



Electrodialytic remediation of copper mine tailings using bipolar electrodes

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ABSTRACT

In this work an electrochemical remediation (EDR) cell for copper mine tailings with bipolar stainless steel plates was analyzed. The bipolar plates were inserted inside the tailings, dividing it into independent electrochemical cells or sections, in order to increase the copper removal efficiency from mine tailings. The bipolar plates design was tested on acidic copper mine tailings with a fixed: applied electric field, liquid content, initial pH, and remediation time. The laboratory results showed that inserting bipolar plates in EDR cells improves the remediation action, even though the applied electric field is reduced by the electrochemical reactions on the plates. Basically three aspects favor the process: reduction of the ionic migration pathways, increase of the electrode surface, and in-situ generation of protons (H^+) and hydroxyls (OH^-). Furthermore, the laboratory results with citric acid addition significantly improve the remediation actions, reaching copper removal of up to nine times better, compared to conventional EDR experiments without any plates or citric acid addition.

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1. Introduction

In 2007 in Chile, copper mine production was 5.56 MMT (Million Metric Tons) of copper content [1], 36% of world production that year. Approximately 3.27 MMT of copper content was obtained by sulfide's ore flotation, generating mine tailings as a waste product. According to these figures, considering copper ore with an average-grade of 1% and copper concentrate with 30%, around 790 MMT of mine tailings pulp with 40% weight solids, were generated in 2007.

Mine tailings contain high concentrations of chemicals and elements that are of concern to the environment. Mine tailings are transported as pulp form to specially conditioned sites called tailing ponds.

Mine tailings, not only have a damaging effect on hydro resources by the natural leaching of chemicals, but also generate effects on flora and fauna, and have serious effect on air quality by the generation of fugitive emissions of fine particles.

Due to the magnitude of the mining activity in Chile, it becomes necessary to find solutions to mitigate the impact of mine tailings on the environment. One aspect of the solution to the problem is to give stability to the mine tailings by heavy metal's remediation processes. Between the processes applicable to this type of waste, electroremediation is a technique which has raised interest over the last 20 years. This type of remediation is based on the application of an electric field to a humid solid sample using two electrodes. Electro-kinetics remediation (EKR) has proven to be a good method

to remove heavy metals in fine solid waste such as grounds, clays, ashes and mud [2–4]. On the other hand, electrochemical remediation (EDR) [5,6] improves the process with the introduction of ionic interchange membranes, isolating the phenomena of remediation from electrode reactions.

The electro-remediation of mine tailings, as in the case of the treatment of other solid waste, requires enhancement method which had been already reported in literature, among others, additions of complexing agents [7], addition of bacteria [8], pH control [9], and use of ultrasound [10].

In order to further enhance EDR, in this work stainless steel plates are inserted inside mine tailings, as is shown in Fig. 1. When an electric field is generated between these plates, they behave in bipolar form [11]: one of its faces acts as an anode and the other as a cathode according to the position they have with respect to the electrode work (monopolar) of the EDR cell. In these bipolar electrodes the following electrode reactions will occur: oxidation and reduction of water, oxidation and reduction of the pair Fe^{2+}/Fe^{3+} , reduction Cu^{2+}/Cu^0 , and other electrodes reactions, depending on electrolyte availability. On the other hand, as a consequence of the electrochemical reactions of water, chemical reactions in the neighboring zones to the electrodes (bipolar plates), such as oxidation Fe^{2+}/Fe^{3+} and precipitation of $Cu(OH)_2$, could be produced.

By installing several stainless steel plates, the original EDR cell will be divided into independent sections according to the number of installed plates. The main objective is to increase the active surfaces of the electrodes of the original EDR cell, to reduce the migration pathway of the polluting agents, and to generate in-situ, protons (H^+), hydroxyls (OH^-), and oxidizing agents like the Fe^{3+} , that will increase the dynamics of the process.

