



Electrokinetic Treatment Of Mercury-Polluted Soil Facilitated By Ethylenediaminetetraacetic Acid Coupled With A Reactor With A Permeable Reactive Barrier Of Iron To Recover Mercury (II) From Water



I. Robles^a, M.J. Lozano^a, S. Solís^b, G. Hernández^b, M.V. Paz^a, M.G. Olvera^a, E. Bustos^{a,1,*}

^a Centro de investigación y Desarrollo Tecnológico en Electroquímica S. C., Parque Tecnológico Querétaro, Sanfandila, Pedro Escobedo, 76703 Querétaro, Mexico

^b Centro de Geociencias, Universidad Nacional Autónoma de México, Campus Juriquilla, Boulevard Juriquilla 3001, Querétaro 76230, Mexico

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ABSTRACT

A soil sample polluted with mercury was treated with electrokinetic treatment (EKT). Seventy-two h was necessary to remove most of the mercury present in soil from the cathode to the anode with a removal percentage of 76.30%. During this period, we measured the interfacial potential and current. We analyzed the physicochemical properties before and after EKT where physical properties such as color, soil particle, real density, porosity and fraction of organic matter did not change; chemical properties such as pH and electric conductivity changed due to the electric field applied to the soil and the addition of ethylenediaminetetraacetic acid as a facilitating agent. Finally, the remaining solution obtained from the EKT was treated in a reactor with a permeable reactive barrier of Fe⁰, where 84.47% of the Hg²⁺ was recovered from water.

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1. INTRODUCTION

Soil is a complex matrix in which a significant number of processes are carried out for the development of plants and organisms. One of the biggest problems in soils is the accumulation of metals [1]. Metals present in soils are distributed through the different soil fractions (organic and inorganic) and metals can attached in different ways: dissolved in the soil solution, occupying exchange sites on inorganic soil constituents, adsorbed in specific inorganic components, associated with insoluble soil organic matter, precipitated, and present in the structure of some minerals [2–4]. Mercury is a non-essential metal that can be easily accumulated in plants and living organisms. Mercury can cause different toxic effects in humans, affecting mainly the nervous system [5–7]. Soil is an important receptor of mercury, where it can be adsorbed in the different ways outlined above.

Electroremediation or electrokinetic treatment (EKT) has been successfully applied in a variety of polluted soils; this technology refers to the application of an electric field or direct current through a pair of electrodes (anode and cathode). These

electrodes are inserted in the soil, where an electrolyte improves the conductive properties of the electric field [8,9]. Enhancing solutions can be added to the soil to improve the efficacy of the treatments [10–16]. The most common enhancing solutions are ethylenediaminetetraacetic acid (EDTA), KI, and NaCl, among others [17,18]. EDTA is a compound with four carboxylic and two associated amino groups, which can act as electron pair donors or Lewis bases. EDTA, in an aqueous solution, forms an octahedral complex with mostly divalent metal cations (M²⁺). A very strong mercury-EDTA ([Hg-EDTA]²⁻) complex is formed, with a log β of 21.8 [19]. Considering the negative charge of this complex, most mercury moves towards the anode side, where it can be recovered.

Reports in the literature note the use of EDTA to improve the electrokinetic remediation of Pb, Zn, Cd, and Cr, with efficiencies from 20–100% at laboratory scale. There are no available reports about the extraction of mercury using this treatment [20–28]. Once the [Hg-EDTA]²⁻ is transported to the anode, which can be removed from the water coupling, a second treatment is possible. Literature reports indicate that adsorption, permeable reactive barriers (PRB), and electrocoagulation are effective treatments [29–32] with efficiencies higher than 90% for recovering heavy metals such as Pb and Cr. In this sense, the EKT of mercury-polluted soil facilitated by EDTA could be coupled to a second treatment to recover Hg²⁺ from water.

* Corresponding author. Tel.: +52 442 2 11 60 59; fax: +52 442 2 11 60 01.
E-mail address: ebustos@cideteq.mx (E. Bustos).

¹ ISE member.

The novelty of this paper is that we combine the EKT of mercury-polluted soil facilitated by EDTA as a complexing agent and the ensuing removal of mercury from water using PRB of Fe⁰.

2. EXPERIMENTAL

2.1. Reagents

Mercury-polluted soil was taken from a mine site located in San Joaquín, Queretaro, Mexico with an initial concentration of 656.27 mg·Kg⁻¹. The soil sample was mixed before EKT [9]. EDTA, disodium salt dehydrate from J. T. Baker brand was used to enhance the EKT. After EKT, a PRB of Fe⁰ obtained from Sigma Aldrich was used to recover the mercury.

2.2. Methods

For the EKT, an acrylic rectangular cell 64 cm³ in volume and Ti rectangular plate electrodes (2 × 5.5 cm, Tepsa, Mexico City) were used as the anode and cathode, respectively. Their separation was 6 cm. Twenty volts was applied as the potential of the cell during EKT to 25 g of polluted soil using a DC Power Supply GP 4303DU (EZ Digital Company, Korea). The experiments were carried out at room temperature. During EKT, the current and corresponding interfacial potential were measured at each electrode.

Mercury was quantified in the extracted solution on the electrode side, and also measured in the soil before and after treatment. To this end, the soil was divided into six equal-sized parts 1 cm away from the cathode to the anode (Fig. 1).

