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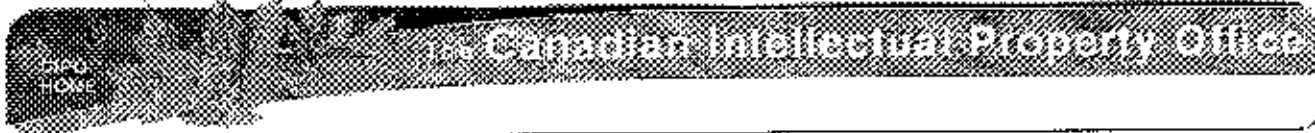
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(12) **Patent:**

(54) CARBOXYLIC ACID

(54) ACIDE CARBOXYLIQUE

Summary of Rights

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ABSTRACT:

CLAIMS: [Show all claims](#)

*** Note: Data on abstracts and claims is shown in the official language in which it was submitted.

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3 This invention relates to a process for the preparation of formic acid from carbon monoxide and steam and is directed particularly to the use of a new catalyst for the reaction.

It has been known that carbon monoxide and steam will react, in the presence of a suitable catalyst, to give formic acid. As this process is one which employs raw materials that are relatively inexpensive, it should, under favorable conditions, produce the acid at an exceptionally low cost. Its commercial success, however, will in no small part be determined by the catalyst used. Those which have been proposed heretofore have not been entirely satisfactory for commercial operation due to low yield, short life, and other economic considerations. Efforts of investigators in this art have been directed, therefore, to the discovery of catalysts having high activity and which, furthermore, favor the production of formic acid while tending to inhibit the formation of undesirable side products.

An object of the present invention is to provide new catalysts for the preparation of formic acid from carbon monoxide and steam having the above desirable characteristics. Other objects will hereinafter appear.

According to the present invention formic acid can be prepared from carbon monoxide and steam by passing a mixture of these gases over a catalyst which comprises an acid salt of the acidic oxides of elements such as, for example, phosphorus, arsenic, tungsten, molybdenum, uranium, chromium, vanadium, boron, silicon, and zirconium. As examples of such catalysts my invention covers the following and similar compounds; primary and secondary phosphates of zinc, calcium, magnesium, sodium, potassium, copper, cerium, thorium, etc., and the acid salts formed between these metals and the oxides of tungsten, molybdenum, etc. These catalysts may be used alone or as admixtures and may be either unsupported or supported upon the usual type of

catalyst support such, for example, as activated charcoal, Fuller's earth, kieselguhr, etc.

The carbon monoxide required for this synthesis may conveniently be derived from various commercial sources, such as, for example, water-gas, producer gas, etc., by liquefaction or other methods, and should likewise for the best results be relatively pure.

Inert gases, such as nitrogen, carbon dioxide etc., may be included with the reactants, this being advantageous in some cases from the standpoint of controlling the temperature of the exothermic reaction and of limiting the extent thereof, or it may be desired to restrict the overall conversion of the reaction for the sake of enhancing the relative yield of formic acid.

I prefer generally to conduct the reaction at pressures in excess of atmospheric, say from 25 to 900 atmospheres. The reaction proceeds over a wide range of temperatures employing the above described catalysts, depending upon the gaseous composition employed. Generally the desired conversion of the carbon monoxide and steam to formic acid can be obtained at a temperature of from 100-400°C, although I generally prefer to conduct the reaction in the range of from 200-300°C.

The following example will illustrate one method of practicing the invention, although the invention is not limited thereto.

Example 1 - A gaseous mixture consisting of 80 parts by volume of carbon monoxide, and 20 parts by volume of steam is passed at a pressure of 700 atmospheres and a temperature of 325°C. over an acid zinc phosphate catalyst. This catalyst may be prepared by adding to an aqueous phosphoric acid solution a suitable amount of zinc oxide and subsequently impregnating a support, such as activated charcoal, with this solution. After drying the

catalysts at approximately 120°C, it is ready for use. Upon condensation of the reaction product a condensate will be obtained containing formic acid.

Example 2 - A gaseous mixture comprising 95 parts by volume of carbon monoxide and 50 parts by volume of steam is passed over a primary calcium phosphate catalyst at a pressure of 200 atmospheres and a temperature of approximately 325°C. A good yield of formic acid admixed with other aliphatic carboxylic acids will be obtained.

The apparatus which may be employed for conducting the reaction may be ^{of}any conventional type and preferably one in which the temperature of exothermic reactions can be readily controlled at the desired temperature. Owing to the corrosive action of formic acid, the interior of the converter and conduits therefrom should preferably be protected. This may be accomplished by using glass or glass-lined apparatus or by coating the inner surfaces of the apparatus with chromium or silver or using for the construction of this equipment acid-resisting alloys of, for example, molybdenum, cobalt, tungsten, chromium, manganese, or nickel.

Various changes may be made in the method of employing the above catalysts for the preparation of formic acid from carbon monoxide and steam without departing from this invention or sacrificing the advantages that may be derived therefrom.

Having thus described my invention, what I claim is:-

1. In a process for the preparation of formic acid from a gaseous mixture of carbon monoxide and steam, the step which comprises passing the gaseous mixture over a catalyst comprising an acid salt of the acidic oxides of the elements selected from the group consisting of phosphorus, arsenic, tungsten, molybdenum, uranium, chromium, vanadium, boron, silicon and zirconium.

2. In a process for the preparation of formic acid from a gaseous mixture of carbon monoxide and steam, the step which comprises passing the gaseous mixture over an acid phosphate catalyst.

3. In a process for the preparation of formic acid from a gaseous mixture of carbon monoxide and steam, the step which comprises passing the gaseous mixture over a primary acid phosphate catalyst.

4. In a process for the preparation of formic acid from a gaseous mixture of carbon monoxide and steam, the step which comprises passing the gaseous mixture over a calcium acid phosphate catalyst.