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54 **Method of altering contaminants in a high-temperature, high-pressure raw synthesis gas stream.**

57 A process for the partial combustion of finely divided coal or related fuels at high temperatures to make synthesis gas containing sticky particles of fly ash and vaporized contaminants which is mixed with a much cooler quench gas under conditions of turbulent flow in an elongated straight quenching chamber of selected dimensions so that the fly ash particles are no longer sticky and the vaporized contaminants have been condensed so as to pass readily with the gas through a downstream heat exchanger.

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## METHOD OF ALTERING CONTAMINANTS IN A HIGH-TEMPERATURE, HIGH-PRESSURE RAW SYNTHESIS GAS STREAM

The invention relates to a process for the partial combustion of finely-divided solid fuel, such as pulverized coal, in which the latter is introduced together with oxygen-containing gas via a burner into a reactor or gasifier from which a stream of high-temperature raw synthesis gas is discharged together with a minor amount of contaminating material, some of which may be in the form of particles having a sticky outer surface, that tend to adhere to equipment located downstream of the reactor.

Partial combustion is the reaction of all of the fuel particles with a substoichiometrical amount of oxygen, either introduced in pure form or admixed with other gases, such as a transport stream of nitrogen, whereby the fuel is partially oxidized to hydrogen and carbon monoxide. This partial combustion differs from complete combustion wherein the fuel would be completely oxidized to carbon dioxide and water.

During the process of partial combustion of pulverized coal in a gasifier, the mineral matter in the coal splits into two streams when the coal is gasified. Molten slag which is formed falls to the bottom of the gasifier where it is discharged. Lightweight particles of fly ash or fly slag which are also formed are carried out through the top of the gasifier by the stream of synthesis gas which is piped to a gas cooler, heat exchanger or waste heat boiler where steam may be generated.

During discharge of the product synthesis gas from the gasifier, a problem arises that the sticky fly ash particles deposit on the walls of the outlet duct from the gasifier where they solidify and tend to clog the duct. Additionally, in some cases the sticky fly ash particles deposit on fins or tube bundles of the cooler and reduce the efficiency of the cooler. In either case, the process must be interrupted to clean equipment periodically, which action is unacceptable for the desired continuous operation of the process.

In an attempt to solve the foregoing problem, it has been previously proposed forming a gas shield on the inner wall of outlet duct from the gasifier to protect the inner wall from sticky particles adhering thereto. The product gas stream passing through the outlet duct to the cooler would be surrounded throughout the length of the duct with a cooler gas sheath. Thus, sticky particles in the gas flow stream near the duct wall could then be cooled when they encountered the surrounding cool gas sheath to such an extent as to cause the ash particles to solidify and lose their stickiness before they hit a wall. According to that proposal, the

protective sheath of cooler gas was to be introduced via an annular slit at the upstream end of the outlet duct from the gasifier. It was found, however, that in certain cases the shield or sheath of cooling gas was prematurely disturbed or disrupted in a manner so as to be unsatisfactory in operation of cooling sticky fly ash particles.

The present invention is directed to a process for the partial combustion of finely divided carbonaceous fuel containing at least 1% by weight ash in a reactor or gasifier to produce a product gas (mainly carbon monoxide and hydrogen) which carries along with it, as it leaves the reactor, sticky particles of fly ash or fly slag, or ash-forming constituents which may consist of alkali metal chlorides, silicon and/or aluminium oxides or other mineral species. At the temperature prevailing in the reactor, the ash is usually sticky. In particular, when the partial combustion takes place by entrained gasification in the burner flames, the residence time in the gasifier or reactor is very short compared with gasification in a fluidized or moving bed process, and the temperature is very high.

The ash that is formed during the present gasification process is at least partly in liquid form at the conditions that prevail in the reactor, usually temperatures from 1050 to 2200 °C. If the ash particles are not fully in liquid form, they will generally consist at least partly of a molten slag or be a combustion product or residue having a partly molten consistency. The high temperature of a reactor is sufficient to vaporize certain other by-products which may assume a sticky form when cooled in the process equipment.

It is an object of the present invention to solidify at least the outer surface of particles which are normally sticky when they are contained in a synthesis gas effluent flow stream leaving a reactor at temperatures above 1200 °C.

