

[Second Edition.]

NOTE.—The application for a Patent has become void.

This print shows the Specification as it became open to public inspection.

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PATENT SPECIFICATION



Convention Date (France): Mar. 22, 1917.

157,047

Application Date (in United Kingdom): Aug. 25, 1917. No. 12,234 / 17.

Complete not Accepted.

COMPLETE SPECIFICATION.

Improvements in or relating to the Manufacture of Formic Aldehyde or Methyl Alcohol or Mixtures thereof.

I, HENRY DREYFUS, of 59, Schutzenmattstrasse, Basle, Switzerland, Chemist, 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 the Specification of my Application for Patent Number 10,591 filed 23 July 1917 I have described a process for the manufacture of formic aldehyde or methyl alcohol, or mixtures thereof, according to which carbon monoxide is passed through one or more vessels, tubes or chambers in presence of hydrogen under suitable conditions to effect the hydrogenation of the carbon monoxide to formic aldehyde or methyl alcohol or a mixture of these products according to the quantity of hydrogen employed, the hydrogen being employed in about the quantity theoretically required for the formation of the formic aldehyde or methyl alcohol or mixture, or slightly in excess of such quantity, the vessels, tubes or chambers being lined filled or provided with platinum or other suitable contact substances, or such substances being in some cases omitted. In particular the reaction was therein stated to be conducted at temperatures not exceeding about 400° C., and preferably between about 100° and 250° C.—300° C., the resulting vapours of formic aldehyde or methyl alcohol or both being condensed after leaving the reaction tubes, vessels or chambers, and the carbon monoxide and

hydrogen being preferably introduced into the reaction tubes, vessels or chambers by leading a current of hydrogen into a current of carbon monoxide gas at a suitable temperature and conducting the mixed gases into the reaction tubes, vessels or chambers in a suitably regulated current. The mixed gases may, moreover, be heated to a suitably regulated temperature before entering the reaction vessels, tubes or chambers.

In the Specification of another Application for Patent Number filed 25 August 1917 I have proposed according to one method to carry out such reactions at a temperature between about 20° C. and about 100° C., and according to another method to carry out the hydrogenation with or without contact substances, at a low temperature, and even cooling if required, for example at temperatures between 20 and -20° C.

In the before mentioned processes the speed of the reaction is increased in proportion as the quantity of contact substance is increased.

I have found that a very efficient contact substance for effecting the reaction at ordinary temperature is constituted by platinum sponge or platinum black, such as is obtained by reducing the chloroplatinates with aldehydes (formaldehydes) and then precipitating the product with alkali; it has likewise been found that nickel obtained by precipitation of the nitrate with alkali and reduction at

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ordinary temperature or below about 100° C. is highly effective in regard to its reducing power.

Further it has been found advantageous for effecting the desired reaction between the carbon monoxide and the hydrogen, to mix the contact substance, such as nickel, platinum *etc.* with a very porous supporting material, in effecting the reduction to nickel, platinum, *etc.* upon the porous material, in order thus to secure the largest possible surface for the contact substance.

For example pumice stone may be taken and impregnated with a solution of the salt of nickel or platinum, then dried and reduced. Or better, a solution of silicate of sodium or sulphate of magnesium may be taken and mixed for example of nitrate of nickel; the whole is then evaporated as rapidly as possible and the resulting mass dried at a high temperature, a very porous material being thus obtained. This material is then reduced before employment, preferably in the reaction tubes or vessels.

The hydrogenation of the carbon monoxide may be effected in cylindrical iron vessels, tubes or chambers filled with the porous material containing the contact substances.

The carbon monoxide and hydrogen may be admitted at the bottom of the contact or reaction apparatus, taking care to have a good mixture. The mixed gases combine and the formic aldehyde or methyl alcohol or mixture thereof distils off at the top if the temperature is higher than the boiling point of the reaction product or products. The mixture may evidently be introduced at top, but in this case, where contact substances are used and methyl alcohol is formed, it is preferable to employ a temperature above the boiling point of the methyl alcohol, as otherwise the contact substance would be successively washed towards the bottom of the apparatus.

In cases where heating is employed for the reaction I may with advantage employ, instead of cylindrical vessels of more or less large diameter for the contact or reaction vessels,—tubes, such as iron tubes providing together the same surface of contact; these tubes are placed vertically and connected together at top and bottom by a collector. The lower or upper part of the tubes carries a

perforated plate the perforations of which have such a diameter that the plates of the tubes together ensure the homogeneous distribution of the passing gases.

In the present invention, as in my said other applications for patent, the carbon monoxide may be replaced by carbonic acid, but such employment is not so satisfactory from an economical point of view, as the carbonic acid requires more hydrogen for the hydrogenation to formic aldehyde or methyl alcohol or mixture thereof.

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. In processes such as hereinbefore referred to for the hydrogenation of carbon monoxide or carbon dioxide by hydrogen to formic aldehyde or methyl alcohol or a mixture thereof, in presence of a contact substance, the employment as contact substance of platinum sponge or platinum black such as is obtained by reducing chloroplatinates with aldehydes and precipitating the reduction product with alkali; or of nickel obtained by precipitation of nitrate of nickel with alkali and reduction at ordinary temperature or below about 100° C.

2. In processes such as hereinbefore referred to for the hydrogenation of carbon monoxide or carbon dioxide by hydrogen to formic aldehyde or methyl alcohol or a mixture thereof, in presence of contact substance, the employment of a contact substance mixed with a porous material serving as a support.

3. A process according to Claim 2, characterised in that the nickel, platinum or other contact substance is reduced in or upon the porous supporting material.

4. In processes such as hereinbefore referred to for the hydrogenation of carbon monoxide or carbon dioxide by hydrogen to formic aldehyde or methyl alcohol or a mixture thereof, the employment of contact substances prepared in a manner substantially as described.

Dated this 25th day of August, 1917.

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