W. E. GREENAWALT.

METALLURGICAL PROCESS.

APPLICATION FILED AUG. 11, 1917.

1,344,030.

Patented June 22, 1920.

2 SHEETS—SHEET 1.

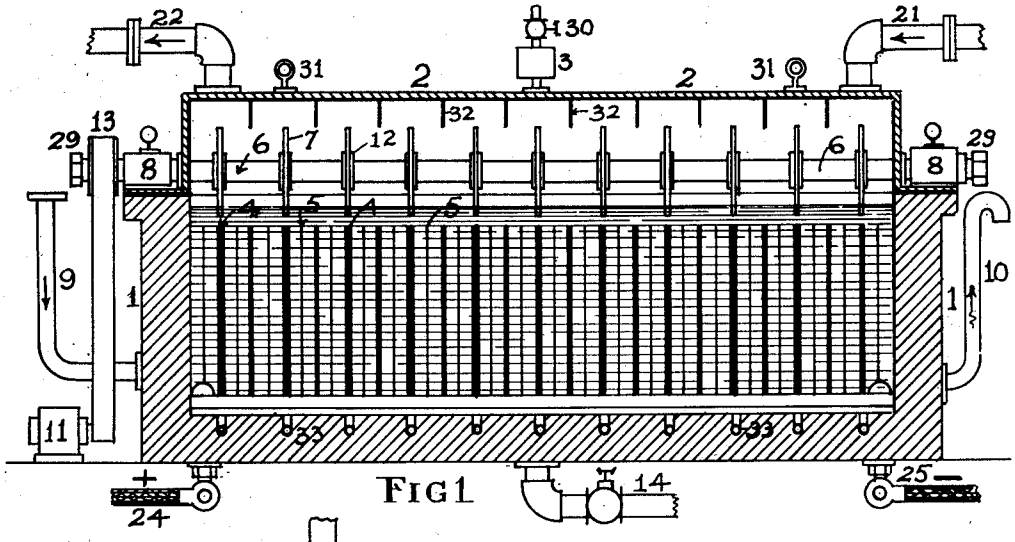


FIG 1

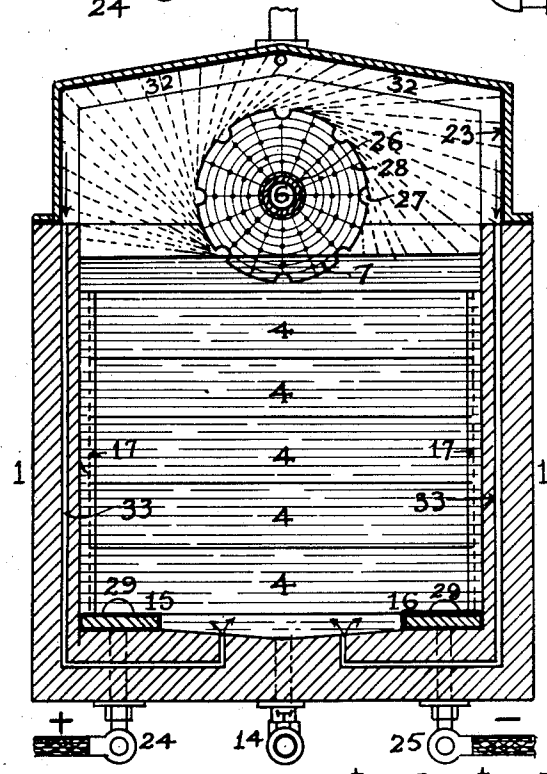


FIG 2

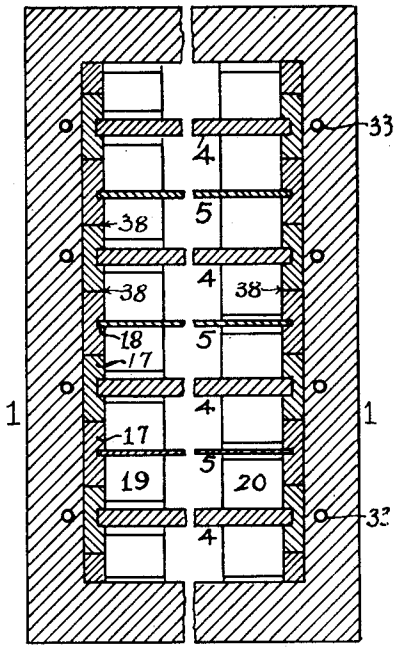


FIG 3

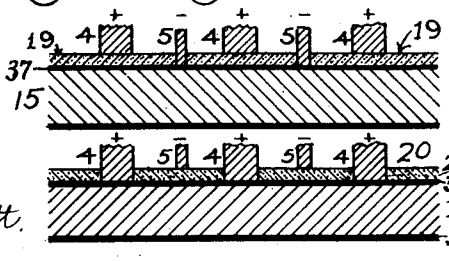


FIG 4

WITNESSES

Irma Greenawalt.