The invention therefore provides a process for the production of synthesis gas wherein coal is partially oxidized at an elevated temperature by feeding substantially dry particulate coal and oxygen to a gasification zone of a reactor, the ratio of coal to oxygen being such as to maintain a reducing atmosphere, and producing raw synthesis gas, said gas also carrying with it a minor amount of at least one contaminating impurity having a sticky outer surface when in a solid particle form which tends to adhere to equipment downstream of the reactor, characterized by

- the step of reducing the sticky characteristic of sticky particles of contaminants carried by the effluent stream of synthesis gas from the reactor by

injecting a flow of lower temperature quench gas into the higher temperature synthesis gas stream substantially immediately upon leaving the reactor to cool the sticky contaminating particles; and  
 - flowing the combined mixture of synthesis gas containing contaminating sticky particles and injected cooling quench gas through a substantially straight conduit under conditions of turbulent flow for a time sufficient to thoroughly mix the combined flowing stream to reduce the sticky characteristic of at least the outer surface of the particles to a form that does not interfere with downstream equipment.

In accordance with the present invention it is desired to provide a long, straight quench section of pipe which forms the first section of the discharge duct from the reactor. The temperature of the product gas at this point may be, say 1400 °C for example. A stream of product gas, which has been cooled several hundred degrees, is recycled from a selected point in the process and injected as a quench gas into the upstream end of the quench section of the reactor discharge duct. By mixing the cool quench gas with the hot reactor effluent as it enters the quench section, and flowing the mixture through a straight quench section of sufficient length and under conditions of turbulent flow, the hot synthesis product gas and the sticky particles carried thereby are thoroughly mixed with the cooler quench gas thereby allowing the molten or sticky particles to "freeze" or at least cause the outer surfaces of the particles to become non-sticky to the extent that they do not stick to the walls of any downstream equipment or piping.

Accordingly, the invention is designed for use in a synthesis gas generation complex comprising

a) a coal gasification plant, including at least one gasifier or reactor for the gasification of coal to produce synthesis gas at a temperature of 1050 °C to about 2200 °C, the gasifier having heat exchange surfaces adapted for indirect heat exchange with steam and water and adapted to utilize dry particulate coal which is mixed with oxygen;

b) a long straight cooling or quench section or conduit mounted at the gas discharge port of the gasifier and in flow communication therewith whereby a quenching gas of lower temperature may be injected into and mixed under conditions of turbulent flow with the hot effluent synthesis gas and the particles carried thereby;

c) a heat exchange section comprising at least one heat exchanger in gas flow communication with said gasifier, said heat exchanger comprising at least one segment adapted to further cool the gas and the particulates carried thereby;

d) a gas cleanup section in flow communication with said heat exchanger, said cleanup section comprising means for removing substantially all of

the particulates and various impurity gases such as H<sub>2</sub>S and other contaminants from said synthesis gas; and

e) a source of quenching gas at reduced temperature and reduced particle content for recycling back to the quench section.

In particular the invention provides a process for the production of synthesis gas comprising the steps of

a) partially oxidizing coal at an elevated temperature by feeding dry particulate coal and oxygen to a gasification zone, the gasification zone preferably comprising at least one burner for oxidizing the coal, the ratio of coal to oxygen being such as to maintain a reducing atmosphere, and producing raw synthesis gas having a temperature of from about 1050 °C to about 2200 °C, and removing heat from said synthesis gas in said gasification zone by indirect heat exchange with steam and water at a temperature of from about 100 °C to about 350 °C;

b) passing raw synthesis gas and the sticky particles carried thereby through a long straight quench chamber formed at the upstream end of the discharge duct from said gasification zone;

c) injecting a cooling quenching gas into said quench chamber and mixing the cooling quenching gas with the hot synthesis gas under conditions of turbulent flow to change at least the outer surfaces of the sticky particles to a non-sticky state;

d) passing raw synthesis gas from step c) to a heat exchange zone of any suitable form well known to the art, and removing heat from said synthesis gas and the particulate material carried thereby; and

e) removing particulates from said raw synthesis gas, producing a synthesis gas substantially free of particles, a portion of the gas being adapted to be recycled back to and injected into the quench chamber.

In an advantageous embodiment of the invention the raw synthesis gas also contains minor amounts of vaporized mineral matter and said reducing step includes condensing the vaporized mineral matter by the injection of a volume of quench gas at a lower temperature than the synthesis gas stream into which it is injected.

In another advantageous embodiment of the invention the quench gas stream is injected into the raw synthesis gas stream at a plurality of points around the periphery thereof so as to form at said gas injection area a sheath of cooler quench gas around the hot stream of synthesis gas and the sticky particles carried thereby.

In still another advantageous embodiment of the invention the quench gas is synthesis gas taken from a point downstream which contains a

minor amount of lower temperature solid particles of the contaminating impurity, said particles being of the same composition as at least one of the impurities in the effluent raw synthesis gas stream leaving the reactor.

