



Removal of Silver (Ag) from Artificially Contaminated Soil Using Acetic Acid as Electrolyte with Electrokinetic Remediation



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Abstract

The electrokinetic remediation of artificially silver (Ag) contaminated soils was evaluated. In this study, the soil sample was taken from locations A and C near silver handicraft workshop assuming has similar natural geochemistry with silver contaminated soil. While control soil was taken from locations B and D at Kotagede, Yogyakarta, Indonesia. All soil samples were contaminated with a silver (100 mg AgNO₃/Kg soil) and they were treated in six cycles of saturation process using de-ionized water and air-dried before being aged for one year and subjected to electrokinetic (EK) remediation treatment. The physical and chemical properties of soil were measured using flame atomic absorption spectrophotometric. Soil components such as organic matter, total nitrogen, CEC, soil texture, and soil buffer capacity were also evaluated. Desorption of silver with different acetic acid concentrations (i.e 0.1, 0.5, 1.0 and 1.5 M) was also studied in a batch system. Electrokinetic remediation treatment was conducted by a graphite electrode in a constant voltage of 1 Vcm⁻¹ for 7 d. The current flow during EK remediation treatment was recorded by data logger for every 10 mins. The results showed that high removal of silver from soil was achieved when using 1.0 M acetic acid as the electrolyte. In this regard, the EK remediation treatment could achieve high silver removal from soil A (92 to 93%) and soil C (82 to 92%) compared with soil B (75 to 96%) and soil D (26 to 74%), respectively.

Keywords: Acetic acid; Electrokinetic remediation; Silver (Ag); Soil

1. Introduction

There is concern about trace metals in natural water and soils which have generated in the aqueous transportation, retention in the water column and sediment burial rates. The mobility of metals in the soil environment had controlled by adsorption and ion-exchange which helps in predicting the migration of these metals in the contaminated environment and for evaluating the feasibility of a contaminated soil for the disposal or recovery [1].

Silver can enter the environment in a variety of chemical forms or compounds, depending on the waste discharged from the fabrication, use, and

disposal of those various products. Silver concentration in the soil is reported as much as < 0.50 µg/g, while the concentration limit in the freshwater is 0.20 µg/L and seawater is 0.25 µg/L [2]. The concentration of available forms of silver in soils is strongly controlled by various physical and chemical processes such as the prevailing pH and redox conditions, and interactions with soil organic matter [1]. Infield soils, silver tends to accumulate in the surface, organic-rich horizons. When silver accumulated in the soil surface layers, it was persistent, and the leaching was slow [3-4]. According to the Indonesian Government Regulation,

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No. 85/1999 on the quality standard of Toxicity Characteristic Leaching Procedure (TCLP) for pollutants in wastewater, the silver concentration limit is 5.0 mg/L. Therefore, above the results, the low concentration of silver must be presented in the environment.

Some jewellery and silver tableware manufacturers operate electroplating processes. The metal is normally plated from alkaline cyanide solutions that usually generate metallic cyanide, such as silver dicyanide $[\text{Ag}(\text{CN})_2^-]$ as listed hazardous waste under the Resource Conservation and Recovery Act (RCRA) in the US [5]. Silver dicyanide may be accompanied by other silver species and is a chemically stable and extremely toxic metal complex of high mobility in the environment [6]. Since the metal has been controlled under the RCRA, the environmental exposure should be minimal.

Kotagede is a special region in the Yogyakarta province of Indonesia whose silver handicraft is produced for souvenirs and jewellery. Other than this area, a similar product is also produced in Indonesia at Koto Gadang (West Sumatera), Bangil (East Java) and Celuk (Bali). Consequently, activities in those areas have increased the volume of a waste stream into the environment from silver handicraft production. Therefore, the decontamination of silver contaminated sites is one of the most important technological challenges. Removal of silver from contaminated water has been reported by physical and chemical methods such as using precipitator reagents, e.g. FeCl_2 , $\text{Ca}(\text{OH})_2$ and NaOH [7], modified chitosan resin [8], clay absorbent [9-10], concrete particle [11] and phytoremediation [12-13], cross-flow microfiltration [14]. However, for silver contaminated soil, maybe there is no effective method other than dig and dump for ex-situ system as it is commonly used to remove heavy metal from contaminated soil [15].

