

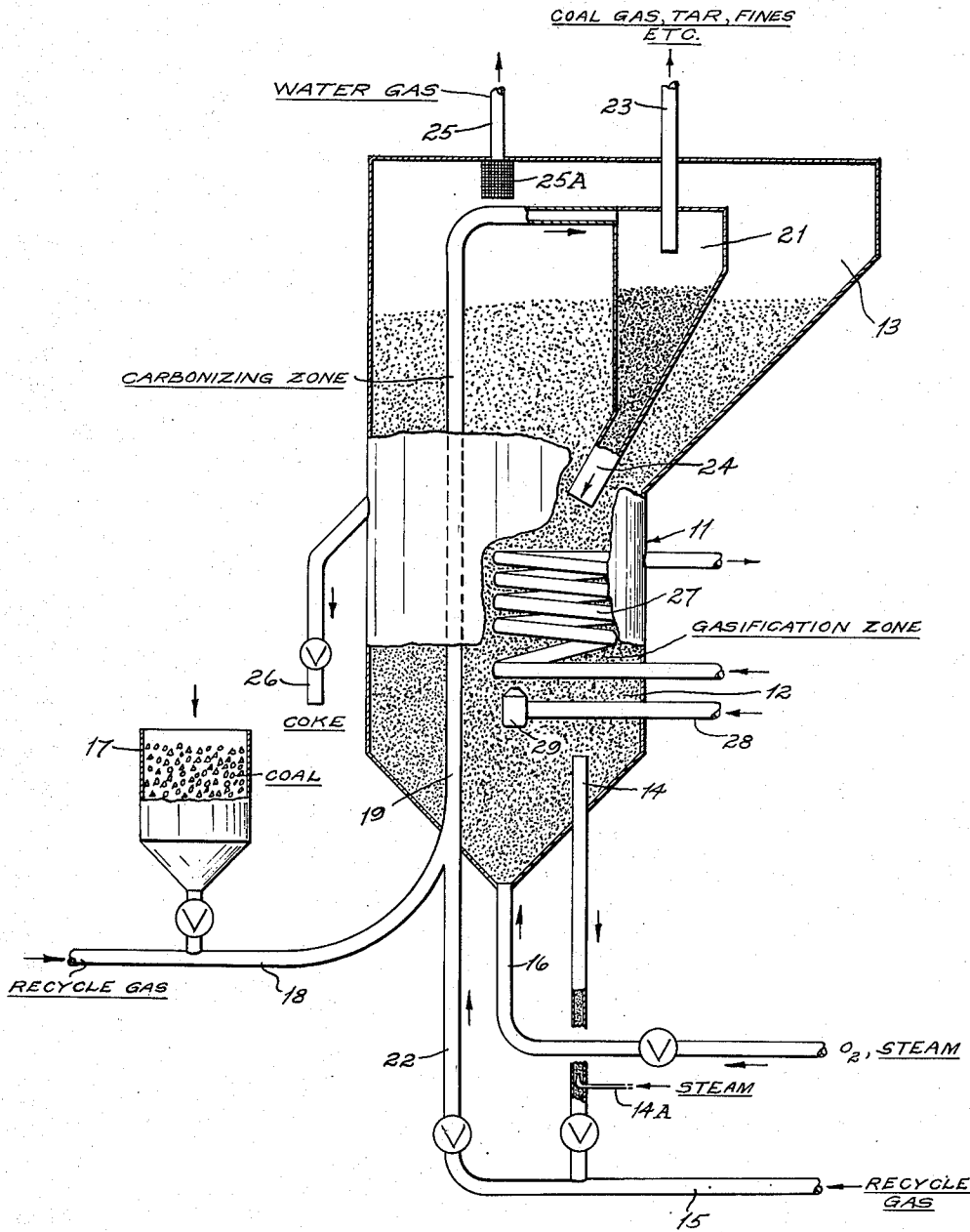
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COAL CARBONIZATION AND GASIFICATION

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COAL CARBONIZATION AND GASIFICATION

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This invention relates to a process for the pyrolysis of solid carbonaceous material. In one of its more specific aspects, this invention relates to a process for the carbonization and gasification of solid carbonaceous materials, such as coal, lignite, and the like, which is particularly useful for the gasification of such materials for the preparation of synthesis gas for synthesis of hydrocarbons, oxygenated compounds, and the like.

