The Pressure Gasification of Solid Fuels with Oxygen

By Dr. Ing. Friedrich Danulat, Frankfort-on-Main

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Introduction.

Gas is the thermal energy-carrying medium in demand as fuel for industrial, commercial and domestic purposes. The original process of gas making was that of de-gasifying coal by heating, with exclusion of air, as a result of which, in addition to a high-grade gas (maximum calorific value 4000 to 5500 kilogram calories per cubic metre N. T. P.), coke was obtained. The endeavour to convert solid fuel completely into gas led to coal gasification processes. By gasification with air it became possible to produce a nitrogenous gas of low calorific value (maximum 1000—1500 kilogram calories per cubic metre N. T. P.). The water gas process, in which the fuel is gasified alternately with air and steam, enabled atmospheric nitrogen to be excluded from the gas produced and thus furnished a gas of a higher calorific value (maximum 2500—3000 kilogram calories per cubic metre N. T. P.). The addition of water gas, produced from coke, to the pure gas obtained from coal rendered it possible to increase the yield of gas with only slight depreciation of the calorific value, and thus to bring the output of gas and of coke more into proportion with the markets existing for the two products. In view of the high calorific value required, the manufacture of town gas remained dependent on the process of degasifying high-grade coal and on the supplementary production of water gas from coke, while the application of the air gasification process was restricted to the internal requirements of industrial plants, where use in the producer's own works rendered a low calorific value admissible. A process for the complete conversion of the solid fuel into a gaseous fuel of high calorific power, of the nature of town gas, thus remained a goal to be achieved by the gas making industry.

The large-scale industrial utilisation of chemical synthesis processes imposed on gas manufacturers, for the first time, the requirement of producing gases of a certain composition suitable for the synthesis. Thus, gases high in hydrogen are required for the synthesis of ammonia and liquid motor spirits by the I.-G.-Bergius or Fischer-Tropsch process. For the production of these gases the water gas process at first appeared suitable, despite the disadvantages of intermittent working and the use of high-grade fuels.

When Linde[1]) was developing the process for resolving air into its constituents, oxygen and nitrogen, at the turn of the century, he proposed the use of the oxygen produced in this manner for the gasification of coal in order to replace the water gas process by a continuously working and therefore more perfect process. For many reasons it was not possible at that time for this suggestion to be acted on in practice. Drawe[2]), his vision widened by subsequent technical progress, was the first to recognise afresh the possibilities of the use of oxygen as a gasifying medium, and in 1927, by means of a plant of semi-industrial size, he supplied proof that a gas free from nitrogen, with simultaneous complete gasification of the fuel, could be produced in the manner suggested by him. Following on this preliminary work, the Lurgi Gesellschaft für Wärmetechnik[3]) continued to develop the process. In 1930 they succeeded in evolving the

[1]) Gesellschaft für Linde's Eismaschinen, Wiesbaden, German patent No. 108 158 of the 21st May 1898.

[2]) Drawe: "Hochwertiges Gas und flüssige Brennstoffe als Endziel der Kohleveredelung" (High-grade Gas and Liquid Fuels as the Final Aim of Coal Refining), Gas- und Wasserfach 69 (1926), P. 1013—1015.

Drawe: "Neue Wege der Schwelung und Vergasung" (New Methods of Low Temperature Carbonisation and Gasification), Gas- und Wasserfach 70 (1927), P. 904, and Braunkohle 26 (1927), P. 573.

[3]) Drawe: "Starkgas durch Brennstoffvergasung mit Sauerstoff" (Rich Gas by Fuel Gasification with Oxygen), Gas- und Wasserfach 76 (1933), P. 541—545.

