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CONVERSION OF CARBONACEOUS MATERIALS INTO A FUEL GAS

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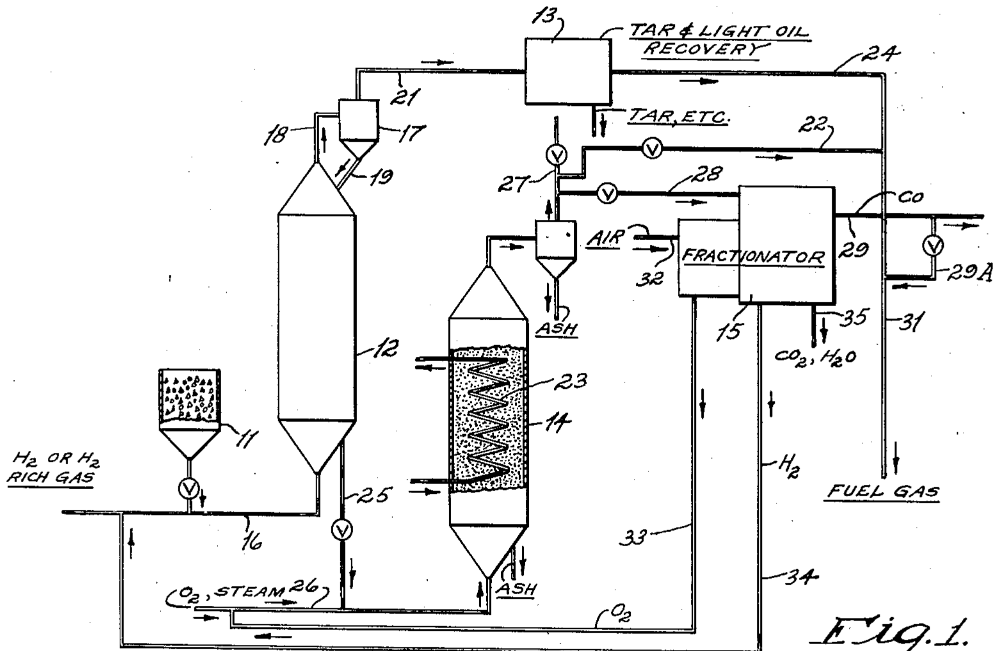

Fig. 1.

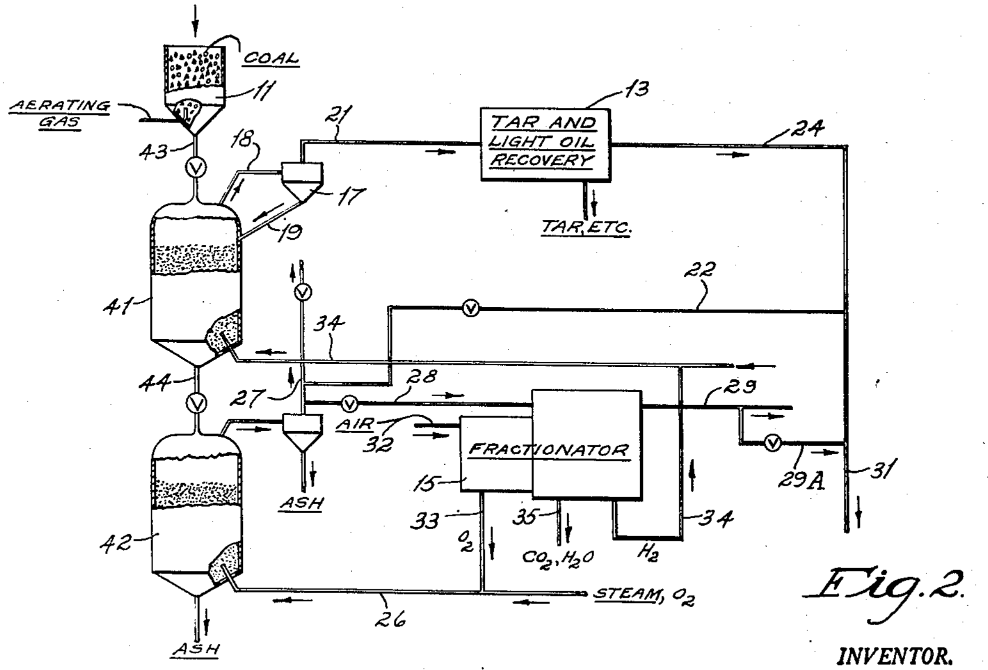

Fig. 2.

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CONVERSION OF CARBONACEOUS MATERIALS INTO A FUEL GAS

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4 Claims. (Cl. 48—197)

This invention relates to the carbonization and gasification of solid carbonaceous materials such as coal, oil shale, lignite, and the like to produce fuel gases of relatively high heating values.