Previously studies have demonstrated that the EK remediation had been used in the removal of heavy metal from electroplating contaminated soil in a laboratory scale [16] and field soil [17]. EK remediation is one of the promising soil decontaminations that could be used to separate and extract heavy metals, and organic contaminants from low permeability clay soil, muddy ground, sludges, sediments, and groundwater [18]. Electrokinetic remediation has high removal efficiency and time-effectiveness and can be done in situ process by applying a low direct current through the electrodes that placed in the clay soil to remove the

contaminant. When the electric field is applied on the clay soil, the electrolysis reactions are induced at the electrodes and electrokinetic transport phenomena (i.e. electroosmosis, electromigration, and electrophoresis) occurred in the porous medium to remove non-polar organics as well as ionic contaminants [19]. In general, the electrode reactions can be considered to produce O_2 gas and H^+ ions in solution at the anode and H_2 gas and OH^- ions in solution at the cathode (see the Eq-4 and Eq-5). In addition, H^+ ions travel at a velocity of 1.8 times faster than OH^- ions to enhance the desorption of heavy metals from the soil [20]. Hydroxide ions (OH^-) in the soil significantly induced a forming of metal hydroxide which declined the mobility of the metal ions. Therefore, the soil pH increased more alkaline from anode to the cathode area [21-22].

Importantly, so far the EK remediation process had not been tried to remove silver from contaminated soil. This study was to investigate the enhancement removal of silver from the soil by EK remediation treatment through the acetic acid as a flushing electrolyte solution. Artificially silver contaminated soil from four different locations at Kotagede of Yogyakarta province, Indonesia was prepared as a model of soil in this EK remediation treatment. Laboratory scale horizontal soil column studies are used according to previously reported [23] in EK remediation for a while of the only 7d, allowing for a comparison of silver removal efficiency. The physicochemical of soil is also evaluated.

2. Experimental Section

2.1. Soil sampling and characterization

Clean soil samples were collected from four different locations e.g. soils A, B, C and D at Kotagede district in Yogyakarta Province, Indonesia. The soil at locations A and C were collected near silver handicraft workshop assuming has similar natural geochemistry with silver contaminated soil. While soil at locations B and D were collected far from silver handicraft workshops and used as control soil. The sampling location of soil samples was shown in [Figure 1](#).

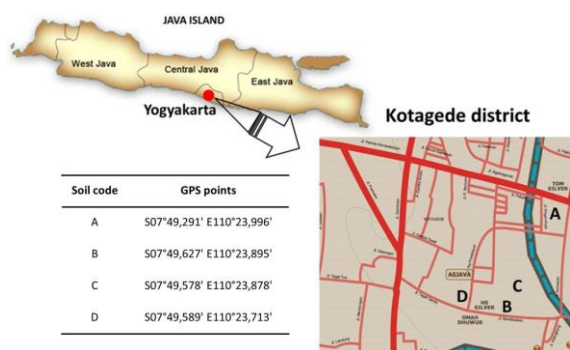


Figure 1. Sampling location of soil samples at Kotagede, Yogyakarta Province, Indonesia

All soil samples were artificially amended by 100 mg/Kg silver concentration as AgNO_3 salts. Soil samples were prepared and characterized according to previously reported paper [23]. Briefly, clean soils were contaminated silver and then they treated with six cycles of saturation treatment using de-ionized water and air-dried before being aged for one year. The soil pH was measured in water suspensions by a ratio of 1:10 (mass/volume) after for 4 h equilibration using a pH meter (Mettler-Toledo FE 20, Switzerland) as described in the paper [24]. Acid-base titration was used to determine the soil buffer capacity as described in the paper [25]. About 1.0 g of dry soil was digested overnight with 10 mL of HNO_3 and then the supernatant was measured for total metal concentration by Flame-AAS (Perkin-Elmer, PinAAcle 900T, USA). The surface area and pore volume of soil were measured by nitrogen adsorption (SSA-N_2 at 77 K) methods using a Bel Sorp mini (Bel, Japan). The SSA-N_2 was calculated using the Brunauer-Emmet-Teller (BET) equation [26].