The conversion of solid carbonaceous materials, such as coal, lignite, brown coal, and the like to a gas or gas mixture of predetermined B. t. u. content has been a major problem because of the quantity of heat involved, the large masses of material handled, the presence in the material of volatiles such as heavy oils and tars leading to agglomeration of the large particles, and other factors. Such factors have compelled the use of relatively complicated processes and apparatus, usually involving a number of heat exchange steps in all of which heat is wasted and the overall efficiency of the elements reduced. Furthermore, in such cases, the gas or gas mixture produced has been of relatively poor quality and it has been difficult to control its production to secure final products of the desired composition.

An object of this invention is to provide an improved process for the pyrolysis of a solid carbonaceous material.

Another object of this invention is to provide an improved method for the pyrolysis of a carbonaceous material containing volatile components.

Another object is to provide an improved method for the carbonization of a carbonaceous material containing volatile components, which carbonaceous material, upon heating to vaporize the volatile components, leaves residual carbon.

Another object is to provide an improved method for the gasification of carbonaceous material containing volatilizable constituents.

Still another object of this invention is to provide an improved method for treating a carbonaceous material containing volatilizable constituents to effect carbonization of said material or removal of volatile constituents therefrom and the exothermic reaction of residual carbon.

Still another object is to provide such a process wherein the heat liberated in the gasification or reaction step is efficiently utilized to effect volatilization of volatile components from the charge material.

A further object of this invention is to provide apparatus for carbonizing and gasifying a car-

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bonaceous material containing volatilizable constituents.

Other objects and advantages of the invention will appear from the following description and claims taken in connection with the attached drawing wherein a preferred embodiment of the invention is shown diagrammatically.

In a preferred specific embodiment of this invention, a solid carbonaceous material is gasified by a novel method wherein the heat for the necessary carbonization and gasification is produced from the material itself, the heat exchange phases being carried out in a manner whereby substantially all the heat is conserved and put to efficient use. The volatile components, such as tar, oil, and coal gas are volatilized in a carbonizing step by a highly efficient transfer of heat from the actual gasifying step, such volatile components being kept out of physical contact with the material being gasified to avoid contamination and interference with the production of the final product gas. The carbonaceous material in finely divided form is dispersed in a gaseous medium and subjected to a flash carbonization, the heat therefor being supplied by oxidation of residual coke from the carbonization. The entire process including the carbonization and the gasification is carried out in a single reactor, means being provided for separating the operations from one another to prevent mixing of the several gaseous products produced thereby.

In brief, in a preferred specific embodiment, the present invention contemplates the maintenance of a mass of devolatilized or carbonized material, for example, coke, preferably in the form of a fluidized mass, in contact with a reactant gas or gases whereby the carbonized material is gasified with the generation of heat to produce the desired gaseous products, the material being arranged in indirect heat exchange relation with an incoming mass of fresh carbonaceous material containing volatile components whereby the heat generated by the gasification of the carbonized or devolatilized material is made available without substantial loss to the fresh material to effect the distillation of the volatile components therefrom, these volatile components being separately withdrawn to avoid mixing with the gaseous products of gasification.

Preferably, the fresh carbonaceous material in the form of a fine powder is suspended in a non-oxidizing gas, suitably recycled coal gas, and conveyed through a heat exchange device heated by the mass of coke being gasified, the residual solid

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heated fresh material being discharged in the form of coke into a separator, also receiving heat from the reacting zone, wherein the gasified volatile components are separated from the coke. The separator is arranged to discharge the gaseous volatile matter in a manner to avoid mixing with the gases from the gasifying zone and to discharge the resultant carbonized material or coke into the fluidized mass of coke already in the gasification zone of the reactor. To minimize mixing of the products of gasification with vapors of volatilized components from the carbonizing step, the solids in the outlet of the separator and the level of the coke mass in the gasification zone are arranged so that there is a substantial resistance to gas flow therebetween. The gaseous reactant or reactants fed into the fluidized mass of coke may be of any desired type which will react therewith exothermically, e. g., oxygen or hydrogen. Limited proportions of endothermically reacting agents, e. g., steam or carbon dioxide, may also be introduced but care must be exercised to ensure that the gasification step as a whole is sufficiently exothermic to provide the heat requirements of the carbonization step. Where synthesis gas is the desired product of gasification, the proportions of carbon monoxide and hydrogen in the product gas may be varied by the proportions of oxygen and steam used.

