Jan. 9, 1923. 1,441,408

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METHOD OF AND APPARATUS FOR ELECTROLYZING LIQUID.

FILED AUG 12, 1915

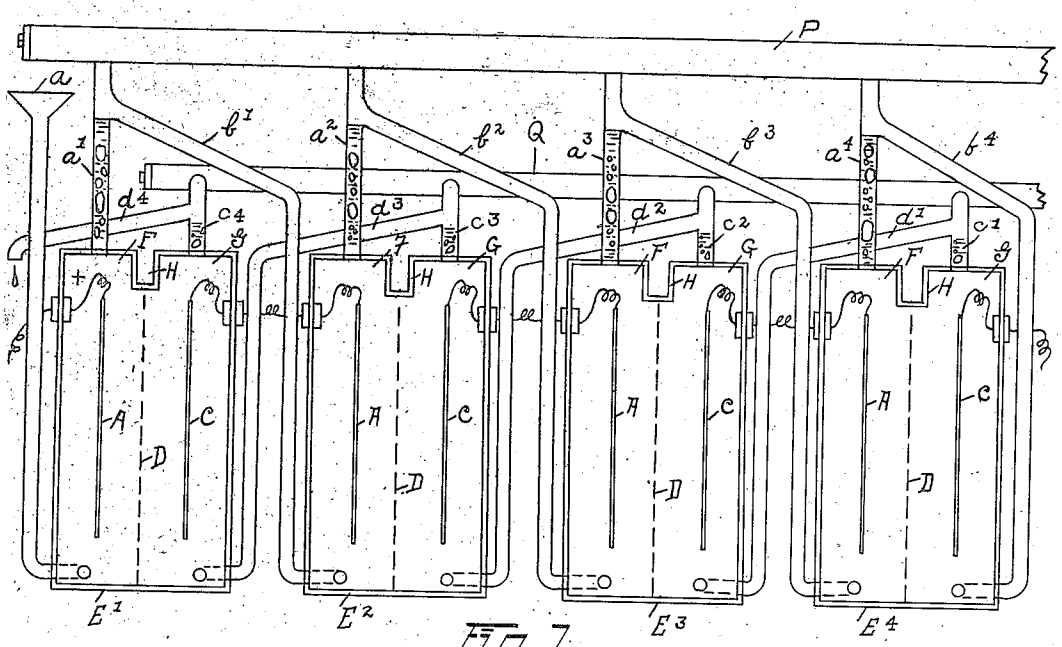


FIG. 1

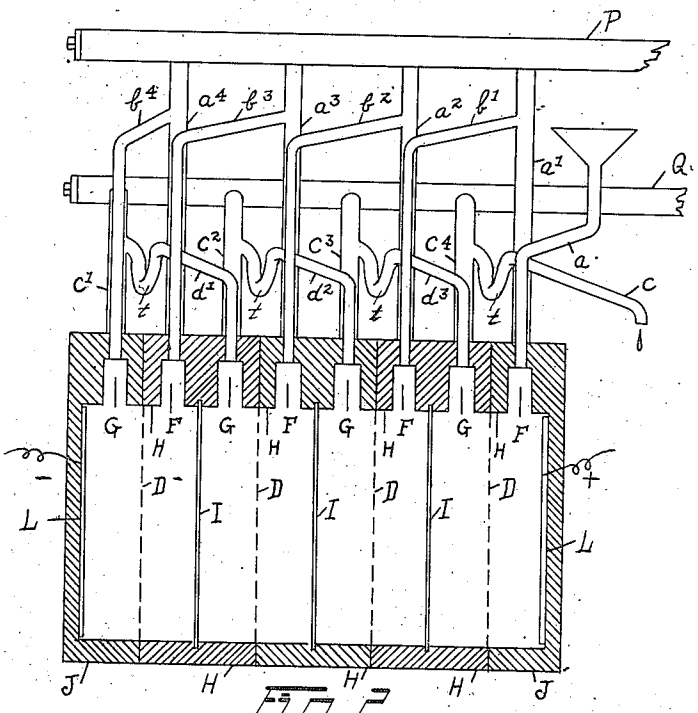


FIG. 2

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Patented Jan. 9, 1923. 1,441,408

UNITED STATES PATENT OFFICE.