Fred L. Scott.

INVENTOR

William E. Greenawalt.

W. E. GREENAWALT.

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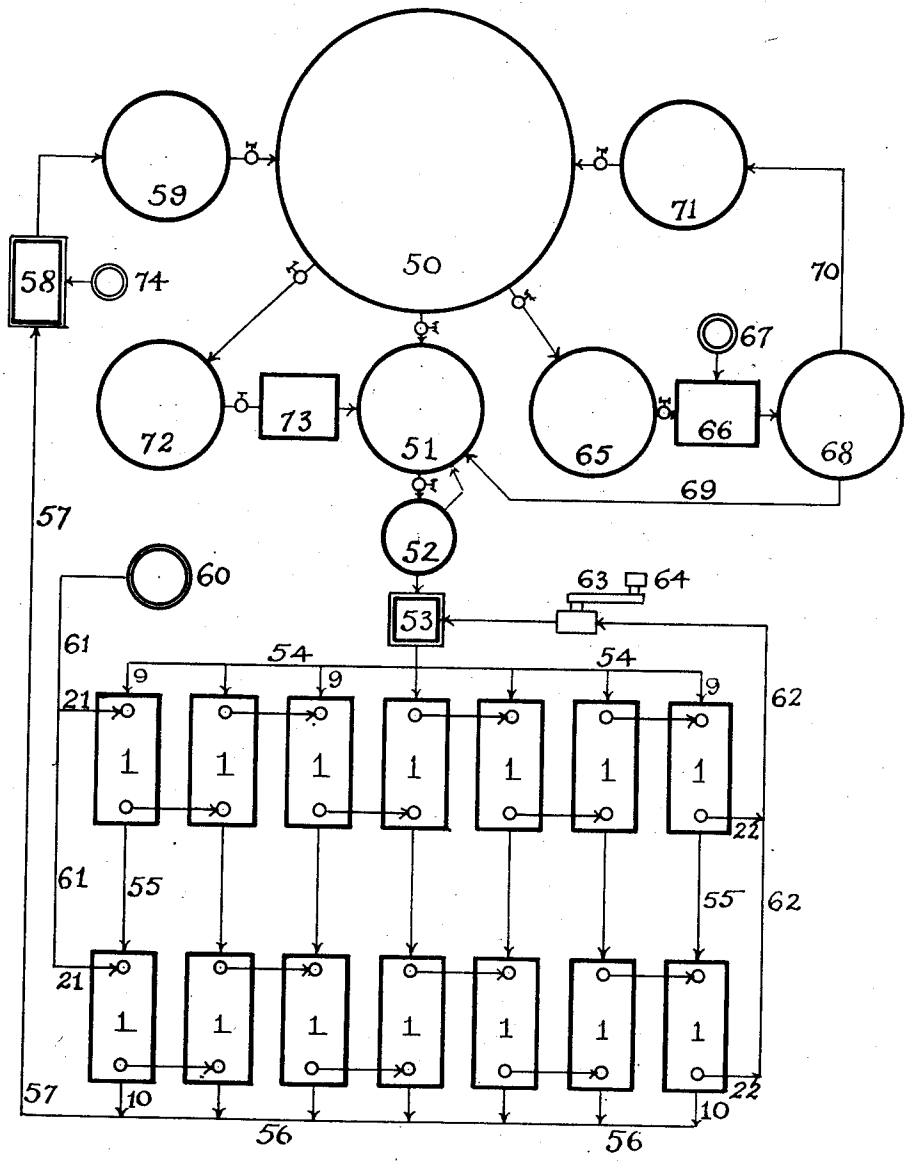


FIG 5

WITNESSES

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UNITED STATES PATENT OFFICE.

WILLIAM E. GREENAWALT, OF DENVER, COLORADO.

METALLURGICAL PROCESS.

1,344,030. Specification of Letters Patent. Patented June 22, 1920.