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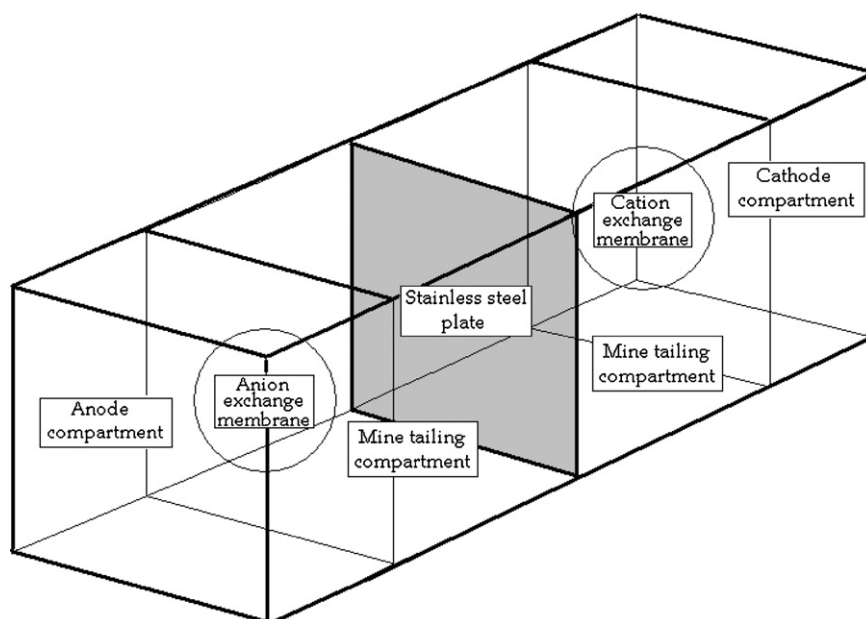


Fig. 1. Schematic description of the EDR cell with one bipolar plate.

Table 1

Summary characteristics of the mine tailings investigated.

| | |
|-------------------------|--|
| Ore deposit type | Porphyry copper |
| Primary ore minerals | Pyrite, chalcocopyrite, bornite, molybdenite, galena, tennantite, magnetite, hematite |
| Secondary ores minerals | Chalcocite, covellite |
| Gangue minerals | Quartz, albite feldspar, biotite, calcite, anhydrite, tourmaline-schorl, rutile, apatite, sericite-muscovite, chlorite, epidote, kaolinite, monmorillonite |
| Grain size distribution | 50% (w/w) smaller than 200 μm |
| Cu content | 1610 \pm 22 mg/kg dry matter |
| pH | 3–4 |

2. Experimental

2.1. Experimental tailings

The mine tailing used for remediation experiments was sampled from the Cauquenes impoundment at Codelco-El Teniente copper mine in the VI Region of Chile. Table 1 shows general characteristics of mine tailings used in this work.

2.2. Analytical methods

2.2.1. Copper concentration

The copper content of the tailings was determined by adding 20 mL 1:1 HNO_3 to 1.0 g of dry material and treating the sample in autoclave, according to the Danish Standard DS 259:2003 (30 min at 200 kPa (120 °C)). The liquid was separated from the solid particles by vacuum through a 0.45 μm filter and diluted to 100.0 mL. The metal content was determined by AAS in flame. At least three analysis of each sample were measured and an average was used in this work.

2.2.2. pH

pH was measured by mixing 5.0 g dry matter and 25.0 mL distilled water. After 1 h of contact time, pH was measured using a pH electrode.

2.3. Tailings pre-treatment

Before remediation experiments, the tailings were stove-dried for two days at 70 °C. Once dried, the material was pulverized in a mortar and sieved with meshes #4 and #20, until its

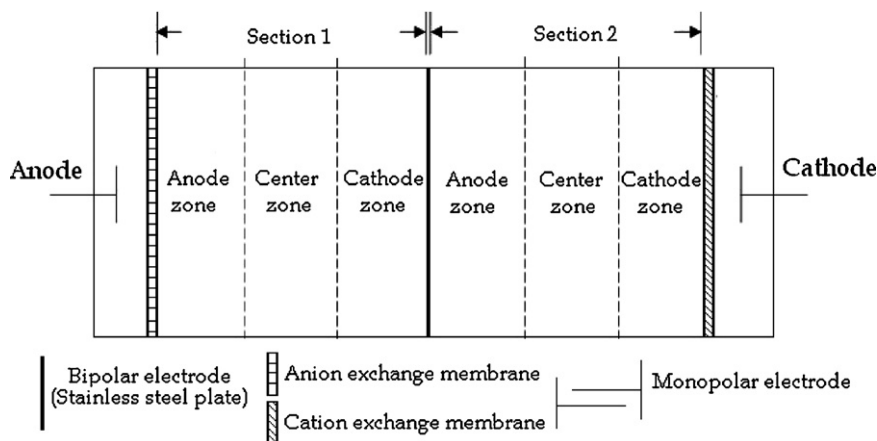


Fig. 2. EDR cell with one bipolar plate showing division in sections and zones.

