AMENDED SPECIFICATION

Reprinted as amended under Section 8, and in accordance with the Decision of the Superintending Examiner, acting for the Comptroller-General, dated the seventh day of February, 1940, under Section 11, of the Patents and Designs Acts, 1907 to 1939.

PATENT SPECIFICATION

Convention Date (Germany): Nov. 30, 1935.

473,932

Application Date (in United Kingdom): Nov. 26, 1936. No. 32419/36.

Complete Specification Accepted: Oct. 22, 1937.

COMPLETE SPECIFICATION

Improvements in the Manufacture and Production of Valuable Hydrocarbons and their Derivatives containing Oxygen from Carbon Monoxide and Hydrogen

We, I. G. FARBENINDUSTRIE AKTIENGESELLSCHAFT, of Frankfort-on-Main, Germany, a Joint Stock Company, organized under the Laws of Germany, 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:—

It has already been proposed to produce gaseous, liquid and/or solid hydrocarbons and their derivatives from carbon monoxide and hydrogen at elevated temperature and under ordinary or slightly increased or reduced pressure in the presence of catalysts. Among catalysts hitherto used for this reaction are the metals of the 8th group of the periodic system, especially iron, cobalt or nickel or their compounds, together with activating additions. In order to attain a good distribution of the catalysts the latter are preferably employed together with carrier substances, such as kieselguhr, silica gel and the like. Before use the catalysts are in some cases subjected to a reducing treatment with hydrogen at temperatures up to about 450° Centigrade. When carrying out the said conversion of carbon monoxide with hydrogen under increased pressures, however, the use of the said catalysts has proved unsatisfactory in some respects. Thus they give rise to a strong formation of methane and also rapidly become destroyed by the formation of carbonyl compounds. Furthermore, carbon block is frequently formed and causes clogging up of the apparatus.

It is already known from specification No. 317,808 to deposit 1 part of ferric nitrate, 2 parts of cupric nitrate and 0.1 part of potassium carbonate on 10 parts of pumice stone and then to decompose the nitrates by a brief calcination. This catalyst is reduced at 300° Centrigrade for 10 hours at ordinary pressure in a current of hydrogen whereupon a mixture of 32 per cent. of carbon monoxide, 64 per cent. of hydrogen and 4 per cent. of nitrogen is passed over it at 200 atmospheres and at 350° Centigrade, higher hydrocarbons and benzines being *inter alia* thus produced.

We have now found that the conversion of carbon monoxide and hydrogen, at an elevated temperature, into liquid, solid and/or gaseous hydrocarbons or their derivatives containing oxygen, such as alcohols, proceeds smoothly and without trouble when working in the presence of catalysts obtained, before the said conversion, by thermally treating iron compounds (which expression is understood to include also materials containing the same) in the presence of reducing gases at temperatures above 600° Centigrade, but below the melting or sublimation point of the particular compound employed or of the resulting iron.

Suitable iron compounds are in particular the oxides or those compounds, (hereinafter referred to as oxidic compounds) which, like nitrates, hydroxides, or carbonates, may be converted into oxides by merely heating them to high temperatures, if necessary up to 1000° Centigrade and above. The thermal treatment of the said iron compounds takes place at temperatures above 600° Centigrade, for example at 800°, 1000° Centigrade or more, but always below the melting point of each particular iron compound or resulting iron. The treatment is performed in the presence of gases hav-

[*Price* 1/-]

ing a reducing action, in particular hydrogen or gases containing hydrogen.

The thermal treatment of the iron compounds may take place at any desired pressure, as for example at ordinary pressure as well as at elevated pressures of 2, 5, 10 or 100 atmospheres or more.

Of the iron compounds the precipitated hydroxides or the oxides obtained, for example, by the decomposition of the nitrates are preferably employed. The activity of the catalysts may be increased by the addition of other substances, such as the oxides or hydroxides of aluminium, silicon dioxide, kieselguhr and the like. Other additions having an activating action, as for example the compounds of copper, titanium, manganese, tungsten, molybdenum, chromium, thorium, cerium, zirconium or other rare earths may also be employed with advantage. It has been found that in many cases the presence of suitable amounts of oxides, hydroxides or carbonates of alkali metals or alkaline earth metals may favourably influence the formation of hydrocarbons.

The conversion of carbon monoxide and hydrogen is effected at temperatures above 150°, preferably between 200° and 450° Centigrade. Atmospheric pressure or reduced pressure may be employed, but it is preferable to employ elevated pressures, such as 5, 10, 20, 50, 100 atmospheres or more.

The following Example will further illustrate how the said invention may be carried out in practice, but it is to be understood that the invention is not restricted to this Example.

EXAMPLE.

Precipitated iron hydroxide to which 5 per cent. of aluminium hydroxide have been added, is heated to 850° Centigrade in a current of hydrogen. After cooling, a mixture of carbon monoxide and hydrogen (in the ratio of 1:2) is led over the catalyst thus obtained at a pressure of 12 atmospheres and at a temperature of between about 220° and 320° Centigrade. In this manner, not only gaseous but also liquid and solid hydrocarbons are formed in good yields, the said hydrocarbons containing a small amount of compounds containing oxygen.

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 we are aware of our Patent No. 465,668 and of our Patent No. 468,434 and do not claim the use in the present process of a catalyst obtained by treating previously fused ferrosoferric oxide with a reducing gas at a temperature above 300° Centigrade but subject to this disclaimer what we claim is:—

1. A process for the conversion of carbon monoxide and hydrogen into liquid, solid and/or gaseous hydrocarbons or their derivatives containing oxygen which comprises contacting carbon monoxide and hydrogen at a reacting temperature with a catalyst prepared, before the said conversion by thermal treatment in the presence of reducing gases of an iron compound at a temperature above 600° Centigrade but below the melting or sublimation point of the iron compound or of the iron resulting from this treatment.

2. In the process as claimed in claim 1, preparing the catalyst from an iron oxide or from an iron compound which by mere heating may be converted into the oxide.

3. In the process as claimed in claim 1 or 2, carrying out the thermal treatment in the presence of hydrogen.

4. The process for the conversion of carbon monoxide and hydrogen into hydrocarbons and their oxygen-containing derivatives substantially as described in the foregoing Example.

5. Hydrocarbons and their oxygen-containing derivatives when obtained by the process particularly described and ascertained.

Dated this 26th day of November, 1936.

J. Y. & G. W. JOHNSON,
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Agents.

Leamington Spa: Printed for His Majesty's Stationery Office, by the Courier Press.—1940.