The steam generated in the gasifier shell may be passed to a heat exchange zone where it may be superheated and then sent for utilization. The gasification may be carried out utilizing techniques suitable for producing a synthesis gas having a gasifier outlet temperature of from about 1050 °C to about 1650 °C.

Although some fluidized bed oxidizers are capable of producing such gas temperatures under the conditions mentioned herein, the process is advantageously carried out with a gasifier comprising at least one burner. Such a process will advantageously include combustion, with oxygen, of dry particulate coal, i.e., coal having less than about 10 per cent water content. Steam may be added in some instances to assist in the combustion. The type of coal utilized is not critical, but it is an advantage of the invention that lower grade coals, such as lignite or brown coal, may be used. If the water content of the coal is too high to meet the requirements mentioned, supra, the coal should be dried before use. The atmosphere will be maintained reducing by the regulation of the weight ratio of the oxygen to moisture and ash free coal in the range of about 0.6 to 1.2, in particular 0.9 to 1.0. The specific details of the equipment and procedures employed form no part of the invention, but those described in U.S. patent specification No. 4,350,103, and U.S. patent specification No. 4,458,607, may be employed. In view of the high temperatures required, however, suitable structural materials, such as the Inconels and Incoloy 800, i.e., high chrome-molybdenum steels, should be employed for superheating duty for long exchanger life. It is an advantage of the invention that, by carrying out the advantageous procedure described herein, or utilizing the advantageous structural aspects mentioned, as described, a synthesis gas stream is produced free of particles of sticky material that might adhere to and/or clog flow lines or equipment.

The particular types of equipment employed, within the limitations mentioned, are not critical. The key to the invention is, as mentioned, the judicious integration of a particular type of coal gasification technique or practice with operations or structure to achieve clean flowlines and equipment and improved energy efficiency. Pressures employed are not critical, those skilled in the art being capable, given the temperatures specified, of providing suitable pressure levels in the various units of the invention.

The invention will now be described by way of

example in more detail with reference to the accompanying drawing wherein

- Fig. 1 is a schematic flow diagram of one portion of the process equipment to be used to carry out the method of the present invention; and

- Fig. 2 is a schematic diagram of an alternative arrangement of equipment to be used in carrying out the method of the present invention.

The drawing is a schematic representation of the process flow type, and illustrates the efficient integration of the specialized gasifier with equipment for substantially eliminating the particles of sticky material that are produced in a gasifier. All values specified in the description relating thereto hereinafter are calculated, or merely illustrative.

Referring now to Fig. 1, dry particulate coal (average particle size about 30 to 50 microns and moisture content of less than about 10 per cent by weight) is fed via a line (1) to burners (2) of a gasifier (3). For reasons of clarity only one burner (2) has been represented. The gasifier (3) may be a vertical oblong vessel, for example cylindrical in the burner area, with substantially conical or convex upper and lower ends, and is defined by a surrounding membrane wall structure (4) for circulation of cooling fluid. Advantageously, the generally cylindrical reactor wall will comprise a plurality of heat exchange tubes, spaced apart from each other by "membranes" or curved plates, the tubes being connected at their extremities for continuous flow of a heat exchange fluid, such as water, and also having multiple inlets/outlets for the fluid, in a manner well known to the art. Concomitantly, oxygen is introduced to the burners (2) via a line (5), the weight ratio of oxygen to moisture and ash free coal being about 0.9, for example. The combustion produces a flame temperature of about 2200 °C, with a gas temperature at the outlet of the gasifier being about 1250 °C to about 1450 °C. Regulation of gasifier and outlet temperature is assisted by coolant in the membrane wall structure (4). Slag is discharged at the outlet (1a).

Hot raw synthesis gas with impurities leaves the gasifier (3) through a straight elongated quench line (8) of selected length the interior of which forms a quench chamber in which the raw synthesis gas and the impurities carried thereby are quenched by cooler synthesis gas through line (6) from any suitable point in the process. The quench gas may be from 140 °C to about 540 °C. The quench line (8) may also be jacketed for heat recovery, although this is not illustrated. The quenched gas then passes to a cooler or heat exchanger (7). The heat exchanger (7) is advantageously a multiple section exchanger, the quenched synthesis gas being cooled by fluid in the tubes.

The raw synthesis gas, now cooled in the low

temperature section of the heat exchanger (7) to a temperature of about 315 °C to 140 °C, passes via a line (14) to a cleanup section (15) or solids separator where particulates and various impurity gases, such as H<sub>2</sub>S, may be removed. The details of the gas cleanup form no part of the invention. Steam requirements for cleanup activities may be supplied from that generated by the overall process. The purified synthesis gas passes from the section (15) in the line (17), and is ready for use. Dry solids impurities are discharged at the outlet (17a).