Referring to the drawing which is to be understood as illustrative and not limiting of the invention, a reactor 11 is provided with a fluidizing section 12 and a separating section 13 of enlarged cross-section to reduce the velocity of the gases and thus facilitate the separation of coke particles therefrom. A standpipe 14 is connected to a line 15 for discharging hot coke into a stream of recycle gas or similar gas. Steam is bled into standpipe 14 through pipe 14A to keep the solids therein in a free-flowing condition. A gas inlet 16 supplies an oxygen-containing gas with or without additions of carbon dioxide and/or steam to the base of the reactor to effect fluidization and gasification. The carbonaceous material to be treated, such as coal, lignite, and the like, preferably in the form of a fine powder, is charged from a hopper 17 into a line 18 and fed upwardly through a heat exchange element 19 into a separator 21, suitably of the cyclone type or the equivalent, in the top of the reactor. A recycle stream of coke from line 14 carried by recycle gas from line 15 may be fed through line 22 to assist in preventing agglomeration of the coal particles during devolatilization and in supplying heat to the coal particles. Separator 21 is formed with a gas outlet 23 and a discharge conduit 24 for solids. An outlet 25 is provided for the gaseous product of gasification and an outlet 26 for product coke in reactor 11. A filter element 25A serves to remove entrained solids from the gases leaving by way of outlet 25.

In operation, the carbonized carbonaceous material or coke is fluidized in section 12 of reactor 11 by gases introduced through line 16. For such fluidizing, it has been found that satisfactory results can be secured with coke particles of less than 40-60 mesh in size, preferably about 40 to 50 per cent of the particles finer than 200 mesh in size, and a gas velocity of about 0.5 to 3.0 feet per second. The flow of gas is so arranged that the level of the coke extends above the discharge port 24 of the separator for a purpose to be later described.

The oxygen-containing gas, preferably at least 95 per cent oxygen, reacts exothermically in the

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reactor to produce carbon monoxide. The addition of steam leads to the generation of water gas, i. e., hydrogen and carbon monoxide, the composition of the final product being variable by varying the proportions of the gases fed in accordance with the well-known practice.

The product gas is taken off through line 25 and conducted to storage facilities, a synthesis reactor, or any other desired point. Product coke may be removed as desired through line 26. Gasification may be carried to any desired extent including substantially complete consumption of the carbon in the residue from the carbonization step; otherwise, high-ash coke may be withdrawn from the gasification zone for treatment in a separate zone.

Fresh powdered uncarbonized material, preheated, if desired, is fed from hopper 17 into line 18, the material being caused to travel through line 19 by reason of the gas fed through line 18. Line 19 constitutes a heat exchanger entirely surrounded by the reacting coke, and hence directly heated thereby without the possibility of any loss of heat in the transfer. Line 19 may be in the form of a spiral, a series of tubes, or any other suitable form, it being desirable that the maximum heat exchange surface be provided consonant with the rate of flow of the material conveyed therethrough. In passing through line 19, volatile components such as water, coal, gas, tar, etc., are distilled from the material, leaving coke. The volatile matter, together with coked fines from the mass, is separated from the residual solid material or coke in separator 21 and discharged through line 23 to suitable recovery means.

While the material in line 19 is heated by the surrounding fluidized mass, it can be considered to be in indirect heat exchange relation with said mass by reason of its being maintained separate therefrom as regards physical contact, the volatilized constituents likewise being maintained out of physical contact with the fluidizing mass or the gases discharged therefrom.

