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Description

The invention relates to a process for the gasification of coal to produce synthesis gas comprising the steps of feeding particulate coal to a gasification zone, and partially oxidizing coal with oxygen in said gasification zone, producing a stream comprising synthesis gas and flyash, and producing bottom impurity solid materials which are collected in a water bath as char, fine slag, and coarse slag; quenching and cooling the said produced stream, and separating flyash from the said stream, producing a separated flyash; removing a material comprising char from said water bath; fluidizing removed char in a fluidized bed in a fluidization zone, and oxidizing said char in said fluidized bed with oxygen at a temperature below the fusion temperature of the mineral components of the char and the fluidized bed, producing a substantially carbon-free residue and heated gases.

Usually in processes for the gasification of coal to produce synthesis gas, significant quantities of unreacted impurity matter, variously referred to as char and slag, and produced during combustion or oxidation in the gasification zone. Upon separation of the char and/or slag from the gasification zone, a disposal problem exists, since these materials have undesirable properties which limit options for their disposal. At the same time, the char, and at least some of the fine slag, contains, in addition to mineral matter or components, a residual carbon content that has significant energy value. For example, the char and fine slag may contain from about 5 per cent to about 90 per cent by weight carbon, based on the total weight of the char and fine slag. If the char, or char and fine slag are disposed of, e.g., as land fill, this energy value is lost. Accordingly, a procedure which captured this energy value, at the same time reducing the volume of material to be disposed of, could have economic importance. The invention provides such a procedure.

Additionally, if coal is dried to remove the bulk of the moisture therein before gasification, the removal of this moisture involves a significant energy expenditure. If coal is combusted to provide the heat necessary for the drying, from about 2 per cent to about 12 per cent by weight of the total coal supplied to the process may be utilized, thus lowering the efficiency of the process. The invention, in a further embodiment, is directed to reducing this energy expenditure.

A process for gasification of coal characterized by utilization of heat values obtained from flyash, with concomitant reduction of material for disposal has already been proposed in the related European patent application No. 0227196. The present invention is complementary thereto, such that a common

fluidized bed oxidation zone for heat energy recovery may be employed.

Therefore, it is an object of the invention to provide a process for the partial oxidation or combustion of coal in which disposal problems for the char and fine slag are reduced, and energy costs for the process are reduced. The invention, further, combines a procedure for reducing char and fine slag disposal with a coal drying procedure, so that improvement in both techniques is achieved.

Therefore, the process of the invention is characterized in that the said removed char is oxidized at a temperature from 500°C to 1150°C and a pressure of about atmospheric or higher in the said fluidized bed concomitantly with at least part of the said separated flyash, which is injected into the said fluidized bed and oxidized, and utilizing outside the gasification zone the heat thus recovered to dry the coal to be fed to the gasification zone.

It is remarked that US-A-4,508,542 discloses a slag separator for a coal gasification installation. This separator permits the classification of slag particles by decantation in a water bath, the precise selection of the desired grain of slag affording an advantage for the slag recycling. Further, US-A-2,633,416 discloses the gasification of carbonaceous solids wherein the slag is burned in a waste heat boiler by oxidation in a fluidized bed for recovering its energy as heating gas for drying the most coal before use.

However, there is no indication at all in US-A-2,633,416 or US-A-4,508,542 that separated flyash from the synthesis gas can be injected into a fluidized bed wherein the char removed from the gasification zone, is oxidized.

In an advantageous form the present invention comprises a process, as described, in which the heat in said heated gases is utilized by contacting moist feed coal with a solid or gas heated directly or indirectly by said heated gases.

Pressures in the gasification zone may be varied widely, but will advantageously range from 10 to 100 atmospheres, more advantageously from 20 to 50 atmospheres, while pressures in the fluidization zone will range from atmospheric to 10 atmospheres or higher. A rough separation of char and fine slag from coarse slag may be made in the gasification zone, or char and fine slag and all or a portion of the coarse slag may both be sent to the fluidization zone. As used herein, the term "char" refers to those materials, whether solid or molten, which remain in the gasifier upon production of synthesis gas, and have an average particle size (on solidification, if molten) of less than 250 microns in width. "Fine slag" refers to those materials, whether solid or molten, which remain in the gasifier upon production of synthesis gas, and have an average particle size (on solidification, if molten)

of greater than 250 microns, but less than 1 millimetre in size. As used herein, the term "coarse slag" refers to those materials which remain in the gasifier upon production of synthesis gas, and which have an average particle size upon solidification of greater than 1 millimetre in width. As indicated, char and fine slag contain varying portions of residual carbon, e.g., from 5 per cent by weight to 90 per cent by weight of residual carbon, based on the total weight of the char or fine slag, while coarse slag, which is generally a vitreous type of material, contains less than 1 per cent by weight of residual carbon.

