PATENT SPECIFICATION



Application Date: March 4, 1926. No. 6103 26.

275,284

Complete Accepted: Aug. 4, 1927.

COMPLETE SPECIFICATION.

Improvements in the Manufacture and Production of Organic Compounds Containing Oxygen.

I, JAMES YATE JOHNSON, a British subject, of 47, Lincoln's Inn Fields, in the County of London, Centleman, do hereby declare the nature of this invention (which has been communicated to me from abroad by the L. G. Farben-industrie Aktiengesellschaft, of Frankfort-on-Main, Germany, a corporation organized according to German laws) 10 and in what manner the same is to be performed, to be particularly described and ascertained in and by the following statement :-

It is already known that methanol and 15 organic compounds containing oxygen, mainly alcohols of a higher order than methanol can be manufactured by the hydrogenation of carbon catalytic monoxide or carbon dioxide or mixtures 20 of both under pressure and at an elevated

temperature.

My foreign correspondents have now found that the said organic compounds of a higher order than methanol, for 25 example isobutyl alcohol and other higher alcohols, can be obtained in a specially advantageous manner by limiting the amount of carbon dioxide in the gases employed for the catalytic treatment. The amount of carbon dioxide in the gases should generally not exceed about 3 per cent. The removal of carbon dioxide may be carried out by absorbing it by means of solvents, for example water, or binding it chemically while maintaining the gases under the high pressure used in the reaction. As the reaction proceeds with a mixture of carbon monoxide and hydrogen at an elevated temperature passed over a contact mass in such a manner that only port of the gases is transformed into organic compounds containing oxygen, the unaltered gases are carried back to the contact mass in a circular manner after the reaction product has been separated. Owing to

the fact that in the production of higher alcohols and the like carbon dioxide is formed, it is necessary, according to the present invention even when the fresh gas has been practically freed from carbon dioxide, to remove carbon or periodically continuously dioxide from the circulating gases either completely or down to a small percentage in 55 order to continuously obtain good yields of the higher molecular products. As the fresh gas is in most cases freed from carbon dioxide by washing with water under pressure, it is sufficient in such cases to 60 add only part of the circulating gases to the fresh gases prior to their purification either continuously or periodically; the total percentage of carbon dioxide in the reacting gases is by this means kept suffi-ciently low.

In the gases subjected to catalytic treatment, the carbon monoxide may be replaced in part and the hydrogen completely or partially by alcohols, such as 70 ethyl alcohol, which means that the gas mixture may consist of alcohol and carbon monoxide with or without hydrogen. The hydrogen may also be replaced com-pletely or partially by hydrocarbons, especially unsaturated or readily decom-

posable hydrocarbons. The following examples will further illustrate how the invention may be carried out in practice, but the invention is 80 not limited to these examples.

EXAMPLE 1.

Water gas consisting of 30.2 per cent. of carbon monoxide, 65.6 per cent. of hydrogen and 4.2 per cent. of nitrogen is circulated under a pressure of 200 atmospheres in a system comprising a contact mass consisting of an intimate mixture of equi-molecular proportions of zinc oxide, chromium oxide and manganese oxides heated to a temperature of from 475° to 500° Centigrade. The carbon dioxide formed in the reaction is continuously removed from the circulating 5 gas by means of absorption with water under a pressure of 200 atmospheres. The gases leaving the reaction vessel are cooled whereby higher alcohols, chiefly isobutyl alcohol, are obtained with a permanently excellent yield jointly with methanol and other organic compounds containing oxygen and small quantities of water.

Example 2.

15 A gas mixture consisting of 10.4 per cent. of carbon monoxide, 85.6 per cent. of hydrogen and 4 per cent. of nitrogen is charged under a pressure of 200 atmospheres with so much of ethyl alcohol 20 vapour that the partial pressure of the latter amounts to about 15 to 20 atmospheres. The resulting gas and vapour mixture is introduced into a high pressure reaction vessel and passed at from 25 480° to 490° Centigrade over a contact mass as described in Example 1.

The gases leaving the reaction vessel which now contain carbon dioxide, are cooled under pressure, whereby ample quantities of higher alcohols, especially isobutyl alcohol are separated along with methanol, unaltered ethyl alcohol and some water. The gases are then reintroduced into the reaction vessel, after the carbon dioxide is absorbed by pure water, or water containing some ammonia, under a pressure of about 200 atmospheres, so that the gases contain at the most 2 to 3 per cent. of carbon dioxide.

I am aware of Specifications Nos. 20,488/13 and 229,714 and do not claim anything described or claimed therein; nothing is said in the said specifications as to the necessity of continuously keep-

ing the gases free from or low in earbon 4 dioxide.

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 50 claim is:—

1. A process for the manufacture and production of organic compounds containing oxygen consisting in circulating a mixture of hydrogen and carbon 55 monoxide with only a small amount of or completely free from carbon dioxide under a high pressure and at an elevated temperature over a contact mass suitable for the manufacture of methanol and other organic compounds containing oxygen and removing continuously or periodically from the circulated gas the carbon dioxide formed by absorbing it by means of solvents, or by binding it chemically under a high pressure, so as to keep the said gas low in carbon dioxide or completely free therefrom.

2. In the process claimed in the preceding claiming clause employing a gas mixture consisting of carbon monoxide and yapour of an alcohol with or without hydrogen.

3. In the process claimed in the preceding claiming clauses substituting part or all of the hydrogen by hydrocarbons.

or all of the hydrogen by hydrocarbons.

4. The process for the manufacture and production of organic compounds containing oxygen substantially as described in the foregoing examples.

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5. Organic compounds containing oxygen when produced in accordance with the preceding claiming clauses.

Dated this 4th day of March, 1926.

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W.C. 2,

Agents.

Abingdon: Printed for His Majesty's Stationery Office, by Burgess & Son. [Wt. 56a. -125/11/1928.]