STABILIZATION/SOLIDIFICATION OF LEAD CONTAMINATED SOIL USING CEMENT INCORPORATED WITH SUGARCANE BAGASSE

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ABSTRACT

Soil that is contaminated with heavy metals, especially lead (Pb) has become a major issue worldwide. Pb is reported to be a metal that affects human health and is related to have caused serious diseases that interrupts the nervous system, blood vessels and kidneys. However, proper treatment techniques such as Stabilization/Solidification (S/S) method can be employed and is capable of controlling these heavy metals from contaminating the soil strata and groundwater resources. This research is to investigate the effect of soil strength and leachability of lead in S/S method when sugarcane bagasse (SCB) is added to remedy contaminated soil. Synthetic contaminated soil was prepared in bulk by mixing soil samples with lead nitrate, Pb $(NO_3)_2$ to achieve the concentration of 500 ppm. After that, cement is added at a proportion of 5%, 10% and 15% in sample weights without SCB while in another sample, the cement replaces SCB at a proportion of 2.5%, 5% and 7.5%. All samples were allowed to harden and cured at room temperature for 7, 14 and 28 days. The effectiveness of the treatment was assessed by conducting physical testing such as Unconfined Compression test, Density test and Water Absorption test. In addition, leaching tests were performed to identify the leachate criteria of lead during treatment. Two leaching tests were conducted and they were the Toxicity Characteristic Leaching Procedure (TCLP) and Synthetic Precipitation Leaching Procedure (SPLP). Results indicate that pH and leachability are found to have major influence on metal release. The final pH after leaching tests showed improvements especially samples containing SCB. In addition, the concentration of lead in the TCLP and SPLP test after the curing period of 28 days were detected to be below the leachability limit as regulated by the United States Environmental Protection Agency (US EPA). As a whole, the results obtained from testing showed that soil samples : 7.5% cement : 7.5% SCB is the most effective and is the optimum mix since this proportion succeeded in minimising the leachability of Pb as low as 2.11 mg/L or a total reduction by 99%, and it even produced the strength of 1389 kPa within 28 days. In conclusion, partial replacement of cement with SCB in the binder system has been successful in increasing the strength and reducing the leachability compared to the controlled sample.

ABSTRAK

Tanah yang dicemari dengan logam berat khususnya Plumbum (Pb) merupakan isu yang hebat diperkatakan di seluruh dunia. Pb dilaporkan menyumbang kepada masalah kesihatan manusia yang semakin serius seperti gangguan sistem saraf, kapilari darah dan buah pinggang. Walau bagaimanapun, teknik rawatan tanah yang tepat seperti teknik Penstabilan/Pemejalan (P/P) boleh digunakan dan ia terbukti mampu mengawal logam berat ini dari mencemarkan strata tanah serta sumber air bawah tanah. Objektif utama kajian ini adalah untuk mengkaji kesan penambahan hampas tebu terhadap kekuatan dan larut resap Pb dari tanah yang distabilkan mengunakan teknik (P/P). Tanah tercemar sintetik telah disediakan secara pukal dengan mencampurkan sampel tanah dengan Pb nitrat (Pb $(NO_3)_2$ untuk mencapai kepekatan 500 ppm. Seterusnya, simen ditambah pada kadar 5%, 10% dan 15% untuk sampel tanpa hampas tebu manakala simen diganti sebahagian dengan hampas tebu pada kadar 2.5%, 5% dan 7.5%. Kesemua sampel dibiar mengeras dan diawet pada suhu bilik selama 7, 14 dan 28 hari. Keberkesanan rawatan telah dinilai dengan melakukan ujian fizikal seperti ujian mampatan tak terkurung, ujian ketumpatan dan ujian penyerapan air. Selain itu, ujian pengurasan juga dilakukan bagi mengenalpasti kriteria larut resap Pb semasa rawatan. Dua ujian pengurasan telah dijalankan iaitu Prosedur Pengurasan Ciri Ketoksikan (PPCK) dan Prosedur Pengurasan Hujan Tiruan (PPHT). Hasil kajian menunjukkan pH dan larut resap didapati mempunyai pengaruh yang besar terhadap pelapasan Pb. Nilai pH akhir selepas ujian pengurasan menunjukkan peningkatan terutamanya sampel yang mengandungi hampas tebu. Selain itu, kepekatan Pb di dalam ujian PPCK dan PPHT selepas tempoh pengawetan 28 hari dikesan berada dibawah had larut resap yang dikeluarkan oleh US EPA. Secara keseluruhannya, hasil daripada ujian yang dijalankan mendapati sampel tanah yang mengandungi 7.5% simen dan 7.5% hampas tebu yang dirawat merupakan campuran paling berkesan kerana berjaya meminimumkan larut resap Pb serendah 2.11 mg/L atau penuruan sebanyak 99% pada kekuatan 1389 kPa dalam tempoh 28 hari. Kesimpulannya, penggantian sebahagian simen dengan hampas tebu did alam sistem bahan pengikat dilihat telah berjaya setelah dibandingkan dengan sampel yang tidak dirawat.

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LIST OF SYMBOL AND ABBREVIATION

µm	-	micro meter
AAS	-	Atomic Absorption Spectroscopy
Al_2O_3	-	Alumina
ANOVA	-	Analysis of Variance
ASTM	-	American Society for testing and materials
BA	-	Bottom ash
BDAT	-	Best Demonstrated Available Technology
C_2S	-	dicalcium silicate
C_3S	-	tricalcium silicate
CAC	-	Calcium Alite cement
CAH	-	calcium aluminate hydrates
CaO	-	Calcium oxide
CBR	-	California Bearing ratio
CO_2	-	Carbon dioxide
C-S-H	-	Calcium Silicate Hydrate
DOE	-	Department of Environment
e.g	-	for example
EK	-	Electrokinetic
EPA	-	Environment Protection Agency
EPT	-	Extraction Procedure Toxicity
EU	-	European Union
FA	-	Fly ash
HCL	-	Hydrochloric acid
i.e	-	in other word
IQ	-	intelligence quotient
JMR	-	Jisim molekul relatif

