

# Numerical Implementation of Electrokinetics for Removal of Heavy Metals from Granite Waste

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**ABSTRACT:** *The goal of the study is to incorporate the electrokinetic models and estimate the remediation time for maximum removal of Heavy Metals (HMs) from polluted soils. Most of conventional electrokinetic technologies have not considered the electrokinetic models in the removal of HMs from polluted soils. We addressed this problem and incorporated the electrokinetics and applied it to the experimental ElectroKinetic Soil Remediation (EKSR) process particularly, to extract the numerical data between the removal performance of HMs versus remediation time with the help of the MATLAB program. In the experimental study, chelating chemical agents (citric acid and ethylenediaminetetraacetic acid (EDTA)) were used in EKSR process under a constant voltage gradient (2V/cm) for the removal of Chromium (Cr), Cobalt (Co), Nickel (Ni), Copper (Cu), Zinc (Zn) and Manganese (Mn) ions from granite dump soil. We experimentally investigated that the removal performance of HMs in chelating agents enhanced EKSR was about 6 to 7 times more than when unenhanced in 20 days of treatment. Furthermore, we estimated the remediation time of about 52 to 54 days for the complete removal of HMs using electrokinetic models. The study may be useful for the researcher particularly, in soil decontamination studies to overcome the uncertainty in the process optimization and scale up the process to the pilot plant and field level.*

**KEYWORDS:** *Electrochemical technology; Chelating agents; Heavy metals, Electrokinetics, MATLAB Software Program.*

## INTRODUCTION

In the past few decades, a vast amount of polluted waste has been produced by various types of anthropogenic activities such as mining, industrial and chemical. The generated waste becomes the major source of potentially hazardous heavy metals (HMs) and which may have adversely affected the environment. Recent studies reported that heavy metals such as Chromium (Cr), Cobalt (Co), Nickel (Ni), Copper (Cu), Zinc (Zn)

and Manganese (Mn) ions at elevated concentrations in granite mining waste [1]. During the mineral and milling process, a vast quantity of granite waste is produced and accumulated at dump yards, which may cause environmental pollution [2]. Recently, star-shaped microfluidic channel techniques have been used to detect HMs [3]. The concentrations of HMs assessed with advanced remote sensing and Geography Information

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**Table 1: Recently Reported the Enhanced EKSR for the Removal of HMs from Polluted Soils.**

HM	Contamination source	Enhancement Agents	Duration (days)	*Removal (%)	Reference
Cr	Industrial waste	Acetic acid, Citric acid, Hydrochloric acid, Hydrogen peroxide	15	92	[17]
	Seabed area	Stirred suspension (1300 rpm)	20	18-21	[18]
Co	Mine tailing	Nitric acid, Humic acid, Fulvic acid	20	29-38	[19]
Cu	Mine tailing	Pulse current frequency	10	1.5	[20]
	Industrial waste	Diffuse double-layer processes	5	-	[21]
	Seabed area	Stirred suspension (1300rpm)	20	41-67	[18]
	Municipal sludge	Nitric acid, Sodium nitrate	6	83	[22]
Ni	Seabed area	Stirred suspension (1300rpm)	20	24-49	[18]
	Mine tailing	Nitric acid, Humic acid, Fulvic acid	20	41-70	[19]
	Harbour sediment	Nitric acid	120	42	[23]
	Municipal sludge	Nitric acid, Sodium nitrate	6	73	[22]
Zn	Seabed area	Stirred suspension (1300 rpm)	20	71-74	[18]
	Mine tailing	Nitric acid, Humic and Fulvic acid	20	25-47	[20]
	Harbour sediment	Nitric acid	120	28	[23]
Mn	Seabed area	Stirred suspension (1300rpm)	20	36-37	[18]
	Mine tailing	Nitric acid, Humic acid, Fulvic acid	20	38-64	[19]

$$\text{*Removal efficiency (\%)} = \frac{C_0 - C_f}{C_0} \times 100 \quad (C_0 : \text{Initial Concentration}, C_f : \text{Final Concentration})$$

System (GIS) particularly for groundwater [4,5]. The environmental impact assessment of HM polluted water and soil were estimated in order to mitigate the environmental pollution [6]. Conventional methods such as biosorption, phytochemical oxidation, coconut coir, saxaul tree ash, and nickel oxide/carbon nanotube composites (NiO/CNT) were investigated to remove HMs and organic contaminants from wastewater [7-11]. Most of the reported conventional remediation studies are ineffective and not feasible on a large scale at the field level. The electrochemical behavior of different metals and metal alloys was studied in sodium chloride solution for environmental and biomedical applications [12,13]. Recent studies investigated the chelating and reducing agents enhanced electrokinetic remediation for the removal of HMs from polluted soil [2]. The metal or HM ions would be desorbed under an acidic environmental condition which is generated at the anode during the electrokinetic process [14]. An electrokinetic soil remediation technology has emerged

and successfully applied for the removal of toxic HMs and organic contaminants from water and soil [15]. The removal of toxic HMs from clayey soil was applied in several field applications by using electric current [16].

Many studies performed well in the electrokinetic removal of HMs on a laboratory scale as reported in Table 1. The removal of HMs in the enhanced ElectroKinetic Soil Remediation (EKSR) process by controlling pH with aid of enhancing chemical solutions.

As seen in Table 1, the removal of chromium(Cr) ions was difficult from mine tailing soils under normal conventional electrokinetic methods, however, enhanced EKSR was applied to remove about 92% of Cr ions by adding strong oxidizing agents such as citric acid and hydrogen peroxides (H<sub>2</sub>O<sub>2</sub>) in field-scale [19,20]. The quantity of cobalt (Co) ions about 38% were successfully removed from mine tailing by adding enhancement chemical reagents such as nitric acid and Humic substances in a pilot plant scale. The removal of copper (Cu) ions about 83%

from municipal sludge by the addition of nitric acid and sodium nitrite reagents within 6 days of operation [21,22]. The amount of nickel (Ni) ions, is removed by almost 73% from mine tailing soils in 6 days of treatment under enhanced conditions [18-23]. In the same way, the amount of zinc (Zn) and manganese (Mn) ions were removed around 25-47% and 38-64% from mine tailing soil by the addition of nitric acid, humic acid, and fulvic acid as enhancement reagents within 20 days of operation on a laboratory scale [18,19].