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METHOD OF AND APPARATUS FOR ELECTROLYZING LIQUID.

Application filed August 12, 1915. Serial No. 45,096.

To all whom it may concern:

Be it known that we, HERBERT H. DOW, THOMAS GRISWOLD, JR., and EDWIN O. BARSTOW, all citizens of the United States, residing at Midland, in the county of Midland and State of Michigan, have invented a certain new and useful Improvement in Methods of and Apparatus for Electrolyzing Liquid, of which the following is a full, clear, and exact description, reference being had to the accompanying drawings.

This invention relates to the art of electrolyzing liquids and has particular connection with the electrolysis of alkali halide for the production of halide and caustic, although not limited to such use. The objects of the invention are the provision of a method and apparatus whereby power loss is minimized and whereby the purity of the effluent is increased; the provision of means for propelling electrolyte from cell to cell of a series of cells placed upon the same level while maintaining the same liquid level in adjacent cells; the diminution of short circuiting and current loss; while further objects and advantages will hereafter appear.

It has heretofore been proposed to conduct the electrolysis of alkali halide in a plurality of cells of the diaphragm type, transferring the catholyte successively from cell to cell so as to obtain an increment of causticity in each cell, while diminishing the average difference in liquid composition upon the opposite side of the diaphragm in the various cells, thus decreasing the tendency to diffusion and with it the power loss. It is well settled that diffusion of the cathode liquid not only causes a loss of power but also contaminates the product and seriously injures the anode plates of the cells. For the most efficient operation the liquid pressure in the anode chambers of the various cells is therefore maintained above that in the cathode chambers, thus tending to set up a percolation of liquid through the diaphragms which counteracts the diffusion tendency of the cathode product and also directly diminishes current loss. However, when the catholyte is caused to flow from cell to cell successively throughout the series, it has been noted that the frictional resistance to the flow causes the liquid level in each cell normally to stand a slight distance below that in the preceding cell so that in case a large number of cells is employed the difference in height of this liquid surface from first to last becomes rather an appreciable amount. Mechanical considerations, however, render a level arrangement of cells desirable, added to which is the fact that it is sometimes desired to maintain a flow of anolyte as well as of catholyte. In addition, in practical use, it is sometimes desirable to employ cells of the bipolar electrode type, in which the electrode plates themselves form the walls between adjacent cells, and this arrangement renders difficult or awkward any arrangement of cells other than on a level.

According to our invention we arrange the various cells substantially level, and make use of the gases evolved by the electrolysis for elevating the electrolyte (either or both anolyte and catholyte) sufficiently to enable it to flow into the next cell, thus overcoming the flow resistance otherwise observed. This arrangement offers the further advantage that the gas bubbles in the pipes increase the electrical resistance therein to such an extent as greatly to decrease the shunting of the current from cell to cell and thus cut off a material waste of power. An additional advantage of this arrangement resides in the fact that the velocity of the liquid flow is caused to depend with very substantial accuracy upon the rapidity of electrolysis, so that the amount of decomposition in each cell is automatically gauged; and, further, the flow of electrolyte ceases immediately upon the interruption of the electric current, so that the cells are always filled with electrolyte and are ready for operation without danger of becoming accidentally drained or overfilled.

In the drawings accompanying and forming a part of this application we have illustrated certain apparatus whereby the processes and objects of this invention may be realized, although it will be understood that these drawings are merely diagrammatic, and that they are not intended to be exhaus-

tive of all the arrangements, connections, or systems whereby the principle of our invention may be utilized. In these drawings: Fig. 1 represents a vertical, diagrammatic, sectional view through a series of cells connected and arranged in accordance with our invention; and Fig. 2 represents in idealized form an arrangement of our invention adapted for use with cells of the bipolar type.

First describing Fig. 1, E^1, E^2, E^3 and E^4 represent the various cells, four of the same being shown here, although it will be understood that the arrangement shown could be extended to any length desired. Each of these cells is separated into two chambers by means of a porous diaphragm D and with the anodes A and cathodes C located upon opposite sides thereof and connected electrically in any suitable manner. The upper portions of the cells are preferably chambered upon opposite sides of the diaphragm as shown at F and G, these chambers being separated by an impervious septum H. Leading upwardly from the chambers F are the discharge pipes a^1, a^2, a^3 and a^4, respectively, and communicating at their upper ends with a manifold P. Projecting laterally from these pipes are the conduits b^1, b^2, b^3 and b^4, respectively, all excepting the last of which discharges into the anode chamber of the succeeding cell. The conduit b^4 of the last cell is shown as communicating with the cathode chamber of the same cell.

