decompose either too early or too late to make a substantial contribution to the cementation of the coal particles. By bringing all parts of the coal charges simultaneously to the molten state no highly viscuous plastic seams are formed through which gases can escape only by exerting high pressures. Consequently, such coals as in the horizontal high-temperature coke ovens exhibit damaging expansion pressures against their heating walls, can be coked in the metallic cells without any serious pressures developing. Such highly expanding coals as those having 19.5 to 21% volatile-matter content have been coked in these oven-cells without grinding or any reduction in bulk density and without any resultant deleterious effect on the metal walls of the coking cells.

Reference is made to a Report by Drs.Hall and Powell on CIOS Target 30/5.02 for further data on the low-temperature carbonization ovens.

VII. HIGH PRESSURE GASIFICATION.

In the high pressure gasification process developed by Lurgi Wärmetechnik, solid fuel is continuously gasified at from 20-30 atm. pressure with oxygen and super-heated steam. The generator is of special construction and is designed as a pressure vessel having spaced double walls that provide a water jacket and those portions of the generator that are subjected to pressure are protected from overheating by a lining of refractories. Double-valved charging and discharging pockets at the top and the bottom of the generator serve respectively to charge the fuel and discharge the granular ashy residue in much the same fashion employed in gas-producers operating at atmospheric pressure or slightly thereabove. The charged fuel rests in the generator on a variable-speed rotary grate; the oxygen and steam are flowed upwardly through the grate and the fuel bed. The reaction between the fuel, steam and oxygen proceeds with decreasing temperature from its hottest part immediately above the ash-bed to the upper part of generator and passes respectively through the phases of combustion, gasification, distillation and drying of the fuel.

Gasification of coal under pressure with oxygen produces a gas-product having a relatively high content of carbon dioxide and methane along with carbon monoxide and hydrogen. With increasing pressures up to about 20 atm., the proportions of methane and carbon dioxide increase in their contents in the crude gas, whereas the hydrogen and carbon monoxide contents decrease. Since the crude gas leaving the generator is at about 20 atm. pressure the carbon dioxide is to a great extent easily removed by scrubbing with water leaving a residue-gas of high calorific value (4000-4600 Kcal/Nm^3) that is, substantially equivalent to coke-oven gas. A typical crude gas and its purified product, as prepared from brown coal at 20 atm. pressure with oxygen is as follows;-

	Crude Gas	Purified Gas.
CO_2	30.6% by vol.	3.0% by vol.
C_nH_m	0.6	0.5
O_2	0.1	0.1
CO	16.5	22.8
H_2	34.0	48.7
CH_4	16.3	22.6
N_2, etc.	1.9	2.3

The oxygen consumption is relatively low and because of the exothermic heat of reaction of the produced methane, hot spots within the fuel bed are avoided. This in turn makes it possible to control slag-formation and consequently to maintain uninterrupted operation in a closed generator without any disturbances.

Normally, only solid fuels that are weakly-coking or non-coking are employed in the process; for example, lean bituminous coals, and all brown coals. Easily coking coals should be given a pre-treatment to destroy some of the coking properties. The water content of the employed fuels should not be over about 25%.

The preferred sizes of the fuel are about 5-25 mm. and especially about 5-15 mm. The fuels should be practically free of dust; that is, no constituents less than 2 mm. Throughputs of up to 1000 kg. per hour per m^2 of generator cross-section have been reached with fuel consisting of grain sizes of 2-8 mm.

The heating value of the produced gas depends on the reactivity of the fuel. At 20-25 atm., a gas having a gross heating value of 4300-4600 Kcal/Nm^3 is obtained when using brown coal. Bituminous coal or coke at the same pressure gives gas of a heating value about 200 Kcal. lower. The high conversion of carbon to carbon dioxide does not mean any loss of heat because, in the process, Hydrogen and methane are formed and the involved reactions converting CO to H_2 or CH_4 are always accompanied by the production of the inerts CO_2 and H_2O; and the high conversion of chemical energy is thus not influenced by large CO_2 formation.

The pressure-generator gas has the following characteristics:-

a) Relative Density (Air = 1) less than 0.5;
b) Oxygen content " " 0.3% by vol.;
c) It is substantially free of H_2S, NH_3, Naphthalene and tar;
d) Its content of organic sulphur when employing high-sulphur coals is less than 20g/100 Nm^3 and by strong aeration of the water employed for removing the CO_2 it can be further reduced.

