REMOVAL OF ARSENIC FROM LANDFILL LEACHATE BY NATURAL SOIL ADSORPTION FOR POTENTIAL USE AS A PERMEABLE REACTIVE BARRIER

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A thesis submitted in fulfilment of the
requirements for the award of the degree of
Doctor of Philosophy

Malaysia-Japan International Institute of Technology
Universiti Teknologi Malaysia

OCTOBER 2022

DEDICATION

This thesis is specially dedicated to my mother, and my daughters, Iman, Nadine and Sumayya, who taught me not to give up, despite the struggles, and for loving me unconditionally.

ACKNOWLEDGEMENT

In the name of Allah, the most gracious and most merciful. I thank Allah for giving me the patience and ability to accomplish this work. I would like to convey my sincere thanks to my mother, and siblings, for all the du'as, their unwavering love and tremendous support throughout my journey. My brother Riyadh, for helping with the kids, and who has been supportive throughout the process of my PhD. My dearest friend Yamuna, who has never tired in listening to my dramas, and understood me without pretension. And most importantly to my daughters, Iman, Nadine and Sumayya, who inspired me to work harder, loved me without conditions and continue to be my source of joy and motivation.

I would like to express my special thanks to many individuals whose supports and helps make this study possible. I would like to take this opportunity to extend my deepest gratitude to my supervisors, Assoc. Prof. Dr Kenichi Yoneda, Dr Nor'Azizi Othman, Dr Zuhaida Mohd Zaki and Dr Amnorzahira Amir for their guidance, suggestions, and their patience and understanding during the course of this study. Also for their detailed advice in all aspects of the work completed. Special thanks also to Dr Amnorzahira for giving me permission to use the facilities in BIOREC, UiTM.

I have been grateful to Assoc. Prof. Dr Kenichi Yoneda, for the overall scope and focus, for all the tireless efforts in correcting my thesis and journal papers and the advice for further improvements. I would like to thank Dr Nor'Azizi, for giving me this chance and understanding my situations, and to Dr Zuhaida Mohd Zaki and Dr Amnorzahira Amir for their academic advice and teaching. Lots of thanks to University Teknologi Malaysia, for giving me the opportunity to complete my post-graduate studies and for financial support.

ABSTRACT

Landfills poses a potential threat to human health and environment due to detrimental effects of toxic pollutants. Leachate is one of the most serious environmental hazards associated with landfills. This study aims to identify the most significant heavy metal pollutant in leachate from landfills in Malaysia and to assess the potential of natural and locally available soil as reactive media for permeable reactive barrier (PRB) systems for leachate treatment. The data collection sites consist of seven landfills in Negeri Sembilan and Melaka. The leachate and soil from the seven landfills were analysed. The analyses of the leachate contamination potentials and environmental impacts of the landfills area involved the investigation of these indexes; quality rating scales (QRS), leachate pollution index (LPI), geo-accumulation index (Igeo), pollution index (PI) and integrated pollution index (IPI). In this study, ten types of locally available soil were screened for their capability to remove arsenic. Batch and column studies were conducted to determine the most effective soil for reactive media in PRB system in terms of adsorption capacity, retention and dispersion properties. Maximum QRS values of arsenic (787) and LPI of 15.28 in Ulu Maasop landfill denoted progressive deterioration of leachate contamination especially in non-sanitary landfills (dumpsites). The difference in IPI values for sanitary (IPI > 1) and for non-sanitary landfill soil (IPI < 1) confirmed advanced decline of the soil quality in non-sanitary landfills. Arsenic was identified as the pollutant of concern based on the contamination potential in leachate and the impacts to the soil in vicinity of the landfill. Ulu Maasop soil (UMS3) was selected based on high arsenic adsorption capacities (0.31 mg/g for initial concentration of 50 mg/L) and also removal of heavy metals in real leachate via batch adsorption test. From column studies, UMS3 soil showed to have retardation factor of 32.7 which may reduce mobility of arsenic in the contaminant plume. Mineral compounds such as hematite, baumite and calcite in UMS3 soil proved to have significant effects on arsenic removal. XPS findings revealed that mechanisms of arsenic adsorption onto UMS3 soil were probably via oxidation, surface complexation and ligand exchange with hydroxyl groups. Hence, UMS3 soil is a good candidate for reactive barrier system proposed for arsenic mitigation in landfill due to its high adsorption capacities, high retention and dispersive characteristics.

