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THE ELECTROKINETIC IMPACT ON HEAVY METALS REMEDIATION OF TASIK CHINI IRON ORE MINE TAILINGS AT PAHANG STATE, PENINSULAR MALAYSIA

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Abstract— The improper disposal of mining tailings is a severe threat to the surrounding environment because it comprises high concentrations of heavy metals contamination. Any precious metal extraction (mining) produces millions of tons of waste; iron ore extraction is common globally, unlike other metal extraction. The iron ore tailings contain heavy metals such as Arsenic (As), Cobalt (Co), Manganese (Mn), Lead (Pb), Copper (Cu), and Zinc (Zn). This study focuses on extracting hazardous metals

Mining, Metal, Geotechnical, Electrokinetic Remediation Technique, ICP-OES Analysis	such as As, Co, Mn, Pb, Cu, and Zn from the disposed waste and improving its geotechnical properties. Nine samples were collected from Tasik Chini Iron ore mine, Pekan district, state of Pahang, Malaysia. The initial data were prepared for elemental analysis by following ICP-OES analysis. The results showed that As, Co, Mn, Pb, Cu, and Zn concentrations were exceeded the standard guidelines. The sustainable electrokinetic remediation techniques (EKR) method was applied to extract these metals from iron ore tailings specimens. A comprehensive approach of EKR shows an outstanding result where the highest removal efficiency of As was 68.4 %, Co 64.5%, Mn 67.8%, Pb 67.1%, and Cu was 64.1% and Zn 64.9% with the voltage gradient of 100 and 150 V for 4 and 8 hours constantly. Increasing the voltage gradient could be a cost-effective long-term solution for the remediation of iron ore tailings. The existing method was experienced as an effective and green technique for extracting heavy metals and recycling mining waste materials.

I. Introduction

Tasik Chini lake is located in Pahang State and is considered one of the largest natural water reservoirs in Malaysia with a total area of 69.22 km^2 as stated by Malaysian Government Gazette No 9, Vol 6, 1914); [1]. UNESCO declared Chini Lake the first status of Rizab Biosfera (Biosphere Reserve) in 2009, further Malaysian government developed this place as a safe eco-tourism destination for local and international tourists [1]. Around the lake, some illegal bauxite and iron ore mining activities were started, and the mining waste was deposited around the lake which consist of different toxic metals including As, Pb, Zn, Mg, etc. The rainwater carried these waste materials down to the lake which makes them contaminated. Most of the local communities depend on Tasik Chini lake as their main source of water supply for houses, and agricultural purposes, which could directly affect their health as well as it also harms aquatic flora and fauna [2-3]. This lake is not only an essential source for fish breeding, but it also plays a vital role in the surrounding agricultural activities. It has been declared that water is highly contaminated and not suitable for drinking purposes [4, 2]. The aquatic sediments are a significant compound entering the environment via chemical transformation which slightly contaminates the water [5-6]. The concentration of As, V, and Zn in Chini Lake sediment was analyzed as higher than the standard guideline by the Canadian Ministry of Environment (2009), Australian Department of Environment and Conservation 2010, Dutch list, Kelly indices, and Soil Quality Guidelines [1].

A. Heavy Metals Contamination – Malaysia

According to the Environmental Quality Act 1974 (EQA) and Contaminated Land Management and Control Guidelines, there is no specific explanation of contaminated land in the Environmental Quality Act 1974. The EQA defines soil and pollution. The soil is defined under the EQA to mean the earth, sand, rock, shale, minerals, and vegetation in the soil. Simultaneously, pollution is explained, which means any act or process, whether natural or anthropogenic activities resulting in the introduction of any pollution into the environment in the contravention of acceptable conditions as stated in the regulation ack under the EQA Malaysia has some highly contaminated sites due to anthropogenic sources such as industrial activities and domestic wastewater, mining, smelting, and agrochemicals are various anthropogenic entering points of heavy metals into agricultural and non-agricultural soil. Previously, some researchers have examined the heavy metal concentration in mining soil, sediments, and groundwater. Iron ore tailings are raw solid waste materials produced during iron ore extraction, due to high output and low utilization, iron ore tailings are considered one of the most common solid wastes in the world [7-8]. Iron is considered an essential metallic metal that plays a critical role in the modern era [9].