2.2. Silver (Ag) desorption

Different concentrations of acetic acid were used to assay the ability of electrolyte on silver desorption to enhance the EK remediation treatment. About 0.5 g of silver contaminated soil was weighed into a polycarbonate tube containing 40 mL of 0.1, 0.5, 1.0 or 1.5 M CH_3COOH . The suspensions were shaken for 24 h and then filtrated using Whatman No.42 prior silver concentration measurement using Flame-AAS (Perkin-Elmer, PinAAcle 900T, USA) at λ 328.1 nm.

2.3. Electrokinetic remediation (EKR) treatment

Bench-Scale electrokinetic remediation was conducted using a clear acrylic box oriented horizontally according to previously reported paper [23] as shown in Figure 2. Graphite was used for the cathode and anode, respectively. In this study, 1.0 M CH_3COOH was used throughout the experimental works as a purging solution since organic acid has been determined to be adequate with the aim of removal silver from the soil in terms of eco-friendly and green process. The potential gradient was applied for 7 d experimental at a constant voltage of 1.0 V/cm. Electric currents were recorded for every 10 mins. during the evaluation periods using midi logger (GL200A, Graphtec USA).

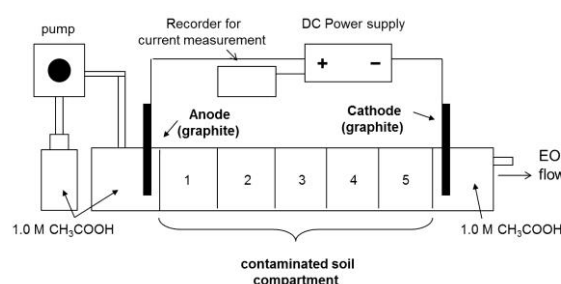


Figure 2. Electrokinetic remediation cell set-up used in this study

At the end of the experiment, power supply and electrodes were carefully disconnected. Treated soil was extruded from the compartment and sectioned into five parts. Soils were dried overnight in the ambient air and then dried at 110 °C for 24 h in the oven to obtain a dry soil sample. About 0.5 g of dried soil from each section was extracted by 10 mL of concentrated nitric acid for 24 h and the aliquot further heated until \pm 5 mL volume. The solution was then cooled and filtered (Whatman No. 42). The filtrate was then transferred into a 25 mL volumetric flask and diluted with deionized water to the mark. Remaining silver concentration in the soil after EKR treatment was determined by Flame-AAS (Perkin-Elmer, PinAAcle 900T, USA) at λ 328.1 nm. Eq-1 shows the calculation of silver removal efficiency from the soil.

$$2.4. \text{Removal efficiency (\%)} = (C_0 - C_t / C_0) \times 100\% \quad (1)$$

Where C_0 was initially concentration of Ag in soil (100 mg/Kg) and C_t was the remaining Ag concentration in the soil after the EK remediation treatment [27].