The heat liberated is used to dry feed coal to the process. That is, the invention relates to a process for the gasification of coal to produce synthesis gas in which particulate coal is dried in at least one drying zone, the coal is fed from at least one drying zone to a gasification zone, and partially oxidized or combusted, as hereinbefore described, to produce synthesis gas, char, fine slag, and coarse slag, which is treated as described, and the heat generated by oxidizing the char or char and fine slag and at least part of the said separated flyash is utilized in drying coal in at least one drying zone. As used herein, the term "oxygen", unless otherwise evident from the context where employed, is taken herein to include air, pure oxygen, air enriched with oxygen, and other oxygen-containing gases.

While the particular gasification or partial oxidation or combustion procedure employed is not per se part of the invention, the invention is particularly applicable to high pressure coal gasification slagging processes in which the coal is partially oxidized or combusted to produce, in addition to the synthesis gas, flyash, a bottom "char" material, fine slag and a vitreous or coarse slag which are collected in water bath in or associated with the gasification zone. The oxygen and coal flows are controlled so that little CO_2 is produced in the gaseous product. In general, these procedures are carried out by feeding coal, advantageously dry coal, e.g., coal having a moisture content of below about ten per cent by weight, entrained in a gas, such as an inert gas, into a gasification zone, and combusting or oxidizing the coal with oxygen. Flame temperatures may reach 3500°C , with zone outlet temperatures advantageously from 1100°C to 1800°C . Pressures in these processes will range from 10 atmospheres to 100 atmospheres, advantageously from 20 atmospheres to 50 atmospheres. In one such process, the synthesis gas and flyash are removed from the upper portion of the gasification zone, and the char, fine slag, and coarse slag are collected from the lower portion in a water bath.

A wide variety of coals are suitable for use in

the invention. For example, anthracite, bituminous coal, lignite, and so-called brown coal may be used in the invention. A real advantage of the invention is the ability to use lower grade coals, such as lignite. As used herein, the term "coal" is taken to include these inferior grade carbonaceous fuels, as well as the higher quality coals. The coal will advantageously be fed to the gasification zone in a particle size suitable for boiler furnace operation, e.g., 80 to 90 microns in diameter, although those skilled in the art may select the appropriate particle size, as desired. In an advantageous embodiment, the invention is practised with "dry" coal, that is, coal having a moisture content of less than 10 per cent by weight, based on the weight of the moist coal.

Synthesis gas and entrained matter, which is known as, or becomes what is known as, flyash, is removed from the gasification zone and treated as described therein. Heavier solid particulate matter and molten matter remaining in the gasification zone are allowed to fall into the lower part of the reactor, and thence for example into a water bath, and are collected in the lower portion of the gasification zone. As those skilled in the art will recognize, a significant distinction between "flyash" and "char" is the density. Fine slag is somewhat denser, while coarse or vitreous slag has, as noted, little carbon content, and has a "glassy" appearance.

The char and fine slag may be separated roughly in the water bath by this density differential, although there will be some intermixture. That is, the coarse slag particles tend to fall to the bottom of the water bath, the fine slag above the coarse slag, and the char generally above the fine slag. Means, e.g., a sluice, may be provided for achieving the separation of the char, and fine slag, if desired, from the coarse slag. Alternately, all material may be removed, either intermittently or continuously, from the bottom or lower portion of the gasification zone water bath. It is thus not a requirement of the invention that the particles in question be separated in the gasification zone water bath; the portions may be separated by sizing equipment external to the bath. Moreover, it is within the concept of the invention that all of the "bottoms" material, i.e., char, fine slag, and coarse slag, be sent to the fluidization zone. If desired, the bottoms material may be crushed to ensure the desired size for fluidization.