ABSTRAK

Tapak pelupusan sampah adalah ancaman kepada kesihatan manusia dan kelestarian alam sekitar, terutamanya daripada bahan toksik dan berbahaya. Larut lesapan pula adalah antara bahan pencemar utama yang terhasil terutamanya dari tapak pelupusan bukan sanitari. Kajian ini bertujuan untuk mengenalpasti bahan pencemar utama (logam berat) yang terhasil dari tapak pelupusan di Malaysia dan mengkaji potensi bahan semulajadi (tanah) sebagai bahan reaktif di dalam sistem rawatan larut lesap tembok penahan tereaktif (PRB). Pengumpulan data untuk kajian ini melibatkan tujuh buah tapak pelupusan di Negeri Sembilan dan Melaka. Larut lesapan dan tanah dari tujuh tapak pelupusan ini dianalisa. Untuk mengenalpasti kadar dan impak pencemaran tapak pelupusan ini, beberapa indeks iaitu skala indeks pencemaran larut lesap (QRS), indeks pencemaran larut lesap (LPI), geo-akumulasi indeks (Igeo), indeks polusi tanah (PI) dan indeks polusi terintegrasi (IPI) telah ditentukan. Bagi kajian ini, sepuluh tanah lokal telah disaring untuk mengenalpasti tahap kebolehserapannya terhadap arsenik. Kajian *batch* dan kolum dijalankan bagi menentukan tanah yang berpotensi untuk digunakan sebagai bahan reaktif di dalam system PRB dari segi keupayaan penyerapan, juga faktor retensi dan kadar penyebaran arsenik. Kadar QRS yang maksimum (787) dan LPI (15.28) didapati di tapak pelupasan Ulu Maasop menunjukkan kadar pencemaran yang progresif terutamanya dari tapak pelupusan bukan sanitari. Perbezaan kadar IPI di tapak pelupusan sanitari ($IPI > 1$) dan tapak pelupusan tidak sanitari ($IPI < 1$) memaparkan kesan pencemaran tanah di sekitar tapak pelupusan tidak sanitari. Tanah UMS3 dipilih berdasarkan keupayaan penyerapan arsenik yang tinggi di dalam larutan arsenik (0.31 mg/g untuk konsentrasi awal 50 mg/L) juga penyerapan logam berat di dalam larut lesap dari tapak pelupusan. Dari kajian kolum pula, tanah UMS3 menunjukkan faktor retensi arsenik sebanyak 32.7 yang mana ia berpotensi mengurangkan penyebaran arsenik di dalam liang tanah. Mineral seperti hematit, baumit dan kalsit di dalam tanah ini juga dilihat memainkan peranan yang penting di dalam penyerapan arsenik yang tinggi. Data daripada analisis XPS membuktikan mekanisme yang terlibat bagi penyerapan arsenik adalah reaksi pengoksidaan As (III) kepada As (V), kompleksiti permukaan, dan penukaran ligan dengan kumpulan hidroksil. Keseluruhannya, tanah UMS3 berpotensi untuk digunakan sebagai bahan reaktif untuk penyerapan arsenik kerana kadar penyerapannya yang tinggi, dan berpotensi mengurangkan penyebaran kadar pencemaran arsenik.

