PATENT SPECIFICATION

636,206

Date of Application and filing Complete Specification: March 31, 1947.

No. 8718/47.

Complete Specification Published: April 26, 1950.

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Index at acceptance:—**Class 55(i),** B(4a: 11: 13: x).

COMPLETE SPECIFICATION

Process for the Production of Carbon Monoxide and Hydrogen

SPECIFICATION NO. 636,206

By a direction given under Section 17(1) of the Patents Act 1949 this application proceeded in the name of Standard Oil Development Company, a corporation duly organized and existing under the laws of the State of Delaware, United States of America, having an office at Linden, New Jersey, and 100, West 10th Street, Wilmington, Delaware, both in the United States of America.

THE PATENT OFFICE,
26th May, 1950

DS 39046/1(19)/3397 160 5/50 R

tained in and by the following statement:—

The present invention relates to the manufacture of gas mixtures containing carbon monoxide and hydrogen from non-volatile carbonaceous materials such as coal, coke, peat, oil shale, heavy oil residues, cellulosic materials, including lignin and the like, and more particularly to the manufacture of such gas mixtures as are suiable for the catalytic synthesis of hydrocarbons from carbon monoxide and hydrogen.

It has long been known that non-volatile carbonaceous materials such as coal, coke, and the like may be converted into water gas by a high temperature reaction with steam. The process yields mixtures of CO and H_2 in varying proportions, depending mainly on the conversion temperatures and the feed ration of steam. The flexibility of the process may be illustrated by a series of possible chemical reactions about as follows:

$$\begin{aligned} C+2H_2O &\longrightarrow 2H_2+CO_2 - 24{,}700 \text{ cal.}\\ C+H_2O &\longrightarrow H_2+CO - 32{,}500 \text{ cal.}\\ C+CO_2 &\longrightarrow 2CO \quad - 40{,}200 \text{ cal.} \end{aligned}$$

The overall water gas reaction being endothermic, heat must be supplied. This is usually accomplished by the combustion of a portion of the carbonaceous feed with an oxidizing gas, such as air and/or oxygen, at about 1600°—3000° production of gas mixtures of varying composition and BThU content. Furthermore, the process as such is extremely well suited not only for the production of fuel gases but also for the production of feed gases for hydrogenation processes and particularly for the catalytic synthesis of hydrocarbons and/or oxygenated organic compounds from CO and H_2. Depending on the products desired, the latter synthesis requires H_2:CO ratios varying within the wide limits of 0.5—5 volumes of H_2 per volume of CO.

However, the technical utilization of the water gas process, particularly for hydrogenation processes and the production of synthesis feed gas, has been appreciably impeded by difficulties encountered in heat supply and continuous operation as well as in the substantial removal of sulfur compounds from the gas, the latter being imperative for the utilization of the gas in the hydrocarbon synthesis.

The problem of supplying heat of reaction with continuity of operation has been materially eased by the application of the so-called fluid solids technique wherein the carbonaceous charge is reacted in the form of a dense turbulent mass of finely-divided solids fluidized by the gaseous reactants and products. In this procedure heat is supplied to the gas generator by the combustion of carbonaceous constituents of the charge with air and/or oxygen either in the gas

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COMPLETE SPECIFICATION

Process for the Production of Carbon Monoxide and Hydrogen

I, Conrad Arnold, a British subject, of 29, Southampton Buildings, Chancery Lane, London, W.C.2, do hereby declare the nature of this invention (a communication from Standard Oil Development Company, a corporation duly organised and existing under the laws of the State of Delaware, United States of America, having an office at Linden, New Jersey, United States of Amerca), 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 the manufacture of gas mixtures containing carbon monoxide and hydrogen from non-volatile carbonaceous materials such as coal, coke, peat, oil shale, heavy oil residues, cellulosic materials, including lignin and the like, and more particularly to the manufacture of such gas mixtures as are suiable for the catalytic synthesis of hydrocarbons from carbon monoxide and hydrogen.

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The overall water gas reaction being endothermic, heat must be supplied. This is usually accomplished by the combustion of a portion of the carbonaceous feed with an oxidizing gas, such as air and/or oxygen, at about 1600°—3000° F., in accordance with the following overall reactions:

$$C+O_2 \longrightarrow CO_2+94{,}400 \text{ cal.}$$
$$2C+O_2 \longrightarrow 2CO+54{,}200 \text{ cal.}$$

The combustion reaction may be carried out either simultaneously with the water gas reaction or alternately in a make and blow fashion.