KPa	-	Kilopascal
L/S	-	Liquid to solid ratio
MEP	-	Multiple Extraction procedure
MPa	-	megapascal
MSW	-	Municipal solid waste
NPL	-	National Priority List
OMC	-	Optimum moisture content
OPC	-	Ordinary Portland cement
$Pb(NO_3)_2$	-	Lead nitrate
PC	-	Pozzolanic cement
POFA	-	Palm Oil fuel ash
RECESS	-	Research Centre for Soft Soils
RHA	-	Rice Hush ash
S/S	-	Stabilization/Solidification
SCB	-	Sugarcane bagasse
SCBA	-	Sugarcane bagasse ash
SEM	-	Scanning electron microscope
SiO_2	-	silica
SPLP	-	Syntactic Precipitation Leaching Procedure
TCLP	-	Toxicity Characteristic Leaching Procedure
UCS	-	Unconfined compression strength
UCT	-	uniaxial compression test
UK	-	United Kingdom
US EPA	-	United States Environmental Protection Agency
UTHM	-	Universiti Tun Hussein Onn Malaysia
WHO	-	World Health Organization
XRD	-	X-Ray Diffraction
XRF	-	X-ray Fluorescence

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CHAPTER 1

INTRODUCTION

1.1 Introduction

In the past few decades, there has been excessive growth in global population, industrial development, usage of energy resources and civil infrastructure development. The growth in one sector often incur problems in other areas. Consequently, debates have been intensified of industrialization and its association to environmental issues such as waste management, ecosystem and human health risk assessments. The issues stated are quite detrimental to the green environment and somehow, it has led to contamination in Malaysia as well. With the increasing concern towards environmental pollution and growing interest in suitable development, the problems of heavy metal contaminations have become more significant (Gollmann et. al., 2010). The rise of contamination rates, especially soil contamination, is considered to create a significant threat to humans and the earth's ecosystem. According to these problems, the EU and the UK legislation has recently encouraged the use of remediation techniques in order to ensure the site or land in safe condition for human activities (Harbottle et al., 2007).

Remediation techniques that are not too high in technology and low input are urgently required to provide cost-effective and environmentally effective solution for soil contamination (Fauziah et al., 2013). Primarily, there are a lot of remediation techniques that have been practiced, such as the stabilization/solidification (S/S) technique, electro-kinetic technique, phytoremediation technique and in-situ immobilization technique. Among the techniques mentioned, the S/S technique has been utilized effectively, and is extensively used in developed countries for the past decade for treatment of heavy metal wastes and contaminated soils (Yin et al. 2006).

1.2 Background of Study

Stabilization/Solidification (S/S) is an established technique used for treating industrial waste sludge prior to proper landfill disposal. “Solidification” refers to improving physical integrity of waste sludge in order to facilitate handling, while “stabilization” refers to the reduction of the mobility of contaminants via various mechanisms such as precipitation, chemisorption, encapsulation and ion exchange (Kumpiene et. al., 2008). S/S technique was first used for treatment of radioactive waste in the 1950s and has demonstrated the best available technique by the US Environmental Protecting Agency (U.S EPA) for land disposal of toxic waste (Voglar & Lestan, 2010). Additionally, S/S technique is routinely used for the final treatment of hazardous waste to reduce contaminant leaching prior to land disposal.

S/S technique consists of binders mixing with sludge and the addition of water which is then cured for several days (Erdem & Ozverdi 2011). Among various types of binders, cement-based systems are the most widely used, due to its relatively low cost, wide availability and versatility (Gollmann, et. al., 2010). For example, a study by Bonen & Sarkar, (1995) stated that incorporating metals such as Ni, Pb and Cd with cement results in the decreasing of the $Ca(OH)_2$ content which increases its vulnerability. Another research from Voglar & Lestan, (2010) shows that the application of Ordinary Portland Cement (OPC) in S/S technique has decreased the concentration of Cd, Pb, Zn and Ni compared to the original soil.

However, in recent years, due to the consequence of high energy consumption in manufacturing cement and the air pollution caused by the release of high quantities of greenhouse gases during its production, the cement industry has been pointed out as one of the major contributors to anthropogenic CO_2 emissions of about 5% globally (Oh et. al., 2013). In this respect, several researches have been directed towards partial or total substitution of Portland cement by pozzolanic binders such as lime, fly ash, and natural pozzolan.

By referring to Massardier et. al., (1997), fly ash is currently one of the most common binders in waste stabilization, and it is available in mainly two kinds of mixture which is Portland cement added with fly ash or lime added with fly ash. Fly ash was selected because of the ettringite formed in the solids in long-term leaching experiments and the associated reduction in leachate concentration in the trace element. Another research by Pereira et. al., (2001) it describes the S/S technique adopted in steel industry to treat the waste using a common type of fly ash has successfully stabilized the concentration of Pb, Cd and Zn in TCLP leachate.

However, when the availability of fly ash is limited, the use of other waste materials are necessary, for example, the physical and mechanical properties of a sandy soil mixture with rice husk ash (RHA) and lime cured for 28 days, as reported by (Alhassan & Mustapha, 2007). In the same study, the author stated that the compressive strength of the mixture containing the RHA was several times higher than the controlled sample, and wetting and drying tests showed improvement with the use of RHA. In addition, the XRD results confirmed the formation of cementing products such as C-S-H as a result of the reaction between the Ca ions with the amorphous silica of the ash. These products were suggested to be responsible for the stabilization of the soil.

Nevertheless, the latest research shows interest in replacing cement with agricultural waste to substitute cement and lime in S/S technique. To enable a more cost-effective S/S treatment design, a lignocellulose and non-lignocellulose crop residue has been used which is typically free of charge (Madurwar et. al., 2013). Therefore, the utilization of this material in making cement-bonded materials offer an attractive alternative at their disposal. For this purpose, lignocellulose and non-lignocellulose crop residue such as straw, corn cobs, sugarcane bagasse, banana waste, pineapple waste, coffee pulp and others have a number of suitable criteria such as low density, low requirements of processing equipment, negligible abrasion to the processing machinery and abundant raw material availability (Asan et al. 2008). Furthermore, these materials can be effectively encapsulated in a cementitious matrix as it is known that plant-based fibers have been used with considerable success with inorganic binders like ordinary Portland cement (OPC).

1.3 Problem Statement

The waste management sector is one of the main contributors to environmental pollution in Asia. The intensity of the issue is immense in developing countries such as India, Indonesia and Malaysia. Whereas, developed countries such as Korea and Japan have sustainable waste management in practice (Agamuthu et. al., 2013). Due to the increasing numbers of waste generated by industries in Malaysia, it has contributed to the illegal dumping. Although Malaysia has a lot of landfills, the number still not sufficient to accommodate the increase of waste produced. Most of these sites will be full or cannot be used within two years and not surprising, it may contribute to hazardous contamination in the water, air and soil (Tarmizi, 2009).

