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METHOD AND APPARATUS FOR PRODUCING QUINONE AND QUINOL.

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1,322,580. Patented Nov. 25, 1919.

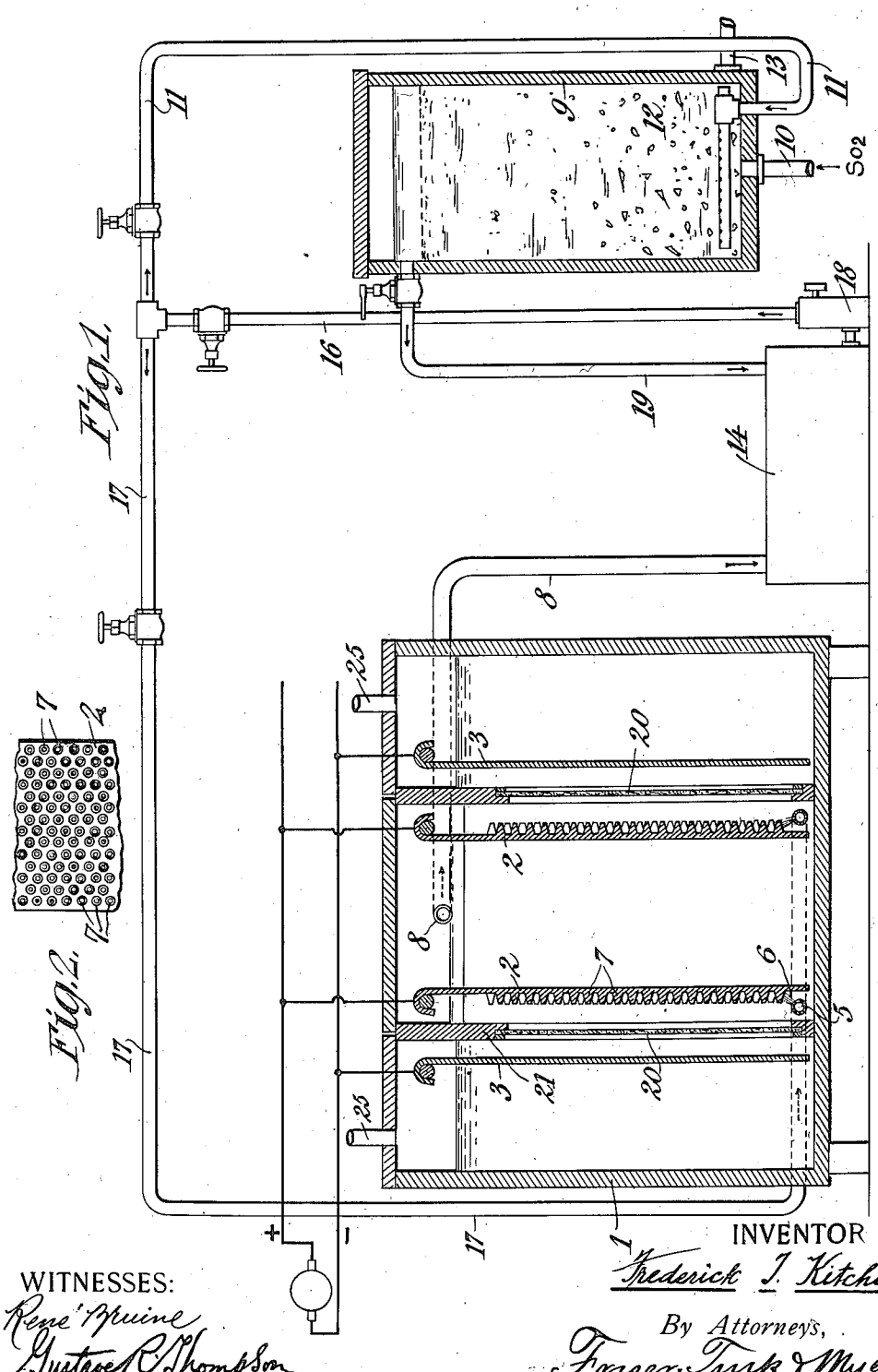



Fig. 1.




Fig. 2.

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# UNITED STATES PATENT OFFICE.

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## METHOD AND APPARATUS FOR PRODUCING QUINONE AND QUINOL.

1,322,580. Specification of Letters Patent. Patented Nov. 25, 1919.

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*To all whom it may concern:*

Be it known that I, FREDERICK T. KITCHEN, a citizen of the United States of America, residing in West New Brighton, borough of Richmond and State of New York, have invented certain new and useful Improvements in Methods and Apparatus for Producing Quinone and Quinol, of which the following is a specification.

This invention relates to a method and apparatus for producing quinone, also quinol (hydroquinone), and aims to provide improvements therein.

