## PATENT SPECIFICATION



Convention Date (France): Feb. 7, 1925.

247,177

Application Date (in United Kingdom); Jan. 28, 1926. No. 2580 26. Complete Accepted: April 28, 1927.

COMPLETE SPECIFICATION.

## Method for the Synthetic Production of Higher Alcohols.

I, Georges Patart, 50, rue Spontini, Paris, 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 relates to a process of catalytic reduction of carbon monoxide under high pressure which permits to direct the reaction towards the formation of alcohols having a greater molecular weight than methyl alcohol.

It is a known fact that the reaction 15 products of the catalytic reduction of carbon monoxide under high pressure usually contain, in addition to methyl alcohol, a certain proportion of higher alcohols, chiefly when the contact between the gas mixture and the catalyzer lasts for a sufficiently long time. But as a rule the proportion of higher electrols thus obtained is much smaller than the amount of methanol 25 which is formed at the same time.

In certain cases, it may be desired to transform the major part or almost the whole of the gas mixture into alcohols of a high molecular weight.

For this purpose, the method according to the invention consists essentially in separating, from the condensed products resulting from the catalytic treatment under pressure of the initial mix-Bu ture of carbon monoxide and hydrogen, the inethanol and the other products whose molecular weight is below that of the alcohols sought for, and in subjecting the products separated from the condensate, in the vapour form, to a further catalytic treatment under pressure, either by introducing said vapour under pressure into the original mixture of carbon monoxide and hydrogen before it

enters the methanol synthesiser, or into a similar mixture of carbon monoxide and hydrogen before it passes into another methanol synthesizer or by circulating said vapour alone and under pressure through a reaction chamber containing the same catalyzer or a similar methanol forming catalyzer.

In carrying out the invention, a gas mixture containing carbon monoxide and hydrogen is circulated under pressure through a reaction chamber or synthesiscr containing a methanol forming catalyzer. The gaseous mixture issuing from the synthesiser is passed through a condenser where the condensable products, containing methanol, higher alcohols, and other products of varying molecular weights are condensed. The non condensed gas containing carbon monoxide and hydrogen is then passed, in a well known 65 manner, through the same or another synthesiser in order to undergo a further catalytic treatment under pressure, and to yield further amounts of condensable products; the portion 70 of gas which entered into reaction is replaced by fresh gas, according to the usual practice.

The condensate is treated in order to separate from the higher alcohols, the methanol and the products whose moleonlar weight is below that of the alcohols sought for, and the products thus separated are subjected to a further cutalytic treatment under pressure.

For this purpose, and in order to obviate all condensation, or even all variations in temperature, which would disturb the reaction, care must be taken to preliminarily vaporize the products 85 thus separated and heat said vapour to the temperature of the methanol synthesiser; the latter will have a temperature above the critical tempera-

ture of the vapour thus introduced, so that no condensation will take place.

In carrying out the invention into practice, the liquid may be forced-pre-5 ferably in a continuous manner—into a closed chamber which is heated to a point slightly above its critical tempera-ture, and thence, the liquid thus vaporized is supplied into the reaction 10 chamber.

The second treatment of these vaporized products of lower molecular weight may be performed in different manners:

1.) The hot vapors of methanol and 15 other lower products are re-introduced into the initial gas mixture of carbon monoxide and hydrogen under pressure before it enters the methanol synthesiser.

2.) The hot vapors of methanol and other lower products are introduced into a similar mixture of carbon monoxide and hydrogen under pressure which is passed through methauol a. then synthesiser.

S.) The hot vapors of methanol and other lower products are circulated alone and under high pressure through a reaction chamber containing the same catalyzer as the methanol synthesiser, or 30 a similar methanol forming catalyzer.

The gas mixture issuing from the methanol synthesiser where the second treatment has been performed are then condensed and the further amounts of higher alcohols produced are recovered

by the known methods. It has been already claimed a process for the manufacture of oxygenated organic compounds of a higher order 40 than methanol which consists in passing a mixture of an oxide of carbon and a gas containing hydrogen at an elevated pressure and temperature over a contact mass capable of reacting with the said gas mixture to form methanol and preferably such masses as contain substantial amounts of a non-reducible metallic oxide, but which are substantially free from iron, nickel and cobalt, the opera-50 tion being performed at such a moderate speed as to lead to the predominating formation of oxygenated organic compounds of a higher order than methanol,

mainly higher alcohols. It has also been claimed the manufac-

ture of valuable organic compounds consisting in subjecting the oily liquids which can be obtained by the catalytic hydrogenation of carbon oxides under pressure and containing oxygenated oxygenated organic bodies chiefly alcohols of a higher order than methyl alcohol, or fractions of such liquids, to treatment with condensing agents, I do not claim these processes as my invention.

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. Method for the synthetic production of higher alcohols, by the capalytic reduction of carbon monoxide under pressure, which consists in separating, from the products of the condensation following the reduction of the initial mixture, the methanol and the other products having a lower molecular weight than the higher alcohols which are to be produced, and in subjecting the products thus separated from the condensate, in the vapour form to a further catalytic treatment under pressure, either by introducing said vapour under pressure into the original mixture of carbon monoxide and hydrogen before it enters the methanol synthesiser or into a similar mixtuare of carbon monoxide and hydrogen before it passes into another methanol synthesiser, or by circulating said vapour alone and under pressure through a reaction chamber containing the same cetalyzer, or a similar methanol

forming catalyzer. 2. Method according to Claim wherein the separated condensation products are heated to the temperature of the reaction chamber before they are

supplied into the latter.

3. Method according to Claim 2 where- 100 in the temperature of the reaction chainber is higher than the critical temperature of the mixture of the separated condensation products.

4. Method for the synthetic produc- 105 tion of higher alcohols substantially as

described.

Dated this 29th day of January, 1926. MARKS & CLERK.