Mining, smelting and various industry activities were identified as the factors that contribute to land contamination (Yukselen & Alpaslan, 2001). Emphasizing on land contamination, inorganic waste have high revenue potential in disrupting the ecosystem, soil and groundwater. Prior to 1960s, research was focused on enhancing the plant uptake or availability of selected heavy metals or minor elements from the soil. Recently, concerns regarding heavy metal contaminations in the environment affects all ecosystem components, including aquatic and terrestrial systems, and they have been identified with increasing efforts on limiting their bioavailability in the dangerous zone (Bolan et. al., 2014). Additionally, many sites have been identified as hazardous waste sites because of the presence of high concentrations of heavy metals in the soil. The total mass of metals in surface soils is an important factor which influences their migration in the soil to the groundwater. Although some of them act as essential micronutrients for living beings, at higher concentrations they can lead to severe poisoning (Kim, 2003). The most toxic forms of these metals in their ionic form are the most stable oxidation states e.g. Cd^{2+}, Pb^{2+}, Hg^{2+}, Ag^{+} and As^{3+} in which, they react with the body's bio-molecules to form extremely stable bio-toxic compounds that are difficult to dissociate (Duruibe et. al., 2007). Unlike organic contaminants that can be destroyed (or mineralized) through treatment technologies, such as bioremediation, metals contaminants still persist in the environment. Once a metal has contaminated the soil, it will remain as a threat to the environment until it is removed or immobilized (Harbottle et. al., 2007).

To address this problem, several techniques have been developed such as Stabilization/Solidification (S/S) technique, incineration technique, Electro kinetic remediation technique, immobilization technique and biological treatments. From all the technologies mentioned, Paria & Yuet, (2006) suggested that the S/S remediation technique provides a viable and relatively economical technique and are particularly effective to heavy metal fixation and immobilization.

The widespread application of S/S technology is due to the widely available common and inexpensive additives and reagents used. The results obtained from solidified materials may require more or no further treatment if proper conditions are maintained. However, the volume of treated materials may increase due to the addition of binder (Awal & Abu Bakar, 2011). As the amount of binder increases, the cost of operations also increase. Due to this situation, most of the waste generators neglect to apply this technique. As such, most of them refuse to do so and illegally store their sludge within their premises or dispose their sludge in nearby areas (Yin et. al., 2006).

The S/S studies in recent years focuses on the usage of recyclable waste materials to substitute cement and lime. In order to enable a more cost-effective S/S treatment design, S/S specialist often substitute portions of S/S binder with industrial wastes such as incinerator bottom ash (Wang et. al., 2015) and fly ash (Tarmizi, 2009). Nevertheless, latest research has shown interest in replacing cement with agricultural waste such as corn cob ash, banana waste, coffee pulp, palm ash, rice husk ash, compost teas, sugarcane bagasse, pineapple waste and others. Therefore in this research, the agricultural waste of sugarcane bagasse will be investigated for its suitability to replace cement in S/S technique. The usage of these wastes would represent a two-pronged approach in solving disposal problems as well as providing a cost-effective cement replacement material.

1.4 Research Objectives

The aim of this study is to evaluate the performance of sugarcane bagasse as a binder. Research objectives to be achieved in this study are:

1) To determine the chemical and physical characteristics of clay soil, cement, and sugarcane bagasse.

2) To investigate the physical behavior (strength, density and water absorption) of cement incorporated with sugarcane bagasse as an additive in the S/S method to immobilize lead contaminated soil.
3) To determine the relationship of strength, density and water absorption of cement incorporated with sugarcane bagasse as an additive in the S/S method.
4) To examine the pH and leachability of lead from the contaminated soil through the S/S method using sugarcane bagasse as partial replacement of cement.

1.5 Research Scope

This research mainly focuses on the remediation of artificially contaminated clay soil, where lead is chosen as the contaminant. The soft clay has been chosen and taken at Research Center for Soft Soil (RECESS), UTHM. The combination of cement with agricultural waste of sugarcane has been selected as the binders by using S/S technique. The characteristics of clay soil, cement, and sugarcane bagasse by using X-Ray Fluorescence (XRF) and Scanning Electron Microscopy (SEM) were noted. In this study, sugarcane bagasse was partially added to the cement in ratio of cement as additive to remediate the contaminated soil. To achieve the second objective, unconfined compressive test, density test and water absorption test was conducted in order to obtain the strength and water absorption of the sample. Furthermore, leaching test were conducted to obtain the leaching characteristics of the each sample. All the leaching tests were discussed briefly in the research methodology. The samples were cured for 7, 14 and 28 days prior to the unconfined compressive test and the leaching crush leaching test.

1.6 Significant of Study

This study was carried out to investigate the effectiveness of cement as an additive incorporated with sugarcane bagasse using the S/S method to remediate lead contaminated soils. This research generates a number of findings which can be applied towards improving the S/S technique. The application of sugarcane bagasse, from the agro-waste in its raw form is predominant in developing countries such as

Malaysia, as an alternative approach in solving disposal dilemma as well as providing an inexpensive cement replacement material.

Furthermore, this study is important to scientists and environmental engineers in public or private sectors to plan the best way to dispose hazardous waste containing heavy metals to landfill. Moreover, this research might also assist the local authorities to find alternative solutions in protecting the environment from hazardous pollutants such as lead. It also can be used as a guideline for other researchers to find the effective materials that can be used as an additive in the S/S technique for soil that is contaminated by lead.

1.7 Concluding remarks

This research was to investigate the performance of using agricultural waste product as a partial replacement material for the cement in the S/S remediation method for soil contaminated by lead. The performance of S/S samples were determined in terms of strength and water absorption as well as the leachability of heavy metals. It is expected that this study will practically reduce the amount of cement used by adding an amount of agricultural waste while increasing the effectiveness of the S/S technique. The concentration of lead contamination in the soil is expected to be reduced as agricultural waste is added to the cement base in S/S technique.

CHAPTER 2

LITERATURE REVIEW

2.1 Introduction

Environmental contaminations has become a serious issue worldwide. It involves contamination in various medium such as soil, water and air. Between these three medium, soil contamination has been reported to be the most dangerous and most threatening due to the fact that contaminants have the capacity to affect human health and destroy the food chain (Fauziah et. al., 2013). The contamination is mainly due to the large number of industrial activities, disposal of municipal solid wastes, urbanization activities and agricultural wastes.

Nowadays, over 80% of hazardous wastes come from industrial activities (Napia, 2012). Sludge, heavy metal, oil and other hazardous wastes are noted to be found in abundance. The amount of these industrial wastes from industrial plants increase every year. Among these hazardous wastes, heavy metal contamination is considered to be the worst due to their harmful effects and long-term persistence in the environment (Kamari et. al., 2011).