It will be appreciated from the above that the water gas process permits the production of gas mixtures of varying composition and BThU content. Furthermore, the process as such is extremely well suited not only for the production of fuel gases but also for the production of feed gases for hydrogenation processes and particularly for the catalytic synthesis of hydrocarbons and/or oxygenated organic compounds from CO and H_2. Depending on the products desired, the latter synthesis requires H_2:CO ratios varying within the wide limits of 0.5—5 volumes of H_2 per volume of CO.

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The problem of supplying heat of reaction with continuity of operation has been materially eased by the application of the so-called fluid solids technique wherein the carbonaceous charge is reacted in the form of a dense turbulent mass of finely-divided solids fluidized by the gaseous reactants and products. In this procedure heat is supplied to the gas generator by the combustion of carbonaceous constituents of the charge with air and/or oxygen either in the gas

[*Price 2/-*]

generator itself or in a separate heater from which it is transferred to the gas generator as sensible heat of hot combustion residue.

The procedure involving combustion within the gas generator requires the use of highly expensive pure oxygen as the oxidizing gas in order to prevent dilution of the product gas with notrogen. When a separate heater is used, considerable loss of potential heat is incurred as a result of high carbon concentrations in the heater requiring greater quantities of air to provide the necessary heat for water gas generation at heater temperatures of about 1800°—2200° F. Moreover, the sensible heat of the combustion residue being the only heat source for the water gas reaction, the solids circulation rates from the heater to the gas generator are quite considerable, i.e., usually around 4 lbs. or 0.2 cu. ft. of solids per cu. ft. of water gas produced. In addition, it is necessary to operate the heater within a few pounds pressure, say 25, of the generator, thereby requiring compression of large amounts of air over a considerable pressure range if water gas of more than 25 lbs. pressure is to be produced. In any case, the water gas produced contains organic sulfur compounds which complicate considerably the gas desulfurization. Finally, the steam and cooling water requirements are relatively high. These difficulties result in a relatively high cost of water gas or synthesis gas from coal, averaging more than $.10 per 1,000 cu. ft. of gas produced as compared with about $.05 per 1,000 cu. ft. of synthesis gas produced by modern methane conversion processes.

The present invention overcomes the aforementioned difficulties and affords various additional advantages. These advantages, the nature of the invention and the manner in which it is carried out will be fully understood from the following description thereof read with reference to the accompanying drawing.

It is, therefore, the principal object of the invention to provide improved means for converting carbonaceous materials into gas mixtures suitable for the catalytic synthesis of hydrocarbons from carbon monoxide and hydrogen.

A more specific object of the invention is to provide improved means for supplying heat to a water gas generator using a carbonaceous charge in the form of a dense fluidized mass of finely divided solids.

Other and further objects will appear hereinafter.

In accordance with the present invention the gasification of solid carbonaceous materials with steam is carried out in the presence of metal oxides supplying the oxygen required for heat generation by partial combustion. Employing the fluid solids technique the fluidized finely divided metal oxide is intimately contacted with, and gives off its oxygen to, the fluidized solid carbonaceous charge of the gas generator. The resulting combustion may generate at least a substantial portion of the heat required by the water gas reaction. In addition, the metal oxide is reduced so that it may be reoxidized in an exothermic reaction to produce heat required by the gasification reaction.

A substantial separation of solid gasification residue from reduced metal oxide may be effected without appreciable heat losses and reduced metal oxide may be continuously or intermittently circulated to a separate oxidation zone to be regenerated by oxidation with air at temperatures within the range of about 1400° to 2500° F. Regenerated metal oxide may be returned to the gas generation zone substantially as the temperature of the oxidation zone thus supplying sensible heat of solids to the gas generation zone in addition to the heat generated by partial combustion within the gas generation zone.