Leading upwardly from the chambers G are the discharge pipes c^1, c^2, c^3 and c^4 connected at their upper ends to a manifold Q, and to the sides of these pipes are connected the conduits d^1, d^2, d^3 and d^4, respectively, all excepting the last of which communicate with the cathode chamber of the adjacent cell. The last of these conduits d^4 forms the ultimate effluent discharge.

It will be noted that the point of connection between the pipes a and b is higher than that of the pipes c and d in every case, this being for the purpose of maintaining a higher head of electrolyte in the anode chamber than in the cathode chamber.

With the arrangement shown in this view the operation is as follows: Supposing that sodium chloride is to be electrolyzed for the production of chlorin and caustic soda. A solution of sodium chlorid being introduced into all of the cells, the application of an electric current in such wise as to render the plates A anodes and plates C cathodes, will cause a depletion of the sodium chlorid content in the anode chambers with an evolution of chlorin gas, and a simultaneous production of caustic soda in the cathode chambers with the evolution of hydrogen gas. The cells are normally filled with solution as high as possible, in this case to the point defined by the intersection of the conduits a^1, a^2, a^3, a^4, and b^1, b^2, b^3, b^4, on the anode side and c^1, c^2, c^3, c^4 and d^1, d^2, d^3, d^4 on the cathode side. The chlorin gas collecting in the chambers F, F rises through pipes a^1, a^2, a^3, a^4 elevating the liquid therein and propelling it into the connecting conduits b^1, b^2, b^3, b^4, respectively, whence it is discharged into the anode chamber of the next cell. Likewise the hydrogen collecting in the chambers G rises in bubbles in the pipes c^1, c^2, c^3, c^4 elevating the catholyte therein so as to propel it into the tubes d^1, d^2, d^3, d^4, respectively, whence it flows to the cathode chamber of the adjacent cell. In the case of the last cell the liquid from the pipe a^4 flows to the cathode compartment of the same cell, but without danger of short circuiting the cell through the liquid because of the fact that the bubbles in the pipe a^4 are not conductors and also ordinarily because of the fact that there is a very noticeable difference in level between the catholyte and anolyte. The liquid ejected through the last conduit c^4 is that richest in caustic and is ultimately treated for recovery of this material in any suitable or desired manner. Raw brine to replace that withdrawn is added at the inlet a. The chlorin gas evolved is recovered from the manifold P and hydrogen gas from the manifold Q, while the purpose of the septa H—H is to keep the gases apart, rendering them better suited for use and preventing the danger of an explosion.

In Fig. 2 we have shown the adaptation of our improvement to electrolytic cells of the bipolar electrode type, in which the cell bodies H—H consists of open-ended boxes or casings traversed interiorly by plates I—I of conducting material tightly secured therein, said casings being secured together face to face in any convenient manner with diaphragms D—D interposed between their adjacent ends. The terminal cells shown at J—J are similar to the cells H—H excepting that each comprises only a single chamber and contains a single conducting plate L—L arranged to serve as an electrode. The upper parts of these cells are preferably formed as before with chambers F and G spaced from the forward faces of the bodies, which form in effect the septa H—H. In the present arrangement it is intended that the plate L at the right hand end shall be made the anode and the similar plate at the left hand end the final cathode, the opposite sides of the intermediate plates serving anode and cathode functions in the usual manner.

Communicating with the chambers F are the upright pipes a, a^1, a^2, a^3, a^4, b^1, b^2, b^3, while communicating with the chambers G are the upright pipes b^4, c^1, c^2, c^3, c^4, d^1, d^2 and d^3. The pipes a^1, a^2, etc., communicate with the manifold P, and the pipes c^1, c^2, etc., with the manifold Q. The operation

of these pipes is the same as that heretofore described and will probably be clear without further elucidation.