In consequence of this scrubbing of the crude gas with water under pressure, the effluent gas contains not more than 5-10 grams H_2S/100 Nm^3; complete purification thus requires only a small oxide-purification installation. Cooling the gas under pressure also provides it with a very low partial-pressure of water vapour.

Installations.

The following plants have been built by Lurgi, all of which are built around the high-pressure generator:

1. Hirschfelde near Zittau: This was the first commercial plant and had a capacity of 5 million m^3 per year of City gas.

2. Böhlen near Leipzig: Two plants having a total capacity of 150 million m^3 per year.

3. Brüx in Czechoslovakia: This plant was designed for a capacity of 100 million m^3 per year.

The first plant at Böhlen had 5 generators built in 1939 and to this the second plant, built in 1942, added 5 more. The Brüx plant was built in 1943.

The Generator.

The generator is a pressure vessel lined with bricks and is furnished with a water-cooled jacket. Despite the high pressure of operation, the coal-charging and ash-discharging pockets are so designed and provided with valves that the generator is continuously operative during both the charging and the ash-removal steps.

The outside shell of the generator is of ordinary steel and is 50 mm. thick, whereas the inner shell of the water-jacket is boiler steel about 25 mm. thick. One of the important points in design for successful operation is the refractory lining; it must be just thick enough to permit reaction-heat to be sufficiently rapidly carried away to prevent slagging of the coal ash. In those generators having a diameter of 2.5 meters (O.D.3.m), it is about 250 mm. thick. The latest generators have the above-stated diameter,but that of the Hirschfelde installation was 1 meter. Dr.Danulat said that the only limits, however, to diameter was transport considerations.

The rotatable grate that supports the fuel-bed is formed of cast chrome-nickel steel; its speed of rotation determines the amount of ash delivered to its discharge-pocket.

The valves employed for sealing these pockets from the generator during their operation are provided with rubber sealingring so-formed and supported as to expand against the valve seat under the influence of the gas pressure. It was stated that the Brüx generators, which have not been seen, have an internal hydraulic ram in the coal pocket for opening the upper valve. The hardened-steel cone valve is of V_2A steel. Incidentally, it was also stated that bituminous coal has been gasified successfully, but optimum conditions had not been established, and Dr.Oetken stated that a true coking coal had never been employed in the pressure generator; however, Ruhr anthracite and the so-called "gas flammkohlen" can be used. Too strongly coking coals are precluded from use.

In the operation of the generator, the carbon dioxide content of the raw gas issuing therefrom is the control index of operating conditions. The temperature of gasification as measured by Seger cones in the fuel bed is 1050-1100°C.

The outlet temperature to the generator is controlled by the moisture content of the brown coal - with 20% water the temperature is 300-320°C. The velocity of the outlet gases is maintained at about 2.5 meters per second so as to prevent precipitation of tar and dust in the upper part of the generator.

The super-heated steam continuously flowed into the lower part of the generator is prepared by combustion of the high-pressure gas discharged from the generator's coal-pocket during its charging. A special relief holder is provided for its storage.

Coal Capacity of a Generator.

A pressure-generator having a 5 m^2 cross-section when operating at 20-30 atm. can gasify:-

100 to 130 tons of dried brown coal per 24 hrs, or
60 to 70 tons of bituminous coal.

This corresponds to a capacity per generator of 2750-3500 Nm^3 city gas per hour, or 20,000,000 Nm^3 per year.

Normal operating rates can be reduced to one-third of capacity and temporary fluctuations are possible without giving rise to any operational difficulties, thus making operation very elastic.

Gas Yield of a Generator.

The attainable gas yield depends on the characteristics of the fuel. Fuels high in moisture and ash naturally give a lower yield;

the same obtains for the brown coals since the higher yields of liquid products - tar, oils and gasoline - reduce the amount of gasifiable carbon of the coal.

1 ton dry brown coal (20% moisture,10% tar) gives 680-580 Nm^3 gas having a gross heating value of 4300-4500 Kcal/m^3.

1 ton lean coal yields 1570-1670 Nm^3 having a gross heating value of 4100-4300 Kcal/m^3.

