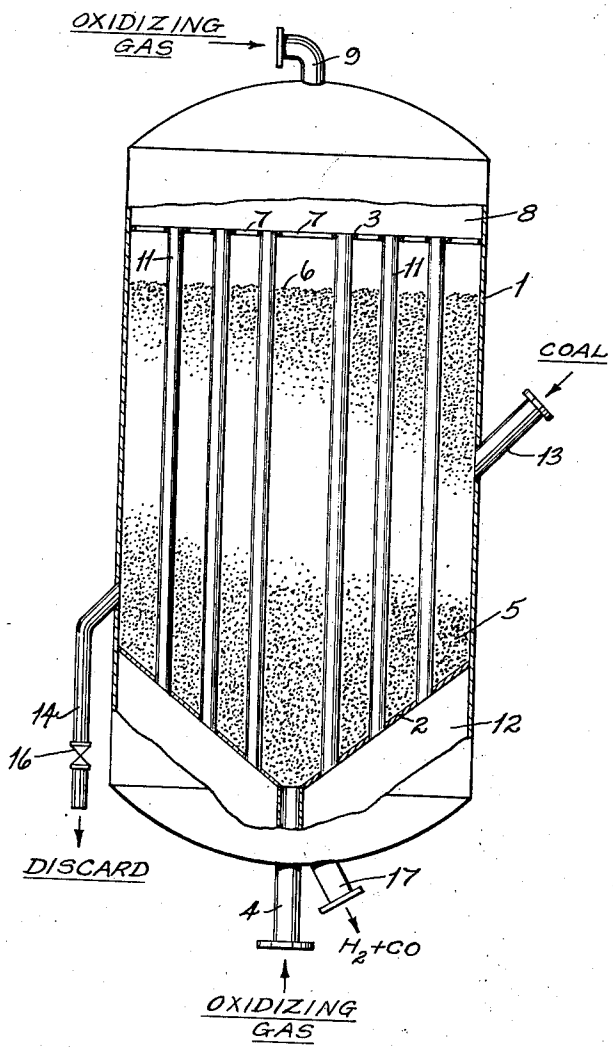
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PROCESS FOR THE PREPARATION OF CARBON MONOXIDE AND HYDROGEN

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PROCESS FOR THE PREPARATION OF CARBON MONOXIDE AND HYDROGEN

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This invention relates to a process for the production of carbon monoxide and hydrogen from solid carbonaceous materials. Coal, lignite, peat and oil shale are examples of suitable carbonaceous materials which may be converted to carbon monoxide and hydrogen in accordance with the present invention.

This invention involves subjecting a solid carbonaceous fuel containing volatilizable constituents, e. g., hydrocarbons, in particulate or granular form to contact in a gasification zone with an oxidizing gas comprising oxygen, preferably admixed with steam and/or carbon dioxide, under conditions suitable for conversion of combustible constituents of said fuel into a gas comprising a substantial amount of carbon monoxide and hydrogen and compounds containing both carbon and hydrogen atoms in the same molecule, including gaseous hydrocarbons. The resulting gaseous products, including the above-mentioned compounds, are immediately subjected to reforming by reaction with an oxidizing gas in a controlled amount and at an elevated temperature to convert the hydrocarbons to carbon monoxide and hydrogen. The gasification of the solid particles is conducted in a dense phase fluidized bed while the reforming of the hydrocarbon gases to carbon monoxide and hydrogen is conducted in an unpacked reaction zone in heat exchange relation with the fluidized bed of solid particles.

An important feature of the present invention is in the treating of the gaseous effluent from the gasification zone, which contains hydrocarbons in addition to carbon monoxide and hydrogen, in a reforming zone with oxygen to convert the constituents containing both hydrgen and carbon atoms in the same molecule into carbon monoxide and hydrogen. The process permits a wide latitude in control of the composition of the resulting product gas to produce a mixture of carbon monoxide and hydrogen in the desired proportions for the synthesis of hydrocarbons and oxygenated hydrocarbons. By conducting the gasification of the solid particles in heat exchange relation with the reforming of the gasification products, it is possible either to supply heat to the gasification zone from the reforming zone or to supply heat from the gasification zone to the reforming zone. If a product gas of high hydrogen-to-carbon monoxide ratio is desired, it is advantageous to supply heat to the gasification zone from the reforming zone. This permits gasification with a relatively large quantity of steam as compared with the amount of oxygen employed and permits conversion of a greater percentage of steam to hydrogen by the water gas

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reaction. On the other hand, if a product gas relatively rich in carbon monoxide is desired, it is advantageous to transfer heat from the fluidized bed gasification zone to the reforming zone. In this manner carbon dioxide may be reacted with hydrocarbons in the reforming zone to increase the production of carbon monoxide relative to hydrogen.

