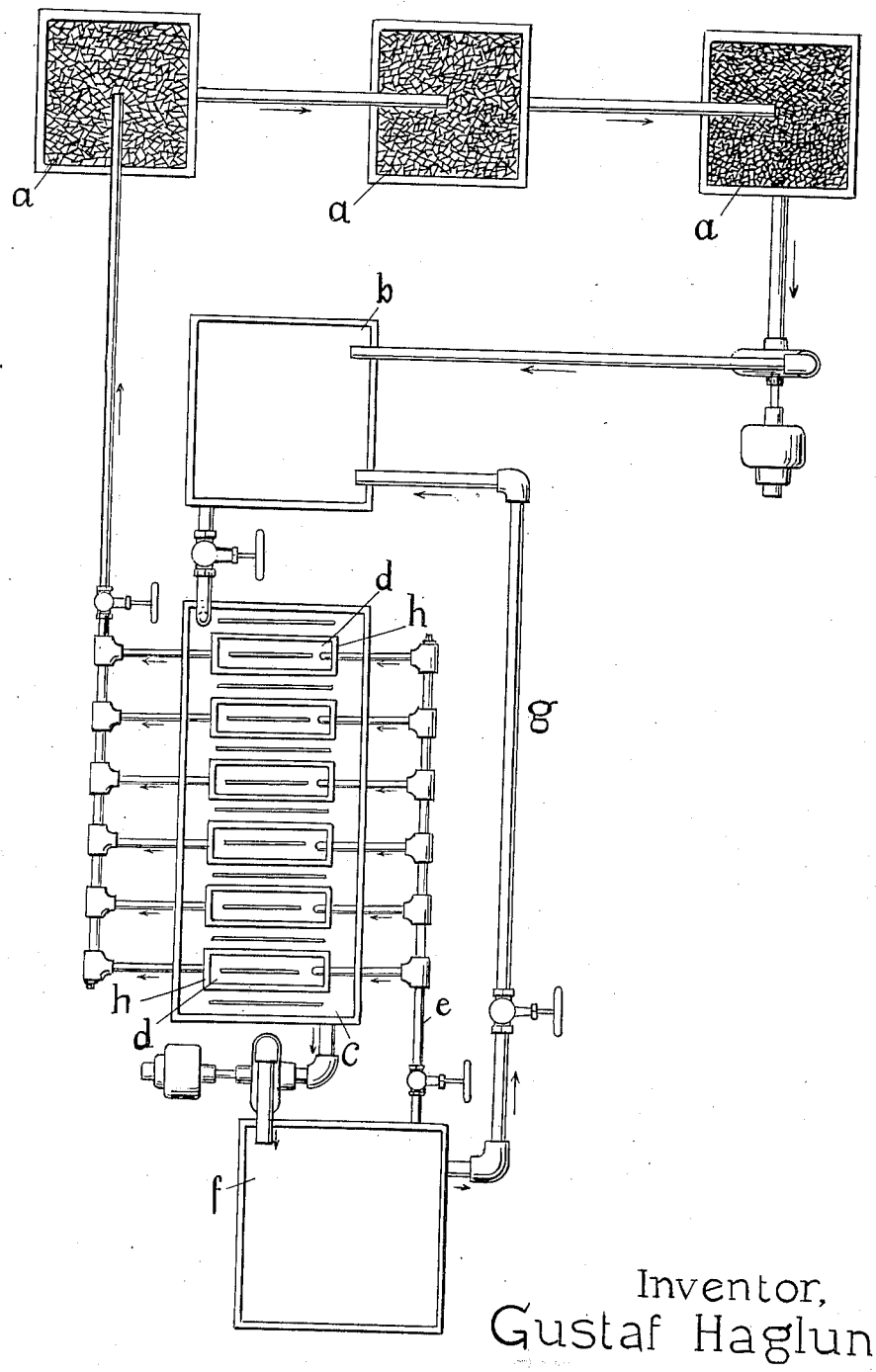
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PROCESS OF SEPARATING AND REFINING METALS.