It will be understood that the above are intended to be only diagrammatic views, and that the foregoing description is intended to set forth only a few of the great numbers of embodiments to which our invention is susceptible and to point out only a part of the apparatus by means of which our improved process may be utilized. In both of the foregoing structures it will be observed that the pipes a^1, a^2, etc., have a diameter which bears such proportion to their length as that the evolved gases during the ascent therethrough convey with them portions of the electrolyte. First of all, the upper portion of the cell is divided by the septa H into separate chambers wherein the gases from the anode and cathode compartments, respectively, may collect, and then it is from these chambers that the pipes or ducts of either reduced diameter or cross sectional area rise.

For example, in the electrolysis of a halide alkali for halogen and caustic the advantages gained by propulsion of the catholyte from cell to cell far outweigh the advantages gained by similar propulsion of the anolyte. So far as the electrical and electrochemical features of the process are concerned, the anode compartments might be fed in parallel and only the cathode compartments operated in series; and, conversely, in the electrolytic production of other substances it might be more desirable to treat the anolyte in successive cells without moving the catholyte. In the present application we have illustrated the movement of both liquids, partly to show how it may be accomplished and partly from the fact that the automatic propulsion of the anolyte exhibits certain advantages of regulation, such as maintaining the anolyte in all the cells at a uniform level during the flow of current and instantly stopping the flow upon interruption of the current. However, in the case of a long series of cells it may be desirable to add fresh liquor at intervals to prevent impoverishment and consequent increase of resistance. Such variations are not modifications but rather different aspects of our same invention, and all such variations and modifications are intended to be included in the following claims.

Having thus described our invention, we claim:—

1. In the process of producing caustic alkali which consists in passing an aqueous solution of a halide salt of said alkali successively through the cathode chambers of a plurality of electrolytic cells of the diaphragm type, the step which consists in utilizing the buoyancy of the gases liberated in each cathode chamber to elevate the catholyte treated therein to a higher level than that of the catholyte in the succeeding cathode chamber.

2. In a method of electrolyzing an alkali halide solution for caustic alkali and free halide, in which the electrolyte is treated successively in a series of electrolytic cells of the diaphragm type, the step which consists in utilizing the buoyancy of the gases liberated in each cell to elevate the electrolyte treated therein to a level equal to that of its introduction into that cell.

3. In a method of electrolyzing an alkali-chloride solution for chlorine and caustic in which the electrolyte is treated successively in a series of electrolytic cells, whereby an increment of causticity is produced in each cell, the step which consists in utilizing the buoyancy of the liberated gases to expel the electrolyte from each cell at a rate substantially proportional to the rate of caustic production therein.

4. In a method of electrolysis in which the electrolyte is treated successively in a plurality of cells with the production of a solute and an evolved gas, the step which consists in utilizing the buoyancy of said gas to expel the electrolyte from that cell at a rate substantially proportional to the rate of production of solute, whereby the concentration of the solute is limited.

5. The process of producing caustic alkali which consists in electrolyzing a solution of a halide salt of said alkali in a series of closed cells of the diaphragm type, discharging the evolved gases through upwardly extending conduits communicating with the upper part of each cell, whereby a certain portion of the electrolyte is expelled continuously from each cell, conveying such expelled electrolyte to the next cell, and supplying new solution to take its place.

6. In a method of electrolysis in which an electrolyte is treated successively in a plurality of electrolytic cells, the liquid treated in each cell being passed to other cells for further treatment, the step which consists in discharging the evolved gases through the same conduits as the liquid, whereby an active movement of the liquid is produced and the passage of electric current along the liquid is retarded.

7. In a method of electrolysis in which the electrolyte is treated successively in a plurality of electrolytic cells with the production of a solute and an evolved gas, the steps which consist in discharging such gas through a restricted passage formed below the surface of the electrolyte, whereby the electrolyte will be expelled from the cell, and transferring such electrolyte to a succeeding cell.

8. The process of electrolyzing an alkali-halide solution for halide and caustic alkali which consists in adding to the anode chamber of a diaphragm cell a greater quantity of solution than said diaphragm can transmit by percolation, discharging the evolved gases through a restricted passage located below the surface of the analyte, whereby the remainder of the solution is ejected, conveying such remainder to the cathode chamber of the same or a similar cell, discharging the cathode gases through a restricted passage located below the surface of the catholyte, whereby the catholyte is ejected and conveying the ejected catholyte to the cathode chambers of succeeding cells until the desired causticity is attained.

