the expert there being Dr.Ipfelkopfer. Lurgi has had no experience with the use of charcoal or other absorbents for selective fractionation of different types of hydrocarbons.

## VI. CARBONIZATION.

Lurgi Gesellschaft Für Wärmetechnik has developed improvements in the carbonization of fuels employing the "Spülgas" or direct heating principle and, in association with Krupp of Essen, a low-temperature carbonization effected by indirect heating of the fuel in closed metal-walled cells or ovens having each an average width of between 70 to 100 mm. According to Dr.Heine's expressed opinion, the former process is the most satisfactory for briquettes and the latter method and apparatus for the processing of weakly-coking coals to provide coke of rugged structure having a residual content of about 10% volatile matter, or less.

## A. Spülgas Process.

This process of Lurgi was developed primarily for the low-temperature carbonization of lignite, brown coal, xylite and lump fuels having a water content of 25-30%. Earthy German brown coals having a water content of 50-55% have been also treated but, since they are crumbly and disintegrate rapidly on heating, they are first briquetted (without a binder) and then carbonized. Oil shales, torbanite and cannel coal can be carbonized with very satisfactory results. Long-flame younger coals can be processed ranging from those that disintegrate to those which cake slightly on heating.

Dr.Heine also explained that the Spülgas process had been applied successfully to bituminous coal. At Blechhammer, the 4,200 tons plant (300 tons per generator per day) had 5 generators operating on slightly-coking coal making hard coke for transport producers and the production of hydrogen. The Meurice index of the coal was 12 and the fines were briquetted with 4% sulphite lye and pitch before carbonizing. The tar (10% yield) was divided into asphalts, acids and "Navy oil" over 230°C. (6/7 per cent). The economics of the process were termed "doubtful", when hard coke sold for 22 RM, Lurgi coke brought 25 RM and producer fuel 28 RM; the cost of the coal was 15 RM. Obsolescence was taken at 2 to 3 years.

In the Lurgi Spülgas process, the employed coal is treated successively to drying, carbonization and cooling steps in a retort of special design, the latter step being so arranged that the heat removed from the cooled coke is returned to the carbonization zone. The above three process steps are carried out in zones that are vertically disposed in respect of each other in the retort which

is open at the top (the hopper) and closed at the bottom by the coke-discharging mechanism. In both the drying zone and the carbonizing zone, the descending flow of fuel is treated successively in direct contact to separate circulation of gases that are appropriately previously by admixture with hot combustion products formed extraneously and thereafter introduced into the circulated gases. Manifestly, there is considerable difference in temperature between the two circulated gas streams; the circulated gases of the drying zone are usually preheated to about 200-250°C. whereas the chosen temperature of those gases circulated into the carbonization zone depend upon the characteristics of the fuel being carbonized and the temperature at which it is best treated, but the usually employed temperature is within the range of about 495° to 650°C. although in special instances, where it is desired to produce a coke having a volatile-matter of from 3-5%, it is feasible to employ preheating temperature of about 800°C. which increases the hardness of the coke; the tar-yield however is not appreciably affected by the temperature of carbonization. Gases leaving the carbonization zone are passed through condensers, tar-extractor and light-oil scrubbers before their return to the carbonization zone. The greater portion of that recirculated gas recycled into the carbonization zone is first, by special construction of the automatic coke-withdrawing pocket, admitted into contact with the hot coke leaving the carbonization zone for cooling the latter; a small portion of said circulation gas supplies the two combustion chambers whereby those gases, admitted to the carbonization zone and the drying zone, are heated.

In consequence of the above method of heating the circulated gas-streams, the formed carbonization gases are of course diluted with combustion products, but they comprise a relatively minor proportion of the total, and the mixed product, in the case of brown coals, burns quite easily as it has a calorific value of 140-200 BTU per cu-ft. In those special instances where there exists a good market for the distillation gases in undiluted form, heating of the recycled gases is effected by combustion-products in indirect contact in tubular heaters. This modification of the process is of value only for those fuels yielding gases containing little carbon dioxide.

The raw coal being thoroughly dried and preheated before it enters the carbonization zone and, in addition, the recycled gas itself being preheated by the hot coke before it also enters the carbonization zone, the carbonization-heat requirement is thus reduced to a minimum. In result, the amount of combustion-products required adding to the recycled gas for heating purposes is relatively small and the volumes of gas that are required to be handled in the condensing, scrubbing and light-oil recovery apparatus are not substantially greater than those encountered in externally-heated retorts.

