Cost Estimation of Electrokinetic Soil Remediation for Removal of Six Toxic Metals from Contaminated Soil

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ABSTRACT

The purpose of the study is to estimate the operating cost of the electrokinetic methods in the removal of toxic metals (TMs) from granite mine tailing soil with the help of the proposed cost estimation models. The conventional electrokinetic technologies have not considered the cost estimation in the removal of TMs from polluted soils. In this study, we incorporated the chelates such as citric acid and ethylenediaminetetraacetic acid (EDTA) enhanced electrokinetic soil remediation process followed by a cost estimation of the processes. Our study proposed the cost estimation models to determine the operating cost of the conventional and enhanced electrokinetic treatment processes, specifically for the removal of six TMs such as chromium (Cr), cobalt (Co), copper (Cu), nickel (Ni), zinc (Zn) and manganese (Mn) from granite mine tailing soil. We investigated the chelating enhanced electrokinetic removal of TMs about six times more than the conventional process for 20 days of operation. Furthermore, we estimated that the operating cost of the conventional and enhanced electrokinetic processes was about US$ 110 to US$ 508 per cubic meter of treated soil. The total operating cost was about US$ 110 to US$ 1006 per cubic meter of treated soil including enhancers cost. We believe the chelating enhanced electrokinetic treatment of soil was more effective than conventional treatment for removal of TMs from contaminated soil.

INTRODUCTION

Soil environment has been polluted by incorporation of toxic heavy metals and organic pollutants. Improper management and human activities cause soil contamination (Shukla & Chandel 2005). Recent studies reported the toxic metals(TMs) such as chromium (Cr), cobalt (Co), nickel (Ni), copper (Cu), zinc (Zn) and manganese (Mn) at elevated concentrations in granite mining waste (Reddy & Yarrakula 2019). During the mineral and milling process, a vast quantity of granite waste is produced and accumulated at dump yards, which may affect the environment (Santhosh et al. 2019). Recently, star-shaped microfluidic channel techniques have been used to detect the TMs (Satish et al. 2017). The concentrations of TMs assessed with advanced remote sensing technique and geography information technology system (GIS) particularly for groundwater (Asadi et al. 2017, Monica et al. 2018). The environmental impact assessment of TM polluted water and soil was determined to minimize environmental pollution (Koteswara & Kiran 2019). Conventional methods such as biosorption, phytochemical oxidation, coconut coir, saxaul tree ash and nickel oxide/carbon nanotube composites (NiO/CNT) investigated for the removal of TMs and organic pollutants from industrial waste

disposal (Aravind et al. 2017, Pratapa et al. 2019). Most of the conventional remediation studies are ineffective and not feasible on a large scale at the field level. Electrochemical behaviour of different metals and metal alloys studied with sodium chloride solution for environmental applications (Suresh et al. 2018). The solubility of metals and TMs in the soil affected by the chemical composition of the soil as well as groundwater (Sposito 2008).

An electrokinetic soil remediation technology has emerged and successfully been applied for removal of TMs and organic contaminants from industrial wastewater and soil (FAO/WHO 2000). The removal of toxic TMs from clayey soil applied in several field applications by using electric current (Collins & Kinsela 2011). Electrokinetic remediation is an advanced technique used in-situ and ex-situ removal of toxic metals, organic pollutants, and radio-nuclide materials from contaminated soil. Recent studies investigated the reducing agents enhanced electrokinetic remediation for removal of TMs from polluted soil (Reddy et al. 2019). The metal or TM ions would be desorbed under an acidic or low pH environment at the anode during the electrokinetic treatment (Sivapullaiah et al. 2015). Recently, many studies have focussed on zero waste/mitigation of waste/prevention

of waste using reducing and chelating enhanced reagents in electrokinetic soil remediation technologies (Peng & Tian 2010, Liu et al. 2017).