Table 2
Electrolytic remediation conditions.

| Experiment | Pre-treatment | Duration (days) | No. of bipolar plates | Initial pH | Solid dry weight (kg) |
|------------|-----------------|-----------------|-----------------------|------------|-----------------------|
| 1 | Distilled water | 18 | 0 | 4.1 | 1.40 |
| 2 | Citric acid | 18 | 0 | 4.1 | 1.50 |
| 3 | Distilled water | 18 | 1 | 4.2 | 1.41 |
| 4 | Citric acid | 18 | 1 | 4.2 | 1.46 |
| 5 | Distilled water | 18 | 3 | 4.0 | 1.50 |
| 6 | Citric acid | 18 | 3 | 4.1 | 1.44 |
| 7 | Distilled water | 3 | 1 | 4.1 | 1.42 |

homogenization. Depending on the experiment, distilled water or 1 M citric acid was added to the tailings until an average humidity of 20% was reached.

2.4. Experimental cell design

The experiments were carried out in an opened acrylic straight parallelepiped cell with a rectangular base of 28 × 10 [cm] and 10 [cm] height, divided in three compartments. Inside the center compartment, 20 [cm] in length, the mine tailing was located. In the two side compartments, 4 [cm] in length each, titanium electrodes were located (anode and cathode) immersed in the electrolyte (see Fig. 1). The bipolar plates are made of AISI 316 stainless steel, 9.6 × 9.6 [cm] square shape, with a thickness of 1 mm each. In order to separate the central compartment from the lateral ones, ionic interchange membranes were used: cationic CMI-7000 and anionic AMI-7001. The solutions of the electrodes were initially distilled water, and later sulfuric acid was added to adjust the pH of the solution under 4.0 to avoid precipitation in the cathode compartment.

After the experiments were carried out, mine tailings remaining in the compartments were segmented into three equal slices, where copper concentration, pH and water content were measured. In this work anode zone is defined as the slice closest to the anode, center zone the slice in the middle, and cathode zone the slice closest to the cathode. Depending on the number of bipolar plates used in the experiments, a greater number of the already defined zones: anode, center and cathode are obtained. Thus using one bipolar plate two sections are obtained each one with its anode, center and cathode zone (see Fig. 2).

2.5. Experimental plan

Seven EDR experiments were carried out with the initial conditions given in Table 2. In all of the experiments, a voltage of 20V

was applied, to an acid sample of mine tailing whose initial humidity was adjusted to approximately 20%. The pH in the tailing was initially adjusted to around 4.0, either with distilled water or citric acid solution.

The objective of experiments 1, 3 and 5 was to evaluate the effect of the number of bipolar plates in the cell (0, 1 and 3, respectively) with distilled water addition, with an EDR remediation time of 18 days. Experiments 2, 4 and 6 were the same experiments as 1, 3 and 5, except that citric acid solution was used to adjust pH to 4.0. Experiment 7, with a remediation time of 3 days, was carried out under similar conditions to experiment 3 to verify the effect of the remediation time.

In order to evaluate the copper removal, the experiments with plates inserted in the mine tailings, were divided in sections of equal size according to the number of bipolar plates used (for instance, see Fig. 3 with three bipolar plates). The subdivision in zones of Fig. 2 was applied for all sections. Consequently, the results presented here for the copper removal in an individual zone, is the average in the cell(s) or section(s) defined as:

$$\overline{\text{Zone } M} = \frac{\sum_{n=1}^z \text{Zone } M_n}{z}$$

where M_n is anode, center and cathode, zone of mine tailing in the section n and z is number of sections of mine tailing (1, 2, and 4).

3. Results and discussion

General remediation results are given in Tables 3 and 4. Table 3 includes EDR results in terms of copper concentrations using zero, one and three bipolar plates, and Table 4 shows the details of the copper mass balance for all the sections of mine tailings defined in each experiment.