By-product and waste hydrocarbons, oxygenated hydrocarbons or the like, for instance, methane-containing tail gas produced in the synthesis of hydrocarbons, may also be converted in the reforming zone to additional carbon monoxide and hydrogen.

When coal, oil shale, lignite, or similar solid fuel is fed directly into the gasification zone in the present invention, tars, oils, and resinous compounds distilled from the solid fuel are converted in the reforming zone directly into carbon monoxide and hydrogen. The reforming step is also advantageous from the standpoint of converting sulphur compounds, particularly organic sulphur compounds, from the fuel into a form more readily removable from the synthesis gas.

The drawing is a more or less diagrammatic elevational view, partly in cross-section, of apparatus suitable for carrying out the process of the present invention. In describing a specific modification of the present invention in detail in connection with the accompanying drawing, reference will be made to the treatment of coal wherein the coal is fed in raw or uncarbonized form directly into the gasification zone of the apparatus. The coal is in crushed or divided form; all of the particles will usually pass through a 40-mesh screen and, preferably, about 40 to 50 per cent by weight of the particles will pass through a 200-mesh screen.

With reference to the drawing, the apparatus comprises a closed pressure-resistant vessel having an outer shell 1. The vessel is divided into sections by partition 2. Preferably the outer shell of the vessel is covered with suitable insulating material, not illustrated in the drawing, to conserve heat within the vessel. The partition 2 divides the vessel to provide a gasification zone 5 within the vessel intermediate and spaced from each end of the vessel. The partition 2 is preferably in the form of a frusto-conical section with the small end of the frustum nearest the lower end of vessel 1. A conduit 4 extends through the lower end of the vessel and is connected with partition 2. This provides even distribution of gases from conduit 4 through the solid material maintained within the vessel in gasification zone 5.

The particles of solid material undergoing gas-

ification are fluidized by gases flowing upwardly from conduit 4. The upper level 6 of the dense phase fluidized bed of particles is maintained somewhat below grille 3.

Grille 3 is provided with a series of openings 7 through which gases escape into the upper section 8 of the vessel. In the upper section 8, gaseous products from the gasification zone are admixed with an oxidizing gas stream introduced into the vessel through line 9. A plurality of vertical conduits 11 extending from grille 3 through partition 2 establish communication between upper end section 8 and section 12, the section of vessel 1 below partition 2. Reforming of the hydrocarbon constituents of the gas evolved in the gasification zone with the oxidizing gas takes place within sections 8 and 12 of the vessel and within conduits 11. Thus the gaseous reactants and reaction products within the reforming zone are in indirect heat exchange with the solids undergoing gasification in the gasification zone 5.

Fresh coal is introduced into the gasification zone 5 through conduit 13 while residue or char is withdrawn from the gasification zone through line 14 at a rate determined by valve 15, to maintain the desired level 6 of the fluidized bed within the gasification zone. Product gases from the reforming zone are withdrawn from section 12 through line 17. These gases consist essentially of hydrogen and carbon monoxide.

In operation, coal is continuously supplied to the gasification zone through conduit 13 while oxygen with or without steam and/or carbon dioxide is introduced into the reaction zone through conduit 4. The reactant gases may be introduced as a mixture and preheated to a temperature as high as about 1,000° F. Steam may be separately heated to higher temperatures. Sufficient oxygen is added to supply the amount of heat required to maintain a temperature within the gasification zone, preferably, within the range of from about 1,400° F. to about 2,000° F.