The prior art relating to the production of fuel gases from coal is replete with diverse processes. However, all of such processes fall into two general classes, namely, the cheaper processes productive of fuel gases of relatively low calorific value such as producer gas, and the more complicated processes productive of fuel gases of higher calorific value but too expensive for practical application. One of the latest proposals in this art involves the processing of the coal of separate carbonization and gasification steps, the coal being carbonized by hot coke from the subsequent gasification step, wherein a part of the coke is oxidized to bring the remainder of the mass to a sufficiently high temperature for the recycle. However, this process has the disadvantage of requiring the recycling of a large stream of hot coke or other heat carrier to provide the heat for the coal carbonization, and the further disadvantage of the carbonizing and gasifying temperature being dependent variables, controllable only to a limited extent and materially influenced by the ratio of recycle coke to fresh coal or a compromise between desired conditions in the two steps.

In improving over the prior art, the present invention has as a major object the provision of a novel carbonization and gasification process wherein the heat required for carbonization is generated in situ by hydrogenation of the material within the carbonization zone under predetermined pressure and temperature conditions, no cycling or recycling of hot coke or other heat carrier being required.

Another object of the invention is the provision of a novel process wherein the carbonization of the material is accomplished with simultaneous hydrogenation, the gaseous products of the carbonization and the hydrogenation forming a fuel gas of a relatively high calorific value.

A further object of the invention is to provide a novel process wherein the carbonization of the material is effected along with hydrogenation, the products of the overall operation being processed in a manner to supply substantially all the hydrogen requirements for the process.

Other objects and advantages of the invention will be apparent from the following description and claims taken in connection with the accompanying drawing wherein:

Fig. 1 is a diagrammatic showing of a process illustrating a preferred application of the invention.

Fig. 2 illustrates a modified process embodying the invention.

In its broader aspect, the present invention is directed to the carbonization and gasification of a solid carbonaceous material in particles of relatively fine sizes, capable of being fluidized, by contacting the fresh material in the fluidized phase with hydrogen or a hydrogen-containing gas to effect the hydrogenation of material in the carbonization zone with the generation of heat, the removal of the volatiles and the formation of coke; the subsequent oxidation or gasification of the coke in the fluidized phase; and the selection and combination of constituents of the gaseous products from both operations to produce a final fuel gas of high calorific content. More specifically, the invention contemplates the supplying of the heat requirements for the carbonization by the exothermic hydrogenation, and the production of the hydrogen requirements in connection with the preparation or separation of the oxygen for the gasification step, the costs of the gases being thus brought down to practical limits.

Referring to Fig. 1, the apparatus shown therein in diagrammatic fashion includes a feed hopper 11, a combined carbonization-hydrogenation reactor 12, a tar and light oil recovery unit 13, a gasification reactor 14, and a combined oxygen and hydrogen separation unit 15. Suitable connections and auxiliary apparatus are described later in connection with the manner of operation of the apparatus.

In carrying out the process, coal in finely divided form and capable of being fluidized by a gaseous medium is fed from hopper 11 into a line 16 carrying hydrogen or a hydrogen-rich gas at a sufficient velocity to carry the coal into reactor 12 and maintain the coal particles therein in a fluidized, ebullient mass having the appearance of a boiling liquid. The coal particles are preferably less than 46–60 mesh in size, 40–50 per cent thereof being 200 mesh or smaller, the fluidizing medium being introduced at a rate such that its rate of flow in the reactor is in the order of about 0.5–3.0 feet per second.

The hydrogen and the contents of reactor 12 are caused to react exothermically, the fluidizing action ensuring excellent contact between the gas and the particles. At the same time, distillation takes place wherein the volatile material is driven from the coal and under the prevailing reaction conditions there is little or no tendency for the volatiles to become cracked or degraded as is the usual case in conventional carbonization processes. Furthermore, methane is generated by the hydrogenation reaction so that the product gases are further enriched in hydrocarbon content. The distillation is relatively fast compared with the hydrogenation reaction, the rate of feed being such that only a small part of the solid carbon has time to react with the hydrogen but that part being adequate to provide the heat required for the distillation. Therefore, excess coke is transferred to the gasification chamber **14**. The rates of feed to the reactor **12** are controlled so that the temperature therein is in the range of 1000° to 1500° F., the pressure being in the range of 300 to 750 pounds per square inch gauge. At lower temperatures, the methane production is relatively low.

A cyclone separator **17**, connected to the reactor **12** by lines **18** and **19**, serves to separate entrained solids from the reaction gases and to return them to the reactor, the gaseous products exiting through line **21**. The latter, including methane, hydrogen, carbon monoxide, light oil, tar and hydrogen sulfide pass through a liquid recovery unit **13** wherein tar, light oil and other liquid constituents are separated from the reaction gases. The separated gas mixture is taken off through line **24**.

Coke is taken from adjacent the bottom of reactor **12** by line **25** and fed into a stream of an oxygen-containing gas (preferably not less than 95 per cent oxygen) in line **26**, usually supplemented with steam. The coke particles are thus transported to and fluidized in reactor **14**, the steam and oxygen reacting with the coke to form the desired gaseous mixture. Preferably the gasification reaction is conducted at a temperature in the range of 1400° to 2400° F. and a pressure in the range of atmospheric to 300 pounds per square inch gauge. The proportion of steam and oxygen to the coke can be varied in well-known manner to vary the composition of the product gas. Thus, synthesis gas consisting essentially of one part of carbon monoxide to two parts of hydrogen for synthesis of hydrocarbons, oxygenated compounds or the like can be prepared, the gas being taken off through a line **27** and passed to a synthesis reactor or storage facilities. Otherwise, the gas may flow through line **22** and mix with the gas in line **24** to provide a fuel gas of desired characteristics.