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AC	-	Activated carbon
AOP	-	Advanced oxidation processes
APHA	-	American Public Health Association
BET	-	Brunauer-Elmer-Teller
BTC	-	Breakthrough curve
BOD	-	Biochemical oxygen demand
BP	-	Bukit Palong landfill
CEC	-	Cation exchange capacity
COD	-	Chemical oxygen demand
D	-	Dispersion coefficient
DO	-	Dissolved oxygen
EC	-	Electrical conductivity
EPA	-	Environmental Protection Agency
EQA	-	Environmental Quality Act
HCl	-	Hydrochloric acid
HNO_3	-	Nitric acid
ICP-OES	-	Inductively Coupled Plasma – Optical Emission Spectrometry
Igeo	-	Geo-accumulation Index
IPI	-	Integrated Pollution Index
JPSPN	-	Jabatan Pengurusan Sisa Pepejal Negara
KK	-	Kampung Keru landfill
KS	-	Kampung Keru soil
LPI	-	Leachate Pollution Index
LTM	-	Ladang Tanah Merah landfill
LTMS	-	Ladang Tanah Merah soil
M	-	Mass
ORC	-	Oxygen releasing compounds
PTFE	-	Polytetrafluoroethylene
PRB	-	Permeable reactive barrier
PI	-	Pollution Index

PJ	-	Pajam landfill
QRS	-	Quality Rating Scales
R	-	Retardation factor
SEM	-	Scanning Electron Microscope – Electron Dispersive X-Ray Spectroscopy
SG	-	Sungai Udang landfill
SGS	-	Sungai Udang soil
SSE	-	Sum of square error
TDS	-	Total dissolved solids
USEPA	-	United State Environment Protection Act
UTM	-	Universiti Teknologi Malaysia
UiTM	-	Universiti Teknologi MARA
UMO	-	Ulu Maasop (Active) landfill
UMC	-	Ulu Maasop (Closed) landfill
UMS	-	Ulu Maasop soil
UPW	-	Ultrapure water
V	-	Volume
XRD	-	X-ray Diffractometer
XPS	-	X-ray Photoelectron Spectroscopy
ZVI	-	Zero-valent iron

LIST OF SYMBOLS

B_n	-	Geochemical background value
C_{eq}	-	Equilibrium concentration
C_n	-	Mean concentration
C_o	-	Initial concentration
F	-	Flowrate
L	-	Length
ppm	-	Part per million
%	-	percentage
q	-	Adsorption capacity
rpm	-	Revolutions per minutes (rotational speed)
ν	-	Average pore velocity
V_{sol}	-	Volume of solution

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CHAPTER 1

INTRODUCTION

1.1 Background of study

The rising of global generation of waste (municipal and industrial wastes, ranging from biodegradable to synthetic) is a consequences of rapid growth in population, increasing urbanization and resource consumptions, industrialization, as well as development of economies, and Malaysia is not excluded (Aja et al., 2014; Mor et al., 2018; Vaccari et al., 2018; Zainol et al., 2012). With the current growth of population and urbanization trend, the generation of waste in the world is expected to increase from 2.01 billion tonnes (in 2016) to 3.40 billion tonnes (in 2050) (Kaza et al., 2018). Unfortunately, around 40 percent of the waste generated will end up in dumpsites which are mostly are located in Africa, Latin America and Asia and other low and middle income countries, disposed in unregulated manner or openly burned (ISWA, 2019). In Malaysia, the generation of waste per kapita is about 1.1 kg per day, resulting to over 26500 tonnes of waste being disposed to only 162 operational landfills (sanitary and non-sanitary) throughout the countries (JPSPN, 2015; Kamaruddin et al., 2017).

