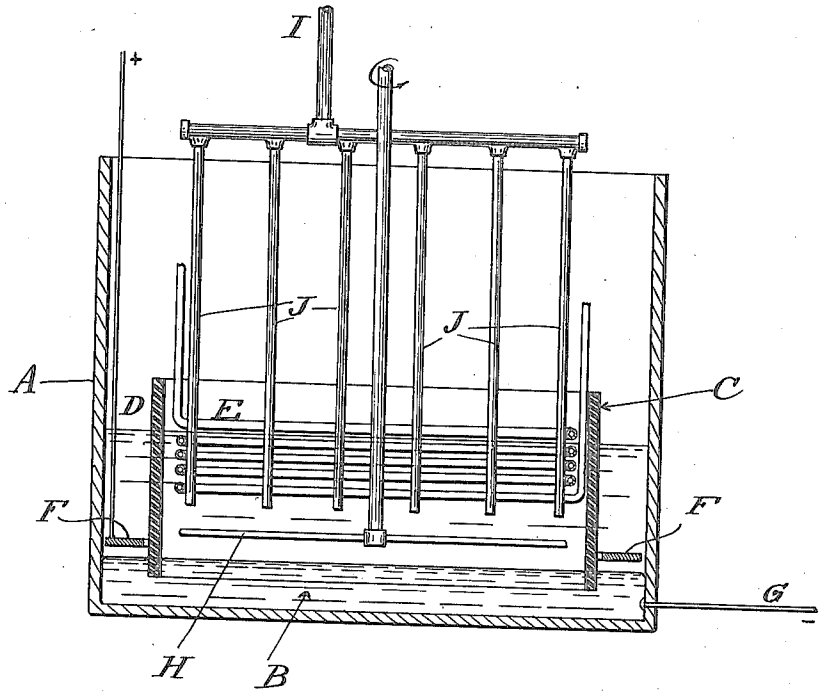
M. VAYGOUNY.

PROCESS OF MAKING FORMIC ACID OR ITS COMPOUNDS.

APPLICATION FILED JULY 23, 1910.

1,185,028.

Patented May 30, 1916.



Witnesses:
W. G. Leard
Rosina Jacobs

Mooshegh Vaygouny Inventor
By his Attorneys
Dinsmore, Wastick

UNITED STATES PATENT OFFICE.

MOOSHEGH VAYGOUNY, OF BERKELEY, CALIFORNIA, ASSIGNOR TO ROYAL BAKING POWDER COMPANY, A CORPORATION OF NEW JERSEY.

PROCESS OF MAKING FORMIC ACID OR ITS COMPOUNDS.

1,185,028. Specification of Letters Patent. Patented May 30, 1916.

Application filed July 23, 1910. Serial No. 573,546.

To all whom it may concern:

Be it known that I, MOOSHEGH VAYGOUNY, a citizen of the United States, residing at Berkeley, in the county of Alameda, State of California, have invented a new and useful Process of Making Formic Acid or Its Compounds, of which the following is a specification.

This invention relates to a process of making formic acid or its compounds from carbonic acid or its compounds and has as its objects the making of formic acid or its compounds in a manner which is commercially available, and more economical and efficient than any heretofore known.

My process embraces the reduction of carbonic acid by means of amalgams properly generated and used.

I describe my process in connection with the accompanying drawing showing a form of apparatus wherein the amalgam desired is made electrolytically in one chamber and used in another chamber.

The drawing is a diagrammatic representation of a double compartment cell wherein both the formation of the amalgam and its decomposition are carried out in a compact manner though still in separate chambers. It is intended to be illustrative only as it is obvious that the particular form of apparatus is not an essential feature of the process.

In the drawing, A indicates a suitable vessel, which may be made of any desirable non-conducting material, such as wood, of cylindrical, rectangular or other suitable shape, here shown as cylindrical.

B indicates a layer of mercury completely covering the bottom of vessel A.

C indicates a vessel conforming in shape with vessel A, open top and bottom, the bottom portion dipping into mercury B and reaching to nearly the bottom of vessel A. Vessel C may be supported in this position in any convenient manner.