In this manner, the volume of solids to be circulated through the system for the purpose of heat supply may be reduced, and air rather than pure oxygen may be used for heat generation. It should be noted that, as indicated above, in the system in which a separate heater is used and hot ashes from the heater supply the heat for the water gas reaction, the carbon concentration in the heater must be low in order to prevent excessive carbon monoxide formation therein. To accomplish this it has been necessary to operate the water gas generator likewise at a relatively low carbon concentration which results in a low rate of the water gas reaction. In accordance with the present invention the necessity of low carbon concentration in a heater is obviated and the water gas generator may be operated at highest carbon concentrations and rates of reaction. Moreover, the metal oxide may act simultaneously as a cracking catalyst for organic sulfur compounds, thus simplifying the desulfurization of the product gas. In addition, it may catalyze the water gas shift reaction, thereby allowing the production of larger proportions of H_2 by reaction of CO with steam, the supply of the latter being controlled, if necessary, to establish concentrations suitable for high H_2 content at equilibrium, toward which the catalytic effect is directed. On the other hand, any methane formed by cracking of the carbonaceous feed is more easily reformed in the presence of metals such as iron and nickel.

The metal oxides which may be used in the present process are quite generally such oxides as have such an affinity for oxygen at the temperatures of our process that their oxygen partial pressures at equilibrium with both higher and lower stages of oxidation present are less than about 0.10 atmospheres and preferably less than 0.01 atmospheres so that substantially all the oxygen of the air used for regeneration can be bound by the lower stage of oxidation. The metal oxides should also be capable of oxidizing carbonaceous constituents of the solid carbonaceous charge at the temperatures and pressures of the water gas reaction. While certain oxides which are reduced to metals such as ferrous oxide, cuprous oxide, and the like, are useful for our process, other suitable oxides are the higher oxides of metals which are capable of forming both higher and lower oxides. Typical of these are cupric oxide, ferric oxide and vanadium pentoxide. Suitable also are mixtures of these oxides and mixtures with carrier materials such as kieselguhr, alumina or silica gels, bentonites, and the like.

As indicated above, spent metal oxide may be concentrated in the solids withdrawn from the gas generation zone for regeneration. While this circulating material need not be free from ash introduced with the carbonaceous charge, it should be freed of significant concentrations of oxidizable material other than the spent metal oxide without incurring appreciable heat losses. For this purpose any suitable means such as flotation, magnetic separation or the like may be used. In accordance with a preferred modification of the invention the linear velocity of the gases in the gas generation zone may be so adjusted as to accomplish a desired separation of solids by elutriation. This method may be replaced or supplemented by packing the gas generation zone with relatively large sized non-fluidizable refractory aggregate such as ceramic tower packings, Raschig rings or the like whereby a segregation or classification of the fluidized solids by density may be accomplished. Such segregation may be further enhanced by maintaining carbonaceous solids and metal oxide in different size distribution, the carbonaceous material having preferably a smaller size, and/or by using a metal oxide having a density significantly different from that of the carbonaceous solid.

In spite of the effective separation of ashes and carbonaceous solids from the metal mass withdrawn from the gas generator by the above procedure, the metal mass accomplished in the gas generator for regeneration in a separate oxidation zone frequently may contain quantities of oxidizable carbonaceous matter which interfere with efficient operation of the oxidation zone. In order to eliminate these difficulties it is preferred to carry out the reoxidation of the metal mass in a fluidized oxidation zone packed with relatively large-sized non-fluidized refractory aggregate such as the above-mentioned ceramic tower packings and to adjust the superficial velocity of the fluidizing gas so as to remove overhead from the oxidation zone, ash and carbonaceous materials having a lower density than the metal mass while retaining the concentrated metal mass in the oxidation zone at oxidation conditions.

Having set forth the general nature and objects, the invention will be best understood from the more detailed description hereinafter, in which reference will be made to the accompanying drawing which shows a semi-diagrammatic view of apparatus suitable to practice a preferred embodiment of the invention.

Referring now in detail to the drawing the system illustrated therein essentially comprises a water gas generator (10), a metal oxide regenerator (20) and a separation system (45), (55), (60), the functions and cooperation of which will be presently explained. For the purpose of the following description, the carbonaceous charge will be referred to as coal and the metal oxide as iron oxide, but other materials may be used.