Conventional electrokinetic methods focussed on the removal of TMs from contaminated water and soil, but not considered the cost estimation of the process (Rosestolato et al. 2015). Several electrokinetic soil remediation studies not considered the cost estimation of the process however, each remediation step involved a cost in a real process (Gao et al. 2013, Reddy et al. 2019). We cope with this problem and proposed the operating cost estimation models soil remediation processes particularly in the electrokinetic removal TMs from mine tailing soils. Generally, an electrokinetic soil remediation process depends upon the treatment time (Ma et al. 2010), by means that longer treatment may also increase the operating cost of the process in terms of cost of chemical reagents and cost of the electrical energy (Pedersen et al. 2015). The basic idea of this study is to determine the operating cost of the electrokinetic treatment of contaminated soils particularly in the remediation of toxic metals. Our previous study investigated the removal of these six TMs using reducing agents enhanced electrokinetic soil remediation process (Reddy & Yarrakula 2019). The TM ionic species would easily migrate from the soil surface to the electrolytic solutions in the presence of electric current (Giannis & Gidarakos 2005, Giannis et al. 2009). Our previous study investigated the removal of these six TMs using reducing agents enhanced electrokinetic soil remediation process (Reddy et al. 2019). In this study, we incorporated the chelating agents (Citric acid and EDTA) for enhanced electrokinetic soil remediation process followed by the estimation of operating cost of the process.

The purpose of the study is to estimate the operating cost of the electrokinetic experiments for removal of TMs from granite mining soil with the help of proposed cost estimation models. The study is classified into two sections, the first section dealt with the electrokinetic removal of TMs by using most efficient chelating agents such as Citric acid and EDTA and subsequent section dealt with the estimation of the operating cost with help of proposed cost estimation models in this study.

MATERIALS AND METHODS

Experimental Design

Recent studies used the electrokinetic reactor with a design of 30cm × 20cm × 15cm in laboratory scale and fabricated with Plexiglas along with two electrode chambers with dimensions of 5cm × 20cm × 15cm and a working volume of 1.5L (Fig. 1) (Reddy et al. 2019). We used the same design of the electrokinetic reactor, and two graphite electrodes act as anode and cathode with a length of 15cm and a diameter of 1.5cm. We conducted four different EKSR experiments with prior prepared anolyte and catholyte solutions as represented in Table 1.

Determination of TMs Concentration and Electricity Consumption

Atomic absorption spectroscopy (AAS) Varian AA110 spectrophotometer used to measure TMs concentration of the soil in pre-treatment and post-treatment of four electrokinetic experiments (Wang et al. 2014, Reddy et al. 2019). The removal efficiency or removal performance can be determined using the Equation (1) (Tang et al. 2014, Bahemmat et al. 2016).

$$\text{Removal efficiency (\%)} = \frac{C_0 - C_f}{C_0} \times 100 \qquad ...(1)$$

Where, C_0: Initial concentration, C_f: Final concentration (mg/kg) of the soil pre-treatment and post-treatment of electrokinetic removal of TMs.

The expenditure of electrical energy is directly related to the remediation time using the integral time for the passing of current during the electrokinetic process across the electrokinetic reactor. The consumption of electricity to be determined with the Equation (2) (Ma et al. 2010, Pedersen et al. 2015).

$$\text{Energy expenditure (E}_e) = \quad P = \frac{1}{V_S} \int_0^t VI dt \qquad ...(2)$$

Where,

E_e: The energy expenditure (Wh/m^3),

P: The electrical power (W)

Table 1: Experimental pattern of four different EKSR experiments.

Exp. No.	Anolyte	Soil saturation	pH	Catholyte	Duration (Days)	Electric potential (V/cm)
Exp.1	Distilled water	Distilled water	9.7	Distilled water	5, 10, 15, 20	2
Exp.2	Distilled water	0.1M Citric acid	5.7	0.1M Citric acid	5, 10, 15, 20	2
Exp.3	Distilled water	0.1M Citric acid	5.9	0.1M EDTA*	5, 10, 15, 20	2
Exp.4	0.1M NaOH	Distilled water	9.7	Distilled water	5, 10, 15, 20	2

* EDTA: Ethylenediaminetetraacetic acid

V_S: The volume of the soil (m^3)

I: The current passage across the electrokinetic reactor (A)

V: The voltage (V)

t: The treatment time (h)

Proposed Cost Estimation Models

The operating cost of the electrokinetic experiments was estimated through the consumption of enhancing solutions and electrical energy during electrokinetic removal of TMs from mine tailing soil. The total cost of the process was estimated by adding the individual costs of enhancers and cost of electricity expenditure. Generally, the total cost of the electrokinetic process is directly related to the remediation time which includes the electricity supplied across the electrokinetic reactor. The cost of the enhancers was estimated by the quantity of enhancers consumed in the process multiplied by the cost of one unit. In the same way, the cost of electricity was estimated by the electricity consumption charges in India. Our study considered the operating cost of the electrokinetic experiments rather than the capital investment of the process.