9. Apparatus for electrolyzing liquids to produce a solute and an evolved gas comprising the combination, with an electrolytic cell, of means for collecting the gases evolved therein, a restricted passage communicating with the electrolyte therein, means for discharging such gases through said passage so as to induce a flow of electrolyte therethrough, and means for conveying said electrolyte to another cell.

10. Apparatus for electrolyzing liquids to produce a solute and an evolved gas, comprising in combination, a plurality of electrolytic cells arranged substantially upon a level, the top of each of said cells being closed to provide a collecting chamber, a restricted conduit communicating with each chamber, and a side branch communicating with said conduit above the top of each cell and leading to the next cell, said branch communicating with the succeeding cell at a point below the level of the conduit therein.

11. In apparatus for electrolyzing liquids to produce a solute and an evolved gas, the combination with an electrolytic cell of the diaphragm type, the top of said cell being closed to provide independent collecting compartments upon opposite sides of the diaphragm therein, of a restricted conduit leading upwardly from each of said compartments, a side branch communicating with each of said conduits at a point above said cell, the point of connection between the conduit leading from the anode chamber and its side branch being higher than the point of connection between the conduit leading from the cathode chamber and its side branch, and means for feeding electrolyte to each of said chambers.

12. Apparatus for electrolyzing liquids to produce a solute and an evolved gas, comprising, in combination, a plurality of electrolytic cells of the diaphragm type, the top of each cell being closed to provide independent gas collecting compartments upon opposite sides of the diaphragm therein, a restricted conduit leading upwardly from each of said compartments, a side branch communicating with each of said conduits at a point above said cell, the branch from each anode conduit communicating with the anode chamber of the succeeding cell and the branch from each cathode conduit communicating with the cathode chamber of the preceding cell, and means for feeding electrolyte continuously to the first anode chamber.

13. Apparatus for electrolyzing liquids to produce a solute and an evolved gas, comprising, in combination, a plurality of electrolytic cells of the diaphragm type, the top of each cell being closed to provide independent gas collecting compartments upon opposite sides of the diaphragm therein, a restricted conduit leading upwardly from each of said compartments, a side branch communicating with each of said conduits at a point above said cell, the branches from the anode conduits discharging at a higher level than the branches from the cathode conduits, each of the cathode branches communicating with the cathode chamber of the succeeding cell, and means for delivering electrolyte to each anode chamber.

14. Apparatus for electrolyzing liquids to produce a solute and an evolved gas, comprising, in combination, a plurality of electrolytic cells of the diaphragm type, the top of each cell being closed to provide independent gas collecting compartments upon opposite sides of the diaphragm therein, a restricted conduit leading upwardly from each of said compartments, a side branch communicating with each of said conduits at a point above said cell, the branches from the anode conduits being arranged at one level and the branches from the cathode conduits being also disposed upon a level but lower than said anode branches, each of said anode branches communicating with a succeeding anode chamber and each of said cathode branches communicating with a succeeding cathode chamber.

15. In apparatus for electrolyzing alkali halide solutions for the production of caustic alkali and free halide, the combination, with a plurality of electrolytic cells, said cells having diaphragms between the electrodes and the tops of said cells being covered to provide independent gas-collecting compartments on each side of the diaphragm, of a restricted conduit leading upwardly from each compartment, and a side branch communicating with each conduit at a point above the cell, the branches from the anode conduits being led to other anode chambers, and the branches from the cathode conduits being led to other cathode chambers.

16. In an apparatus for electrolyzing liquid to produce a solute and an evolved gas, the combination with a plurality of electrolytic cells, each of which is divided by a porous diaphragm into a pair of com-

partments, said cells having the tops of similar compartments closed to form collecting chambers, of a restricted conduit leading from each chamber, and means for conveying any liquid propelled through said conduit into the similar compartment of a succeeding cell.

In testimony whereof, we hereunto affix our signatures in the presence of two witnesses.

HERBERT H. DOW.
THOMAS GRISWOLD, JR.
EDWIN O. BARSTOW.

Witnesses:
JAMES T. PARDEE,
A. W. SMITH.