According to the invention, at least the char, and advantageously the fine slag, are oxidized concomitantly with at least part of said separated flyash in a fluidization zone comprising at least one fluidized bed to remove the carbonaceous matter therein, producing a denser, more easily disposal of material. The heat liberated during oxidation is

used either directly or indirectly to dry the coal to be fed to the gasifier. As will be appreciated by those skilled in the art, the amount of heat generated from the bottoms material will be insufficient to dry the large volume of coal needed for the process. The bottoms material is combined, as mentioned, with at least part of the said separated flyash, in the manner provided in the above-mentioned application. To supplement the heat generated by the bottoms material and flyash, other combustible or partially combustible materials or fuels, such as coal or coal fines, may be added to the fluidized bed and oxidized. Obviously, a separate drying technique may also be used, but it is an advantage of the invention that an independent dryer or drying zone may be eliminated.

The type of fluidized bed or beds employed is a matter of choice. What is required, however, is that the bed or beds be operated at a temperature below the fusion temperature of the mineral matter in the bed, including that of a coal or fuel supplement. In general, the mineral content of flyash comprises silica, alumina, and other inorganic components in varying quantities, and the melting point of the mixture may be determined routinely. Since these temperatures are normally above 1482 °C (although some may be less), problems with clogging will normally not be encountered if the bed is operated below this temperature. As indicated, the fusion temperature or range of temperatures may be determined routinely for each flyash or fuel utilized, and the temperature in the fluidized bed may be controlled accordingly. Advantageously, the temperatures are operated well below the fusion temperature, e.g., from 500 °C to 1150 °C, more advantageously from about 650 °C to about 1120 °C. Pressures in the fluidization zone may be varied to the extent suitable for fluidized beds, but may be ranged from atmospheric to 10 atmospheres, for example from atmospheric pressure to 5 atmospheres. The fluidized bed or beds may be jacketed, or may have coils to absorb the heat produced by the oxidation. The bed may contain other particulate matter, including catalysts. The combustion gases generated are removed from the fluidization zone, and transfer their heat, either directly or indirectly, as desired, advantageously to the coal feed. The coal can be ground and dried in a combination procedure, and to allow conventional equipment to be used, a moderating gas, such as air, or nitrogen, at a temperature of, for example, 15 °C to 40 °C, is added to the combustion gases to lower the temperature to 200 °C to 500 °C.

The invention will now be described in more detail by way of example with reference to the accompanying drawing, in which the figure is a schematic illustration of the process flow type. All

values are calculated or exemplary.

Accordingly, reference numeral (1) designates a supply line from, for example, a storage vessel, not shown, in which coal having an average particle size diameter of 1.3 cm is fed to a drying zone (2) which contains a combination pulverizer-dryer wherein the coal is crushed to an average particle size of from 80 to 90 microns and dried by the gas stream in a line (3) to a moisture content of 10 per cent by weight, based on the weight of the moisture and the coal. In this illustration, the gas stream in the line (3) has a temperature of 250 °C, contacts the coal directly in the combination pulverizer-dryer, and exits the drying zone (2) via a line (4). The exit gas may be treated for control of pollutants, or utilized (not shown). Concomitantly, dried particulate coal is removed from the drying zone (2) through a line (5) and forwarded to a gasifier (6). Means may be provided (not illustrated), for raising the pressure of the coal and entraining gas up to the level employed in the gasification zone. In the gasification zone (6), the coal is injected in an entraining gas, e.g. nitrogen, through nozzles and combusted in a reducing atmosphere or partially oxidized with pure oxygen at 25 atmospheres and at a flame temperature of 3400 °C to 3500 °C. Synthesis gas and impurity particles are removed overhead from the gasification zone at a temperature of 1400 °C, and sent to a quenching and cooling zone (7). Char and slag, which are heavier impurity materials, fall downward into the gasifier, and are removed from the lower portion thereof. The quench and cooling zone (7) can be connected directly to the outlet of the gasifier (6). The synthesis gas can be first quenched and cooled with cold recycle gas. The temperature of the synthesis gas can be lowered to 900 °C, and molten impurities in the gas are solidified to what is known as flyash. The quenching and cooling sequence can be carried out in more than one stage, the final temperature before separation of the flyash being from 235 °C to 320 °C.

