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AMENDED SPECIFICATION

Reprinted as amended in accordance with the decision of the Assistant-Comptroller, acting for Comptroller-General, dated the first day of October, 1937, under Section 21, of the Patents and Designs Acts, 1907 to 1932.

(The Amendments are shown in erased and italic type.)

489

PATENT SPECIFICATION

Convention Date (Germany): July 22, 1935. **461,402***

Application Date (in United Kingdom): April 21, 1936. No. 11378/36.

Accepted: Feb. 16, 1937.

COMPLETE SPECIFICATION (AMENDED)

Process for the Production of a Gas of Definite Stoichiometric Composition by the Carbonisation of Fuels

We, DIDIER-WERKE AKTIENGESELLSCHAFT, a Company organised according to the Laws of Germany, of Westfälische Strasse 90, Berlin-Wilmersdorf, Germany, do hereby declare the nature of this invention, and in what manner the same is to be performed, to be particularly described and ascertained in and by the following statement:—

The present invention relates to a process and apparatus for the production of a gas of definite stoichiometric composition capable of direct utilisation for chemical synthesis, by the thermal treatment of fuels. In particular those fuels which cannot be coked, and which are known as the " younger " fuels, can be used as the initial materials, and can be converted into a valuable gaseous product in a single operation.

Hydrogen and carbon monoxide are the chief initial materials used for the synthetic production of liquid hydrocarbons, and the object is therefore to obtain an initial gas mixture as free as possible from undesirable substances, and containing the largest possible quantities of hydrogen and carbon monoxide. Such undesirable substances are in this case not only CO_2 and H_2O but also other CH compounds, such as CH_4, C_2H_4, C_nH_m, &c. Moreover, the initial gas for synthesis purposes should be free from injurious substances or should contain these only in extremely small quantities. First among these injurious substances are the sulphur compounds which originate from the thermal treatment of a fuel, i.e., H_2S and CS_2. A final requirement of the initial gas is that the components, hydrogen and carbon monoxide, should bear a definite and regulable stoichiometric relation to each other.

Gas produced by known carbonisation processes from fuels has already been used for chemical synthesis purposes, but it was necessary first to subject the gas so obtained to a transformation, e.g., CO conversion, and CH_4 decomposition, and to free it from injurious substances, e.g., H_2S. The present process makes possible the production of an initial gas suitable for chemical synthesis purposes which possesses on production the properties required of a synthesis gas. The process is based upon a thermal treatment of fuel, by external heating with the addition of steam to the fuel, whereby the fuel is converted into the desired gas in a unitary continuous operation, and a carbonisation residue obtained as coke which suffices to generate all the heat required by the whole process. The fuel used may be brown coal, lignite, flame coal &c.

According to the present invention, a process for the production of a gas of definite stoichiometric composition from fuels is characterised in this, that the fuel is first preheated by means of external heating, the products of the preheating are removed, and the fuel is then carbonised at high temperature in an externally-heated chamber, and the high temperature carbonisation gases are led, in admixture with steam, through incandescent coke resulting from the carbonisation of the fuel, to effect the substantial conversion of the gases into the desired mixture of

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(The Amendments are shown in erased and italic type.)

ERRATUM

AMENDED SPECIFICATION
No. 461,402*.

At the end of the Specification, *for* " E. GEORGE LEWIN & CO." *read* " E. GEORGE LEWIN."

THE PATENT OFFICE,
November 6th, 1937.

is to be performed, ~~to be particularly de~~scribed and ascertained in and by the following statement:—

The present invention relates to a process and apparatus for the production of a gas of definite stoichiometric composition capable of direct utilisation for chemical synthesis, by the thermal treatment of fuels. In particular those fuels which cannot be coked, and which are known as the " younger " fuels, can be used as the initial materials, and can be converted into a valuable gaseous product in a single operation.

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According ~~to the~~ present invention, a process for the production of a gas of definite stoichiometric composition from fuels is characterised in this, that the fuel is first preheated by means of external heating, the products of the preheating are removed, and the fuel is then carbonised at high temperature in an externally-heated chamber, and the high temperature carbonisation gases are led, in admixture with steam, through incandescent coke resulting from the carbonisation of the fuel, to effect the substantial conversion of the gases into the desired mixture of hydrogen and carbon monoxide.