Coal ground to a fluidizable particle size, for example, of the order of 50% having a size of less than 100 mesh per linear inch is fed, preferably continuously, from feed hopper (1) through line (3) which may have the form of an aerated standpipe, to water gas generator (10). Steam, which particularly during the starting period, may be preheated to temperatures of about 1200—1600° F., is supplied through line (5) to the bottom portion of generator (10) and passes through a distribution grid (7) into gas generation zone (9) to react with the coal at a temperature of between 1400° and 2400° F., preferably about 1600° to 1800° F. whereby water gas is formed. About 0.5 to 3.0 lbs. of steam per lb. of coal to be gasified is generally sufficient for this purpose, although if high CO gas is desired, steam addition may be dispensed with, except as required for fluidization, gasification being accomplished by the CO_2 and steam formed by oxidation of the carbon and hydrogen constituents of part of the coal.

Gas generation zone (9) also contains finely divided iron oxide of a particle size slightly larger than that of the carbonaceous solids in the gas generator, e.g., a particle size of say about 50 mesh, which is supplied from metal oxide regenerator (20) as will appear more clearly hereinafter. The amounts of metal oxide required to supply the oxygen necessary to support the heat-generating combustion in generator (10) vary between about 1 and 25 lbs., preferably about 2—10 lbs., per lb. of coal to be gasified, depending on the character of the coal or other carbonaceous solid used, the amount of inert material circulating, and the temperature desired in generator (10). However, this amount is preferably not less than that which gives up about 7—8 cu. ft. of oxygen (measured at standard conditions) per lb. of coal charged (on an ash and moisture-free basis). In addition to supplying oxygen to the gas generation zone, the circulating solid may also act to carry heat evolved in reoxidation zone (25) to the generation zone. The amount of heat so evolved will depend on the metal oxide used, but up to 500 lbs. of solids may be circulated per lb. of coal charged to accomplish this. A wide range of operable ratios of circulating solids to fresh coal addition, therefore, exists in operation of this process.

The linear velocity of the steam is preferably so selected that the charge of gas generation zone (9) forms a dense ebullient mass of fluidized solids having a well defined upper level (L_{10}) and that coal ash is carried overhead from generator (10), suspended in the product gases. Steam velocties of about 0.5—10 ft. per second, preferably 1—4 ft. per second are suitable for this purpose. At these conditions of particle sizes and gas flow, a crude classification of fluidized solids may be accomplished with the result that the solids charge of zone (9) gradually changes from a lower layer consisting substantially of reduced and unreduced iron oxide of high specific gravity to a middle layer consisting of a reacting mixture of coal and iron and iron oxides and an upper layer consisting mainly of coal undergoing gasification with steam. While there is no sharp separation between these layers the classification is sufficiently pronounced to permit the withdrawal of substantially spent metal oxide containing minor proportions of carbonaceous material from the bottom of gasification zone (9) while at the same time allowing for sufficient contact between coal and iron oxide to produce by combustion heat required for the gasification reaction. If desired, this effect may be furthered by arranging a refractory tower packing of relatively large-diameter ceramic Raschig rings in zone (9).

As previously indicated, spent oxide is withdrawn from the bottom of zone (9) and passed downwardly through standpipe (12) aerated through taps (15) by small amounts of a suitable fluidizing gas such as air and/or flue gas and under some conditions, steam, and provided with a slide valve (17). This fluidizing gas may be added in sufficient quantities to strip the spent oxide of occluded water gas.

If desired, a reverse standpipe as illustrated at (12a) may be used in place of standpipe (12) to transport spent metal oxide from generator (10) to regenerator (25). This arrangement has the advantage that the spent metal oxide may be conveyed to regenerator (25) using low pressure air while, if desired, the generator may be kept under a high pressure when using a conventional standpipe of sufficient height for the feed of regenerated metal oxide from regenerator (25) to generator (10). This may be particularly desirable from an economic point of view when high-pressure synthesis gas is to be produced. It should also be noted in this connection that the use of metal oxide in place of ashes or other light inert heat carriers permits the use of standpipes of considerably reduced height for the purpose of establishing the desired pressures. The spent fluidized iron mass, if conveyed through a downflow standpipe, enters pipe (19) wherein it is picked up by air to form a dilute suspension which is passed under the pseudo-hydrostatic pressure of standpipe (12) to the lower portion of metal oxide regenerator (20) and through distributing grid (23) into oxidizing zone (25).

