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COMPLETE SPECIFICATION

Process and Apparatus for the Gasification of Fine-Grained Carbonaceous Substances

We, DE DIRECTIE VAN DE STAATSMIJNEN IN LIMBURG, handelend voor en namens de Staat der Nederlanden, of 2, van der Maesenstraat, Heerlen, The Netherlands, do hereby declare the nature of this invention and in what manner the same is to be performed, to be particularly described and ascertained in and by the following statement:—

The present invention relates to process and installation for the gasification of fine-grained carbonaceous substances.

In the well-known processes for gasifying fine-grained carbonaceous substances, such as fine coal, small coke, sawdust, and coal slime, usually called "dust-gasification", the yield of gases at a definite capacity of the reactor employed is considerably lower than in the case of the gasification of coarse-grained material under comparable conditions in a reactor of the same capacity, where capacity means the number of cubic metres of carbon monoxide and hydrogen produced per hour per cubic metre of the reaction space. This unfavourable result is obtained despite the fact that the rate of gasification is proportional to the surface area of the particles which are gasified and is chiefly due to the fact that in prior processes of dust gasification the conditions have not been realised which enable the same principles to operate as are in operation when coarse-grained fuel is gasified.

The conditions realised in the case of the gasification of coarse-grained fuel are obtained by providing a comparatively small reaction zone at a high temperature in which the exothermic reactions takes place and above this zone or between it and the outlet for the gases, a much larger and cooler zone in which the endothermic reactions occur. The starting material is then introduced in the larger and cooler zone in counter-current with the gases produced which pass through this larger zone and out of the producer, the ash being discharged preferably in the form of a liquid slag from the bottom of the producer. However, if the dust-gasification is carried out on the counterflow-principle usually observed in shaft producers with coarse-grained fuel the rate of gasification at high temperatures is so high that the average speed of the gas becomes considerably higher than the speed of fall of the small particles. Consequently the counterflow-principle can hardly be applied without objections in the case of dust-gasification, which has consequently been carried out on the concurrent principle with a floating bed (see, for instance, B.I.O.S. final report No. 1142, item No. 30, The Wintershall-Schmalfeldt Process, Figure 2), and to methods founded on a combination of this concurrent principle with a reaction under whirling conditions (see the Koppers process—Gas Journal 251 (1947), p. 617, and the Winkler process in B.I.O.S. final report 333, item No. 30). The use of whirling conditions has the advantage that a high dust-concentration and a fine transmission of heat can be accomplished, but the thermal efficiency obtained is low, because the gases produced leave the reactor at almost the same high temperature as that obtained at the gasification, and many solid particles are carried along by the gas mixture. In this connection it has been suggested to connect behind the gasifying reactor a second chamber, usually called the secondary producer, in which the gasification of the particles which were carried along takes place. In the so-called Winkler-process this secondary gasification is performed after adding oxygen to the gas mixture, with the drawback, however, that the temperature of the gas-mixture becomes still higher than the temperature of gasification. According to the so-called Koppers-process an improvement is gained by establishing a number of "boiling beds" one on top of the other, causing the reactor to resemble a plate-column usually employed in distillations, so that the dust-gasification is operated in more or less uniform reaction spaces fitted on top of each other, behind which reactor a secondary producer is placed for the purpose of gasifying the particles which are carried along.

The well-known methods of dust-gasification are characterised by the employment of such a second producer, in which the gasification takes place at a much lower dust concentration than that which is present in the first reactor, as a result of which the capacity of the gasification is low.

Now it has been found that favourable results are obtained with the gasification of fine-grained carbonaceous material in two reactors connected in series, by the process according to the invention, according to which the volume of the reaction space in one reactor (the large reactor) is a multiple of the volume of the reaction space in the other reactor (the small reactor) and the gasification in the small reactor is carried out under whirling conditions so that mainly the exothermic combustion reactions take place therein at a temperature above the melting temperature of the ash constituents and the contents of the small reactor after separating from the liquid slag are passed into the large reactor, in which mainly the endothermic gas reactions occur with an artificially increased concentration of the particles to be gasified and a lower temperature than the temperature in the small reactor.

It will thus be seen that in the process according to the invention the conditions obtaining in the gasification of coarse-grained fuel in a shaft-producer working according to the counterflow principle, are realised for it will be observed that at the bottom of such a shaft-producer a zone of high temperature and of a high capacity, which is short in proportion to the remaining part of the shaft, is being maintained. This is due to the incoming supply of fuel which has to preheated up to the gasification temperature by the gases coming from the bottom of the shaft and the high temperature and the high oxygen percentage required in the gasifying medium. That this zone of high temperature and high concentration is but short is because as a result of the endothermic character of the gasifying reaction the temperature falls rapidly at first and more slowly afterwards, due to the decrease of the rate of reaction of the gasification as a result of the lower temperature and the decrease of the concentration of the active components of the gasifying medium. In this way the short zone high temperature and high capacity is followed by a much longer zone of lower temperature and lower capacity. The longer the latter zone, the higher the efficiency of the producer.