Several efforts have been focused on waste minimization or reduction by introducing enhanced EKSR methods. However, the conventional electrokinetic technologies concentrated on the removal of HMs from polluted soils but not considered the cost of the process [24-26]. However, from a practical point of view, each remediation step has a cost and it is highly dependent on treatment time. Longer remediation has also increased the cost of the EKSR process in terms of enhancers, electrical energy, and operating costs. Because the electrical energy consumption is directly proportional to the time of electric current usage across the electrokinetic reactor [27]. Several recent studies face uncertainty in the remediation time, particularly in the removal of HMs from contaminated soil. We addressed this problem and used electrokinetics to estimate the remediation time for the complete removal of HMs from contaminated soils.

The goal of the study was to estimate the remediation time in the electrokinetic removal of Chromium (Cr), Cobalt (Co), Nickel (Ni), Copper (Cu), Zinc (Zn), and Manganese (Mn) ions from granite mining dump soils. In the present work, we incorporated electrokinetics to estimate the treatment time for the complete removal of HMs and further, the MATLAB software program was used to solve the electrokinetic models with encoding programs. Our study is classified into two main sections, First, the variation of concentration profiles and removal efficiency w.r.t time under chelating agents. Subsequent sections focussed on the estimation of numerical data between removal efficiency of HMs versus remediation/treatment time using the MATLAB coding program.

## EXPERIMENTAL SECTION

### *Characteristics of the soil and enhancing agents*

The soil samples were collected in the vicinity of the granite mining industry at Chimakurthy, Andhra Pradesh, India.

The random sampling method was used to collect the top layer of soil from a depth of 30cm [1]. Further, sieved in a series of 4.75mm to 0.02mm mesh nylon fiber sieves to remove any vegetation material and large particles and kept in polythene zipped covers for further analysis [28,29]. Prior to electrokinetic soil remediation, the physicochemical properties of soil were characterized in the laboratory and reported in Table 2.

The chelating agents can capture the metal ions due to their bi-dentate (two bonds) or multi-dentate (more than two bonds) ligand (the chelant), and a single metal central atom or iron. However, metals or heavy metals are easily desorbed and separated from the soil surface and get water-soluble metal ion complexes through chelating agents such as hydroxy carboxylates (Ex: citric acid) and amino polycarboxylates (Ex: EDTA(diethylene triamine), pentaacetic acid (DTPA) and pentaacetic acid (DTPA)). Sodium hydroxide (NaOH) is alkaline in nature and enhances the electroosmotic flow due to its ionization under low-voltage electric fields [30]. In this study, chelating agents such as citric acid and EDTA were used to investigate the enhanced ElectroKinetic Soil Remediation (EKSR) process.

### *Experimental design of the electrokinetic reactor*

Recent studies used the electrokinetic reactor with a design of 30cm x 20cm x 15cm on a laboratory scale and fabricated with Plexiglas along with two electrode chambers with dimensions of 5cmx20cmx15cm and a working volume of 1.5L as shown in Fig. 1 [1]. We used the same design of an electrokinetic reactor, and two graphite electrodes act as anode and cathode with a length of 15cm and diameter of 1.5cm. We conducted four different EKSR experiments with prior prepared anolyte and catholyte solutions as represented in Table 3.

### *The removal efficiency of HMs*

Atomic absorption spectroscopy (AAS) Varian AA110 spectrophotometer was used to measure the concentration of HMs of soil in pre-treatment and post-treatment of four EKSR processes. The removal efficiency or removal performance can be determined using Eq. (1) [31-33]:

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

Table 2: The Physicochemical Characteristics of Soil.

Soil characteristics		Units	Value	Method
Gravel (>4.75 mm)		%	36	Sieve analysis
Sand	Coarse (4.75-2.00 mm)	%	12.81	Sieve analysis
	Medium (2.00-0.425 mm)	%	13.06	Sieve analysis
	Fine (0.425-0.075 mm)	%	11.91	Sieve analysis
Silt (0.075-0.002 mm)		%	13.02	Sieve analysis
Clay (<0.002mm)		%	14.12	Sieve analysis
Porosity		-	0.44	Permeability method
pH		-	9.8	1:2.5 soil/water slurry
Electrical Conductivity		* $\mu\text{S/ppm}$	147.97	1:5 soil/water slurry
Zeta Potential		**mV	-26.5	Electrokinetic method
Total dissolved salts (TDS)		$\dagger\text{mS/ppm}$	173.18	1:5 soil/water slurry
Moisture		%	0.92	Drying method at 125 $^{\circ}\text{C}$
Total Inorganic Matter		%	97.57	Drying method at 500 $^{\circ}\text{C}$
Total Organic Matter		%	1.44	Drying method at 500 $^{\circ}\text{C}$
Chromium		mg kg $^{-1}$	192	Acid digestion
Cobalt		mg kg $^{-1}$	175	Acid digestion
Nickel		mg kg $^{-1}$	204	Acid digestion
Copper		mg kg $^{-1}$	363	Acid digestion
Zinc		mg kg $^{-1}$	505.1	Acid digestion
Manganese		mg kg $^{-1}$	704	Acid digestion

\* $\mu\text{S/ppm}$ : micro siemens/ parts per million; \*\*mV: milli volts;  $\dagger\text{mS/ppm}$ : milli siemens/ parts per million

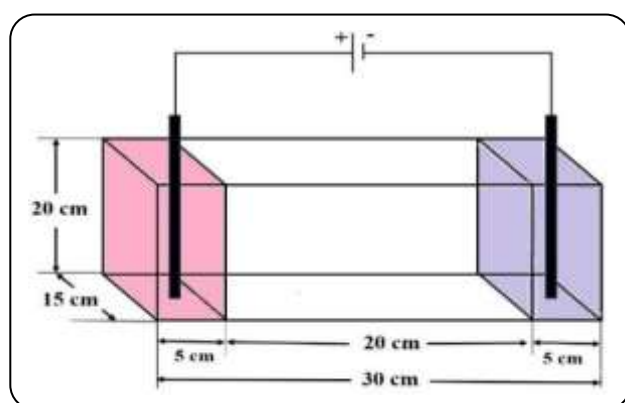


Fig. 1. Design of Electrokinetic Reactor used in this Study

Where,  $C_0$ : Initial concentration,  $C_f$ : Final concentration (mg/kg) of the soil before and after electrokinetic treatment.