Application filed August 11, 1917. Serial No. 185,652.

To all whom it may concern:

Be it known that I, WILLIAM E. GREENAWALT, a citizen of the United States, residing in the city and county of Denver and State of Colorado, have invented certain new and useful Improvements in Metallurgical Processes, of which the following is a specification.

This process may be regarded as a modification and continuation of the processes described in my co-pending applications, Serial No. 15,583, filed March 15, 1915, and Serial No. 140,738, filed January 5, 1917, and the apparatus used in carrying out the process, a continuation and modification of that described in my co-pending apparatus application, Serial No. 145,884, filed February 1, 1917.

The process has for its object the effective application of reducing gases in the electrolysis of impure electrolytes, and the effective reduction of the variable valent salts in the electrolyte by a suitable reducing agent, such as sulfur dioxid.

The process will be described more particularly in reference to the extraction of metals from their ores with chlorid solutions. The ores may be assumed to be copper ores carrying variable quantities of the base and the precious metals. It is not intended, however, to limit the process to any particular use.

If copper ores containing precious metals are treated with a chlorid solution, such as a dilute solution of hydrochloric acid containing in solution various other chlorids, such as the chlorids of iron and sodium, the copper will be extracted as the cupric and cuprous chlorids, and the gold and silver as their respective chlorids. Some of the copper will be in the cuprous condition, but in practical operations, most of it will be in the cupric condition.

The solution of the metals from the ore usually involves no particular difficulty. The trouble commences when an attempt is made to electrolyze the chlorid solutions to obtain the copper. If such a solution is electrolyzed, copper is deposited at the cathode and chlorin liberated at the anode. This chlorin combines with the ferrous salts in the solution to convert it into the ferric salts, and the ferric salts and free chlorin, acting on the copper deposited at the cathode, re-dissolves it, thus causing a loss of efficiency: This loss under aggravated conditions, may be so great that the copper is re-dissolved as rapidly as it is deposited.

On the other hand, it is known that, theoretically at least, twice as much copper is deposited, per unit of current, from a cuprous chlorid solution as from a cupric chlorid or cupric sulfate solution. The advantages in this respect, however, have been largely outweighed by the disadvantages due to the free chlorin and ferric salts formed by the chlorin. One of the most important objects of this invention is to maintain the electrolyte so reduced as to effectively realize the advantages, without being bothered by the disadvantages.

If a chlorid solution, resulting from treating ores as described, is treated with sulfur dioxid, the cupric chlorid is reduced to the cuprous condition and the ferric salts reduced to the ferrous condition, thus:

1. $2CuCl_2+SO_2+2H_2O=$
$2CuCl+$
2. $2FeCl_3+SO_2+2H_2O=$
$2FeCl_2+2HCl+H_2SO_4.$

If this reduced solution is electrolyzed, and if it were possible to maintain the variable valent salts at their lowest valence, a high cathode efficiency could be obtained, and, as stated, twice as much copper deposited as from a cupric solution. The essence of the trouble, however, is in maintaining the solution reduced.

I accomplish this effectively with my process, by using an apparatus, as shown in the accompanying drawings, in which Figure 1 represents a longitudinal section of an electrolyzer in which the process may be carried out, Fig. 2 a transverse section, Fig. 3, a plan, Fig. 4, electrode details, and Fig. 5 a diagrammatic sketch of the preferred method of carrying out the entire process.

In the drawings, 1 is an electrolytic tank, having a removable cover 2, and containing the electrolyte and vertical electrodes 4 and 5, resting on submerged conducting bars 15 and 16 at the bottom of the tank. The elec-

trolyte flows into the tank through the pipe 9 and flows out through the pipe 10. The gas with which the electrolyte is treated, flows in through pipe 21, and the excess, if any, flows out through pipe 22. Above the electrolyte, and within the hood, or cover, is a spraying, or atomizing, device, consisting of a shaft 6, rotated in the bearing 8, when driven by the motor 11, and having mounted on its disks 7 which dip into the electrolyte, and spray, or atomize, the liquid into the gas confined over it and submerge some of the gas in the liquid, when rotated at a proper speed by the motor 11. The walls of the tank are arranged with a series of pipes, or ducts, 33, so that the liquid sprayed into the gas will run down the sides of the cover and through the pipes, and thus cause a circulation of the electrolyte, due to the difference in level of the top of the pipes and the surface of the liquid.