The contamination in soil by heavy metals, particularly lead (Pb), is a common problem throughout the world (Halim et. al., 2005). Lead has been reported as a metal that affects the human nervous system, blood vessels and kidneys. Currently, governments are trying to minimize the adverse impact of lead which affects mental and physical development in humans, even at the lowest level of exposure (Aslam et. al., 2013). Therefore, remediation of these contaminated soil becomes a great concern for both engineers and researches.

Currently, there are several remediation methods that have been implemented, such as the stabilization/solidification (S/S) technique, electro-kinetic technique, phytoremediation technique and in-situ immobilization technique. Among these methods, the S/S technique has been utilized as a promising technology with the addition of binding agents to encapsulate and reduce the mobility of hazardous waste elements at low cost, in wide availability and versatility (Luna Galiano et. al., 2011).

According to Grega & Domen (2011), the S/S method has emerged as an efficient technique for the treatment of sites that are contaminated with potentially toxic metals. Other research that were done by Lasheen et. al., (2013) stated that heavy metal wastes normally needs S/S method processes to reduce contaminant leaching prior to landfill disposal. Another research by Yao et. al., (2012) mentioned that the S/S technique is commonly used to reduce the mobilization of contaminants within a hardened mass (solidification) and chemical conversion of contaminants into less soluble form (stabilization). In the same way, Hunce et. al., (2012) defines the S/S method as a technique that aims in immobilizing contaminants by converting them into a less soluble form and encapsulating them with the creation of durable matrix.

2.2 Soil contamination

Soil is a basic environmental element that constitutes the ecosystem and is an important basic material for the survival and development for human beings (Yao et. al., 2012). Thus, this medium is considered a highly potential medium that is easily exposed to contamination. A study by John et. al., (2011), describes land contamination as areas with high concentration (above normal background level) of substance, which may have arise from previous land use. Soil contamination especially by heavy metals pose a major environmental and human health problem that is still in need of an effective and affordable technological solution. The main causes for these problems are from a large number of industrial activities which produce wastes and contaminants that reach the soil through direct disposal, emissions and other pathways (Grega & Domen, 2010). Figure 2.1 shows the schematic of soil contamination.

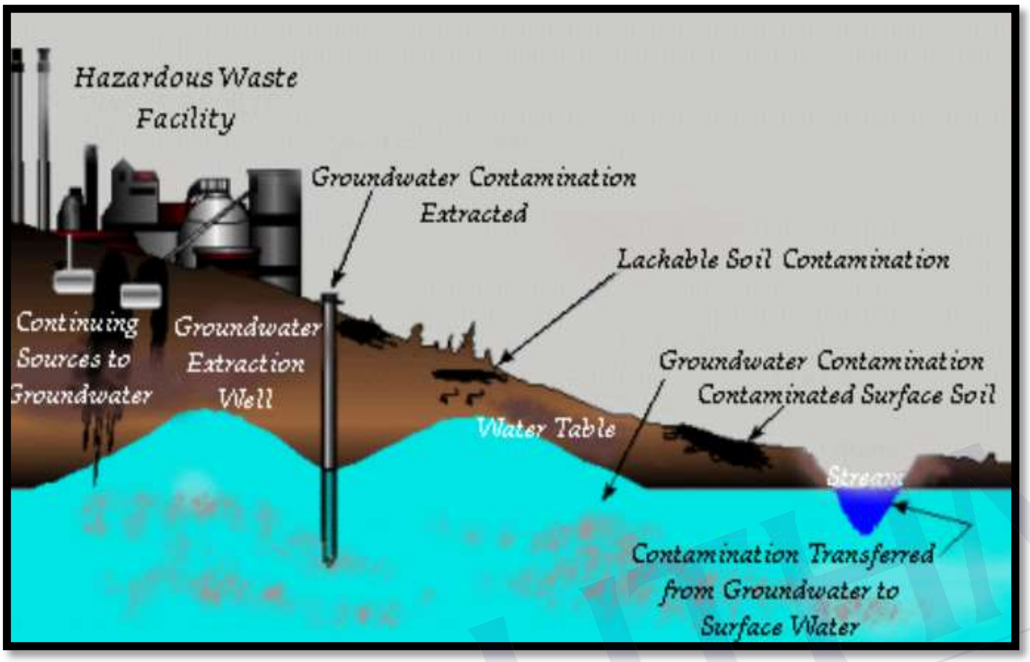

Figure 2.1: Schematic of soil contamination (Guo et., al. 2007)

Moreover, soil properties are affected by past land use, current activities on the sites and the nearness to pollution sources. Human activities have intentionally added substances such as pesticides, fertilizers and other amendments to soil (Du et. al., 2014). Additionally, accidental spills and leaks of chemicals used for commercial or industrial purposes have been sources of contamination.

Furthermore, improper waste disposal and mismanagement of soil is one of the main contributors to environmental pollution (Foo & Hameed, 2009). In Asia, sustainable waste management are being practiced especially in developed countries such as Korea and Japan. While in the developing countries such India, Indonesia and Malaysia, these issues are still dilemmas that seems hard to be solved. In Malaysia particularly, current rate of municipal solid waste (MSW) has exceeded 19,000 tonnes daily. With the lack in waste recycling has made matters worse (Agamuthu & Fauziah, 2012).

2.3 Impact of contaminants in soil composition

Contaminants mostly contribute to negative impacts to the soil. Even the contaminants occur naturally in soil, the amount of substances may exceed the levels recommended for the health of humans, animals and plants. Once contaminants are in the soil composition, where they go and how quickly they travel depends on many factors. Some organic (carbon-based) contaminants can undergo chemical changes or degrade into product that may be more or less toxic than the original compound (Antemir et. al., 2010). In the same way, chemical elements such as metals cannot be destroyed but the characteristics will be changed and may be more or less easily taken up by plants or animals (Du et. al., 2014).

In addition, different contaminants vary in their tendency to end up in water held in the soil or in the underlying groundwater from leaching through the soil. There are certain characteristics of the soil that shows it has been affected by contaminants (Akcil et. al., 2015). The important characteristics that may be affected by contaminants include soil mineralogy and clay content (soil texture), pH (acidity) of the soil, amount of organic matter in the soil, moisture levels, temperature and presence of other chemical (Fauzi et., al. 2013).

2.4 Heavy metals

Heavy metals is referred to any metal or metalloid that is of environmental concern. The term originated from the harmful effects of Arsenic (As), Cadmium (Cd), Chromium (Cr), Mercury (Hg), Lead (Pb) and Thallium (Ti) (Guo-li et. al., 2007). Heavy metals in the environment cannot be decomposed by organism. According to Fu & Wang, (2011), heavy metals can be accumulated gradually and transformed into more toxic metal compounds, which produces adverse reactions through bio-magnification of the food chain at all levels of organism in the ecosystem, being harmful to humans and other life forms.

