

## PATENT SPECIFICATION



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342,817

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Complete Accepted : Feb. 12, 1931.

## COMPLETE SPECIFICATION.

### Improvements in or relating to the Distillation of Solid Carbonaceous Materials, and to the Manufacture of Gas.

I, HARALD NIELSEN, a subject of the King of Denmark, of 78, Hayes Road, Bromley, in the County of Kent, and BRYAN LAING, a subject of the King of Great Britain, of York Mansion, Petty France, Westminster, in the County of London, 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:—

This invention relates to an improvement in or modification of the invention claimed in our prior patent No. 292,060.

In our prior patent No. 292,060 we have claimed a process of distilling carbonaceous materials of a coking or caking character for the purpose of gas manufacture, which process consists in first subjecting carbonaceous materials to the action of hot gases at a temperature of about 150° C., said gases containing a controlled percentage of oxygen or an oxidising constituent of the order of from 2% to 8% so as to oxidise some of the so-called coking constituents of the coking materials and thereby influence their coking power. The result of this treatment is to cause a large proportion of the coking carbonaceous material, either at this stage of the process or during the subsequent step of distillation, to form relatively large lumps of from 2" to 8" cube or larger instead of the entire mass fusing together as is normally the case during so-called tumescence stage, which stage occurs when coking coals are subjected to low temperature distillation. The lump coke so produced, or the carbonaceous material subjected to this preliminary treatment is then subjected to distillation. This distillation process may be carried on at temperatures not exceeding 600° C. and during distillation at this temperature the condensable oils are driven off in the form of vapour and recovered. Thereafter the lump coke may, if desired, be subjected to a process of high temperature distillation so as to drive off the remaining volatile constituents of the lump coke, and if desired reduce its volatile content to from 1% to 4%. The lump coke so produced

may then be fed into a water gas producer and treated with steam for the purpose of water gas production.

In the specification of our prior patent No. 292,060 we have stated that the pieces of carbonaceous material subjected to the aforesaid treatment should preferably not exceed 2" cube.

According to the present invention, the pieces of coking or caking coal subjected to the aforesaid treatment may range in size from 1/32" cube to approximately 2" cube, and it is to be understood that mixtures containing pieces of any of the aforesaid and intermediate sizes with or without dust particles may effectually be used for carrying on the aforesaid process. The semi-coke produced by the aforesaid process or selected or graded pieces therefrom may be gasified in a water gas generator. Gasification may be carried on at a high rate, e.g., 50 to 80 lbs. of coke per hour employing an excess of steam and a relatively shallow fuel bed, e.g., 12 inches. The semi-coke containing approximately 10% to 15% of volatile matter may be fed direct from the low temperature retort to the water gas generator, or in cases where the material is subjected to distillation at higher temperatures so as to reduce its volatile content, the lump coke or the coke containing from 1% to 4% of volatile matter may also be fed direct to the water gas generator. The water gas so produced from the last mentioned coke and which contains methane may be treated with steam at a high temperature in the presence of a catalyst adapted to cause the conversion of methane to carbon monoxide and hydrogen, in a manner well known in the art. Furthermore, the resultant gaseous mixture obtained as aforesaid may be treated in the presence of a catalyst or catalysts with steam at a lower temperature to cause the conversion of carbon monoxide to carbon dioxide with production of a corresponding amount of hydrogen, all as well known in the art.

The lump semi-coke produced by varying the coking properties of coking or caking coals and subsequently subjecting the

[Price 1/-]

same to carbonisation, may be gasified with steam to produce water gas and the water gas so produced may be subjected to treatment, at temperatures between 200° C. and 300° C. with catalysts favouring the production of higher paraffin hydrocarbons. The paraffins so produced may then be separated. The methane may also be removed separately from the higher paraffin by diacarbons and the residual gas may be employed for methanol or ammonia synthesis, in the latter case after conversion of the carbon monoxide, the removal of carbon dioxide and the addition of nitrogen, all as well known in the art. Furthermore, the water gas from the low temperature coke may be treated at high temperatures with excess of steam to convert the methane present in the gas mixture into hydrogen, carbon monoxide and carbon dioxide gases, the gas being then utilised for the synthesis of methanol with passage of the gases over a suitable catalyst and utilisation of the residual gas for ammonia synthesis, with conversion of carbon monoxide to  $\text{SO}_2$ , subsequent removal of carbon dioxide and addition of nitrogen.

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. An improvement in or modification of the invention claimed in our prior patent No. 292,060, which consists in starting with pieces of carbonaceous material ranging in size from  $1/32$ " cube to approximately 2" cube, or with mixtures of materials containing pieces of any of the aforesaid and intermediate sizes with or without dust particles, preheating said carbonaceous material in the presence of oxygen so as to control its coking properties and carbonising the product at temperatures of approximately 800° C. thereby producing a large proportion of lump coke and finally treating said coke, or selected pieces or graded pieces thereof, with steam in a water gas generator.

2. The process claimed in claim 1 wherein after low temperature carbonisation proper has been carried out with recovery of the gas and oils, the semi-coke still containing volatile matter is heated to a higher temperature to drive off further quantities of volatile matter and to produce a lump coke suitable for use in water gas generators.

3. The process claimed in claim 1 in which the semi-coke is gasified at a high rate in a water gas generator employing an excess of steam and relatively shallow fuel beds.

4. The process claimed in claims 1 or 2

in which the semi-coke is fed direct from the low or high temperature retort to the water gas generator.

5. The improvement in or modification of the invention claimed in our prior patent No. 292,060, or in any of the preceding claims, wherein the lump semi-coke resulting from distillation at higher temperatures is treated with steam, and in which the water gas so produced and which contains methane is treated with steam at a high temperature in the presence of a catalyst adapted to cause the conversion of the methane to carbon monoxide and hydrogen.

6. The process as claimed in claim 5 in which the resultant water gas is treated in the presence of a catalyst with steam at a lower temperature to cause the conversion of carbon monoxide to carbon dioxide with the production of a corresponding amount of hydrogen.

7. The improvement in or modification of the invention claimed in our prior patent No. 292,060, or in any of the preceding claims 1 to 4, wherein the water gas produced by gasifying with steam the lump semi-coke resulting from distillation at lower temperatures is treated at temperatures between 200° and 300° C. with catalysts favouring the production of higher paraffin hydrocarbons, separating said paraffins, removing methane and employing the residual gas for methanol or ammonia synthesis, in the latter case after conversion of the carbon monoxide, removal of carbon dioxide and the addition of nitrogen.

8. The improvement in or modification of the invention claimed in our prior patent No. 292,060, or in any of the preceding claims 1 to 4, wherein the water gas produced by gasifying with steam the lump semi-coke resulting from distillation at lower temperatures is treated at high temperatures with excess of steam to convert the methane into hydrogen, carbon monoxide and carbon dioxide, the resultant gases being utilised for the synthesis of methanol with passage of the gases over a suitable catalyst and utilisation of the residual gas for ammonia synthesis, with conversion of carbon monoxide, removal of carbon dioxide and addition of nitrogen.

Dated this 21st day of November, 1929.

J. McCLARY,

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Agent for the Applicants.

Reference has been directed, in pursuance of Section 7, Sub-section 4, of the Patents and Designs Acts, 1904 to 1928, to Specification No. 292,573.