World's dependencies to land disposal and dumpsites has directly impacted the health of human, ecosystems as well as the environment surrounding these sites. In most developing and under-developed countries, landfilling has become the ultimate methods and primary facilities for municipal solid waste disposal (Xaypanya et al., 2018). Such method is considered as most cost-effective waste disposal option as compared to incineration and composting, though it consequently require more land spaces and critical environment approaches (Agamuthu and Izyani, 2017). In Malaysia, presently there are 176 operational and 114 non-operational landfills in each state and the Federal Territories (JPSPN, 2015). A great number of uncontrolled landfills without appropriate bottom liners and leachate collection systems have been wide spread throughout the country. Most of the landfills were not under the sanitary landfill classification because there are no facilities to collect and treat the leachate as well as there is no infrastructure to capture the landfill gas. There are only a few sanitary landfills with leachate treatment and gas exploitation facilities (Alkassasbeh et al., 2009; Fauziah and Agamuthu, 2012).

Non-sanitary landfill in Malaysia refers to waste disposal sites which is constructed or operated without a proper engineering plan and landfill bottom liner system (Fauziah and Agamuthu, 2012). These refers to controlled dumps which are unlined with proper liner system to prevent leachate percolation into groundwater and soil which would poses a serious pollution threat due to the migration of the pollutants in leachate. Open tipping sites were also falls into same category which are considered as non-sanitary landfills where liner system, gas control, leachate collection and treatment were all not provided. Nevertheless, some of these non-sanitary landfills were upgraded and facilitate with leachate collection system and treatment during the closure procedures. Some of the open tipping sites were also upgraded and provided with road accessibility, designated tipping areas, weekly waste spreading, compaction and soil cover, however there no gas control, leachate collection and also treatment available (Rahim et al., 2010).

Landfilling is definitely not a ‘problem free’ solution for disposal of solid waste. One of the problems associated with it is the leachate generation and its uncontrolled infiltration into the environment, which may result with serious contamination of ground and surface water. It is stated that around 38 out of 50 of the world’s largest uncontrolled dump located in coastal areas, where the waste and leachate can easily enters water courses and sea (ISWA, 2019), thus, being another stringent issues globally. This leachate can easily seep through the ground or mix with the runoff and pollute adjacent ground and surface water sources (Abhayawardana, 2015). This issues is evident around landfills without proper lining materials or when the lining is punctured or leaky (Agamuthu and Fauziah, 2010). In Malaysia itself, most of the closed landfills and other non-sanitary landfills were not installed with landfill bottom liner which is crucial to confine and collect leachate from polluting the environment (Fauziah and Agamuthu, 2012).

Many researchers agree that leachate from active and closed municipal solid waste landfills, which is formed as a result of multiple chemical and biological reactions of solid waste within the landfill, can be significant source of contamination and toxicity for ground water and surface water if not controlled properly (Abu-Daabes et al., 2013; Ashraf et al., 2013; Naveen et al., 2017). The production of leachate is due to contact of moisture with waste content, and may aggravate especially in tropical countries with high amount of rainfall like Malaysia (Suleman, 2016). The concentration and level of contaminants in the leachate may be influenced by waste compositions and characteristics, age of the deposited waste and also age of the landfills, site operation methods, and also quantity of water entering the landfill (Kjeldsen et al., 2002; Xaypanya et al., 2018).

Leaching from paints, mercury-cointaining wastes, batteries, pesticides and fertilizers and other hazardous substances may imposed to the presence of heavy metals in landfill leachate and constitute a serious threat to the environment and human health not only because it is the most toxic contaminants (Abu-Rukah and Abu-Aljarayesh, 2002) but also due to the facts that they do not degrade. Leaching of metals may not only occur due to anthropogenic causes such as leaching from landfill wastes, but also naturally through the reducing conditions due to microbial degradation process in the landfill leachate which caused the release of metals such as iron and manganese, and concomitant release of other metals which are bound onto iron, such as arsenic (Di Palma and Mecozzi, 2010; Nguyen et al., 2015; Wang et al., 2012). The presence of heavy metals in the environments will also perturb the natural biological balance and also inhibits the self-purification processes in the natural systems (Gworek et al., 2016; Mor et al., 2006; Talalaj, 2015). Moreover, according to Adeloppo et al.(2018), heavy metals can remain within the landfills for around 150 years if it leached at a rate of 400 mm/year. Mobility and transport of contaminant constituents from landfill leachate especially heavy metal pollutants in surface and groundwater has also been an emerging issues as it poses serious threat to the environment (uptake in soil and plants) and also human health (due to the facts that it pollutes drinking water sources). A study by Emenike et al. (2012) reported that heavy metals can even mobilize kilometers away from landfill site.

