REMOVAL OF HEAVY METAL IN MEDICAL INSTUTIONAL WASTEWATER USING ULTRASONIC ASSISTED VARIOUS LEACHING SOLUTION

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NUR SYUHADA BINTI OMAR

A report submitted in partial fulfillment of the
requirement for the award of degree the of
Bachelor of Chemical Engineering

Faculty of Chemical & Natural Resources Engineering
Universiti Malaysia Pahang

JANUARY 2011

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I dedicate this to:

My parents:
En Omar bin Ismail and Pn Rohaya binti Mansor
My siblings:
Nur Safawiah, Arsyaddul Ibad, Nur Saiyidah and Alawiyah
and
My supervisor and friends.

Without their patience, understanding and support and most of all love,
the completion of this work would not have been possible.

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ABSTRACT

Daily, hospital generates a significant amount of wastewater which is more than the consumption of domestic water. The hospital wastewater may contaminate with micro-organisms, heavy metal, toxic chemicals and radioactive elements. In fact, the hospital wastewater is generating hybrid wastewater at the same moment domestic, industrial and effluents of care and medical research. Throughout this research our main concern is to remove targeted heavy metal that found to contain in the hospital wastewater, Pb and Ni. The wastewater will be treated using the ultrasonic technology assisted with several of leaching solution, HNO_3, citrate and EDTA. Ultrasonic process has been found as reliable solution which applies intense, high frequency sound to liquids, producing intimate mixing and powerful chemical and physical reactions. The best performance of these leaching solutions (HNO_3, citrate and EDTA) will be studied through the main parameter such as type of solution, sonication time and concentration of the solution. The sample obtained from the hospital is filtered and spike with amount of targeted heavy metal prior to ultrasonic process and AAS analysis. As the result, the best time to remove Pb is 3 minutes while 6 minutes for Ni removal. The suitable concentration to remove Pb is 0.06 M while 0.1 M for Ni. The percentage removal is 54% for Pb and 35.93% for Ni.

ABSTRAK

Setiap hari, hospital menghasilkan sejumlah air kumbahan yang besar berbanding yang dihasilkan dari penggunaan domestik. Air kumbahan dari hospital dicemari dengan mikro-organisma, logam berat, bahan kimia toksik dan unsur-unsur radioaktif. Bahkan, komposisi air kumbahan dari hospital adalah sisa hibrid yang sama seperti sisa domestik, sisa industri serta sisa dari kajian rawatan dan perubatan. Sepanjang kajian ini perhatian utama kami adalah untuk menyingkirkan logam berat, Pb dan Ni yang ditemui dalam air kumbahan hospital. Air kumbahan dari hospital ini akan dirawat dengan menggunakan teknologi ultrasonik dibantu dengan beberapa larutan, HNO_3, sitrat dan EDTA. Proses ultrasonik telah ditemui sebagai penyelesaian yang berlaku kuat, getaran frekuensi tinggi ke atas cecair serta menghasilkan percampuran bahan kimia secara tindak balas fizikal dan kimia. Prestasi optimum daripada larutan-larutan (HNO_3, sitrat dan EDTA) akan dikaji melalui parameter utama seperti jenis larutan, masa sonikasi dan kepekatan larutan. Sampel yang diperoleh dari hospital akan ditapis dan 'spike' dengan jumlah logam berat yang disasarkan sebelum proses ultrasonik dan analisis AAS. Sebagai hasilnya, waktu optimum untuk menyingkirkan Pb adalah 3 minit sementara 6 minit untuk menyingkirkan Ni. Kepekatan sesuai untuk menyingkirkan Pb 0.06 M sedangkan 0.1 M untuk Ni. Peratus penyingkiran adalah 54% untuk Pb dan 35,93% untuk Ni.

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EDTA	-	Ethylenediaminetetraacetic acid
HNO3	-	Nitric acid
AAS	-	Atomic Adsorption Spectrometer
Pb	-	Lead
Ni	-	Nickel
ml	-	Milliliter

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CHAPTER 1

INTRODUCTION

1.1 Background of study

The increasing of various illness and virus worldwide had led to innovation of new remedies and their consumption in medical institution nowadays. Research had been done widely to produce new medicine and pharmaceuticals product. The chemical used during the research is channeled out of the hospital may contaminate the environment. Typically, health care waste consists of both organic and inorganic substances including pathogenic microorganism (Ekhaise and Omavwoya, 2008). Amount of antibiotics, cytostatics and psycho tropics are among the drugs that had been swallowed and excreted by the patient might find their way into the sewer (sn daily). The use of many types of chemical to decontaminate the tools and equipments in hospitals are contributing to heavy metal content disposed in domestic water and sites. Medical waste in particular has increased greatly because hospitals use relatively more disposable items, such as syringes, cutlery, food trays, bed pans, etc. (Morrison, 1992). Source of hospital wastewater can be originated from clinical laboratories, research laboratories, medical waste incinerators and hospital laundries (Niumpang, 2006). The activities from the hospital facility indirectly release many toxic substances into the aquatic environment (Emmanuel et al., 2004). The hospital wastewater contains with hazardous medical waste which posses serious environmental health risk due to their carcinogenic natural (Ekhaise and Omavwoya, 2008), pathogenic microorganisms,

pharmaceutical partially metabolized, radioactive element and pollutants such as anti-tumor agent, antibiotics and organohalogen compound (Rezaee et al., 2005). Several studies had been done on hospital wastewater treatment. It has been studied the removal efficiency of pollutants and significant removal of pathogenic bacterial using an integrated anaerobic aerobic fixed film bioreactor (Rezaee et al., 2005). The effect of addition of disinfecting agent in hospital wastewater also has shown in reducing the water bacteria pollution and give rise to toxicity effects on aquatic organisms (Emmanuel et al., 2004). The hospital waste has been studied to pose negative effect on the microbiological and physiochemical parameters on the environment (Ekhaise and Omavwoya, 2008). However, the specific study on removal heavy metal from hospital wastewater is not founded.

Numerous studies had been done on heavy metal removal. Various of method had been used to remove heavy metal. Phytoextraction is one of the methods to remove heavy metal from municipal swage sludge in which plants uptake heavy metals from the soil sludge, followed by harvesting the aboveground biomass. In absence of chemical used for extraction, the phytoextraction effectiveness of Pb, Cd and Zn can be enhanced (Pogrzeba et al.). PCB wastewater is treated using sulfuric acid to remove heavy metal in treating the sludge generated from the wastewater treatment plant (Kuan et al., 2009). Instead of sulfuric acid, the nitric acid also had been used in extraction from sewage sludge which is proved that extraction efficiencies of heavy metals increased with increasing sonication time and acid concentration (Deng et al., 2009). In acid treatment of sludge, the sludge can be dissolved and then exist in solution. As the acid concentration, temperature and contact time increased, the percentage of extraction also increased (Stylianou et al., 2007). Another method to remove heavy metal is by remediation. Examples of remediation process such as washing, electrochemical remediation, flotation and ultrasonic-assisted extraction. The limitation of remediation technology is that they are time consuming (Peng et al., 2009)

Ultrasound excites high-energy acoustic cavitation which results in formation, growth and implosive collapse of bubbles in liquid. Intense heating of the bubble occur

during the cavitational collapse. The impact is sufficient to melt most metal. With the solvent assisted the ultrasonication, this will result in localized high shear forces which can remove the material adhering to particle surface (Peng et al., 2009). The removal efficiency of Ni ions from Ni-EDTA complex is more rapid when using Fenton reagent assisted with ultrasonic treatment and result in more Ni ion is released (Fu et al., 2009). In ultrasonic-assisted acid digestion (UAD), the sonication time is reduced from 3 to 4 hour of conventional wet acid digestion method to 20-30 min of UAD and low volume of acid consumption (Kazi et al., 2009). The ultrasonic-assisted leaching method (ULM) of trace metals from sediments also proved that the use of ultrasonic leaching method reduce the time of conventional leaching from 12 hr to 25 min (Elik, 2007). In recovery of copper and iron from PCB waste sludge, higher recovery efficiency of the ultrasonic assisted metal recovery process had achieved which is copper recovery efficiency of 95.2-97.5% and iron recovery efficiency of 97.1-98.5%. This technology is capable of providing higher separation and recovery efficiencies, lower recovery cost, higher quality product and zero process waste emission (Xie et al., 2009).