The shafts of the retorts, as above-mentioned, are open at
the top and some of the circulated gases from the drying zone are
continuously discharged therefrom - they consist principally of com-
bustion products and steam and contain little free oxygen. The retort
shafts are operatively divided into three zones and the fuel moves
freely from the one to the other. There are no closing devices or
moving parts in or between the zones so that the retorts are relative-
ly compact. The gas off-take must be large to minimize the amount of
dust carried into the condensing equipment.

The preferred size of carbonization zone has a capacity
not in excess of 250-350 tons per day. Larger size installations
present problems of distribution of materials and heat so that there
may result unequal carbonization throughout the charge. For larger
capacities, it is advisable to build several unit retorts having each
the above capacity. It has proven feasible to supply at least two
retorts from the same combustion apparatus and with the same off-take
mains.

In the cases of younger coals which disintegrate on heat-
ing, and the formed small-sized coke has only low market value, the
fuel is briquetted in, for example, a Krupp ring-roll press; the pro-
duced briquette coke is of lump size and is a more desirable product.
In the briquetting, after the fuel is ground and dried by contact with
hot combustion-products at a temperature of 900-1000°C. to a moisture
content of 6-10%, it is introduced into the ring-roll press as a
powder and briquetted at a pressure of 30,000 to 35,000 lbs/sq.in.
These briquettes are then used as the charging stock to the Spülgas
retorts. They either retain their shape during the carbonization
process or break into a few large pieces. It is claimed that the re-
sultant product is a dense, abrasion-resistant fuel. This method of
briquetting is claimed to be more economical than by briquetting after
carbonization.

In large plants of the "Spülgas" type, it is customary
fractionally to condense the tar. The gases leaving the retorts are
first freed of dust and then cooled to about 100°C. and the heavy tar
removed by Cottrell precipitators. The gas is next cooled to 20-30°C.
and the light-tar fraction condensed and finally the light oil is
removed in an oil scrubber. Thereafter, circulating gas is returned
to the carbonizing ovens for recirculation through the fuel charge.

Successful operation of Spülgas plants depends upon uni-
form distribution of heat over the treated fuel, and to effect this
result, maintenance of definite particle size and the treatment of a
fuel that is either non-coking or only weakly so, are important
factors.

The following data are given as characteristic Material and Heat Balances for a Lurgi Spülgas plant operating on brown coal briquettes containing 16% water and 14.3% tar, as determined in the Fischer-Hempel assay.

| Input | Lb. | B.T.U. | % Heat |
|---|---|---|---|
| Brown coal Briquettes | 1.000 | 9876.6. | 99.92 |
| Air for combustion | 0.425 | 8.1. | 0.08 |
| Total : | 1.425 | 9884.7 | 100.00 |

Output.

| | Lb. | B.T.U. | % Heat |
|---|---|---|---|
| Semicoke | 0.445 | 5627.5 | 56.96 |
| Surplus gas * | 0.253 | 676.7 | 6.84 |
| Refined tar | 0.116 | 2086.9 | 21.10 |
| Light oil (below 200°C) | 0.023 | 434.0 | 4.39 |
| Liquor | 0.081 | 2.9 | 0.03 |
| Residue | 0.009 | 122.2 | 1.23 |
| Gas loss | 0.001 | 12.4 | 0.13 |
| Heat in cooling $H_2O$ | | 225.9 | 2.28 |
| Waste gases | 0.487 | 185.3 | 1.88 |
| Loss | 0.010 | 510.9 | 5.16 |
| Total: | 1.425 | 9884.7 | 100.00 |

* 7400 cu-ft./ton (30 m.60°F.)

## Characteristics of a Typical Spülgas-Process Tar

| Carbonization Temperature (Gas entering carbonization zone) | Distillation | | | |
|---|---|---|---|---|
| | Excluding Light Oil | | Including Light Oil. | |
| | Up to 180°C. | Up to 320°C. | Up to 325°C. | Up to 325°C. |
| 650°C | 1.4% vol. | 43.8% | 47.4% | 52.0% |
| 590°C | 1.4% vol. | 44.5% | 46.5% | 46.5% |
| 495°C | 1.6% vol | 45.3% | 48.1% | 48.1% |

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| Carbonization Temperature (Gas entering carbonization zone) | Creosote & Asphalt Contents of Tar. (Excluding Light Oil). | |
|---|---|---|
| | Creosote | Asphalt |
| 650°C | 14.5% wt. | 2.3% wt. |
| 590°C. | 14.3% wt. | 1.6% wt. |
| 495°C. | 15.2% wt. | 1.3% wt. |

The low-temperature tar obtained by the carbonization of lignite and bright brown coals is mostly distilled to give fuel oil, Diesel oil and impregnating oils; it is also used as a source for pure phenols. Considerable quantities of the tar are used as raw stock for high pressure hydro-genation by the I.G.Farben methods to produce motor fuel, Diesel oil and lubricating oil.