3. Results and Discussion

3.1. Soil characteristics

All soil samples have the same texture as sandy loam, and they all have high CEC values and organic matter content. Therefore, the soil has a high permanent charge due to the variable charge components [28]. The quantity of buffering capacity of the soil samples was represented by plotting the $d\log V\text{NaOH}_{\text{added}}$ and the $d\log V\text{HCl}_{\text{added}}$ versus $d\text{pH}$. The slopes of the curves indicated that the larger the slopes are, the smaller the buffer capacity was [25]. The calculated slopes showed that soil at locations A and C have low buffering capacity against acid causes the soil pH changed slightly when acid was added to (e.g. $d(\log V\text{HCl}_{\text{added}})/d\text{pH} = -0.422$ and -0.423), but soil at locations B and D have low buffering capacity against the base (e.g. $d(\log V\text{NaOH}_{\text{added}})/d\text{pH} = -0.479$ and -0.382). Therefore, it seems that both soils at locations A and C relatively easy to acidify than that soil B and D. Beside, high amounts of heavy metals are retained in soil when the buffer capacity is high enough to resist a change under the alkaline condition [29]. The soil properties were summarized as shown in [Table 1](#).

3.2. Profiles of silver (Ag) desorption

For a selection of proper electrolytes in EK remediation, the toxicity, buffering range, and economic effects of electrolyte are the main criteria. [Figure 3](#) shows the profiles of Ag desorption concentration carried out using acetic acid (CH_3COOH). Generally, for all soil samples the desorption of silver concentration from soil increases with the order of acid concentration in the sequence of $1.0 > 1.5 > 0.5 > 0.1$ M. Therefore, the highest silver desorption from the soil as much as 38.98 to 44.48 mg/Kg was achieved for 1.0 M acetic acid concentration. These phenomena can be explained that organic acids like acetic acid are milder leaching agents that cannot desorb as much as macro-minerals because the macro-minerals have a smaller hydrated radius and are therefore more tightly bound to soil particles [30]. In addition, as a weak acid, acetic acid was not as effective as strong acid like nitric acid, but it was biodegradable and lowers soil pH to some extent without causing secondary pollution to groundwater [31]. Acetic acid also partially dissociates in aqueous solution into the acetate ion (CH_3COO^-) and a proton (H^+), so that Ag^+ ions in the soil bond to acetate ions formed CH_3COOAg as shown the overall reaction in Eq-2 and Eq-3. Therefore, in this study, 1.0 M acetic acid was applied in all the EK remediation of silver contaminated soil.

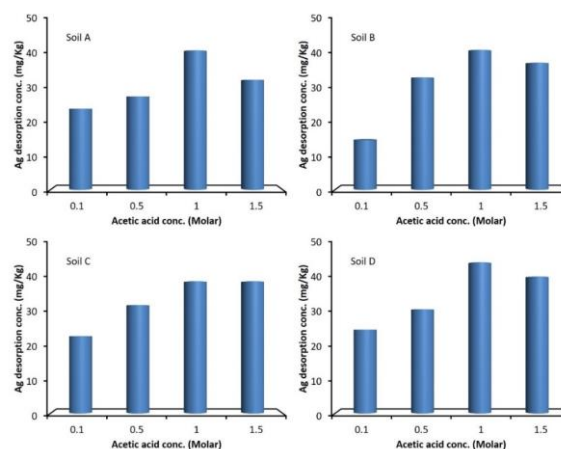
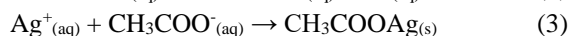


Figure 3. Desorption concentration of silver from soils A, B, C, and D using acetic acid as an electrolyte. The initial concentration of silver was 100 mg/Kg.

There are several advantages in using acetic acid to depolarize the hydroxyl ions generated by the cathodic electrolytic reduction processes: (i) most metal acetates are highly soluble; (ii) the concentration of ions generated by dissociation of the acid is very low due to the low pKa value of acetic acid, and thus the electrical conductivity of the soil was not increased drastically; (iii) it was environmentally safe and biodegradable; and (iv) acetate ions will prevent the formation of other insoluble salts in the vicinity of the cathode, thus preventing the development of a low electrical conductivity zone and dissipation of excess energy in the soil near the cathode [32].