We measured mercury in an aqueous solution using anodic stripping voltammetry (ASV), and we also corroborated our results using atomic absorption (AA). ASV was measured in an electrochemical cell with three electrodes: vitreous carbon, Ag|AgCl, and a Pt wire. These electrodes function as working, reference, and counter electrodes, respectively. A pre-concentration potential of -0.6 V, a deposition time of 5 min, and a rest period of 40 s were applied before a scan potential of 20 mV·s⁻¹. A Potentiostat - Galvanostat Bas - Epsilon (Bioanalytical Instruments, USA) was used for this purpose. To quantify the mercury content using AA, the samples were pre-treated with microwave digestion using 5 mL of 2% HNO₃ solution; deionized water was added to each sample to obtain a final volume of 50 mL of solution. These liquid samples were digested using a MarsX microwave according to protocol 3015A-EPA-2007 [33]. In the case of solid samples, 0.5 g of soil was digested according to protocol NOM-004-SEMARNAT-2002 [25]. At the end of the digestion procedure, deionized water was added to the supernatant of each sample to obtain 100 mL of solution; for

the analysis of these samples, an AA Aa400 Perking Elmer coupled to FIAS 100 equipment was used. We employed a hydride generator technique via cold vapor following the methodology reported in protocol NOM-004-SEMARNAT-2002 [34].

We conducted a physicochemical characterization of the soil before and after EKT. pH, electric conductivity, color, size particle, real density, porosity, and fraction of organic matter were compared. pH and electric conductivity were estimated according to procedures NMX-AA-008-SCFI-2000 [35] and NMX-AA-093-SCFI-2000 [36], respectively, using a Hanna pH-meter (Hanna Instruments, USA) and a YSI-3200 conductimeter (Cole-Parmer, USA). Color, size particle, real density, porosity, and fraction of organic matter were analyzed according to NOM-021-SEMARNAT-2000 [37] and ISRIC [38] protocols. All soil analyses were conducted in duplicate.

The electrodes were also characterized before and after EKT using scanning electron microscopy (SEM). SEM was conducted using 15 kV of voltage acceleration and 100x with a Jeol JSM-5400LV scanning electronic microscope (JEOL, USA). After EKT, the extracted solution with [Hg-EDTA]²⁻ was treated to recover Hg using a flux reactor with a PRB of Fe⁰ [39], where 18 g of iron was needed to treat 250 mL of Hg solution. We also used a flux of 120 mL·min⁻¹, using a MasterFlex peristaltic pump (Cole-Parmer, USA). The sample was collected and analyzed using ASV.

3. RESULTS AND DISCUSSION

After 72 h, the mercury in the cathodic side was negligible, which indicates that the [Hg-EDTA]²⁻ complex was formed and transported to the anodic side. The current (Fig. 2A) should be proportional to the mercury concentration close to the anode, as can be seen in Fig. 2A. Seventy-two h was sufficient to remove most of the mercury present in the soil. After that time, the remaining mercury is removed too slowly due to there being specific soil fractions where mercury bonds very strongly. These soil types include exchangeables, clays, oxides, sulfurs and residual fractions [40], which is why 72 h is sufficient to remove mercury electrochemically. pH during EKT varied between 4 and 5 (Fig. 2B) due to the physicochemical composition of the soil and the addition of EDTA. The electric conductivity changed and increased from 10 to 15 mS·cm⁻¹ in the presence of the [Hg-EDTA]²⁻ complex generated during EKT (Fig. 2C).

During the 72-h EKT, an average current of approximately 0.15 A was observed. The interfacial potential was measured on both sides of the electrodes. The potential was -1.42 V on the cathodic side and 1.41 V on the anodic side, although the latter slowly increased to attain a value of 2.75 V. This increment can be attributed to the reaction of mercury movement towards the anode compartment, such as the [Hg-EDTA]²⁻ complex.

After the EKT, we measured the mercury content using ASV. These results were corroborated by AA on the anodic side and the different soil portions between both electrodes. A mercury recovery rate of 76.30% was obtained from the soil. We measured the remaining mercury in the soil to the anode to determine how mercury was moving towards the anode. Table 1 lists the percentage of mercury remaining in the soil fractions; one can clearly see that a larger fraction of mercury is present near the anodic side because the [Hg-EDTA]²⁻ complex moves towards the anode. A percentage of 2.8% was recovered close to the cathode and 4.8% close to the anode. This trend can be described by a logarithmic increase represented by the equation: $Hg = 1.161 \ln(d) + 2.677$ with $R^2 = 0.981$, where Hg is given in percent and d represents the distance from the cathode to the anode. This equation was obtained from these data based on the different transport phenomena generated during EKT [9–17]. Our results accordingly explain why more mercury remains on the anodic side.

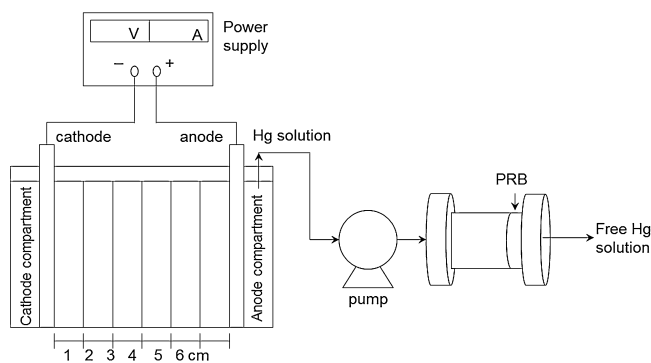


Fig. 1. Rectangular system employed to carry out the EKT with an acrylic electrochemical cell, including a Ti cathode and anode separated by 6 cm. The solution containing mercury was removed from the anodic compartment with a pump to an acrylic cylindrical cell containing a PRB of Fe⁰ to remove the mercury from the solution.

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