Aziman et al. [1] studied the As concentration was 43 – 56 mg/kg, Co 3 – 5 mg/kg, Pb 36 – 80 mg/kg, and Zn 61 – 301 mg/kg in Tasik Chini lake deposits was observed to exceed its recommended guidelines. Krishnankutty et al. [2] also conducted a study on the metals' chemical speciation in surface sediments' risk assessment due to their surroundings' mining activities. The result indicated that the 30 concentrations of As, Cd, Co Pb, and Ni were high in the mining area, which is considered a very high ecological risk category. These toxic metals are released near water and make it contaminated. Mn, Zn, and Cr concentrations were increased with high environmental risk, while Al and Fe revealed low to moderate ecological risk. The highest risk-possessing metals are particularly crucial because of their instability, transforming into the water and becoming more toxic for human health and the ecosystem [2].

Madzin et al. [10] studied heavy metal contamination in Bukit Ibam, and Kuala mining sites in Lapis, Pahang. The study was focused on heavy metals such as (Fe, Pb, Cr, Cd, Zn, Cu, Mn, As, Cd, Cr, and Ni). Bukit Ibam surface soil the concentration of Pb (lead) lowest value was measured at 96 mg/kg, and the highest value was measured at 2195 mg/kg according to the Canadian Councils of Ministers of Environment (CCME), 2011, the standard value for Pb is 35 mg/kg which highly exceeds then the expected value. The lowest value for Cu was measured at 67.5 mg/kg, and the highest value was 2845 mg/kg, which also exceeded 35.7 mg/kg of the (CCME 2011) expected value as shown in Table 1. The lowest value for Zn was measured at 1542.5 mg/kg, and the highest was 1682 mg/kg, while the standard value is 123 mg/kg (CCME 2011). Similarly, the Mn concentration was measured at 920.5 mg/kg and 5285 mg/kg, where the standard value is 400 mg/kg (CCME 2011). As concentration was measured between 54 mg/kg and 632.5 mg/kg, the standard value is 5.9 mg/kg (CCME 2011).

II. Methods and Materials

A. Site Description

Tasik Chini lake is considered the second largest freshwater reservoir in Peninsular Malaysia (3° 22′ 30" to 3° 28′ 00" N and 102° 52′ 40" to 102° 58′ 10" E), located in Mukim Penyor in Pekan district, Pahang state with the total land of 202 ha [11-13]. This lake is a biologically rich biosphere reserve history instigated by UNESCO. A small network of 12 lakes in the surrounding lake Chini covered 150 hectares (ha) in the dry season and approximately 300 ha in the rainy season, depending on the weather condition [14]. The lake serves as an important breeding and spawning ground for fish as it connects to the Pahang River through the Chini River. It is not only a livelihood for the native indigenous people nearby but also an important food source Excessive erosion of soil from nearby mining sites increases the sedimentation in the lake, therefore, high bioavailable metal concentrations are released which is harmful to the health of aquatic species in the freshwater. There are several active iron ore and bauxite mining activities in the Laut Jemerau area near Chini Lake. Deforestation has existed for bauxite and iron ore mining; thus, the soil has been eroded to Chini Lake as can be seen in Figure 1. Due to the surface mining activities, the soil has been directly washed to the Sungai Jemberau (Feeder River) and Laut Jemberau close to Chini Lake.

B. Sample Collection

A total of 9 soil samples were collected around Chini Lake as can be seen in Figure 2, which were analyzed further in the laboratory for chemical purposes as presented in Table 1. The soil samples were collected from 3 different points, each site with 3 specimens with a depth of 10 to 20 cm below the surface level using a sharp spade, hand fork, and trowel. The pH value was measured at the site by using a pH meter. The specimens were further stored in tight-capped polyethylene bags. The specimens were moved to the laboratory carefully for further analysis. Each specimen was air-dried until a constant mass is achieved. Further, the dried sample was pulverized by using a motorized grinder followed by sieving through a mesh of 500 μm before analysis.