3.3. Removal of silver (Ag) by EK remediation

3.3.1 Effect of soil pH

Electrolyte composition changes during EK remediation when dissolved substances get removed from the soil sample either by entering the catholyte/anolyte or by depositing on the cathode in the case for many metal ions [22]. The electrolysis of pore water was a predominant process in moist soils, which results in the formation of H^+ ions and oxygen at the anode and OH^- ions and hydrogen at the cathode as shown in Eq-4 and Eq-5. Hindered migration would result in the local change in pH of the pore fluid and therefore acidity increases with time at the anode and alkalinity increased at the cathode. [Figure 4](#) shows the changes in soil pHs under EK remediation treatment.

All soil samples had a neutral initial pH 7.08 to 7.59 but changed as a result of water hydrolysis as

shown in Eq-4 and Eq-5, soil pH increased with a distance from anode to cathode in the EK remediation cell. For each soil, the final soil pHs changed after EK remediation with varied pHs in each soil from 4.07 to 6.30, 4.58 to 6.88, 4.04 to 6.04 and 4.34 to 6.35, respectively for soils A, B, C, and D. Those profiles were in the pH range of acid to mild acid which was a suitable pH on the enhancement of metal ions desorption from the soil into pore solution. The results were in accordance with the findings reported

in another study [22]. The H^+ ions were transported towards the cathode by electromigration and electroosmosis, while OH^- was moved towards the anode. However, H^+ travels at velocity 1.8 times faster than OH^- [33]. Whereas H^+ enhanced desorption of heavy metals, OH^- significantly declined the mobility of the metal ions because of $Me(OH)$ precipitation [21].

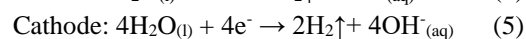
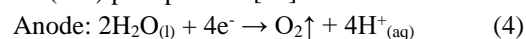


Table 1. Characteristics of soils used in the study

Parameters	Soil code			
	A	B	C	D
Particle size distribution (%) :				
Sand	66.43	64.35	51.84	68.16
Silt	23.09	24.35	36.05	23.68
Clay	10.48	11.31	12.11	8.16
Soil texture	Sandy loam	Sandy loam	Sandy loam	Sandy loam
Organic C (%)	0.83	1.64	1.47	4.27
Organic matter (%)	1.43	2.83	2.54	4.27
Total N (%)	0.09	0.05	0.11	0.14
CEC (meq/100 g)	12.35	16.83	18.06	16.95
Surface area (N_2 -BET), m^2/g	25.672	35.095	40.688	28.456
Pore volume, m^3/g	52.372	64.015	64.109	50.168
pH (1:20, w/v)	7.7	7.8	6.9	7.4
Buffer capacity :				
$d(\log V_{HCl\text{added}})/dpH$	-0.422	-0.310	-0.423	-0.367
$d(\log V_{NaOH\text{added}})/dpH$	-0.594	-0.479	-0.391	-0.382
Heavy metals (mg/Kg):				
Ag	NA	NA	NA	NA
Pb	0.25	0.34	0.37	0.22
Cu	1.01	4.88	18.56	2.09
Cr	NA	NA	0.37	NA
Ca	7.91	15.80	11.02	10.60

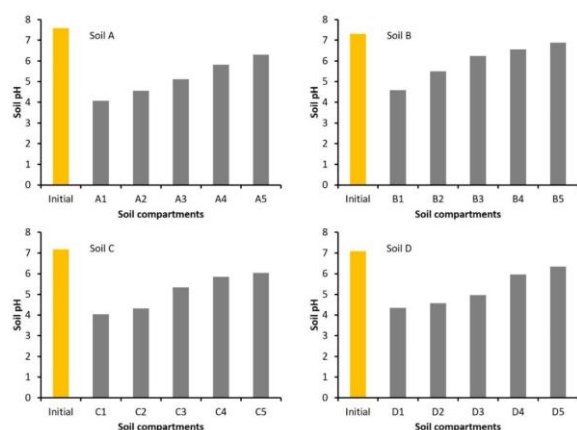


Figure 4. Profiles of pHs in soils A, B, C, and D after EK remediation treatment using 1.0 M acetic acid as an electrolyte.