Ultrasonic-assisted extraction has been proved by many of studies to be a promising technology in heavy metal removal since it is an expeditious, inexpensive and efficient alternative to conventional extraction technique as demonstrated by application to both organic and inorganic analytes in a wide variety of sample (Elik, 2007). Ultrasonic is a preferred extraction technique because of its cavitation effect that in effect “cold boiling” and result in creation of the chemical and physical reactions such as surfactancy. The intensity of cavitation accelerates physical and chemical reactions and not the power applied to the system (Hwang et al., 2007). Besides, ultrasonic provides cost effective way to recover heavy metal from industrial waste sludge and reduces public health risk (Xie et al., 2009).

1.2 Problem Statement

The amount of hospital wastewater generates daily increase. The effluents that also contain many harming substances might pose a risk and environmental issue. Less study had been done specifically focusing on heavy metal removal in hospital wastewater. The previous study is focusing on biological treatment of the wastewater and removal of pathogenic bacterial (Rezaae et al., 2009). Other study on analysis microbiological and physiochemical analysis of the hospital wastewater (Ekhaise and Omavwoya, 2008) but it is not provided with the appropriate method to treat the heavy metal contain in the hospital wastewater. The heavy metals detoxification from medical waste fly ash had been done using supercritical water in addition of hydrogen peroxide, H_2O_2 (SCWH). SCWH treatment is believed to be effective alternative for hazardous elements detoxification in medical waste fly ash (Bo et al., 2009).

Previous study had used numerous type of chemical prior to sonication in heavy metal removal and had shown a significant removal. Ultrasound assisted solvent had proven that ultrasound alone is not effective enough to remove heavy metal. In the study of MTBE degradation by ultrasonic-assisted chemical process (UACP), it has shown that in the present of Fenton reagent and ultrasonic cavitation, UACP can effectively degrade MTBE with efficiency of 78% and only require low dosage of hydrogen peroxide to initiate the reaction (Chang, 2006). With the aid of nitric acid and hydrogen peroxide prior to sonication, this can give maximum extraction in the shortest possible time in which the time of sonication is reduced to 15 to 20 minutes compare with conventional single extraction, 16 hour (Arain et al., 2009).

1.3 Objectives

The proposed research was studied to find the appropriate leaching solution (EDTA, HNO3 and citrate) in ultrasound-assisted leaching solution to remove Pb and Ni from hospital wastewater.

1.4 Scopes of Study

In order to achieve the objectives, the following scopes have been studied

i. To study the effect of type of solvent and compare the removal of Pb and Ni at same concentration of EDTA, HNO3 and citrate.
ii. To study the effect of sonication time within a range at same concentration of EDTA, HNO3 and citrate.
iii. To study the effect of various concentration of the leaching solutions (EDTA, HNO3 and citrate) in removal Pb and Ni

CHAPTER 2

LITERATURE REVIEW

2.1 Ultrasonic Treatments

Ultrasonic can be define as vibrational or stress waves in elastic media that have a frequency above 20 kilohertz, the highest frequency of sound waves that can be detected by the human ear. They can be generated or detected by piezoelectric transducers (*see* piezoelectricity). In real media, the propagation of ultrasonic waves is accompanied with their attenuation dependent on geometrical factors such as divergence of an acoustic beam, scattering, and energy dissipation. In the ideal media – gases, liquids or solids, it is assumed that the disturbances caused by these vibrations were relatively small and vibrations were considered to be of a low amplitude or intensity (1). Sonication is a process of dispersing, disrupting and deactivating various biological materials by the use if sond waves. In fluids such was water produces cavitation bubbles which upon collapse, due to major surface or other asymmetry results in formation of microjets (Chung et al., 2006). Ultrasonic has been studied widely in many researches. Ultrasound is used together with multistage elution to elute super heavy metal oil from weathered soils. It is shown that ultrasound increase the elution of super heavy oil by 13-14% (Ji et al., 2010). In extraction of active ingredient *F. suspensa,* the ultrasound is adopted in this research which indicated that ultrasound-assisted extraction is a powerful tool for the extraction of important phytochemicals from plant materials (Xia et al., 2010). In the study of influence of ultrasound on cadmium ion removal by sorption

process, the ultrasonic process is an effective method for the removal of cadmium ion from solution quickly and efficiently under proper condition; intensity and sorbate/sorbent ratio (Entezari and Bastami, 2007). In the study of activity of allinase from fresh garlic, the ultrasonic irradiation increase the activities of free enzymes instead of their deactivation when the shear force, temperature, pressure and production of radicals are limited (Cao et al., 2010). Previous research had reviewed that ultrasonic applies acoustic energy which can be transmitted to any medium with elastic properties, including water, gas-saturated water and aqueous particle suspensions, in the form of mechanical energy in which can influence the medium through which it passes (Hindu and Takemura, 2006).

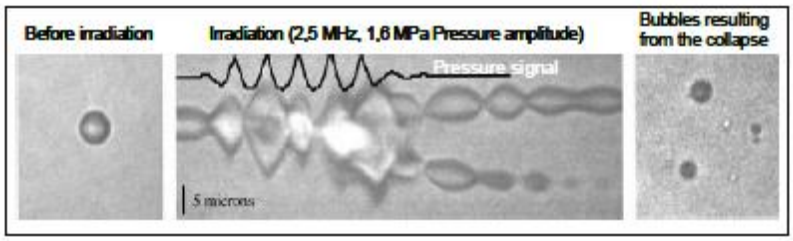

Figure 2.1 Optical images of micro-bubble during and after sonication (Jhony, 2008)

The separation of copper and iron in printed circuit board (PCB) waste sludge has achieve leaching efficiencies of 97.83% and 1.23% each, with the proper leaching procedure and ultrasonic is applied (Cai et al., 2009). Ultrasound also reduces the amount of heavy metals and enhances the dewaterability of sludge significantly (Kim et al., 2004). In study of different approaches for heavy metal partitioning, total operation of ultrasonic assisted extraction only require 15-20 min than 16 hour for conventional single extraction. The extraction is increased when both ultrasound and microwave energy were applied (Arain et al., 2009).

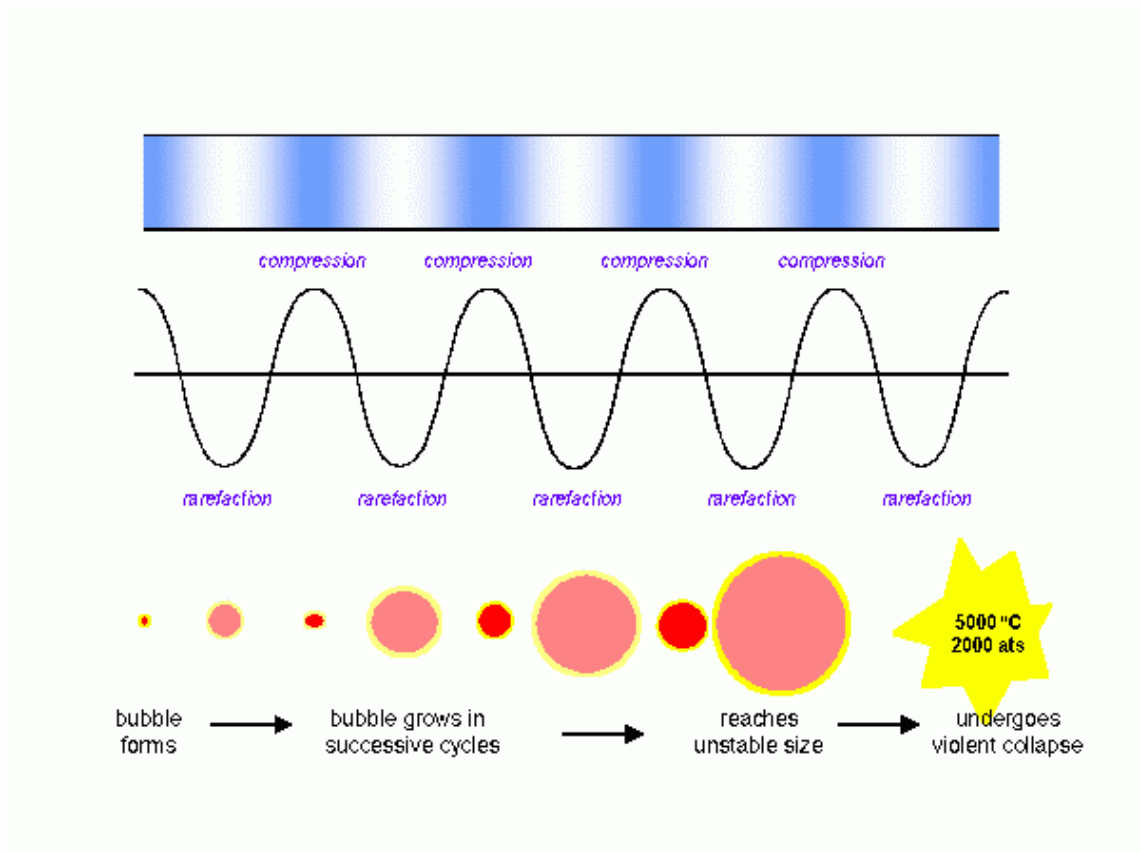

Figure 2.2 Generation of acoustic bubble

2.2 Hospital Wastewater

Wastewater can be defined as any water, whose quality has been adversely being abused by anthropogenic influence which includes liquid waste discharged from domestic home, industries, agricultural and commercial sectors (Ekhaise and Omavwoya, 2008). Hospitals produce a significant volume of wastewater per day. Previous study had shown that the volume of water generally generated from hospitals varies from 400 to 1200 L/day/bed. This volume of water produces by the hospital contain micro-organisms, heavy metals, toxic chemicals and radioactive elements which results in the production hybrid wastewater as the combination of domestic, industrial and effluents of care and medical research. The hospital wastewater also found to