The invention also consists in the provision of suitable apparatus for carrying out the process of the present invention, as hereafter described and claimed.

Further features of the invention will be apparent from the Specification and from the appended claims.

The fuel, advantageously in the form of small pieces, medium-sized grains or small briquettes, is fed into a pretreatment chamber (hereinafter called the "preheating" chamber) which is in direct connection with a gasification chamber. Passing through the former continuously, the fuel enters the gasification chamber, through which it also passes continuously. At the lower end of the gasification chamber, the residue of the fuel is removed continuously as coke. The preheating chamber is externally heated (preferably by the waste gases from the heating of the gasification chamber), the temperature increasing from top to bottom, so that in its passage through the preheating chamber, the fuel is gradually heated more strongly, to a temperature of approximately 400° C. The final temperature, however, depends upon the particular fuel used. The fuel which is being subjected to an increasing temperature in the preheating chamber yields first steam, and then steam and carbon dioxide. At the same time, mainly in the lower portion of the preheating chamber, hydrogen sulphide is formed from the steam and the organic sulphur contained in the fuel; tar vapours are also formed from the low-boiling hydrocarbons. These various gases are now drawn off from the preheating chamber, either collectively or separately. The outlet for the said gases is preferably arranged as far as possible at the lower end of the preheating chamber, so that all the gases evolved may be led out into one main. This is of advantage if the gases are not to be subjected to any special treatment for the recovery of by-products therefrom. If, however, sulphur or the tar is to be recovered from the gases from the preheating chamber, outlets are suitably arranged at different levels in the preheating chamber, whereby, in particular, a separation of the tar vapours from the remaining gases can be effected. The height of the preheating chamber is such that the coal, in its passage from the inlet to the outlet into the gasification chamber, is heated long enough and to such a degree that, more especially, all the sulphur present in the coal can be converted into gaseous sulphur compounds, which are then drawn off. None of that portion of the sulphur which causes the formation of hydrogen sulphide should remain in the fuel when the latter passes from the preheating chamber into the gasification chamber. By suitable regulation of the heating in the preheating chamber and arrangement of the gas offtakes therefrom, a fractional separation of the products formed in the preheating chamber can be effected. This is necessary when, for example, extensive recovery of distillation tar and sulphur is to be practised. Moreover, as the gases evacuated contain combustible gases, they can be used for heating the oven installation, if necessary, after recovery of the by-products. The whole of the gases from the preheating chamber can, without hesitation, be led into a gas producer. In the combustion of this gas, the injurious substances, which represent only a very small proportion of the total amount of combustion gas produced, can no longer have any detrimental effect.

The fuel heated in the preheating chamber and freed from sulphur compounds, water-vapour and carbon dioxide then enters the externally-heated gasification or treatment chamber proper, which includes several successive zones, as follows: a carbonisation zone, a reaction zone and a water-gas zone. In the carbonisation zone, or first zone immediately adjacent to the preheating chamber, the fuel is, as far as possible, at once raised to a high temperature. In this zone, the temperature of the fuel should not fall below 850° C. The fuel in the carbonisation zone is heated strongly, so that the carbonisation zone need only be comparatively short. Between the lowermost gas offtake from the preheating chamber and the carbonisation zone, there must be a sufficient quantity of fuel to prevent the passage of gases from the preheating chamber into the gasification chamber, and vice versa. The fuel in this intermediate portion thus forms a partition between the preheating zone and the carbonising zone. The carbonisation zone lies approximately in the upper third of the gasification chamber, and does not extend beyond half-way down the chamber. The heating of the gasification chamber commences immediately at the top of the chamber. The chamber is heated strongly in this region, and the degree of heating should not diminish to any large extent in the downward direction, so that throughout the carbonisation zone and also in the adjacent reaction and water-gas zones the temperature should not, as far as possible, fall below about 850° C. The gasification chamber should have a pronounced taper in the carbonisation zone, and should be suitably enlarged in the downward direction.

