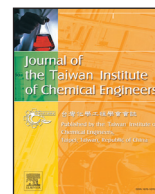




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Enhanced electrochemical removal of arsenic and heavy metals from mine tailings

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ABSTRACT

Electrochemical remediation process can be used to separate arsenic and heavy metals from mine tailings and prevent them from leaching into the environment. Citric acid, EDTA, NaOH, HCl and H₂O were used as chemical extractants in the electrokinetic process. The electrochemical extraction with citric acid as extractant was the most effective method to remove arsenic, iron and manganese from mine tailings highly contaminated. As, Fe and Mn removal from mine tailings was influenced for the citric acid concentration, the liquid/solid ratio, the current intensity and the extraction time. Different values for each of these parameters were tested in order to check their effects on the removal efficiency. The highest efficiency and smallest residual concentrations of contaminants were reached for concentrations of citric acid of 0.8 mol/L, liquid/solid ratio of 10, current intensity of 800 mA and extraction time of 6 h. Too high current intensities or too long extraction times caused corrosion problems in the steel parts of the system.

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1. Introduction

Arsenic and heavy metal toxicity occurs far more often and people are exposed to toxic metals on a day-to-day basis in our environment. Mining activities are one of the primary contributors to arsenic and metals contaminated soil problems. Mining operations such as those developed in the tungsten mines have resulted in vast tracts of land being contaminated with arsenic and heavy metals.

The recovery of degraded areas is becoming a challenge because remediation trials have shown that a full recovery is not possible in the majority of cases due to the technical or the economic limitations.

The soil structure and the difficulty of degradation or extraction and transport of some contaminants make the existing techniques not suitable to the removal of these contaminants particularly from fine grained soils.

Chemical extraction [1–3] and electrokinetic treatment [4–6] have been used to remediate soil; the results obtained by these different methods were not satisfactory. Nevertheless, these techniques can be combined to improve soil remediation.

Electrokinetic remediation involves applying a low direct current or a low voltage gradient to a pair of electrodes that are inserted into the soil. To remove contaminants from soil by electrokinetic techniques, the contaminants should be in pore water in soluble ionic form so that they are transported to the electrodes depending on the nature of their charge. The contaminant removal efficiency depends on how great a percentage of the contaminant can be transformed into a soluble form and how rapidly [7].

In order to increase electrokinetic remediation efficiency, the conditioning of electrodes with the addition of suitable aqueous solutions (extractants) is necessary. The conditioning solutions enhance the solubilization of arsenic and metals and favour the contaminant migration. This combination of chemical and electrokinetic extractions (electrochemical extraction) has also been used to remediate soil [8–10]; however, the results have been unsatisfactory until now.

The purpose of this work was to increase the efficiency of electrochemical arsenic and heavy metal removal in difficult soils highly contaminated such as mine tailings. The heavy metals studied were iron and manganese. These metals (essentials for biological processes) become toxic in high concentrations. They are elements frequently associated to As with influence on As removal efficiency from soils [11,12]. The mine tailings used in this study contain high concentrations of As as main contaminant but also of Fe and Mn. The effects of process variables on As, Fe and Mn

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Table 1
Characteristics of mine tailings.

Property	Value
Moisture (%)	1.37 ± 0.06
pH	4.32 ± 0.02
Particle size distribution (%)	
Sand	21.93 ± 0.48
Silt	65.53 ± 0.39
Clay	14.53 ± 0.30
Cation exchange capacity (meq/100 g)	13.52 ± 0.33
As (mg/kg)	1020.51 ± 88.84
Fe (mg/kg)	26,474.80 ± 1860.23
Mn (mg/kg)	126.64 ± 11.98

All properties are expressed as percentages and on a dry matter basis except moisture.

extraction including extractant type, extractant concentration, soil to liquid ratio, electric current and time were investigated.

2. Materials and method

2.1. Mine tailings samples

Samples were obtained from the tungsten mine tailings of Barreco (Spain). The sampling procedure was carried out so that aliquot was a composite sample consisting of seven subsamples randomly taken in homogenized tailings and collected from a depth of approximately 0.2–1.5 m below the surface. The subsamples were air dried for a week and then sieved through a 2.0 mm opening sieve and mixed to homogenize uniformly.

