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(54) GAS-PREPARATION PROCESS

(71) We, SHELL INTERNATIONALE RESEARCH MAATSCHAPPIJ B.V., a company organised under the laws of The Netherlands, of 30 Carel van Bylandtlaan, The Hague, The Netherlands, do hereby declare the invention, for which we pray that a patent may be granted to us and the method by which it is to be performed to be particularly described in and by the following statement:—

The present invention relates to a process for the preparation of hydrogen — and/or carbon monoxide-containing gases, in which process

(i) a feed containing carbon and/or one or more hydrocarbons is subjected to incomplete combustion in an empty reactor, resulting in a stream of crude product gas,

(ii) the crude product gas obtained according to (a) is, optionally, cooled in a indirect heat exchanger to a temperature not lower than 200°C,

(iii) solid particles such as ash and/or soot are removed from the crude product gas, after which, if desired, the product gas is subjected to further purification and processing.

In a gas-preparation process as described above special steps have to be taken to free the crude product gas from by-products such as soot, ash, volatile sulphur compounds, and traces of HCN, which by-products can be present in varying amounts, depending on the process conditions and the nature of the starting material for the gas-preparation process.

Several processes are known for the removal of the said by-products from the crude gas. If it is done by washing the gas with water, then a water stream is obtained in which a large proportion of the said by-products are present. This water cannot be re-used without purification since the concentration therein of solid particles and dissolved gases would become higher and higher. Moreover, the amount of water increases continuously, due to condensation of water vapour contained in the crude product gas which arises from the partial combustion and, when steam is added as a moderator therefore, from the added steam. It is therefore necessary that water should be removed from the process, and it is this water, in particular, that should be free from undesirable components. If, for the separation of solid particles from the crude gas, use is made of cyclones, the same problems are encountered because washing of the gas leaving the cyclones remains necessary to remove the remaining solid particles from the gas. This water also contains dissolved gases; and in this case also, water must be removed from the process. The present invention provides an improved process for the preparation of hydrogen- and/or carbon monoxide-containing gases.

According to the present invention there is provided a process for the preparation of hydrogen- and/or carbon monoxide-containing gases, which comprises (a) subjecting a feed containing carbon and/or one or more hydrocarbons to incomplete combustion in an empty reactor to form a crude product gas containing entrained solid particles of ash and/or soot, (b) combining with said crude product gas an aqueous suspension of such solid particles of ash and/or soot in an amount which is such that the water content of said suspension vaporises and mixes with said crude product gas, (c) passing the resulting gas mixture through at least one gas-proof cyclone to separate therefrom a proportion of the solid particles entrained therein, and (d) contacting the gas from said cyclone(s) with an aqueous suspension of said solid particles in which suspension the water has a temperature at least equal to the dew point of said crude product gas to remove the remaining entrained solid particles as an aqueous suspension, part of which aqueous suspension is returned to step (b) and the remainder of which is used as a source of water for step (d), if necessary after cooling.

The process can be started-up by supplying water per se to steps (b) and (d) until sufficient aqueous suspension is available for recycling from step (d).

Said crude product gas can be cooled in an indirect heat exchanger to a temperature not lower than 200°C, which preferably is at least 250°C, prior to effecting step (b), or simultaneously therewith. Such cooling can be effected in a waste-heat boiler.

The removal of solid particles, ash and/or soot from the crude product gas is effected in two steps. With the aid of one or more cyclones it is possible to remove about 90% of the solid particles as present in the crude product gas as obtained in step (a), with or without said optional cooling thereof. The remaining solid particles are removed by washing with water in step (d). This washing treatment also serves to remove part of the gaseous by-products such as sulphur compounds and traces of HCN. The volume of the gas is not increased in the washing step (d) because the temperature of the wash water is at least equal to the dew point of the gas. In fact, water condenses from the gas in the washing step. All of the wash water remains in the process, being recycled, in part to the crude product gas, and in part to water washing step (d). During the process of the present invention an equilibrium concentration of solid particles in the recycled water is achieved. Preferably the amount of water recycled to step (d) is five to twenty times as large that recycled to step (a). The solid particles in the aqueous suspension obtained in step (b) are removed from the system in the cyclone(s), so that in due course all the solid particles originally entrained in the crude product gas are removed by the cyclone or cyclones.

The water which passes to step (b) is combined with a gas stream which has a temperature of at least 200°C and preferably at least 250°C. This temperature is much higher than the dew point of the gas, which, in the gasification of hydrocarbons with air, is about 70°C. The water vaporises completely, the dew point of the gas rises, and an amount of water equal to the amount vaporised will condense in the washing process of step (d) if the temperature of the wash water added in step (d) is equal to the first-mentioned dew point. Such condensation of water vapour during washing promotes efficient removal of solid particles from the gas.

If gasification is effected with oxygen, the dew point of the crude product gas can be 170 - 180°C. It is then preferred to cool the stream of crude product gas upstream in an indirect heat exchanger ("waste-heat boiler") and in such case the aqueous suspension added in step (b) can be added to the crude product gas upstream of the heat exchanger because the temperature of the gas will be high enough (1300 - 1400°C) to evaporate all of the water added and it can easily be arranged that the temperature of the gas leaving the waste-heat boiler is at least 200°C and preferably at least 250°C, so that there is enough heat in the gas to fully evaporate the added water without cooling the gas to the dew point.