contain chlorinated molecules in high concentrations and heavy metal such as mercury and silver. The differences of hospital wastewater compared to municipal wastewater is there are some patient therapy activities which make the wastewater containing more in organic matters, chemicals and pathogenic organisms. The occurrence of pharmaceuticals in wastewater also being reported which is including antibiotics, hormones, strong painkillers, tranquilizers and chemotherapy chemicals given to cancer patients excreted by humans and domestic animals are distributed into the environment by flushing toilets and spreading manure and sewage sludge onto and into soil (Emmanuel et al., 2002). From WHO report, 85% of hospital waste is non-hazardous, 10% infective and 5% not infective but hazardous (Ekhaise and Omavwoya, 2008). Waste water from hospital may come from few category of hospital facility which are clinical and research laboratories, medical waste incinerators and hospital laundries (Niumpang, 2006).

Table 2.1 Source of hospital wastewater (Niumpang, 2006)

Hospital facility	Description
Clinical laboratories	• Process performed such as anatomic pathology (including routine histology and cytology), chemistry, drug monitoring and toxicology, hematology, immunology, microbiology, transfusion medicine and urinalysis • The wastewater from this facility might contain ionic mercury and organumeric compounds, other heavy metals, organic chemicals, blood products and body fluids, phosphates, oxidizers and particulate materials
Research laboratories	• Research laboratories conduct studies in infectious disease control, blood chemistry, pathology, animal research and inorganic chemistry • The waste contains pollutants such as oxidizers (disinfecting media),

	radionuclides, proteins, oil and grease (vacuum pumps and other rotating equipment), heavy metals (analytical reagents), organic solvents, blood products and other body fluids, formaldehyde, phosphates and detergents (from glass cleaning and instrument strelizing process) and photographic imaging chemicals
Medical waste incinerators	• On-site medical waste incinerators is installed to reduce the cost of off-site waste disposal to burn the waste • The liquid waste from the incinerator usually has relatively low concentrations of organic material, oxidizers and also significant concentrations of particulate matter and heavy metals (including mercury)
Hospital laundries	• The laundries usually process linens, gowns and lab coats • The waste stream might contain with starch, particulate (including lint), proteins (blood products), detergents and oxidizers (bleach or other disinfectant) • Laundry chemicals (sodium hydroxide and bleach) are known often to contain significant levels of mercury contamination

Hazardous medical waste primarily consists of chemicals and discarded cytotoxic drugs in which spread into the environment due to improper usage and undiscriminate disposal (Ekhaise and Omavwoya, 2008). Their carcinogenic natural might pose serious environmental health risk (Ekhaise and Omavwoya, 2008). Leaving the wastewater treatment plants, these chemical compounds could provoke the pollution of natural environment by entailing biological imbalance. This might lead to negative effect on the receiving waters and the living species (Rezaee et al., 2005). The medical waste pose major health risks to the inhabitants of the terrestrial and aquatic ecosystem such as contamination of dirty water that possibly the leachate entering the aquifers, surface water, accumulation of toxic non-biodegradable hospital waste products, which could lead to the blockage of the sewage system, release of toxic substance into the air

due to burning, accumulation of heavy metals and unprotected landfill as well as inefficient sorting of waste materials (Ekhaise and Omavwoya, 2008). The hospital pollutants that come in contact with the elements of the aquatic ecosystem endanger them which are bound to existence of hazardous substances that have the potential of negative effects on the living species and environment (Rezaee et al., 2005). There is also the probability of toxic effects appearance after the exposure of the living organisms to hazardous objects (Rezaee et al., 2005).

2.3 Heavy metals

The source of heavy metal in hospital wastewater can be originated from numerous sources such as from feces, cleaners, paints and wear and tear of utensils and equipment (Niumpang, 2006). Certain heavy metals are micronutrients essential for plant growth (e.g., copper and zinc) and provide benefit to the crop. However, they can present problems for plant growth at excessive amount. Some are also not essential plant or animal and toxic to animals and human such as arsenic, cadmium and mercury. Heavy metals pose negative effect on plant growth and health, animal health, human health and other environmental impacts (Niumpang, 2006). Heavy metals will not degrade over time and relatively immobile in soil and remain indefinitely. The physiochemical study of hospital wastewater discharged from University of Benin Teaching Hospital (UBTH) shown that the concentration of sulphate, nitrate and potassium are high (Ekhaise and Omavwoya, 2008).

Table 2.2 Heavy metal analysis of UBTH, Benin City Hospital Wastewater (Ekhaise and Omavwoya, 2008)

Element	Concentration (µg/l)
Sulphate	22.76
Nitrate	1.00
Potassium	9.45
Cadmium	<0.01
Lead	0.10
Chromium	<0.01
Zinc	0.02
Iron	0.06

Table 2.3 Heavy metal effects (Krogman, 2006)

Element	Essential for plant growth	Toxic*
Arsenic	No	P, A, H
Cadmium	No	P, A, H
Copper	Yes	P
Lead	No	A, H
Mercury	No	A, H
Molybedium	Yes	P, A, H
Nickel	Yes	P, A
Selenium	No	P, A, H
Zinc	Yes	P, A

*P = plants; A = animals; H = humans

2.3.1 Lead (Pb)

Lead is a naturally occurring bluish-gray metal found in small amounts in the earth's crust. It has no characteristic taste or smell. Metallic lead does not dissolve in water and does not burn. Lead can combine with other chemicals to form what are usually known as lead compounds or lead salts. Some lead salts dissolve in water better than others. Some natural and manufactured substances contain lead but do not look like lead in its metallic form. Some of these substances can burn - for example, organic lead compounds in some gasolines. In medical field, lead has been used in a large variety of medical equipment especially for radiation shields for protection against X-rays, electronic ceramic parts of ultrasound machines, intravenous pumps, fetal monitors and surgical equipment. Expose to lead will interfere several viral enzyme activities if it is chronically inhaled or ingested in sufficient quantity to accumulate. Lead poses most vulnerable effect to developing fetuses, infants and young children. Severe childhood lead poisoning can cause neurological and kidney damage, anemia, and even death. Moderate poisoning can produce stomach upsets, vomiting, convulsions, an abnormal walk, headaches, irritability, hyperactivity, learning and behavioral disorders, delayed mental development, and perhaps retardation. In adults, toxic levels of the metal can do extensive damage, especially to the nervous system and kidneys. Its symptoms include high blood pressure, headache, loss of appetite, intestinal upset, memory loss, an impaired gait, and a metallic taste in the mouth. During pregnancy, lead poisoning can cause miscarriage, severe congenital defects, or death of the fetus.

2.3.2 Nickel (Ni)

Pure nickel is a hard, silvery white metal, which has properties that make it very desirable for combining with other metals to form mixtures called alloys. Some of the metals that nickel can be alloyed with are iron, copper, chromium, and zinc. These alloys have important uses such as in the making of metal coins and jewelry and in

industry for making items such as valves and heat exchangers. Most nickel is used to make stainless steel. Compounds of nickel combined with many other elements, including chlorine, sulfur, and oxygen, exist. Many of these compounds dissolve fairly easily in water and have a characteristic green color. Nickel and its compounds have no characteristic odor or taste. Base on the study on animal, rats and mice may die after eating large amounts of nickel. Eating levels of nickel very much greater than the levels normally found in food causes lung disease in dogs and rats and affects the stomach, blood, liver, kidneys, and immune system in rats and mice. Effects on reproduction and birth defects also were found in rats and mice eating or drinking very high levels of nickel. The studies in animals were completed using high levels of soluble nickel which is more readily absorbed by the gastrointestinal tract than the nickel compounds usually found in water and food. The most serious effects of nickel, such as cancer of the lung and nasal sinus, have occurred in people who have breathed nickel dust while working in nickel refineries or in nickel processing plants (Habeck, 1995)

2.4 Leaching Solution

In this research, the leaching solution is used to assist the ultrasonic process to remove heavy metal. Leaching itself is the process of extracting minerals from solid by dissolving them in a liquid, either in nature or through an industrial process. Leaching has a variety of commercial applications such as separation of metal from ore using acid and sugar beets using hot water. Using chemical to remove heavy metals has received extensive attention due to its simple operation, short extraction time and high removal efficiency. The efficiency of heavy metal removal is typically depends on pH, temperature, contact time, and extracting agent type (Deng et al., 2009). Heavy metals in this hospital wastewater will be removed using various of leaching solutions; ethylenediaminetetraacetic acid (EDTA), nitric acid (HNO_3) and citrate ($C_3H_5O(COO)_3^{3-}$).