There are many sources that are subjected to heavy metal production. As mentioned by Xi et. al., (2014), heavy metals are widespread in urban/rural and industrial areas as a consequence of industrial and agricultural activities such as metal mining, smelting and refining, gasoline processing, automotive exhaust emissions, as well as the application of fertilizers and agricultural chemicals.

2.4.1 Toxicity of heavy metals

Heavy metals such as Lead (Pb), Cadmium (Cd), Mercury (Hg) and Arsenic (As) pose environmental and human health problems that are still in need of an effective solution. In small quantities, certain heavy metals are nutritionally essential for a healthy life but it can become toxic when the heavy metals are not mobilized by the body and accumulate in the soft tissues. In the same way, heavy metals may enter the human body through food, water, air or absorption through the skin when they come in contact with human in agriculture and manufacturing industries, or even in industrial and residential settings (Habib et. al., 2012). Table 2.1 shows the permissible limit and health effects of various toxicity from heavy metals poisoning.

Table 2.1 Permissible limit and health effect of various toxic of heavy metals

Metal Contaminant	Permissible limits by International bodies (mg/L)		Health Hazard
	WHO limit for drinking water (2010)	US EPA (1993)	
Arsenic	0.01	5	Carcinogenic, producing liver tumors, skin and gastrointestinal effect.
Mercury	0.001	0.2	Corrocive to skin, eyes and muscle membrane, dermatitis, anorexia, kidney damage and severe muscle pain.
Cadmium	0.003	1	Carcinogenic, cause lung fibrosis, dyspnea and weight loss
Lead	0.01	5	Suspected carcinogen, loss of appetite, anemia, muscle and joint pains, diminishing IQ, cause sterility, kidney problem and high blood pressure
Chromium	0.005	5	Suspected human carcinogen, producing lung tumors, allergic dermatitis
Nickel	0.02	-	Causes chronic bronchitis, reduced lung function, cancer of lungs and nasal sinus
Zinc	3	-	Causes short-term illness called “metal fume fever” and restlessness
Copper	2	-	Long term exposure causes irritation of nose, mouth, eyes, headache, stomachache, dizziness, diarrhea

2.4.2 Heavy metal contaminated soil

Heavy metal contaminated soil is a worldwide problem that urgently needs to be solved. Heavy metal in soil can threaten people's health either by accidental soil ingestion, by breathing the contaminated soil dust particles or by the ingestion of polluted drinking water or farm product associated with contaminated soil (Yin & Shi, 2014). On the other hand, heavy metal contaminated soil is mainly due to the subsequent migration of leachate forms and within the landfill waste cells. According to Agamuthu and Fauziah (2012), natural processes such as infiltration within the boundaries of the waste cells can accelerate the process of heavy metal leaching from waste components that are sources of heavy metals within the landfill system. The released of heavy metals into the adjacent environment is a serious environmental concern and a threat to public health and safety.

Kamari et. al., (2011), stated that heavy metal behaviour in soil and biological effect caused by their presence in elevated concentrations are in fact strongly determined by the processes of metal released from the solid phase into soil solution as well as the factor that influences the chemical forms of the metal in soil.

2.4.3 Lead (Pb)

Lead is one of the chemical element in the carbon group with the symbol (Pb) and atomic number 82. Lead is usually a bluish-white lustrous metal which is very soft, high malleable, ductile and a relatively poor conductor to electricity. Lead is a major constituent of the lead-acid battery used extensively in car batteries. It has been used as a coloring element in ceramic glazes, as projectiles and in some candles to treat the wick (Omar et al. 2012). Additionally, lead is traditionally a base metal for pipes and used as electrodes in the process of the electrolysis. Due to its malleability, lead is considered as one of the oldest metals used by humans for different purposes like in the manufacture of lead-acid storage batteries, alloys, plumbing, cable covering, heavy machinery and recently, it is used as an important source in gasoline combustion (Hale et. al., 2012).

2.4.4 Pb in environment

Lead is persistent in the environment and accumulates in soils and sediments through deposition from air sources, direct discharge of waste streams to water bodies, mining, and erosion (Kim, 2003). Ecosystems that are near point sources of lead demonstrate a wide range of adverse effects including losses in biodiversity, changes in community composition, decreased growth and reproductive rates in plants and animals, and neurological effects in vertebrates.

Pb is found in ore with zinc, silver and copper and has been extracted together. According to Yan et. al., (2014), lead occurs naturally in the environment. However, most of the lead concentration that is found in the environment is the result of human activities such as application of lead in gasoline and an unnatural lead-cycle. Furthermore, in car engines, the lead that was burned, produces lead oxide. This oxide will enter the environment from the car exhaust (Ogundiran et. al., 2013). The process suggests that the largest particles will drop to the ground immediately and pollute the soil or water surface, while the smaller particles will travel long distance through air and remain in the atmosphere.

Finally, parts of this lead, either small or large particles, will return back to earth when it rains. Furthermore, this cycle caused by human activities and production is considered more detrimental than the natural lead cycle and becomes the biggest worldwide issue (Gollmann et. al., 2010). Kamari et. al., (2011) studied lead contamination in mango, guava and papaya grown on ex-mining land in Malaysia and found that the concentration of lead in the fruits exceeded the Malaysian Food Act permissible limits.

2.4.5 Pb impact to the human health

Humans may be exposed to lead and chemicals that contain lead via air, drinking water and food. Once taken into the body, lead distributes throughout the body through the blood and is accumulated in the bones (Gollmann et al. 2010). Depending on the level of exposure, lead can adversely affect the nervous system, kidney function, immune system, reproductive and developmental systems and the cardiovascular system. Lead exposure also affects the oxygen carrying capacity of the blood. Yap et al. (2002) mentioned the lead effects that are most commonly encountered in current populations are neurological effects in children and cardiovascular effects such as high blood pressure and heart disease in adults. Infants and young children are especially sensitive to even low levels of lead, which may contribute to behavioral problems, learning deficits and lowered IQ.

Moreover, lead is distributed to many tissues and organ systems of the body. It is important to note that lead cannot be destroyed or changed to something else in the body. The amount of lead stored in the body has been described as a "body burden" by lead. Among adults, over 95% of lead is stored in bones. Meanwhile, for children, about 70% of lead is stored in bones (Ismail et al. 2013). This lead is not simply stored away in bones forever, but moves in and out as the body functions normally. For example, as children grow their bones restructure to permit normal shapes as they develop.