Hubmann: "Production of Gas rich in Hydrogen for Municipal Supply and (chemical) Synthesis", Metallgesellschaft's Periodic Review No. 8 (1934).

process of gasifying fuel with oxygen under high pressure and, after exhaustive preparatory experiments, in showing by means of a plant working on an industrial scale, that it was possible to produce directly a gas suitable for town supply. Further promising work dealt with the production of synthesis gas (i. e. gas suitable for synthetic processes) by the same process.

about 20 kg. per sq. cm. The gas producer is a pressure vessel lined with brick and furnished with a water-cooled jacket. To it are connected the lock chambers for filling in the coal and for removing the ash. As is also the case with normal gas producers, the fuel bed lies on a rotary grate, the rotational speed of which determines the amount of ash discharged. It is possible to build the lock

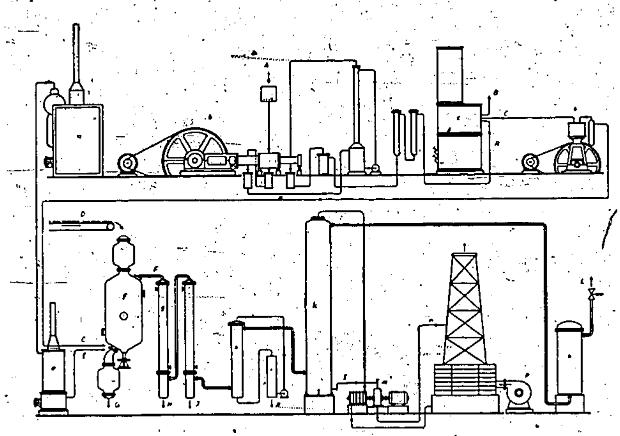


Fig. 1: Production of gas high in hydrogen for town supply and synthesis by the Lurgi Pressure Gasification Process.

A Air	*G Ash*	*a Steam boiler*	*i Stripping apparatus*
B Nitrogen	*H Tar*	*b Air compressor*	*k Pressure water wash*
C Oxygen	*I Middle oil*	*c Separator*	*l Pump*
D Fuel	*K Benzine*	*d Oxygen compressor*	*m Turbine*
E Steam	*L Pure gas*	*e Steam superheater*	*n Aeration tower*
F Crude gas		*f Gas poducer*	*o Extraction of sulphur residue*
		g Cooler	*p Air blower*
		h Benzine scrubber	

The Process.

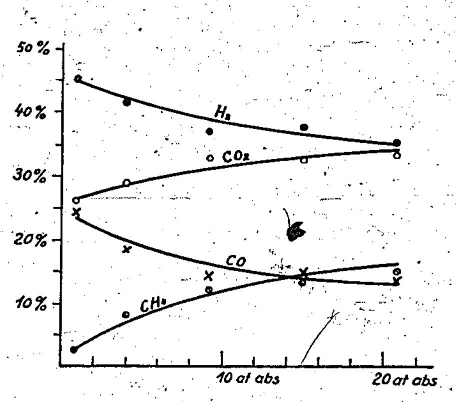
The mode of operation of the process is illustrated diagrammatically in Fig. 1. The atmospheric air is resolved into its constituents, oxygen and nitrogen, and the oxygen is thereupon compressed to about 20 kg. per sq. cm. gauge pressure. In addition to this, steam is produced at the same pressure and superheated to 500° C. Oxygen and superheated steam are mixed and introduced as gasifying medium into the gas producer, in which the gasification is carried out at a gauge pressure of

chambers in such a manner that the coal can be introduced into the gas producer in the usual way, despite the high pressure, so that its gasification may take place continuously. From the gas producer the crude gas flows to the condensing plant, where the tars and oils contained in the gas, and also the steam, are condensed in several stages. The gas leaves the coolers at a temperature of 20—30° C. and is then freed from gas benzine (light naphtha) in a scrubbing tower by means of wash oil. The dissolved benzine is expelled from the wash oil by distillation at atmospheric pressure; a high pressure pump then returns the generated wash oil to the scrubbing tower. The cooling of the gas and the separation of the condensable substances therefrom are followed by scrubbing with water under pressure, which removes the carbon dioxide and most of the hydrogen sulphide from the gas. The water is recirculated, being first delivered by a high pressure pump to the scrubbing tower; on contact with the gas it absorbs from the latter the carbon dioxide and hydrogen sulphide, is then expanded to atmospheric pressure in a turbine, regenerated in a tower by aeration and returned by a high pressure pump to the scrubbing tower. The gas purification ends with dry cleaning with bog iron ore, which takes the remaining traces of hydrogen sulphide out of the gas. The pure gas is passed on, under the pressure of gasification, for further use. The gas containing carbon dioxide resulting from the scrubbing with water under pressure is mixed with the expansion gas from the coal lock chamber and serves to superheat the steam for the gasification.