The low-temperature tar from the Spülgas process can be, by hydrogenation, to the extent of 80% converted into gasoline. It can, at a pressure of 250 atm. and 400-450°C. and with only 10% loss, also be converted into -

| | |
|---|---|
| Gasoline | 20% |
| Diesel Oil | 39% |
| Lubricating Oil | 17.5% |
| Paraffin | 13.5% |

The largest Spülgas Plant built by Lurgi has a throughput of 12,500 tons per day of brown coal. The coke from this particular plant is used primarily for the production of steam in an adjacent power plant. The remainder of the coke is gasified to produce water gas and hydrogen which is used for the hydrogenation of the low-temperature tar.

Cold Extraction Methods for Tar Refining.

Experimental work has been done by Lurgi on cold extraction methods for tar. Plants were under construction at Hirschfelde and Altenburg. The process had been applied only to brown coal tar and involved:-

(1)  Extraction of the tar with amyl and butyl acetates to remove phenols;

(2)  Extraction of the 200-350°C. fraction with
methyl alcohol giving 50/50 phenols and neutral
oil, the latter being a Diesel oil of 35 cetane
number and was suitable for blending with Fischer-
Tropsch oil.

Dr.Herbert explained that this research work had been done
his laboratory on Mouson Strasse in Frankfurt.

## Low-Temperature Carbonization by Indirect Heating.

In association with Friedrich Krupp A.G., Essen, Lurgi
developed a new type of oven for distilling at low-temperature
static vertically-disposed beds of coking or weakly-coking coals
in small or nut sizes, or of briquetted fine coals, by indirect
contact with hot combustion gases. This new distillation oven is
based on employment of metal-walled cells as the coking chambers.
As the basic unit has been used successfully, a group of metallic
cells or an oven, each comprising six rectangular coking spaces
that are disposed in alternation with seven similar heating spaces;
the structure is thus reminiscent of the well-known high-tempera-
ture horizontal coke-oven in its configuration. Each such unit is
entirely surrounded by a housing through which is circulated
horizontally along the heating spaces for the cells extraneously
produced combustion-products that are maintained at an elevated
temperature. The fuel to be carbonized is charged into the top
of the cells as is common practice in the horizontal coke oven out
the formed coke is withdrawn from the bottom thereof by gravity. A
charging machine that moves over the tops of the units is provided
with a device for stamping the charged fuel; this device can be
used, if necessary, to assist in discharge of the coke.

The coking spaces of the metallic cells have each an
average width of about 85 mm; they are about 75 mm. wide at the top
and 100 mm. at the bottom. In height and length they are respect-
ively 2.1 and 3.1 meters and the six coking spaces comprising an
oven unit have coal capacity of 2.75 metric tons.

The ovens are formed of normal heavy cast steel parts that
are welded in a special manner (not learned by the investigators).
This subject and technology should be further investigated. Some
years of operation in large-scale, it is claimed, have occasioned
no bending or warping of the cell walls and periodic metallurgical
tests made on them have shown that their original structure has

remained unaltered and that no alteration in the carbon content of the steel has been determined.

Heating of the metal ovens and their individual coal cells is effected by circulating hot products of combustion through those heating spaces that alternate with the coking spaces. These circulated hot gases enter the ovens at a temperature of about 600-620°C. at one face and suffer a drop in temperature of about 40° to 60°C. at their outlets at the opposite face; the gases are then returned to the inlet side of the ovens and there reheated by admixture with hotter combustion-products before returning to the coking chambers. Thus, all required combustion for heating purposes is performed out of contact with the metallic walls and the gases coming into contact with them are limited to an oxygen content of less than 0.8 to 1.05%; by means of oxygen-recording apparatus the oxygen content of the combustion-products is constantly supervised in order to protect the cell walls. Also there has been no evidence that sulphur compounds in the heating gases have attacked the cell metal.