2.4.1 Ethylenediaminetetraacetic acid (EDTA)

EDTA or ethylenediaminetetraacetic acid is a polyamino carboxylic acid and a colourless, water-soluble solid. It is widely used to dissolve scale. EDTA becomes very useful due to its role as hexadentate (six-toothed) ligand and chelating agent as well as its ability to sequester metal ions. After being bounded by EDTA, metal ions remain in solution but exhibit diminished reactivity.

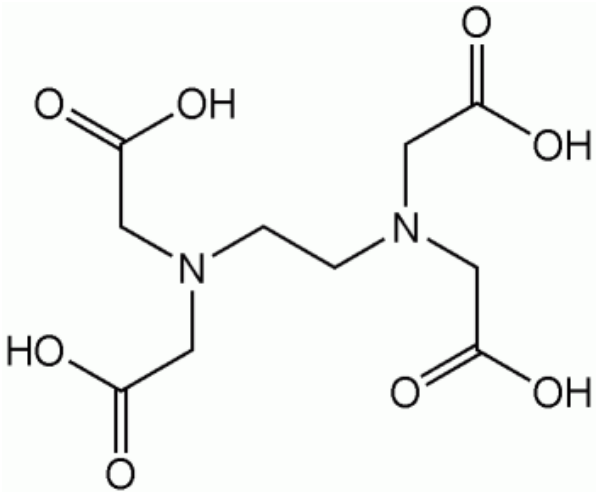

Figure 2.2 EDTA structure

Previous research had widely used EDTA as leaching solution. EDTA is widely used on photographic industry, in textile and paper manufacturing, industrial cleaning and also use with micronutrients in agriculture and horticulture to increase their bioavailability. EDTA also has the ability to bring heavy metals into solution to remediate contaminated soil (Thayalakumaran et al., 2003). EDTA also easily coupled with another metal that formed complexes, such as Ni-EDTA and Cu-EDTA which are commonly found in

electroplating wastewater (Fu et al., 2009). It is also used as leaching solutions to remove heavy metals in contaminated soil (Hwang et al., 2007). In medical treatment, EDTA has been used in chelating therapy that is used to treat acute and chronic lead poisoning by pulling toxins including heavy metal from bloodstream. In soil-washing process, EDTA is used as the chelating agent for the decontamination of lead-contaminated soil and capable of being recycled several times without losing its extractive power (Kim and Ong, 1999). The biosorption of few heavy metals such as Cu, La, U, Ag, Cd and zPb are severely affected in the presence of EDTA (Bishnoi and Garima, 2004). It has been reported that EDTA is used to bound with Ca(II) and form hydroxide precipitation of the complex metal and major component to determine Cd(II) solubility (Kurniwan et al., 2006). EDTA also has been used as chelator agent to enhance phytoextraction of heavy metals from contaminated irrigated by industrial wastewater by increasing the metal accumulation (Sun et al., 2009).

2.4.2 Nitric acid (HNO_3)

Nitric acid, chemical compound, HNO_3, colorless, highly corrosive, poisonous liquid that gives off choking red or yellow fumes in moist air. It is miscible with water in all proportions. It forms an azeotrope (constant-boiling mixture) that has the composition 68% nitric acid and 32% water and that boils at 120.5°C. The nitric acid of commerce is typically a solution of 52% to 68% nitric acid in water (wiki).

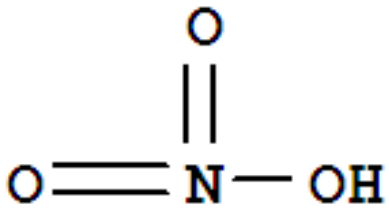

Figure 2.3 HNO_3 structure

Nitric acid had been used together with ultrasonic treatment in removal of heavy metals from sludge and proved to remove significant amount of heavy metal (Deng et al., 2009). In separation of metal ions from a surfactant solution by acidification, HNO_3 had shown the highest percentage of metal separation compare to H_2SO_4 and HCl (Kim et al., 2006). Maximum extraction of heavy metal can be achieved when using together with hydrogen peroxide in shorter sonication time (Arain et al., 2009).

2.4.3 Citrate ($C_3H_5O(COO)_3^{3-}$)

Citrate is the first intermediate of the citric acid cycle and tricarboxylic acid (TCA) cycle. It also can be refered either to the conjugate base of citric acid, ($C_3H_5O(COO)_3^{3-}$), or to the esters of citric acid. An example of the former, a salt is trisodium citrate; an ester is triethyl citrate.

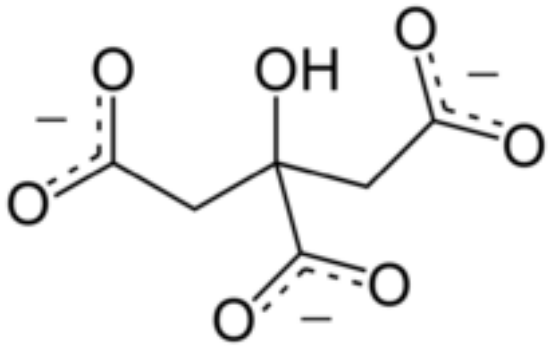

Figure 2.4 Citrate structure

From previous study, citrate has been used as a chelating agent in combination with electrodialytic remediation. It is a naturally occurring chelating agent which forms stable chelates with several heavy metal and used for extraction heavy metals from polluted soils (Elik, 2007). It also has been used to assist ultrasonic process in extraction to release heavy metal from contaminated soil (Hwang et al., 2007).

CHAPTER 3

METHODOLOGY

3.1 Introduction

Currently, the study on medical wastewater most revolve around the biological treatment and more on the analysis of microbiological and physiochemical of the wastewater itself. Numerous studies had been done on removal of heavy of metal from several samples such as industrial wastewater, soil and sludge.

As the procedure, prepared sample will be added with amount of various leaching solutions (EDTA, HNO_3, citrate), thus acidification process occurred that make the heavy metal soluble. The ultrasonic will be used in which the cavitation will excite disintegration of heavy metal particle into solution. The sample then analysed using AAS.

3.2 Sample

The 10L tank is used to collect the sample from Hospital Kemaman, Terengganu from its septic tank. The sample is filtered using vacuum pump. For the experimental process, 10 ml sample is diluted in 100 ml volumetric flask. The sample is spiked with targeted heavy metal, 5ppm of Pb and 20ppm of Ni.

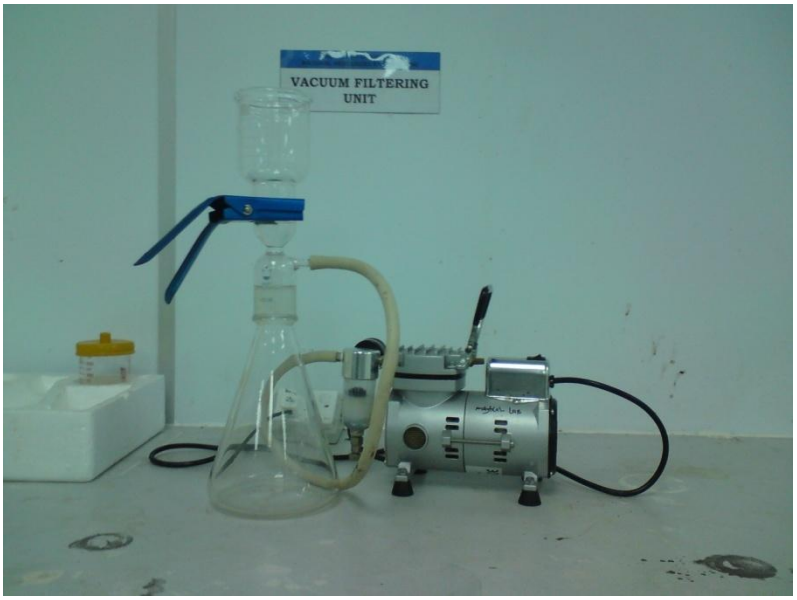

Figure 3.1 Vacuum filtering unit

3.3 Material

Lead (Pb) and nickel (Ni) was obtained from Merck in analytical grade. Stock solutions are prepared by dissolving lead and nickel in ultrapure water at various concentrations.