The linear velocity of the air supplied to regenerator (20) is so selected that a dense turbulent mass of fluidized solids having an upper level (L_{20}) is formed above grid (23). Sufficient air must be charged to permit reoxidation of the iron mass at the rate prescribed by the heat requirement of gasification zone (9). In general, 40 to 130 cu. ft. of air per lb. of coal to be gasified is sufficient for this purpose.

The spent metal oxide mass supplied through line (19) may contain carbonaceous constituents resulting from an in-complete separation in gas generator (10). This carbonaceous material may be sufficient in amount to consume more oxygen than would be permissible for efficient operation of regenerator (20). It is preferred, therefore, to provide reoxidizing zone (25) with a packing of large-sized non-fluidizable refractory Raschig rings while maintaining a superficial gas velocity of about 1—4 ft. per second as described in connection with gas generator (10). In this manner, a substantial concentration of metal oxide in zone (25) and elutriation of any material of lower specific gravity including the carbonaceous constituents are accomplished.

The oxidation temperature in zone (25) is maintained between 1500° and 2500° F., preferably at about 1700°—1900° F. by a proper control of the air supply. Residual air consisting essentially of nitrogen and small proportions of CO_2 is withdrawn overhead through a conventional gas-solids separator (27) provided with solids return pipe (29), and through line (31) to be either vented or passed to any desired use such as aeration of standpipe (12) or nitrogen recovery. If desired, cyclone separator (27) may be located downstream of means for cooling the flue gases. Separator (27) is preferably operated so as to return to zone (25) only metal oxide entrained in the residual air.

Regenerated oxidized iron is withdrawn downwardly from regenerator (20) and passed substantially at the temperature of zone (25) into gas generation zone (9) of generator (10) through a standpipe (33), wherein it is stripped with steam admitted through taps (35). The rate of metal oxide circulation through pipe (33) may vary within wide limits depending on the metal oxide and carbonaceous solid used and the desired temperature gradient between zones (9) and (25). It should, however, be sufficient to make about 0.01 to 0.5 mols of O_2 available in zone (9) per lb. of carbon to be gasified. In the case of iron oxide and a common bituminous coal the solids circulation through pipe (33) may be about 1 to 25 lbs., preferably about 5—10 lbs. or 0.02—0.07 cu. ft. of water gas produced.

Fresh metal oxide may be supplied to regenerator (20) through line (37) and metal oxide fines of undesirably small size may be withdrawn from the system through line (39).

Returning now to gas generator (10), product gas consisting essentially of CO and H_2 and small amounts not exceeding about 5—15% of CO_2 and containing entrained coal ash together with a small amount of unconverted coal and iron or iron oxide is withdrawn through line (41) and passed to a conventional gas-solids separator (45), preferably of the centrifugal type, wherein a major proportion of the heaviest entrained solids, particularly metal and metal oxide, is separated and returned through line (47) to gas generator (10). The gas containing the remaining entrained solids may be passed through line (50) to a water scrubber (55) wherein it is completely freed of solids by water supplied through line (57) for countercurrent flow. Product gas is withdrawn overhead from scrubber (55) and passed through line (58) to conventional gas purification equipment (not shown) and ultimately to a synthesis reactor or other use.

Water containing coal ash and minor amounts of metal or metal oxide and unreacted coal may be withdrawn from the bottom of scrubber (55) through line (59) and passed to a settling zone (60) wherein a further crude classification of the suspended solids by density takes place. A suspension enriched in coal ash may be decanted and removed from settler (60) through line (63). The lower layers of settler (60) contain a suspension enriched in metal or metal oxide and some carbonaceous material which may be withdrawn through line (65) and passed into steam feed line (5) to be returned to generator (10).

If desired, tail gas from the hydrocarbon synthesis may be recycled to generator (10), for example, via steam feed line (5) in order to modify the composition of the water gas produced by the conversion of the gaseous hydrocarbons and CO_2 contained in the tail gas.

While no specific pressures have been mentioned above it should be understood that pressures varying from slightly subatmospheric to about 400 lbs. per sq. in. gauge may be applied. If fuel gas of high B.Th.U. value is desired, the methane content of the gas may be increased by using pressures of at least 200 lbs. per sq. in. A particular advantage of the invention resides in the fact that synthesis gas may be produced in generator (10) at any desired elevated pressure while reoxidizer (20) may be operated at substantially atmospheric pressure, thus savng the cost of compressing large volumes of air to high pressures.