Another aspect to consider is the state of leachate parameters as the landfill stabilises during the decomposition phase. Several parameters change dramatically as the landfill stabilises. According to Kjeldsen et al. (2002), there is a strong relationship between the state of refuse decomposition with leachate characteristics. It is indicated that during landfill stabilisation phase, level of heavy metals in leachate will decrease. Nevertheless, several inorganic parameters such as Cd, Cr, Co, Cu, Pb, Ni and Zn will not be affected by landfill stabilisation, thus the concentrations will not reduce significantly (Christensen et al., 2001). Thus, leachate characterization serves as a guideline for the implementation of an appropriate leachate treatment procedure.

Scarcity of studies regarding the heavy metal contamination in landfill leachate and soil specifically has resulting in less attention focussed on the removal of these pollutants. With indiscriminate distribution of uncontrolled landfills and dumpsites in Malaysia, it is important to realize that landfill leachate which were untreated and loaded with heavy metals may endangered environment and ecosystems impacted. Pathogens, toxins, and carcinogenic metals, such as arsenic, chromium, and nickels are classified as pervasive environmental pollutants. Uncontrolled widespread of these pollutants are not only detrimental to the surrounding environment and ecosystems, but also increase risks of numerous cancers and other health hazards such as kidney and liver damage, mental retardation in children, and gastrointestinal disorders (Alluri et al., 2007; Chervona et al., 2012; Nilanjana et al., 2008).

Thus, the leachate generated from solid wastes should be treated in order to reduce the concentration of pollutants to the acceptable limits before being discharged into the environment. In Malaysia, the traditional source of drinking water had been surface water. It is therefore very important that municipal landfill is properly sited, designed, managed and maintained so that the sources of water are protected from leachate pollution (Agamuthu et al., 2015). Nevertheless, unlike sanitary landfill, leachate in dumpsites or unlined landfill cannot be collected. Traditional pump and treat methods are not possible, thus, permeable reactive barrier (PRB) system is an alternative and good option.

Permeable reactive barrier (PRB) (Figure 1.1) is one of the innovative, yet sustainable methods of environmental protection which effectively prevent migration of contaminants in leachate from landfills. As compared to any other traditional treatment methods of landfill leachate which adopt the concept of pump and treat, PRB system is more applicable especially in the situation where leachate cannot be collected due to the absence of landfill liner and leachate collection system as it provides *in-situ* plume capture and treatment method (Chung et al., 2007; Fronczyk and Garbulewski, 2009; Kumarasinghe et al., 2018; Sewwandi, 2014; Wang et al., 2016; Zhou et al., 2014). The principle of PRB treatment system is that contaminated water will flow under hydraulic gradient through a reactive media that will efficiently degrade the contaminant into less harmLess forms or easily degradable compound (Snow and Jones, 1999) (Figure 1.1). Not only effective in reducing the level of multiple contaminants in contaminated water and soil, PRB system is also very cost effective (for operation and maintenance), requires very less maintenance, low energy input, and cleans up only the area of contamination. Other than that, this system also promotes the use of low cost and readily available materials as reactive media, such as zero valent irons, industrials or agricultural wastes or mulch, in the form of single media or combined media, based on the contaminants to be treated.