In order to separate the flyash from the synthesis gas, the stream is forwarded via a line (8) to a cyclone (9) (or the cooling zone may discharge directly thereto). The great bulk of the flyash is separated in the cyclone (9), and the flyash is removed from the cyclone (9) via a line (10). The gas stream is removed from the cyclone (9) via a line (11) and sent for further processing, such as for H₂S removal, and for product use. Because the cyclone is not totally effective, a final solids cleanup stage (12), e.g., bag filters, is provided in the line (11). Solids are removed from a unit (12) via a line (13). Solids from the cyclone (9) and the solids removal stage (12) is utilized for energy recovery according to the invention.

The impurity molten materials and solids not removed with the synthesis gas from the reaction zone fall to the lower portion of the gasifier (6), and through a slag tap (20) into a water bath (21). Primarily because of differences in the density of the particles, a rough separation of the particles takes place, as mentioned previously. A sluice (22) is provided for removal of fine slag and char, and coarse slag may be removed from the bottom of the bath at a line (23). The material separated in the sluice (22) is passed via a line (24) to a fluidization zone (14), wherein it is treated, as described hereinafter.

Accordingly, solids in the lines (10), (13) and (24) are injected into the fluidized bed (14), and are reacted with oxygen in excess from a line (15) at a temperature of 800 °C to 900 °C, which is well below the fusion temperature of the mineral components of the bed. The oxygen may serve as the fluidizing gas, or a separate fluidizing gas may be provided. The substantially carbon free residue may be removed via a line (16). Residence times of the solids will depend on the operating conditions, such as the temperatures, pressures, and specific equipment, and may be adjusted suitably by those skilled in the art. The combustion gas is removed from the bed (14) via the line (3), and is utilized as described previously. As noted previously, the total carbon present in the bottoms solids is insufficient to provide the required heat for drying the coal to the process. The bottoms material is combined with at least part of the separated flyash. To supplement the heat requirements, additional carbon-containing materials, such as coal fines, may be added to the fluidized bed (14) via a line (25).

In order to determine if the bulk density of a sample of fine slag and char remaining from the gasification of coal might be increased, the sample was heated in a quartz dish for at least one hour in air at 750 °C in a muffle furnace. The sample weighed 69.11 grams and had a volume of about 82 ml. After the heat treatment, the sample weighed 61.2 grams, had a volume of about 55 ml. The loss on ignition was 11.4 per cent by weight, based on the weight of the original sample.

While the invention has been illustrated with particular apparatus, those skilled in the art will recognize that, except where indicated as material herein, other equivalent or analogous equipment may be employed. For example, crushing and drying equipment may be separate, and the gasifier need not have the specific configuration shown. Means other than a sluice may be provided for fine slag and char removal.

Again, as used herein, the term "zone", as employed in the specification and claims, includes, where suitable, the use of segmented equipment

operated in series, or the division of one unit into multiple units to improve efficiency or overcome size constraints, etc. For example, a series of cyclones may be employed, and the quench and cooling operations are preferably carried out in multiple units, utilizing different techniques. Parallel operation of units, is, of course, well within the scope of the invention, and all equipment, such as pumps, valves, control units, etc. has not been illustrated.

Claims

1. A process for the gasification of coal to produce synthesis gas comprising the steps of feeding particulate coal to a gasification zone, and partially oxidizing coal with oxygen in said gasification zone, producing a stream comprising synthesis gas and flyash, and producing bottom impurity solid materials which are collected in a water bath as char, fine slag, and coarse slag; quenching and cooling the said produced stream, and separating flyash from the said stream, producing a separated flyash; removing a material comprising char from said water bath; fluidizing removed char in a fluidized bed in a fluidization zone, and oxidizing said char in said fluidized bed with oxygen at a temperature below the fusion temperature of the mineral components of the char and the fluidized bed, producing a substantially carbon-free residue and heated gases; characterized in that the said removed char is oxidized at a temperature from 500 °C to 1150 °C and a pressure of about atmospheric or higher in the said fluidized bed concomitantly with at least part of the said separated flyash, which is injected into the said fluidized bed and oxidized, and utilizing outside the gasification zone the heat thus recovered to dry the coal to be fed to the gasification zone.
2. The process as claimed in claim 1 characterized in that the heat in said heated gases is utilized by contacting moist feed coal with a solid or gas heated directly or indirectly by said heated gases.

