NOTE.—*The application for a Patent has become void.*
This print shows the Specification as it became open to public inspection.

PATENT SPECIFICATION

Convention Date (France): Feb. 7, 1925. **247,176**

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Complete not Accepted.

COMPLETE SPECIFICATION.

Method for the Utilisation of Methane.

I, GEORGES PATART, 50, rue Spontini, Paris, France, a citizen of the French Republic, 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:—

Methane, which is the most common hydrocarbon gas, whether obtained as natural gas escaping from the ground in coal or oil fields, or as forming the main part—after hydrogen—of the gases from coal distillation, or as resulting from other natural or artificial chemical reactions, has only been utilised on an industrial scale by the combustion—as complete as possible—for lighting or heating purposes, or by its combination with chlorine.

A known method has been proposed for the transformation of methane into methyl alcohol or formaldehyde by an oxidation, which is more or less restricted, by the action of molecular oxygen, but this method, in its proposed form, has not as yet afforded industrial results.

This invention relates to an indirect process of a most reliable nature by which a chemical combination of great value can be obtained by the use of methane. For this purpose, the methane is first burned incompletely by means of a volume of oxygen which is equal to or very slightly in excess of one-half the volume of the methane and this affords an incomplete combustion which is represented as follows:

1) $CH_4 + {}^1/_2O_2 = CO + 2H_2 + 11.4$ calories

[*Price* 1/-]

and the resulting mixture ($CO + 2H_2$) is itself utilised to produce methyl alcohol, or homologous oxygenated compounds of carbon, according to known methods of catalysis under pressure, and the reactions may be represented by an equation having the form

2) $n(CO) + 2nH_2 = C_nH_{2n+2}O + (n-1)H_2O$ which, for $n=1$, will produce methyl alcohol, so that I obtain, in two stages, the following reaction:

3) $n(CH_4) + n/2\ O_2 = C_nH_{2n+2}O + (n-1)H_2O$ and this, for $n=1$, will afford $CH_4 + {}^1/_2O_2 = CH_4O$ (methyl alcohol), which reaction has already been above mentioned but cannot be obtained directly without practical difficulties of a most serious nature.

To obtain in a regular manner the incomplete combustion of the methane according to equation 1), certain special precautions are to be taken, according to the present invention, and in particular the combustion must take place in the presence of a mass which is maintained at a very high temperature (about 1000 degrees C.) so that the oxygen will not preferably combine with the hydrogen, which should produce water vapour with a deposit of carbon, or that the carbon shall not be oxidised to form carbone dioxide rather than carbon monoxide. To obviate these two drawbacks, the most practical method consists in injecting the mixture of the two gases (methane and oxygen) in the above indicated proportions, upon an incandescent mass which might consist of any suitable refractory substance but which prefer-

ably consists of a solid fuel such as wood, charcoal, or coal, but coke is to be preferred for this purpose. The gases pass through this incandescent mass, which is given a proper thickness, for instance 0,50 to 0,75 metres, and preferably in the horizontal direction, between two apertures which are symmetrically disposed at the lower part of a chamber containing the fuel, such as coke, and whose size is as large as desired. At the gas outlet there will be collected a gaseous product, the composition whereof corresponds substantially to the above mentioned composition of two volumes of hydrogen for one volume of carbon monoxide and whose temperature is about from 800° to 1000° C. Should carbon monoxide be slightly in excess, this can be readily compensated by adding the corresponding amount of hydrogen which can be either supplied from a separate source or obtained by the treatment—by any suitable known process—of the whole or a part of the gas originally produced. The heat produced by the combustion (even incomplete) of the methane will usually be sufficient to compensate for the heat losses and to maintain the mass of coke in the incandescent state. The incandescent mass is to consist of a fuel for the case in which the heat produced by the combustion of the methane should not compensate for the heat losses, and in such case it will be sufficient to slightly increase the amount of oxygen in the insufflated gas, and should it be necessary, the proportion of the hydrogen supplied will also be increased in proportion.

From this standpoint, it will be advantageous to preliminarily heat the gases employed (i.e. methane and oxygen) to the highest possible temperature, and preferably by means of the hot gases discharged from the apparatus. I may further prevent the excessive heating of the nozzles serving for the injection of the said gases, by a water circulation around the said nozzles.

The coke (or other fuel constituting the incandescent mass) also serves to compensate, by the combustion of a corresponding part thereof, for the other heat losses which may occur; but if the operation is properly performed, the said losses will be very small so that only a very small amount of fuel will be thus consumed.

The ash from this combustion, may be either drawn off by the current of gas, and then retained by any known means, or may be melted directly in the apparatus by means of a periodical injection of an excess of oxygen and may be discharged, in the liquid state, through a suitable conduit disposed at the bottom of the fuel recipient. In this latter case, it is advisable to add a small amount of flux according to the composition of the ash.

In the case in which—as may sometimes occur in the catalytic synthesis of alcohols at a high temperature—there are formed new quantities of methane during the combination of the carbon monoxide with the hydrogen, this methane may be separated by any known means, for instance by partial liquefaction, and may then be treated as above mentioned in order to again form the mixture of hydrogen and carbon monoxide.

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

1. Method for utilising methane by transforming the same into a hydro-oxygenated compound of carbon, which consists in first effecting an incomplete combustion of the methane so as to obtain a gaseous mixture containing substantially two hydrogen molecules for one carbon monoxide molecule, and then in causing the combination of said hydrogen and carbon monoxide by any known method whereby one or more hydro-oxygenated compounds of carbon is or are produced, for instance by catalysis under pressure.

2. Method according to Claim 1 wherein the incomplete combustion of the methane is effected in the presence of a mass of refractory material maintained at a very high temperature, e.g. 1000° C.

3. Method according to Claim 2 wherein the mixture of methane and oxygen is injected upon a mass of an incandescent material and passes through the same preferably in the horizontal direction.

4. Method according to Claim 3 wherein the said mass consists of a solid fuel, preferably of coke.

5. Method according to Claim 1 wherein a small additional amount of hydrogen is added to the gaseous mixture produced by the incomplete combustion of the methane, so as to compensate for any amount of carbon monoxide in excess.

6. Method according to Claim 4 or 5 wherein an additional amount of oxygen is added to the initial mixture of methane and oxygen, so as to provide for the combustion of a predetermined portion of said fuel mass, the amount of additional hydrogen being proportionally increased if necessary.

7. Method according to Claim 1 where-

in the initial gases (methane and oxygen) are preliminarily heated to the highest possible temperature.

8. Method according to Claim 6 whereing a further additional amount of oxygen is injected in a periodic manner so as to provide for the melting of the ash produced during the combustion of the fuel mass.

9. Method for utilising methane substantially as described.

Dated this 28th day of January, 1926.

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