1 ton lean coal yields 1570-1670 Nm^3 having a gross heating value of 4100-4300 Kcal/m^3.

<u>Results of Pressure-Generator Operations on Various Fuels.</u>

		Brown Coal (Lausitz)	Brown Coal. (Middle Germany)	Lean Coal Fines (Ruhr)
1. Composition of the Fuel				
Combustible Substance	%	67.5	76.3	88.4
Water	%	27.4	14.8	6.6
Ash	%	5.1	8.9	5.0
Tar (Fischer Method)	%	10.2	12.6	
Gross Heating Value Kcal/kg.		4730	5260	7600
Grain Size	mm.	2.10	2.10	3.10
2. Operating Pressure at.		20	20	20
3. Throughput Kg/m^2h		750	770	310
4. Composition of Purified Gas.				
CO_2	%	3.0	2.3	1.0
C_nH_m	%	0.5	0.9	0.3
O_2	%	0.1	0.2	-
CO	%	22.8	22.0	27.9
H_2	%	48.7	50.7	52.4
CH_4	%	22.6	21.8	16.9
N_2	%	2.3	2.1	1.5
Density Gas (air = 1)		0.448	0.435	0.432
Gross heating value Kcal/Nm^3		4280	4500	4100
5. Gas Yield Nm^3/to coal		760	680	1690

Results of Pressure-Generator Operations on V arious Fuels. (Contd.)

	Brown coal. (Lausitz)	Brown Coal Middle Germany)	Lean Coal Fines (Ruhr)
6. Oxygen Requirement Nm^3/Nm^3 Gas	0.15	0.145	0.198 *
7. Steam requirement Kg/Nm^3 gas	0.01	1.06	1.40

* For comparative purpose, it is noted that the Winkler Generator employs slightly more than twice as much oxygen per m^3 of gas produced.

If, in the case of the above bituminous coal, the pressure is raised to 30 atm., the calorific value of the gas is raised from 4100 to 4300 $Kcal/Nm^3$

Recovery of Liquid Hydrocarbons.

The crude gas leaving the pressure-generator at 300-350°C. is cooled in two steps. The first step is performed in a direct-spray cooler wherein the temperature is reduced to 120-140°C. and the second step in a tube-cooler that reduces the gas temperature to 30°C. This condenses much of the tar and water.

From the above description of the process, it is manifest that this gasification method is broadly a counter-current process which naturally protects somewhat the original hydrocarbons in the coal and makes it possible to recover, as tar, up to 85% of the tar content of the coal, as determined analytically.

The total tar is different from that obtained by atmospheric-pressure low-temperature distillation by the high proportion of hydrocarbons boiling up to 200°C. which is increased about 20%. The increased pressure has little effect on the other characteristics (Creosote, Paraffins, etc.) since the produced gas acts as a Spülgas. Only the high-boiling resins and asphalts are in part decomposed.

Dust and water content of the tar are similar to those of a good low-temperature tar. The gasoline fraction has an octane number of 90-100, and after refining, is a good motor fuel. The good quality of the tar makes a preliminary low-temperature carbonization of bitumen-containing fuel superfluous.

The benzine of the light-oil fraction is recovered by scrubbing the gas cooled to 30°C., or the like, with a scrubbing oil while it is still under pressure. The concentration of benzine is the scrubbing oil is 6-8 percent by volume.

Purification of the Gas.

After the above oil scrubbing, the gas passes, under pressure, to a water-scrubber wherein it is treated with about 700-800 m^3 of water per hour per 6000 m^3 of clean gas. The scrubbing water is then expanded in Pelton pumps, aerated and recirculated. This removes carbon dioxide and a large amount of the hydrogen sulphide. Residual hydrogen sulphide amounting to about 5 g. per 100 m^3 is removed in iron oxide.

If the employed coal has a high content of sulphur, the gas issuing from the water scrubbing will contain less then 20 g/100 Nm^3 of organic sulphur; it can be removed by the Ruhrchemie process employing alkaline iron oxide.

Heat Balance of the Process.