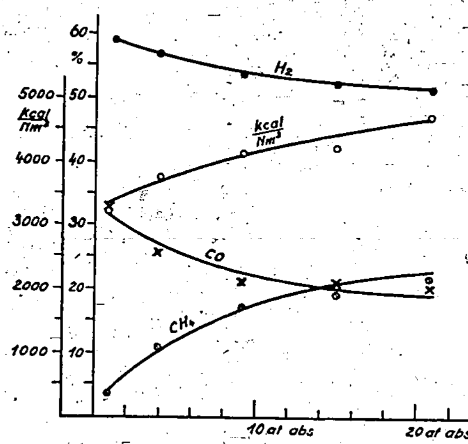
As compared with earlier gas production processes, the high pressure gasification process has the following particular features in respect of the actual gasification operation.

In gasifying a solid fuel with a mixture of oxygen and steam, the following reaction products are obtained: carbon monoxide, hydrogen, carbon dioxide and methane, while a portion of the steam remains undecomposed. The relative proportions of the reaction products are substantially determined by the pressure, temperature and time of reaction. Apart from the absence of nitrogen from the gas, the use of pure oxygen renders the composition of the gas capable of regulation within a wide range by means of the gasification pressure, the amount of steam in the gasification medium, and the charging of the shaft. These possibilities of modifying the method of operating the process allow the production of both gases of high calorific power for town supply purposes and gases of definite composition for synthesis purposes. The increased gasification pressure, as compared with gasification at atmospheric pressure in general, displaces the gas equilibria in such a manner that gases of multi-atomic composition are preferentially formed.

In the production of town gas the gasification process is carried out, under high pressure and with an adequate proportion of steam in the gasification medium, in such a manner that methane and carbon dioxide are preferentially produced in addition to carbon monoxide and hydrogen (see Fig. 2). The heat liberated in the exothermal formation of methane and carbon dioxide results in a reduction of the amount of oxygen required for the gasifica-



Composition of the crude gas plotted against the gasification pressure.



Composition and heat of combustion of the pure gas plotted against the gasification pressure.

Fig. 2: Influence of the gasification pressure on the formation of gas.

30824

tion, so that the oxygen required amounts to only about 60% of that needed in the case of gasification at atmospheric pressure.

The process has for the first time proved the possibility of methane synthesis in the gasification process and has rendered this synthesis itself practicable. The latter raises the calorific value of the gas to such an extent that, after removing the carbon dioxide, a gas equivalent in quality to town gas is obtained. The long-sought process of pure gas production and complete gasification of solid fuel into high-grade gaseous fuel is thus a reality.

A modified method of operating the gasification process renders it possible moreover to produce gases suitable for synthesis. If in a gas producer, working at a moderate pressure and with a forced output of gas, the amount of steam added is increased beyond a certain limit, the temperatures of the gasification will fall to such an extent that the formation of carbon monoxide is reduced, while at the same time the formation of methane sharply declines on account of the lack of reactivity. In this process the proportion of carbon monoxide to hydrogen in the gas may be varied within the range of from 1:2 to 1:4.5. A process for making synthesis gas is thus created, which is in many respects superior to the ordinary water gas process.

Town Gas Manufacture.

In 1936 the first industrial plant was constructed to the order of the A.-G. Sächsische Werke. In this plant brown coal is completely gasified into a town gas of normal quality, which is conveyed by a long distance pipe-line, under the production pressure, to the town of Zittau (40,000 inhabitants) and there used for the gas supply in place of the mineral coal gas formerly consumed. The plant, the annual output of which amounts to 3.5 million cubic metres of town gas, is shown in Fig. 3.

