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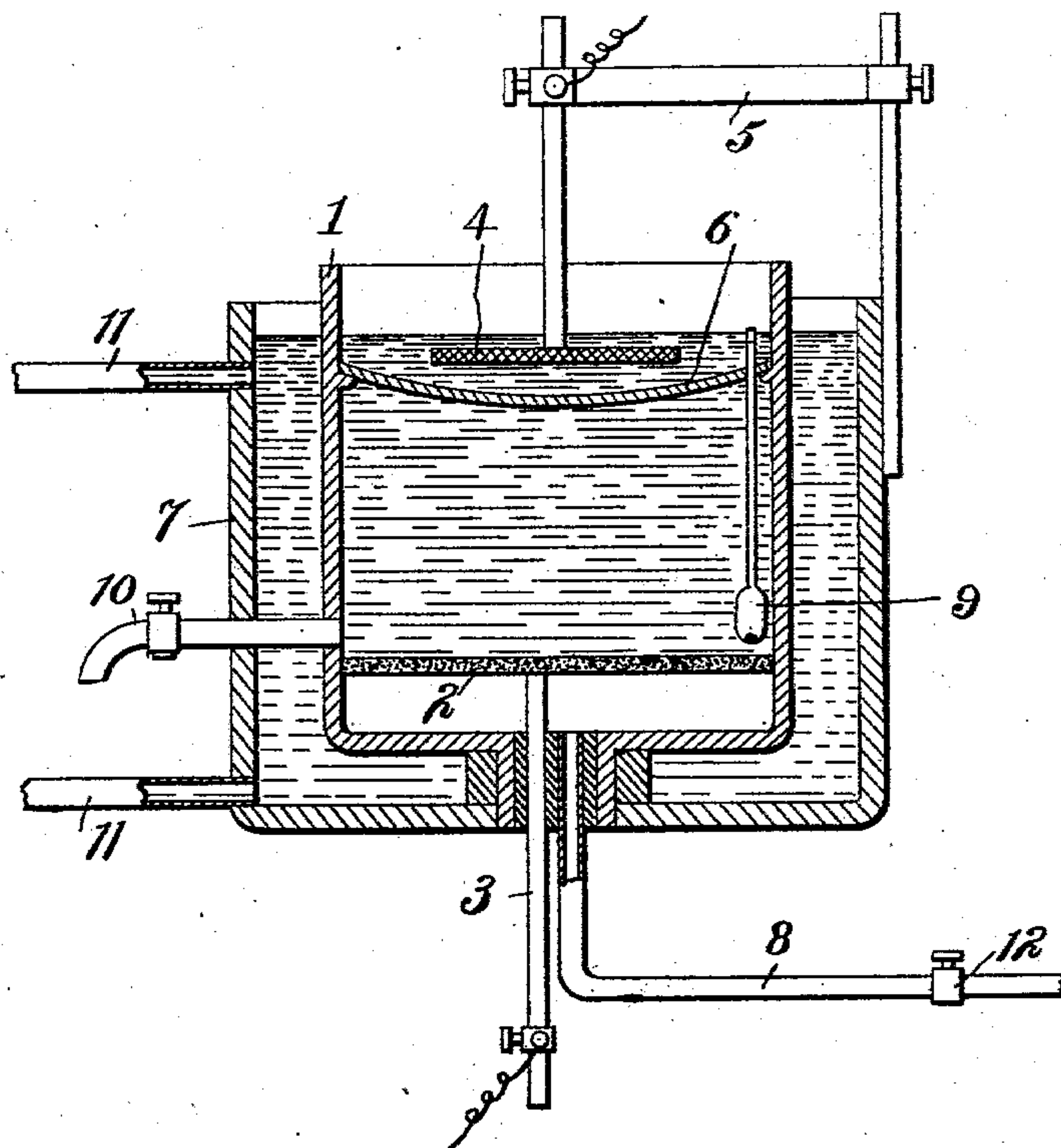
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C. B. JACOBS.

PROCESS OF MANUFACTURING SULFURIC ACID FROM SULFUR DIOXID IN  
AQUEOUS SOLUTION BY ELECTROLYSIS.

(Application filed June 1, 1901.)

(No Model.)



Witnesses.

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# UNITED STATES PATENT OFFICE.