2.2. Chemical analyses

Moisture content was measured on samples of approximately 10 g on the basis of the weight lost after oven drying at 105 °C until constant weight [13]. The pH value of the samples was analysed in a suspension of 10 g of sample in 25 mL of distilled water using a pH meter (CRISON-GLP22) [14]. The particle size distribution was determined by the Robinson pipette method [15] separating the fractions of sand (>50 µm), silt (50–2 µm) and clay (<2 µm). Cation exchange capacity (CEC) was measured using the ammonium acetate method [16]. The content of arsenic and heavy metals was determined by inductively coupled plasma-optical emission spectrometry according to method 200.7 of the Environmental Protection Agency [17]. All tests and analyses were conducted in triplicate, and the results were expressed as mean ± standard deviation. Table 1 shows the basic properties of the mine tailings samples.

2.3. Electrokinetic experiments

Experiments were conducted in a stirred glass vessel of 9.5 cm (Ø) × 12 cm (L) where sample and extractant solution were introduced. Graphite bar electrodes 120 × 8 mm, 60 mm apart, partially immersed in extractant solution to a depth of 60 mm were used for the anode and the cathode electrodes in this unit. Stirred glass vessels of 13 cm (Ø) × 18.5 cm (L) and 6 cm (Ø) × 12 cm (L) were used for experiments with more than 500 mL or less than 100 mL of extractant solution, respectively; the separation and the depth of immersion of electrodes were adapted to the size of the vessels. Anode and cathode were connected to a power pack (TENMA 72-7245) capable of delivering 0–3 A at up to 30 V. The electric current intensity, voltage and temperature were monitored during the test periods.

Five separate sets of experiments have been conducted to determine the effect of different operating parameters on the process: extractant type, extractant concentration, liquid/solid ratio, electric

current intensity and time. Although temperature and pH are important factors affecting the extraction, their influence was not determined in this work. Operating conditions for the tests are listed in Table 2.

After electrokinetic experiments, treated mine tailings were separated from solution by filtration and sequential washing of the solid.

3. Results and discussion

Reference operating conditions were selected according to the literature: chemical concentration of 1.0 mol/L [18,19], extraction time of 6 h [3,18], liquid/solid ratio of 10/1 [3,20] and electric current intensity of 600 mA [21]. The stirring rate was fixed at 600 rpm to generate a 2 cm vortex necessary to prevent particle deposition.

3.1. Influence of extractant type and extractant concentration

Extractants were chosen from literature on arsenic removal [1,22,23]. The extractant solutions were prepared by dissolving weighed amounts of the desired compounds in tap water. The presence of minimal quantities of metals in tap water did not interfere in the extraction process. Tap water was also used as extractant.

Results (Fig. 1) showed that citric acid is the best extractant for As (56.06 ± 0.45%), HCl is the best extractant for Fe (20.20 ± 9.22%) and Mn (63.15 ± 14.06%), and water is the worse extractant in all cases (0%). The extraction sequence is:

As: C₆H₈O₇ > EDTA > NaOH > HCl > H₂O

Fe: HCl > C₆H₈O₇ > EDTA > NaOH > H₂O

Mn: HCl > C₆H₈O₇ > EDTA > NaOH > H₂O

Since the key element of the mine tailings remediation is As, the choice of extractant is determined by the As removal. Accordingly, the most suitable extractant is citric acid that has also been proved successful in Fe and Mn removal.

In order to check the influence of the extractant concentration on the electrochemical remediation, a citric acid concentration range of 0.0–1.0 mol/L was studied.

Extraction of As was highly dependent on the citric acid concentration (Fig. 2a). The amount of As extracted increased as the citric acid concentration increased to 0.6 mol/L reaching a maximum As removal of 66.5%, but above 0.8 mol/L of citric acid a decrease was observed in the As removal. Fe and Mn electrochemical extraction by citric acid showed a similar trend with respect to As extraction (Fig. 2b and c), and a maximum extraction of Fe and Mn was obtained at a citric acid concentration of 0.8 mol/L corresponding to 17.0% Fe removal and 53.5% Mn removal. According to these results, 0.8 mol/L citric acid seems to be the most suitable concentration for the maximum extraction of As, Fe and Mn.

Although there are no studies on the influence of the concentration of citric acid on the electrochemical extraction of As, Fe and Mn, a study on chemical extraction of As with citric acid demonstrated that an increase in the citric acid concentration from 0.05 mol/L to 0.10 mol/L raises around 10 times the chemical extraction of As in contaminated soils [18]. Similar results were found for extractants other than citric acid: all studies show a clear relationship between the extractant concentration and the As, Fe or Mn removal. This relationship can be direct or indirect [12,24,25] which agrees with our results.

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