In Fig. 2 the same reference numerals as used in Fig. 1 are applied. Instead of the quench line (8) of Fig. 1 running horizontally from the gasifier (3), the quench line (8) from the gasifier (3) in Fig. 2 is illustrated as extending vertically from the top of the gasifier for a calculated distance. The length of the quench line (8) depends on many factors such as flow rates or volumes in the quench lines and the recycled quench gas supply lines, the temperatures of the raw synthesis gas from the gasifier and that of the quench gas, the nature of the vaporized impurities in the raw synthesis gas to be condensed, the nature of the sticky ash particles in the synthesis gas to be rendered non-sticky, etc.

Tests were conducted in a pilot operation in which 200 tons per day of finely-divided coal were burned with an equal weight of oxygen. The gasifier in one test was operated at about 25 bar (10 to 100 bar range) with the product gas being discharged from the gasifier with contaminants at about 1450 °C. After being cleaned and cooled, a portion of the product gas flow stream at about 315 °C was recycled and injected as a quench gas into the upstream end of the quench section which forms substantially the first section of gas discharge line from the gasifier.

Equal masses of product gas and quench gas may be used although this ratio may be varied with from 0.5 to 4.0 mass of quench gas being used for each mass of product gas depending on operating conditions. To achieve adequate cooling and turbulent flow in the quench chamber, it is necessary for the length of the quench line to be between 5 and 20 times the inner diameter of the line. It is advantageous that this minimum length of quench line have no bends in it until the particles have passed a point in the line where they are no longer sticky. At the high operating temperature of the gasifier, certain impurities such as the lower oxides of silica and many of the alkali metals are vaporized. The materials are then condensed and cooled in the quench section of the process equipment. A quench section having a straight run of 14 times the section diameter handled a 9 metres per sec flow of hot synthesis gas in a system burning 200 tons of coal per day.

The remaining equipment shown in Fig. 2 are similar to the components of Fig. 1. Fig. 2 is included to show that the quench line (8) may leave the gasifier in a vertical direction and enter the heat exchanger (7) from the top, if desired.

## Claims

1. A process for the production of synthesis gas wherein coal is partially oxidized at an elevated temperature by feeding substantially dry particulate coal and oxygen to a gasification zone of a reactor, the ratio of coal to oxygen being such as to maintain a reducing atmosphere, and producing raw synthesis gas, said gas also carrying with it a minor amount of at least one contaminating impurity having a sticky outer surface when in a solid particle form which tends to adhere to equipment downstream of the reactor, characterized by

- the step of reducing the sticky characteristic of sticky particles of contaminants carried by the effluent stream of synthesis gas from the reactor by injecting a flow of lower temperature quench gas into the higher temperature synthesis gas stream substantially immediately upon leaving the reactor to cool the sticky contaminating particles; and
- flowing the combined mixture of synthesis gas containing contaminating sticky particles and injected cooling quench gas through a substantially straight conduit under conditions of turbulent flow for a time sufficient to thoroughly mix the combined flowing stream to reduce the sticky characteristic of at least the outer surface of the particles to a form that does not interfere with downstream equipment.

2. The process as claimed in claim 1 characterized in that the raw synthesis gas also contains minor amounts of vaporized mineral matter and said reducing step includes condensing the vaporized mineral matter by the injection of a volume of quench gas at a lower temperature than the synthesis gas stream into which it is injected.

3. The process as claimed in claims 1 or 2 characterized in that the quench gas is low temperature product gas free of water and particles which was made by the present process.

4. The process as claimed in any one of claims 1-3 characterized in that the quench gas stream is injected into the raw synthesis gas stream at a plurality of points around the periphery thereof so as to form at said gas injection area a sheath of cooler quench gas around the hot stream of synthesis gas and the sticky particles carried thereby.

5. The process as claimed in any one of claims 1-4 characterized in that the mass of quench gas injected into the raw synthesis gas stream is from 0.5 to 4 times the mass of the synthesis gas.

6. The process as claimed in claims 1-5 characterized in that the length of the straight section of conduit downstream of the quench gas injection point is from 5 to 20 times the diameter of the conduit.

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7. The process as claimed in any one of claims 1-6 characterized in that the quench gas is synthesis gas taken from a point downstream which contains a minor amount of lower temperature solid particles of the contaminating impurity, said particles being of the same composition as at least one of the impurities in the effluent raw synthesis gas stream leaving the reactor.

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