[Second Edition.]

PATENT SPECIFICATION

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Complete Accepted: Mar. 10, 1921.

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COMPLETE SPECIFICATION.

Improved Manufacture of Methane.

We, FARBWERKE VORM. MEISTER LUCIUS & BRUNING, a German company, of Hoechst a. Main, Germany, 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 a modification of or improvements in that described in Specification No. 146,110, that is to say in a process for obtaining methane in a high concentration or pure from carbon monoxide and hydrogen by gradually introducing the carbon monoxide while periodically eliminating the water formed during the reaction and, if required, subsequently eliminating the residue of the hydrogen by any known method. In order to ensure that the process proceeds smoothly, it is important that the amount of carbon monoxide contained in the mixture should never exceed about $^1/_5$th of the amount of hydrogen, because otherwise carbon will separate.

Special care must be taken to maintain this quantitative proportion; particularly in the later stages in which increasingly smaller quantities of gases are concerned, the necessary continual strict control of the working of the reaction causes some difficulties.

By the present invention these difficulties can be avoided by using for converting into methane the hydrogen still present, certain dioxide instead of carbon monoxide when the mixture has already been enriched in methane to a certain extent. The use of an excess of carbon dioxide does not in any way disturb the reaction; when the reaction is finished the excess may easily be removed, for instance by absorption by lime.

As is known, hydrogen is converted into methane by carbon-dioxide according to the gross equation; $CO_2 + 4H_2 = CH_4 + 2H_2O$. We have discovered that in the present process, as in that of the aforesaid specification, the periodical elimination of the water vapour promotes the reaction. According to the above equation a volume of carbon-dioxide amounting to $^1/_4$ of the volume of the hydrogen present is required; in practice a certain excess is used. Any residue of hydrogen may be removed by any known method.

For the pure components of the reaction there may be substituted technical gases, such as illuminating or water gas.

The following example illustrates the invention:—Water gas in which, by introducing hydrogen, the proportion of carbon monoxide; hydrogen has become 1 : 5, is conducted at about 290° C. over a nickel contact mass prepared in the usual manner and then the amount of methane is increased by adding carbon monoxide in further contact furnaces while periodically eliminating water vapour and restoring the required proportion of carbon monoxide; hydrogen. At desired places in the system of contact furnaces there is then introduced instead of a further quantity of carbon monoxide a quantity of carbon dioxide corresponding with $^1/_4$ of the hydrogen still present. The mixture is then exposed in the following contact furnaces to a temperature of about 320° C. while between the several furnaces the water vapour is eliminated by cooling or in any other manner. The excess of carbon dioxide may be removed by absorp-

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tion by lime or the like, and the residue of hydrogen by heated cupric oxide, for instance.

Ordinary illuminating gas may be used after it has been freed from heavy hydrocarbons and any substance impeding the catalysis, regard being had to the composition of the illuminating gas which is generally 30 *per cent.* of methane, 10 *per cent.* of carbon monoxide and 50 *per cent.* of hydrogen.

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 :—

The modification of the manufacture described in Specification No. 146,110, wherein, after a high concentration of the methane has been attained, the excess of hydrogen is converted into methane by conducting the mixture together with a quantity of carbon-dioxide corresponding with about $^1/_4$ of the amount of hydrogen, over heated contact bodies while periodically separating the water vapour, whereupon, if required, the residue of the hydrogen and the excess of carbon-dioxide are removed by any known method.

Dated this 22nd day of June, 1920.

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