Table 3.1 Standard solution of Pb and Ni

Pb	Ni
0 (blank)	
10ppm	5ppm

20ppm	10ppm
30ppm	20ppm

The chemical used in this experiment are summarized in the table below.

Table 3.2 Type of Chemical

Chemical	Provider
EDTA Sigma Grade	SIGMA
Citric acid anhydrous	Fluka
Sodium hydroxide	Merck
Nitric acid 65%	Fisher Chemicals

3.4 Equipment

In this experiment, the equipment used for ultrasonic treatment is DAIHAN Digital Ultrasonic Cleaner with the temperature control up to 90°C and the range of sonication time 0-99 minutes. The determination of Pb and Ni analysis is carried using HITACHI Z-5000 Polarized Zeeman Atomic Adsorption Spectrometer.

Figure 3.2 DAIHAN Digital Ultrasonic Cleaner

Figure 3.3 HITACHI Z-5000 Polarized Zeeman Atomic Adsorption Spectrometer

3.5 Experimental Procedures

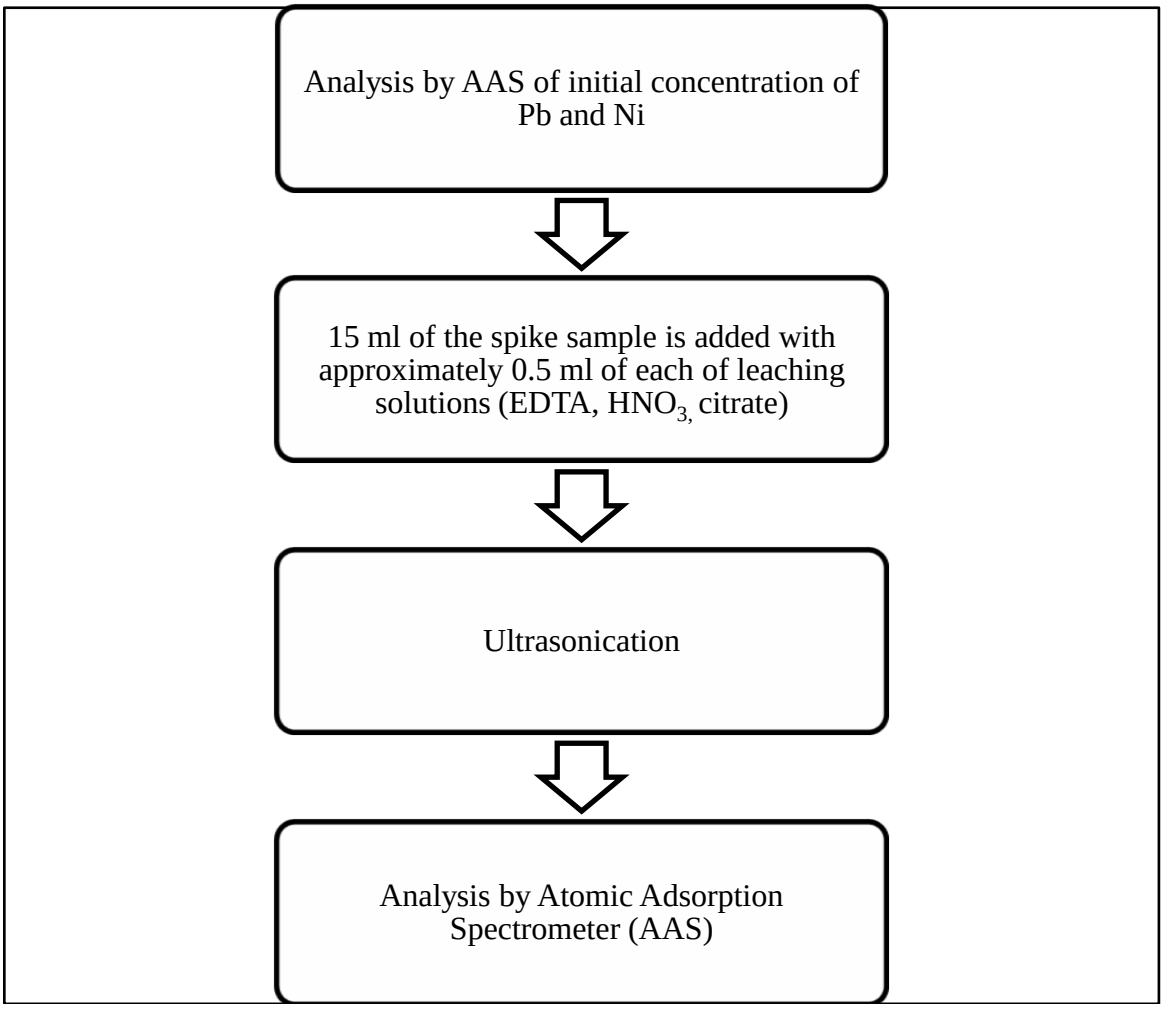

Figure 3.4 Experimental Procedure

Table 3.3 Summarization of procedure according to parameter

PARAMETER	Type of leaching solution	Sonication time	Concentration of leaching solution
PROCEDURE	Sample is added with leaching solution	Sample is added with leaching solution	Sample is added with varied of concentration of leaching solution
	Ultrasonication for 10 minutes	Ultrasonication in varied of time	Ultrasonication according to previous parameter
	Analysis by AAS		

From table 3.3, for the first parameter, 10 ml sample is transferred into conical flask 100 ml and added with approximately 0.5 ml of fixed concentration of 0.1 M EDTA, 0.1 M HNO_3 and 0.1 M citrate. Then, the sample is put into the ultrasonic cleaner for 10 minutes. The analysis of Pb and Ni removal is analysis by AAS.

For the second parameter, the same method as the first parameter is adopted. However, ultrasonication is run at varied of time which for the Pb sample within the range of 0 to 15 minutes and Ni, 0-20 minutes. The analysis of Pb and Ni removal is analysis by AAS.

For the third parameter, the sample is added with various of concentration which are ranged from 0.02 M to 0.12 M. The sample is put into ultrasonic cleaner and run at the time which had gave the higher removal of Pb and Ni from previous parameter. The analysis of Pb and Ni removal is analysis by AAS.

CHAPTER 4

RESULT AND DISCUSSION

4.1 Type of Leaching Solutions

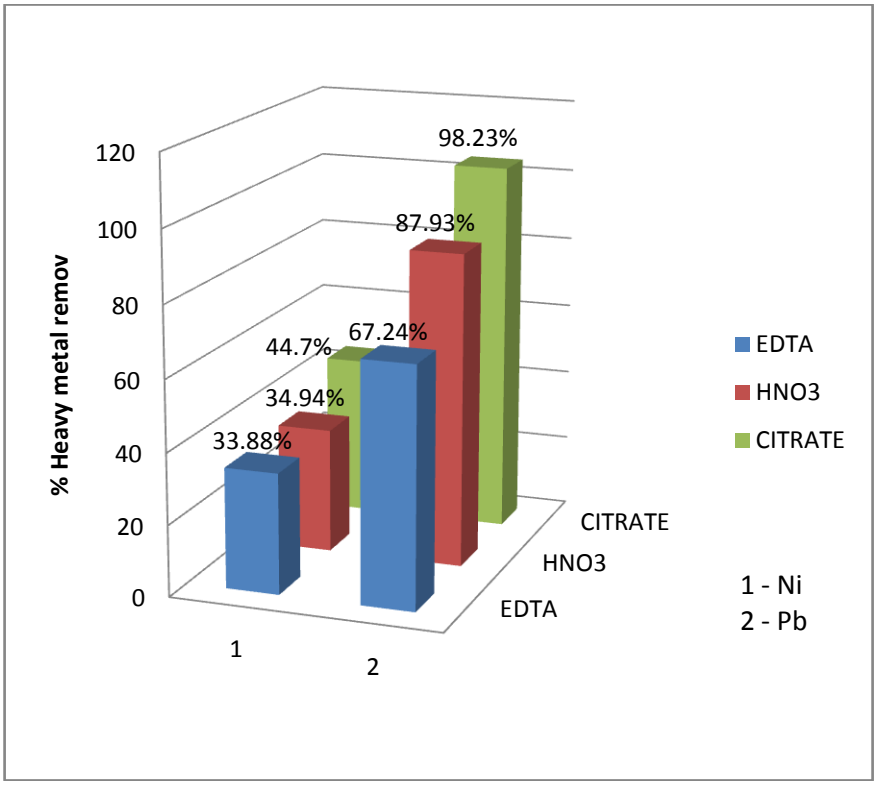

Figure 4.1 Effect of Type Of Solvent

At different type, the Pb and Ni removal at 0.1M of each of the leaching solution and 10 minutes of sonication time, citrate has shown the highest removal for both Pb and Ni which is 98.23% and 44.7% each. While for nitric acid and and EDTA, the removal

of Pb is 87.93% and 67.24% each and Ni is 34.94% and 33.88% each. In previous study, the extraction of efficiencies of Pb from contaminated soil is higher for EDTA than citrate (Hwang et al., 2009). The percentage of metal removal is depending on metal and sample type (Elik, 2009).