D represents an electrolytic chamber constituted by the space between vessels A and C. E indicates a purely chemical or reaction chamber within vessel C. Chambers D and E are separated and kept so separated from each other by the layer of mercury B, which acts as a seal, so that the solutions in the two chambers cannot communicate with each other.

F indicates a suitable insoluble anode, such as platinum, and G the cathode terminal, the mercury B acting as the cathode.

H indicates a suitable stirrer, supported in any convenient manner, the function of which is to keep the solution to be reduced in a thorough state of agitation.

I is a pipe for conveying a stream of carbonic acid gas to a series of distributers J, J, discharging beneath the surface and within the body of the liquid to be contained in chamber E.

With a cell thus constituted, a suitable electrolyte, such as a strong solution of potassium hydrate, is introduced into the electrolytic chamber D. E is substantially filled with water kept saturated with carbonic acid from the distributers J, J, or otherwise. As a direct current is made to flow from the anode F to cathode B, an amalgam will form in the mercury within the chamber D. By diffusion, assisted mechanically by stirrer H, or by rocking the cell, if necessary, this amalgam is caused to travel from chamber D to reaction chamber E. Here the amalgam is rapidly attacked by the water and by the free-acidic carboxyl groupings

$$\left(-C\begin{matrix}O\\OH\end{matrix}\right)$$

present in the solution, tending thereby to generate potassium hydrate and hydrogen. The potassium hydrate combines with the excess of carbonic acid present, forming bicarbonate, while the hydrogen, tending to be liberated, acts at once in that nascent state and under the conditions obtaining, upon the carboxyl grouping

$$\left(-C\begin{matrix}O\\OH\end{matrix}\right),$$

thereby converting or reducing the latter into the grouping

$$\left(-C\begin{matrix}O\\H\end{matrix}\right)$$

of the formate molecule, in a very satisfactory manner. During the process of this reduction the contents of the reaction chamber are kept well stirred while simultaneously they are kept cool, if necessary, by means of a cooling device applied either externally or internally, as may be most convenient, the object of the cooling being merely to keep the temperature of the reacting agents from rising much above 40°

centigrade throughout the operation of the reduction.

When the current has passed for a sufficient length of time and a sufficient quantity of formate has thus been formed in the reaction chamber by means of the amalgam, the solution is withdrawn and may be further worked up in order to recover the formate present in any suitable manner. I have discovered a novel method of accomplishing this, consisting in removing the excess of bicarbonate by concentrating the solution until it becomes very strong in formate, when the bicarbonates are precipitated in consequence of the fact, which I have discovered, that in such stronger solutions of formates such bicarbonates are practically insoluble. At this point the bicarbonate is filtered off while the filtrate may further be evaporated down in order to obtain the formate in crystallized or solid form, when it can be utilized for any desired purpose; for instance, the formate may be decomposed by sulfuric acid, and the free formic acid liberated may be recovered by distillation in any suitable manner.

In practice, instead of having recourse to such partial evaporation to remove the excess of bicarbonate contained in the foregoing solution, the operation of reduction may be carried to such a point that the solution in the reaction chamber becomes wellnigh saturated with formate, in which case the bicarbonate precipitates in consequence of its insolubility under the conditions above mentioned. It is obvious that by having recourse to such a means, a solution is arrived at directly which is substantially a pure saturated solution of a formate, which may be worked up further as desired.

While I have illustrated my process in connection with the use of potassium hydrate, which I have found to be preferable, I do not limit myself to the use of that or any other compound of potassium, as the salts of other alkali and alkaline-earth metals, such as sodium, ammonium, calcium or magnesium, may be used. And where I use the term "alkali" in the claims, I intend to include alkaline-earth metals as well as alkali metals.