It will be appreciated from the above description that the system illustrated in the drawing permits a fully continuous conversion of carbonaceous solids into a mixture of carbon monoxide and hydrogen of improved purity, affords optimum heat economy and requires a minimum of solids circulation. While preferred means have been shown for recovering metal oxide suspended in the product gas or admixed with other solids, the invention is not limited to these specific means. Numerous variations and modifications thereof may appear to those skilled in the art without departing from the spirit of the invention.

The invention will be further illustrated by the following specific example.

EXAMPLE

For the production of 415 million cu. ft. of synthesis gas per day containing 175 million cu. ft. of H_2, 175 million cu. ft. of CO, and 65 million cu. ft of CO_2 from a coal containing 4.2 lbs. of H_2 per 100 lbs. of carbon using iron oxide as the oxygen carrier, the following operating conditions have been found to be suitable.

Coal feed	188 tons per hour
Process steam feed	440,000 lbs. per hour
Air feed to regenerator	315,000 standard cu. ft. per minute
Solids circulated from regenerator to generator	1,200 tons per minute
Generator temperature	1800° F.
Generator pressure	100 lbs./sq. in. gauge
Regenerator temperature	1900° F.
Regenerator pressure	1 lb./sq. in. gauge
Approximate height of standpipe from regenerator to generator	100 ft.
Preheating temperature of steam and air	1100° F.

While the foregoing description and exemplary operations have served to illustrate specific applications and results of the invention, other modifications obvious to those skilled in the art are within the scope of the invention.

Having now particularly described and ascertained the nature of the said invention, and in what manner the same is to be performed, as communicated to me by my foreign correspondents, I declare that what I claim is:—

1. A process for the production of mixtures of carbon monoxide and hydrogen by treating solid carbonaceous material in the form of a dense turbulent fluidized bed of finely divided particles with steam at an elevated temperature wherein a substantial part of the heat required for the reaction between the said carbonaceous material and the steam is supplied by the combustion of a portion of the said carbonaceous material simultaneously with the reaction with the steam, said combustion being supported by oxygen liberated from a finely divided solid oxygen carrier in the reaction chamber in direct contact with the said carbonaceous material.

2. A process as claimed in Claim 1, wherein the amount of the said finely divided oxygen carrier in the reaction chamber is sufficient to supply the major portion of the heat required for the reaction between the carbonaceous material and the steam.

3. A process as claimed in Claim 1 or 2, wherein the particle size of the carbonaceous material is smaller than that of the oxygen carrier whereby the mixture of solids in the reaction chamber is caused to segregate into a plurality of layers, the lower of which comprises relatively pure reduced and unreduced oxygen carrier.

4. A process as claimed in Claim 3, in which the reaction zone is also provided with packing elements to assist in effecting the separation of the oxygen carrier from the reaction mixture.

5. A process as claimed in any one of Claims 1—4, wherein the spent oxygen carrier is withdrawn from the lower part of the gasification reaction zone and is passed to a reoxidation zone in which it is treated in the form of a dense turbulent fluidized mass of finely divided solid particles with air or an oxidizing gas to reoxidize it to its initial state, and the thus regenerated carrier is recycled to the gasification reaction zone.

6. A process as claimed in Claim 5, wherein the reoxidation of the oxygen carrier is carried out at a temperature higher than that in the gasification zone, and the regenerated carrier is recycled to the gasification zone at such higher temperature whereby part of the endothermic heat requirement of the gasification reaction is supplied as sensible heat from the hot oxygen carrier.

7. A process as claimed in Claims 5

and 6, wherein the gasification reaction is carried out at a temperature between 1600 and 1800° F., and the reoxidation of the oxygen carrier is carried out at a temperature between 1700 and 1900° F.

8. A process as claimed in any one of Claims 1—7, wherein the gasification reaction is carried out under a pressure of at least 200 pounds per square inch, and the reoxidation of the oxygen carrier is carried out at substantially atmospheric pressure.

9. A process as claimed in any of Claims 1—8, wherein the oxygen carrier is a metal oxide capable of oxidizing carbonaceous materials under the conditions of gasification, and is preferably an oxide of iron or copper.