The electric current is supplied to the electrodes by means of the terminals 24 and 25, and the conducting bars 15 and 16 in the bottom of the tank. Electrical connections is made with the electrodes by letting the electrodes rest on the conducting bars. The electrodes are held in position by the side pieces 17, which may be of wood, and having saw-cuts, or grooves, 18 to receive the ends of the electrodes. The arrangement for insulation of the electrodes is shown in detail in Fig. 4. One end of the electrode rests on its corresponding conducting bar, say 15, while the other end rests on an insulation plate 19, and this plate, in turn, rests on the opposite conducting bar. The electrode 4, for example, rests on the conducting bar 16 while the opposite end rests on the insulation plate 19, which in turn, rests on the bar 15. The electrode 5 rests on the conducting bar 15, but is insulated from bar 16 by the plate 20. A current, therefore, entering through the bar 16 would pass to the electrode 4, and, being insulated from bar 15, would pass through the liquid from electrode 4 to electrode 5, and then to the bar 15 on which electrode 5 rests and with which it is in electrical contact, while electrode 4 is insulated from bar 16. The conductor bars 15 and 16 are preferably made of copper, with a lead casing 37. The copper gives the desired conductivity, while the lead has a much greater resistance to deterioration than the copper. The spraying disks are preferably made of rubber, with serrations 27 and small perforations 28, to facilitate the spraying and mixing of the electrolyte and gas. The disks are held in position on the shaft by washers 12 and annular rings 26 inserted between the washers, and arranged so that the whole can be made tight and rigid by the screws 29 at the ends of the shaft. The cover 2, is provided with a lead lining 23 to resist the corrosive action of the liquid and gases.

In operating the process the leach solution from the ore, and, preferably, having its variable valent salts reduced to their lowest valency, is introduced into the electrolyzer 1, through the pipe 9 and is exhausted through the pipe 10. Sulfur dioxid gas is introduced through the pipe 21 and exhausted through the pipe 22 in such a way as to always maintain a slight inward suction to prevent the corrosive gases from escaping from the cell and contaminating the atmosphere.

When the current is turned on, copper is deposited on the cathode and chlorin liberated at the anode. Some of the chlorin is consumed in converting the variable valent elements from their lower to their higher valencies, and some escapes from the liquid into the hood, above the liquid, where coming in contact with the sulfur dioxid gas and sprayed liquid, the chlorin combines with the sulfur dioxid to form acid. These reactions may be shown by the following equations:

3. $2CuCl + \text{electric current} = 2Cu + 2Cl$.
4. $2CuCl + 2Cl = 2CuCl_2$.
5. $2FeCl_2 + 2Cl = 2FeCl_3$.
6. $2Cl + SO_2 + 2H_2O = 2HCl + H_2SO_4$.

If this action were to go on indefinitely, the process would soon become commercially inoperative. To obviate this, the sulfur dioxid is maintained in excess above the electrolyte, and as the atomizers spray the electrolyte into the sulfur dioxid gas and submerge some of the gas into the electrolyte, the cupric and ferric salts are maintained reduced as shown by equations 1 and 2, and the chlorin converted into acid as shown by equation 6. Thus the copper is deposited, the acid regenerated, and the cupric and ferric salts reduced to the cuprous and ferrous condition as rapidly as formed. In this way the process may go on efficiently indefinitely. It is very desirable to always maintain the sulfur dioxid in excess in the cell. Fortunately this can readily be done, as sulfid ores are almost always available, and the sulfur dioxid from roasting is otherwise usually a waste product. The excess of sulfur dioxid, issuing from the cells, may be used in reducing the ferric iron and cupric copper in the solution before electrolyzing it. With a hot or a warm electrolyte the space in the electrolyzer above the liquid will contain much of the liquid sprayed into it, as a fine mist and steam, and thus the reactions between the gases and the liquid are rapidly effected. The atomizers are rotated at a fairly high speed so as to

effectively spray the liquid into the gas, while at the same time submerging some of the gas in the liquid: In this way it is quite possible to always maintain a slight excess of sulfur dioxid in the electrolyte. It has been observed that when there is an excess of sulfur dioxid in the electrolyte a high current efficiency can always be expected. The reaction between the sulfur dioxid and chlorin is practically instantaneous.