In order that the process illustrated by the foregoing may be the more successfully practised in the various forms which it may be desirable to give it to meet particular commercial conditions, I direct attention to certain special precautions or conditions which should be observed to secure the best results:—First: The amalgam employed should be applied in a dilute form, otherwise the reduction is very uneconomical and unsatisfactory. Second: The amalgam of potassium I find to be preferable to others in many respects. Third: The solution to be reduced should at all times contain an excess of carbonic acid in order to insure the presence at all times of an excess of the acidic or free carboxyl grouping in the solution, otherwise no satisfactory production of formates will take place under these conditions. Fourth: The temperature at which the reduction is carried out had best be kept below 40° centigrade or so, room temperatures being very satisfactory. Fifth: The solution in the reduction chamber should be kept agitated during the reduction to prevent loss of hydrogen which would otherwise tend to evolve in a free condition, should the said carboxyl groupings be absent from the immediate vicinity of the reacting surfaces of the amalgam by local impoverishment of the solution.

Among the important advantages of practising my process in the preferred form above described are:—First: By carrying on the reduction of the carbonic acid in a chamber separated from the electrolytic action, instead of directly in an electrolytic cell wherein the mercury would act as a cathode, I avoid the destructiveness of the current at the anode, which the use of such cells necessarily would involve (even if porous diaphragms were introduced), to such an extent as to render that arrangement unsuited for practical manufacturing purposes. Second: So, too, by employing amalgams formed electrolytically, I avoid the disadvantages which would necessarily be involved in using purely chemically prepared amalgams, such as the further oxidation products both of the mercury and the other metal employed, which would tend to diminish the efficiency of the hydrogen generated, and moreover this would involve cumbersomeness, lack of economy and unsuitability to practical operation.

In order to obtain the most satisfactory results, the amalgam should be made electrolytically and decomposed chemically in a chamber wherein electrolysis does not take place.

What I claim is:—

1. A process of making formates comprising the introduction of an alkali amalgam to an aqueous solution maintained supplied with free carbonic acid in excess.

2. A process of making formates comprising the introduction of an electrolytically-formed alkali amalgam to an aqueous solution maintained supplied with free carbonic acid in excess.

3. A process of making formates comprising the introduction of an electrolytically-formed alkali amalgam to an aqueous solution maintained supplied with free carbonic acid in excess and kept free from the amalgam-forming current.

4. A process of making formates comprising the reduction of free carbonic acid by means of hydrogen resulting from the

action of an electrolytically-formed alkali amalgam.

5. A process of making formates comprising the reduction of free carbonic acid by means of hydrogen resulting from the action of an alkali amalgam formed electrolytically.

6. A process of making potassium formate comprising the reduction of free carbonic acid by means of hydrogen resulting from the action of potassium amalgam formed electrolytically.

7. A process of making formates, comprising producing electrolytically an alkali amalgam in the solution in one chamber, preserving said amalgam substantially intact and transferring it to a solution comprising a hydrogen-containing substance and carbonic acid in another chamber, and decomposing said amalgam in said chamber and thereby reducing the carbonic acid therein.

8. A process of making formates, comprising producing electrolytically a potassium amalgam in the solution in one chamber, preserving said amalgam substantially intact and transferring it to a solution comprising a hydrogen-containing substance and carbonic acid in another chamber, and decomposing said amalgam in said chamber and thereby reducing the carbonic acid therein.

9. A process of making formates comprising the reduction of carbonic acid by the action of an alkali amalgam in dilute form.

10. A process of making formates comprising the reduction of a solution of carbonic acid by means of the action of an alkali amalgam generated electrolytically and continuing said reduction to such an extent that the solution becomes saturated with formate and bicarbonate is precipitated out.

11. A process of making formates comprising the reduction of a solution of carbonic acid by means of the action of an alkali amalgam generated electrolytically and introduced into the solution containing the same, concentrating the resulting solution and thereby precipitating out the bicarbonate.

12. In the production of formates the process which comprises electrolyzing an aqueous solution of an alkali metal compound with a mercury cathode and contacting portions of metal-charged mercury from such cathode with another solution containing free carbonic acid.

13. In the production of formates the process which comprise electrolyzing an aqueous solution containing a potassium compound with a mercury cathode and contacting portions of metal-charged mercury from such cathode with another solution containing free carbonic acid.

In testimony whereof I have signed this specification in the presence of two subscribing witnesses.

MOOSHEGH VAYGOUNY.

Witnesses:
K. G. Le Ard,
Lester H. Jacobs.