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PROCESS OF MANUFACTURING SULFURIC ACID FROM SULFUR DIOXID IN AQUEOUS SOLUTION BY  
ELECTROLYSIS.

SPECIFICATION forming part of Letters Patent No. 704,831, dated July 15, 1902.

Application filed June 1, 1901. Serial No. 62,711. (No specimens.)

*To all whom it may concern:*

Be it known that I, CHARLES B. JACOBS, a citizen of the United States, residing at East Orange, county of Essex, State of New Jersey, have invented a certain new and useful Process of Manufacturing Sulfuric Acid from Sulfur Dioxid in Aqueous Solution by Electrolysis, of which the following is a specification.

10 The object of this invention is to provide a cheap and effective process for the manufacture of pure sulfuric acid without the use of acid-chambers, Glover's tower, or nitrous fumes.

15 The invention is based upon a discovery made by me that when sulfur dioxid in aqueous solution is subjected to electrolysis oxidation of sulfur dioxid to sulfur trioxid takes place at the anode-plate and that the sulfuric anhydrid thus formed combines with the  
20 water of the electrolyte to form sulfuric acid and that the concentration of the sulfuric acid may be carried to 50° or 67° Baumé by a continuation of the electrolysis under the  
25 conditions about to be described.

In carrying out the process any simple electrolytic cell may be used whose electrodes are not acted upon by sulfurous or sulfuric acid; but in order to make the process commercially efficient and to avoid liberations of free sulfur and loss of sulfur dioxid to any great extent by the reduction of sulfur dioxid by the hydrogen liberated at the cathode-plate and to control the operation, so that any degree of concentration of the sulfuric acid may be obtained in the cell, and also to promote an efficient and rapid oxidation of the sulfur dioxid by the oxygen liberated at the anode-plate and to effect the practical maximum absorption of any sulfur dioxid which may at  
40 first escape oxidation at the anode by the water of the electrolyte surrounding the anode I operate in a cell the construction of which is shown in cross-section in the accompanying drawing, in which—

45 1 is a square glass jar, about two inches from the bottom of which is cemented a porous

plate 2, which serves as the anode. The plate is made of porous carbon, preferably a section or sections of coke cemented together 50 the dynamo connection of which is a carbon rod 3, countersunk and cemented with the lower side of the plate and passing through a stopper in the neck of the glass jar, as shown in the drawing. The cathode-plate 55 4 is a rectangular sheet of platinized gauze wire relatively much smaller in section than the anode-plate. Thus I prefer to use anode and cathode surface whose relative areas are about thirty-three to one, and I prefer to use 60 a current of such strength that the current density at the anode is about eight one-hundredths of an ampere. The cathode is held in the cell just below the surface of the electrolyte by the wooden arm 5. Just below the 65 cathode-plate a porous diaphragm 6, of asbestos cloth, is placed to prevent free sulfur liberated at the cathode from falling down and contaminating the sulfuric acid formed at the anode. This porous diaphragm has also 70 another use in that it prevents the hydrogen liberated at the cathode from diffusing downward through the bath and causes it to escape from the surface of the liquid above the cathode, thus preventing reduction and loss 75 of  $\text{SO}_2$  in the bath below the porous diaphragm. Instead of the porous diaphragm I may use an ordinary porous battery-cup of unglazed earthenware to serve the same purpose. 80

The cell 1 is surrounded by a wooden jacket 7, through which a solution of brine cooled to zero degree centigrade is caused to flow by pipe connections 11 11 to a source of such cold brine, the object of this being to 85 keep the contents of the cell cooled to about zero degrees centigrade, at which temperature water will dissolve its maximum weight of  $\text{SO}_2$  and retain its saturation. The cell 1 is provided with an inlet-pipe 8 for the delivery of sulfur dioxid beneath the porous anode under sufficient pressure to force it 90 through the pores of the anode when it comes in contact with the oxygen set free on the



anode and is oxidized to sulfur trioxid, which combining with the water of the electrolyte results in the formation of sulfuric acid. More or less of the sulfur dioxid escapes oxidation until it has been dissolved by the water and is thus oxidized to sulfuric acid.