The gasifier may be operated at a pressure within the range of from about atmospheric to about 50 atmospheres or higher. A pressure within the range of from about 20 to about 30 atmospheres is usually desirable. The temperature in the reforming zone is maintained, preferably, within the range of from about 2,000° F. to about 2,500° F. The temperature of the gasification zone may be regulated by control of the rate of solids feed and the proportions of oxygen supplied to the gasifier through conduit 4 and the temperature within the reforming zone may be controlled similarly by controlling the proportions of oxygen supplied through conduit 9.

Since the mixture of hydrogen and carbon monoxide which is produced by the process of this invention is often required for utilization in a plant producing synthetic gasoline by the reaction of hydrogen and carbon monoxide in the presence of an iron catalyst or the like at elevated pressures of the order of 20 atmospheres and higher, it becomes desirable to generate the hydrogen and carbon monoxide mixture by the process of this invention at such elevated pressures in order to avoid the difficulties and expense of compressing the product gas from this process prior to its introduction into the catalytic synthesis reactor. In this connection it is well to note that the operation of the gasification zone in the process of this invention at elevated pressures of the order of 20 atmospheres and higher generally tends to effect hydrogenation and methane-forming reactions with the result that the presence of hydrocarbons in the gases emerging from the fluidized bed in the gasification zone is increased. Accordingly, the present invention is particularly valuable in converting the hydrocarbons present in the gaseous stream leaving the gasification zone to additional carbon monoxide and hydrogen when the gasification is being conducted at elevated pressures to supply a synthesis gas comprising essentially hydrogen and carbon monoxide to a synthesis reactor operated at elevated pressures.

Obviously, many modifications and variations of the invention as hereinbefore 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 the production of carbon monoxide and hydrogen from a solid carbonaceous fuel containing volatilizable hydrocarbonaceous constituents, the improvement which comprises subjecting said carbonaceous fuel in particle form to the action of an oxidizing gas in a dense phase fluidized bed gasification zone, discharging from said gasification zone gaseous products comprising compounds containing both carbon and hydrogen atoms in the same molecule, and admixing oxidizing gas with said gaseous products from the gasification zone in a reforming zone maintained at a temperature of at least about 2,000° F. wherein said compounds are converted into carbon monoxide and hydrogen while passing the gaseous mixture therein in indirect heat exchange relationship with the particles of carbonaceous fuel undergoing gasification in said gasification zone.

2. A process according to claim 1 wherein the temperature in the gasification zone is maintained within the range of from about 1,400° F. to about 2,000° F.

3. In a process for the production of carbon monoxide and hydrogen from a solid carbonaceous fuel containing volatilizable hydrocarbonaceous constituents, the improvement which comprises subjecting said carbonaceous fuel in particle form to the action of an oxidizing gas in a dense phase fluidized bed gasification zone, discharging from said gasification zone gaseous products comprising hydrocarbons, and passing said gaseous products in admixture with an oxidizing gas through a reforming zone extending through said dense phase fluidized bed gasification zone and in indirect heat exchange relation with the particles of carbonaceous fuel undergoing gasification in said gasification zone and maintained at a temperature within the range of from about 2,000° F. to about 2,500° F., thereby effecting conversion of said hydrocarbons into carbon monoxide and hydrogen.

4. A process as defined in claim 3 wherein said solid carbonaceous fuel is coal.

5. A process as defined in claim 3 wherein said oxidizing gas supplied to said gasification zone is a mixture of substantially pure oxygen and steam.

6. In a process for the production of carbon monoxide and hydrogen from a coal containing volatilizable constituents, the improvement which comprises subjecting said coal in particle form to the action of a mixture of steam and substantially pure oxygen in a dense phase fluidized bed gasification zone maintained at a temperature within the range of from about 1,400° F. to

about 2,000° F., separating the resulting gaseous products comprising hydrocarbons from the solid particles in the gasification zone, passing said products in admixture with substantially pure oxygen into a reforming zone maintained at a temperature within the range of from about 2,000° F. to about 2,500° F. in indirect heat exchange relation with the particles of coal undergoing gasification in said gasification zone, thereby effecting conversion of said hydrocarbons into carbon monoxide and hydrogen, and discharging the resulting mixture of carbon monoxide and hydrogen from said reforming zone.

7. A process as defined in claim 6 wherein the gasification and reforming zones are maintained at a pressure of at least 20 atmospheres.

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