4.2 Effect of Sonication Time

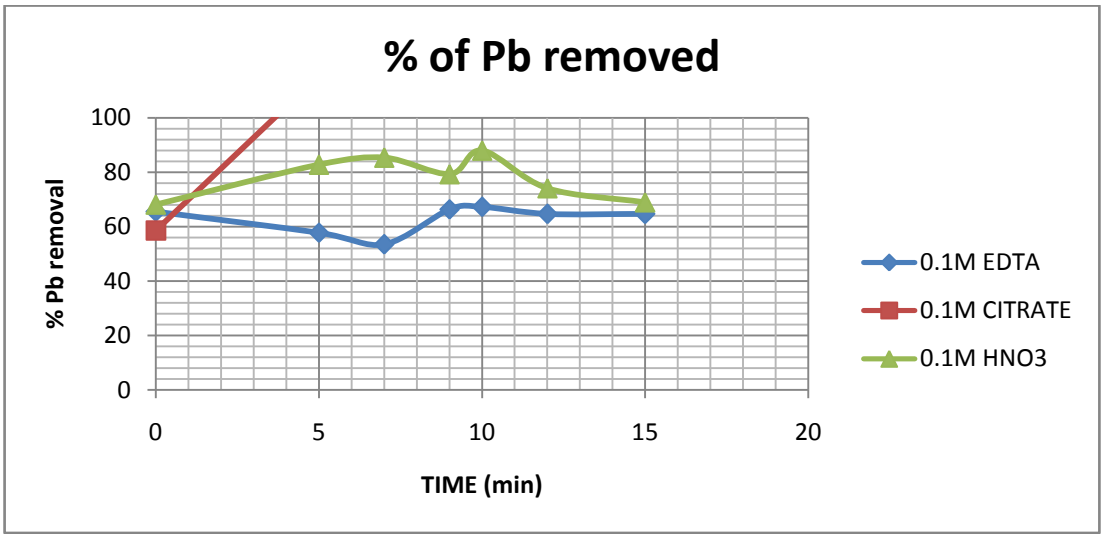

Figure 4.2 Effect of Sonication Time in Pb Removal

At 0.1M of each of the leaching solution, it is shown that the removal of Pb using citrate require only less than 4 minutes to remove higher percentage of Pb. The percentage of Pb removed is 97%. Using EDTA, the percentage is slightly decreased at 10 minutes and increase at 10 minutes with significant percentage of removal 67.24%. Previous study the higher extraction efficiency is achieved at 12 minutes of sonication time (Hwang et atl, 2007) Further sonication time, it is observed that percentage is decreased. Using HNO_3, the percentage removal is decrease within the range of time 0 to 7 minutes. The curve is increased until 9.5 minutes where the percentage of removal

is significant which is at 87.93%. The removal of heavy metal is not significant at longer sonication time and become constant.

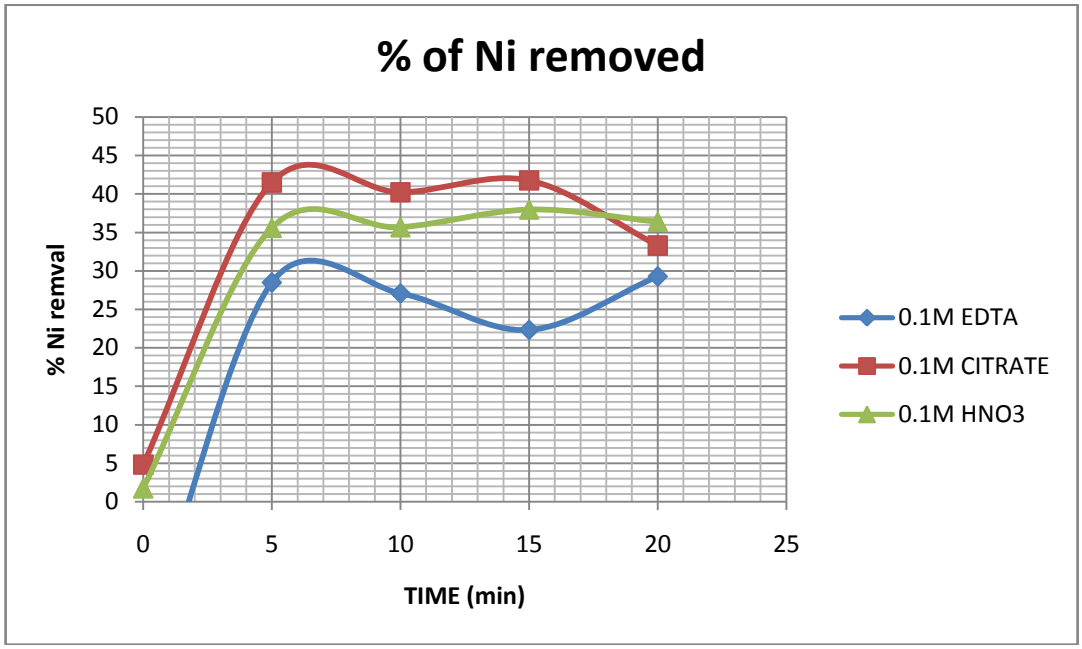

Figure 4.3 Effect of Sonication Time in Ni Removal

For Ni removal, citrate also shown the highest removal of Ni which is at 41.48% within 6.5 minutes of sonication time. Further the sonication time, it is observed that the percentage of removal is decreased. The higher removal of Ni also occurred within 6.5 minutes for both EDTA and HNO3, with 0.1M EDTA, 32% removal and 0.1M HNO3, 38% removal. However, for HNO3, the percentage of removal decreased within the range of 15 to 20 minutes of sonication. For EDTA, the curve is decreased from 7 to to 15 minutes and increased from 15 to 20 minutes.

Citrate is a chelating agent which can form stable chelates with several heavy metals (Pedersen, 2002). Chelating agent removes a metallic ion from a solid salt and holds it in solution. Citrate form stable complex with selected heavy metal (Pedersen,

2002) which for this research is Pb and Ni. In the presence of ultrasonic, the cavitation process is occurred which is a rapid and repeated formation and resulting implosion of micro bubbles in a liquid resulting in the propagation of microscopic shock waves. Shock waves form cavitation which produce high-velocity particle collisions and remove the heavy metal (Hindu and Takemura). Previous study had shown that extended ultrasonication, may release the heavy metal from the sludge . With the addition of leaching solution, it improved the efficiency of heavy metal removal. Longer sonication time will remove more heavy metal but it will pose a negative effect such as higher energy consumption (Deng et al., 2009).

4.3 Effect of Concentration

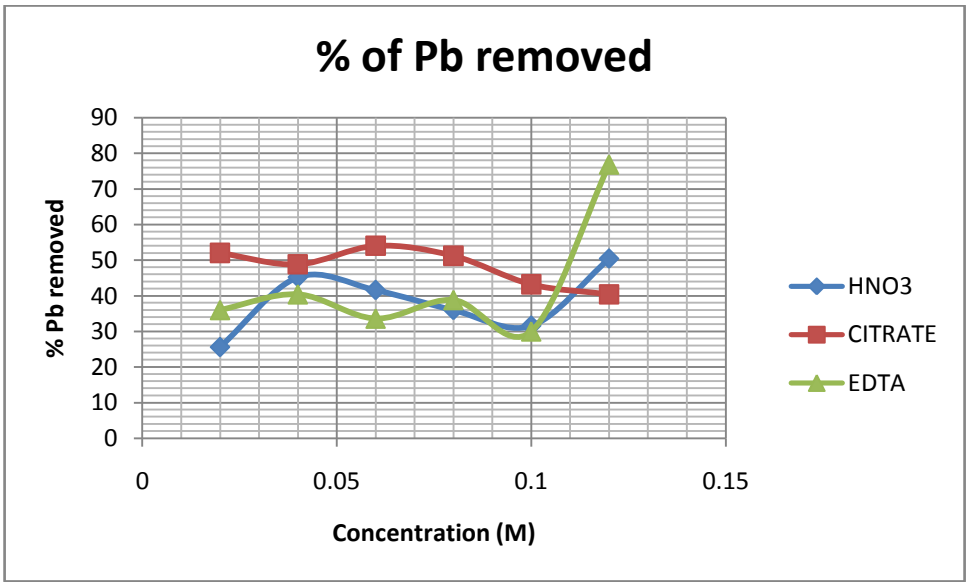

Figure 4.4 Effect of concentration in Pb removal

In Pb removal, the significant percentage is observed when using 0.06 M citrate with the percentage of 54% removal. At higher concentration, the percentage removal is lower and not significant. Meanwhile, EDTA had shown higher removal at 0.12 M which is76.8%. At lower concentration within the range of 0.02 M to 0.12 M, there is no

significant removal is observed. Similar with EDTA, HNO3 also removed higher percentage of heavy metal (50.4%) at higher concentration (0.12 M).