The coke collects in the bottom of the separator and is gradually fed into the fluidized mass in the gasification chamber 12 through conduit 24 at a rate generally corresponding to the rate of consumption of the coke in the fluidized mass, the feed being substantially automatic. It will be noted that while the coke in the bottom of the separator and the coke in the gasifying section 12 are of substantially the same density per se, the fluidizing of the mass in section 12 tends to decrease the effective density in section 12, thus creating a pressure differential operating to cause coke to flow from the separator into the major body of coke in section 12. Because of the resistance to gas flow offered by the relatively packed char in conduit 24, it is difficult for the gases introduced through line 16 to pass into separator 21 or the volatile material distilled in line 19 to escape into sections 12 and 13 of the reactor to become mixed with the product gas or enter into reactions therewith.

By feeding fresh carbonaceous materials to the apparatus at a suitable rate, depending upon various factors, such as the cross-sectional area of lines 18 and 19, the material charged into line 18 may be so treated in heat exchanger 19 as to be in a substantially carbonized condition and ready for reaction with the oxygen and steam in the gasification chamber when it is discharged from line 19 into separator 21.

By means of section 13 being of enlarged cross-section relative to section 12 there is a consider-

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able reduction in the velocity of the fluidizing medium therethrough. Accordingly, the larger particles in the mass tend to fall back into the reactor, only some fines being entrained and separated by filter 25A from the gases leaving through line 25.

By reason of the conveyance of the fresh material through line 19 with a gaseous medium, the carbonization is effected in a widely dispersed phase which is in rapid motion. This high velocity insures an effective transfer of heat received from the surrounding fluidized mass of coke; accordingly, there is little or no chance for agglomeration in line 19 regardless of the original volatile content of the material. It will also be noted that the coal gas and other volatiles are in contact with the solid material for only a short period of time, thereby enabling a minimum of cracking or degradation of these products.

With a bed depth in the gasification section of about 40 feet and an overhead gas space about 5 feet deep and a temperature in the range of about 1200-1600° F., within pipe 19, the coking of the fresh feed in line 19, when in finely divided form, can be accomplished in a travel or residence time of about 1 to 4 seconds. In such case, the temperature of the fluidized bed in the reactor is maintained in the range of about 1300-1800° F., dependent upon the rate of travel or residence time of the fresh material in line 19. Obviously, the temperature as well as the travel time and the rate of flow of the fresh material will vary with different type of carbonaceous materials.

The present invention requires only a single reactor and a minimum of external piping through which heat may be lost. No difficulty is involved in determining the correct coal grind as with the conventional two chamber design and the problem of agglomeration is substantially eliminated. In cases where more heat is generated in the gasification step than is required for the carbonization step, the excess heat may be recovered in useful form, for instance, by making steam in coil 27 or equivalent heat exchanger disposed in contact with the carbon-containing bed which is being exothermically gasified. Alternatively, excess heat may be utilized to crack a heavy hydrocarbon fraction, say gas oil, introduced through line 28 and distributing nozzle 29; this method of utilizing excess heat is particularly appropriate where a carburetted water gas is the desired gaseous end product of the gasification step.

In the treatment of oil shale and like material containing a large proportion of relatively inert material of the same waste character as coal ash, suitable means is provided for the removal of the waste material such as separators in the product gas lines or a solid discharge outlet in reactor 11.

An important field in which my invention finds application is the manufacture of city gas. In such instance, synthesis gas produced in the gasification step is reacted in the presence of a catalyst to form methane which is mixed with coal gas from the carbonization step to yield the desired city gas.

Where the term "carbonaceous material" is employed herein, it is intended to include coal, lignite, oil shale, brown coal, and the like, containing varying proportions of volatile constituents which, after distillation, leave a solid carbon-containing residue that is capable of reacting with an oxygen-containing gas or hydrogen at elevated pressure in an exothermic manner.

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Obviously, many modifications and variations of the invention as above set forth may be made without departing from the spirit and scope thereof and only such limitations should be imposed as are indicated in the appended claims.