In the heating-gas system, the recirculating fans for the heating gases are of special construction and the movable parts such as the shaft and fan-blades are formed from special steel which it is claimed has been satisfactory. The construction, manufacture and identity of the employed steels should be determined by investigators. The cells are provided, in the heating system, with guide-plates (Führungsblechen) adapted so to direct the hot gases that the lower wider portions of the single cells with their larger content of coal are heated to a higher temperature than the upper portion but in accordance with the vertical taper of the cell to produce uniform heating from the top to the bottom thereof and a uniform carbonization throughout the cells. Cooler combustion-products that are outlet to the heating spaces are displaced from the heating system in a quantity equivalent to the volume of fresh-combustion products that are added for reheating purposes. The hot purged gases still have a considerable heat content that can be employed for a wide variety of purposes.

The low-temperature coke is discharged from the cells as large flat plates of uniformly rugged structure throughout. It was stated that in the drum test 76.5% of the coke was over 40 mm. and produced only a minor amount of breeze on crushing. The coke has 8-10% volatile matter and depending of course on the characteristics of the treated coal a heating value of 8000-8400 kcal/kg. based on the ash-free coke. It is smokeless burning; also depending upon the characteristics of the coal, the coke yield is 75 to 85% by wt. of treated coal. The statement is interesting that, in contrast to high-temperature coking procedure, the best low-temperature coke made in the metal ovens is that prepared from the

larger sized coal rather than from the finely ground.

The following table shows the size composition of gas or gasflammkohle suitable for use in the Krupp-Lurgi cells:-

| | |
|---|---|
| 0 to 1 mm | 30 to 40% |
| 1 to 3 mm. | 30 to 40% |
| 3 to 6 mm. | 20 to 30% |
| 6 to 10 mm. | 5% |

## Other Products

The low-temperature tar is rich in oils and is practically free of dust and has a low content of pitch.

The distillation gas has a gross heating value of 7800-8200 Kcal/m$^3$ and a relatively low hydrogen content. Depending upon the coal, the gas-yield is 70-120 Nm$^3$/ton.

The recovered benzin amounts to 0.9-1.3% by wt. of the carbonized coal. It has a high content of aromatics and consequently is good motor fuel of high octane number and after washing by known methods shows good stability in storage.

The ammonia content is so low that no provision is made for its recovery; the gas is merely water-washed for its removal.

The following table gives a comparison between yields obtained in the Krupp-Lurgi metal cells and in high-temperature coke ovens processing one ton of the same dry Ruhr-Gas-Fettkohlen mixture having 27% volatile matter:-

## Carbonization.

| Low-Temperature. | High Temperature |
|---|---|
| 820 kg. Coke (dry) | 750 kg. Coke (dry) |
| 90 Nm$^3$ Gas | 320 Nm$^3$ Gas |
| 8000 Kcal/Nm$^3$ (gross) | 4600 Kcal/Nm$^3$ (gross) |
| 11 kg. Crude Gasoline | 11 kg. ammonium sulphate. |
| 60 kg. Tar usable as heating oil. | 9 kg. Crude light oil |
| | 34 kg. tar. |

| Normal Distillation of the Tar. | Normal Distillation of the Tar. |
|---|---|
| About 1% water | About 2-4% Water |
| 3-4% Crude Gasoline | 0.8-1.2% Crude light oil |
| 15-20% Acid Oils | 4-6% Middle oil |
| 37-47% Fuel Oil | 7.10% Heavy oil |
| 30-40% Pitch | 20-25% Anthracene oil |
| | 4-6% Naphthalene |
| | 55-60% Pitch |

The values in the following table show the characteristics of the low-temperature coke obtained in the Krupp-Lurgi cells when operating on a high-volatile coal of low-ash content.

### The Coal:-

Approximate analysis:-

| | |
|---|---|
| Water | 8.2% |
| Ash | 3.24% |
| Volatile matter | 22.4% |

Screen analysis:-

| | |
|---|---|
| 0 to 1 mm. | 38.7% |
| 1 to 3 mm. | 34.4% |
| 3 to 6 mm. | 23.2% |
| 6 to 10 mm. | 3.7% |

