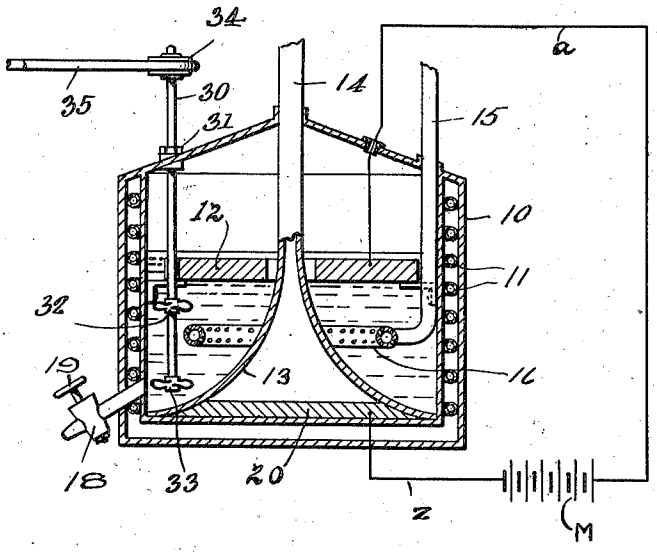
G. O. CURME, JR.

METHOD OF PREPARATION OF ACETIC ACID.

APPLICATION FILED JUNE 5, 1916. RENEWED MAR. 28, 1918.

1,315,546. Patented Sept. 9, 1919.



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UNITED STATES PATENT OFFICE.

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METHOD OF PREPARATION OF ACETIC ACID.

1,315,546. Specification of Letters Patent. Patented Sept. 9, 1919.

Application filed June 5, 1916, Serial No. 101,837. Renewed March 28, 1918. Serial No. 225,366.

To all whom it may concern:

Be it known that I, GEORGE O. CURMÉ, Jr., a citizen of the United States, residing at Pittsburgh, Allegheny county, and State of Pennsylvania, have invented and discovered certain new and useful Improvements in Methods of Preparation of Acetic Acid, of which the following is a specification.

Ethylene and a salt of mercury react in aqueous solution to form a compound of the type $HgSO_4.C_2H_4$, such as is formed by the interaction of mercuric sulfate and ethylene gas in aqueous solution; this compound being readily soluble in water, offers the possibility of obtaining a considerable concentration of ethylene in the aqueous solution. This compound above specified is capable of oxidation and, by suitable means, can be transformed into acetic acid, regenerating the original mercury salt.

My present invention, therefore, consists in the method and means for preparing acetic acid by the anodic oxidation of ethylene in an electrolytic cell in the presence of a salt of mercury.

The accompanying drawing illustrates an apparatus which is found suitable for the purpose. Said apparatus consists of a cell 10 formed with double walls between which is mounted a heating coil 11. Within the cell is mounted a ring-shaped anode 12 connected by wire *a* with battery *m*, and a cone-shaped diaphragm 13 dividing the cell into an anode chamber above said diaphragm and a cathode chamber below said diaphragm. Said diaphragm terminates at the top in a discharge tube or central outlet 14 for the hydrogen which is formed simultaneously in the process of electrolysis. An inlet pipe 15 connects with a perforated ring 16 mounted beneath the anode and above the cathode in the cell, while a discharge pipe 18, with a controlling valve 19, leads from the bottom thereof. The cathode 20 is connected by wire *z* with battery *m*. A device for stirring the contents of the cell is preferably provided, and I have illustrated such a device consisting of a vertical shaft 30, extending through a stuffing box 31 in the top of the cell, and having stirring blades 32 and 33 on the lower end thereof, with a pulley 34 on its upper end, which may be connected by a belt 35 with any convenient driving power.

It will be understood, of course, that the apparatus thus illustrated is merely a suggested arrangement, found suitable for the purpose, but may be modified materially as may be desired for varying commercial uses. The cone diaphragm 13 is of porous material pervious to liquids but not to gases when wet, and separates the cathode chamber from the anode chamber; the chambers are filled with solutions described below.