It consists of two gas producers, each of a shaft diameter of 1.2 metres. The gasification is carried out at a gauge pressure of 20 kg./sq. cm., and the gas is purified at the same pressure. The oxygen employed for the gasification, which is supplied by the air-resolving plant of a calcium cyanamide factory, is drawn in by compressors in the gas plant and compressed to 20 kg./sq. cm. gauge pressure. Before admission into the gas producer it is mixed

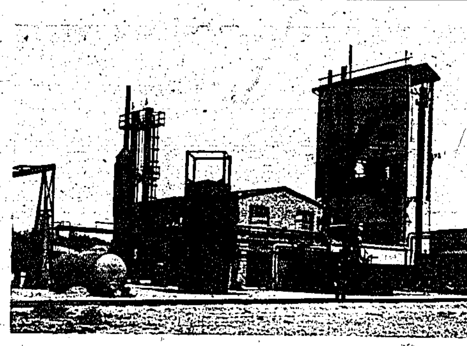


Fig. 3: Gas plant of the A.-G. Sächsische Werke — Regional gas supply.

with superheated steam. For the mean daily output of about 10,000 cubic metres it is sufficient to operate one gas producer which runs at about two-thirds load. The condensing plant attached corresponds in construction to the diagrammatic representation of the process given on a preceding page (Fig. 1). The gas producers are erected in a special building, whilst the compressor and pump house is separate from this building. The towers for scrubbing the gas with water under pressure and the containers of the dry purifiers are in the open. In consequence of the high pressure at which the gasification process and the succeeding purification are carried out, the plant is of very small dimensions in comparison with those of ordinary gas producing plants. Four men are sufficient to run it. Figs. 4 and 5 show individual parts of the plant.

The results obtained at the acceptance trials with the plant are reproduced in the following tables:

Result of the Performance Test:

A. General.

I. Coal (Screened Dry Brown Coal).

1. *Low Temperature Distillation Analysis.*

	in %
Tar	10.2
Water of distillation	7.4
Moisture	27.4
Coke	44.9
Gas + residue	10.1

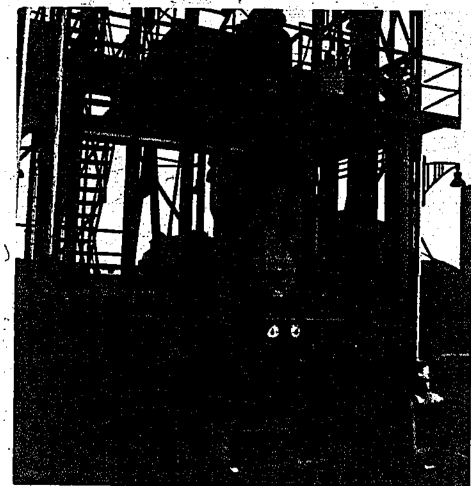


Fig. 4: Pressure gas producer.

2. *General Composition:*

	in %
Combustible substance	67.5
Ash	5.1
Water	27.4

3. *Elementary Analysis:*

	in %
Carbon	50.30
Hydrogen	3.45
Sulphur (combustible)	0.70
Oxygen + nitrogen	14.74
Ash	5.21
Water	25.60

4. *Screen Analysis:*

	in %
Over 5 mm	39.2
2 —5 mm	56.8
1 —2 mm	2.5
0.5—1 mm	0.7
Below 0.5 mm	0.8

5. *Calorific Value:*

Gross calorific value 4,730 kg.-cal./kg.
Net calorific value 4,390 kg.-cal./kg.

6. *Amount of coal gasified:*

21,500 kgs. per 24 hours.

7. *Shaft performance:*

746 kgs. per square metre per hour.

