

# PATENT SPECIFICATION



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## COMPLETE SPECIFICATION.

### Improved Manufacture of Methane.

We, FARBERWERKE VORM. MEISTER LUCIUS & BRÜNING, of Höchst a/Main, Germany, a German company, 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:—

In Specification No. 146,110 is described a process for manufacturing methane of high concentration or pure from carbon monoxide and hydrogen, which consists in conducting the mixture (obtained in the known manner by passing carbon monoxide and an excess of hydrogen over heated contact substances) through a series of further contact furnaces; the water being eliminated between the several furnaces and carbon monoxide being introduced in such quantity that the amount of carbon monoxide in the mixture does not exceed about  $\frac{1}{5}$  of the amount of hydrogen present. The present invention relates to a modification of, or an improvement in that described in the said specification.

Owing to the strongly exothermic reaction between carbon monoxide and hydrogen the regulation of the temperature is not very simple, it requires, if it is to be effected by external cooling, very efficient cooling devices. By the present invention the temperature is regulated entirely or partly by sufficiently diluting the gases undergoing reaction by means of gases which play no part in the process. In the last phases of the process described in the said specification, that is to say, in the reaction occurring in the last furnaces, the gases have already been considerably diluted by the finally formed

methane; however, if required, the reaction may in this case be still better regulated by a further dilution. In the earlier stages of the process there is greater need to pay attention to the development of heat. The means adopted may be of two kinds. They may consist in adding gases which are indifferent to the process, preferably methane of high concentration or pure, or in adding less carbon monoxide than the quantity indicated by the proportion carbon monoxide : hydrogen = 1:5. In the latter case the dilution is effected by the gases present in the mixture, namely, in the first reaction phase substantially by hydrogen which while the reaction is in progress, is exchanged in ever increasing proportion for methane. The dilution of the gases by methane renders it possible to apply the process of the said specification continuously in a single furnace. Indeed, we have found that the process in question may be carried out in one single furnace by circulating the mixture, in which case it repeatedly passes through one and the same contact furnace, the water present being always eliminated after the contact furnace while carbon monoxide is introduced, before the mixture re-enters the contact zone, in a quantity not exceeding  $\frac{1}{5}$  of the hydrogen contained in the mixture. In the present process the temperature can also be regulated as above described, by suitable dilution of the gases entering into reaction. This process can be applied continuously by introducing into a gas mixture diluted with methane and circulated as above described, before it enters the contact zone, not only the quantity of carbon monoxide correspond-

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ing with the hydrogen present in the mixture, but also by introducing further quantities of carbon monoxide and hydrogen (in the proportion of about 1:5) into the mixture. The quantity of highly concentrated methane corresponding with the increase of the total volume can then be drawn off continuously after the contact furnace. In order that the composition of the methane thus produced may be uniform, the quantities of the whole of the carbon monoxide and hydrogen to be introduced continuously before the contact furnace must be taken in a proportion corresponding with the proportion 1:3 theoretically required for the reaction, while a further small excess of hydrogen is added, corresponding with very small quantity of hydrogen which is continually drawn off together with the concentrated methane after the contact furnace. By using a correspondingly large quantity of the methane serving as diluent, it is possible to produce methane of any desired percentage strength, from which the last remainder of hydrogen may then be removed by any known method.

When eliminating the water of the reaction by cooling, it is advantageous to use the heat removed for pre-heating the gases to be re-introduced into the reaction.

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. In a process described in Specification No. 146,110 the modification which consists in regulating the temperature of the reaction partly or entirely by diluting the mixture with gases which are indifferent to the process.

2. In the modification referred to in Claim 1, diluting the mixture with methane.

3. In the modification referred to in Claim 1, diluting the mixture by reducing the quantity of carbon monoxide introduced.

4. In the processes described in Specification No. 146,110, or in the modification referred to in Claim 1, re-introducing the mixture repeatedly into the same contact furnace instead of introducing it into a larger number of contact furnaces, the water being separated and the necessary quantity of carbon monoxide and, if required, hydrogen, being introduced during each circuit.

5. In the process conducted as referred to in Claim 4, taking the quantities of the carbon monoxide and hydrogen to be freshly introduced at each circuit in the approximate proportion of 1:3.

Dated this 10th day of August, 1920.

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