According to the present invention, quinone is produced by oxidizing benzene. The present invention provides an improved, simplified and economical process for producing the quinone, and an improved apparatus wherein the method may be carried out. The process is one which may be continuous in its operation.

Particular features of improvement will be pointed out in connection with the particular description of the invention.

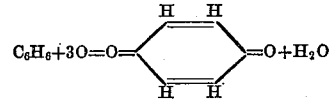
An embodiment of the apparatus in which the method is carried out is illustrated in the accompanying drawing.

Referring to said drawing, numeral 1 designates a suitable electrolytic tank having one or more anodes 2 therein, and one or more cathodes 3 therein. The electrodes and the electrolyte are such that when an electric current is passed through the bath, oxygen will be liberated at the anode. The electrolyte is preferably a dilute-aqueous sulfuric acid solution, for example, a 10 per cent. solution, and the electrodes are preferably of lead.

At the lower portion of the tank, in proximity to the lower end of the anode, is a pipe 5, adapted to conduct benzene to the bath. The pipe 5 has openings 6 therein, the openings being preferably quite small, through which openings small streams or drops of the benzene are adapted to percolate or enter the bath. The openings are formed so as to direct the drops or small streams of benzene against the anode. The benzene being immiscible in, and of less specific gravity than, the electrolyte, rises or flows upwardly to the surface of the electrolyte.

When an electric current is passed through the electrolyte and oxygen liberated at the anode, the particles of benzene rising upwardly in the bath against the plate, are acted upon by the oxygen, and the benzene oxidized to quinone.

The action may be expressed by the following equation:




In order that the oxidation of the benzene may be efficiently performed, the effective surface of the anode is increased to a large extent. One mode of increasing the surface of the anode which has been found to give good results, in practice, has been to form the anode with a large number of projections, as indicated at 7. The projections are preferably staggered, so that the particles of benzene take a devious course in rising. The nature of the surface should be something different from that of a porous body, the irregularities being of greater size, and substantially all points of the irregular surface being in direct line position with the cathode.

The efficiency of the process is also promoted by using a relatively small current density. A current density of 0.5 ampere per square decimeter has been found to give good results. By observing a proper relation between the benzene susceptible to anodic oxidation and the amount of oxygen liberated at the anode, formation of gas in the electrolyte, which may rise to the surface is completely or largely diminished. It has been found that it is advantageous to carry out the oxidation at a temperature below 12° C. (50° F.).

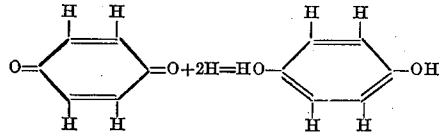
The quinone is also immiscible in the electrolyte, but is soluble in benzene, and the benzene arising from the pipe 5 is preferably in excess, so that the excess of benzene which rises from the bottom of the tank dissolves the quinone, which is formed in the bath, and carries it with it to the surface. The benzene solution of quinone may be withdrawn from the electrolytic tank, as by means of an outlet 8.

The quinone may be separated from the benzene solution, as quinone, in a suitable manner.

To produce quinol, the benzene solution of quinone is preferably carried to a suitable tank or vessel 9, in which the quinone is reduced to quinol, and the quinol preferably extracted from the benzene solution by a solvent, such a water, which is much cheaper than the benzene and in which the quinol is more soluble than in the benzene.

The quinone is preferably reduced by sulfurous acid, which may be formed by introducing sulfurous anhydrid ($SO_2$) through a suitable pipe 10, into water contained in tank 9. The reduction and extraction is preferably carried out by introducing the quinone solution into the bottom of the tank 9 and allowing it to rise or percolate through the reducing solution, preferably in the form of small streams or drops. The quinone solution may enter the tank 9 through a suitable pipe 11. To facilitate the intimate contact of the quinone solution with the reducing agent, a mass of broken material 12, such as glass, may be placed in the tank 9.

The reduction of the quinone may be expressed by the following equation:



The aqueous solution of quinol which is formed in tank 9 may be withdrawn and crystalized in a suitable manner, as for instance, in an evaporating apparatus. A suitable outlet 13 may be provided for withdrawing the aqueous solution.

The reduction of the quinone to quinol is preferably carried out without previous separation of the quinone from the benzene solution. The process is thereby simplified, and a better product obtained. Changes in the quinone, as for instance those produced by light, being diminished or avoided.

The benzene which rises to the surface in the vessel 9, after the conversion of the quinone and the extraction of the quinol, may be returned to the electrolytic tank 1, to undergo the previously described treatment therein.

The process is preferably carried out in a continuous manner, the benzene (or the benzene solution of quinone) being repeatedly subjected to the oxidation action in the tank 1, and portions of the quinone solution being continuously or intermittently led off for reducing the quinone and extracting the quinol, the benzene from which the quinol has been extracted being returned to the oxidizing cycle.