In the process according to the present invention the two reactors can be compared with the two reaction zones respectively which occur in a shaft-producer for coarse-grained fuel. In the small reactor according to the invention a reaction zone is maintained which can be compared with the small high temperature zone of the shaft-producer and in the larger reactor of the invention a reaction zone is maintained which can be compared with the larger and cooler zone of the shaft-producer for coarse-grained material with the advantages of the shaft-producer. Furthermore, the use of two reactors enables several modifications to be made in the process according to the invention, for example, in the manner of introduction of the gasifying medium and the starting material.

In the small reactor in accordance with the invention it is necessary for the purpose of maintaining a high temperature that the time of stay of the reacting gases be small in proportion to the total time of contact, which can be obtained in a simple way because the volume of this reactor is smaller than of the large reactor. In order to otain a fine contact between the particles to be gasified and the gasifying medium and a long time of stay of the particles to be gasified, the whirling principle is made use of in this reactor.

By maintaining a high temperature in the small reactor the following advantages are obtained:—

1. The ash parts of the particles can be drained off as molten slag which presents a special interest at the dust-gasification, i.e., because in this way the percentage of fly ash of the produced gas mixture can be kept very low. The drain of the molten slag can, if required, also be effected from the big reactor;

2. Because of the relatively small dimensions of the reactor the same can be constructed in a simple way for high temperatures;

3. Because of the high temperature the proportion between the quantity of O_2 and the quantity of H_2O and/or CO_2 in the gasifying medium can be adjusted at such a value, preferably a proportion of 1:0.9—1.5, that a gas-mixture is produced having a very slight quantity of ballast of CO_2 and H_2O.

The requirements of a high temperature, a short time of stay, the whirling-principle, molten slag and a relatively small reactor can be complied with by employing a vortex chamber or preferably a cyclone as the small reactor; by operating the process in accordance with the invention with a cyclone as the small reactor notably fine results are obtained.

In the large reactor, which is also employed in the process in accordance with the invention, the components, which act as gasifying agents and are still present in the gas-mixture coming from the small reactor, react with the parts of the material to be gasified which are not yet completely converted, and at a lower temperature than that which prevails in the small reactor. For this reason the time of stay of the gas in this large reactor has to be longer in order to ensure a sufficient contact between the gasifying medium and the particles to be gasified.

This contact is furthermore promoted by increasing artificially the concentration of the particles, which can be done, e.g., by using one or more of the well-known "boiling bed" systems as used in the first reactor in some of the well-known methods, e.g., of Koppers, or by using a system as described in the British Patent Application No. 6172/49 (Serial No. (669,906), where the gasification of the bigger particles takes place in accordance with the counterflow-principle and the smaller particles, insofar as they are not gasified are carried along out of the reactor and after separating from the gas mixture can be fed to the small reactor.

The separation of solid particles from the reaction mixture leaving the large reactor is of particular interest when carrying out the process according to the invention in such a manner that the initial material to be gasified is first introduced into the large reactor, and the particles not gasified therein, are separated from the gases, advantageously by means of a cyclone, and are fed to the small reactor.

Moreover the large reactor may be arranged, for the purpose of increasing the concentration of the particles, in the way as described in the British Patent Application No. 33431/48, by which a favourable effect can be obtained in a simpler way than in the above-mentioned system according to Koppers. In that case a special advantage can be obtained at the same time if coke is applied as filling-material, because the surface of carbon in the reactor is then increased, beside other advantages, such as avoiding losses by wear and tear, while the refractoriness of the filling material does not present any problem.

A further advantage can be obtained by introducing water vapour and/or CO_2 and/or hydrocarbons into this reactor in one or more places Catalysts can be provided inside the reactor for the conversion of CO with water vapour, as well as catalysts for the formation of CH_4, so the composition of the produced gas-mixture can be regulated.

The composition of the gas produced may also be influenced by adding to the mixture coming from the small reactor the carbonaceous substances to be gasified and/or gasifying agents, such as flue gases and/or gases which contain oxygen. Water vapour and/or carbonic acid and/or hydrocarbons can also be added to the same. Besides, the composition of the gas-mixture can be regulated by connecting two or more small reactors, preferably cyclones, in parallel and by connecting them with one large reactor.

When working with a molten slag-drain, the produced gas-mixture can be wholly freed by a cyclone of the fly ash which can still be present therein, which fly ash, when containing still ungasified parts, can be returned to the small reactor, if required.

