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PATENT SPECIFICATION

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COMPLETE SPECIFICATION.

Improved Manufacture of Methane.

We, FARBWERKE vorm. MEISTER LUCIUS & BRÜNING, 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:—

It is known that carbon monoxide can be reduced to methane by hydrogen in presence of a catalyst, such as palladium, nickel or cobalt, at a suitable temperature; also that by using only the theoretically necessary proportion of hydrogen a part of the carbon monoxide decomposes with separation of carbon and formation of carbon dioxide, and that this inconvenience can be avoided only by using at least five volumes of hydrogen to one of carbon monoxide. However, in this manner there is obtained only a mixture containing methane and hydrogen in the proportion of 1 : 2.

We have found that by re-conducting the mixture of gases, obtained by the known process, through reaction furnaces after having, before the entry into each furnace, introduced carbon monoxide in a quantity not exceeding $^1/_5$ of the volume of hydrogen still present, there can be attained an increase of the amount of methane up to an approximate proportion of methane: hydrogen =3 : 1. A further treatment with freshly introduced carbon monoxide has no practical effect. However, if before each renewed catalysis, the water vapour is removed from the mixture, the hydrogen can be reduced to a very small proportion. At the same time by thus removing the water vapour there is obtained the advantage of diminishing the volume and consequently lessening the space required for further reaction.

The present invention is based on the foregoing observations.

In practice, the process is for instance carried out in the following manner:—

A constant stream of hydrogen is conducted at about 300° C. through a set of, say, eight contact-furnaces placed in series and charged with pumice carrying nickel rendered active in the usual manner; between every two furnaces is a well-acting condenser for separating the water-vapour, and before each contact furnace there is a conducting pipe for carbon monoxide and a measuring device through which this gas enters in quantity which in no case exceeds $^1/_5$ of the quantity of hydrogen entering that furnace. For easy control of working it is advantageous to introduce into each of the first furnaces the same quantity of carbon monoxide and to diminish this quantity in the remaining furnaces. The following illustrates the working method:—

Into the first furnace constantly enters for instance 100 litres of hydrogen per minute and within the same time there are introduced into each of the first six furnaces 5 litres of carbon monoxide. The quantity of hydrogen then diminishes in each consecutive furnace as follows:—

Furnace No.	1	2	3	4	5	6	7	8 residue
Litres of H_2	100	85	70	55	40	25	10	4 1.6
„ „ CO	5	5	5	5	5	5	2	0.8

[Price

Thus if into each of the furnaces 1—6 are introduced 5 litres of carbon monoxide, the quantity of hydrogen originally used diminishes in each furnace by $3 \times 5 = 15$ litres according to the equation: $CO + 3H_2 = CH_4 + H_2O$. Into furnace 7 only 10 litres of hydrogen enter. This quantity is not sufficient to reduce 5 litres of carbon monoxide without decomposition of a part of the carbon monoxide. Therefore only 2 litres of carbon monoxide are introduced into furnace 7 and only 0.8 into furnace 8, that is to say $^1/_5$ of the available amount of hydrogen. Thus, 32.8 litres of methane and 1.6 litres of hydrogen issue per minute from furnace No. 8, which corresponds with a percentage of 95.4 of methane and 4.6 of hydrogen. The introduction of ever decreasing quantities of carbon monoxide may, of course, still be continued. The last remainder of hydrogen can easily be removed in any other manner, for instance by passing the gas over heated cupric oxide or by liquifying the methane by cooling.

In the place of pure hydrogen, purified illuminating gas may be used containing about 30 *per cent.* of methane, 10 *per cent.* of carbon monoxide and 50 *per cent.* of hydrogen. For the carbon monoxide may be substituted purified water gas consisting of about equal parts of carbon monoxide and 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:—

1. The manufacture of methone by causing a mixture of carbon monoxide and hydrogen to react by passing it through a set of furnaces containing heated contact bodies, care being taken that in this gas mixture entering each furnace the amount of the carbon monoxide does not exceed about $^1/_5$ of the hydrogen present and that the water vapour is eliminated between every two furnaces.

2. The process referred to in Claim 1, conducted with technical gases containing carbon monoxide and hydrogen.

Dated this 21st day of June, 1920.

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