In carrying out the process, as so far described, by making the initial reduction with sulfur dioxid, sodium sulfate is formed in considerable quantities from the sodium chlorid which is assumed to be in the solution, and, while this does not appear to exert any influence on the electrolysis, nevertheless, it would soon accumulate in amounts to give trouble by crystallization. Fortunately, the acid regenerated in the process, is in such excess in the treatment of suitable ores, that some of the solutions can be diverted at each cycle, and thus the impurities kept within bounds. To facilitate this, it is preferred to precipitate this diverted portion with hydrogen sulfid, and then use the resulting copper sulfid in the initial reduction. In this way the solution may be reduced, and no sodium sulfate formed by the reactions, thus:

7. $CuCl_2+Cu_2S=2CuCl+CuS$.
8. $CuCl_2+CuS=2CuCl+S$.
9. $2FeCl_3+Cu_2S=2CuCl+2FeCl_2+S$.
10. $FeCl_3+CuS=CuCl+FeCl_2+S$.

This not only serves to reduce the cupric and ferric salts, but also serves to convert the copper in the washwaters and lean foul solutions into the electrolytic metal in the regular process of deposition.

The important step in the process is confined to the treatment of the solutions in the electrolyzer. The process, as a whole, is best described by referring to Fig. 5, in which 50 represents a leaching vat for the treatment of the ores with an acid chlorid solution. The rich solutions flow into tank 51 and the lean solutions into tank 65. The lean solutions are drawn from tank 65 into the precipitator 66, where, being treated by hydrogen sulfid from the gas generator 67, the copper is precipitated as the sulfid with a simultaneous regeneration of an equivalent of acid. The mixture of liquid and sulfid precipitate flows into the separator, or thickener, 68, where the sulfid sludge is separated from the liquid. The weak regenerated acid solution flows into tank 71, and from there applied to the ore as desired.

The copper sulfid precipitate is introduced into the reducing tank 51, where it is agitated with the rich solutions, and the cupric and ferric salts reduced; it then flows into the separator 52, where the copper sulfid is separated from the reduced solution and returned to the agitating tank 51. The clarified solution flows into a tower 53, where the excess of sulfur dioxid from the electrolyzers is applied to it, and it then flows into the electrolyzers.

Since the solution in the electrolyzers is at all times reduced and agitated and circulated, there is no particular object in causing a rapid flow through the cell, or in flowing the solution through a series of cells before returning it to the ore. The solution may, therefore remain in the electrolyzer until the copper has been sufficiently extracted from it, or say, down to one or two per cent. from a three to four per cent. solution. It is therefore preferable to feed the first row of cells in multiple, and then pass the solution on to a second series of cells with perhaps a lower current density, on account of the impoverished electrolyte from the first series of cells. The gas, from the sulfur dioxid generator 60 is distributed to the cells by the pipe 61, and enters the cells by the connection pipe 21, and then flowing from one end of the cell to the other end, flows into the next cell, and so on through the series, finally issuing through the pipes 22, where it flows into the main 62, and by means of the exhauster 63 is forced into the tower 53, where the excess sulfur dioxid is applied to the new electrolyte. The solution from the first series of electrolyzers flows into the second series through the pipes 55, and on issuing from the second series of electrolyzers, is collected by the pipe 56, and through pipe 57 flows into the storage tank 59, where it may be applied to the ore as desired. If it is desired to raise the valency of the variable valent elements, after electrolyzing the solution and before applying it to the ore, the electrolyzed solution is passed through an oxidizer 58 where chlorin or oxygen can be applied to it from the gas generator 74. If the ore contains gold the solution should also contain free chlorin, before it is applied to the ore. If the ore contains precious metals, the solution from the leaching vat flows into the tank 72, and from there, in a regulated stream, through the precipitator 73, where the gold and silver can be precipitated with copper, before entering the storage tank 51. It is usually preferred, however, to omit this step and precipitate the gold and silver with the copper sulfid in the reducing tank 51, and then from time to time extract the gold and silver from the sulfid residue.

In electrolyzing solutions by the method purposed in this process, it is evident that the usual electrical connections will not suffice. In the swirl of the gases and electrolyte the usual electrical connections

would be too complicated and would be very quickly corroded, and on account of the difficulty of getting at the connections, the practicability of the apparatus might be questioned. These difficulties are overcome by having two conducting bars in the bottom of the tank; one for the positive and one for the negative conductor. These bars are bolted tight to the bottom so that there will be no leak, and the terminals on the outside arranged so that connections with the electric dynamo are easily made. The electrodes rest on the conducting bars, and their weight suffices to make the proper contact for the electric current. The edges of the anodes are insulated from the cathode bar, and the edges of the cathodes are insulated from the anode bar. The anodes may be either of lead, carbon, or magnetite, and the cathodes either lead or copper.