### Electrokinetics

In this study, the main three ion transport processes considered such as diffusion, electromigration, and electroosmosis. Furthermore, we considered the permeable soil medium is saturated and isothermal. At these conditions, the flux density per unit cross-sectional area of porous medium  $J_i$  (mol/m $^2$ s) of a dissolved chemical  $i^{\text{th}}$  species can be expressed in Eq. (2) as [34]:

$$J_i = -D_i^* \nabla c_i - D_i^* c_i \nabla \phi - k_{eo} c_i \nabla \phi \quad (2)$$

Where,

$D_i^*$ : Effective diffusion coefficient of  $i^{\text{th}}$  chemical species (m $^2$ /s)

$c_i$ : The concentration of  $i^{\text{th}}$  chemical species (mol/m $^3$ )

Table 3: Experimental Pattern of Four Different EKSR Experiments.

Exp. No.	Anolyte purging solution	Soil saturation	pH	Catholyte purging solution	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

$U_i^*$ : The effective ion mobility of  $i^{\text{th}}$  chemical species ( $\text{m}^2/\text{sV}$ )

$\phi$ : The electric potential (V)

$k_{eo}$ : The coefficient of electroosmotic permeability ( $\text{m}^2/\text{Vs}$ ).

$\nabla c$ : The connection gradient of  $i^{\text{th}}$  chemical species with x-direction of soil matrix  $i^{\text{th}}$  chemical species i.e concentration/ length of the soil matrix between electrodes ( $\text{mol}/\text{m}^2$ ).

$\nabla \phi$ : The electrical potential gradient of  $i^{\text{th}}$  chemical species with x-direction of soil matrix  $i^{\text{th}}$  chemical species i.e electrical potential/length of the soil matrix between electrodes ( $\text{mol}/\text{m}^2$ ) variation/difference (V/cm).

The porous medium allows the tortuous path of ions, hence the effective diffusion coefficient/diffusivity and effective ion mobility used in Equation (2) can be rewritten in terms of the effect of porosity ( $\phi$ ) and tortuosity ( $\tau$ ) in Eqs (3,4):

$$D_j^* = \tau \phi D_j \quad (3)$$

$$U_i^* \tau \phi U_i \quad (4)$$

Where,

$D_i$ : The diffusion coefficient ( $\text{m}^2/\text{s}$ )

$U_i$ : Ion mobility at infinite dilution ( $\text{m}^2/\text{sV}$ ).

According to the Nernst-Townsend-Einstein (NTE), the relationship between diffusivity and ionic mobility is expressed in Eq. (5):

$$U_j^* = \frac{D_j^* z_j F}{RT} \quad (5)$$

Where,

R: The universal gas constant (8.314 J/mol K),

T: The absolute temperature (K)

F: The Faraday's constant (96485 C/mol).

The most widely used electroosmotic flow equation for the soil system was proposed by Casagrande [30-34]. The coefficient of electroosmotic permeability coefficient ( $k_{eo}$ ) by electroosmotic flow rate ( $Q_{eo, i}$ ) in the case of one dimensional (1D) in x-direction given by Equation (6) [34,35].

$$Q_{eo, i} = k_{eo} \phi_x A \quad (6)$$

Here,  $\phi_x$ : The electric field (V) and  $\partial \phi / \partial x$ : The electric field in the x-direction (V/m),

Both anode and cathode compartments are considered Continuous flow Stirred-Tank Reactors (CSTRs), further considered as the electrochemical reactions carried out on the surface of both electrodes. Hence, the initial and boundary conditions were defined by the law of mass conservation equations for anode and cathode chambers and reported in Table 3.

The mass balance Eqs (7 and 8) can be defined for the  $i^{\text{th}}$  chemical species concentration inside each anode and cathode compartment was uniform:

$$V_a \frac{dc_i^a}{dt} = -J_i A - Q_{f, a} c_i^a = R_i^a \quad (7)$$

$$V_c \frac{dc_i^c}{dt} = -J_i A - Q_{f, c} c_i^c = R_i^c \quad (8)$$

Where,

$V_a$ : The volume of the anode compartment ( $\text{m}^3$ )

$V_c$ : The volume of the cathode compartment ( $\text{m}^3$ )

$c_i^a$ : The concentration of chemical species in the anolyte ( $\text{mol}/\text{m}^3$ )

$c_i^c$ : The concentration of chemical species in catholyte ( $\text{mol}/\text{m}^3$ )

$J_i$ : The flux density ( $\text{mol}/\text{m}^2\text{s}$ ) of the  $i^{\text{th}}$  species

A: The cross-sectional area ( $\text{m}^2$ )

$Q_{f, a}$ : The flushing flow rates of the anolyte ( $\text{m}^3/\text{s}$ )

$Q_f^c$ : The flushing flow rates catholyte ( $m^3/s$ )

$R_i^a$ : The net production of the  $i^{\text{th}}$  species due to electrochemical reactions at the anode (mol/s)

$R_i^c$ : The net production of the  $i^{\text{th}}$  species due to electrochemical reactions at the cathode (mol/s)

By integrating the mass balance Eqs (7, 8) with a lower limit is considered zero which means that an initial concentration of HMs species was zero in both electrolytes such as anolyte and catholyte. Then the simplified final concentration of HMs species in anolyte and catholyte can be written in Eqs (9, 10):

$$c_{i,f}^a = \left[ \frac{R_i^a - J_i A}{Q_{f,a}} \right] \left( 1 - e^{-\frac{Q_f^a}{V_a} t} \right) \quad (9)$$

$$c_{i,f}^c = \left[ \frac{R_i^c - J_i A}{Q_{f,c}} \right] \left( 1 - e^{-\frac{Q_f^c}{V_c} t} \right) \quad (10)$$

The simplified equation for determining the final concentration of HM species in anolyte and catholyte can be rewritten in Eq. (11) by adding both Eqs (9,10):

$$c_{if} = \left[ \frac{R_i^a - J_i A}{Q_{f,a}} \right] \left( 1 - e^{-\frac{Q_f^a}{V_a} t} \right) + \left[ \frac{R_i^c - J_i A}{Q_{f,c}} \right] \left( 1 - e^{-\frac{Q_f^c}{V_c} t} \right) \quad (11)$$

Here,

$C_{i,f}^a$ : The final concentrations of  $i^{\text{th}}$  species in the anolyte solution ( $\text{mol}/m^3$ ),

$C_{i,f}^c$ : The final concentrations of  $i^{\text{th}}$  species in the catholyte solution ( $\text{mol}/m^3$ ),

$C_{i,f}$ : The final concentrations of  $i^{\text{th}}$  species in both anolyte and catholyte solution ( $\text{mol}/m^3$ ).