In the washing process of step (d), water from the wash liquid, ie the recycled aqueous suspension, also evaporates and the evaporation will be the greater the higher the temperature of the recycled aqueous suspension is above the dew point of the gas. Since the water evaporated is carried off with the gas stream, it is necessary to add water to the washing process of step (d) from an outside source to compensate for the water evaporated.

In this way all solid particles removed from the crude product gas are obtained in the dry state, and no water need be removed in liquid form from the system.

That part of the aqueous suspension which is recycled to the washing step (d) can, if necessary, first be cooled to the temperature conditions required in step (d). The need for this will be the greater the more the temperature of the gas entering step (d) exceeds that of the recycled stream.

The volume ratio of the recycled streams of aqueous suspension from step (d) may be chosen within very wide limits and is determined in particular by the nature and the amount of the solid particles in the gas leaving the cyclone or cyclones. The higher the content of the solid particles of this gas, the larger will be the amount of aqueous suspension recycled to step (b).

In general, it is desirable that the gas obtained from step (d) should be cooled to a temperature below the dew point referred to in step (d), and that the water formed by such cooling should be separated from the gas stream. Before such cooling the gas will generally have a temperature of about 250°C, and the temperature to which the gas will be so cooled will depend on the use to which the gas will be put. The aqueous condensate formed in such cooling will contain dissolved gases such as sulphur compounds, and traces of HCN. No metal ions or metal compounds will be present. The condensate can be freed from dissolved gases by stripping and/or oxidation with oxygen or an oxygen-containing gas, such as air. The absence of metal ions or compounds in the condensate is of great importance, because HCN, in particular, can form complex metal compounds which are difficult to oxidize in aqueous medium.

Complex metal compounds of HCN will certainly be present in the recycled streams of aqueous suspension obtained in step (d), but an equilibrium concentration in the aqueous phase will be reached, due to the recycling of one of the streams to the gas upstream of the cyclone or cyclones. Metal compounds are removed in the solid material separated by the cyclone or cyclones. The volume of water separated from the gas after the cooling referred to above, is, at most, equal to the amount of water leaving the reactor with the crude product gas. The water that has been separated and subsequently been freed from dissolved gases is suffciently pure to be disposed of, or re-used. Part of this water can be used, in particular, to maintain the required amount of wash water in step (d), if this is necessary.

The solid particles separated in the cyclone(s) can be passed to the reactor for incomplete combustion (step (a)) with the aid of a transport gas, which can be steam or pressurized product gas. In this way the feed is substantially fully converted into gas, only ash particles, if present, being unconverted. Such ash particles will be able to accumulate in the reactor, from which they can be removed.

Through the use of different cyclones it is also possible to separate the solid particles into ash and soot particles, if the nature of the ash particles permits. In this case only the soot will be recycled, in order to prevent the accumulation of ash in the reactor. The soot, separated or not from ash particles, if present, can also be used for purposes other than recycling to the reactor. It can serve as fuel. It can also be processed to active-carbon preparations.

The process of the present invention will be described with reference to the accompanying drawings, which are block diagrams of two embodiments of the process according to the invention.

In figure 1 the numeral 1 refers to a reactor to which are fed a feed stream 2 containing coal and/or one or more hydrocarbons and a stream 3 of free oxygen-containing gas. Steam or a different gaseous moderator can be supplied as well. The crude product gas 4 leaving the reactor 1 is passed to an indirect heat exchanger 5, in which a stream of water 6 is converted into steam 7. Cooled crude gas 8 is combined with a stream 10 in an evaporator 9, which will be described in more detail further on and which consists of a suspension of solid particles in water. The evaporator 9 can be a venturi. The heat content of the gas 8 is partially used for evaporation of the water of the stream 10. The gas 11 from the evaporator 9 travels to a cyclone 12. This cyclone 12 produces a stream 13 of separated solid material and a gas stream 14, which last-mentioned gas stream still contains some solid particles. This gas stream 14 is passed to a washer 15, which can consist of a column with gas-liquid contact discs, or of one or more venturis. The washer 15 produces a suspension 16 of solid particles in water. A partial stream 17 thereof is passed via a cooler 18 back to the washer 15, and the other partial stream is the aforementioned stream 10. At its entry into the washer 15, the partial stream 17 has a temperature which is at least equal to the dew point of the gas stream 8. (In fact, the streams 4 and 8 have the same dew point). All of the volume of water evaporated in the evaporator 9 condenses in the washer 15 if the temperature of the water 17 after the cooler 18 is equal to the dew point of the gas streams 4 and 8. The condensation promotes the take-up of the solid particles from the gas by the water.

The stream 13 of solid particles—soot and possibly ash—are passed to a mixer 19, where they are dispersed in a gas stream 20 and travel as a stream 21 to the reactor 1.