II. Crude Gas.

1. *Analysis:*

	in %
$CO_2 + H_2S$	30.6
C_nH_m	0.6
O_2	0.1
CO	16.5
H_2	34.0
CH_4	16.3
N_2 + remainder	1.9

2. *Hydrogen sulphide content:*

0.45 % by volume

III. Town Gas.

1. *Analysis:*

	in %
CO_2	3.0
C_nH_m	0.5
O_2	0.1
CO	22.8
H_2	48.7
CH_4	22.6
N_2 + remainder	2.3

2. *Gross calorific value of gas:*

4,280 kg.-cal./cub. m. N. T. P.
(i. e. at 0° C. and 760 mm. Hg)

3. *Specific gravity (referred to air = 1):* 0.448

4. *Hydrogen sulphide:*

0.0 gram per 100 cubic metres N. T. P.

5. *Ammonia:*

0.28 gram per 100 cubic metres N. T. P.

6. *Amount of gas produced:*

16,020 cubic metres N. T. P. per 24 hours.

B. Performance Figures.

I. *Gas yield (reduced to 4,200 kilogram calories per cubic metre N. T. P.)*

1. *Gas yield per metric ton of dry coal:*

760 cubic metres N. T. P.

2. *Gas yield per metric ton of pure carbon:*

1,128 cubic metres N. T. P.

II. *Oxygen consumption:*

0.15 cubic metre N. T. P.
per cubic metre N. T. P.

III. *Steam consumption:*

1.01 kg. per cubic metre N. T. P.

IV. *Yield of tar:* . . . . 72 %

C. Special measurements.

1. *Hydrogen sulphide content after scrubbing with water under pressure and before sulphur residue cleaning:*

   0.15 grams per 100 cubic metres N. T. P.

2. *Nitric oxide in town gas:*

   0.052 cubic centimetre per cubic metre N.T.P.

3. *Total sulphur in town gas:* (principally organic sulphur)

   1.0 gram per 100 cubic metres N. T. P.

The heat balance compiled with the aid of the foregoing figures shows the following distribution:

Heat Balance of the Process.

A. Heat brought in:

	in %
1. In the coal	89.9
2. In the saturated steam	10.1
	100.0

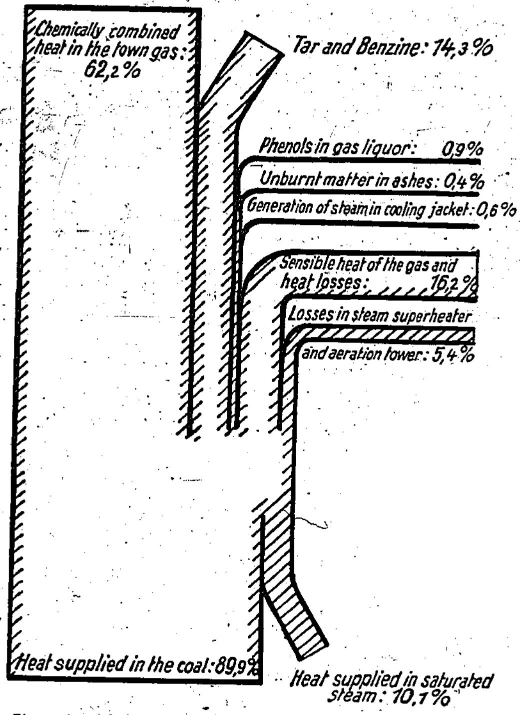


Fig. 6: Heat balance of the Lurgi pressure gasification process.

Fig. 5: Ash lock gate of the pressure gas producer.

B. Heat taken out:

	in %
1. Chemically combined heat in the town gas	62.2
2. In tar + benzine	14.3
3. Phenols in gas liquor	0.9
4. Unburnt matter in ashes	0.4
5. Generation of steam in cooling jacket	0.6
6. Sensible heat of gas and heat losses .	16.2
7. Losses in the steam superheater and aeration tower	5.4
	100.0

The efficiency of fuel conversion is as follows:

$$\frac{B_1 + B_2}{A_1} = 85.2\ \%$$

The advantages of the new process over previous methods of gas production are apparent from the foregoing description of the process and the results of the performance test with the plant.