3.3.2. Profiles of silver (Ag) concentrations

Electrochemical treatment of contaminated subsurface was probably one of the most promising in situ soil decontamination technologies capable of removing organic and inorganic contaminants and radionuclides. By using a low-level direct current (DC) electrical potential difference across electrode pairs placed in-ground, contaminants in the soil are moved under the action of the electrical field. The extraction of contaminants by the EK remediation assumed that the metal ion was in the liquid phase on the soil pores [34]. The variation in the amount of remaining silver concentration after EK remediation in the soil cell for each test was shown in [Figure 5](#).

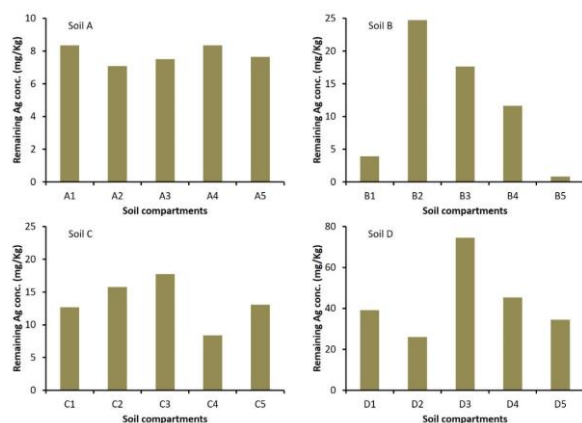


Figure 5. Profiles of remaining silver concentrations in soils A, B, C, and D after EK remediation treatment using 1.0 M acetic acid as an electrolyte. The initial concentration of silver in the soil was 100 mg/Kg.

Generally, for all soil samples, the retained concentration of silver was lower than the initial concentration (100 mg/Kg) with a varied concentration in each soil from 7.08 to 8.03 mg/Kg, 3.90 to 24.72, 8.36 to 17.25 mg/Kg, and 25.99 to

74.49 mg/Kg respectively for soils A, B, C and D. Based on these results, it can be concluded that higher removal efficiency of silver from soil has achieved as much as in the soil A (92 to 93%) and soil C (82 to 92%) than that in soil B (75 to 96%) and soil D (26 to 74%) after the EK remediation. Low remaining of silver concentration in the soil compartments for soil A and C have indicated that the soil has low acid buffering capacity so that acetic acid as electrolyte easy to promote the removal of silver ions by lowering the soil pH (i.e. ~ 4 to 6 as shown in [Figure 4](#)). Acid front during the EK remediation was produced by electrolysis of water at the anode compartment (Eq-4) and then migrated toward the cathode along with the desorption of silver ion into pore solution, enabling silver transport in the soil by electromigration and electroosmosis. These processes were repeated in the soil cell and the amount of silver concentration appear to be gradually decreased toward the cathode in the electric field as the time went on. However, high remaining silver concentration after the third compartment in the soil B and D (i.e. compartment B3 and D3 as shown in [Figure 5](#)) had attributed to the low buffering capacity of the soil against base due to migration of the OH^- ions generated at the cathode (Eq-5) into soil toward the anode. In addition, the pH junction had occurred when acidic and alkaline fronts meet in the soil and lead the precipitation of metal hydroxide to decrease the efficiency of silver removal from soil. Similar results for EK remediation on the heavy metal contaminated soil have been previously reported [35]. A high content of organic carbon and organic matter in the soils B and D affect the stronger bond formed of silver with the soil so that metal ion migration was impeded by its sorption onto the soil. Besides, soils A and C where easily acidified since they have a low buffer capacity against acid as shown in [Table 1](#). Those circumstances greatly influenced the variation of silver concentration in the compartment after the EK remediation.