Table 4 was prepared considering for each section: (a) the initial and final copper concentrations, (b) the mass of tailings in each zone, and (c) the copper leaving each zone. The mass tailing in each

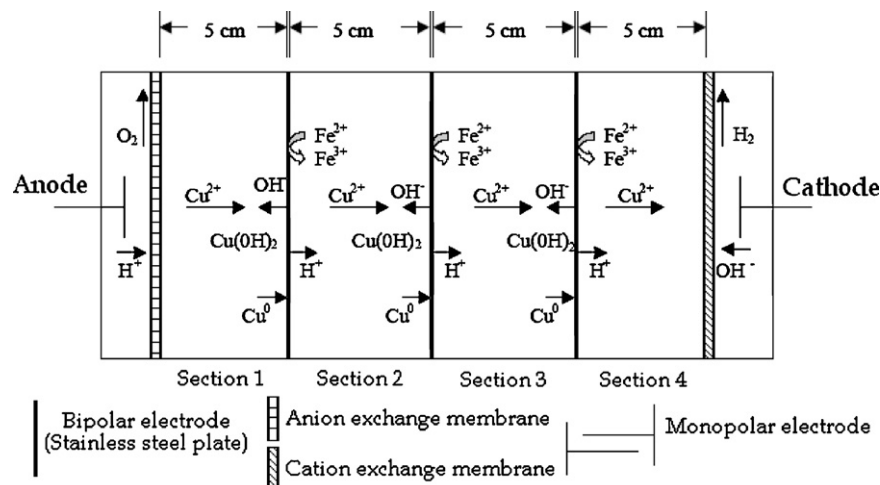


Fig. 3. EDR cell with three bipolar plates.

Table 3
Initial C_0 and final C_F copper concentrations (mg/kg) in mine tailings.

| Exp | Plates | Section | C_0 | Anode zone | | Center zone | | Cathode zone | | Cathode zone* | |
|-----|-----------|-----------|-----------|------------|-------------|-------------|-------------|--------------|-------------|---------------|-------------|
| | | | | C_F | $1-C_F/C_0$ | C_F | $1-C_F/C_0$ | C_F | $1-C_F/C_0$ | C_F | $1-C_F/C_0$ |
| 1 | 0 | Section 1 | 1623 | 1470 | 0,09 | 1613 | 0,01 | 1741 | -0,07 | 1786 | -0,10 |
| 2 | | | 1604 | 1446 | 0,10 | 1580 | 0,01 | 1767 | -0,10 | 1786 | -0,11 |
| 3 | 1 | Section 1 | 1607 | 1462 | 0,09 | 1547 | 0,04 | 1711 | -0,06 | 1798 | -0,12 |
| 4 | | | 1625 | 1394 | 0,14 | 1567 | 0,04 | 1735 | -0,07 | 1891 | -0,16 |
| 7 | | | 1584 | 1430 | 0,10 | 1605 | -0,01 | 1671 | -0,05 | 1732 | -0,09 |
| 3 | | Section 2 | 1607 | 1406 | 0,13 | 1607 | 0,00 | 1715 | -0,07 | 1811 | -0,13 |
| 4 | | | 1625 | 1430 | 0,12 | 1599 | 0,02 | 1692 | -0,04 | 1896 | -0,17 |
| 7 | | | 1584 | 1462 | 0,08 | 1608 | -0,02 | 1695 | -0,07 | 1696 | -0,07 |
| 5 | | 3 | Section 1 | 1583 | 1374 | 0,13 | 1575 | 0,01 | 1663 | -0,05 | 1809 |
| 6 | 1641 | | | 1350 | 0,18 | 1615 | 0,02 | 1687 | -0,03 | 1952 | -0,19 |
| 5 | Section 2 | | 1583 | 1438 | 0,09 | 1567 | 0,01 | 1711 | -0,08 | 1754 | -0,11 |
| 6 | | | 1641 | 1398 | 0,15 | 1575 | 0,04 | 1683 | -0,03 | 2025 | -0,23 |
| 5 | Section 3 | | 1583 | 1422 | 0,10 | 1480 | 0,07 | 1530 | 0,03 | 1892 | -0,20 |
| 6 | | | 1641 | 1440 | 0,12 | 1520 | 0,07 | 1600 | 0,02 | 2005 | -0,22 |
| 5 | Section 4 | | 1583 | 1374 | 0,13 | 1486 | 0,06 | 1583 | 0,00 | 1945 | -0,23 |
| 6 | | | 1641 | 1326 | 0,19 | 1518 | 0,07 | 1644 | 0,00 | 2210 | -0,35 |

* Recalculated values.

zone was calculated as the initial solid dry weight of the section divided by 3. The copper leaving each zone was calculated as:

Copper leaving = initial copper + entering copper – final copper.