APPLICATION FILED DEC. 5, 1918.

1,375,631. Patented Apr. 19, 1921.



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PROCESS OF SEPARATING AND REFINING METALS.

1,375,631. Specification of Letters Patent. Patented Apr. 19, 1921.

Application filed December 5, 1918. Serial No. 265,383.

To all whom it may concern:

Be it known that I, GUSTAF HAGLUND, a subject of Sweden, and a resident of Falun, Kingdom of Sweden, have invented certain new and useful Improvements in Processes of Separating and Refining Metals, of which the following is a specification.

This inventiton relates to the treatment of alloys or metal compounds such as are obtained by smelting of complex ores like copper-nickel ores and copper-zinc ores.

The object of the invention is a method whereby the metals contained in such alloys or metal compounds may be recovered separately and in a practically pure condition.

My process may be used for the treatment of various metal compounds, but I have found that it is especially suitable for the treatment of copper-nickel materials such as the matte obtained from the smelting of copper-nickel ores.

In order to recover the metals contained in such matte according to my invention I first subject the matte to a roasting and subsequent reduction process such as is well known and commonly practised, and granulate the resulting product which principally consists of metallic copper and nickel

These granules are brought into towers in which they are treated with an acid solution. Sulfuric acid is generally the most suitable acid for this purpose but other acids capable of dissolving copper and nickel may be used. In the following description of my process I will however for convenience only refer to sulfuric acid. During the treatment of the granules in the towers air is introduced at the same time as the acid which is periodically passed through the mass of granules. As the treatment progresses the acid used as a solvent will be neutralized and if sulfuric acid is used there will result a neutral solution containing principally copper sulfate and nicket sulfate. I have found that as long as the solution contains acid, both copper and nickel will enter into solution, the amount of each metal dissolved being in proportion to the amount contained in the granules. But when the solution has become neutral a cementation process takes place, the nickel contained in the granules plating out the more electronegative copper. Thereafter only nickel will enter into solution, and an equivalent amount of copper will precipitate.

I can continue this treatment until a solution of nickel sulfate is obtained which is free from copper.

I subject this solution to electrolysis using insoluble anodes, separated from the cathodes by means of diagrams, whereby the nickel sulfate is decomposed, metallic nickel being deposited on the cathode and sulfuric acid being formed at the anode.

I have found that this electrolytic process may advantageously be combined with above mentioned dissolution process, thereby utilizing the acid formed during electrolysis for the dissolution of granules.

I prefer to arrange several electrolytic vats each containing a plurality of anodes and cathodes. The anode compartments are furnished with inlets and outlets for the anolyte and may be connected in such a manner that the solution may pass through one anode compartment after the other. The cathode compartments are connected in similar manner. By such arrangement of the two independent systems of circulation I obtain great advantages. First, the contents of acid of the anolyte can be regulated and raised to the desired amount by regulating and reducing the speed of the solution passing through the anode compartments, second, the circulation of the catholyte may be arranged so as to give the best conditions for a smooth deposit of nickel on the cathode.

The acid solution leaving the anode-compartments is led to the towers for dissolution of granules. By the treatment of the granules as above set forth the solution is again neutralized and freed from copper, whereupon it is returned to the electrolytic vats.

In the tower or towers in which the cementation takes place a considerable amount of cement copper will collect. I remove this copper in any convenient manner and either sell it as cement copper or subject it to a refining process according to known methods.

In the diagram forming part of this application is shown the arrangement of the various apparatus by means of which my invention is carried out. In the specific example chosen for illustration, *a* designates three towers filled with granules. The

granules contain approximately 60% Ni and 40% Cu including small amounts of impurities.