If the anodes are made of carbon, which is the material preferred for chlorid solutions, the construction of the anode is very much simpler than in an ordinary cell where the electrodes are suspended from above. The carbon electrodes are usually manufactured in slabs, but there has been some difficulty in assembling these slabs to make a large anode, to overcome corrosion and resistance. I overcome these difficulties very easily by laying the first slab on the conductor bars with one end properly insulated from the cathode conducting bar, and by means of wooden grooved pieces at the sides, lay one slab vertically on top of another with the adjacent edges contacting, as shown in Fig. 2. In this way an electrode may be built vertically as large as desired, while the electrical contact between the slabs is all that is necessary. If perfect abutting edges are desired to give a better electrical contact, the abutting edges may be planed to a true surface. This will not ordinarily be necessary, for, even if the contact in the beginning is not perfect, it will soon be made so by the electrolytic action of the current in depositing copper between the gaps.

In chlorid solution, especially, the conducting bars are more likely to corrode than in sulfate solutions, although in any case the corrosion is quite slow. It is almost entirely avoided by surrounding the conducting bars with a covering of lead.

In practically all electrolytic processes, hydrogen is, or may be, liberated. The hydrogen is undesirable in the upper part of the cell: It may act to free sulfur from the sulfur dioxid, or it may, under certain conditions, become explosive. These difficulties are overcome by having a small hydrogen outlet in the top of the cover of the cell. Hydrogen, being by far the lightest gas likely to be in the cell, will by its own buoyancy in a quiescent space 3, displace the other gases downwardly. The hydrogen may be continuously and automatically allowed to escape through the valve 30 in the amounts formed, or used as desired.

To avoid the difficulty, in deep cells, of a stagnant and impoverished electrolyte at the bottom of the tank, provision is made to cause a positive circulation and distribution of the electrolyte, by means of the pipes, or ducts, 33, the atomizers, or spraying disks, 7, and the cover 2. The amount of electrolyte sprayed into the gas over it is easily regulated by the speed and submergence of the disks, and the disks spraying the electrolyte into the gas also causes it to hit the sides of the cover, and this electrolyte, effectively treated with the gas, passes down the sides of the cover, and enters the pipes 33, and is again introduced into the body of the electrolyte at the bottom, and causes a circulation upward, and brings freshly reduced electrolyte in contact with the electrodes in the lower as well as in the upper portion of the cell. This same procedure is observed in flowing the electrolyte from one cell to another: the electrolyte is preferably taken from the top of the first cell and introduced into the bottom of the next.

It might be thought that the covered electrolyzers would complicate the electrolysis and increase the cost of operation. It is more likely to simplify the operation and cheapen the cost of deposition. The idea, of course, is, to use fairly large units, and while the cost of the individual cells is greatly increased, the cost of the electrolytic installation as a whole is materially decreased. Electrolytic dynamos with a capacity of 10,000 amperes are regularly made as a standard size, and 38x8x2 inches is a stock size carbon slab from which anodes 38x48x2 inches could easily be constructed, as described. Electrodes having a total active area of 25 square feet may therefore be considered a fair standard. With a current density of 10 amperes per square foot, each electrode would take 250 amperes, and with 40 cathodes and 41 anodes there would be a total current of 10,000 amperes.

Assuming a current efficiency, under the conditions, of about 96 per cent. or 2.5 lbs. of copper from cupric solutions, per 1000 ampere-hours, there would be deposited 25 lbs. of copper per hour with 10,000 amperes, or 600 lbs. per day of 24 hours, per cell. If the deposit on each side of the cathode is one half inch thick, the total weight of the copper on each cathode would be approximately 1155 lbs. and for 40 cathodes it would be 46,200 lbs. Depositing at the rate of 600 lbs. per day, 77 days would be required to make this deposit Or in other words, the cell would not have to be disturbed or the cover lifted for a period of 77 days, when it would be cut out of the circuit, and 46,200 lbs. of copper removed

therefrom. It is true that trees may form, but with a low current density and adequate circulation and agitation of the electrolyte, this is not likely to cause trouble.