In the passage of the coal or fuel through the carbonisation zone, high temperature carbonisation products which consist principally of hydrocarbons, CO, H_2, CH_4 and high temperature tar vapours are evolved. The height of the carbonisation zone is so proportioned that the fuel on issuing therefrom is completely carbonised. The resulting carbonisation gases now pass downwards out of the carbonisation zone into the adjacent reaction zone and traverse this in the same direction as the moving fuel. Simultaneously, steam is introduced at the commencement of the reaction zone, which also traverses the reaction zone in the same direction as the carbonisation gases and in admixture with them. The reaction zone is filled with incandescent coke from the preceding carbonisation zone, and is likewise heated externally, in order to supply the heat necessary for the maintenance of the reaction temperature. While the carbonisation gases and the steam pass together through this zone, a gas conversion takes place in conjunction with the catalytic cooperation of the incandescent coke, and as a result the hydrocarbons contained in the carbonisation gases, especially the methane, are converted into carbon monoxide and hydrogen. In addition, the hydrocarbons resulting from cracking of the tar vapours, and consisting essentially of methane (as the most stable hydrocarbon compound) are decomposed, together with the primarily-formed methane referred to above. At the same time, water-gas is formed from the excess steam and the carbon of the incandescent coke, this water-gas consisting principally of carbon monoxide and hydrogen. The process in the reaction zone is so conducted that during the formation of the water-gas. *an excess of hydrogen is* ~~hydrogen is mainly~~ obtained,. ~~and the formation of carbon dioxide is avoided as far as possible—~~ This is achieved by the addition of a quantity of steam greater than that theoretically necessary for the formation of water-gas; preferably, a quantity of steam many times in excess of the amount theoretically necessary is mixed with the carbonisation gases at the beginning of the reaction zone.

In this connection, the proportioning of the height of the reaction zone as well as the flow velocity of the gas and of the steam are of importance. Both are so adjusted in relation to one another that the gas conversion proceeds in the direction of increased formation of hydrogen. This is possible because of the previous elimination, in the preheating chamber, of the carbon dioxide yielded by the fuel itself; the prior elimination of the carbon dioxide tends to influence the equilibrium of the reaction in the direction of the production of hydrogen. As no more carbon dioxide and no steam will be generated in the gasification chamber from the fuel itself, the gas conversion in the reaction zone can be regulated as a catalytic conversion, by suitable adjustment of the ratio between steam and carbonisation gases, since a regulable catalytic conversion is now made possible by the elimination of the uncontrollable amounts of carbon dioxide and steam originating from the fuel. For the admission of steam, valve controlled inlet pipes are provided at various levels. It is thus possible to adjust the height of the reaction zone.

In order to obtain the desired results, it is particularly important to provide a controlled and adjustable supply of heat to the various zones in the gasification chamber. This requirement can be fulfilled by a graduation of the heat supplied through the walls of the gasification chamber. With external heating similar in other respects, varying quantities of heat can then be supplied to the individual zones, and it is possible to adjust or arrange the successive zones suitably for different fuels, for the same height of the chamber. Thus, for example, if brown coal is the initial fuel, the preheating chamber will be of iron or other suitable heat-resisting metal, the carbonisation zone of silicon carbide material, the reaction zone of silica material, and the water-gas zone of fire-clay ("Chamotte") material, so that the thermal conductivity of the material of the chamber walls decreases from zone to zone, from the preheating zone, through the carbonising and reaction zones, to the water-gas zone. The transfer of heat in the preheating zone is very rapid, as also in the carbonisation zone, because silicon carbide has good thermal conductivity. The carbonisation zone can be quite short. Moreover, by the rapid carbonisation resulting from efficient heat transfer, products are formed whose subsequent transformation is easier than with the products resulting from a slower carbonisation. In the reaction zone, which must possess a suitable length in order that the gas traversing it is properly converted, a smaller rate of heat supply will be sufficient, and therefore a material of lower thermal conductivity than silicon carbide, e.g., silica material, may be used here. For the water-gas zone a fire-clay material will, in general, suffice. Other constructional materials with different thermal conductivities may, of course, also be used, but the carbonisation zone should be of a refractory material of good thermal conductivity.

