Minerals Engineering 55 (2014) 52–56

Contents lists available at ScienceDirect

Minerals Engineering

journal homepage: www.elsevier.com/locate/mineng

Electrokinetic remediation of mine tailings by applying a pulsed variable electric field

Adrián Rojo *, Henrik K. Hansen, Omara Monárdez

Departamento de Ingeniería Química y Ambiental, Universidad Técnica Federico Santa María, Casilla 110-V, Valparaíso, Chile

ARTICLE INFO

Article history:
Received 9 May 2013
Accepted 8 September 2013
Available online 5 October 2013

Keywords:
Electrokinetic remediation
Heavy metals
Copper mine tailings
Pulsed electric field

ABSTRACT

Following the flotation step of copper sulphide mineral processing, a considerable amount of ground ore in the form of a pulp containing heavy metals and other polluting compounds is discarded as waste, known as “mine tailings”. This waste is deposited behind dams, and unless it is treated, it represents a danger to the environment because the natural oxidation of heavy metals makes the waste chemically unstable.

Electrokinetic remediation (EKR) is a technology used to remove contaminants from soils. In recent years, the technology has been of research interest for stabilising mine tailings from the copper industry.

Nine EKR experiments with pulses of sinusoidal electric fields (by applying simultaneously DC and AC voltages) and an AC voltage frequency of the order of kHz were performed to improve conventional EKR and stabilise synthetic tailings samples. The synthetic tailings were prepared based on representative data for tailings from a combined Cu–Mo concentrator plant.

It was found that, in general, the use of a pulsed sinusoidal electric field favoured overall copper removal in the EKR cell, and particularly good results were observed when this type of electric field produced periodical polarity reversal in the electrodes.

The best results in terms of overall cell removal and specific energy consumption were obtained under the following conditions: (i) effective voltage of 14.6 V (VDC = 10 V and VAC = 15 V), (ii) AC voltage frequency 1000 Hz, (iii) electrical field applied in pulses with a time ratio of 25.

1. Introduction

Electrokinetic remediation (EKR) is an in situ treatment technology for restoration of contaminated hazardous waste sites (Acar and Alshawabkeh, 1993). The conventional application of this alternative treatment uses direct current (DC) applied across electrodes inserted in the soil to generate an electric field for the mobilisation and extraction of contaminants (Probstein and Hicks, 1993). In the case of waste from copper mining, previous work (Rojo et al., 2006) has shown that the conventional DC system was limited with regard to metal removal efficiency and had a very high electrical energy consumption. Under this scenario sinusoidal EKR was applied by an electric field through the simultaneous application of DC and AC voltages.

The driving mechanisms for EKR are transport phenomena in an electric field, particularly electro-migration (ionic migration) and electro-osmosis, coupled with electrolysis and geochemical reactions (Alshawabkeh and Bricka, 2000).

Electro-migration is the transport of charged ions in the pore fluid liquid toward the electrode that is opposite in polarity to the ions. The rate of electro-migration depends on the values of the effective ionic mobility of contaminants, considering soil porosity and tortuosity. Electro-osmosis uses an electric field to move contaminants from one location to another within flowing water, typically from the anode toward the cathode. Depending on the flow direction, the migration of some ions may be enhanced, while others with the opposite charge may be slowed.

The relative contribution of electro-migration and electro-osmosis to overall transport in an electric field varies for different soil types, water content, types of ions, pore fluid chemistry, and boundary conditions. For all of the above, EKR refers to coupled charge and fluid transport.

Water is electrolysed by the electric field electrodes in a remediation cell. Oxidation at the anode and reduction at the cathode generate, respectively, an acid and a base front in the remediation cell. The acid generated at the anode moves through the soil towards the cathode by electro-migration and electro-osmosis, while the base generated at the cathode moves towards the anode by electro-migration and diffusion. Acid production generally enhances the process, and a high pH zone adversely affects the extraction of heavy metals from soils. In relative terms, the acid front dominates the base front due to the greater mobility of H^+ ions and backflow due to electro-osmosis that retards the base front.

* Corresponding author. Tel.: +56 322654463; fax: +56 322654278.
E-mail address: adrian.rojo@usm.cl (A. Rojo).

0892-6875/$ - see front matter © 2013 Elsevier Ltd. All rights reserved.
http://dx.doi.org/10.1016/j.mineng.2013.09.004

Geochemical reactions in the soil pores can enhance or retard the process; precipitation/dissolution, sorption, redox and complexation reactions are all dependent on the ph. In this case, the predominant acid front assists in desorption and dissolution of metals, while the base front assists in immobilisation and precipitation of metal hydroxides. Innovative methods are required to enhance the EKR technique to avoid the immobilisation of metals and to reduce the high electrical energy consumption. Due to the large amount of accumulated tailings from copper mining in Chile, the energy expenditure could become a major issue restricting widespread field application of the EKR technology.

The main goal of this work is to evaluate the technique of applying a pulsed sinusoidal electric field to enhance EKR of mining waste from the copper industry. This investigation is part of the search for a remediation technique to environmentally stabilise the large amount of solid waste generated by the Chilean copper industry (Government of Chile, 2011; Minería Chilena Magazine, 2010).