2.5 Remediation of heavy metal

Heavy metal contaminations in soil are causing a serious threat to the environment and human health. However, there are several technologies that have been developed to treat and remediate the contaminated soil. Remediation technologies can be classified according to immobilization or extraction (action that is applied to metals), in-situ or ex-situ (location that is applied to metals) and other types of technologies (Dermont et al. 2008). Figure 2.2 shows a schematic of remediation technologies for metal contaminated soil.

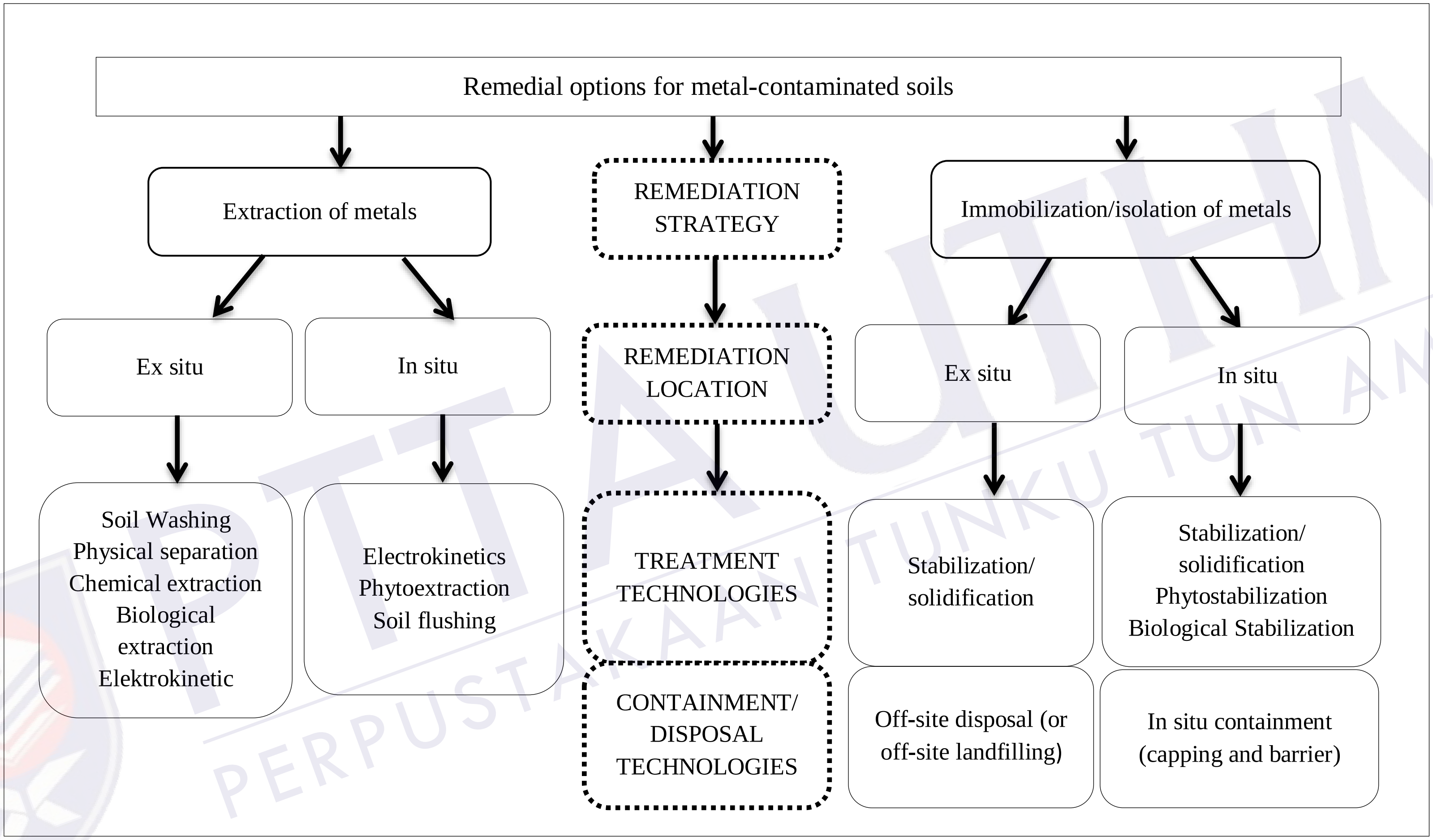

Figure 2.2: Schematic of remediation technologies for metal contaminated soil

The treatments of heavy metal contaminated soils are limited to two main strategies which is immobilization and extraction. Immobilization approaches aim at stabilizing the metals by minimizing the leaching characteristics of the soil matrix and change the metals to less soluble, toxic or bioavailable form in the soil to reduce the risks of human health and the environment (Tantawy et al. 2012). While, extraction approaches are referred to a process that separates the metals from the soil's composition, reduce the concentration of metals as well as reduce the volume of the entire contaminated medium. According to Wang et al. (2014), extraction treatments aim in completely decontaminating the soil by removing the metals from the soil matrix. However, when the metals and the soil matrix are strongly bound together, the extraction treatment is emphasized on reducing the metals concentrations to an acceptable level.

2.5.1 Type of Available Remediation Technique

According to Yao et al. (2012), there are three types of remediation techniques that are suitable for heavy metal contaminated soil and they are physical remediation, chemical remediation and biological remediation. Physical remediation mainly includes soil replacement method and thermal desorption (Chen et. al., 2010). Soil replacement means using clean soil to replace or partly replace the contaminated soil with the aim of diluting the concentration of pollutants, increase the soil environment capacity and thus, remediate the soil. While, Shi et. al., (2009) stated that thermal desorption is based on pollutant volatility where and the contaminated soil is heated using steam, microwave, or infrared radiation until the pollutant is volatile. The volatile heavy metals are then collected using vacuum with negative pressure to remove the heavy metals.

Chemical remediation is divided into 3 types which are chemical leaching, chemical fixation and Electrokinetic remediation. Firstly, chemical leaching is a process of washing the contaminated soil using fresh water, reagents and other fluids or gases that can leach the pollutant from the soil. According to Khan et. al., (2004), heavy metals in soil are transferred from soil to liquid phase through ions exchange, precipitation and adsorption process in chemical leaching remediation. Secondly, chemical fixation is a process of adding reagents or materials into the contaminated soil and using them with heavy metals to form insoluble or hardly movable, low toxic matters, thus decreasing the migration of heavy metals into water, plants and other environmental media and achieving the remediation of soil (Yao et al. 2012).