To this end a tank 14 may be provided from which the benzene (or the benzene solution of quinone) may be pumped to the pipe 5 in the tank 1 through suitable pipes 16 and 17 by a pump 18. The supernatant benzene solution of quinone in the tank 1 may be run off to the tank 14 through the overflow pipe 8.

The benzene containing liquid may undergo repeated treatment in the electrolytic tank by circulating it through the overflow 8, tank 15, pump 18, and pipes 16, 17 and 5.

When the circulating liquid has obtained a sufficiently high content of quinone, it may be run off through the pipe 11 into vessel 9, where the quinone may be reduced to quinol and the quinol extracted, as heretofore explained.

The benzene in the tank 9 remaining after the extraction process has been carried out, may be conveniently run off into a tank 14 through a pipe 19.

The anode is preferably separated from the cathode by means of a porous diaphragm or partition 20, which may be carried in a suitable frame 21 arranged within the tank.

This diaphragm does not materially impede the passage of the ions, but acts to confine the rising particles or stream of benzene to a restricted area in proximity to the anodes.

The hydrogen which may be formed in the cathode chamber may be conveniently led away through suitable outlets 25. The space above the anodes is preferably inclosed, to prevent evaporation of the benzene. The benzene is in fact inclosed at all points of its travel in the apparatus, so that loss by evaporation, and the formation of explosive mixtures are prevented.

The inventive ideas herein set forth may receive other mechanical or formal expressions than those which have been specifically set forth.

What I claim is:—

1. A method of producing the product or products herein set forth, comprising introducing benzene into a suitable electrolyte of greater specific gravity than benzene in such manner that it rises in the electrolyte, and passing an electric current through the electrolyte to produce oxygen to oxidize the benzene during its rise in the bath.

2. A method of producing the product or products herein set forth, comprising introducing benzene into a suitable electrolyte of greater specific gravity than benzene, in such manner that it rises in the electrolyte, and passing an electric current of low current density through the electrolyte to produce oxygen to oxidize the benzene during its rise in the bath.

3. A method of producing the product or products herein set forth, comprising introducing benzene into a suitable electrolyte of greater specific gravity than benzene in such manner that it rises in the electrolyte, and passing an electric current through the electrolyte to produce oxygen to oxidize the benzene during its rise in the bath, and con-

fining the rising benzene during its rise in the bath in proximity to an electrode.

4. A method of producing the product or products herein set forth, comprising introducing benzene into a suitable electrolyte of greater specific gravity than benzene, in such manner that it rises in the electrolyte, and passing an electric current through the electrolyte to produce oxygen to oxidize the benzene during its rise in the bath, the benzene being introduced in excess, so that the quinone formed may be dissolved by the benzene.

5. A method of producing the product or products herein set forth, comprising introducing benzene into a suitable electrolyte of greater specific gravity than benzene, in such manner that it rises in the electrolyte, and passing an electric current through the electrolyte to produce oxygen to oxidize the benzene during its rise in the bath, the benzene being introduced in excess, so that the quinone formed may be dissolved by the benzene, converting the quinone to quinol, extracting the quinol from the benzene solution, and recovering the quinol from the extract.

6. A method of producing the product or products herein set forth, comprising introducing benzene into a suitable electrolyte of greater specific gravity than benzene, in such manner that it rises in the electrolyte, and passing an electric current through the electrolyte to produce oxygen to oxidize the benzene during its rise in the bath, the benzene being introduced in excess, so that the quinone formed may be dissolved by the benzene, and repeatedly subjecting the benzene solution to the electrolytic oxidation until the solution becomes high in concentration of quinone.

7. A method of producing the product or products herein set forth, comprising introducing benzene into a suitable electrolyte of greater specific gravity than benzene, in such manner that it rises in the electrolyte, and passing an electric current through the electrolyte to produce oxygen to oxidize the benzene during its rise in the bath, and preventing evaporation of the benzene to the atmosphere.

8. A method of producing the product or products herein set forth, comprising electrolytically oxidizing benzene in a state of aggregation greater than a colloid.

9. A method of producing the product or products herein set forth, comprising electrolytically oxidizing benzene at a temperature not substantially above 12° C.

10. A method of producing the product or products herein set forth, comprising electrolytically oxidizing benzene with a current density not substantially greater than five-tenths ampere per square decimeter.

11. A method of producing the product or products herein set forth, comprising electrolytically oxidizing benzene, the benzene being in excess, repeatedly subjecting the benzene solution to the electrolytic oxidation, diverting portions of the solution from the oxidizing cycle, and further treating it.