3.3.3. Current profiles

In this study, the constant voltage was applied between the electrodes. The current profiles throughout the EK remediation treatment on the soil were shown in [Figure 6](#). From these figures, it was revealed that the variance of current profiles has significantly correlated to the number of ions in the pore fluid. Generally, for all experiments current increased from initial time to maximum intensity at

the first of two days process, suggesting that more ions migrated through pore fluid in the soil. Furthermore, when the soil resistance increased the current densities in the soil decreased at the 3rd to 5th day and then remaining in the constant current until the last of the 7th day. The occurrence of a drastically lower current plateau in the soil A than other soil has reflected high silver ions were removed from the soil. This observation was also in agreement with another paper [36].

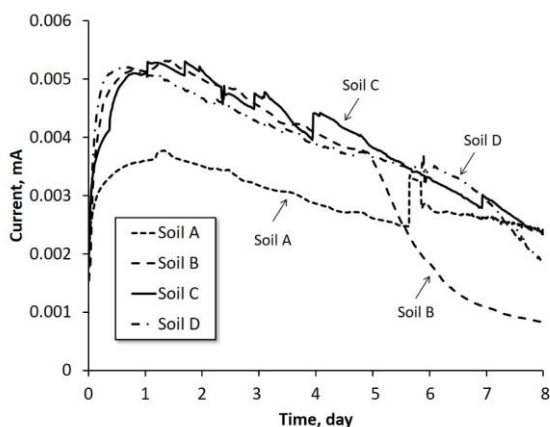


Figure 6. Profiles of current flow in soils A, B, C, and D during the EK remediation using 1.0 M acetic acid as an electrolyte.

The heavy metal content in the soil as shown in Table 1 (e.g Ca, Pb, Cu, and Cr) had precipitated as metal hydroxide in the soil matrix and at the surface of the cathode electrode as shown in Figure 7 due to production of hydroxide ion at the cathode. A similar result has also been reported by another study [22].

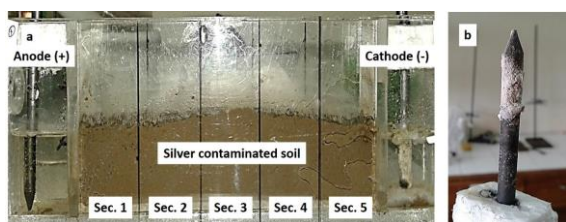


Figure 7. EK remediation cell with anode and cathode electrodes. Graphite electrode in the anolyte catholyte well (a), and the surface of the graphite electrode at cathode well was covered by white solid precipitation (b). The soil compartment was sectioned as S1 to S5 that is normal distance from anode to cathode.

This clogging of a precipitate could increase the electric resistance in the soil which showed by the decreasing of a current plateau. In addition, resistance at the interface between electrodes and electrolyte might increase because of concentration polarization and water dissociation, and ions with positive or negative charges move to the two ends of

the electrode cell, which results in the drop of ionic strength in soils and the current as well [37].

4. Conclusions

The main conclusions obtained from this study are as follow:

1. The application of electric across the silver contaminated soil, causes silver ions migrated from anode to cathode in the EK remediation cell, accompanied by marked changes of silver concentration in the normal distance from anode to cathode in the soil compartment.
2. Regarding the removal of silver ions from the soil, 1.0 M of acetic acid (CH_3COOH) effectively can be used as a flushing electrolyte solution in the EK remediation treatment.
3. High removal of silver has achieved from the soils A and C respectively as much as 92 to 93% and 82 to 92% than that compared with the soils B and D respectively as much as 75 to 96% and 26 to 74%.
4. The current profiles showed that silver ions have effectively migration in the soils A and C which indicated by low resistance (i.e. high peak current) in the soil compartment. While the soils B and D have high resistance (i.e. low peak current) in the soil due to high carbon contents (i.e. %C and OM) and low buffer capacity of soil against the base.

5. Conflict of Interest

The authors declare that there is no conflict of interest in any manner concerning the research that gave rise to the data herein discussed and published.

6. Acknowledgements

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