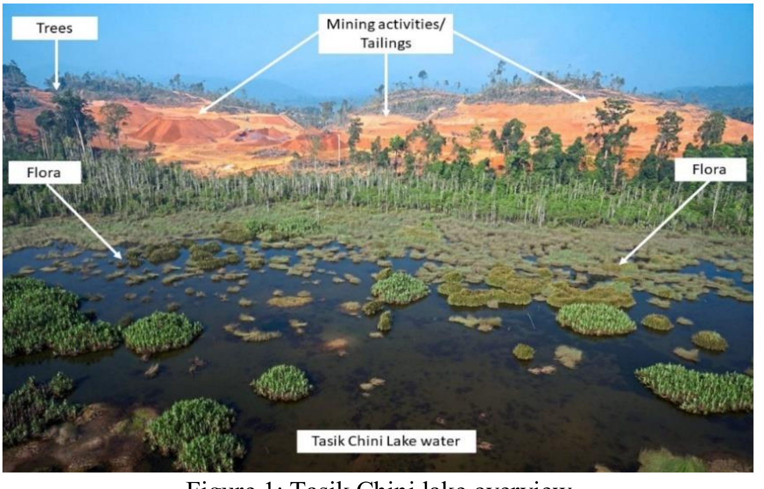

Figure 1: Tasik Chini lake overview

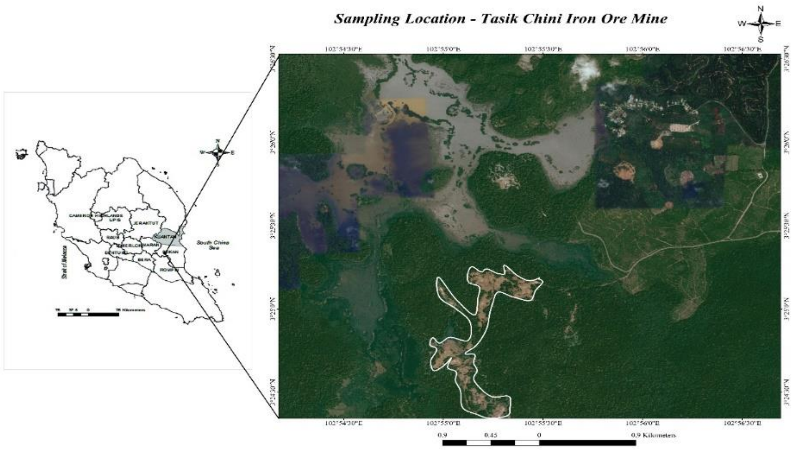

Figure 2: Sampling location Tasik Chini mining tailings

C. Sample Analysis

All the soil specimens were shifted to the laboratory carefully, where the samples were air-dried and grinded for chemical analysis. The concentration of As, Co, Mn, Pb, Cu, and Zn was determined by Inductively Coupled Plasma Optical Emission Spectroscopy (ICP-OES, Agilent 5800 ICP-OES) was performed to detect elemental identification.

D. Experimental Setup

The electrokinetic cell was designed for this experiment, the cell was made of an acrylic glass plate with a rectangle shape with dimensions of 30 cm in length, width, and height of 10 cm. The transparent acrylic plate is being used to prevent short-circuiting, monitor the soil level during consolidation, and monitor the water level. The EKR cell is divided into three significant portions: an anode, untreated soil, and cathode compartments. The anode and cathode section are 5 cm, while the untreated soil compartment is 15 cm, as shown in Figure 3. Two electrodes are inserted vertically into the soil compartment with 20 cm between each electrode. Aluminum was subjected to the anode and the cathode compartment to pass the direct current (DC) to the soil. The voltage gradient of 150 V is operated for the operational period of 8 hours for post-EKR phase I, while in phase II, the voltage gradient of 100 V was applied for 4 hours continuously.

Subsequently, putting the soil in the central section of the electrokinetic cell, the soil specimen is permitted to be placed in the cell for approximately 8 hours. The solidification process is carried out by putting a Perspex sheet on the soil specimen's exterior part. After that, a total load of about 100 kg is subjected to the Perspex sheet. During the soil specimen preparation, the shaking process is intended to homogenize the soil specimen and exclude air pockets. The purpose of solidifying the soil specimen is to remove metals from the load and further standardize the soil specimen. The soil is removed carefully from the EKR cell and placed in a polyethylene plastic bag. Later, the treated soil proceeded for geotechnical properties and elemental analysis.

III. Results and Discussion

A. Pre and Post-Chemical Analysis

The results show the concentration of As, Co, Pb, Mn, Cu, and Zn highly exceeded the recommended guidelines by the Canadian Council of Minister of Environment (2001), Canadian Ministry of Environment (2009), Pre-Industrial Level, Crust Average, Threshold level, Soil Quality Guidelines (SQG), and Soil Quality Guidelines for Environmental Health (SQGEH) as can be seen in Table 1. The pre-elemental composition analysis for untreated soil samples was performed to evaluate the heavy metal composition/concentration in iron ore and bauxite mine tailings soil around Chini lake. The pH value was measured at 7.0 during the soil sampling analysis. While inductively coupled plasma mass spectrometry ICP-OES analysis was performed at Jabatan Mineral dan Geosains (JMG) laboratory, Kuantan, Pahang, Malaysia.

