DETERMINATION OF ARSENIC, LEAD AND MERCURY IN CONTAMINATED WATER VIA ELECTROKINETIC PROCESS

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DEDICATION

This thesis is dedicated to my family and my friends.

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ABSTRACT

Electrokinetic remediation is a method to remove heavy metals such as arsenic (As), lead (Pb) and mercury (Hg) by application of a constant direct current connected to the cathode and anode in the electrolyte solution. This experiment is conducted to remove the heavy metals in contaminated water under controlled time, types of electrodes, distance between electrodes and voltage applied in a laboratory scale. The parameters considered in electrokinetic experiments were zinc (Zn), aluminium (Al) and stainless steel as types of electrodes, electrodes spacing of 3 cm, 7 cm, and 9 cm, and the applied voltage of 30 V, 60 V, and 90 V in one hour treatment. The heavy metals were then extracted with several series of electrokinetic experiments to obtain the maximum removal of the metals. Heavy metal depositions in both electrodes were analysed using X-ray fluorescence (XRF) spectroscopy. The concentrations of heavy metals in the simulated contaminated water were analysed using inductively coupled plasma - optical emission spectroscopy (ICP-OES). It was found that the stainless steel as electrode, 5 cm for the distance between electrodes and 90 V for the voltage provide the optimal condition. When the optimal parameters were applied to contaminated water of Sungai Masai, Johor, the study found the presence of As and Hg at 4.46 ppm and 2.08 ppm concentration, respectively, but Pb was not detected. Later, all Hg was removed in the electrokinetic remediation experiment. In summary, voltage, spacing and type of electrode affect the heavy metals removal in contaminated water.

ABSTRAK

Pemulihan elektrokinetik adalah satu kaedah untuk menyingkirkan logam berat seperti arsenik (As), plumbum (Pb), dan merkuri (Hg) dengan menggunakan arus terus malar yang disambungkan kepada katod dan anod dalam larutan elektrolit. Eksperimen ini dijalankan untuk menyingkirkan logam berat dalam air tercemar di bawah masa terkawal, jenis elektrod, jarak antara elektrod dan voltan yang digunakan dalam skala makmal. Parameter yang dipertimbangkan dalam eksperimen elektrokinetik adalah zink (Zn), aluminium (Al) dan keluli tahan karat sebagai jenis elektrod, jarak antara elektrod iaitu 3 cm, 7 cm dan 9 cm, dan voltan yang digunakan iaitu 30 V, 60 V dan 90 V dalam rawatan satu jam. Logam berat kemudian diekstrak dengan beberapa siri eksperimen untuk memastikan penyingkiran maksimum logam. Pengendapan logam berat di kedua-dua permukaan elektrod dianalisis menggunakan spektroskopi pendarfluor sinar-X (XRF). Kepekatan logam berat dalam simulasi air yang tercemar dianalisis menggunakan spektroskopi pemancaran optik - plasma berganding aruhan (ICP-OES). Ia didapati bahawa keluli tahan karat sebagai elektrod, 5 cm untuk jarak antara elektrod dan 90 V untuk voltan memberikan keadaan optimum. Apabila parameter optimum digunakan terhadap air tercemar di Sungai Masai, Johor, kajian mendapati kehadiran As dan Hg masing-masing pada kepekatan 4.46 ppm dan 2.08 ppm, tetapi Pb tidak dikesan. Kemudian, semua Hg disingkirkan dalam eksperimen pemulihan elektrokinetik. Kesimpulannya, voltan, jarak dan jenis elektrod mempengaruhi penyingkiran logam berat dalam air tercemar.

TABLE OF CONTENTS

LIST OF TABLES

LIST OF FIGURES

LIST OF ABBREVIATIONS

BOD	-	Biochemical oxygen demand
EQS	-	Environmental Quality Standards
FAO	-	Food and Agricultural Organization
ICP-OES	-	Inductively Coupled Plasma Optical Emission Spectroscopy
NWQMP	-	National Water Quality Monitoring Program
NWQS	-	National Water Quality Standard
RO	-	Reverse osmosis
UF	-	Ultrafiltration
UIRL	-	University Industry Research Laboratory
WHO	-	World Health Organization
XRF	-	X-ray fluorescence