At the anode zone, the amount of copper entering was considered always zero. In the cathode zone copper leaving corresponds to the copper removed from the tailings across the cation exchange membrane or to electro-deposition of the copper on the surface plate which acts as cathode, depending on the section under analysis.

To quantify the copper removal from the cathode zones in all the sections, the copper leaving was considered as part of the final content of the metal in it. Therefore, the final copper concentration (C_F) was recalculated and copper removal was quantified as $(1 - C_F/C_0)$ as it is shown in Table 3. As it can be realized from the negative values in the last column of Table 3, as expected there was accumulation of copper in the cathode zone.

The assumption, that copper leaving the cathode must be considered as accumulation, is a conservative assumption since in a full-scale remediation application of this method, the solution near the cathode will be pumped out from the electrode wells.

The values of copper analysis detailed in Tables 3 and 4 were used to calculate the average copper removal as percentage of total

copper content. Tables 5 and 6 show the results obtained. Table 5 shows the effect of inserting zero, one and three bipolar plates in EDR results for 18 days remediation time and pre-treatment by using distilled water and citric acid. Table 6 shows the effect of remediation time in EDR results inserting one bipolar plate and pre-treatment by using distilled water.

3.1. Effect of number of bipolar plates

The removal of copper in the anode zone increases proportionally with the number of bipolar plates, independently of the type of pre-treatment of the sample. In the center zone, the tendency of copper removal follows the same tendency, and in the cathode zone, the tendency is similar but in terms of copper accumulation. In general, the results with citric acid are higher than those with distilled water, mainly because citric acid is a complexed agent which facilitates the dissolution of copper from mine tailings. All this resulted from the analysis of data in Table 5, for 18 days EDR remediation time.

The increase in the removal of copper when using more bipolar plates (0–3) can be explained:

- by the increase in active surface of the anode and cathode,

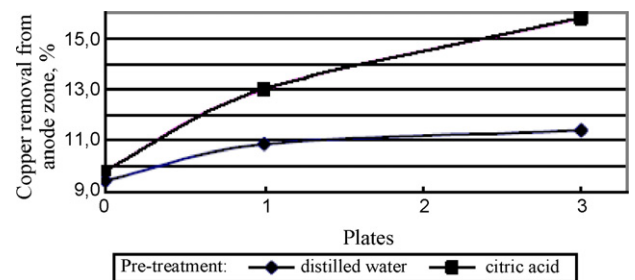
Table 4
Copper mass balance.