Table 1: Heavy metals concentration in Tasik Chini iron tailing

Location	Heavy metals Concentration mg/kg					
(Tasik Chini Iron Ore)	As	Co	Mn	Pb	Cu	Zn
Fresh Tailing Sample	99.1	61.62	2516.00	116.53	204	310.50
	↑	↑	↑	↑	↑	↑
(Tasik Chini Iron Ore) Abandoned Tailing Sample	99.0	76.86	2903.00	127.70	98	423.60
	↑	↑	↑	↑	↑	↑
	Standard Guidelines					
Canadian Council of Minister of Environment (2001)	5.9	*	*	35	35.7	123
Canadian Ministry of Environment (2009)	11	*	*	45	63	290
Pre-Industrial Level	15	*	*	70	50	175
Crust Average	2	*	527	70	50	175
Threshold level	5.9	*	*	35	36.7	123
Soil Quality Guidelines (SQG)	30	40	*	70	63	200
Soil Quality Guidelines for Environmental Health (SQGEH)	12	*	*	*	63	200

The concentration of Arsenic (As) in fresh untreated Tasik Chini Iron ore tailings was recorded at 99.1 mg/kg and 99.0 mg/kg in abandoned mine tailing. According to the Canadian Council of Ministers of Environment (2011) standard value of As is 35 mg/kg, the Canadian Ministry of Environment (2009) 11 mg/kg, the Pre-Industrial Level is 15 mg/kg, the Crust average is 2 mg/kg, Trash hold value is 5.9 mg/kg, Soil Quality Guidelines is 30 mg/kg and Soil Quality Guidelines for Environmental Health (SQGEH) is 12 mg/kg [10].

The Cobalt (Co) concentration was also measured exceeded the recommended value, where it was observed at 61.62 mg/kg for fresh tailings while it was observed at 76.86 mg/kg for abundant Tasik Chini tailing. The Cobalt concentration in soil is 40 mg/kg recommended by Soil Quality Guidelines where it was observed to exceed its suggested value.

Manganese (Mn) is considered the 12th most abundant element in the earth's crust which is comprising approximately 0.1% and is the 5th most plentiful metal [15]. The heavy metals concentration of Mn in Chini Lake fresh iron ore tailing was measured at mg/kg while in the abandoned untreated tailing the concentration of Mn was 2903.00 mg/kg. The concentration of Mn exceeded the standard guidelines recommended by the Predicted Effects Level Background for Mn is 500 mg/kg. The Pb concentration was measured at 63.5 mg/kg and 72.5 mg/kg, while the Canadian Council of Ministers of Environment (2011) standard value is 35 mg/kg. The Cu concentration was measured from 97 mg/kg to 165.5 mg/kg, while the standard value is 35.5 mg/kg.

Lead (Pb) was also detected in Tasik Chini iron mine tailing, where the concentration of Pb in fresh tailing was measured at 116.53 mg/kg and 127.70 mg/kg in abundant iron mine tailing. According to the Canadian Council of Minister of Environment (2001), the acceptable value for Pb is 35, the Pre-Industrial Level recommended value is 70 mg/kg, and the Threshold level acceptable range is 35 mg/kg, while Soil Quality Guidelines (SQG) recommended value is 70 mg/kg.

Copper (Cu) concentration in measured in Tasik Chini Iron fresh tailing which was 204 mg/kg whereas it was observed at 98.0 mg/kg in abandoned mine tailings. The recommended value is 35.7 by the Canadian Council of the Minister of Environment (2001), the Pre-Industrial Level acceptable value is 63 mg/kg and the Soil Quality Guidelines (SQG) recommended value is 63 mg/kg. The concentration of Zinc (Zn) was observed to exceed its recommended guidelines, where the Zn concentration was 310.50 mg/kg in fresh tailing while it was measured at 423.60 mg/kg in abundant tailing. Canadian Council of the Minister of Environment (2001) suggested acceptable value for Zn in the soil is 123 mg/kg, Soil Quality Guidelines for Environmental Health (SQGEH) acceptable value is 200 mg/kg.

