[Second Edition.]

PATENT SPECIFICATION

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PROVISIONAL SPECIFICATION.

Improvements in or relating to the Manufacture of Methyl Alcohol.

I, HENRY DREYFUS, of 8, Waterloo Place, London, S.W. 1, a citizen of the Swiss Republic, do hereby declare the nature of this invention to be as follows:—

This invention relates to the manufacture of methyl alcohol from mixtures of or containing carbon monoxide and hydrogen.

It has been known since 1913—1914 (Badische Anilin & Soda Fabrik in No. 20,488 of 1913) that when mixtures of carbon monoxide and hydrogen, usually mixtures in which the carbon monoxide is preponderant or largely preponderant, are submitted to high temperature and high pressure, hydrocarbons together with a series of oxygenated aliphatic products are obtained. In this process a large number of products are produced together, each in relatively small amounts, and the industrial recovery and separation of these products is a matter of extreme difficulty and even impossibility. A number of processes have since been proposed, according to which, by the selection of a particular proportion of carbon monoxide and hydrogen, particular classes of catalysts, and particular conditions of working, it has been attempted to modify the reaction so as to produce particular organic compounds as main products.

Now, whereas the main basis of the theory of the previous researches has been that the production of methyl alcohol as exclusive or practically exclusive product is due to the use of excess hydrogen, I have now found that for the production of methyl alcohol as exclusive or main product, it is not necessary to have an excess of hydrogen, and that, subject to the choice of catalyst and temperature employed, it is possible to obtain methyl alcohol as exclusive or principal product even with a reduced amount of hydrogen.

In order to perform this reaction in the sense indicated it is sufficient that the quantity of hydrogen relatively to the quantity of carbon monoxide present in the mixture should be in about the proportion ordinarily present in water gas, or, if the hydrogen and carbon monoxide are separately produced, that one uses them in such proportions as about one molecule of carbon monoxide to about one molecule of hydrogen, and further that one employs zinc oxide as catalyst, which substance may be supposed to be capable of initiating combination of the hydrogen and carbon monoxide to form formaldehyde and owing to its having at the same time a basic character, is capable of transforming this into methyl alcohol by splitting off carbon monoxide.

It is understood that the proportion of hydrogen relatively to carbon monoxide may be lower than that above mentioned, but round about this.

It is likewise understood that the hydrogen and carbon monoxide may be separately produced or that one may employ water gas or other industrial or other gaseous mixtures containing the gases in the desired proportions.

The reaction is performed under heat and pressure.

The temperature should not exceed about 350°—450° C. and is preferably kept between about 200° and 300° C.

The pressure employed varies according to the catalysts used and may be up to 200 atmospheres and more, but generally, a pressure of about 50 to 150 atmospheres is used.

Based on the foregoing method of pro-

cedure, it is possible to produce in a continuous process and practically quantitatively methyl alcohol.

The reaction is preferably executed in a copper system, implying copper-lined apparatus or apparatus of other suitable metals, such as aluminium, etc., or other materials, according to the temperatures used.

In executing the process for the production of methyl alcohol the speed of passage of the gases has also an influence, as, if the gases are passed through at relatively low speed, hydrocarbons and higher alcohols may be formed in substantial quantities or even as main products.

Dated this 13th day of June, 1925.

T. L. WHITEHEAD,
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COMPLETE SPECIFICATION.

Improvements in or relating to the Manufacture of Methyl Alcohol.

I, HENRY DREYFUS, of 8, Waterloo Place, London, S.W. 1, a citizen of the Swiss 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:—

This invention relates to the manufacture of methyl alcohol from mixtures of or containing carbon monoxide and hydrogen.

It has been known since 1913—1914 (Badische Anilin & Soda Fabrik in No. 20,488 of 1913) that when mixtures of carbon monoxide and hydrogen, usually mixtures in which the carbon monoxide is preponderant or largely preponderant, are submitted to high temperature and high pressure, hydrocarbons together with a series of oxygenated aliphatic products are obtained. In this process a large number of products are produced together, each in relatively small amounts, and the industrial recovery and separation of these products is a matter of extreme difficulty and even impossibility. A number of processes have since been proposed, according to which, by the selection of a particular proportion of carbon monoxide and hydrogen, particular classes of catalysts, and particular conditions of working, it has been attempted to modify the reaction so as to produce particular organic compounds as main products.