The gas converted in the reaction zone is drawn off at the end of this zone. The residual fuel now passes from the reaction zone into a water-gas zone adjoining the former zone, in which water-gas is generated by the introduction of steam. ~~Here, too, by~~ *By* regulating the quantity of steam supplied and the height of the zone, the production of water-gas may be so conducted as to favour an increased formation of hydrogen and only small quantities of carbon dioxide. This is possible by continually maintaining a high reaction temperature in the zone, with a corresponding supply of the heat of reaction necessary for water-gas production, through the walls of the chamber. The water-gas is drawn off from the water-gas zone, and is then admixed with the final gas issuing from the reaction zone. In the example shown in the accompanying drawings, and hereinafter described in detail, the water-gas is drawn off from the upper end of the water-gas zone, together with the final gas from the lower end of the reaction zone. In doing this, however, care must be taken that the water-gas comes in contact with the final gas from the reaction zone only when the latter is fully converted. No water-gas should enter the actual reaction zone, where it would disturb the balance of the catalytic reaction, to the detriment of the reaction favouring the formation of carbon monoxide and hydrogen. If the reaction zone and the water-gas zone are formed as separate chambers, then, clearly, the water-gas may be taken off at either end of the water-gas chamber.

The gasification chamber, i.e., the carbonising zone, the reaction zone and the water-gas zone are advantageously maintained under a pressure exceeding atmospheric. The entry into these zones of combustion products from the heating flues, in particular nitrogen, is thereby avoided.

The residual coke from the water-gas zone is discharged and serves as a fuel for the production of the heating gas for the plant. Advantageously, the whole process is so conducted that the quantity of coke left over is just sufficient to cover the total heat requirements of the plant.

The final gas from the reaction zone and the water-gas from the water-gas zone together form the synthesis gas. A composition is thereby obtained, containing comparatively little carbon dioxide and in which the ratio of hydrogen to carbon monoxide is at least 2 : 1. It is thus possible to produce a gas in which the carbon monoxide and hydrogen together amount to from 85—90 per cent. of the total, while the ratio of hydrogen to carbon monoxide is not less than 2 to 1. By increasing the temperature, lengthening the reaction zone and correspondingly dimensioning the water-gas zone, a quantitative ratio of hydrogen and carbon monoxide exceeding 2 to 1 can be obtained.

The accompanying drawing illustrates, by way of example, an arrangement for carrying out the process of the present invention. In the drawing:—

Figure 1 shows a vertical section on the minor axis of a preheating chamber and a gasification chamber suitable for carrying out the process of the present invention, and

Figure 2 shows a vertical section through the major axis of the chambers.

The fuel is contained in the bunker 1 and is fed through the valve 2 into the preheating chamber 3. From the preheating chamber, the preheated fuel enters the gasification chamber 4, traverses the various zones of the latter, and after passing through the coke-cooling chamber 5, the residue is discharged by the discharge device 6. The preheating chamber 3 is heated externally by means of a heating wall system 7. Waste gases from the heating system of the gasification chamber enter the lower part of the heating system 7 through the inlet 9 and escape through the outlet 10, after traversing the heating flues provided on each side of the preheating chamber. An offtake 11 for the removal of the products of the preheating is provided in the lower part of the chamber 3, and additional gas offtakes 12, 13, 14 and 15 are also provided at various levels in the chamber. These various offtakes may be used simultaneously, or individually, or in groups, as required.

Combustion air for the heating flues 20 of the gasification chamber 4 is supplied through the duct 16, and heating gas through the duct 17; the air and the combustion gas enter the heating flues 20 through the ports 18 and 19 respectively. Hot waste products of combustion leave the heating flues through the waste gas flue 21 and are transferred through the distributing channels 21*a* to the recuperator 22, in which the combustion air and/or heating gas is/are preheated. After traversing the recuperator the waste gases are collected in the collecting space 23 and from thence pass through the duct 8 to the inlet 9 to the heating flues of the preheating chamber.

As shown in Figure 2, a vertical brickwork wall 24 extends part-way down and across the smaller dimension of the gasification chamber. The wall 24 contains ducts or pipes, 25 and 26, opening into the chamber at different levels, for supplying steam to the reaction zone in the chamber. In the construction shown, the uppermost of these steam inlets is about one-third way down the gasification chamber. Further steam supply openings 27 are provided near the lower end of the chamber, in order to supply steam to the water-gas zone, as already described.

The gas from the reaction zone and the water-gas from the water-gas zone are drawn off together through the lateral opening 28 into the gas offtake main 29. The upper edge of the gas offtake 28 is about two-thirds way down the gasification chamber. This gas is the initial synthesis gas required.