The number of  $i^{\text{th}}$  species (HM ions) accumulated w.r.t Time can be expressed in equation (12):

$$m_i = c_{i,f} \times mw_i \times V_{A+C} \quad (12)$$

Where,

$m_i$ : The amount of metal accumulated in the anolyte and the catholyte ( $mg$ ),

$mw_i$ : The molecular weight of the  $i^{\text{th}}$  species

$V_{A+C}$ : The total volume of anolyte and catholyte collected (liters)

$M_i$ : The initial amount of HM in the soil ( $mg$ ),

The removal efficiency or removal performance ( $\eta_e$ ) of HMs can be estimated by the following equation (13):

$$\eta_e = c \left( \frac{m_i}{M_i} \right) \times 100 \quad (13)$$

### Estimation of remediation time for complete removal of HMs

In this study, the main three ion transport equations are considered such as diffusion, electromigration, and electroosmosis to estimate the removal of HMs completely from contaminated soil [42]. We considered the electrochemical reaction rates are at equilibrium for four EKSR experiments. By considering these assumptions we numerically simplified the transport equations. Several studies considered these assumptions and investigated that well in agreement with electrokinetic experimental data [42,43].

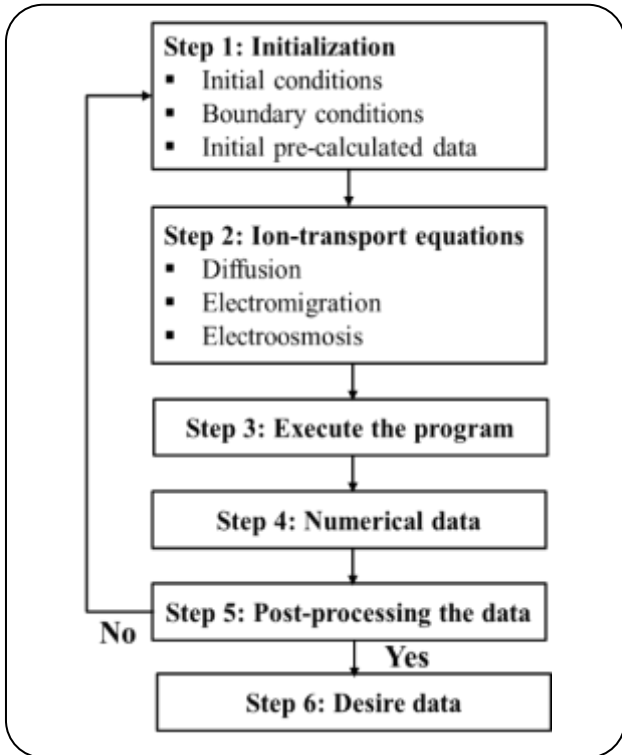
In the program coding in MATLAB, the initial, boundary conditions, and pre-calculated kinetic parameters were specified in Table 3. Then, the three-transport process such as diffusion, electromigration, and electroosmosis kinetic equations (Eqs (2-11)) was described as shown in Fig. 2. Further, executed the program with a major loop to solve the kinetic equations and predict the numerical data between removal efficiency versus various time intervals. After achieving desired numerical data by means of the removal of HMs completely that time was recorded. We estimated the remediation time for the complete removal of HMs for four EKSR experiments using electrokinetic models with the help of the MATLAB program.

### MATLAB Code for Electro-Kinetics

```
%% Matlab code for Electro-Kinetics
close all
clear
clc
disp('Author Name: Koteswara Reddy G')
disp('Job: Assistant Professor, KL University,
Vijayawada.')
```

**Table 3: Initial, Boundary Conditions, Pre-calculated Data and Kinetic Parameters used to Solve the Electrokinetic Equations with the MATLAB.**

Parameter	Value	Unit	Description
$\tau$	0.7	-	Tortuosity
$\phi$	0.44	-	Porosity
F	96485	C mol <sup>-1</sup>	Faraday's constant
T	298	K	Room temperature
R	8.314	J K <sup>-1</sup> mol <sup>-1</sup>	Universal gas constant
V <sub>a</sub>	3.75 X 10 <sup>-4</sup>	m <sup>3</sup>	Volume of anode compartment
V <sub>c</sub>	3.75 X 10 <sup>-4</sup>	m <sup>3</sup>	Volume of cathode compartment
A	3 X 10 <sup>-2</sup>	m <sup>2</sup>	Cross sectional area of soil matrix
$\varphi$	50	V	Electrical potential
$\varphi_x$	200	V/m	Voltage gradient
Cr(VI)	1.14X10 <sup>-9</sup>	m <sup>2</sup> /s	Diffusion coefficient [36]
Co <sup>2+</sup>	1.18X10 <sup>-9</sup>	m <sup>2</sup> /s	Diffusion coefficient [37]
Ni <sup>2+</sup>	6.79X10 <sup>-10</sup>	m <sup>2</sup> /s	Diffusion coefficient [38]
Cu <sup>2+</sup>	1.16X10 <sup>-9</sup>	m <sup>2</sup> /s	Diffusion coefficient [39]
Zn <sup>2+</sup>	7.02X10 <sup>-10</sup>	m <sup>2</sup> /s	Diffusion coefficient [40]
Mn <sup>2+</sup>	1.25X10 <sup>-9</sup>	m <sup>2</sup> /s	Diffusion coefficient [41]