10. A process as claimed in any of the preceding claims wherein the steam is passed upwardly through the carbonaceous material and metal oxide in the gasification reaction zone at such velocity that the fine particles of ash formed from the carbonaceous material are carried overhead with the product gases, from which they are separated in known manner.

11. A process as claimed in Claim 10, in which any oxygen carrier separated from the said product gas is recycled to the gasification zone.

12. A process as claimed in any of the preceding claims in which packing elements are arranged in the oxygen carrier regeneration zone whereby the oxygen carrier is caused to separate into layers of varying density and particle size, and the velocity of the oxidizing gas is regulated to carry any solids of lower specific gravity than the regenerated oxygen carrier overhead with the waste gases therefrom.

13. A process as claimed in Claim 5, wherein the reoxidation of the oxygen carrier is carried out at substantially lower pressure than the gasification reaction, and the spent oxygen carrier withdrawn from the gasification zone is passed to the reoxidation zone by means of a reverse standpipe.

Dated this 31st day of March, 1947.
D. YOUNG & CO.,
29, Southampton Buildings,
Chancery Lane,
London, W.C.2,
Agents for the Applicant.

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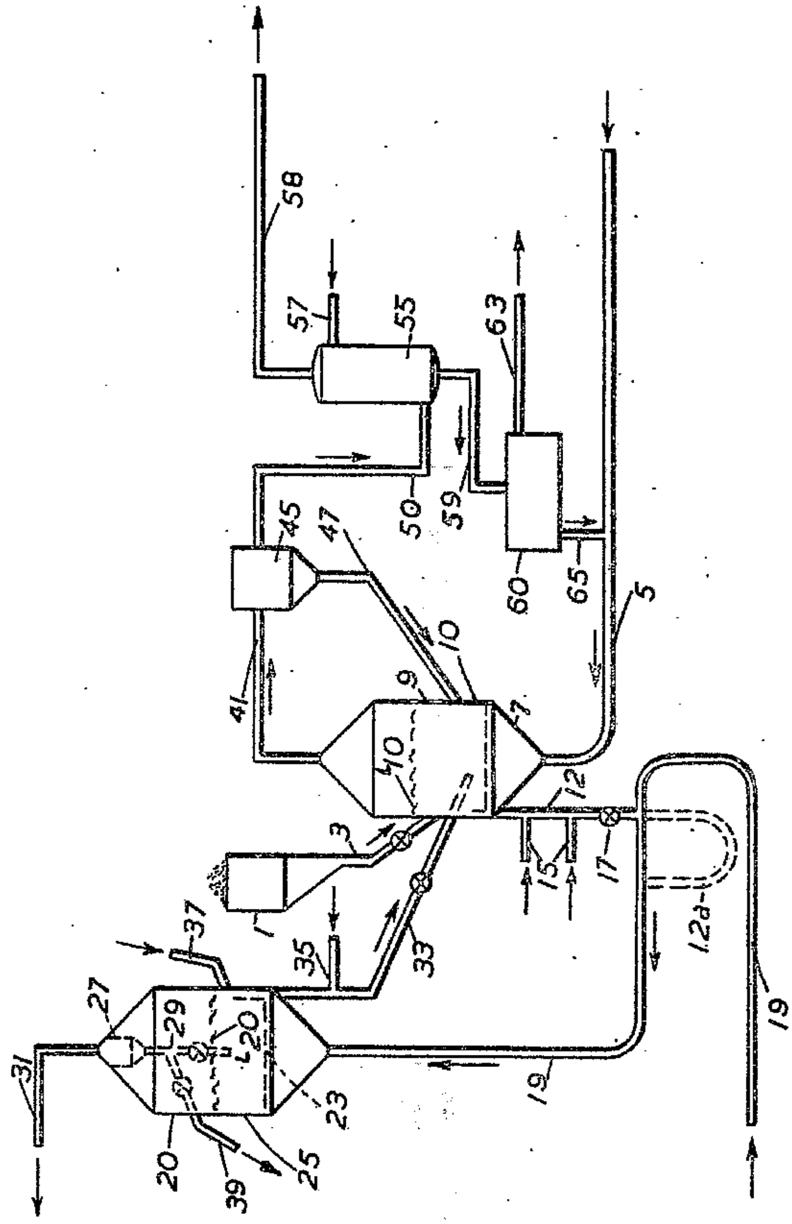

H.M.S.O.(Ty. P.)