Any non-caking fuel may be converted, with complete gasification and a high degree of efficiency, into a gas of high calorific power, the composition of which complies with current standards for town gas. As the increase in calorific value is obtained by the formation of methane in the gasification process, which in turn can be widely varied by the gasification in pressure, the process is not restricted to the use of brown coal but may be applied to other fuels also. This is proved by further tests. For the same reasons it is possible to dispense with tar cracking for the purpose of improving the calorific value, as is necessary in other brown coal processes. In consequence of the pressure it is possible to gasify fine-grained coal with a high throughput rate amounting to a multiple of that hitherto found with atmospheric pressure gasification. By adding a high proportion of steam to the gasification medium, a low gasification temperature, eliminating all danger of slagging, is obtained. At the same time this addition of steam influences the gasification in the direction of the desired reactions, so that a crude gas high in hydrogen, methane, and carbon dioxide is produced, the carbon monoxide content of which is adequately lowered. As the removal of carbon dioxide takes place under the pressure of gasification, the cost of this purification is low; so that the formation of carbon dioxide, necessarily involved in the production of a gas of the desired composition, does not affect the overall efficiency of the process. The advantage of pressure also becomes manifest both in the cooling of the gas, in the form of a better heat exchange, and in the extraction of benzine, through facilitated absorption of the latter by wash oil. As the gasification process is continuous, the composition of the gas and also its calorific value are hardly subject to fluctuations. It is astonishing that, despite the pressure, in normal working — depending on the load — tar yields of 70 to 80% were attained. The removal of the large quantities of carbon dioxide from the crude gas entails the circulation of relatively large quantities of water, which affords the advantage that the hydrogen sulphide is at the same time removed from the gas to a great extent. In view of the nature of the coal gasified, dry cleaning would hardly have been necessary, since even after the scrubbing with water under pressure the hydrogen sulphide content was below the admissible limits, so that after the pure gas issued from the final purification its content of hydrogen sulphide could not be determined qualitatively by the usual methods. The amount of nitric oxide and organic sulphur contained in the pure gas was unexpectedly low; this should be particularly noted in view of the diminished corrosive action of the products of combustion of the gas on gas appliances. With the first plant in operation it was proved that the process works dependably, and that a town gas can be produced by this process that is equal to that previously used and obtained by coking mineral coal.

The process has particularly good prospects of future development in regional gas supplies, since its advantages become especially apparent in plants working on a larger scale. The use of the process on a brown coal basis enables the large scale supply of gas to Germany to be rendered independent of the mineral coal previously exclusively used for that purpose, and to ensure the supply of gas in the central parts of the country without having recourse to the mineral coal districts situated on the frontiers of the country. In this connection it is of particular importance that the restrictions imposed on the sale of gas by the production of coke, necessarily involved in ordinary gas manufacture, are eliminated, since the process permits the complete conversion of the solid fuel into the gaseous form. The liquid fuels, tar, oil and benzine, obtainable as by-products, afford a welcome addition to national motor spirit production.

The Production of Synthesis Gas.

It has been stated in the opening remarks that the requirements of chemical synthesis, in respect of the nature of the gas are fundamentally different from those of town gas manufacture. The hydrogenation of solid and liquid fuels by the I.-G.-Bergius process for the purposes of synthesising motor spirit and lubricating oil, and also the synthesis of ammonia, require a high per cent hydrogen gas. On the other hand, the Fischer-Tropsch synthesis needs a carbon monoxide-hydrogen mixture containing these gases in the very definite proportion of $CO:H_2 = 1:2$. The Lurgi pressure gasification process now enables synthesis gas for these syntheses to be produced directly.

