

## PATENT SPECIFICATION

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COMPLETE SPECIFICATION.



### Process for Manufacturing Formaldehyde from Gas Containing Hydrogen and Carbon Monoxide.

I, LUCIEN HENRI ROMAN, residing at 11, rue d'Enghien, Paris (Seine), France, a citizen of the Kingdom of Italy, 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 is known to obtain formaldehyde from gases containing hydrogen and carbon monoxide by processes in which the reaction takes place at low temperature and pressure, said processes involving the use of a catalyst, but the aldehyde formed at the outset of the reaction rapidly polymerises on contact with the catalyst, impregnates the latter and impairs and finally destroys the catalytic action so that the formation of the aldehyde soon ceases.

According to the present invention, the gas containing hydrogen and carbon monoxide, such as water gas, blast furnace gas etc., previously purified, is subjected, under normal pressure and at a temperature included between 130° and 200° C, to the action of a catalyst constituted by one or more of the following reduced metals or the oxides of said metals, viz. nickel, iron, cobalt, copper, platinum and palladium, while in order to regenerate the catalyst, the current of gas to be treated is discontinued periodically, and a current of nitrogen or oxide of nitrogen is then passed through the catalyst, the durations of flow of the reaction gas and regenerating gas being, respectively, 90 seconds (plus or minus 10 seconds) and 30 seconds (plus or minus 5 seconds).

It is current practice to recover gases containing hydrogen and carbon monoxide or to manufacture water gas, and such practices do not require describing. It is known that by means of suitable methods, mixtures of approximately 50% of CO and 50% of H can be obtained.

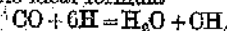
The proportions of the mixtures CO + H may, according to the products to be obtained, undergo any modifications, either by enrichment in H or impoverishment in CO.

Similarly the purification of said gases containing hydrogen and carbon monoxide

for the elimination therefrom of the harmful or useless elements such as H<sub>2</sub>S, H<sub>2</sub>As, CO<sub>2</sub> etc., is well-known and need not be described.

The gas containing hydrogen and carbon monoxide thus obtained and purified is passed into an apparatus heated to a suitable temperature (130° to 200° C.) in order to undergo therein a catalytic action on the following reduced metals or metallic oxides: nickel, iron, cobalt, copper, platinum and palladium. The gas is maintained in the apparatus at a pressure approaching the normal.

It is known that the mixture CO + H may under such conditions produce methane when the proportion of the gases CO and H is favorable. In the case in which the mixture CO + H does not correspond to the ideal formula



methane may still be formed, but the reaction is then only partial.

During the above transformation, it has been observed that formaldehyde is produced. But from this point the action of the catalyst slows down considerably and rapidly tends towards zero.

This action is due to the fact that the aldehyde formed gradually polymerises on the surface of the catalyst and polarises it without any chemical combination taking place however. Due to this polarisation of the catalyst, which prevents contact between it and the gas, the catalytic action gradually falls off and would finally cease unless measures were adopted to depolarise the catalyst.

In order, therefore, to depolarise and reactivate the catalyst, the flow of the gas to be treated is temporarily arrested and the catalyst is subjected to a current of nitrogen or oxide of nitrogen, the effect of which is to depolymerise the aldehyde deposited on the catalyst whereby the latter is depolarised and recovers its catalytic properties.

According to the present invention, the durations of alternate flow of the current of gas to be treated and of the current of catalyst reactivating gas are respectively 90 seconds (plus or minus 10 seconds) and 30 seconds (plus or minus 5 seconds).

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The formaldehyde formed is connected by the known methods and is eminently suitable for the manufacture of alkaline-earth saccharates or saccharates, in particular, and carbohydrates in general.

By way of example the process according to the invention for the manufacture of formaldehyde will be described hereafter, the apparatus used being the one shown in the accompanying drawing.

The water gas from the gas generator 1 is treated in a purifier 2 by an alkaline solution in order to remove its harmful components  $H_2S$  and  $CO_2$ . Said gas is then collected in a gasometer 3. The purified gas is passed through a nickel catalyst 4 of known type and constituted for example by nickel spirals covered with powdered nickel known as "reduced nickel". The catalysis chamber is heated to a temperature approaching  $175^\circ C.$  (+ or  $-10^\circ$ ). In said chamber the purified gas undergoes a transformation and the greater part of it passes into the state of formaldehyde. During said reaction, a slowing down of the catalytic action may be observed, which tends towards zero. The current of water gas through the catalyst is stopped after 90 seconds (+ or  $-10$  seconds) and is replaced by a current of nitrogen for a period of 30 seconds (+ or  $-5$  seconds). Said nitrogen is introduced, for example, by an additional pipe 5. The current of water gas is then passed again and so forth.

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. Process for the industrial manufacture of formaldehyde, according to which gases containing hydrogen and carbon monoxide, such as water gas, blast furnace gas, etc., purified beforehand, are subjected under normal pressure and at a temperature included between  $130^\circ$  and  $200^\circ C.$  to the action of a catalyst constituted by one or several of the following reduced metals or their oxides: viz. nickel, iron, cobalt, copper, platinum and palladium, while in order to regenerate the catalyst, the current of gas to be treated is discontinued periodically, and a current of nitrogen or oxide of nitrogen is then passed through the catalyst, the durations of flow of the reaction gas and regenerating gas being respectively 90 seconds (plus or minus 10 seconds) and 30 seconds (plus or minus 5 seconds).

2. Process for the industrial manufacture of formaldehyde, substantially as hereinbefore described.

Dated this 14th day of October, 1930.

For the Applicant,

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2nd Edition

*[This drawing is a full-size reproduction of the Original.]*