Having now particularly described and ascertained the nature of our said invention, and in what manner the same is to be performed, we declare that what we claim is:—

1. Process for the production of a gas of definite stoichiometric composition from fuels, characterised in this, that the fuel is first preheated by means of external heating, the products of preheating are removed, the fuel is then carbonised at high temperature in an externally-heated chamber, and the resultant carbonisation gases are led in admixture with steam through a body of the incandescent coke resulting from the carbonisation of the fuel, to effect the substantial conversion of the gases into the desired mixture of hydrogen and carbon monoxide.

2. Process for the production of a gas of definite stoichiometric composition from fuels, characterised in this, that the fuel is first preheated at a relatively low temperature by means of external heating to drive off undesirable substances, such as carbon dioxide, sulphur compounds and water vapour, which are removed, the fuel is then heated to a high temperature in an externally heated chamber, and the resultant high-temperature carbonisation gases, free or nearly free from the said undesirable substances, are led together with added steam through a body of the incandescent coke resulting from the carbonisation of the fuel, to effect the substantial conversion of the gases into the desired mixture of hydrogen and carbon monoxide.

3. Process as claimed in Claim 1 or 2, characteried in this, that the fuel is first preheated at a relatively low temperature to free it, as far as possible, from carbon dioxide, sulphur compounds, water vapour and distillation tar, the fuel is then subjected to a high temperature carbonisation in a carbonising zone, and the carbonisation gases resulting therefrom are passed into a reaction zone adjoining the carbonising zone and containing incandescent coke from the carbonising zone, and traverse the reaction zone in admixture with steam which is introduced into the reaction zone, to effect the substantial conversion of the gases into the desired mixture of hydrogen and carbon monoxide.

4. Process as claimed in Claim 1, 2 or 3, characterised in this, that the amount of steam admixed with the carbonisation gases is considerably in excess of the amount which is theoretically required.

5. Process as claimed in Claim 1, 2, 3 or 4, characterised in this, that the process is operated continuously, the fuel passing continuously through the preheating, carbonising and reaction zones, and the gases evolved in the carbonising and reaction zones flow in the same direction as the moving fuel.

6. Process as claimed in Claim 1, 2, 3, 4 or 5, characterised in this, that in the preheating stage the fuel is gradually heated to a temperature sufficient to eliminate the specified undesirable substances.

7. Process as claimed in Claim 1, 2, 3, 4, 5 or 6, characterised in this, that the gaseous products resulting from the preheating of the fuel, such as water-vapour, carbon dioxide and tar vapours, are drawn off either at various points or collectively.

8. Process as claimed in Claim 1, 2, 3, 4, 5, 6 or 7, characterised in this, that the gases evolved in the preheating of the fuel flow in the same direction as the moving fuel and are drawn off at the lower end of the preheating zone.

9. Process as claimed in Claim 1, 2, 3, 4, 5, 6, 7 or 8, characterised in this, that water-gas is admixed with the gas generated in the reaction zone.

10. Process as claimed in Claim 9, characterised in this that the water-gas is generated from the residual coke leaving the reaction zone by the introduction of steam and, if necessary, by supplying heat to the coke.

11. Apparatus when used for carrying out the process according to any one of the preceding claims, comprising an externally-heated vertical preheating chamber, provided with an offtake or offtakes for the gaseous products of preheating, arranged above an externally-heated vertical gasification chamber, the upper portion of the latter forming the carbonising zone and below which is the reaction zone, with a gas offtake from the lower end of the reaction zone.

12. Apparatus as claimed in Claim 11, characterised in this, that below the re-

action zone in the lower part of the gasification chamber is a water-gas zone, in which water-gas is generated by passing steam through the hot coke descending from the reaction zone.

13. Apparatus as claimed in Claim 11 or 12, characterised in this, that the walls of the preheating chamber or zone are of metal and the walls of the gasification chamber, in the carbonising zone, of silicon carbide or of some other refractory material of high thermal conductivity.

14. Apparatus as claimed in Claim 13, characterised in this, that the thermal conductivity of the material of the walls decreases from zone to zone, from the preheating zone through the carbonising zone and the reaction zone to the water-gas zone.

