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(12) Patent:

(54) ALCOHOL PRODUCTION

(54) PRODUCTION D'ALCOOL

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ABSTRACT:

CLAIMS: Show all claims

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It is a known fact that in the catalytic reduction of carbon monoxide by the action of catalyzers such as a mixture of zinc oxide with potassium oxide, or copper oxide or copper, and under high pressures, varying from 100 to 500 atmospheres, there may be obtained, in addition to methyl alcohol, a certain quantity 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 alcohols thus obtained is much smaller than the amount of methyl alcohol which is formed at the same time.

But in certain cases it may be desired to transform the major part or even the whole of the gas mixture into alcohols of a high molecular weight or even into a given alcohol having the maximum obtainable molecular weight.

It has been found, according to the present invention, that the reaction may be directed towards the formation preferably of an alcohol having a greater molecular weight than methyl alcohol.

The method according to the invention consists essentially in separating, from the condensed products resulting from the above mentioned catalytic reduction under pressure, the methyl alcohol as well as the other products whose molecular weight is below that of the alcohol sought for, and in subjecting these separated products to a further catalytic action either by again introducing them in the state of vapour into the gas mixture under reaction or into a different gaseous mixture or by circulating them over another catalytic mass in a special reaction chamber containing the same catalyzer

or any other catalyzer which may be more suitable for the purpose.

To obviate all condensation, or even all variations in temperature, which would disturb the reaction, care must be taken to preliminarily heat the liquid to be introduced so as to bring it to the temperature of the reaction chamber, which will as a rule be above the critical temperature of the liquid to be again introduced, so that no condensation will take place.

For this purpose, the liquid may be forced -preferably in a continuous manner- into a closed chamber which is heated to a point slightly above its critical temperature, and from which, after vaporisation, it will be supplied into the said reaction chamber at a suitable point. The supply of the liquid will be regulated according to the composition of the collected products.

The following example of application of the process will clearly illustrate a method of carrying the invention into practice.

EXAMPLE.- A catalyzer composed of an intimate mixture of:

Chromium oxide	35 %	by weight
Potassium hydroxide	25 %	" "
Zinc oxide	40 %	" "

is kept at a temperature of about 450° C. and water gas is circulated in a closed path upon said catalyzer, maintaining a pressure of about 300 atmospheres. The composition of the water gas used is approximately:

Hydrogen	50 %	by volume
Carbon monoxide	40 %	" "
Carbon dioxide	5 %	" "
Nitrogen	5 %	" "

The portion of the gas which is condensed into hydrogenated compounds is continuously replaced by a cor-

responding addition of fresh water gas in order to maintain the above pressure of 300 atm. The gas issuing from the catalytic apparatus is cooled by a circulation of cooling water and the condensed liquid, after dehydratation, has the following composition:

Methanol and compounds of lower boiling point than methanol	(60% by weight
Ethanol	1.5 % "
Propanol	3% by weight
Isobutanol	10% " "
Pentanol	2% " "
Hexanol and compounds of higher boiling point than hexanol	{ 10% " "
Organic acid in the free or combined state	{ 5% " "
Tars and non-defined products	1% " "

By fractional distillation of the liquid products thus separated from the gas, methanol (or the major part of the methanol) is collected separately. Said methanol which is substantially in the pure state, is sent back under pressure to the gaseous circuit, under the form of vapor, superheated to 300° C, said methanol replacing the corresponding amount of water gas which is capable of maintaining the pressure of 300 atm.

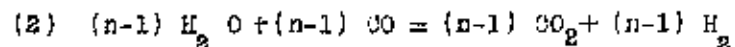
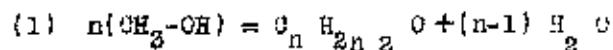
In these conditions, it is found that the composition of the condensate, as well as its total amount is not substantially modified; this result shows that a material portion of the methanol which is supplied to the gaseous circuit (say about 40 %) is transformed into alcohol or hydro-oxygenated compounds of higher boiling point or molecular weight.

If the methanol is thus separated and re-introduced in a continuous manner into the gaseous circuit, the fi-

nal result is that the products formed by the transformation of the water gas are constituted only by products of higher boiling point than methanol.