| Exp | Plates | Section or cell | Anode zone (mg) | | | | Center zone (mg) | | | | Cathode zone (mg) | | | | | |
|-----|--------|-----------------|-----------------|---------|-------|---------|------------------|---------|-------|---------|-------------------|---------|-------|---------|-----|----|
| | | | Entering | Initial | Final | Leaving | Entering | Initial | Final | Leaving | Entering | Initial | Final | Leaving | | |
| 1 | 0 | 1 | 0 | 751 | 681 | 71 | 71 | 751 | 747 | 75 | 75 | 751 | 806 | 21 | | |
| 2 | | | 0 | 802 | 723 | 79 | 79 | 802 | 790 | 91 | 91 | 802 | 884 | 10 | | |
| 3 | 1 | 1 | 0 | 341 | 310 | 31 | 31 | 402 | 387 | 46 | 46 | 384 | 409 | 21 | | |
| 4 | | | 0 | 364 | 312 | 52 | 52 | 384 | 370 | 65 | 65 | 400 | 427 | 38 | | |
| 7 | | | 0 | 383 | 346 | 37 | 37 | 371 | 376 | 32 | 32 | 345 | 364 | 13 | | |
| 3 | | 2 | 0 | 379 | 332 | 47 | 47 | 387 | 387 | 47 | 47 | 374 | 400 | 22 | | |
| 4 | | | 0 | 431 | 379 | 52 | 52 | 406 | 400 | 58 | 58 | 349 | 364 | 44 | | |
| 7 | | | 0 | 396 | 366 | 31 | 31 | 369 | 375 | 25 | 25 | 352 | 376 | 0 | | |
| 5 | | | 3 | 1 | 0 | 174 | 151 | 23 | 23 | 283 | 282 | 24 | 24 | 171 | 180 | 16 |
| 6 | | | | | 0 | 159 | 131 | 28 | 28 | 218 | 215 | 32 | 32 | 167 | 172 | 27 |
| 5 | 2 | 0 | | 176 | 160 | 16 | 16 | 198 | 196 | 18 | 18 | 168 | 181 | 5 | | |
| 6 | | 0 | | 217 | 185 | 32 | 32 | 167 | 161 | 39 | 39 | 166 | 170 | 35 | | |
| 5 | 3 | 3 | 0 | 182 | 164 | 19 | 19 | 261 | 244 | 36 | 36 | 182 | 176 | 42 | | |
| 6 | | | 0 | 226 | 199 | 28 | 28 | 222 | 205 | 44 | 44 | 199 | 194 | 49 | | |
| 5 | | 4 | 0 | 168 | 146 | 22 | 22 | 241 | 226 | 37 | 37 | 161 | 161 | 37 | | |
| 6 | | | 0 | 210 | 170 | 40 | 40 | 236 | 219 | 58 | 58 | 167 | 168 | 58 | | |

- by the reduction in the migration pathway of the polluting agents,
- by the in-situ generation of H^+ which increase dissolution of copper in a direct and also indirect form,
- by the oxidation Fe^{2+}/Fe^{3+} which increase the oxidizing power of the electrolyte.

Fig. 4 shows the copper removal in the anode zone when using bipolar plates (0–3). The pre-treatment with citric acid shows better results in all cases without reaching a maximum removal value. However, when three plates are used for the EDR with distilled water, an asymptotic behavior is observed.

Figs. 5 and 6 show the copper removal at the center and cathode zones, respectively. In the center zone, copper removal is less than in the anode zone as shown in Fig. 4. The accumulation in the cathode

**Fig. 4.** Copper removal from anode zone, with zero, one and three bipolar plates, for 18 days of EDR.**Table 5**
Effect of inserting bipolar plates in EDR results for 18 days of remediation time and pre-treatment with distilled water and citric acid.

| Experiment | Pre-treatment | Plates | Removal anode zone (%) | Removal center zone (%) | Removal cathode zone (%) | Total charge passed (C) |
|------------|-----------------|--------|------------------------|-------------------------|--------------------------|-------------------------|
| 1 | Distilled water | 0 | 9.4 | 0.6 | -10.0 | 76200 |
| 3 | | 1 | 10.9 | 1.9 | -12.3 | 28700 |
| 5 | | 3 | 11.4 | 2.6 | -16.8 | 18000 |
| 2 | Citric acid | 0 | 9.9 | 1.5 | -15.4 | 100000 |
| 4 | | 1 | 13.0 | 2.6 | -16.5 | 44400 |
| 6 | | 3 | 15.8 | 5.2 | -24.7 | 31000 |

Table 6
Effect of remediation time in EDR results with one bipolar plate and pre-treatment with distilled water.

| Experiment | Duration (days) | Removal anode zone (%) | Removal center zone (%) | Removal cathode zone (%) | Total charge passed (C) |
|------------|-----------------|------------------------|-------------------------|--------------------------|-------------------------|
| 7 | 3 | 8.7 | -1.4 | -8.3 | 3900 |
| 3 | 18 | 10.9 | 1.9 | -12.3 | 28700 |

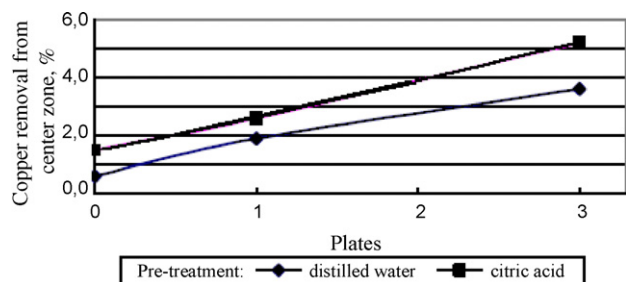


Fig. 5. Copper removal from center zone, with zero, one and three bipolar plates, for 18 days of EDR.

zone follows a similar tendency to the observed values in the anode and center zones.