There is no difficulty in removing the cover from the cells. The cells are easily arranged so that they can be disconnected one at a time, and a traveling crane used to lift the cover and place it on a fresh cell, while the one from which it was taken has its copper removed. In taking the copper out of the cells, the individual cathodes are lifted out with pincers suspended from the crane. If lead cathodes are used, the copper is easily pried off, and the cathode returned to the cell. The anode is not removed until worn out.

If the current density is doubled, say to 20 amperes per square foot, which is by no means excessive, twice the amount of copper would be deposited with the same cells, and in the same time, or, 1200 lbs. per day. If the solution is cuprous, and the deleterious reactions can be held in check by the efficient use of sulfur dioxid made possible in these cells, 1200 lbs. of copper will be deposited per day with a current density of 10 amperes per square foot, and 2400 lbs. with a current density of 20 amperes per square foot.

In the operation of these cells it is unnecessary to resort to expensive absorption towers and an elaborate system of pumping and circulation. All this is eliminated. Manifestly if the variable valent salts can be reduced by sulfur dioxid at all, they can always be maintained reduced under the action of this cell, especially at fairly high temperatures, say from 130 to 160 deg. F. It will not be necessary to pass the electrolyte through more than one cell, although in large plants it is preferred to pass it through two, and possibly three in series, with different current densities.

It is well known that the E. M. F. required in copper deposition from solutions containing considerable ferrous sulfate, is greatly reduced on account of depolarization at the anodes. The difficulty has been to practically maintain the iron in the ferrous condition, for, depolarization is effected by the ferrous salts being oxidized, or reduced, to the ferric condition, and if the reduction to the ferrous salts is promptly made, the advantages gained by the reduced E. M. F. due to depolarization, greatly outweighs the disadvantages due to the corrosive action of the small amount of ferric salts which can possibly exist in the electrolyte.

In this process, it is purposed to reduce the ferric and cupric salts as rapidly as formed, so that all the advantages of depolarization with ferrous iron, cuprous copper, or sulfur dioxid, can be realized without any of the corresponding disadvantages. It is believed that with a current density of 10 amperes per square foot and with an electrolyte at a temperature of 130 to 160 deg. F., the process can be carried out indefinitely with a very high current efficiency, and an E. M. F. of not to exceed one volt. At fairly high temperatures sulfur dioxid is quite active in reducing the ferric and cupric salts; on the other hand, it is not as soluble in the electrolyte. But, as the object is, not so much to absorb large quantities of the gas as to maintain the ferric and cupric salts reduced, the solubility of the gas, as used in this process, is not a matter of much consequence.

If violent agitation is desired, a portion of the gas from the exhaust pipe 22 may be introduced into the cell through the pipe 14. Air agitation cannot well be used, nor is it necessary, as the agitation can just as well, or better, be carried out with the reducing gas, of which only a small amount would be used for agitation.

The process is applicable to sulfate as well as to chlorid solutions. It may also be applied to oxidizing reactions as well as to reducing reactions.

If chlorid solutions are used, there should be sufficient base metal chlorids in the solution to retain the cuprous chlorid in solution.

The cover 2, of the electrolyzer, will usually be lined with lead to resist the corrosive action of the gas and electrolyte. To prevent any possibility of the current short circuiting through the supporting strips 17, thin strips of rubber 38, may be placed in the joints.

I claim:

1. A process which consists in confining a gas over a metal solution containing constituents capable of chemically reacting with the gas, electrolyzing the liquid, spraying the electrolyzed liquid into the gas confined over it, and maintaining a supply of gas over the liquid from which the liquid may be charged with the gas as rapidly as the gas in the liquid is combined with the constituents in the liquid capable of combining with it.

2. A process which consists in confining sulfur dioxid over a copper solution containing constituents capable of combining with it, electrolyzing the liquid, spraying the electrolyzed liquid into the gas confined over it, and maintaining a supply of the gas over the liquid from which the liquid may be charged with the gas as rapidly as the gas in the liquid is combined with the constituents in the liquid capable of combining with it.

3. A process which consists in confining a gas over a liquid containing constituents capable of combining with the gas, charging the liquid with the gas by intimately

mixing the liquid and the gas, electrolyzing the liquid, and maintaining a supply of the gas over the liquid from which the liquid may be charged with the gas as rapidly as the gas in the liquid is consumed in reacting with the constituents in the liquid capable of reacting with it.