The draining of the molten slag can be effected from the small reactor as well as from the large reactor or from both reactors. When draining molten slags from the large reactor, the temperature at the bottom of this reactor should be higher than the temperature at which the slag becomes molten.

The method according to the invention can be performed in an efficient manner by utilising in the usual way the heat which is present in the produced gas-mixture, preferably for the purpose of preheating the material to be gasified. This exchange of heat can also take place by means of a "boiling bed" and/or a column with filling-bodies. When using a cyclone as the small reactor the preheating of the material which is advantageously fed into the said cyclone by means of a feed-cyclone, can be effected in such a manner that the said material is heated in the feed cyclone to about the reaction temperature by leading into the feed cyclone a fuel, preferably gaseous, and burning the same therein with oxygen or a gas containing oxygen.

The method according to the invention may be performed in the way indicated and illustrated by way of example in the diagram on the annexed drawing, according to which oxygen or a gas containing free oxygen, such as air, or air enriched in oxygen is introduced at 2 into the small reactor 1, which has the form of a cyclone whereas by means of a small feed-cyclone 3 with an inlet-opening 4 the material to be gasified is introduced into the reactor. In the large reactor 5 there is a column with filling-bodies 6 on the top of which is a "boiling bed" 7. In this reactor the fine grained carbonaceous substances to be gasified is supplied at 8 and/or 9, whereas gases which contain oxygen are introduced at 10 and water vapour at 11 and 12. The molten slag is drained off at 13, whereas the heavier particles leave the reactor at 14, and at 15 the gas-mixture and the lighter particles, which are separated in the cyclone 16 from the gas which leaves at 17 and are returned into the feed-cyclone at 4 together with the heavier particles.

Having now particularly described and ascertained the nature of our said invention and in what manner the same is to be performed, we declare that what we claim is:—

1. A process for the manufacture of gas-mixtures by gasifying fine-grained carbonaceous material in two reactors connected in series, characterised in that the volume of the reaction-space in one reactor (the large reactor) is a multiple of the volume of the reaction-space in the other reactor (the small reactor) and that the gasification is carried out in the small reactor under whirling conditions so that mainly the exothermic combustion reactions take place therein at a temperature above the melting temperature of the ash constituents and the contents of the small reactor after separating from the liquid slag are passed into the large reactor in which mainly the endothermic gas reactions occur with an artificially increased concentration of the particles to be gasified and a lower temperature than the temperature in the small reactor.

2. A process in accordance with Claim 1, characterised in that the concentration of the particles to be gasified in the large reactor is artificially increased by employing a column containing filling-bodies and/or a "boiling bed" system.

3. A process in accordance with Claim 2, characterised in that ores are employed as filling-bodies and the metal obtained therefrom is discharged.

4. A process in accordance with Claim 2, characterised in that a column of coke is used as the column with filling-bodies.

5. A process in accordance with any one of the preceding Claims 1—4, characterised in that a cyclone is employed as the small reactor.

6. A process in accordance with any one of the preceding Claims 1—5, characterised in that a continuous operation the starting material to be gasified is first introduced into the large reactor and the solids not gasified therein are separated from the gas produced and are passed into the small reactor and subsequently the contents of the small reactor after separation of the molten slag are fed to the large reactor.

7. A process in accordance with any one of the preceding Claims 5 and 6, characterised in that a feed-cyclone is used for introducing the material into the cyclone used as the small reactor, and the material is preheated in the said feed-cyclone to about the reaction temperature by leading therein a fuel, preferably gaseous, and burning the same therein with oxygen or a gas containing oxygen.

8. A process in accordance with one of Claims 1 to 7, characterised by the feature that two or more small reactors connected in parallel are connected to one joint big reactor.

9. An installation for carrying out the process claimed in Claim 1, consisting of two reactors connected in series, characterised by the feature that a cyclone which is a radially symmetrical apparatus with a tangential inlet is connected in series with a reactor, the volume of the reaction-space whereof is a multiple of the volume of the cyclone and means for artificially increasing the concentration of the particles to be gasified are provided by filling the reaction space in said large reactor with irregularly shaped filling bodies leaving free interstitial spaces between said bodies.

10. An installation in accordance with Claim 9, characterised by the feature that a number of cyclones is connected in parallel and that each one hereof is connected in series with a joint reactor, the volume of the reaction-space whereof is a multiple of the volume of each of the cyclones.

11. Process for the gasification of fine-grained carbonaceous substances substantially as herein described.

12. Apparatus for the gasification of fine-grained carbonaceous substances substantially as herein described with reference to and as illustrated in the accompanying drawings.

Dated this 29th day of December, 1948.

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690,214 COMPLETE SPECIFICATION
1 SHEET

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