In the towers, *a*, which may be of any suitable construction the acid solution coming from the anode compartments, *d*, is periodically percolating until all acid is neutralized and further until the nickel of the granules has plated out all copper from the solution. Thus if the solution coming from the anode compartments, *d*, contains about 15 g. Ni per liter and 40 g. H_2SO_4 per liter the neutralized solution will contain approximately 24 g. Ni and 16 g. Cu per liter, and finally after the Cu has been plated out a neutral solution will result containing 40 g. Ni per liter and being practically free from impurities. This solution is pumped into the vat, *b*, in which it is mixed with the solution from the vat, *f*, which runs into the vat, *b*, through the pipe, *g*. The solution from the vat, *f*, has just passed through the cathode compartments, *c*, in which a part of its Ni has been plated out. From the vat, *b*, the solution runs into the cathode compartments, *c*, of the electrolytic cells in which a suitable circulation is provided according to known methods. A small portion of the cathode solution in the vat, *f*, is led by means of pipe *e* into the anode compartments, *d*, which are separated from the cathode compartments by means of the diaphragms, *h*. During electrolysis sulfuric acid is formed at the anode, and the solution will when leaving the anode compartments be comparatively rich in sulfuric acid. It is then returned to the towers, *a*, for dissolution of granules as above described.

The electrolytic cells are of the type generally used for such purposes, the cathodes preferably being made of sheet iron and the anodes being made of a material which will not dissolve or disintegrate under the action of sulfuric acid and oxygen, such as magnetite, hard lead and others. Several cells may be connected in series, each cell requiring a voltage of about 3.2 volts when a current density of 10–15 amperes per square foot is used. Under these conditions a current efficiency of about 90–95% is obtained and the nickel deposited on the cathodes runs 99% Ni or more. The cement copper which accumulates in the towers for cementation is from time to time removed and discarded from the process.

Instead of using metallic granules in the towers, *a*, I may partly substitute these by granulated converter matte, as I have found that the sulfids may be dissolved upon formation of sulfates by the treatment in the towers as above described. Thus a great saving is obtained in the costs of roasting and reduction, as the metallic granules need only be used in such an amount as is required to cement out the Cu dissolved by the acid solution. The cementation will of course not be effected by sufids.

In the previous description I have only referred to copper-nickel materials. It is to be understood however that a similar result will be obtained when operating according to my method on other materials such as copper-zinc or others whose metals have an electrolytic relation to each other equal to that of copper and nickel.

It is further to be understood in referring in the claims hereof to the solution resulting from the acid treatment as being neutral, and to the solution following the cementation step as being pure, that the terms are employed in the technical sense from an industrial standpoint and not in an absolute sense from the standpoint of the chemical laboratory.

The foregoing detailed description has been given for clearness of understanding, and no undue limitation should be deduced therefrom, but the appended claims should be construed as broadly as permissible in view of the prior art.

What I claim as new and desire to secure by Letters Patent of the United States is:

1. In the separating and refining of metals the process of recovering copper and another metal from material in granular form containing the same, which comprises subjecting the said material to the action of an acid solution whereby the copper and the other metal are dissolved and a neutral solution formed, continuing the treatment after the formation of the said solution until the copper is cemented out by the other metal and a pure solution of the last named metal is formed, and recovering the said metal from the last named solution by subjecting the solution to electrolysis whereby the said metal is deposited on the cathode in refined condition.

2. In the separating and refining of metals the process of recovering copper and another metal from material in granular form containing the same, which comprises subjecting the said material to the action of a sulfuric acid solution whereby the copper and the other metal are dissolved and a neutral solution formed, continuing the treatment after the formation of said solution until the copper is cemented out by the other metal and a pure sulfate solution of the last named metal is formed, and recovering the said metal from the last named solution by subjecting the solution to electrolysis whereby the said metal is deposited on the cathode in refined condition.

3. In the separating and refining of metals the process of recovering copper and nickel from material in granular form containing the same, which comprises subjecting the said material to the action of a sul-

furic acid solution whereby the copper and the nickel are dissolved and a neutral solution formed, continuing the treatment after the formation of the said solution until the copper is cemented out by the nickel and a pure solution of nickel sulfate is formed, and recovering the nickel from the last named solution by subjecting the solution to electrolysis whereby the nickel is deposited on the cathode in refined condition.