Several studies used the equation (2) for estimating the amount of electricity consumed during electrokinetic removal of heavy metals from polluted soils (Ma et al. 2010, Pedersen et al. 2015, Reddy et al. 2019). We simplified the equation (2) into equations (4 and 5) for quick estimation of the cost of enhancers and the cost of the electricity for four different electrokinetic experiments. In this study, we proposed the following three cost estimation models to determine the enhancers cost, electricity cost and the total operating cost of the electrokinetic process. The models were well correlated with manual calculations in the estimation of enhancers cost and electricity cost for four different electrokinetic experiments.

Enhancers cost (E_C) can be estimated by the following Equation (3):

$$E_C = \frac{1}{V_S}\left[\sum_i^n (m_i \times c_i)\right] \qquad \dots(3)$$

Electricity cost (EE_C) can be estimated by the following Equation (4):

$$EE_C = \frac{1}{V_S}\left[\left(10^{-3}\int_0^t VIdt\right)C_c\right] \qquad \dots(4)$$

The Total cost (T_C) can be estimated by the following Equation (5):

$$T_C = \frac{1}{V_S}\left[\sum_i^n (m_i \times c_i) + \left(10^{-3}\int_0^t VIdt\right)C_c\right] \qquad \dots(5)$$

Here,

T_C: The total cost of the process including enhancers and energy cost

V_S: The volume of the soil (m^3)

m_i: The amount of enhancing reagent (kg)

C_i: The cost of enhancing reagent ($/kg)

I: The current passage across the electrokinetic reactor (A)

V: The voltage (V)

t: The treatment time (h)

RESULTS AND DISCUSSION

Conventional Electrokinetic Experiment

During the electrokinetic process, the toxic metal ionic species are dissociated from the soil surface and travelled between anode compartment and cathode compartment by electromigration, electrophoresis and electroosmosis. The metallic species was collected through anolyte and catholyte solutions and measured their concentration via acid digestion followed by atomic absorption spectrometry. A conventional electrokinetic experiment was performed for 5-20 days by purging distilled water for both the anode chamber and cathode chamber. Thereafter collected the anolyte and catholyte solution samples for detecting the TM species for every five days of treatment. The removal performance of TM species with respect to time was recorded and can be seen in the Experiment (1) (Fig. 1a). The removal percentage was about 6%, 9%, 16%, 24%, 11% and 32% respectively for Cr, Co, Ni, Cu, Zn and Mn after 20 days of treatment. The average percentage of removal of all metallic species found that around 13% by means that longer treatment time (>20days) is required to remove the TMs completely from mine tailing soil.

