil and dust reach by means of suction and whirls the upper part of the filter box and the isolator itself. It is essential to state that these particles were sedimented on the isolator by mechanical forces and not by condensation, for its temperature is much higher than the surrounding parts. For this reason it is not advisable to switch on the high tension on the filters later than 20-30 hours after the beginning of carbonisation.

The Cotrell plant is switched on normally 10-15 hours after the beginning of the carbonisation. During this time the isolator must be heated at full strength on about 150°C. The heating has to be decreased on 120°C roughly one hour before applying the high tension. As above intimated, these precautions do not give any full results: the first two hours the operation of the Cotrell plant after preceding several hours of carbonization are rather turbulent because of frequent spark-overs. These make the condensated emulsion on the insulator evaporate and dry the surface. The operation becomes then much smoother. If between two meilers there is a short pause only, the heating may be not switched off at all. At longer stops the heating must be interrupted to save the power. The insulators must be cleaned every 7th carbonisation. It is advisable to clean the filter boxes by means of brooms and superheated steam every 12th or 15th carbonization. Otherwise, the no load insulating resistance of the whole filter decreases.

i. Succession of the meilers. As mentioned already the Cotrell-plant can't be used within the first 8-15 hours of every meiler-pile owing to the oxygen content and to the overload by intensive suction. It was foreseen to ignite every 24 hours one meiler. The use of the Cotrell would have been diminished under these conditions to 9-16 hours per one meiler. The profitableness of the Cotrell plant in collaboration with the meiler-pile at such strictly scheduled succession would have been doubtful once more. The Cotrell requests a uniform and as much as possible continuous operation. The meiler, on the other hand, depends on various contingencies. At least in Wurttemberg we failed to overcome the latter.

k. It may be mentioned that the high tension switch room must be kept very clean and dry to avoid trouble by dust and moisture. Especially the high tension rectifier disks are very sensitive towards moisture. The room must be kept under over-pressure to avoid corrosion of contacts if corrosive gases and vapors are present. The heating of the room must be performed by electric stoves.

1. Electric power consumption of the electrostatic tar-precipitation and the rectifying and transforming losses are small. The heating of insulators and the room in a climate like Wurttemberg are power consumers worth while mentioning. These do not depend on the duration of the Cotrell's working time but must be switched on more or less continuously.

The room heating takes during the winter and at wet days 9 Kws, at dry weather at least 3 Kws. The insulator heating takes during the operation of the high tension and during the intervals between the piles 10 Kws and during the carbonisation before the switching on the high tension 15 Kws for a Cotrell plant of 5 filters, i.e. for 10 insulators.

The load of discharge and rectifying fluctuates very strongly in slope of the efficiency (i.e. "feasible load") of the Cotrell's working. Owing to high O_2-content, overloading and other disturbances the current amounted in the average to 25 mA per filter, varying for hours between 10 mA and 60 mA. This corresponds to undervoltage A.C. loads of 5 working filters of 7.5, resp., at bad precipitation 4, resp., at good precipitation 17 Kws.

The power consumption is at a carbonisation duration of 40 hours, good ignition and low O_2-content, for instance:

Rectifying and discharge current	32 hs x 17 Kws	= 550 Kwhs
Heating of insulators	32 hs x 10 Kws	= 320 Kwhs
Heating of room	32 hs x 9 Kws	= 290 Kwhs
Light and fan		= 25 Kwhs
TOTAL		1185 Kwhs

The power consumption is for instance at bad carbonization of 40 hours and dry and hot weather:

Rectifying and discharge current	25 hs x 5 Kws =	125 Kwhs
Heating of insulators	25 hs x 10 Kws =	250 Kwhs
Heating of room	25 hs x 3 Kws =	75 Kwhs
Light and fan	=	25 Kwhs
	TOTAL	475 Kwhs

In each case there must be added the consumption for heating beyond the working time of the Cotrell. Assuming a pause between two meilers of say 20 hours in the first case:

Heating of insulators	20 hs x 10 Kws =	200 Kwhs
Heating of insulators	8 hs x 15 Kws =	120 Kwhs
Heating of room	28 hs x 9 Kws =	250 Kwhs
	TOTAL	570 Kwhs

At a pause of say 40 hours in the second case:

Heating of insulators	40 hs x 10 Kws =	400 Kwhs
Heating of insulators	15 hs x 15 Kws =	225 Kwhs
Heating of room	55 hs x 3 Kws =	165 Kwhs
	TOTAL	790 Kwhs

In the first case we get a total of 1185 + 570 = 1755 Kwhs, whereby the ratio of load current to idle running current is around 2:1. Only 550 Kwhs or 31% of the total consumption serves directly to the oil gain.

We get in the second case a total of 475 + 790 = 1265 Kwhs, whereby the ratio of load to no load is about 1:2. Mere 125 Kwhs or 10% of the total consumption serves to the oil precipitation. The costs of electric power are RMk 105.- resp. 76.- at an average price of RMk 0.06($0.006) per one Kwh.

It may be intimated in this connection that the power consumption is of little importance if the power factor of the respective factory is bad, because the Cotrell load is an ohmic or even reactive load by which the power factor of the whole factory is improved and the Kwh-price lowered. At a good power factor this is not the case and at long pauses of carbonization the Cotrell consumption may rise to several thousands of Kwhs. In the average it amounted on 1300 Kwhs and was about 7% of the exhausters' consumption which took per meiler 18,500 Kwhs.

m. <u>Profitableness of the Cotrell plant at the meiler method.</u> If we calculate the power costs of the two examples per one hour of effective working, we get in the first case 55 Kwhs (i.e. RMk 3.30) and in the second case 51 Kwhs (i.e. RMk 3.06) per one working hour. The total costs are in this case:

Power costs	RMk	3.30	$0.33
Wage costs	RMk	1.50	0.15
Annuity and interest on capital	RMk	6.00	0.60
TOTAL	RMk	10.80	$1.08

per one working hour.

The price of a Cotrell-plant with 5 filters is around RMk 200,000.--. The legal extinction would be possible in 10-15 years. In connection with the meiler carbonization method the duration of life must be calculated mere 5 years because of the limits of the other apparatus. At the best 15 meilers monthly, each with an average Cotrell-working-time of 32 hrs, may be calculated. From which we get 5750 working hours per annum and in 5 years 29,000 working hours of the Cotrell. It means every working hour is debited with RMk. 6.- of capital costs.

The costs of maintenance and repair of the Cotrell plant are very little and may be neglected. The gain of oil by means of the tar-precipitator is according "Lurgi" 12-20% of the total output of one meiler. These data were checked in the Pilot Plant where we took as an average 15%. On the other hand, the precipitation on the standard plant must have been much less due to the mentioned circumstances. If we calculate 10% at the best, we get 3 tons of oil. Then 3 tons are debited with RMk. 345.-, i.e. per one ton RMk 115.- or per one barrel $1.80. Taking in consideration that the cost price per ton was $80.00 to $95.00, or per barrel $12.70 to $15.00, one comes to the conclusion that this additional oil is very "cheaply" gained.

5. CONCLUSION AND SUMMARY

After having read the above report one might have got the impression that the meiler-carbonization method is not worth while mentioning at all. But this is not quite right. I have tried to emphasise that many faults of the Wurttembergian experiment are due to the climate, human and technical insufficiencies. The main faults to be removed are:

a. Erection of such plants in an extremely inopportune climate.

b. Conclusion from a tiny test plant to the industrial stage.

c. Erection according to the principles of war defence.

d. Principle wrong technical outfit.

e. The meiler itself was too little mechanised.