The usual process for the production of gases high in hydrogen starts with the well known water gas process, which, in addition to the disadvantage of intermittent working and unavoidable use of high-grade fuel, has the further drawback that it yields a gas containing too high a proportion of carbon monoxide. This gas, in which the ratio $CO:H_2$ amounts to almost 1:1, must be converted with the addition of steam; the carbon monoxide and the steam then react to form hydrogen and carbon dioxide, the latter being removed. By regulating the amount of carbon monoxide reacting it is possible to obtain a synthesis gas of the desired composition.

For the production of synthesis gas, the pressure gas process is assisted by its ability to influence the nature of the gas depending on the pressure. The ratio of the constituents CO, H_2, CO_2 and H_2O forming the crude synthesis gas is preferentially determined by the conditions of equilibrium of the water gas reaction. It is known that this reaction is not directly dependent on the pressure. This applies as long as the reaction is carried out in a purely gaseous phase. If, however, the formation of gas takes place in the gasification process itself, it is influenced, by reason of the presence of carbon, in such a manner that the pressure displaces the CO/CO_2 equilibrium in favour of carbon dioxide. If, in addition, the gasification process is carried out at a very low temperature, by adding a large proportion of steam to the gasification medium, the gas equilibrium is still further displaced in favour of a reduced formation of carbon monoxide. The pressure gasification thus enables the formation of carbon monoxide to be restricted from two sides — through the gasification temperature and pressure. As the required reduction of carbon monoxide formation is not very great, it is sufficient to apply relatively low pressures of about 5 to 10 kg./sq. cm. for the gasification.

Practical tests for the production of a carbon monoxide-hydrogen gas-mixture suitable for the Fischer synthesis showed that the gasification of low temperature mineral coal coke (10—20 mm. grain size) under a positive gasification pressure of 8.5 kg./sq. cm., with a gasification medium consisting of oxygen and steam, yields a crude gas of the following composition:

Composition of the Crude Gas:

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 succeeding scrubbing with water under pressure, the crude gas yields a pure gas of the following composition:

Composition of the Pure Gas:

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

In this gas carbon monoxide and hydrogen are in the proportion of 1:2, desired for the synthesis. The sum of CO + H_2 amounts to about 92 % of the gas mixture. According to earlier views the methane is certainly a disturbing factor but investigations into its influence in the benzine synthesis have shown that the presence of methane does not impair the reaction.

Proceeding still further, the formation of carbon monoxide may be reduced so extensively, by an increased addition of steam, that a pure gas containing 70—75 % of hydrogen and only 15 % of carbon monoxide is produced. This gas is excellently suited for the synthesis of ammonia and for hydrogenation.

In the production of synthesis gas the advantages of pressure make themselves felt in the purification and cooling of the gas, in the same manner as in the production of town gas. In the scrubbing with water under pressure, in particular the hydrogen sulphide is removed together with the carbon dioxide, to such an extent as to attain the purity of town gas. Tests also showed that the removal of organic sulphur, to attain synthesis purity, may also be substantially simplified by the employment of pressure.

To sum up, the following may be regarded as the particular advantages of the Lurgi pressure gasification process for the production of synthesis gas:

The process permits the direct production of a gas suitable for the synthesis of ammonia or the synthesis of motor spirits by the I.-G.-Bergius or Fischer-Tropsch processes. The fuel is gasified, in this process, with a high shaft performance (600 to 800 kgs. of fuel per square metre per hour) and, unlike the water gas process, is continuous. Cheap fine-grained fuels are preferably directly utilisable *for the gasification. The scrubbing with water* under pressure necessarily involved by the process effects a thorough purifying of the gas from hydrogen sulphide; at the same time the pressure enables relatively simple means to be employed for the separation of organic sulphur.

Summary.

The process developed by the Lurgi Gesellschaft für Wärmetechnik for the complete gasification of solid fuels under pressure, with oxygen, is suitable for the direct production of gases for town supply and for synthesis purposes. In 1936 the first industrial plant was erected for the production of standard quality town gas from brown coal, the excellent results in the operation of which indicate promising development of the process. Further work deals with the production of synthesis gas, for which the new process promises particular advantages over previous methods of gas production.