By regulating the pressure and flow of the sulfur dioxid delivered through pipe 8 and the current pressure and density passing between the anode and cathode the process may be conducted in such a manner that practically all the sulfuric dioxid passed into the cell will be oxidized to sulfur trioxid and there will be no loss of sulfur dioxid by escape from the surface of the liquid in the bath. The electrolysis is carried on without interruption until the small hydrometer 9, which is loaded to rise from the bottom when the acid solution at the anode reaches a strength of 50° or 67° Baumé, as may be desired by the operator, rises. Then the lower layer of acid is drawn off through the glass stop-cock 10 and fresh water added to the bath until the cathode is just below the surface of the liquid. The operation is carried on until the hydrometer again rises, when another portion of the acid liquor is drawn off, thus making the operation continuous.

The cell operates most efficiently with a pressure of two volts and a current density of fifteen to twenty amperes per square foot of anode-surface.

Working under theoretical efficiency with a pressure of two volts and five hundred amperes distributed over the proper anode-surface the cell will produce one ton of 50° Baumé sulfuric acid in three hundred and three hours. As high as fifty-four per cent. of the theoretical efficiency has been obtained with the cell. The sulfur dioxid used in the process may be derived from the waste gases of various processes or by the direct roasting of pyrites or the burning of sulfur.

From whatever source the sulfur dioxid may be obtained by absorbing it in water saturated with the gas at zero degrees centigrade the crystalline hydrate of sulfurous acid  $\text{H}_2\text{SO}_3 \cdot 14\text{H}_2\text{O}$  will separate out, leaving all impurities of the gas behind. The crystalline hydrate will give up its sulfur dioxid by slightly heating in a separate generator. Thus absolutely pure sulfur dioxid, and consequently pure sulfuric acid of any desired strength, may be obtained from the cell.

The advantages of the process over the ordinary methods of manufacturing sulfuric acid are: compactness of plant necessary, avoidance of the use of the leaden acid-chambers, the Glover tower, and the use of nitrous fumes for oxidation of sulfur dioxid, purity of product, rapidity of working, and consequent increased yield for a given time, and reduction of cost for pure acid produced.

As above stated, when the process is prop-

erly carried out there is no escape of sulfur dioxid from the surface of the bath, nor is there any liberation of free sulfur at the cathode; but all of the sulfur dioxid is oxidized. This is due partly to the fact that the sulfur dioxid is passed into or toward the bath through the anode, so that it will be mostly or largely oxidized before it can actually enter the bath. The comparatively large surface of the cathode also contributes to this result in exposing a larger oxidizing surface. On the other hand, the smaller size of the cathode and the consequent higher current density thereat insures a rapid escape of the hydrogen therefrom, whereby reducing action and deposition of sulfur are obviated. Both of these features of the process—namely, the passage of the sulfur oxid through the anode and the concentration of current at the cathode—thus contribute to the same end—namely, the complete oxidation of the sulfur dioxid—and they are mutually coöperative, as the supplying of the sulfur dioxid at the anode—*i. e.*, at a point remote from the cathode—enables the action of the cathode to operate with greater certainty and obviates the risk of reduction and the deposition of sulfur.

Other advantages of my process will be apparent in the passing of the current in an upward direction, whereby I secure a current density uniform over the surface of the cathode, and also by the cathode being near the top of the bath the hydrogen is rapidly and readily eliminated before it can exercise any deleterious reducing action upon the bath.

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

1. The process of making sulfuric acid, which consists in passing sulfur dioxid through a porous anode and setting free the oxygen of water by electrolysis upon said anode.

2. The process of making sulfuric acid by electrolysis, which consists in setting free the oxygen of water upon a porous anode and forcing sulfur dioxid through said anode under pressure.

3. The process of making sulfuric acid which consists in supplying sulfur dioxid through the anode of an aqueous electrolytic bath, setting free the oxygen of the water at the anode by electrolysis of the bath, maintaining at the cathode of such bath a current density sufficient to prevent access of sulfur dioxid to the cathode, and causing the hydrogen to escape from the cathode without acting on the sulfur dioxid.

4. The process of producing sulfuric acid consisting in passing sulfur dioxid into an aqueous electrolytic bath, maintaining at the cathode of said bath in proximity to the an-

ode a current density greater than the current density at the anode whereby rapid escape of hydrogen at the cathode is effected.

5 The process of producing sulfuric acid, which consists in supplying sulfur dioxide at the anode of an aqueous electrolytic bath, passing the electric current through said bath in an upward direction to oxidize the

sulfur dioxide in the lower region of the bath and retain the product of oxidation remote from the cathode.

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Witnesses:

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