A. Heat Input

	in %
1. - Coal	89.9
2. - Steam (saturated)	10.1
	100%

B. Heat Output

1. Chemically combined heat in the town gas.	62.2%
2. In tar and benzine	14.3%
3. Phenols in gas liquor	0.9%
4. Carbon in ash	0.4%
5. Steam from cooling jacket	0.6%
6. Sensible heat of gas & heat losses.	16.2%
7. Losses in steam superheater and aeration tower.	5.4%
	100%

The efficiency of fuel conversion to gas is thus about 85.2%

Production cost of the city gas using brown coal was stated to be about 2.4 pfg. per m^3 as compared with coke-oven gas at 1.8 pfg. delivered to the distributor; that is, if the dried brown coal costs 6.5 RM per ton.

Synthesis Gas.

In the Fischer-Tropsch synthesis of hydrocarbons from CO and H_2 as usually practised with the cobalt-thoria catalyst, the employed Synthesis Gas preferably has a H_2/CO ratio of 2. The lesser the content of inerts, the better. As above indicated, the pressure-generator gas product, after removal of carbon dioxide, contains hydrogen and carbon monoxide in some instances in a little higher than the said ratio of 2 and in the other instances, somewhat less; it also contains somewhat over 20% of methane. Such large proportion of methane would seriously limit the output of any Fischer-Tropsch hydrocarbon plant as it acts as a diluent and does not take part in the synthesis.

However, the pressure-generator with the advantages of continuous operation, as distinguished from the intermittent production of water-gas sets and also the facts that it operates satisfactorily on less expensive fuels and that it produces a gaseous product having a higher H_2/CO_2 ratio than does the usual water-gas generator, can be employed for the manufacture of Synthesis Gas.

As hereinbefore mentioned, the actual pressure existing in the pressure generator influences the nature of the produced gas. At lower pressures, less methane is formed. In the presence of carbon, pressure displaces the CO/CO_2 equilibrium in the favor of carbon dioxide. If also the gasification process is carried out at a relatively low temperature, the equilibrium is further displaced in favor of a reduced formation of carbon monoxide. The pressure-gasification thus enables the formation of carbon monoxide to be restricted from two sides; that is, through the temperature and pressure of gasification. Since the required reduction of carbon monoxide formation is not very great, it is adequate to reduce the pressure from the usual 20 atm. to about 5 to 10 atm./sq.in.

Practical tests have shown that a carbon monoxide-hydrogen gas mixture suitable for the Fischer-Tropsch synthesis with cobalt-thoria catalyst can be prepared by the gasification of low-temperature bituminous coke (10-20 mm grain size) when employing a gasification pressure of about 8.5 atm. and both steam and oxygen as the gasification media. The formed crude gas has the following composition:

CO_2	-	29.3%
CO	-	21.9%
H_2	-	44.0%
CH_4	-	3.3%
N_2	-	1.5%

After removal of the carbon dioxide in the water-scrubbing step under pressure, the crude gas yields a Synthesis Gas having the following composition:-

CO_2	-	1.0%
CO	-	30.7%
H_2	-	61.6%
CH_4	-	4.6%
N_2	-	2.1%

It should be further recorded that by increasing the addition of steam in the pressure-gasification, the formation of carbon monoxide may be so extensively reduced that a water-scrubbed gas containing 70-75% of hydrogen and only 15% of carbon monoxide is produced. This gas is well-suited for use in the synthesis of ammonia and hydrogenation.

The pressure under which the Synthesis Gas is produced has the advantage also that the hydrogen sulphide and some organic sulphur is removed at the same time as carbon dioxide is separated by the water-scrubbing step. The purification of the Synthesis Gas of sulphur compounds is thus importantly cheapened.

The iron catalyst prepared by Lurgi for the preparation of hydrocarbons from Synthesis Gas requires in such gas a H_2/CO ratio differing from the requirement of the cobalt-thoria catalyst; the preferred synthesis pressure with the iron catalyst is 22 atm. and the H_2/CO ratio in the employed gas is 1.2 with recirculation over the catalyst. The equation which seems best to explain the reaction with the iron catalyst is as follows:-

$$2CO + H_2 \longrightarrow CH_2 + CO_2.$$

1. The high pressure of the Synthesis Gas required for the iron catalyst is easily supplied directly by the generator for the high-pressure gasification with oxygen. However, when the generator is normally operated at a pressure of over 20 atm. and a high temperature, as above discussed, the methane (inert) content of the produced gas and the H_2/CO-ratio are too high. The CO content of the produced gas can be increased by employing higher temperatures or lower pressures, but the possible increase in temperature is limited by the fusion-point of the ash in the employed fuel and any sharp decrease in pressure would require provision of means to compress the produced gas up to the above-requires pressure of synthesis. This was the problem presented Lurgi in connection with the design of the SICS-plant project at Valdarno, Italy. The fusion-point of the ash of the employed coal was relatively low. Analyses of the various possibilities available to the technology for its solution, such as:-