Bulk Density Moist Coal 0.846 to 1 $m^3$

### The Low-Temperature Coke:-

Approximate analysis:-

| | |
|---|---|
| Ash | 3.78% |
| Vol. matter | 9.1% |

Ultimate analysis:- (ash-free basis)

| | |
|---|---|
| Carbon | 91.45% |
| Hydrogen | 3.76% |
| Sulphur | 0.95% |

Heating Value (ash-free basis) - 8240 Kcal/kg (gross)

| | |
|---|---|
| Ignition point (air) | 396°C. |
| Porosity | 50.4% |

Drum Test:-

    Over 40 mm.                      76.5%

Size of the Crushed Coke:-

    Over 40 mm. - 77.2%

    40-20 mm.   - 9.8%
    20-10 mm.   - 3.8%
    10-6  mm.   - 2.0%
    Below 6 mm. - 7.2%

A typical low-temperature tar obtained from mixed coals showed the following characteristic analysis:-

| | | |
|---|---|---|
| First Drop to $200^\circ$C. | - | $97.5^\circ$C |
| $200^\circ$ to $230^\circ$C. | - | 3.0% |
| $230^\circ$ to $270^\circ$C. | - | 12.5% |
| $270^\circ$ to $300^\circ$C. | - | 13.5% |
| $300^\circ$ to $360^\circ$C. | - | 10.0% |
| | - | 21.5% |
| | | ———— |
| | | 60.5% |

| | | |
|---|---|---|
| Specific Gravity | - | 1.058 |
| Viscosity $20^\circ$C. | | $35^\circ$ E. |
| $50^\circ$C. | | $4.5^\circ$ E. |
| Pour Point (Stockpunkt) minus | | $1^\circ$ C. |

The low-temperature tar is of such character that after careful de-watering it can be used as a heating oil without further refining and it is claimed that 50-60% of the total tar yield can be easily used for fuel in slow and moderate speed Diesel motors. About 10% of the Diesel oil fractions are carbolic oils having a relatively large content of meta-cresol. The high-boiling fractions of the tar are especially valuable, because of their high hydrogen content, as raw material in high-pressure hydrogenation plants.

Water-gas produced from the coke is said to contain CO and $H_2$ in the ratio of 1:1.5 instead of in the ratio of 1:1.25 obtained from high-temperature coke.

The underfiring requirement per kilogram of coal is 500 kg. cal. which is equal to 900 BTU per pound of charged coal.

The coking time is about 6 hours for the substantially 3.5 in. thick cell charges.

Gas formed by the process of coal distillation on a mixture of 75% gas coal+ 25% Fettcoal has the following analysis:-

| | | |
|---|---|---|
| Carbon dioxide | – | 4.5% |
| $C_n H_{2n}$ | – | 2.5% |
| $C_x H_y$ | – | 0.8% |
| Oxygen | – | 0.2% |
| Carbon Monoxide | – | 3.5% |
| Hydrogen | – | 23.8% |
| Ethane | – | 13.6% |
| Methane | – | 46.1% |
| Nitrogen | – | 5.0% |
| Density | – | 0.605 |
| Gross Heating Value | – | 8137 Kcal/$m^3$ |

The gas driven off the low-temperature coke by heating the same to 1000°C according to the laboratory method of Bauer has the following analysis; it amounts to about 225 $m^3$ per ton of said coke:-

| | | |
|---|---|---|
| $C_x H_y$ | – | 0.3% |
| Carbon Monoxide | – | 7.4% |
| Hydrogen | – | 77.2% |
| Methane | – | 12.5% |
| Nitrogen | – | 2.0% |
| Gross Heating Value | – | 3840 Kcal/$m^3$. |

The Krupp-Lurgi metallic coking cells were developed for coking at low-temperature of between 500-600°C. both coking and weakly-coking coals. Most any such coals as exhibit ability to form coke can be processed in them. Non-coking coals preformed into egg-shaped briquettes by means of bituminous binders, and the like, have also been processed to advantage and the coking time is reduced to about 65% of that required when the cells are charged with the same coal in unbriquetted condition. This increased capacity in many cases covers the cost of the briquetting.

The philosophy behind the development of this type of coking oven or cell has been stated as follows:-

The forming of a good- low-temperature coke depends upon a sufficiently rapid inflow of heat into the cells, between temperatures of 350° and 450°C., to bring all portions of a charge to the melting point about simultaneously. The good heat conductivity of the metal walls and the relative thinness of the charges makes this possible.

The more rapidly the temperature is raised, the more quickly all constituents that take part in the forming of the coke exert their effects and the less opportunity the divers constituents have to exhibit any possible distinctions in their reactivities such, for example, as differences in pre-,principal- and post-gasification temperatures and to