PATENT SPECIFICATION

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COMPLETE SPECIFICATION

A Process for the Manufacture of Alcohols

I, JON CRISTESCU, of the Polytechnic School, Timisoara, Roumania, a Roumanian Subject, 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:—

This invention relates to a process for the manufacture of alcohols from gas mixtures.

The known processes for manufacturing alcohols from gas mixtures have the disadvantage of requiring the use of high pressures and relatively high temperatures.

The object of this invention is to avoid these disadvantages which render difficult to the carrying out of the operations and increase the cost of the products obtained, by providing a process which renders possible the transformation of the gases into alcohols at normal pressure and at temperatures up to about 100°C.

According to the process of this invention for the manufacture of alcohols by mixing gases consisting of or containing hydrogen, methane or other hydrocarbons with gases consisting of or containing oxygen, air, or an oxide of carbon, one or both the gases containing a carbon compound, the mixtures of gases are introduced into boiling water at normal atmospheric pressure, in the presence of catalysts comprising metallic oxides (other than calcium oxide) and/or metal powders, or charcoal or coke washed with alkali and the gases and steam produced are passed before condensation through vessels charged with catalysts comprising metallic oxides (other than calcium oxide) and/or metal powders, or charcoal or coke washed with alkali.

The liquid obtained by the condensation of the vapours is then fractionally distilled in order to separate the water, the alcohols and the ketones.

The free gases are conducted through washing vessels in order to cause them to deposit the aldehydes.

The reactions taking place in the water during the boiling are favoured by the physical phenomenon of the gases becoming loaded with fine drops of liquid, electrified by the friction caused by the molecular stirring in the immediate neighbourhood of the metallic powders, metallic oxides, or charcoal or coke washed with alkali, which act as catalysts.

The accompanying diagrammatic drawing illustrates an elevation partly in section of an apparatus which may be employed for carrying the invention into effect:

The gas mixture is introduced through I, the pressure being controlled by the manometer M. The flow is controlled by the cock R and is verified by the meter G.

The apparatus in which the reactions take place consists of two tanks B and C separated by a perforated metal plate F, the tank C being provided with a thermometer T. The metallic powders, metallic oxides or charcoal or coke washed with alkali are placed in tank B into which the water is introduced through A at the same rate as it is removed by evaporation. At the beginning of the process ⅔ of the volume of the tank B should be filled with distilled water. The additional catalysts are placed in tank C.

The apparatus is heated through the bottom plate E. The vapours formed are cooled by the cooler K and condensed in the vessel D.

The free gases are conducted through the water-washer S in order to deposit the aldehydes therein and are then collected in a vessel (not shown) in which their composition may be determined.

The liquid obtained in the vessel D is fractionally distilled to obtain the alcohols.

The following examples illustrate how the process of the invention may be carried into effect:

1. A mixture of 3 volumes of methane and one volume of air is introduced into boiling water contained in tank B, under ordinary pressure, whereby a mixture of aldehydes and alcohols is formed. Aluminium powder is employed in tank B and granulated copper oxide is employed in tank C. Instead of the granulated copper oxide, a metallic powder, preferably aluminium, alone or mixed with

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other metallic powders, as well as with small pieces of metal alloys, free from arsenic, lead and sulphur may be employed.

When the catalyst in tank B consists of finely powdered lead dioxide, copper oxide or granulated bauxite is used in tank C and methyl alcohol is obtained.

The reactions which take place may be explained by the transformation of methane into carbon monoxide and hydrogen, in the presence of water vapour with subsequent formation of methyl alcohol according to the following equations:

$$CH_4 + H_2O = CO + 3H_2$$
$$CO + 3H_2 = CH_3OH + H_2$$

If charcoal powder or a cobalt oxide or other metallic oxide is used as catalytic material in tank B, a mixture of alcohols is obtained in which mixture ethyl alcohol predominates.

2. A gas mixture consisting of hydrogen and carbon dioxide containing an excess of hydrogen is treated employing in tank B as catalyst, charcoal powder or a metallic oxide preferably cobalt oxide, zinc oxide, chromium oxide, aluminium oxide or mixtures thereof and in tank C charcoal powder washed with sodium hydroxide or other alkali. Ethyl alcohol and acetaldehyde are obtained by a series of reactions in accordance with the following equations:

$$2CO_2 + 2H_2 = 2CO + 2H_2O$$
$$2CO + 3H_2 = C_2H_4O + H_2O$$
$$C_2H_4O + H_2 = C_2H_5OH$$

Instead of the charcoal, coke washed with sodium hydroxide and other alkali may be employed. Similarly metallic oxides may be employed instead of the coke, but in this case lower yields are obtained.