**Fig. 2: Steps in the Prediction of Numerical Data using an Encoding Program with the MATLAB.**

```

d = [3.51*10^-10 3.63*10^-10 2.09*10^-10 3.57*10^-10 2.16*10^-10 3.85*10^-10];
divc = [(3.69*10^-2/0.25) (2.96*10^-2/0.25) (3.47*10^-2/0.25) (5.71*10^-2/0.25) ... (7.72*10^-2/0.25) (1.28*10^-2/0.25)];
u = [8.19*10^-8 2.73*10^-8 1.62*10^-8 2.78*10^-8 1.68*10^-8 2.99*10^-8];
ke = 1.7746*10^-8;
c = [3.69*10^-2 2.96*10^-2 3.47*10^-2 5.71*10^-2 7.72*10^-2 1.28*10^-2];
divs = 200;
n = numel(d);
for i = 1:n;
    J(1,i) = -d(1,i)*divc(1,i)-((u(1,i)+ke)*c(1,i)*divs);
end
V = 3.75*10^-4;
A = 3*10^-2;
Q = 1.0648*10^-7;
R = 1.186*10^-5;
syms x
for i = 1:n;
    f(1,i) = (1/V).*((-J(1,i).*A)-(Q.*x)+R);
    
```

```

end
t = 25;
for j = 0;
  for i = 1:n;
    xf1(1,i) = ((R-J(1,i)*A)/Q)*(1-exp(-(Q*j/V)));
  end
end
for j = 5;
  for i = 1:n;
    xf2(1,i) = ((R-J(1,i)*A)/Q)*(1-exp(-(Q*j/V)));
  end
end
for j = 10;
  for i = 1:n;
    xf3(1,i) = ((R-J(1,i)*A)/Q)*(1-exp(-(Q*j/V)));
  end
end
for j = 15;
  for i = 1:n;
    xf4(1,i) = ((R-J(1,i)*A)/Q)*(1-exp(-(Q*j/V)));
  end
end
for j = 20;
  for i = 1:n;
    xf5(1,i) = ((R-J(1,i)*A)/Q)*(1-exp(-(Q*j/V)));
  end
end
for j = 25;
  for i = 1:n;
    xf6(1,i) = ((R-J(1,i)*A)/Q)*(1-exp(-(Q*j/V)));
  end
end
for j = 30;
  for i = 1:n;
    xf7(1,i) = ((R-J(1,i)*A)/Q)*(1-exp(-(Q*j/V)));
  end
end
xf = [ xf1; xf2; xf3; xf4; xf5; xf6; xf7];

```

## RESULTS AND DISCUSSION

### Un-enhanced EKSR experiment

An unenhanced EKSR experiment was conducted for 5 to 20 days by purging distilled water as a purging solution at anode and cathode compartments. For every 5 days after treatment, the anolyte and catholyte samples were analyzed for tracing the HM content. The concentration

and removal efficiency of HMs w.r.t Time was depicted in Fig. 3a and Fig. 4a.

Exp.1 was conducted with the unenhanced EKSR method which means that without adding any enhanced agents, particularly catholyte pH-controlling chemicals. As seen in Table 4 the removal of HMs in unenhanced experiments was not much significant compared with the previous studies on the same mine tailing soils. It is observed that the more precipitation of HMs in anolyte than catholyte during the unenhanced process for different treatment times. Furthermore, the observed removal of HMs is directly proportional to the treatment time which means that a longer treatment time is required to remove the HMs completely from mine tailing soil.

### Enhanced EKSR experiments

Two different types of enhanced EKSR experiments were conducted for 5 to 20 days by purging citric acid and EDTA as chelating agents at the cathode compartment. For every 5 days of treatment, the anolyte and catholyte samples were analyzed for tracing the HM content. The concentration and removal efficiency of HMs w.r.t Time was depicted in Fig. 3(b, c) and Fig. 4(b, c).

Two different EKSR experiments were conducted adding two different catholyte pH controls with citric acid and EDTA as enhancing agents. As seen in Table 5 the removal of HMs in unenhanced experiments was more significant compared with the previous studies on mine tailing soils. In an experiment (Exp.3) removal efficiency of HMs was more even compared to experiment 2 (Exp.2) which indicates that the EDTA acts as the chelating agent. We observed that citric acid/EDTA was more effective than nitric acid and humic substances which were reported in previous studies. More precipitation of HMs occurred in catholyte during the enhanced process, which might be the reason for the chelating effect of EDTA. The ligands citric acid and EDTA were having more encapsulation and entrapment towards the heavy metal ions due to their chelant nature under an electric field, which might be the reason for the removal of HMs about 6 to 7 times more in enhanced EKSR experiments than in unenhanced experiment.

### Alkali-enhanced EKSR experiment

An alkali-enhanced EKSR experiment was conducted for 5 to 20 days by purging NaOH as a purging solution at anode compartment. For every 5 days of treatment,



**Table 4: The concentration and removal efficiency of HMs w.r.t Time during un-enhanced EKSR experiment and comparison with previous similar studies.**

S.No	HM	Initial Concentration	Final Concentration	Removal (%)	Bahemmat et al. (2016) [19]	Sun et al. (2012) [20]
1	Cr	192	180	6.25%	--	--
2	Co	175	158	9.71%	29-38 %	--
3	Cu	363	279	23.14%	--	1.50%
4	Ni	204	170	16.67%	41-70%	--
5	Zn	505	447	11.49%	--	25-47%
6	Mn	734	496	32.43%	38-64%	--

**Table 5: The concentration and removal efficiency of HMs w.r.t Time during enhanced EKSR experiments and comparison with previous similar studies.**

HM	Initial Concentration	<sup>*</sup> Final Concentration	<sup>†</sup> Final Concentration	<sup>*</sup> Removal Efficiency	<sup>†</sup> Removal Efficiency	Bahemmat et al. (2016) [19]	Sun et al. (2012) [20]
Cr	192	30.1	45	84.3%	76.56%	--	--
Co	175	21.6	4.97	87.7%	97.16%	29-38 %	--
Cu	363	28.46	45.45	92.2%	87.4%	--	1.50%
Ni	204	68.2	14.19	66.6%	93.0%	41-70%	--
Zn	505	185.12	45.36	63.3%	91.0%	--	25-47%
Mn	734	26.86	263.72	96.3%	64.1%	38-64%	--