LIST OF SYMBOLS

g	-	gram
kg	-	kilogram
cm	-	centimeter
m	-	meter
mm	-	milimeter
mg	-	miligram
ml	-	mililiter
L/l	-	liter
Al_2O_3	-	Alumium oxide
H^+	-	Hydrogren ion
OH^-	-	Hydroxyl ion
H_2O	-	Water
H_2O_2	-	Hydrogen peroxide
HPO_3	-	Metaphosporic acid
HNO_3	-	Nitric acid
C_2H_6O	-	Ethanol

LIST OF APPENDICES

CHAPTER 1

INTRODUCTION

1.1 Overview

Poor water quality becomes a major threat to human health and other living organism. In this country, mostly the source of water pollution is in the industrial, domestic area and industrial activities. Even though some action is taken, it still has not solved the issues. As a responsible community, an action must be taken to make sure the water pollution is not worse from time to time.

Water pollution is defined when the quality of water in terms of biological, physical or chemical changes affect the living organisms in a bad way. Unsafe water supply is one of the biggest issues that scared the human being as water is a necessity in daily life. Without clean water, many problems will arise. Once the water is polluted it will spread through the other places.

Water pollutants consist of heavy metals, microorganisms, bacteria and thousands of toxic organic compounds. For this research, heavy metals are the most concern subject to study where the residents in that area are exposed to health hazards cause by water pollution (Danuta et al., 2006).

Heavy metals are toxic and non-biodegradable elements and can accumulate in the living things (Zayadi et al., 2016; Abdullah et al., 2014). These elements are causing a threat to the environment through human activities such drinking of contaminated water with heavy metals. The concentration of heavy metals in natural water resources has been the subject of research for decades (Raeisi et al., 2014). Some of the heavy metals are necessary in small quantities for biological viability such as zinc (Zn), manganese (Mn), copper (Cu) and iron (Fe) but heavy metals such as arsenic (As), lead (Pb), mercury (Hg) and cadmium (Cd) has become hazardous to the human body when exposed with high dose of concentration (Barka et al., 2001). In fact, heavy metals are taken into the body through consumption of food, inhalation and also skin absorption. These actions may lead to serious health effects such as allergic reaction, decreases metabolic rate and cardiovascular disease (Tchounwou et al., 2012).

During the past decades, many treatment techniques have been introduced for the efficient removal of the contaminant of heavy metals, organic and inorganic compounds and other hazardous materials from groundwater, soil and associated ecosystems (Krishna et al., 2009). Many attempts are being made to treat the polluted water with mixed contaminants for both inorganic and organic contaminants. There are several techniques in a range of both in-situ and ex-situ treatment such as nanofiltration membranes, which is an intermediate process between ultrafiltration (UF) and reverse osmosis (RO) (Ortega et al., 2008), hydrocolloid liquid-core capsules (Nussinovitch et al., 2015), chemical precipitation such as hydroxide and sulfide precipitation (Fu et al., 2011) and biosorption system (Amirna et al., 2015). To date, ion exchange (Kang et al., 2004), and electrokinetic remediation (Embong et al., 2017) techniques are the most frequently studied for heavy metal removal treatment. Electrokinetic remediation studies in Malaysia are limited and there is not much research has been done for the past few years.

Electrokinetic remediation is one of the promising technologies to treat the contaminated water by heavy metals (Albert et al., 2011; Embong et al., 2015; Rosestolato et al., 2015; Embong et al., 2017) that can be carried out either ex-situ or in-situ. For a decade, electrokinetic technology has involved large company's investment as the electricity supply is expensive. Hence, due to high investment, it has not been widely applied yet in Malaysia. In this research, the small scale of apparatus and equipment are proposed using an electrokinetic technique which is useful and economical for low concentration of heavy metals. Therefore, the main aim of the study is to improve a system of heavy metals remediation in contaminated water in the river by using electrokinetics method.

1.2 Problem Statement

In Malaysia, electrokinetic remediation technique has not been introduced widely. A study has been conducted using these techniques in Sri Gading Industrial Area, Batu Pahat, Johor where the place is chosen due to high levels of radiation background in soil (Johar et al, 2015). Also, Embong et al. (2017) has conducted a study on Zn(II) removal from Sembrong river, Johor is using electrokinetic remediation. This electrokinetic method is needed to treat the groundwater, sediment and soil with the polluted critical condition.