I claim:

1. In a process for carbonizing and gasifying a solid carbonaceous material containing volatilizable constituents wherein said material is subjected to carbonization by the distillation of volatilizable constituents therefrom in a distillation zone and the resulting carbonized material is subjected to gasification with an oxygen-containing gas in a gasification zone, the improvement which comprises passing said carbonaceous material in finely divided form as a dispersed phase entrained in a stream of non-oxidizing gas flowing at a velocity sufficient to inhibit settling of said carbonaceous material therein through the distillation zone in indirect heat exchange with a reacting dense phase fluidized mass of the carbonized material undergoing gasification in the gasification zone whereby heat required for the carbonization is transferred from the gasification zone to the distillation zone, separating the resulting carbonized material from gases comprising volatilized constituents from the solid carbonaceous material, introducing at least a part of said carbonized material into the gasification zone, and separately discharging the gases from the distillation zone and from the gasification zone.

2. A process as defined in claim 1 wherein carbonized material from the gasification zone is suspended in non-oxidizing gas in admixture with said carbonaceous material supplied to the distillation zone.

3. A process as defined in claim 1 wherein said carbonaceous material is coal.

4. In a process for carbonizing and gasifying a solid carbonaceous material containing volatilizable constituents wherein said material is subjected to carbonization by the distillation of volatilizable constituents therefrom in a distillation zone and the resulting carbonized material is subjected to an exothermic gasification reaction with a reactant gas in a gasification zone, the improvement which comprises passing said carbonaceous material in finely divided form as a dispersed phase entrained in a stream of non-oxidizing gas flowing at a velocity sufficient to inhibit settling of said carbonaceous material therein through a distillation zone contiguous and in direct heat exchange with a reacting dense phase fluidized mass of the carbonized material undergoing gasification in a separate gasification zone whereby heat required for the carbonization is transferred from the gasification zone to the distillation zone, separating the resulting carbonized material from gases comprising volatilized constituents from said solid carbonaceous material, introducing at least a part of said carbonized material into the gasification zone, and separately discharging the gases from the distillation zone and from the gasification zone.

5. A process as defined in claim 4 wherein carbonized material from the gasification zone is suspended in non-oxidizing gas in admixture with said carbonaceous material supplied to the distillation zone.

6. A process as defined in claim 4 wherein the exothermic gasification reaction is carried out with hydrogen at an elevated pressure as the reactant gas.

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7. In a process for carbonizing and gasifying a solid carbonaceous material containing volatilizable constituents wherein said material is subjected to carbonization by the distillation of volatilizable constituents therefrom in a distillation zone and the resulting carbonized material is subjected to an exothermic gasification reaction with a reactant gas in a gasification zone, the improvement which comprises passing said carbonaceous material in finely divided form as a dispersed phase entrained in a stream of non-oxidizing gas flowing at a velocity sufficient to inhibit settling of said carbonaceous material therein upwardly through a distillation zone disposed within a gasification zone containing a reacting dense phase fluidized mass of the carbonized material undergoing gasification whereby heat required for carbonization is transferred by indirect heat exchange from the gasification zone to the distillation zone, introducing the resulting carbonized material and gases comprising volatilized constituents from the solid carbonaceous material into a separation zone disposed within said gasification zone and having

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an outlet for solid carbonized material disposed within said fluidized mass of carbonized material whereby carbonized material from said separation zone is introduced directly into said fluidized mass in said gasification zone, withdrawing said gases comprising volatilized constituents and substantially free from carbonized material from said separation zone, and separately discharging the gases from the gasification zone.

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REFERENCES CITED

The following references are of record in the file of this patent:

UNITED STATES PATENTS

| Number | Name | Date |
|-----------|----------------------|---------------|
| 2,248,196 | Plummer ----- | July 8, 1941 |
| 2,285,276 | Hemminger ----- | June 2, 1942 |
| 2,341,193 | Scheineman ----- | Feb. 8, 1944 |
| 2,378,342 | Voorhees et al. ---- | June 12, 1945 |
| 2,428,872 | Gunness ----- | Oct. 14, 1947 |
| 2,445,327 | Keith ----- | July 20, 1948 |