<sup>\*</sup>Exp.2: Citric Acid enhanced EKSR, <sup>†</sup>Exp.3: EDTA enhanced EKSR

**Table 6: The concentration and removal efficiency of HMs w.r.t Time during Alkali-enhanced EKSR experiment and comparison with previous similar studies.**

S.No	HM	Initial Concentration (C <sub>0</sub> )	Final Concentration (C <sub>f</sub> )	Removal Efficiency (%)	Bahemmat et al. (2016) [19]	Sun et al. (2012) [20]
1	Cr	192	177.64	7.5%	--	--
2	Co	175	155.3	11.3%	29-38 %	--
3	Cu	363	329.82	9.1%	--	1.50%
4	Ni	204	163.87	19.7%	41-70%	--
5	Zn	505	438.02	13.3%	--	25-47%
6	Mn	734	553.74	24.6%	38-64%	--

the anolyte and catholyte samples were analyzed for tracing the HM content. The concentration and removal efficiency of HMs w.r.t Time was depicted in Fig. 3d and Fig. 4d.

Exp.4 was conducted with one more enhanced EKSR method by adding NaOH as an anolyte pH-controlling enhancing agent. As seen in Table 6 the removal of HMs in enhanced experiments was not much significant compared with the previous studies on mine tailing soils. It is observed that the less precipitation of HMs in anolyte and catholyte during enhanced processes for different

treatment times. The pH of the soil matrix was drastically increased during NaOH alkali electrokinetic process due to the acidic font would be converted into the basic font. Furthermore, the observed removal of HMs is directly proportional to the treatment time which means that a longer treatment time is required to remove the HMs completely from mine tailing soil. The results demonstrated that the removal of HMs in alkali-enhanced EKSR experiment is close to unenhanced, by means that the precipitation of HMs is less in alkali conditions.

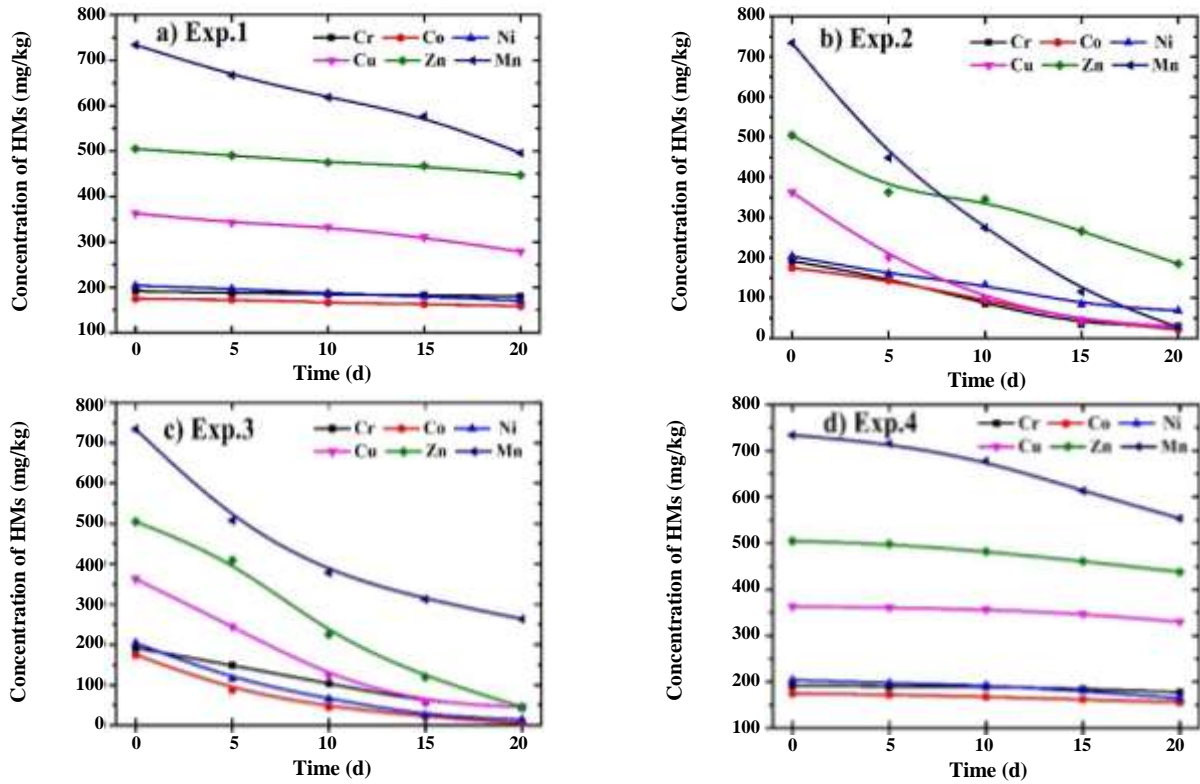


Fig. 3: Concentration profile of HMs w.r.t time for four EKSr process.

