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(12) **Patent:** (11) **CA 281814**

(54) GAS AND HYDROGEN MANUFACTURE

(54) FABRICATION DE GAZ DE COKE

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ABSTRACT:

CLAIMS. Show all claims

*** Note: Data on abstracts and claims is shown in the official language in which it was submitted.

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The growing importance of low temperature carbonisation of fuels has caused some attention to be directed towards the rational utilisation of the carbonised product known as semi-coke. Apart from its use as a smokeless domestic fuel, the active combustible properties of this material have given rise to various suggestions for its employment in industry. Thus it has been suggested to employ semi-coke in pulverised fuel furnaces, and it has also been proposed to make producer gas or water gas from it. However, for this latter purpose ordinary semi-coke exhibits the notable disadvantage that its physical form does not suit the usual type of water gas apparatus, which demands a fairly uniform lump coke, and further when the water gas is to be used as a source of hydrogen, as for example in the case of ammonia synthesis, the hydrocarbons produced from the semi-coke during gasification are very objectionable.

According to the present invention we start with a coal of preferably high caking properties, pre-heat said coal, preferably as a slack, in the presence of oxygen so as to control its caking properties, and carbonise the product at a temperature not exceeding 600°C so as to produce a large proportion of lump semi coke and finally treat said semi coke with steam in a water gas generator. According to a further feature of the invention the lump semi-coke is gasified in a water gas generator or other suitable apparatus by means of steam at a high temperature and the resultant gas, containing undesirable fixed hydro-carbons such as methane derived from the semi-coke, is treated with steam at a high temperature to convert the methane etc. to carbon monoxide and hydrogen.

If desired, the semi-coke may be heated to a higher temperature for example 800°-900°C after low-temperature carbonisation, to drive off remaining volatile matter before treatment in the water-gas generator.

In this way a gaseous mixture comprising carbon

monoxide and hydrogen in about equal proportions and substantially free from methane is obtained, and according to further features of the invention this gaseous mixture is utilised in any of the following ways:

(a) The gas mixture at atmospheric pressure may be passed in known manner over a catalyst favouring the production of higher paraffin hydrocarbons.

Suitable temperatures for this reaction are from 200°C-300°C. Catalysts containing finely divided iron or metals, with or without promoters, may be used.

(b) The original gas mixture or the residue from (a) after separation of the paraffins may be compressed and after removal of the methane, if present, passed over a catalyst such as basic zinc chromate favouring the production of methanol.

(c) The original gas mixture or the single or combined residue from (a) and (b) may be treated with steam in the presence of an iron oxide catalyst at about 500°C in this way converting the carbon monoxide to carbon-dioxide with the simultaneous production of hydrogen. The carbon dioxide is then removed, preferably by dissolution in water under pressure, and the residual hydrogen is mixed with the proper proportion of nitrogen for the synthesis of ammonia.

In order to produce a predetermined quality of lump semi-coke according to the present invention, it is necessary to start with a coal having preferably somewhat pronounced caking properties, e.g. Durham slack. If the coal has a high ash content it may be desirable to remove part of the foreign matter by washing, so as to produce a semi-coke having not too much ash with a view to avoiding clinkering trouble in the gasification apparatus. The raw coal, whether washed or not, is dried by heating, preferably by means of hot flue or combustion gases, and enough oxygen (air) is added to effect partial oxidation

of the material, in this way controlling its caking properties, utilising the existing knowledge of the art. For example a temperature of 200° 300°C is suitable with flue gases containing 5% of free oxygen for say 3 to 4 hours. Heating without oxygen may be employed in some cases.

The coal is then submitted to low temperature carbonisation in a retort of any suitable design employing a temperature not exceeding 800°C. The gas and oils are of course recovered in the usual way. A large proportion of the semi-coke is easily obtained in the form of lumps suitable for use in a water gas generator, especially as larger lumps than usual may be used since the semi-coke carbon is more or less in the active condition and as is well known, the semi-coke may be gasified at a much greater rate than ordinary coke. It is advantageous to feed the semi-coke from the low temperature retort direct to the water gas generator in order to save heat. In making water gas from the semi-coke it is also advantageous to employ shallow beds of the fuel in the generator and to work with rather a high excess of steam since not only are the gasification conditions thereby favourably influenced, but also the steam necessary for the further catalytic treatment of the water gas is conveniently introduced and superheated.