Besides that, the electrokinetic remediation technique or EK is a new remediation technique which is mainly applying voltage at the two sides of the soil and then forming electric field gradient. According to Syakeera et al. (2013), basically, this technique is to improve the volume stability of the soil around and beneath the foundation. This technique involves applying an electrical current across the soil mass to boost the chemical migration from the injection point with the purpose of reacting beneficially with the soil to bring about an improvement in its properties.

Another promising technology in soil remediation is biological remediation. The biological remediation is a process of changing the physical and chemical characteristics through migration and transformation process of heavy metals by microorganisms (Hakeem et. al., 2015). The remediation mechanisms include extracellular complexation, oxidation-reduction and intracellular accumulation. Additionally, the microbial leaching by microorganism is a simple and effective technology for extracting valuable metals from low-grade ores and mineral concentrates. In the same way, Yao et al. (2012) argued that the microbial leaching has some potential in remediation of mining sites, treatment of mineral industrial waste products, detoxification of sewage sludge and for remediation of soils and sediments contaminated with heavy metals.

Besides that, phytoremediation is also a part of biological remediation. According to Oosten & Maggio (2014), phytoremediation is a remediation technique that uses living green plants to fix or absorb and clean the contaminants or reduce the risk provide by heavy metals. The phytostabilization, phytovolatilization and phytoextraction are the main three types of phytoremediation (Surriya et. al., 2015). Phytostabilization is referred to a fixing of metals using plants through adsorption, precipitation and reduction of roots, and thus reducing their migration into the groundwater and food chain. Despite of phytostabilization, phytovolatilization involves transferring heavy metals into a volatile state or adsorption of the metals in gaseous matter by using special agents secreted by the roots of the plants. While, phytoextraction involves adsorption of the heavy metals using tolerant and accumulating plants by transferring and storing at ground parts. Table 2.2 summarizes the advantages and disadvantages of available remediation technologies.

Table 2.2 Summary of advantages and disadvantages of available remediation technologies (Babel & Dacera, 2006)

Technology	Description	Advantages	Disadvantages
Stabilization/ solidification (S/S)	S/S mostly based on cement process aims at stabilized and solidified the metals is a strongly modified soil matrix	S/S is applicable to a wide range of mixed contaminants and soil types	S/S process increases the volume of treated materials
In-situ chemical stabilization	In situ chemical stabilization aims to reduce metal bioavalability/solubility without affecting the soil matrix	In situ chemical stabilization may promote site revegetation and can be applied for a large site	Requires the chemical agent which is expensive and hazardous
Phytoremediation	In situ emerging technology that uses plants to prevent soil erosion (by wind and rain), to stabilized metal in order to avoid metals migration to groundwater	Potentially applicable for many metals. Large area can be treated. No disposal of contaminated biomass required	Application limited to depth of the root zone. Remaining liability issues, including maintenance for an indefinite period of time. Requires controlling of site use.
Electrokinetics	Technique that uses electrochemical processes to remove metals from (saturated) soils. In situ option is more interesting rather than ex situ approach	Metals can be effectively removed from soils via in situ approach. Potentially applicable for broad type of metals	Applicable only for saturated and partially saturated (clay and silt clay) soils. Multi-metals contaminated sites pose problems.
Biological Extraction	In situ emerging technology that uses bio-solids or microbial activity to reduce metals toxicity or bioavailability for the environment. This technology is often associated with chemical stabilization	Metals bioavailability for human and biological receptors is reduced. Potential re-vegetation of the site	Requires more pilot studies to evaluate the efficiency. Remaining liability issues, including maintenance for an indefinite period of time.

2.6 Stabilization/Solidification method

Stabilization/Solidification (S/S) is typically a process that involves a mixing of waste with binders to reduce the volume of contaminant leachability by means of physical and chemical characteristics to convert waste in the environment that goes to landfill or others possibly channels (Hunce et. al., 2012). Stabilization is attempts to reduce the solubility or chemical reactivity of the waste by changing the physical and chemical properties. While, solidification attempt to convert the waste into easily handled solids with low hazardous level (Malviya & Chaudhary 2006). These two processes are often discussed together since they have a similar purpose of improvement than containment of potential pollutants in treated wastes. The combination of stabilization and solidification is often termed as “waste fixation” or “encapsulation” by researchers around the world (Voglar & Lestan, 2010).

Solidification of waste materials is widely used for the disposal of radioactive waste. Many developments relating to solidification originated from low level radioactive waste disposal (Erdem & Ozverdi, 2011). Regulation that relates to the disposal of radioactive waste requires a change of the waste into a free-standing solid with a minor amount of free water. Most of the processes were utilized for nuclear waste, including a step in which granular ion exchanges with the liquid and waste phase are often used in the incorporation of solid matrix with cementing or binding agents such as Portland cement, organic polymer or asphalt. It shows good results with relatively low permeability, low concentration and reduces the surface area across which pollutants was transferred (Yoon et. al., 2010).

In addition, in hazardous waste disposal and site remediation, treated material must achieve certain standards for safe land disposal by removing the hazardous characteristics, especially in Malaysia (Ahmaruzzaman, 2010). For toxic characteristics, this usually requires passing concentration-based standards using the US EPA TCLP test (Harbottle et. al., 2007). To accomplish this goal, a variety of strategies may be used to prevent contaminant leaching, including neutralization, oxidation/reduction, physical and chemical effects from the contaminant. Appropriate treatment strategies must be taken to ensure the effectiveness of this technology, where appropriate binder selection must be benchmarked as the key to success (John et. al., 2011).

In the S/S technique, a binder is often used to stabilize the contaminants in the waste or contaminated site and to remove the free liquid (Paria & Yuet, 2006). In cases where the waste is extremely soluble or no suitable chemical binder can be found, the waste may be contained in an encapsulated condition in some hydrophobic medium such as asphalt or polyethylene. This may be done either by incorporating the waste directly in the partially molten material or by forming jackets of polymeric material around blocks of waste (Ponou et. al., 2011).

Portland cement is the most used binder for the S/S technique. S/S with cement is relatively common due to the universal availability, desirable hydration properties, which is appropriate and suitable for heavy metal immobilization (Harbottle et al., 2007, Chen et al., 2009, Kogbara et al., 2012, Du et al., 2014). It is particularly suitable for heavy metal remediation and has been applied widespread over several decades, especially in the U.S. Figure 2.3 shows the distribution of the latest technology to treat hazardous waste and contaminated soil in U.S.