12. A method of producing the product or products herein set forth, comprising electrolytically oxidizing benzene, the benzene being in excess, repeatedly subjecting the benzene solution to the electrolytic oxidation, diverting portions of the solution from the oxidizing cycle, reducing the quinone in the diverted portion to quinol, separating the quinol from the benzene, and returning the benzene to the oxidizing cycle.

13. A method of producing the product or products herein set forth, comprising electrolytically oxidizing benzene, the benzene being in excess, repeatedly subjecting the benzene solution to the electrolytic oxidation, diverting portions of the solution from the oxidizing cycle, reducing the quinone in the diverted portion to quinol, extracting the quinol from the benzene, and returning the benzene to the oxidizing cycle.

14. A method of producing the product or products herein set forth, comprising reducing quinone in a benzene solution to quinol, and extracting the quinol with water.

15. Extracting quinol formed in benzene solution with water.

16. An apparatus of the character described comprising an electrolytic tank, an electrode having a large surface area relative the dimensions of the electrode, and means for introducing a substance of less specific gravity than the electrolyte into the lower part of the tank, whereby it may rise in the electrolyte to undergo treatment at one of the electrodes.

17. An apparatus of the character described comprising an electrolytic tank, an electrode having projections thereon, and means for introducing a substance of less specific gravity than the electrolyte into the lower part of the tank, whereby it may rise in the electrolyte to undergo treatment at one of the electrodes.

18. An apparatus of the character described comprising an electrolytic tank, an electrode, and means for introducing a substance of less specific gravity than the electrolyte into the lower part of the tank, whereby it may rise in the electrolyte to undergo treatment at one of the electrodes, and means for confining the rising particles in proximity to an electrode.

19. An apparatus of the character described comprising an electrolytic tank, an electrode, and means for introducing a substance of less specific gravity than the electrolyte into the lower part of the tank, whereby it may rise in the electrolyte to undergo treatment at one of the electrodes,

and means for withdrawing and repeatedly introducing the substance undergoing treatment into the electrolyte.

20. An apparatus of the character described, comprising an electrolytic tank, an electrode, and means for introducing a substance of less specific gravity than the electrolyte into the lower part of the tank, whereby it may rise in the electrolyte to undergo treatment at one of the electrodes and an overflow at the level of the supernatant liquid.

21. An apparatus of the character described, comprising an electrolytic tank, an electrode, and means for introducing a substance of less specific gravity than the electrolyte into the lower part of the tank, whereby it may rise in the electrolyte to undergo treatment at one of the electrodes, said tank having a closure above the supernatant liquid to prevent evaporation.

22. An apparatus of the character described, comprising an electrolytic tank, an electrode and means for introducing a substance of less specific gravity than the electrolyte into the lower part of the tank, whereby it may rise in the electrolyte to undergo treatment at one of the electrodes, a tank in communication with said electrolytic tank, and means for introducing a reducing agent therein.

23. An apparatus of the character described comprising an electrolytic tank, an electrode and means for introducing a substance of less specific gravity than the electrolyte into the lower part of the tank, whereby it may rise in the electrolyte to undergo treatment at one of the electrodes, a reducing tank, means for introducing a reducing agent therein, and means for introducing the treated substance into the lower part of said reducing tank.

24. An apparatus of the character described, comprising an electrolytic tank, an electrode and means for introducing a substance of less specific gravity than the electrolyte into the lower part of the tank, whereby it may rise in the electrolyte to undergo treatment at one of the electrodes, a reducing tank, means for introducing a reducing agent therein, and means for introducing the treated substance into the lower part of said reducing tank, said reducing tank containing means for increasing the intimacy of contact between the introduced substance and the reducing agent.

25. An apparatus of the character described, comprising an electrolytic tank, an electrode and means for introducing a substance of less specific gravity than the electrolyte into the lower part of the tank, whereby it may rise in the electrolyte to undergo treatment at one of the electrodes, and an extracting tank in communication with said electrolytic tank.

26. An apparatus of the character described, comprising an electrolytic tank, an electrode and means for introducing a substance of less specific gravity than the electrolyte into the lower part of the tank, whereby it may rise in the electrolyte to undergo treatment at one of the electrodes, and an extracting tank in communication with said electrolytic tank, and means for introducing a reducing agent therein.

27. An apparatus of the character described comprising an electrolytic tank, an electrode and means for introducing a substance of less specific gravity than the electrolyte into the lower part of the tank, whereby it may rise in the electrolyte to undergo treatment at one of the electrodes, an extracting tank in communication with said electrolytic tank, and a conduit for leading off the appropriate liquid in the extracting tank and conveying it to the electrolytic tank to again undergo treatment.

In witness whereof, I have hereunto signed my name in the presence of two subscribing witnesses.

FREDERICK T. KITCHEN.

Witnesses:

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THOMAS F. WALLACE.