In summary, according to the results obtained in the EDR experiments for 18 days, applying a voltage of 20V, there was a proportional increase in the average copper removals by increasing the number of bipolar plates (0–3). Therefore, by increasing the number of plates a better use of energy is obtained. However, for a full-scale remediation with bipolar plates, the number of plates will be an economical decision, because more plates mean less energy consumption for the remediation, but on the other hand also mean an increase in investment because of the need of installation of more electrode wells.

3.2. Effect of remediation time

As expected, the increase in remediation time from 3 to 18 days increases the removal and concentration of copper in the anode and cathode zones, respectively. From the analysis of the results for different remediation time, it can be concluded that 80% of the total copper removed in the anode zone, and 68% of the accumulated copper in the cathode zone, which are obtained with a remediation time of 18 days, are reached in just 3 days. No significant copper removal in the center zone is detected for a remediation time of 3 days; this means that copper is mainly transferred in and out of this zone. Higher remediation times would increase the copper removal in the center zone of the cell and thus a greater amount of remediated material is obtained. The details of the results are shown in Table 6.

3.3. Removal efficiency

The total electric charge passed through the mine tailing is given in Tables 5 and 6, and was obtained from current-time recording. Analysis of the data shows that the using of more bipolar plates (0–3) will produce a lower use of electricity. On the other hand, citric acid pre-treatment increases the use of electric charge because of the increase in copper dissolution as result of complex formation.

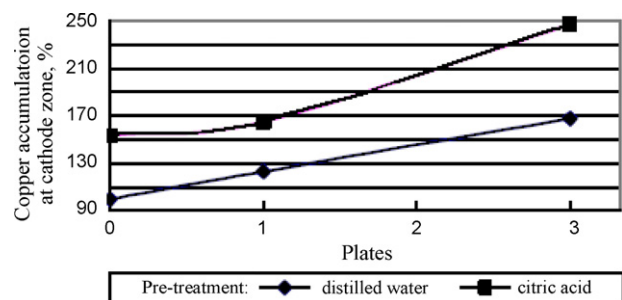


Fig. 6. Copper accumulation at cathode zone, with zero, one and three bipolar plates, for 18 days of EDR.

If the current efficiency is evaluated by the ratio between the true metal removal and the theoretical removal given by the total charge passing, according to Faraday's law, it can be concluded that current efficiency will increase when using more bipolar plates.

The current efficiency reaches a maximum value of 1.3% for a remediation time of 18 days, using three bipolar plates.

The smaller electric charge obtained when using more bipolar plates in the mine tailing is explained by the reasons stated in Section 3.1, above.

3.4. Final discussion

In general, it should be noted that in the original mine tailing, copper could be expected to be found as residual copper sulfide, which was not liberated in the grinding process prior to flotation.

The low copper removals obtained here were due to the low soluble copper content in the tailings. This content, of soluble copper, is variable due to the heterogeneous origin of the mine tailings in the ponds, among others reasons: copper grades depend on the original characteristics of the tails disposed, aging of the tailing in the ponds as consequence of physical–chemical changes due to weathering and bacterial actions in time.

The use of this remediation technology will imply the periodic application of the method in order to remove the additional soluble copper that will be generated with time. Therefore, the remediation action for this heterogeneous solid waste is to remove the soluble copper in the tailings and in this way making the final residue more stable.

4. Conclusions

From the analysis of the results obtained, the following conclusions can be made:

- The use of bipolar plates in the electrochemical remediation of copper improves the process substantially, mainly in terms of increasing the copper removal. As expected, copper removal was higher using citric acid in the pre-treatment of the mine tailings.
- Copper removal could be improved by increasing the remediation time by more than 18 days, and in this way, also a major amount of remediated material can be processed.
- The effective voltage of the process diminishes using more plates, and as a consequence, the electric energy consumption. Nevertheless, better copper removals and current efficiencies are obtained.
- This remediation technology could be promising to remove soluble copper from mine tailings, generating a more stable residue.

Acknowledgements

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