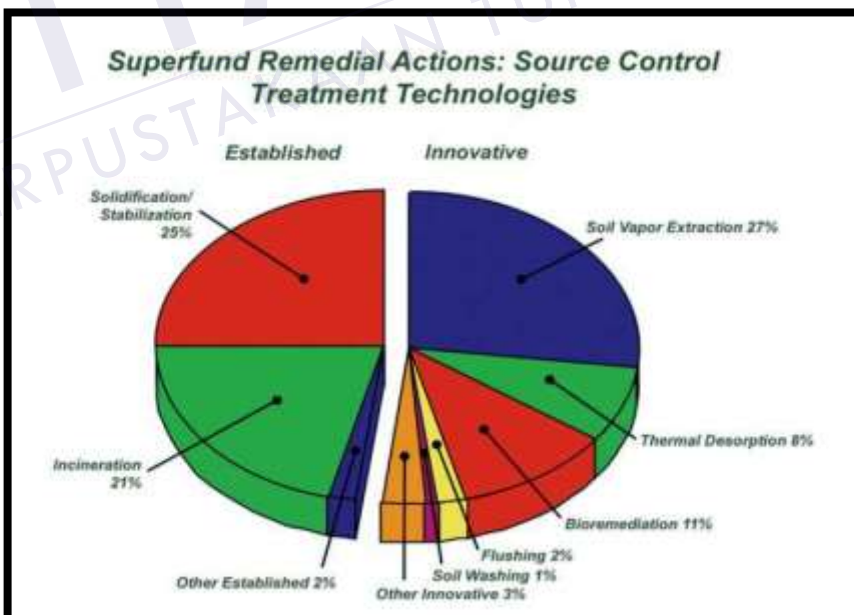

Figure 2.3: Frequency of application of S/S treatment compared to other technologies at U.S. Superfund sites (USEPA, 2001)

U.S EPA has identified the S/S technique as the Best Demonstrated Available Technology (BDAT) for 57 type of hazardous waste listed in Resource Conservation and Recovery Act (Wadanambi et. al.,2008). According to U.S EPA 2001, about 25% of the established superfund remediation sites were treated by S/S technique. Compare with others technologies, cement-based S/S has the following advantages.

- Relatively low cost
- Good long-term stability, both physically and chemically
- Good impact and compressive strength
- Material and technology well known
- Widespread availability of the chemical ingredient
- Non-toxicity of the chemical ingredient
- Ease of use in processing (processing normally conducted at ambient temperature and pressure with any unique or very special equipment)
- High waste loading possible
- High resistance to biodegradation
- Relatively low water permeability
- Good mechanical and structural characteristic
- Low cost because the reagents are widely available and inexpensive
- Can be used on a large variety of contaminants
- Can be applied to different types of soils

2.6.1 Process involved in S/S method

The processes and techniques used in S/S has been accepted and have became an important part of environmental technology worldwide. Stabilization refers to the techniques that reduces the hazard potential of waste by converting the contaminants into their least soluble, mobile or toxic form (Malviya & Chaudhary 2006). The physical nature characteristics of waste are not necessarily changed through stabilization. While, solidification refers to a technique that encapsulates the waste in a monolithic solid of high structural integrity.

The S/S method is usually applied by mixing contaminated soils containing treatment residuals with a physical binding agent to form a crystalline, glassy, or polymeric framework surrounding waste particles (Hebatpuria et. al., 1999). Navarro Blasco et al. (2013) argued that the other form of S/S treatment relies on micro-encapsulation where waste is unaltered but macroscopic particles are encased in a relatively impermeable coating or on specific chemical fixation, where contaminants are converted into solid compound that is resistant to leaching. In addition, the macro-encapsulation involves certain chemical fixation mechanisms to improve resistivity of waste leachate.

Moreover, the S/S treatment can be accomplished primarily through the use of either inorganic binders (cement, fly ash or furnace slag) or by organic binders such as bitumen. Normally, the processes in the S/S method are divided into two parts, chemical processes and physical processes (Babel & Dacera, 2006). Chemical processes actually requires a chemical reaction to take place to allow the process to perform. In this process, the chemical reaction may consist of something as simple as acid neutralization to provide an alkaline environment, or may involve complex speciation reactions (Singh & Pant, 2006). In addition, most chemical processes involve solidification reactions from cement or pozzolanic materials; these reactions are very complex.

Physical processes do not involve chemical reactions. The process operates by adsorbing or absorbing constituents on surfaces or in pores, or encapsulating it in a matrix that coats the constituent particles and disperses them within it, while physically separating the hazardous constituents from the environment (Yukselen & Alpaslan, 2001). Within this comprehension, the polymerization of a thermo-setting polymer that results in microencapsulation of the waste constituents is not considered a chemical process because it does not interact chemically with the waste.

2.6.2 Overview of soil remediation by S/S method

Soil contamination from heavy metals has become a very serious environmental problem, mainly caused by rapid developments due to urbanization around the world (Sun et., al. 2010). Therefore, remediation of these contaminated soils becomes a concern among researchers. As mentioned in chapter 1, the S/S method is widely used in the remediation practice to reduce the release of contaminants and enhance soil strength due to its convenience and cost-effectiveness.

Cement-based S/S technology has been shown to be effective in immobilizing the heavy metals even without additional additives (Napia et. al., 2012). Du et al. (2014) has studied the leaching behavior of Pb contaminants by using OPC as a binder. This research concluded that at pH 2.0, this strongly acidic condition has resulted in substantial lowered leachate pH and significantly increased the amount of Pb leached. Contradictory to the condition, when OPC was added in S/S sample from 12% to 18%, it resulted in a decreased amount of Pb leached. In another study, Li et al. (2014) concluded that Pb concentration has been leached out from the solidified specimens using OPC as a binder at 109, 83 and 71 mg respectively with cement ratio of 0.2, 0.3 and 0.4. Another research by Wang et. al., (2014) has showed an excellent capacity of OPC in remediating the contaminated soil at a 17 year-old site. This research found that the Toxicity Characteristic Leaching Procedure (TCLP) test sample containing Cu, Ni, Zn, Pb and Cd has satisfied the drinking water standard.

Malviya & Chaudhary, (2006) has also used the OPC to remediate soil that is contaminated by Pb, Zn, Cu, Fe and Mn. As a result, they argued that sample containing OPC at pH $\geq$ 12 leached less Pb. Then, it was observed that the concentrations of Zn, Cu, Fe and Mn were also decreased in alkaline conditions. This study concluded that the leachability of heavy metals studied are very pH dependent. Similar observation were reported by Voglar & Lestan (2010) which showed that concentrations of Cd, Pb, Zn and Ni decreased in alkaline conditions on TCLP extraction and met the regulatory limit for heavy metals in soils.

Furthermore, a part from OPC, there is an interesting study using Calcium Aluminate cement (CAC). Voglar & Lestan, (2011) studied the use of CAC and

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