B. Post Chemical Analysis

The chemical analysis was performed in two phases where, in the first phase, the voltage gradient of 100 V was applied for 4 hours while in the second phase, the voltage gradient of 150 volts was applied for 8 hours through the aluminum electrode. It was observed that the heavy metals concentration was decreased with charge particles moment during electrokinetic remediation in both phases. In the first phase, the concentration of As was decreased from 99.1 mg/kg to 20.13 mg/kg in fresh tailings and 9.40 mg/kg in abandoned tailing, while in the second phase it was reduced to 9.34 mg/kg to 3.10 mg/kg. The concentration of Co was observed to exceed its limit for untreated tailing where the concentration of Co was reduced to 0.12 mg/kg and 0.01 mg/kg and 0.1 mg/kg in the second phase. Mn concentration was also observed to reduce with an electric gradient, it was reduced from 2903.0 mg/kg to 700.47 mg/kg and 401.84 mg/kg in the first phase while in the second phase it was decreased to 409.84 mg/kg and 218.38 mg/kg. Similarly, the Pb concentration was decreased with EKR, where it was reduced from 127.70 mg/kg to 80.46 mg/kg and 42.21 mg/kg in the first phase while in the second phase with an applied voltage gradient of 150 v for 8 hours it was reduced to 31.59 mg/kg and 28.49 mg/kg. Cu concentration was observed to be reduced from 204 mg/kg to 71 mg/kg and 34.59 mg/kg in the first phase and 32.45 mg/kg and 28.49 mg/kg in the second phase. Similarly, the Zn amount was observed to exceed its limit in Tasik Chini tailings, where it was reduced with applied the voltage gradient in both phases, in the first phase it was reduced from 423.60 mg/kg to 309.93 mg/kg and 119.80 mg/kg in the first phase while in the second phase it was observed reduce to 113.68 mg/kg and 101.82 mg/kg as can be seen in Table 2. and 3.

IV. Conclusions

This study was conducted to detect the heavy metals concentration in Tasik Chini iron ore mine fresh and abandoned tailings high risk for aquatic, agriculture, and public health. According to Soil quality guidelines, a higher concentration of As, Co, Mn, Pb, Cu, and Zn was detected in Tasik Chini iron tailings. The Tasik Chini lake water is highly contaminated due to the high concentration of As, Co, Mn, Pb, Cu, and Zn. These metals were considered the most toxic causing health issues. The water is not suitable for drinking, due to the movement of the toxic metal toward the lake. A comprehensive approach of EKR shows an outstanding result where the highest removal efficiency of As was 68.4 %, Co 64.5%, Mn 67.8%, Pb 67.1%, and Cu was 64.1% and Zn 64.9% with the voltage gradient of 100 and 150 V for 4 and 8 hours constantly. The EKR technique is highly recommended for metal extraction.

Table 2: Post-EKR Heavy metals concentration in Tasik Chini iron tailing Phase I

Location	Heavy metals Concentration mg/kg											
	As		Co		Mn		Pb		Cu		Zn	
	Pre-EKR	Post-EKR	Pre-EKR	Post-EKR	Pre-EKR	Post-EKR	Pre-EKR	Post-EKR	Pre-EKR	Post-EKR	Pre-EKR	Post-EKR
(Tasik Chini Iron Ore) Fresh Tailings	99.1	20.1	61.62	0.12	2516.0	700.47	116.53	80.46	204	71	310.50	309.9
(Tasik Chini Iron Ore) Abandoned Tailings	99. 0	9.40	76.86	0.01	2903.0	401.84	127.70	42.21	98	34.5	423.60	119.8
Standard Guidelines												
Canadian Council of Minister of Environment (2001)	5.9		*		*		35		35.7		123	
Canadian Ministry of Environment (2009)	11		*		*		45		63		290	
Pre-Industrial Level	15		*		*		70		50		175	
Crust Average	2		*		527		70		50		175	
Threshold level	5.9		*		*		35		36.7		123	
Soil Quality Guidelines (SQG)	30		40		*		70		63		200	
Soil Quality Guidelines for Environmental Health (SQGEH)	12		*		*		*		63		200	

Table: 3: Post-EKR Heavy metals concentration in Tasik Chini iron tailing Phase II

Location	Heavy metals Concentration mg/kg											
	As		Co		Mn		Pb		Cu		Zn	
	Pre-EKR	Post-EKR	Pre-EKR	Post-EKR	Pre-EKR	Post-EKR	Pre-EKR	Post-EKR	Pre-EKR	Post-EKR	Pre-EKR	Post-EKR
(Tasik Chini Iron Ore) Fresh Tailings	99.1	9.34	61.62	0.01	2516.0	409.8	116.53	31.5	204	32.4	310.50	113.6
(Tasik Chini Iron Ore) Abandoned Tailings	99. 0	3.10	76.86	0.1	2903.0	218.3	127.70	28.4	98	28.4	423.60	101.8
Standard Guidelines												
Canadian Council of Minister of Environment (2001)	5.9		*		*		35		35.7		123	
Canadian Ministry of Environment (2009)	11		*		*		45		63		290	
Pre-Industrial Level	15		*		*		70		50		175	
Crust Average	2		*		527		70		50		175	
Threshold level	5.9		*		*		35		36.7		123	
Soil Quality Guidelines (SQG)	30		40		*		70		63		200	
Soil Quality Guidelines for Environmental Health (SQGEH)	12		*		*		*		63		200	

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