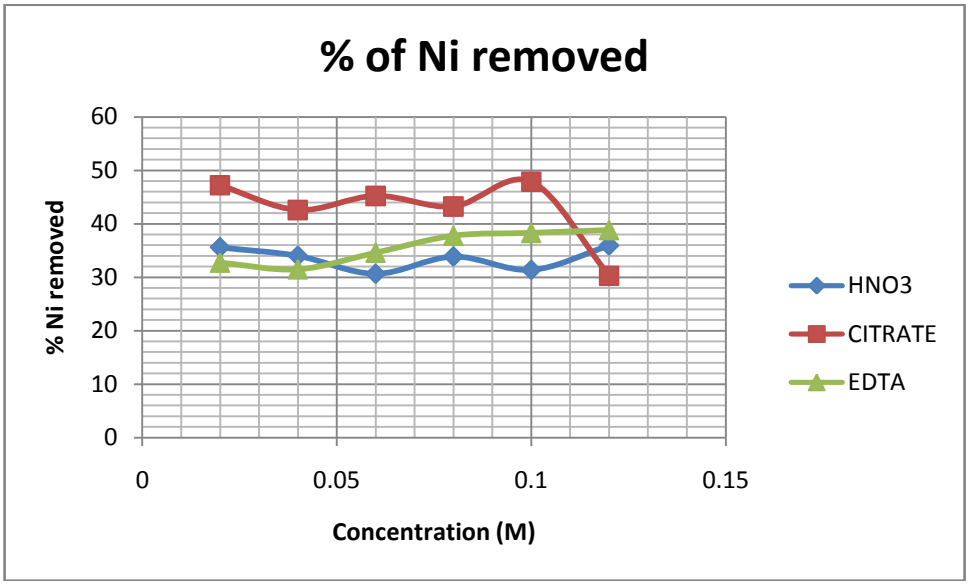

Figure 4.5 Effect of Concentration in Ni Removal

The removal of Ni is higher at 0.1 M of citrate with the percentage removal of 35.93%. At 0.12 M citrate, the percentage of removal is decreased. As for EDTA, the trend looks the same Pb removal since the removal is increased at higher concentration. At 0.12 M EDTA, the percentage of removal is 38.84%. The same goes with HNO3 whereas the removal is higher at higher concentration. At 0.12 M HNO3, the removal of Ni is 35.93%.

Using acid as a solvent for extraction is a common method to remove heavy metals from sludge and type of acid used and other operating condition play an important role in determining the removal efficiency. It has been reviewed that the increased in acid concentration will increased the acid extraction (Kuan et al., 2009). Increase in acid concentration results in corresponding increase of the energy for the breakdown of the chemical bonds of the metal (Stylianao et al., 2007) which improved

the ability to dissolve heavy metal. In addition, high concentrations cause the disintegration of the heavy metal particle into solution and consequently, increased the process of dissolving heavy metals (Hwang et al., 2009). However, higher concentration will require more energy consumption for the breakdown of the chemical bonds of the metal.

CHAPTER 5

CONCLUSION AND RECOMMENDATION

5.1 Conclusion

This study is to find the appropriate leaching solution in ultrasound-assisted leaching solution to remove Pb and Ni from hospital wastewater. The leaching solutions that had been used and compared throughout this experiment are EDTA, HNO_3 and citrate. As the result, citrate had found to become an appropriate leaching solution to remove Pb and Ni with the removal of 54% and 35.93% each. The best sonication time is obtained at 3minutes for Pb removal and 6 minutes for Ni removal. The suitable concentration to be used is 0.06 M for Pb removal and 0.1 M for Ni removal.

It is found that, the ultrasonic treatment is an economically process to remove heavy metal and a powerful tool when it is combined together with the use of leaching solution. Thus, ultrasound is a necessary method for the improvement of heavy metal extraction.

5.2 Recommendation

The study of heavy metal removal from wastewater can be widened by using other available chemical to remove heavy metal such as inorganic acids (HCl and

H_2SO_4) and inorganic acids (oxalic acid) and complexing agent (NTA) which has been proposed as effective extracting agent.

Instead of the effect of sonication time and acid concentration, other effects also can be studied such as pH of solution, power adjustment of the ultrasonic equipment and temperature.

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APPENDIX A

SAMPLE AND CHEMICAL PREPARATION

Appendix A1: Sample preparation

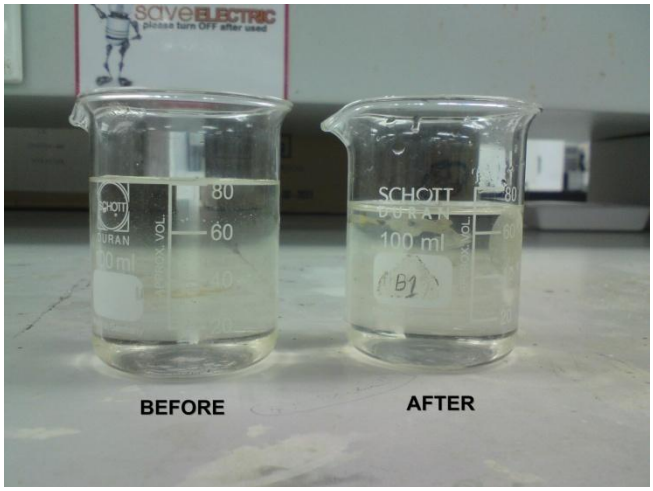

Figure A.1.1 Sample filteration

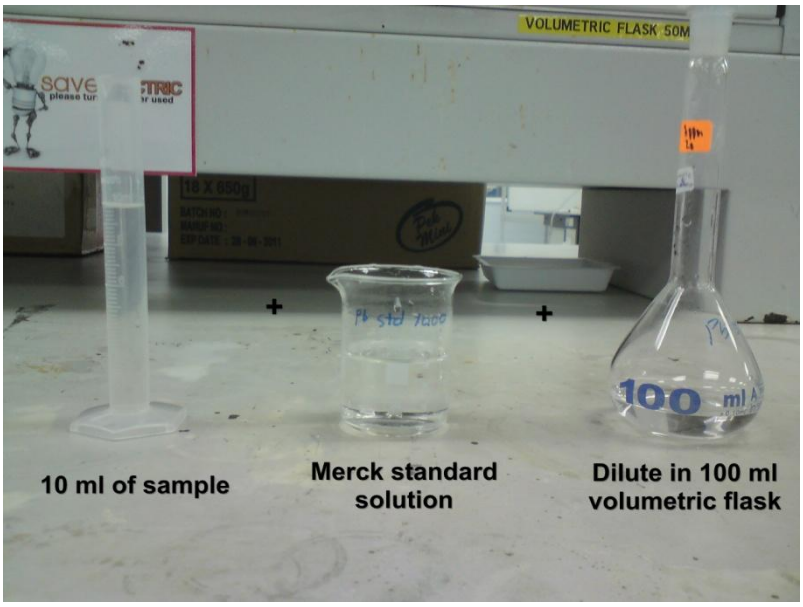

Figure A.1.2 Dilution of sample and amount of standard solution being spike

Appendix A.2 Chemical preparation

Figure A.2.1 Chemical used for the experiment (citric acid, EDTA, sodium hydroxide)