Now, whereas the main basis of the theory of the previous researches has been that the production of methyl alcohol as exclusive or practically exclusive product is due to the use of excess hydrogen, I have now found that for the production of methyl alcohol as exclusive or main product, it is not necessary to have an excess of hydrogen, and that, subject to the choice of catalyst and temperature employed, it is possible to obtain methyl alcohol as exclusive or principal product even with a reduced amount of hydrogen.

In order to perform this reaction in the sense indicated it is sufficient that the quantity of hydrogen relatively to the quantity of carbon monoxide present in the mixture should be in the proportion ordinarily present in water gas, or, if the hydrogen and carbon monoxide are separately produced, that one uses them in the proportion of one molecule of carbon monoxide to one molecule of hydrogen, and further that one employs zinc oxide as sole catalyst, that is to say, without admixture with other methanol-forming catalysts. Zinc oxide without such additions is capable of initiating combination of the hydrogen and carbon monoxide, probably to form formaldehyde, and owing to its having at the same time a basic character, is capable of forming methyl alcohol, probably by splitting off carbon monoxide from formaldehyde.

It is understood that the proportion of hydrogen relatively to carbon monoxide may be somewhat lower than that above mentioned.

It is likewise understood that the hydrogen and carbon monoxide may be separately produced or that one may employ water gas or other industrial gases or other gaseous mixtures which contain the gases in the prescribed proportions, or in which the proportions of the gases have been adjusted to the prescribed proportions.

The reaction is performed under heat and pressure.

The temperature should not exceed about 350°—450° C. and is preferably kept between about 200° and 300° C.

The pressure employed varies according to the catalysts used and may be up to 200 atmospheres and more, but gener-

ally, a pressure of about 50 to 150 atmospheres is used.

Based on the foregoing method of procedure, it is possible to produce in a continuous process and practically quantitatively methyl alcohol.

The invention is illustrated by the following example, but is not limited thereto.

EXAMPLE.

A mixture containing 50 parts by volume of carbon monoxide and 40—50 parts by volume of hydrogen is passed under a pressure of about 80—120 atmospheres through a chamber filled with granules of zinc oxide, or with zinc oxide obtained by precipitation, and maintained at a temperature of about 250—300° C. The products are separated by condensation and the methyl alcohol is purified by distillation if necessary. The undecomposed gases are recirculated through the system for further reaction.

The reaction is preferably executed in a copper system, implying copper-lined apparatus, or in apparatus of other suitable metals, such as aluminium, etc., or other materials, according to the temperatures used.

In executing the process the speed of passage of the gases has also an influence, as, if the gases are passed through at relatively low speed, hydrocarbons and higher alcohols may be formed in substantial quantities or even as main products.

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. Process for the manufacture of methyl alcohol, characterised in that water gas or other gaseous mixtures containing hydrogen and carbon monoxide in the proportion of one molecule or slightly less than one molecule of hydrogen relatively to one molecule of carbon monoxide are subjected to the action of heat and pressure in presence of zinc oxide without admixture with other methanol-forming catalysts.

2. Process according to Claim 1, characterised in that the reaction is performed at temperatures under about 350° to 450° C. and preferably at temperatures of from about 200° to 300° C.

3. Process according to Claim 1 or 2, characterised in that the reaction is performed under pressures of up to 200 atmospheres or more and especially under pressures of 50 to 150 atmospheres.

4. Process for the manufacture of methyl alcohol from carbon monoxide and hydrogen substantially as described.

5. Methyl alcohol when prepared or produced by the processes herein described and claimed.

Dated this 12th day of April, 1926.

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Reference has been directed, in pursuance of Section 7, Sub-section 4, of the Patents and Designs Acts, 1907 and 1919, to Specification No. 20,488 of 1913.

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