For this research, Sungai Masai has been chosen as a place to do the experiment where local residents use as the main route for fisherman to the jetty or sea (Berita Harian, 2018). Moreover, in a rural area people are exposed to the risk of health hazard where the water supply is contaminated, especially by heavy metals (Akoto et al., 2014). Heavy metals must be removed from water resources before it is released to the

environment. To overcome this problem, the planning, design, operation of the remediation process of removal of heavy metals should be considered as the main concerned with this problem. Therefore, in this work, an alternative remediation process needs to perform, known as electrokinetics.

The purpose of the experiment is to explore if an electrokinetic remediation system is applicable to use in river water. The system is simulated by varying several parameters involved to obtain the optimum efficiency. This will provide new data concerning an electrokinetic remediation process for river water from Sungai Masai, Johor.

1.3 Objectives

The objective of this research is:

i. To determine optimized parameters of the electrokinetic system efficiency for heavy metals remediation in terms of treatment time, applied voltage, type of electrodes, and the distance between electrodes.
ii. To measure the amount of heavy metal removed from simulated contaminated water between pre and post electrokinetic process by using X-ray fluorescence spectroscopy (XRF) and inductively coupled plasma optical emission spectroscopy (ICP-OES).
iii. To determine the electrokinetic efficiency for heavy metals removal from contaminated water in Sungai Masai, Johor.

1.4 Scope of Study

The ex-situ experiment is conducted to optimize the efficiency of the electro-kinetic system for heavy metals remediation in various conditions such as treatment time, applied voltage, type of electrodes and distance between electrodes. The system is stimulated by varying the several parameters involved to obtain the optimum efficiency of the system. The study has been done in a longer period of time to get the highest percentage of removal in low potential voltage (Yuan et al., 2003). In these experiments, we test the sample with higher potential voltage in a shorter period. Also, the samples of contaminated water are self-prepared where the distilled water is contaminated with a fixed amount of heavy metals with known concentration. The concentration of heavy metals is set higher than WHO (2008) limits to find out the highest percentage of removal of heavy metals these experiments will achieve. The heavy metals concerned in this study are mercury (Hg), arsenic (As), and lead (Pb). These heavy metals are label as high toxic elements and can affect human health if ingested (Owen, 2012). In addition, the deposition of heavy metals in the cathode and anode is analyzed by XRF and the concentration of heavy metals in contaminated water is analyzed by ICP-OES. Then, the actual contaminated water samples were collected at several areas at river in Sungai Masai, Johor.

1.5 Significance of Study

Exposure to heavy metals can take place in different ways, such as inhalation, ingestion and dermal contact. Therefore, the food and drinking water become the important routes of exposure to the human body. Water, which is a possible route of exposure, comes from two sources such as surface water (e.g. rivers and reservoir) and groundwater (Fawel et al., 2003). Lacks of information about the area of contaminated water in Malaysia become one of the problems with authority to take actions. Besides, levels of exposures are unknown, and the risks to health are uncertain as a result of lack of information of the research in this area.

The data findings from this work, will provide information on health hazard caused by how much heavy metals present in contaminated water and how effective this electrokinetic remediation technique to remove the heavy metals in the research area. This issue is one of the important research fields because it involves the resident's health and the environmental problems in the affected area. Jauharah (2010) reported that many cases involving the effect of heavy metals on the human health such as diarrhea, sensory disturbances, kidney and lung damaged, and may cause fatal. Hence, the findings of this work hopefully will help to improve the efficiency of the existing water treatment system in a particular area through implementation of the suitable technology.

1.6 Outline of the Thesis

This thesis consists of five chapters. Chapter 1 introduces the research, including the problem's statement, objectives of the study, scope and significance of the study. Chapter 2 is a literature review of this research. Few main topics are described in term of basic knowledge such as type of heavy metals; distributions of heavy metals research in worldwide and in Malaysia, and the theory and how the electrokinetic system operated. In Chapter 3, the preparation of samples for the collection, storage, testing and analyses of the samples was described. Also, the instruments used in this research are elaborated briefly. Chapter 4 was focused on collecting data and analysis of the solutions and electrodes. Both samples, electrodes and contaminated water have been analysed using XRF and ICP-OES and the results are obtained. From the data, the percentages of heavy metals removed from the contaminated water are analysed and are predicted for the next research. Chapter 5 is the last chapter of the study. Conclusions are drawn and some recommendations for future study are given.

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