Figure A.2.2 Nitric acid 65% analytical reagent

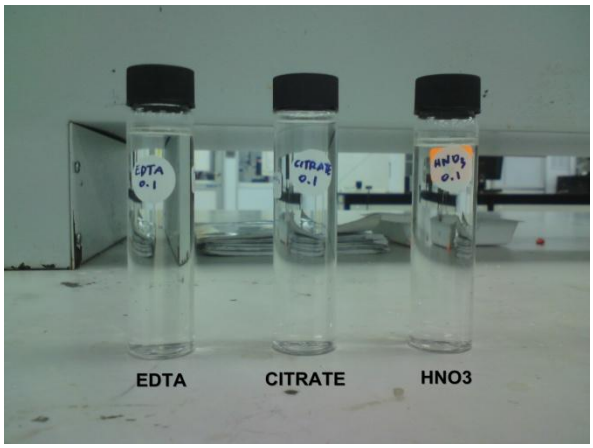

Figure A.2.3 Diluted leaching solution

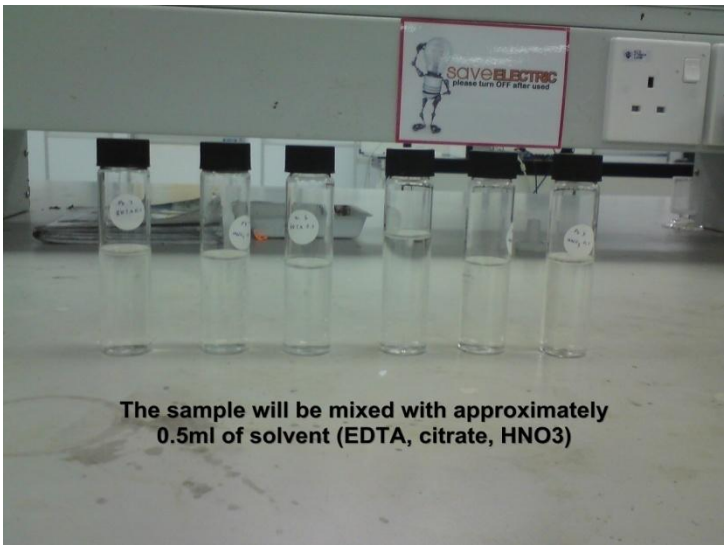

Figure A.2.4 The sample added with leaching solution

APPENDIX B

TABLE OF RESULT

Appendix B1: Equation of removal percentage (%)

$$Percentage\quad (\%) = \frac{(initial\ c\ \ oncentrati\ \ on\ -final\ \ concentrat\ \ ion)}{initial\ co\ \ ncentratio\ \ n} \times 100$$

Appendix B2: Type of solvent

HM	initial conc.	0.1 EDTA	0.1 HNO3	0.1 CITRATE	actual conc.	ACTUAL			% HM REMOVAL		
						0.1 EDTA	0.1 HNO3	0.1 CITRATE	EDTA	HNO3	CITRATE
Ni	35.14	30.01	29.85	28.36	15.14	10.01	9.85	8.36	33.88375165	34.94055	44.78203
Pb	6.16	5.38	5.14	4.95	1.16	0.38	0.14	-0.05	67.24137931	87.93103	104.3103

Appendix B3: Effect of sonication time

Table B3.1 % Pb removal

SOLUTION	TIME (min)	conc. Pb	actual Pb conc.	% removal
0.1M EDTA	0	5.4	0.4	65.5172414
	5	5.49	0.49	57.7586207
	7	5.54	0.54	53.4482759
	9	5.39	0.39	66.3793103
	10	5.38	0.38	67.2413793
	12	5.41	0.41	64.6551724
	15	5.41	0.41	64.6551724
0.1M CITRATE	0	5.48	0.48	58.6206897
	5	4.82	-0.18	115.517241
	7	7.4	2.4	-106.89655
	9	4.89	-0.11	109.482759
	10	4.98	-0.02	101.724138
	12	5.06	0.06	94.8275862
	15	4.97	-0.03	102.586207
0.1M HNO3	0	5.37	0.37	68.1034483
	5	5.2	0.2	82.7586207
	7	5.17	0.17	85.3448276
	9	5.24	0.24	79.3103448
	10	5.14	0.14	87.9310345
	12	5.3	0.3	74.137931
	15	5.36	0.36	68.9655172

Table B3.2 % Ni removal

SOLUTION	TIME (min)	conc. Ni	actual Ni conc.	% removal
0.1 M EDTA	0	37.97	17.97	-18.692206
	5	30.83	10.83	28.4676354
	10	31.04	11.04	27.0805812
	15	31.76	11.76	22.324967
	20	30.71	10.71	29.2602378
0.1 M CITRATE	0	34.41	14.41	4.82166446
	5	28.86	8.86	41.4795244
	10	29.05	9.05	40.2245707
	15	28.82	8.82	41.7437252
	20	30.1	10.1	33.2892999
0.1 M HNO3	0	34.88	14.88	1.71730515
	5	29.75	9.75	35.6010568
	10	29.74	9.74	35.667107
	15	29.39	9.39	37.9788639
	20	29.63	9.63	36.3936592

Appendix B4: Effect of leaching solution concentration

Table B4.1 % Pb and Ni removal

SOLUTION	M	Ni			Pb		
		conc.	actual conc.	% removal	conc.	actual conc.	% removal
HNO3	0.02	29.75	9.75	35.60106	6.86	1.86	25.6
	0.04	29.99	9.99	34.01585	6.37	1.37	45.2
	0.06	30.5	10.5	30.64729	6.46	1.46	41.6
	0.08	30.01	10.01	33.88375	6.6	1.6	36
	0.1	30.39	10.39	31.37384	6.71	1.71	31.6
	0.12	29.7	9.7	35.93131	6.24	1.24	50.4
CITRATE	0.02	27.99	7.99	47.22589	6.2	1.2	52
	0.04	28.69	8.69	42.60238	6.28	1.28	48.8
	0.06	28.29	8.29	45.24439	6.15	1.15	54
	0.08	28.59	8.59	43.26288	6.22	1.22	51.2
	0.1	27.89	7.89	47.88639	6.42	1.42	43.2
	0.12	30.55	10.55	30.31704	6.49	1.49	40.4
EDTA	0.02	30.18	10.18	32.7609	6.6	1.6	36
	0.04	30.37	10.37	31.50594	6.49	1.49	40.4
	0.06	29.9	9.9	34.6103	6.66	1.66	33.6
	0.08	29.42	9.42	37.78071	6.53	1.53	38.8
	0.1	29.34	9.34	38.30911	6.75	1.75	30
	0.12	29.26	9.26	38.83752	5.58	0.58	76.8

APPENDIX C

EDTA PHYSICAL PROPERTIES

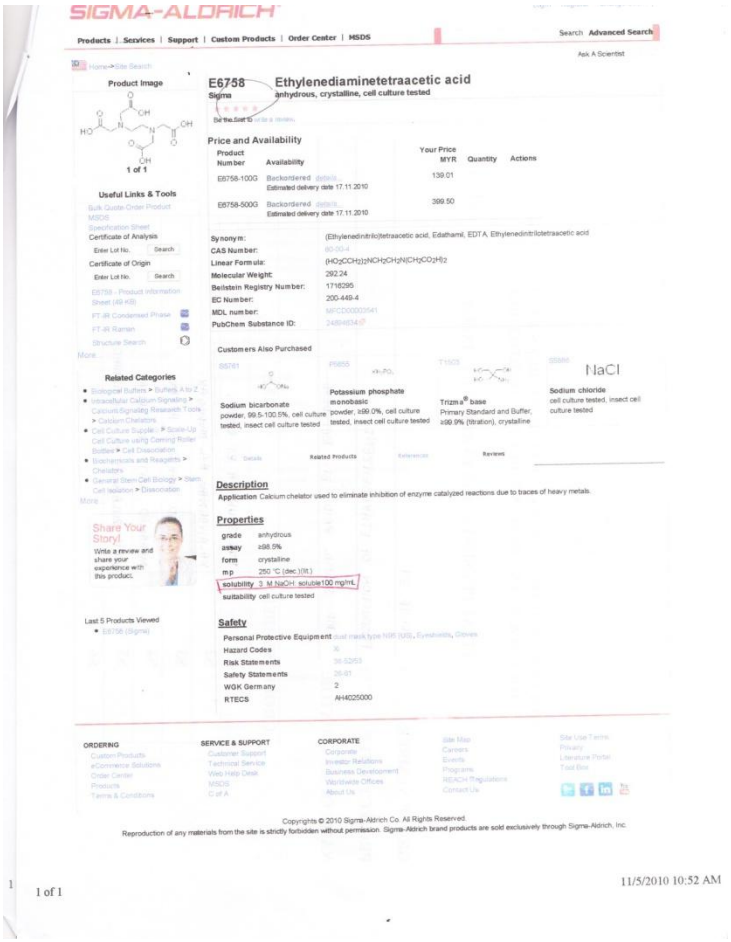

Figure C1: EDTA physical properties