The gaseous products may also be conveyed through line **28** to a low temperature separation unit **15** wherein the carbon monoxide and hydrogen are separated, the carbon monoxide leaving through line **29**. The latter stream by flowing through line **29A** may be mixed with the gas in line **24** to form in line **31** a fuel gas with a heating value of 500 to 550 B. t. u. per cubic foot. This relatively high heating value is secured without the necessity of high temperature carburation or the use of a Fischer-type synthesis unit. Preferably the low temperature separation unit **15** is of a type wherein the steps of separating oxygen from air entering through line **32** and separating hydrogen from the product gases in line **28** are accomplished in a single coordinated unit, the oxygen being fed through line **33** to line **26** and the hydrogen through line **34** to line **16**. Water and carbon dioxide are removed through line **35**. If desired, the gas separations can be accomplished in separate units.

Fig. 2 illustrates a modification of the invention wherein the feed hopper **11**, a combined carbonization and hydrogenation reactor **41** and a gasification reactor **42** are arranged in vertical relation, the remaining components of the system being the same as shown in Fig. 1, like reference characters being applied thereto. The coal, in a finely divided form capable of being fluidized, is fed through line **43** into reactor **41**, the hydrogen-containing gas being introduced at the bottom of the reactor through line **34** and reacting with the fluidized contents in the same manner as described in connection with Fig. 1. The gaseous products are removed through lines **18** and **21** and processed as already described. The coke is conducted through line **44** into reactor **42** for gasification by its reaction with oxygen and steam charged through line **26**, the gaseous products being processed as already described. This arrangement possesses the advantage that the coal and its solid residue follow a vertical path, the flow thereof being accomplished through gravity.

In both Figs. 1 and 2, ash is removed at convenient points, suitable lines therefor being shown. In view of the relatively high pressures employed at some stages in the process, it is to be understood that suitable valve controlled seals will be used wherever necessary in accordance with well known engineering practice.

It will be noted herein that it is unnecessary to recycle a quantity of coke or like heat carrier to supply the heat for the carbonization, the heat requirements therefor with the production of desirable gas products being secured by controlled hydrogenation within the carbonization zone. Additionally the carbonizing and gasifying steps are entirely independent, there being no recycle of coke or other heat carrer therebetween dictating any relation in such conditions such as heat and pressure.

The retardation of the hydrogenation by carbon monoxide in the carbonizing step is avoided or materially minimized by recycling to the carbonization zone hydrogen substantially free of carbon monoxide and excellent controls over the various steps and the final products are obtained, those controls being available to the end that the degrees of carbonization and gasification and the characteristics of the final products can be widely and easily varied.

While coal has been described herein by way of example as the feed material, it is to be understood that any carbonaceous material having volatilizable constituents and leaving a solid carbon-containing residue can be similarly processed. The term "carbonaceous material" as used herein and in the appended claims is intended to define such feed materials. The processing of such materials as oil shales having a relatively high content of inert material is also contemplated since such inert material can be removed in the same manner as ash.

Obviously, many modifications and variations of the invention as above set forth may be made without departing from the spirit and scope thereof. For instance, a coil **23** or equivalent heat exchanger may be placed in any of the reactors as shown in reactor **14** to make steam and thus recover in useful form excess heat generated within the reactor. Also, the fresh coal or like feed material may be partially preheated, for instance, by heat exchange with hot product gases. Only such limitations should be imposed on the invention as are indicated in the appended claims.

I claim:

1. In a process for producing fuel gas of high heating value from a solid carbonaceous material containing volatilizable constituents, the improvement which comprises simultaneously hydrogenating and carbonizing autogenously said solid carbonaceous material by continuously contacting said carbonaceous material in comminuted form in a dense phase fluidized bed with a stream of hydrogen at a pressure within the range of from about 300 to about 750 pounds per square inch gauge and at a substantially uniform temperature within the range of from about 1,000 to about 1,500° F. in a carbonization zone, continuously introducing said carbonaceous material into said carbonization zone, discharging a gaseous product comprising hydrocarbons and unreacted hydrogen from said carbonization zone, continuously withdrawing carbon-containing residue from said carbonization zone and reacting said residue in a gasification zone with an oxygen-containing gas and steam to produce a mixture of carbon monoxide and hydrogen, separating hydrogen from said mixture, passing said hydrogen to the carbonization zone as the source of hydrogen for reaction with said solid carbonaceous material, and admixing carbon monoxide separated from said mixture with said gaseous product from the carbonization zone to produce a fuel gas of relatively high heating value.

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

3. A process as defined in claim 1 wherein said oxygen-containing gas contains not less than 95 per cent oxygen.

4. A process as defined in claim 1 wherein the gasification zone is operated at a pressure within the range of from about atmospheric to about 300 pounds per square inch gauge.

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