2. Experimental details

2.1. Experimental synthetic mine tailings

The synthetic mine tailings used for the EKR experiments were prepared using dry sand (<200 μm), copper concentrate (chalcopyrite) and copper sulphate pentahydrate. Based on data from Minera Los Pelambres, the synthetic sample was adjusted to 820 mg/kg of total copper in the tailings, with 45% soluble copper (Antofagasta Minerals, 2012).

2.2. Analytical methods

The total and soluble copper contents were determined according to the following methods. All analyses were performed in triplicate and the results averaged.

The total copper content of the synthetic tailings was determined by adding 20 mL 1:1 HNO_3 to 1.0 g of dry material and treating the sample in an 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 atomic absorption spectrometry (AAS) in flame. This determination was performed for the original and final tailings, before and after EKR treatment.

The soluble copper content of the synthetic tailings was determined by adding 50 mL H_2SO_4 5% (v/v) to 5.0 g of dry material and stirring the sample in a 250 mL Erlenmeyer flask for 30 min. The liquid was separated from the solid particles by vacuum through a 0.45 μm filter and diluted to 100.0 mL by adding 10 mL concentrated HCl and distilled water. The metal content was determined by Flame Atomic Absorption Spectrometry (FAAS). This determination was performed for the original tailings, before EKR treatment, because the soluble copper in the final tailings was determined by mass balance assuming that only the soluble copper was removed.

The effectiveness of the EKR process applied to soils contaminated with heavy metals such as copper requires a prior geochemical dissolution of the metal in the pore solution. In the case of tailings from copper mining mineral processing, the copper content at the time of disposal is present as soluble or insoluble species in the acidic environment (pore solution) where the remediation occurs. Therefore, only soluble copper is involved in the remedial action or removal process. During remediation under acidic conditions with an electric field, only dissolution and transport of soluble copper occur, while the insoluble copper remains fixed in the tailings. Because insoluble copper remains fixed in all three zones of the cell, the differences in the total copper content are concluded to represent the soluble copper removed.

2.3. Experimental EKR cell

A schematic description of the remediation cell is given in Fig. 1. Experiments were carried out in an opened acrylic 30 cm × 10 cm × 10 cm high rectangular box divided into three compartments. The length of the central compartment was 20 cm, with two electrode compartments with lengths of 5 cm each. The power supply, which combined the AC–DC voltages, was connected to the cell by two titanium rods immersed in the electrode compartments. Both nylon mesh (149 μm) and filter paper (grade 131) were used to separate the central compartment from the electrode compartments. The pre-treated synthetic mine tailings were placed in the central compartment. The initial electrolyte in the anode compartment was distilled water, while the initial electrolyte in the cathode compartment was distilled water with pH adjusted to between 2 and 4 using a dilute (1 M) sulphuric acid solution. To control pH in the cathode compartment, a sample was taken each day for pH monitoring, and a continuous drop addition of concentrated acid to maintain pH below 4 was supplied.

After the experiments were carried out (Hansen et al., 2005), the synthetic mine tailing sample was segmented into three slices of equal size, and the copper concentration was measured. Because of the sand in the synthetic sample, percolation of the pore solution occurred; therefore, each slice was split into a top and bottom part to better assess copper removal. In this work, the anode slice was defined as the zone closest to the anode, the centre slice was defined as the zone in the middle, and the cathode slice was defined as the zone closest to the cathode.

2.4. Experimental EKR plan

Nine EKR experiments, each with a remediation time of 7 days, were carried out with the conditions given in Table 1. In all experiments, a sample of approximately 1.5 kg (solid dry weight) synthetic mine tailings was adjusted to an initial humidity of 20% using sulphuric acid solution. The addition of acid is needed to ensure dissolution of the copper and its subsequent removal by EKR process. This process is possible if the pH of the tailings is below 4.

According to previous work (Hansen and Rojo, 2007; Rojo et al., 2010, 2011, 2012), a pulsed sinusoidal electric field produces good results if a polarity reversal of the electrodes occurs. In the case of pulses (Sun et al., 2012), the ratio of the times for which the electric field is ON and OFF is 25 (t_{ON}/t_{OFF} = 2500/100). As shown in Table 1, in the sinusoidal EKR experiments with a negative $V_{minimum}$, a polarity reversal of the electrodes occurs periodically.

The experimental plan considered a conventional reference EKR configuration with 20 V DC, and eight sinusoidal pulsed electric field (DC + AC) EKR configurations with effective voltages of approximately 20 V (14.6–26.7 V). Experiments were defined based on the partial results that were initially obtained under fixed conditions with an effective voltage of 14.6 V (10 V DC, 15 V AC) and a pulse time ratio of 25 (t_{ON}/t_{OFF} = 2500/100, t in second). In experiments 2, 3, 4 and 7, the effect of an AC frequency in the range of 50–2000 Hz was analysed. Then, to confirm the effect of AC frequency, experiments 5 and 8 were performed with an effective voltage of 26.7 V, pulse time ratio 25 and frequencies of 1000 and 2000 Hz, respectively.

The effect of electrode polarity reversal and the effective voltage was verified in experiment 6 with an effective voltage of 22.6 V and a pulse ratio 25 was performed. Finally, because the results of experiment 4 were the most attractive, a similar experiment 9