Enhanced Electrokinetic Experiments

During the enhanced electrokinetic process, the metallic species were collected through anolyte and catholyte solutions and measured their concentration via acid digestion followed by atomic absorption spectrometry. The enhanced electrokinetic experiments were performed for 5 to 20 days by purging citric acid and EDTA as chelates at the cathode chamber to suppress the increase in pH of catholyte. The acidic environment was most favoured for the dissociation
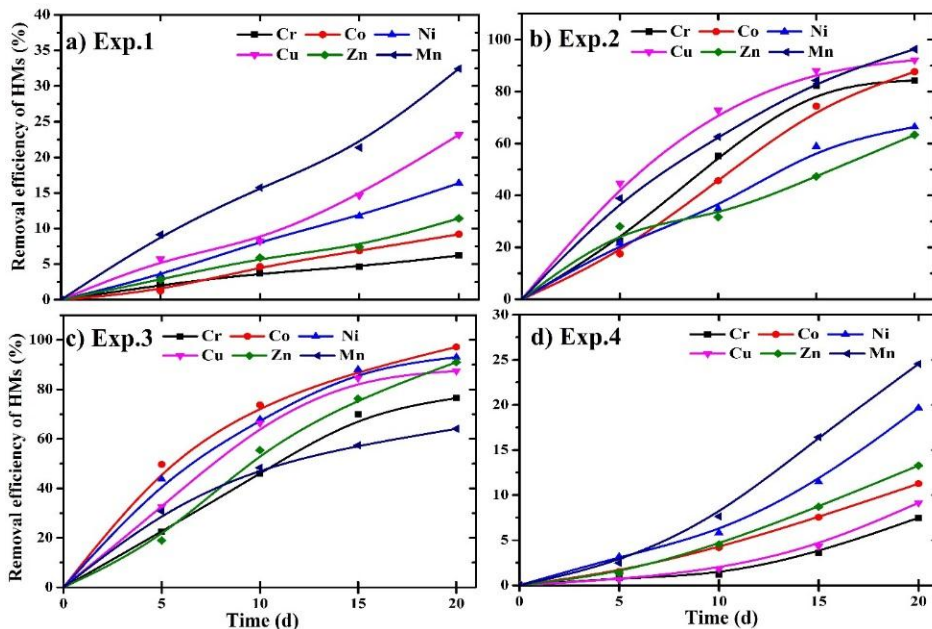
Fig. 1: Removal performance (%) of TMs in experiments (1, 2, 3 and 4) w.r.t. time for 20 days.

of metallic species from the soil surface to anolyte/catholyte electrolyte solutions. Thereafter collected the anolyte and catholyte solution samples for detecting the TM species for every five days of treatment. The removal performance of TM species with respect to time was recorded (Fig. 1b & 1c) and can be observed in Experiments (2 &3). The removal percentage was ranged about 76%-84%, 88%-97%, 66%-93%, 87%-92%, 63%-91% and 64%-96% respectively for Cr, Co, Ni, Cu, Zn and Mn after 20 days of treatment. The average removal performance (%) of all metallic species found that around 84% by means that 25-30 days treatment time is required to remove the TMs completely from mine tailing soil in the case of chelating enhanced electrokinetic experiments. However, the results demonstrated that the enhanced electrokinetic removal of TMs was about six times more than the conventional method.

Alkali Electrokinetic Experiment

The experiment (4) was performed for 5-20 days by purging sodium hydroxide (NaOH) at the anode chamber to increase the pH of the anolyte solution. Thereafter collected the anolyte and catholyte solution samples for detecting the TM species for every five days of treatment. The removal performance of TM species with respect to time was recorded (Fig. 1d). The removal percentage was about 7%, 11%, 19%, 9%, 13% and 24% respectively for Cr, Co, Ni, Cu, Zn and Mn after 20 days of treatment. The average percentage of removal

of all metallic species found that around 14% by means that longer treatment time (>20days) is required to remove the TMs completely from mine tailing soil. The results indicated the removal of TMs in alkali electrokinetic experiment was close to conventional electrokinetic experiment.

Cost Estimation of Electrokinetic Experiments

In the case of the conventional electrokinetic experiment (1), the enhancers were not used, hence the total operating cost was directly related to the electricity consumption for 20 days of operation. The expenditure of electrical energy was about 1104 kWh/m^3 respectively. However, the total operating cost estimated by the proposed model was around 110.4 US$/m^3. The citric acid as enhancing electrokinetic experiment from Table 2 and Fig. 2, the total operating cost was directly related to the electricity consumption and the amount of citric acid utilized for 20 days of operation. The citric acid was consumed about 146 kg/m^3 with a cost of 293 US$/m^3. The consumption of electricity was about 2480 kWh/m^3 with a cost of 248 US$/m^3 respectively. However, the total operating cost was estimated at around 541 US$/m^3. In the case of EDTA, an enhancing electrokinetic experiment, the total cost was related to the electricity consumption and amount of citric acid consumed during removal of TMs for 20 days of operation. The consumption of EDTA was about 267 kg/m^3 with a cost of 800 US$/m^3 and citric acid cost was about 26 US$/m^3 for soil saturation. The consumption of electricity