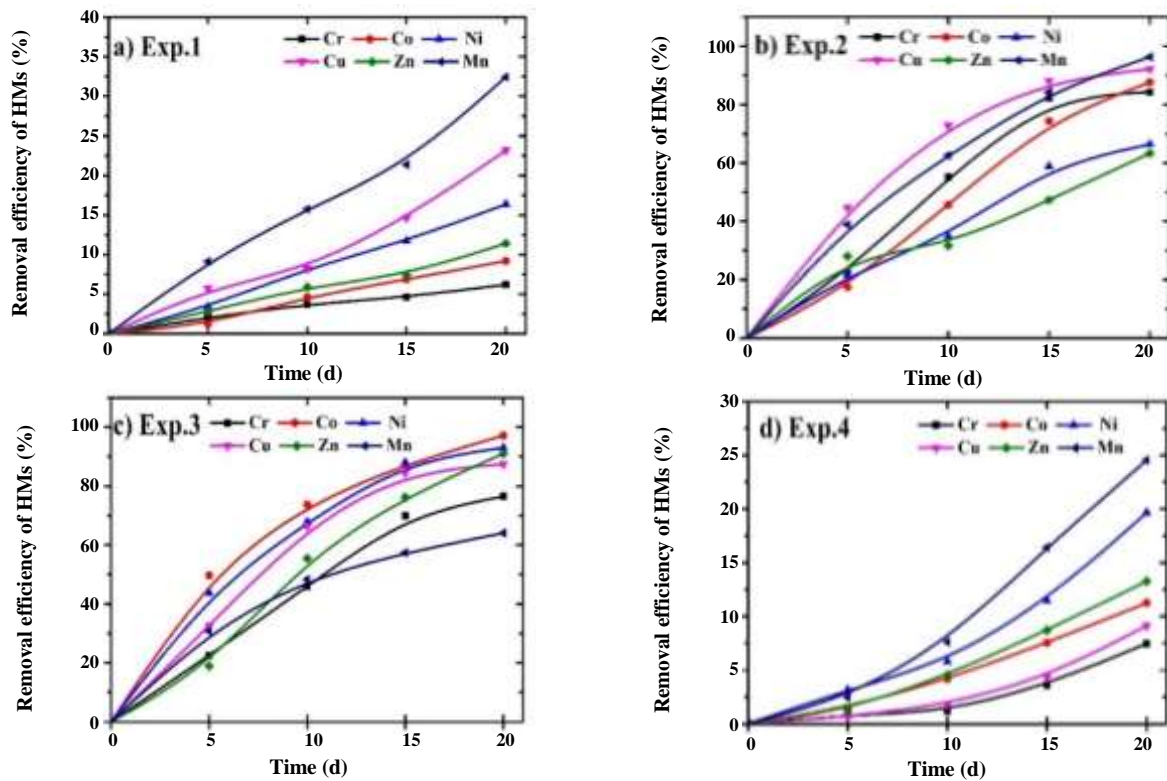


Fig. 4. Removal Performance of HMs w.r.t time for four different EKSr processes.

**Table 7: Estimation of Remediation Time for Complete Removal of HMs for four EKSR Experiments by Solving Electrokinetic Equations using an Encoding Program with MATLAB.**

	Observed (Experiment)			Predicted (ND) <sup>†</sup>			Observed (Experiment)			Predicted (ND) <sup>†</sup>			
	Soil	A+C*	Average	A+C*				Soil	A+C*	Average	A+C*		
Exp.1	20 days			30 days	50 days	200 days	Exp.2	20 days			33 days	41 days	54 days
Cr	6.22	3.49	4.86	5.38	13.39	52.35	Cr	84.32	78.47	81.4	85.4	100	-
Co	9.2	8.04	8.62	6.57	16.34	63.86	Co	87.66	92.13	89.9	100	-	-
Ni	16.37	9.7	13.04	5.6	13.94	54.5	Ni	66.41	50.6	58.51	90.51	100	-
Cu	23.17	10.22	16.7	13.45	18.58	33.55	Cu	92.16	71.52	81.84	55.13	82.62	100
Zn	11.42	8.65	10.04	25.53	36.35	44.83	Zn	63.35	50.45	56.9	40.76	61.09	81.38
Mn	32.45	18.6	25.53	34.5	46.08	54.1	Mn	96.34	69.72	83.03	23.53	35.26	46.97
Avg.	16.47	9.78	13.13	15.17	24.11	50.53	Avg.	81.71	68.82	75.26	65.89	75.79	76.12
Exp.3	20 days			33 days	45 days	52 days	Exp.4	20 days			50 days	80 days	177 days
Cr	76.52	91.32	83.92	100	-	-	Cr	7.48	2.83	5.155	17.63	78.11	100
Co	97.16	100	98.58	100	-	-	Co	11.26	25.9	18.58	38.72	86.79	100
Ni	93.04	100	96.52	100	-	-	Ni	19.67	37.55	28.61	46.11	84.69	97.85
Cu	87.48	84.51	86	68.67	100	-	Cu	9.14	15.76	12.45	50.4	75.53	92.03
Zn	91.02	61.56	76.29	50.78	76.08	101.33	Zn	13.28	17.17	15.225	37.27	55.85	74.41
Mn	64.07	66.43	65.25	29.31	43.91	58.47	Mn	24.56	18.73	21.645	21.49	32.21	69.27
Avg.	84.88	83.97	84.43	74.79	73.33	79.9	Avg.	14.23	19.66	16.94	35.27	68.86	88.93

\*A+C= Removal efficiency (%) of HMs via (Anolyte+Catholyte), <sup>†</sup>ND=Numerical Data.

#### Estimation of remediation time for complete removal of HMs

We estimated the remediation time for the complete removal of HMs for four EKSR experiments using electrokinetic models with the help of the MATLAB program. In this study, we conducted the four EKSR Experiments for 20 days of operation and predicted further in Table 7 the numerical data between removal efficiency versus remediation time by solving electrokinetic equations with an encoding program in MATLAB. Numerical data predicted well correlated to the experimental data in the removal of HMs for four EKSR processes.

Exp.1 was conducted for 20 days and measured the removal efficiency of six HMs. It was noted that the average removal performance of six HMs Cr, Co, Ni, Cu, Zn, and Mn ions was close to 5%, 8%, 13%, 16%, 10%, and 25%, respectively. The encoded MATLAB program was executed for different time periods to find the complete removal of six HMs based on 20 days of experimental data.

The program was iterated for different time intervals 30, 50, and 200 days as remediation time and stopped the program after achieving the numerical data on the complete removal efficiency of six HMs. Numerical data analysis revealed that more than 200 days were required to remediate six HMs completely (Fig. 5).