The washer 15 produces a gas stream 22 which is free from solid particles. This gas is cooled in the cooler 23, resulting in a dry gas stream 24 and a water stream 25. The latter stream contains dissolved gases such as sulphur compounds and traces of HCN, which can be removed in a stripper 26, for instance with a gas stream 27 (air or steam). The noixous components of the resulting gas stream 28 can be converted into harmless compounds by burning them, or they may be removed in some other way. The water stream 29 is free from dissolved gases and can be disposed of.

In figure 2 reference numerals that are also used in figure 1 have similar meanings. Here, the evaporator 9 is positioned between the reactor 1 and the heat exchanger 5, so that at a very high temperature a gas stream 30 is formed into which the partial stream has evaporated and been taken up. The figure further shows two cyclones, of which cyclone 31 separates ash particles, which are removed as stream 33. Ideally, the solid particles contained in the gas stream 34 are only soot particles, which are separated off for the greater part with the cyclone 32 and are removed as stream 35. The temperature of the partial stream 17 at its entry into the washer 15 is now at least equal to the dew point of the gas stream 4. If any water is to be added, then a partial stream 36 taken from the water stream 37 from the stripper 26 can be used, the rest of the water being carried off from the stripper as stream 38.

It will be appreciated that other embodiments of the process of the present invention are possible.

WHAT WE CLAIM IS:—

1. A process for the preparation of hydrogen- and/or carbon monoxide-containing gases, which comprises (a) subjecting a feed containing carbon and/or one or more hydrocarbons to incomplete combustion in an empty reactor to form a crude product gas containing entrained solid particles of ash and/or soot, (b) combining with said crude product gas an aqueous suspension of such solid particles of ash and/or soot in an amount which is such that the water content of said suspension vaporises and mixes with said crude product gas, (c) passing the resulting gas mixture through at least one gas-proof cyclone to separate therefrom a proportion of the solid particles entrained therein, and (d) contacting the gas from said cyclone(s) with an aqueous suspension of said solid particles in which suspension the water has a temperature at least equal to the dew point of said crude product gas to remove the remaining entrained solid particles as an aqueous suspension, part of which aqueous suspension is returned to step (b) and the remainder of which is used as a source of water for step (d), if necessary after cooling.

2. A process as claimed in claim 1, wherein said crude product gas is cooled in an indirect heat exchanger to a temperature not lower than 200°C prior to effecting step (b), or simultaneously therewith.

3. A process as claimed in claim 1 or claim 2, wherein the gas resulting from step (d) is cooled to a temperature below the dew point of said crude product gas, and the water formed thereby is separated from the cooled gas.

4. A process as claimed in claim 3, wherein the separated water is freed from dissolved gases by stripping and/or oxidation with oxygen or a free oxygen-containing gas.

5. A process as claimed in any one of claims 1-4, wherein solid particles separated in said cyclone(s) are carried to said empty reactor with a transport gas for incomplete combustion therein.

6. A process as claimed in claim 1 and substantially as described hereinbefore with reference to Figure 1 or Figure 2 of the accompanying drawings.

7. A hydrogen- and/or carbon monoxide-containing gas prepared by the process claimed in any one of claims 1-6.

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1458448 COMPLETE SPECIFICATION

1 SHEET *This drawing is a reproduction of the Original on a reduced scale*

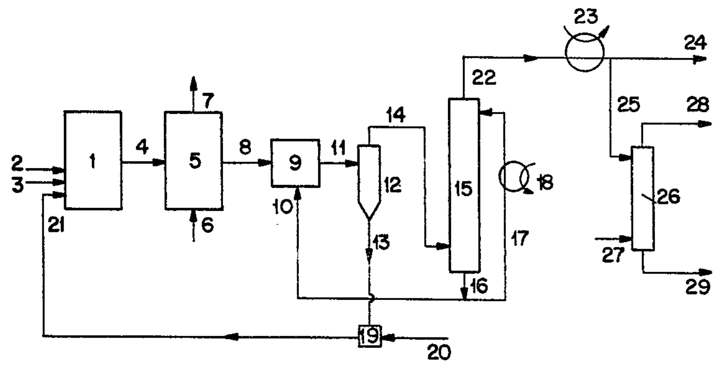

FIG. 1

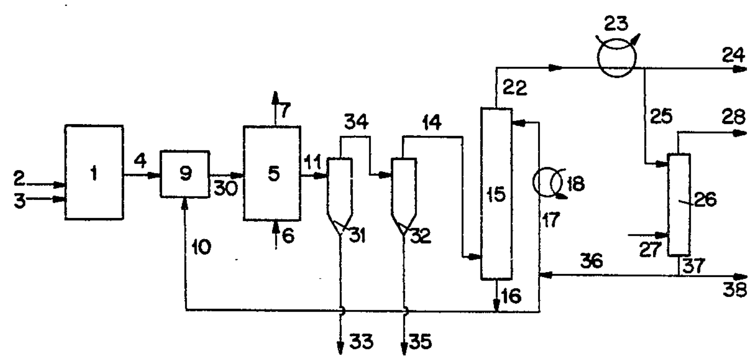

FIG. 2