Revendications

1. Procédé de gazéification de charbon pour produire un gaz de synthèse comprenant les étapes:

- d'alimentation de particules de charbon dans une zone de gazéification, et d'oxydation partielle du charbon avec de l'oxygène dans ladite zone de gazéification, produisant un flux comprenant du gaz de synthèse et des cendres volantes, et produisant des impuretés de matières solides au fond qui sont collectées dans un bain d'eau telles que du produit de carbonisation, des scories fines ou grossières; 5
- de trempe et refroidissement dudit flux produit, et de séparation des cendres volantes dudit flux, produisant des cendres volantes séparées; 10
- d'élimination de la matière comprenant le produit de carbonisation à partir dudit bain d'eau; 15
- de fluidisation du produit de carbonisation éliminé dans un lit fluidisé dans une zone de fluidisation, et oxydation dudit produit de carbonisation dans ledit lit fluidisé avec de l'oxygène à une température inférieure à la température de fusion des composants minéraux du produit de carbonisation et du lit fluidisé, produisant un résidu pratiquement dépourvu de carbone et des gaz chauffés; caractérisé en ce que ledit produit de carbonisation éliminé est oxydé à une température de 500 °C à 1150 °C et une pression d'environ la pression atmosphérique ou supérieure dans ledit lit fluidisé concomitamment avec au moins une partie desdites cendres volantes séparées, qui est injectée dans ledit lit fluidisé et oxydée, et on utilise à l'extérieur de la zone de gazéification la chaleur ainsi récupérée pour sécher le charbon destiné à alimenter la zone de gazéification. 20 25 30 35 40

2. Procédé selon la revendication 1, caractérisé en ce que la chaleur des gaz chauffés est utilisée en mettant en contact le charbon d'alimentation humide avec un solide ou gaz chauffé directement ou indirectement par lesdits gaz chauffés. 45

Ansprüche 50

1. Ein Verfahren zur Vergasung von Kohle zwecks Herstellung eines Synthesegases, umfassend die folgenden Verfahrensstufen: 55
- Einspeisen von teilchenförmiger Kohle in eine Vergasungszone und Teiloxidation der Kohle mit Sauerstoff in der genannten Vergasungszone, Herstellung eines Stroms

,umfassend Synthesegas und Flugasche, und Herstellung von Verunreinigungen als Feststoffmaterialien als Rückstand, die in einem Wasserbad als Kohle, feine Schlacke und grobkörnige Schlacke gesammelt werden;

Abschrecken und Abkühlen des genannten hergestellten Stroms, und Abtrennen der Flugasche aus dem genannten Strom, Herstellen einer abgetrennten Flugasche;

Entfernen einer Kohle umfassenden Materials aus dem genannten Wasserbad; Fluidisieren der entfernten Kohle in einem Wirbelschichtbett in einer Fluidisierungszone, und Oxidieren der genannten Kohle in dem genannten Wirbelschichtbett mit Sauerstoff bei einer Temperatur unterhalb der Schmelztemperatur der Mineralkomponenten der Kohle und des Wirbelschichtbettes, Herstellen eines im wesentlichen kohlenstofffreien Rückstandes und erhitzter Gase; dadurch gekennzeichnet, daß die entfernte Kohle bei einer Temperatur im Bereich von 500 °C bis 1150 °C und einem Druck im Bereich von atmosphärischen Druck oder höher in dem genannten Wirbelschichtbett zusammen mit mindestens einem Teil der genannten abgetrennten Flugasche oxidiert wird, welche in das genannte Wirbelschichtbett eingedüst und oxidiert wird, und Verwendung der so gewonnenen Wärme außerhalb der Vergasungszone zum Trocknen der in die Vergasungszone einzuspeisenden Kohle.

2. Das Verfahren, wie in Anspruch 1 beansprucht, dadurch gekennzeichnet, daß die Wärme in den genannten erhitzten Gasen genutzt wird, indem man feuchte Einspeisungskohle mit einem Feststoff oder Gas kontaktiert, welcher(s) direkt oder indirekt durch die genannten erhitzten Gase erhitzt wird.