Exp.2 was conducted for 20 days and measured the removal efficiency of six HMs. It was noted that the average removal performance of six HMs Cr, Co, Ni, Cu, Zn, and Mn ions is close to 82%, 90%, 58%, 82%, 57%, and 83% respectively. The encoded MATLAB program was executed for different time periods to find the complete removal of six HMs based on 20 days of experimental data. The program was iterated for different time intervals 33, 41, and 54 days as remediation time, and stopped the program after achieving the numerical data on the complete removal efficiency of six HMs. Numerical data analysis Numerical data analysis

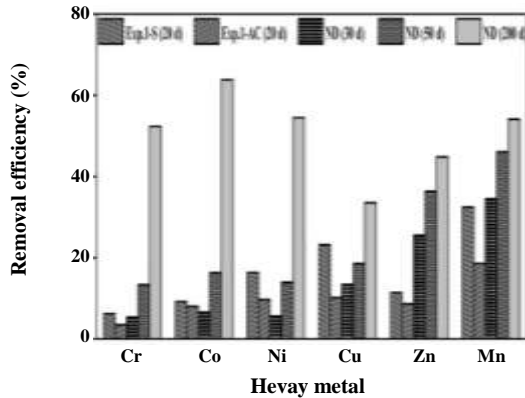


Fig. 5: Estimation of Remediation time for Complete Removal of HMs for Exp.1 and corresponding Numerical Data (ND) for 30,50 and 200 days.

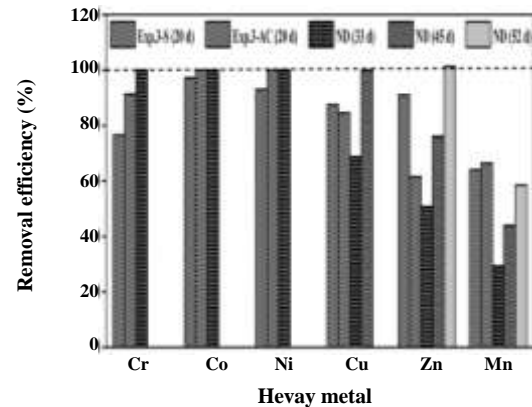


Fig. 7: Estimation of Remediation time for Complete Removal of HMs for Exp.3 and corresponding Numerical Data (ND) for 33,45 and 52 days.

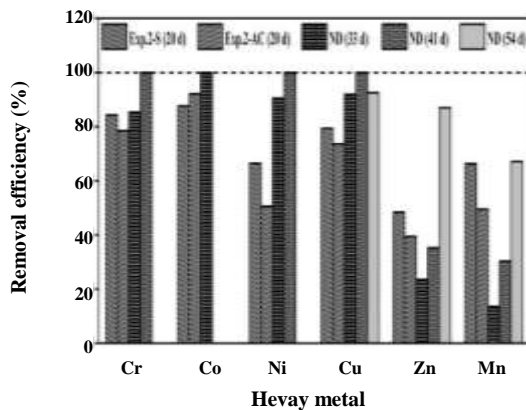


Fig. 6: Estimation of Remediation time for Complete Removal of HMs for Exp.2 and corresponding Numerical Data (ND) for 33,41 and 54 days.

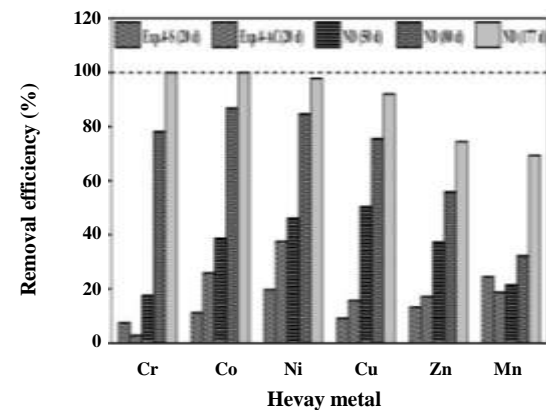


Fig. 8: Estimation of Remediation time for Complete Removal of HMs for Exp.4 and corresponding Numerical Data (ND) for 50, 80, and 177 days.

revealed that the removal efficiency of Co in 33 days, Cr/Ni in 41 days, Cu in 54 days, in the case of Zn and Mn ions required more than 54 days to remediate completely (Fig. 6).

Exp.3 was conducted for 20 days and measured the removal efficiency of six HMs. It was noted that the average removal performance of six HMs Cr, Co, Ni, Cu, Zn, and Mn ions was close to 84%, 98%, 96%, 86%, 76%, and 65% respectively. The encoded MATLAB program was executed for different time periods to find the complete removal of six HMs based on 20 days of experimental data. The program was iterated for different time intervals 33, 45, and 52 days as remediation time and stopped the program after achieving the numerical data on the complete removal efficiency of six HMs.

revealed that the removal efficiency of Cr/Co/Ni in 33 days, Cu in 45 days, Zn ions in 52 days, and Mn required more than 52 days to remediate completely (Fig. 7).

Exp.4 was conducted for 20 days and measured the removal efficiency of six HMs. It was noted that the average removal performance of six HMs Cr, Co, Ni, Cu, Zn, and Mn ions was close to 5%, 18%, 28%, 12%, 15%, and 21% respectively. The encoded MATLAB program was executed for different time periods to find the complete removal of six HMs based on 20 days of experimental data. The program was iterated for different time intervals 50, 80, and 177 days as remediation time and stopped the program after achieving the numerical data on the complete removal efficiency of six HMs. Numerical data analysis revealed that the removal efficiency of Cr/Co/Ni

in 177 days, Cu, Zn, and Mn ions required more than 177 days to remediate completely (Fig. 8).

## CONCLUSIONS

In this study, electrokinetic technology was applied and investigated the chelating agents-enhanced electrokinetic removal of HMs from granite waste soil. We find that an enhanced removal performance of HMs in chelating agents is more when unenhanced. Our study implemented the electrokinetic models for further experimental EKSR process and derived the numerical data between the removal performance of HMs versus remediation time. Moreover, we estimated the remediation time for the removal of HMs completely by solving electrokinetic models with an encoding program in the MATLAB software program. We experimentally investigated that the removal performance of HMs in chelating agents enhanced EKSR was about 6 to 7 times more than when unenhanced in 20 days of treatment. Furthermore, we estimated the remediation time about 52 to 54 days for the complete removal of HMs using electrokinetic models with help of MATLAB software program. Further studies can be focused on process optimization and scaling up the process to the pilot plant scale. We believe that electrokinetics might be useful to estimate the treatment time of the process to overcome the uncertainty in process optimization.

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