4. The process of separating and refining metals comprising the steps of treating granules containing copper and another metal with sulfuric acid solution, whereby copper and the other metal are dissolved, thereby neutralizing the sulfuric acid solution, continuing the treatment after the solution has become neutral, whereby copper is cemented out by the other metal and a pure sulfate solution of said metal is formed, electrolyzing said sulfate solution in a diaphragm cell leading the solution first through the cathode compartment of the cell, whereby metal is deposited on the cathode, then dividing the solution in two parts, one part being led through the anode compartment and the other being mixed with the solution entering the cathode compartment, and using the solution leaving the anode compartment, rich in sulfuric acid, for dissolution of new amounts of granules containing copper and another metal.

5. The process of separating and refining metals comprising the steps of treating granules containing copper and nicked with sulfuric acid solution whereby copper and nickel are dissolved, thereby neutralizing the sulfuric acid solution, continuing the treatment after the solution has become neutral, whereby copper is cemented out by the nickel and a pure sulfate solution of nickel is formed, electrolyzing said sulfate solution in a diaphragm cell, leading the solution first through the cathode compartment of the cell, whereby nickel is deposited on the cathode, then dividing the solution in two parts, one part being led through the anode compartment and the other being mixed with the solution entering the cathode compartment, and using the solution leaving the anode compartment rich in sulfuric acid, for dissolution of new amounts of granules containing copper and nickel.

6. The process of separating and refining metals comprising the steps of treating granules containing copper and another metal in the form of sulfids with a sulfuric acid solution whereby said sulfids are dissolved, thereby neutralizing the sulfuric acid solution, using the neutral solution for treatment of granules containing metallic copper and another metal whereby copper is cemented out by the other metal and a pure sulfate solution of said metal is formed and electrolyzing said solution whereby the metal is deposited on the cathode in a refined condition.

7. The process of separating and refining metals comprising the steps of treating granules containing copper and nickel in the form of sulfids with a sulfuric acid solution whereby said sulfids are dissolved, thereby neutralizing the sulfuric acid solution, using the neutral solution for treatment of granules containing metallic copper and nickel, whereby copper is cemented out by the nickel and a pure nickel sulfate solution is formed and electrolyzing said solution whereby nickel is deposited on the cathode in a refined condition.

8. The process of separating and refining metals comprising the steps of treating granules containing copper and another metal in the form of sulfids with a sulfuric acid solution, whereby said sulfids are dissolved, thereby neutralizing the sulfuric acid solution, using the neutral solution for treatment of granules containing metallic copper and another metal whereby copper is cemented out by the other metal and a pure sulfate solution of said metal is formed, electrolyzing said sulfate solution in a diaphragm cell, leading the solution first through the cathode compartment of the cell whereby metal is deposited on the cathode, then dividing the solution in two parts, one part being led through the anode compartment and the other being mixed with the solution entering the cathode compartment, and using the solution leaving the anode compartment, rich in sulfuric acid, for dissolution of new amounts of granules containing copper and another metal in the form of sulfids.

9. The process of separating and refining metals comprising the steps of treating granules containing copper and nickel in the form of sulfids with a sulfuric acid solution, whereby said sulfids are dissolved, thereby neutralizing the sulfuric acid solution, using the neutral solution for treatment of granules containing metallic copper and nickel whereby copper is cemented out by the nickel and a pure sulfate solution of nickel is formed, electrolyzing said sulfate solution in a diaphragm cell, whereby nickel is deposited on the cathode, then dividing the solution in two parts, one part being led through the anode compartment and the other being mixed with the solution entering the cathode compartment, and using the solution leaving the anode compartment rich in sulfuric acid for dissolution of new amounts of granules containing copper and nickel in the form of sulfids.

Signed at Christiania, Norway, this 6th day of September, 1918.

GUSTAF HAGLUND.