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PATENT SPECIFICATION

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COMPLETE SPECIFICATION.

Catalytic Process for the Manufacture of Hydrogen.

I, RENE JEAN AUGUSTE GRENIER, of 3, Avenue Girardot, Montmorency, Seine-et-Oise, France, a citizen of the French 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:—

The present invention has reference to a catalytic process for the manufacture of hydrogen. It consists of the use of carbon in the porous state, and particularly wood-charcoal, as a catalyser for the chemical reaction in which carbon monoxide reacts upon steam with the production of hydrogen, which reaction is represented by the following equation:—

$$CO + H_2O = CO_2 + H_2$$

This reaction is reversible and exothermic in the direction of the passage from the first member to the second. It is known that a reduction of temperature has for result to displace the state of equilibrium in this same sense favourable to the production.

The applicant is aware that it has already been proposed to utilize porous carbon, alone or in mixture with other bodies in the capacity of catalyser, for catalytic reactions, either between carbon monoxide and ammonia for the manufacture of hydrocyanic acid, or between carbon monoxide and hydrogen for the manufacture of formaldehyde or hydrocarbons, or again in other catalytic reactions of hydrogenation or dehydrogenation.

He is also aware that it is known to obtain hydrogen by catalytic reaction between carbon monoxide and steam, in presence of compounds of iron as catalyser supported upon porous carbon.

The applicant has ascertained that carbon in the porous state, and particularly wood charcoal, constitutes for this reaction a catalyser of which the use affords clearly characterized industrial advantages over that of catalysers hitherto used, as the experimental results hereafter set out will definitely prove:—

In the manufacture of hydrogen, according to the reaction:

$$CO + H_2O = CO_2 + H_2,$$

experiments carried out have enabled me to ascertain that by passing through an iron tube filled with wood-charcoal and heated by a gas burner, a mixture composed of one volume of carbon monoxide and about six volumes of steam, there is collected a mixture of hydrogen, of carbonic acid gas, of carbon monoxide and steam.

As indicated above, the proportion of these gases varies with the temperature. The proportion of carbonic anhydride in particular passes through a maximum at about 350°—380° C., at the same time that that of carbon monoxide passes through a minimum.

In particular and without the least restriction of the generality of the invention by this experiment, quoted like the following ones solely by way of example, if there is passed at a temperature of 375° C. into the catalyser tube a mixture having the following composition:—

CO	99 volumes
CO_2	1 volume
H_2O (as steam)	600 volumes

there is collected a gaseous mixture of which the analysis indicates the following composition by volume:—

CO_2	51%
H_2	49%
CO	Traces

The reaction is thus practically complete. In the course of this experiment, the duration of contact of the gases with the catalyser has not exceeded two minutes, and it will be possible in practice to reduce it in a certain degree.

The slight excess of carbonic acid gas relative to the theoretical quantity corresponding to the complete reaction would seem to arise from the last traces of this gas adsorbed in the wood-charcoal in the normal state, which traces it gives up with difficulty.

It has also been ascertained that the presence of an excess of hydrogen is not capable in practice of hindering the reaction by mass action.

In support of the foregoing and likewise by way of example, the applicant points out that by adding an excess of hydrogen to the gases introduced into the catalyser tube, there is obtained at the

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exit from this tube a gaseous mixture, of which the following composition has been found in the course of two experiments differing from one another as regards the proportion of hydrogen added and the speed of passage of the gases in the catalyser tube :

First experiment :—

CO_2	18.1%
CO	0.4%
H_2	81.5%

Second experiment :—

CO_2	10%
CO	0.6%
H_2	89.4%

The percentage of hydrogen added and the speed of passage of the gases were greater in the second experiment than during the first experiment.

Thus, the use of wood-charcoal as a catalyser permits of lowering the temperature of the reaction and thereby displacing its equilibrium in the direction of increased yield.

Moreover, this new catalyser, compared with those usually employed for this reaction, permits of obtaining a gaseous mixture which contains a greater proportion of carbonic acid gas and consequently of hydrogen, for the same expenditure of steam, or a mixture containing the same proportion of carbonic acid gas and consequently of hydrogen, with a less expenditure of steam, from which it follows that there is an economy of steam.

It being given that an excess of hydrogen does not in practice prevent the reaction from taking place, there may be utilized in place of a mixture of carbon monoxide and steam, a mixture of water gas and steam.

The presence of other gases, in particular nitrogen, ethylene, etc., not being harmful, there might also be utilized a mixture of industrial gases containing carbon monoxide (poor gas, coke oven gas, etc.) and steam.

It results from the foregoing that wood-charcoal presents from the industrial point of view a great advantage as a catalyser, for several reasons of which the principal are the following :—

(1). Lowering of the temperature beyond which the speed of the reaction in question becomes appreciable.

(2). For a given temperature, increase of the speed of the reaction.

(3). Long duration of use of the catalyser by reason of its resistance to chemical agents.

(4). Low cost price of the catalyser.

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 :—

Catalytic process for the manufacture of hydrogen, starting from a mixture of carbon monoxide and steam, characterized by the fact that there is utilized as catalyser carbon in the porous state, for example wood-carcoal.

Dated the 23rd day of May, 1927.

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