It should be noted that the initial composition of the water gas is not altered by the addition of methanol, that is the relative proportions of carbon monoxide and hydrogen remain substantially constant although the proportion of hydrogen relatively to carbon monoxide is larger in methanol (combined) than in water gas (free).

This is due to the formation, which has been experimentally verified, of carbon dioxide, in relatively large proportion, consequently to the production of the heavier alcohols. The reactions which take place in the catalytic chambers, may be represented by the following equations:



The second reaction is a reversible reaction or reaction of equilibrium whereby the proportion of hydrogen which would tend to increase in the gaseous mixture due to the addition of methanol, is limited by the presence of an excess of carbon dioxide. On the other hand, carbon dioxide and water vapor will be condensed as soon as their partial pressure will exceed their tension of vapor at the temperature of the cooling device, (the maximum proportion of carbon dioxide in the gaseous mixture is about 22 to 25 %) in the conditions of composition temperature and pressure above indicated.

The above indications will explain the fact that the gaseous mixture has always the composition indicated for water gas, irrespectively of the amount of methanol

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which is re-circulated through the catalytic chambers, and, therefore, the fact that the amount and the composition of the condensate remain unchanged. Obviously, there will be an increase in the proportion of nitrogen, the latter being periodically discharged at the proper time.

C L A I M S

1.- Method for the synthetic production of higher molecular organic compounds containing oxygen which consists in reducing carbon monoxide by hydrogen in the presence of a catalyzer and under a high pressure, condensing the products thereby obtained, separating from the condensate the methanol and other compounds having a lighter molecular weight than the higher compounds to be produced and subjecting the methanol and other compounds thus separated again to a catalytic reduction under a high pressure.

2.- Method for the synthetic production of higher molecular organic compounds containing oxygen which consists in reducing carbon monoxide by hydrogen in the presence of a catalyzer and under a high pressure, condensing the products thereby obtained, separating from the condensate the methanol and other compounds having a lighter molecular weight than the higher compounds to be produced and re-introducing the methanol and other compounds thus separated into the mixture of carbon monoxide and hydrogen to be treated.

3.- Method for the synthetic production of higher molecular organic compounds containing oxygen which consists in reducing carbon monoxide by hydrogen in the

presence of a catalyzer and under a high pressure, condensing the products thereby obtained, separating from the condensates the methanol and other compounds having a lighter molecular weight than the higher compounds to be produced, vaporising the said methanol and other separated compounds, mixing the vapors thus produced with a gaseous mixture containing carbon monoxide and subjecting the mixture thus obtained again to a catalytic reduction under a high pressure.

4.- Method for the synthetic production of higher molecular organic compounds containing oxygen which consists in reducing carbon monoxide by hydrogen in the presence of a catalyzer and under a high pressure, condensing the products thereby obtained, separating from the condensates the methanol and other compounds having a lighter molecular weight than the higher compounds to be produced, vaporising the said methanol and other separated compounds, mixing the vapors thus produced with a gaseous mixture containing carbon monoxide and hydrogen and subjecting the mixture thus obtained again to a catalytic reduction under a high pressure.

5.- Method for the synthetic production of higher molecular organic compounds containing oxygen which consists in reducing carbon monoxide by hydrogen in the presence of a catalyzer and under a high pressure, condensing the products thereby obtained, separating from the condensates the methanol and other compounds having a lighter molecular weight than the higher compounds to be produced, vaporising the said methanol and other separated compounds, heating the vapors thus produced up to a predetermined temperature and subjecting the said vapors again to a catalytic reduction under a high pressure.

6.- Method for the synthetic production of higher molecular organic compounds containing oxygen which consists in reducing carbon monoxide by hydrogen in the presence of a catalyst and under a high pressure, condensing the products thereby obtained, separating from the condensates the methanol and other compounds having a lighter molecular weight than the higher compounds to be produced, vaporising the said methanol and other separated compounds and heating the vapors thus produced up to a temperature above the boiling ~~point~~ temperature of the heaviest of said separated compounds and subjecting the said vapors again to a catalytic reaction under a high pressure.