1. Pressure Gasification with Cracking of Residue Gas (Bamag);

2. Pressure Gasification with Cracking of the Crude Gas (without Regenerators);

3. Atmospheric Gasification with addition of CO_2;

4. Atmospheric Gasification with addition of CO_2;

5. Pressure Gasification with Cracking of Crude Gas (with Regenerators);

6. Pressure Gasification with Cracking of Residue Gas (Koppers);

7. Pressure Gasification with CO_2 addition; and

8. Winkler Gasification with Prior Low-Temperature Distillation;

were made by Lurgi and it came to the conclusion that the economically most advantageous method for preparing the Synthesis Gas from the lignitic brown coal was to employ oxygen at a pressure of about 23.5 atm. and, after washing the produced gas with water to remove about one-third of the CO_2 and some of the H_2S followed by oxide treatment to remove the remainder of the latter, the resultant gas is heated to about 145° C. and saturated with water. Thereafter, the gas is preheated to 500°C. and by partial combustion with oxygen, without regenerative heating, is heated to 1300°C. in a methane-converter in the absence of a catalyst, thereby to convert methane content thereof to CO and H_2. By heat exchange, the outlet gases from the methane converter are reduced to 350°C. at which temperature, and under pressure, they are scrubbed with water and cooled to about 30°C.; the hot scrubbing water is circulated to the above 145°C water-saturator step where some of its heat is employed to preheat and saturate gas that enters the 500°C. preheater for the methane converter. After another water-scrubbing step, for the substantial removal of CO_2, the formed crude Synthesis Gas has then the following composition:-

CO_2	-	2.0%
C_nH_m	-	0.2
CO	-	42.2
H_2	-	50.7
CH_4	-	2.4
N_2	-	2.5

The Synthesis Gas is then preheated to 250°C. and passed to the apparatus for removal of organic sulphur whereupon it is ready for delivery to the synthesis.

In this scheme of Synthesis-Gas production, there is con-sumed per m^3 thereof, 0.317 m^3 of oxygen. It was stated to be advantag-eous that the high-temperature methane cracking reduced the amount of gums in a synthesis gas; it was considered important by Lurgi to make provision to remove these compounds from a synthesis gas by, for example, active carbon to protect the catalyst.

Dr.Danulat in written statements, has emphasized the import-ance of the slagging generator for solving the above technical problem; its use would provide both the cheapest plant and lowest operating costs, but he indicated in December 1942 that such equipment was only in the development stage and could not be recommended for the Fischer-Tropsch plant at V aldarno, Italy. He also stated that the above pressure-generator operation, coupled with thermal cracking without regenerative heating, is even more favorable than the Winkler gasifi-cation process employed for the same technical problem, and emphasized that such operation had for Lurgi the advantage that it could make the company a competitor of the Winkler process in the field of ammonia synthesis and hydrogenation. Dr. Danulat emphasized that the develop-ment of a slagging pressure-generator was a necessary development for Lurgi to undertake immediately. During one of the interrogations, he stated that such a generator would be built along the lines of the blast furnace.

The accompanying Fig.8 shows diagrammatically a flow-sheet and apparatus as recommended by Lurgi for preparing Synthesis Gas having a H_2/CO ratio of 1.2 as is employed in combination with the Fischer-Tropsch iron catalyst.

Oxygen Production

The pressure-gasification plants erected at Hirschfelde, Boehlen and Brüx are equipped with air-separation plants of the standard Linde type erected by Ges.fuer Linde's Eismaschinen, A.G. of München. Lurgi have not carried out any research on problems connected with the separation of air into components and could supply no detailed informa-tion concerning recent developments in the use of expansion turbines.

The Messer Co. of Frankfurt-am-Main have erected a number of air-separation plants of the Linde type using normal heat exchangers, but were believed to have carried out some work on the design of expansion turbines. The contract for the air separation plant of