15. Apparatus when used for carrying out the process according to Claims 1 to 10 inclusive, characterised in this, that above a vertical high-temperature gasification chamber heated externally, either uniformly or decreasingly from top to bottom, is a preheating chamber, externally heated by the waste heating gases from the gasification chamber, the temperature in the preheating chamber decreasing upwardly to the inlet for the fuel, the preheating chamber having an offtake or offtakes for the removal of the gaseous products generated therein, whilst the upper portion of the gasification chamber is provided with steam supply ducts, for leading steam into the reaction zone, and further steam supply ducts are arranged in the lower portion of the chamber to lead steam into the water-gas zone situated below the reaction zone.

16. Apparatus as claimed in Claims 11 to 15 inclusive, characterised in this, that the lowest gas offtake provided in the preheating chamber is so far removed from the opening of the gasification chamber that the intermediate fuel layer prevents the products of preheating from passing into the high temperature zone, and conversely prevents the high-temperature gases from entering the preheating chamber.

17. Apparatus as claimed in Claims 11 to 16 inclusive, characterised in this, that the outlet for the final gas from the reaction zone and the outlet for the water-gas from the water-gas zone are so arranged that the two types of gases do not mix within the respective zones.

18. Apparatus as claimed in Claims 11 to 17 inclusive, characterised in this, that a pressure exceeding atmospheric is maintained in the gasification chamber.

19. Apparatus when used for the production of a gas of definite stoichiometric composition from fuels by the process claimed in any of the claims 1 to 10 inclusive, comprising an externally-heated vertical gasification chamber, above which is an externally-heated vertical preheating chamber heated by the waste heating gases from the gasification chamber, a gas offtake or offtakes from the preheating chamber, a lateral gas offtake arranged about two-thirds way down the gasification chamber, steam inlets about one-third way down the gasification chamber, and further steam inlets at the lower end of the chamber, a supply of fuel to the inlet of the preheating chamber and extractor gear for the fuel residue at the lower end of the gasification chamber.

20. Apparatus as claimed in Claim 19, characterised in this, that the heating means for the gasification chamber are arranged to heat the upper third (approximately) of the chamber strongly, whilst a coke-cooling chamber is provided between the lower end of the gasification chamber and the extractor gear.

21. Process and apparatus for the production of a gas of definite stoichiometric composition from fuels, substantially as described with reference to the accompanying drawings.

Dated this 21st day of April, 1936.
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Ebury and Allington Houses,
136—150, Victoria Street,
London, S.W.1,
Agent for the Applicants.

Leamington Spa: Printed for His Majesty's Stationery Office, by the Courier Press.—1937.

[This Drawing is a reproduction of the Original on a reduced scale.]

Fig. 1.

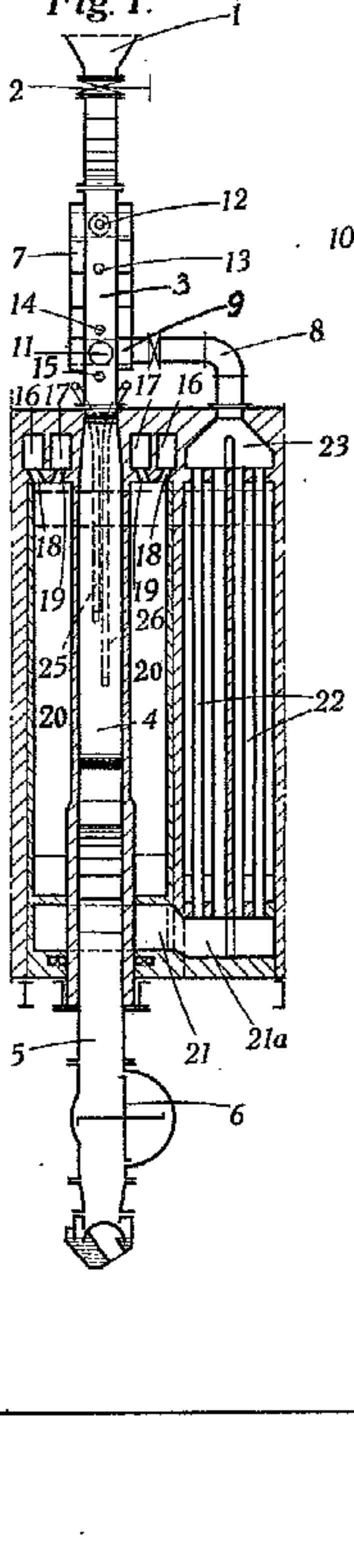

Fig. 2.