was about 1800 kWh/m^3 with a cost of 180 US\$/m^3 respectively. However, the total operating cost was estimated at around 1006 US\$/m^3. In the case of the alkali electrokinetic experiment, the total operating cost was directly related to the electricity consumption and amount of NaOH utilized for 20 days of operation. The NaOH was consumed about 53.3 kg/m^3 with a cost of 112 US\$/m^3. The consumption of electricity was about 5080 kWh/m^3 with a cost of 508US\$/m^3 respectively. However, the total operating cost was estimated around 620 US\$/m^3 (Fig. 2). The electricity cost of experiment 4 with NaOH was almost twice higher than other experiments 1-3, it might be the reason that the increase in pH of the soil matrix during the electrokinetic process.

The results demonstrated that conventional and alkali electrokinetic experiments were costly and not effective in the removal of TMs from contaminated soils. The citric acid enhanced electrokinetic experiment was more effective and economically feasible than the EDTA enhanced electrokinetic experiment in the removal of TMs from contaminated soils.

CONCLUSION

Our study proposed the cost estimation models to estimate the operating cost such as cost of chemical reagents, cost of electrical energy and total cost of the process in the electrokinetic removal of TMs from contaminated soil. We estimated the operating cost of conventional and enhanced electrokinetic treatment processes in specifically for the removal of six TMs such as chromium (Cr), cobalt (Co), copper (Cu), nickel (Ni), zinc (Zn) and manganese (Mn) from granite mine tailing soil. We investigated that the chelating enhanced electrokinetic removal of TMs about six times more than the conventional process in 20 days of operation. Furthermore, we estimated the operating cost of the conventional and enhanced electrokinetic processes was about US\$110 to US\$508 per cubic meter of treated soil. The total operating cost becomes US\$110 to US\$1006 per cubic meter of treated soil including enhancers cost. We conclude that the chelating enhanced electrokinetic treatment of soil was more effective and economically feasible than conventional treatment for removal of TMs from contaminated soil.

Table 2: Cost estimation of four electrokinetic experiments.

Exp. No	Enhanced solution consumption						Energy consumption		[e]Total cost (\$/m^3)
	Citric acid		EDTA		NaOH		Power		
	Consumption (kg/m^3)	[a]Cost (\$)	Consumption (kg/m^3)	[b]Cost (\$)	Consumption (kg/m^3)	[c]Cost (\$)	Consumption (kwh/m^3)	[d]Cost (\$)	
Exp.1	0	0	0	0	0	0	1104	110.4	110.4
Exp.2	146.6	293.2	0	0	0	0	2480	248	541.2
Exp.3	13.3	26.6	266.6	800	0	0	1800	180	1006.6
Exp.4	0	0	0	0	53.33	112	5080	508	620

[a] Cost of citric acid per kg: 2\$; [b]Cost of EDTA per kg:3\$; [c] Cost of NaOH pellets per kg: 2.1\$; [d] Average cost of electricity consumption in India per kilo-Watt-hour(kWh): 0.1\$.
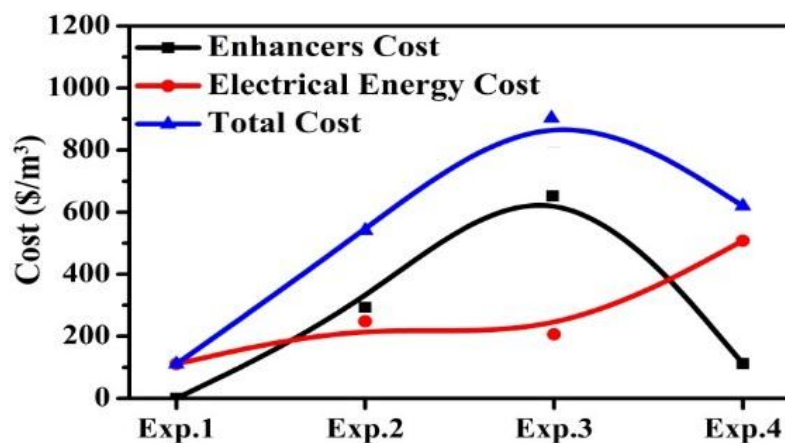

Fig. 2: Cost Estimation of four electrokinetic experiments.

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