The anode may be of platinum, lead peroxid, magnetite, or other suitable substance such as will not be attacked either by the acid solution or by the oxidizing influence of the anode chamber; and the chamber containing the same is charged with aqueous sulfuric acid from 0–50% concentration and the desired amount of mercuric sulfate, or compound of mercury forming the sulfate when in contact with the acid of the strength given. This amount may vary from the smallest to a considerable quantity greater than what will dissolve in the acid of the strength given. The cathode in the cathode chamber, separated from the anode chamber by the porous funnel-shaped diaphragm, may be of mercury, or other suitable material to withstand the acid solution, and is also submerged in a bath of sulfuric acid of the same strength as used in the other chamber.

After the cell is thus prepared, ethylene is introduced into the anode chamber, through the pipe 15 and perforations in ring 16 connected to the lower end thereof, and bubbles through the mercury salt-sulfuric acid solution, or, in other manner, is brought into intimate contact with it, and the electric current is allowed to pass. The ethylene reacts with the mercury salt and the resulting compound dissolves and is subjected to the oxidizing influence of the anode, where it is oxidized to acetic acid, regenerating the mercury salt in its original condition at the same time.

In this process the acetic acid will collect in the anode chamber 12, and may be removed from time to time along with the sulfuric acid and mercury salt solution with which it is admixed, and the mixture purified to give pure acetic acid.

In this process the acetaldehyde, which may be considered the first product in the oxidation, cannot escape and thus is held in the solution and further oxidized to acetic

acid. The current employed is of suitable density and voltage.

The current density on the anode may vary between wide limits. Current densities up to 5.0 amperes per square decimeter will give good results and even higher densities may profitably be used. The voltage required is that necessary to produce the desired current in any given cell where the resistance is approximately constant. It need not exceed 4.0 volts in most cases.

Stirring favors the reaction.

The presence of a cerium salt in the anode chamber acts catalytically in favoring the oxidizing reaction.

Any temperature up to the boiling point of the solution used may be conveniently employed.

Having thus fully described my said invention, what I claim as new and desire to secure by Letters Patent is:

1. The process of forming acetic acid which consists in combining ethylene with a mercury salt solution, and subjecting the compound thus formed to prolonged anodic oxidation in aqueous solution.

2. The process of forming acetic acid which consists in combining ethylene with a mercury salt solution in an electrolytic cell, and subjecting the compound thus formed to anodic oxidation continued until the compound is broken up into acetic acid and the original mercury salt.

3. The process of forming acetic acid which consists in combining ethylene with a mercury salt solution charged with aqueous sulfuric acid from 0–50% concentration, and subjecting the compound thus formed to prolonged anodic oxidation.

4. The process of forming acetic acid which consists in combining ethylene with a mercury salt solution at a temperature maintained at a point below the boiling point of the solution, and subjecting the compound thus formed to prolonged anodic oxidation.

5. The process of forming acetic acid which consists in combining ethylene with a mercury salt solution in an electrolytic cell, and subjecting the compound thus formed to prolonged anodic oxidation, the anode being composed of platinum, lead peroxid, magnetite, or similar substance.

6. The process of forming acetic acid which consists in combining ethylene with a mercury salt solution, and subjecting the compound thus formed to prolonged anodic oxidation in an electrolytic cell having an anode chamber and a cathode chamber separated by a porous diaphragm, the cathode consisting of mercury or other suitable material.

7. The process of forming acetic acid which consists in combining ethylene with a mercury salt solution, stirring the solution of said compound, and subjecting said solution to prolonged anodic oxidation.

8. The process of forming acetic acid which consists in combining ethylene with a mercury salt in the presence of a cerium salt in an anodic chamber of an electrolytic cell, and subjecting the compound thus formed to prolonged anodic oxidation.

9. The process of forming acetic acid which consists in combining ethylene with a mercury salt solution in an electrolytic cell, and passing an electric current through the cell of the proper density and voltage continued to subject the compound to prolonged anodic oxidation.

10. The process of forming acetic acid which consists in combining ethylene with a mercury salt solution, raising the temperature thereof, and subjecting the compound thus formed to prolonged anodic oxidation in aqueous solution.

11. The process of forming acetic acid which consists in combining ethylene with a mercury salt solution and subjecting the compound thus formed to prolonged anodic oxidation in aqueous solution, removing and purifying the acid.

In witness whereof, I have hereunto set my hand and seal at Washington, District of Columbia, this twenty-ninth day of May, A. D. nineteen hundred and sixteen.

GEORGE O. CURME, JR. [L. S.]

Witnesses:
E. W. BRADFORD,
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