3. A mixture of gas consisting of one volume of carbon dioxide, one volume of methane and two volumes of hydrogen is introduced into the apparatus and treated using the same catalysts as in example 2. Ethyl alcohol and acetaldehyde are obtained.

4. A mixture of gases consisting of oxygen, hydrogen, carbon monoxide and carbon dioxide with or without hydrocarbons in which the volume of the hydrogen is equal to the sum of the volumes of carbon dioxide and carbon monoxide is passed into the water of the apparatus, in the presence of the catlysts employed in examples 2 and 3 and ethyl alcohol and acetaldehyde are obtained.

The presence of sulphur and nitrogen, does not disturb the reactions. It is, however, preferable to eliminate the sulphur.

The process of this invention thus enables alcohols to be obtained in an economical manner, by treating methane, suction gas, illuminating gas, as well as any gas produced by combustion.

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. A process for the manufacture of alcohols by mixing gases consisting of or containing hydrogen, methane or other hydrocarbons with gases consisting of or containing oxygen, air, or an oxide of carbon, one or both the gases containing a carbon compound, wherein the mixtures of gases are introduced into boiling water at normal atmospheric pressure, in the presence of catalysts comprising metallic oxides (other than calcium oxide) and/or metal powders, or charcoal or coke washed with alkali and the gases and steam produced are passed before condensation through vessels charged with catalysts comprising metallic oxides (other than calcium oxide) and/or metal powders, or charcoal or coke washed with alkali.

2. A process as claimed in claim 1, wherein the mixtures of gases comprise methane and air, and the catalysts comprise one or more metallic oxides (other than calcium oxide).

3. A process as claimed in claim 1 or 2, wherein the catalysts comprise aluminium powder or aluminium oxide and the catalysts through which the gases and the steam pass before condensation, comprise one or more granulated metallic oxides (other than calcium oxide).

4. A process as claimed in claim 1, wherein the mixtures of gases comprise carbon dioxide and hydrogen, the catalysts comprise aluminium powder, and the catalysts in the vessels through which the gases and steam flow before condensation, comprise pieces of charcoal, washed with an alkaline solution, preferably sodium hydroxide.

5. A process as claimed in claim 4, wherein the catalysts placed in the water comprise pulverised charcoal or one or more metallic oxides (other than calcium oxide).

6. A process as claimed in claim 1, wherein the gas mixtures comprise methane and carbon dioxide, and the catalysts employed are those claimed in claim 4 or 5.

7. A process as claimed in claim 1, wherein the gas mixtures comprise oxygen, carbon monoxide, carbon dioxide, hydrogen and hydrocarbons, and the catalysts employed are those

claimed in claim 4 or 5.

8. A process for the manufacture of alcohols substantially as described with reference to the examples given.

9. Alcohols when manufactured by the process claimed in any of the preceding claims or by an obvious chemical equivalent thereof.

Dated this 16th day of March, 1934.

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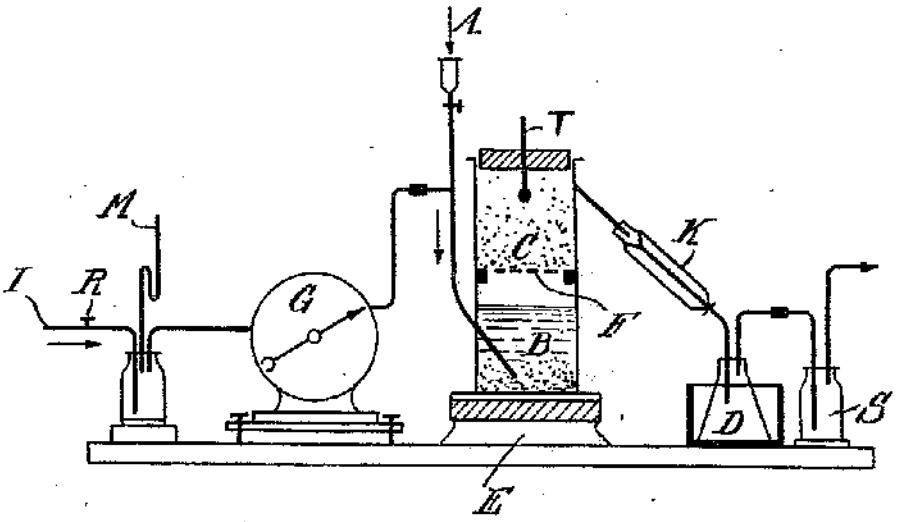
[This Drawing is a reproduction of the Original on a reduced scale.]



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