4. A process which consists in confining sulfur dioxid over a copper solution containing salts of the variable valent elements, electrolyzing the liquid, treating the liquid with the gas during electrolysis, and maintaining a supply of the gas over the electrolyte to maintain the variable valent salts reduced to their lower valencies, and maintaining an excess of sulfur dioxid in the electrolyte.

5. A process which consists in confining a gas over a metal solution containing constituents capable of reacting with the gas, electrolyzing the liquid, and spraying the liquid into the gas above it during electrolysis.

6. A process which consists in confining a gas over a metal solution containing constituents capable of chemically reacting with the gas, electrolyzing the liquid, and intimately mixing the liquid and the gas confined over the liquid.

7. A process which consists in confining a gas over a liquid containing constituents capable of chemically reacting with the gas, electrolyzing the liquid, spraying the liquid into the gas over it, and catching a portion of the liquid sprayed into the gas and by its own gravity through a confined channel introducing it into the lower portion of the electrolyte.

8. A process which consists in confining a gas over a liquid containing constituents capable of chemically reacting with the gas, electrolyzing the liquid, spraying the liquid into the gas confined over it, and causing a circulation of the electrolyte by withdrawing a portion of the electrolyte from the upper portion and introducing it into the lower portion.

9. A process which consists in confining a gas over a metal solution containing constituents capable of chemically reacting with the gas, electrolyzing the liquid, intimately mixing the electrolyte with the gas confined over it, and causing a circulation of the electrolyte by withdrawing some of the electrolyte from the upper portion and introducing it into the lower portion.

10. A process which consists in confining over an electrolyte a gas capable of reacting chemically with the anode products of the electrolysis, intimately mixing the gas and electrolyte, and maintaining a supply of the gas over the electrolyte as rapidly as the gas is consumed in combining with the anode products of electrolysis.

11. A process which consists in confining sulfur dioxid over a metal solution containing constituents capable of chemically reacting with the gas, electrolyzing the liquid, spraying the liquid into the gas over it, and conducting a portion of the sprayed liquid through a confined channel into the lower portion of the liquid.

12. A process which consists in confining sulfur dioxid over a metal solution containing constituents capable of chemically reacting with the gas and capable of releasing hydrogen, electrolyzing the liquid, intimately mixing the liquid and the gas over it, and separating the released hydrogen from the gas over the liquid by displacement in a quiescent space.

13. A process which consists in treating solutions obtained from leaching ores of metals with precipitated copper sulfid, electrolyzing the solution, confining sulfur dioxid over the electrolyte, and intimately mixing the gas and the electrolyte.

14. A process which consists in confining a gas over a liquid containing constituents capable of combining with it, spraying the liquid upwardly into the confined gas, maintaining a flow of gas through the sprayed liquid, and electrolyzing the liquid.

15. A process which consists in electrolyzing a cuprous chlorid solution containing ferrous chlorid, and maintaining the cuprous and ferrous chlorids reduced by spraying the electrolyte during electrolysis into sulfur dioxid gas confined over the electrolyte.

16. An electrolytic process which consists in spraying the electrolyte into a gas confined over it, and circulating the electrolyte within the cell.

17. An electrolytic process which consists in spraying the electrolyte into a gas confined over it, circulating the electrolyte, and agitating the electrolyte.

18. An electrolytic process which consists in spraying the electrolyte into sulfur dioxid gas confined over it, and agitating the electrolyte with sulfur dioxid gas injected into it.

19. A process which consists in confining a gas over a liquid containing constituents capable of chemically reacting with the gas, electrolyzing the liquid, and agitating the liquid with the gas.

20. An electrolytic process which consists in treating an electrolyte with a gas confined over it, causing a flow of electrolyte longitudinally through the cell, and a circulation of the electrolyte transversely of the cell.

21. A process which consists in confining a gas over a liquid containing constituents capable of chemically reacting with the gas, electrolyzing the liquid, spraying the liquid into the gas confined over it transversely of the flow of the liquid, and causing a flow of

gas through the sprayed liquid longitudinally with the flow of the liquid.

22. A process which consists in confining a gas over a liquid containing constituents capable of chemically reacting with the gas under the action of an electric current, spraying the liquid upwardly into the gas confined over it transversely of the flow of the liquid, causing a flow of gas through the sprayed liquid longitudinally with the flow of the liquid, and passing an electric current through the electrodes.

WILLIAM E. GREENAWALT.

Witnesses:
MARY H. WOOLSEY,
HILDA L. GREENAWALT.