The water gas leaving the generator may be cooled and freed from tar in the usual way and employed without conversion of the methane according to method (a) above. Or it may be treated after dust separation, without allowing its temperature to fall appreciably, in a catalytic chamber containing a catalyst such as nickel which promotes the conversion of methane according to the equation $CH_4 + H_2O = CO + 3H_2$. Suitable temperatures for this reaction are above 800°C and an excess of steam is very desirable. As a rule the water gas to be treated contains up to 5 per cent of methane, which the conversion reduces to practically nothing. The resultant gas mixture is

adapted for use according to methods (b) and (c) outlined above.

We do not claim broadly the method of producing the lump semi-coke or of treating the water gas produced therefrom, herein described, as our invention resides in the combination as stated, which possess special economic and technical advantages.

I CLAIM AS MY INVENTION:-

1. The process of low temperature carbonisation and gas manufacture consisting in starting with coal, preheating said coal in presence of oxygen, and carbonising the product at temperatures not exceeding 600°C thereby producing a large proportion of lump semi-coke and finally treating said coke with steam in a water gas generator.

2. Process of low temperature carbonisation and gas manufacture consisting in starting with coal of high caking properties, preheating said coal as a slack in presence of oxygen so as to control its caking properties and carbonising the product at a temperature not exceeding 600°C thereby producing a large proportion of lump semi coke, and finally treating said coke with steam in a water gas generator.

3. Process of carbonisation and gas manufacture consisting in starting with coal of high caking properties, preheating said coal in presence of oxygen so as to control its caking properties carbonising the product at a temperature not exceeding 600°C recovering the rich gas and oils, heating the lump semi coke thus obtained to 800-900°C to drive off further volatile matters and treating the coke thus obtained with steam to generate water gas.

4. Process of low temperature carbonisation and gas manufacture consisting in starting with coal of high caking properties, preheating said coal in presence of oxygen to control its caking properties carbonising the product at a temperature not exceeding 600°C, thereby producing a large proportion of lump semi coke and gasifying said semi coke at a high rate in a water gas generator, using an excess of steam and a relatively shallow fuel bed.

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5.. Process of low temperature carbonisation and gas manufacture consisting in starting with coal of high caking properties, preheating said coal in presence of oxygen to control its caking properties carbonising the product at a temperature not exceeding 600°C thereby producing a large proportion of lump semicoke and feeding the semi coke thus obtained directly from the low temperature retort to a water gas generator and gasifying same with steam.

6. Process of carbonisation and gas manufacture consisting in starting with coal of high caking properties, preheating said coal in presence of oxygen so as to control its caking properties carbonising the product at a temperature not exceeding 600°C recovering the rich gas and oils, heating the lump semi coke thus obtained to 800-900°C to drive off further volatile matters and feeding the coke thus obtained directly from the retort to a water gas generator, and gasifying same with steam.

7. Process of gas manufacture in which lump semi coke obtained by low temperature carbonisation is treated with steam, and the gas so produced is treated with steam at a high temperature in the presence of a catalyst adapted to cause the conversion of the methane etc., to carbon monoxide and hydrogen.

8. Process of gas manufacture in which lump semi coke obtained by low temperature carbonisation is treated with steam and the gas so produced is treated with steam at a high temperature in the presence of a catalyst adapted to cause the conversion of the methane etc., to carbon monoxide and hydrogen, and the resultant mixture of carbon monoxide and hydrogen is treated with steam in the presence of a catalyst at a lower temperature (about 500°C) to produce hydrogen and carbon dioxide.

9. Process consisting in gasifying with steam, lump

semi coke obtained by low temperature carbonisation, treating the resultant gas at temperatures between 200-300°C with catalysts favoring the production of higher paraffin separating said paraffins removing methane and employing the residual gas for methanol synthesis.

10. Process consisting in gasifying with steam, lump semi coke obtained by low temperature carbonisation, treating the resultant gas at temperatures between 200-300°C with catalysts favoring the production of higher paraffins separating said paraffins, removing methane and employing the residual gas after replacement of the carbon monoxide by hydrogen, removal of carbon dioxide, and addition of nitrogen, for ammonia synthesis.

11. Process consisting in gasifying with steam lump semi coke, obtained by low temperature carbonisation, treating the gas with steam at high temperatures to decompose the methane, utilising the gas for the synthesis of methanol with single passage of the gas over a suitable catalyst, and utilising the residual gas for ammonia synthesis, after replacement of the carbon monoxide by hydrogen, removal of carbon monoxide and addition of nitrogen.

IN TESTIMONY WHEREOF I have signed at the City of London, England, this twelfth day of January A.D. 1928.

Herbert Alfred Humphrey

Witnesses:

E. J. Parker

A. D. Diggs