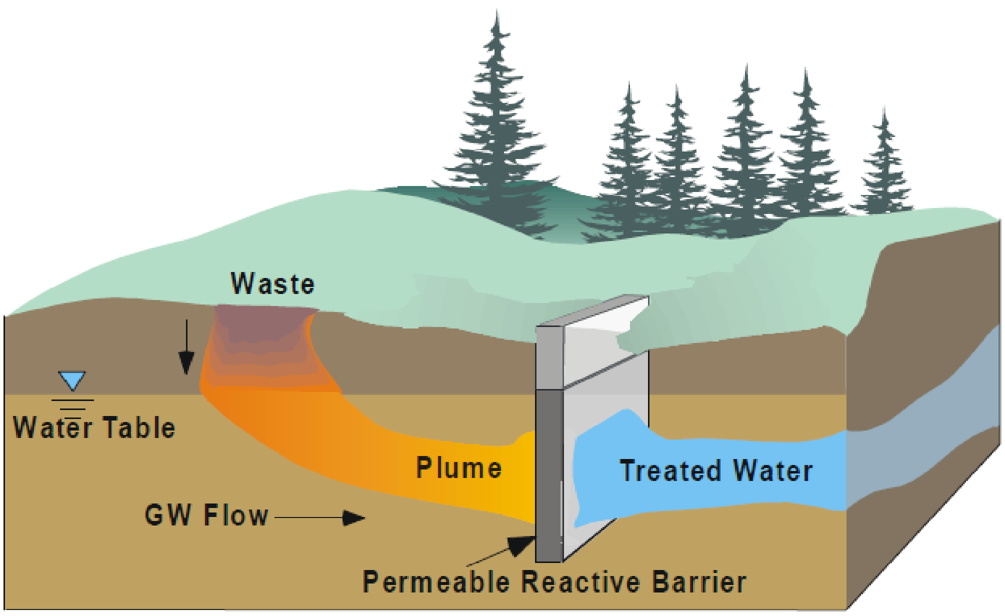

Figure 1.1 Schematic diagram of a Permeable Reactive Barrier (PRB) system (Powell et al., 1998)

1.2 Problem statement

One of the major challenges when exploiting a landfill is the pollution deriving from leachate generation. The waste and leachate problems are also worsened due to the facts that most of these land disposal areas were lacking with proper landfill facilities. Uncontrolled landfills and unregulated dumpsite were mostly not equipped with leachate collection and treatment system and without appropriate bottom liner, thus, increase the possibilities of soil pollution, and severe surface and groundwater contamination (Ashraf et al., 2013; Aziz et al., 2010; Kanmani and Gandhimathi, 2013; Xaypanya et al., 2018). In Malaysia itself, less than 5 percent of the landfills fall within the category of sanitary landfills (carefully designed, constructed, operated and monitored regularly), while the rest are controlled and uncontrolled dumpsites, and some with over-loaded capacities (Noor et al., 2013). Remedial processes such as leachate collection, leachate treatment, and monitoring of landfills are complex, thus, is usually costly (Tyrrel et al., 2002; Youcai et al., 2000). Hence, a very innovative, with a very less maintenance, simple and cost-effective, such as permeable reactive barrier (PRB) system, are required to avoid spreading of contaminated leachate to the environment.

Landfill leachate generally consists of organic matter (biodegradable and non-biodegradable), inorganic pollutants (heavy metals) and other hazardous subtances (Aziz et al., 2010; Slack et al., 2005). Distribution of numerous heavy metal pollutants especially due to the unorganized dumping of solid waste, pose a significant pressure on the environment as several pollutants are very toxic and dangerous to every form of life. Heavy metals are pollutants that are classified as toxic to the environment, even at minute concentrations (Jayanthi et al., 2016).

Most of the implemented treatment process of landfill leachate in Malaysia were focussed on organic compounds and ammonia removal due to the high concentration of the constituents. Therefore, less attention were given to the removal of heavy metals from landfill leachate (Aziz et al., 2004). Surprisingly, studies from Agamuthu and Fauziah (2010), Atta et al. (2015), Jayanthi et al. (2016), Mohd Raihan Taha et al. (2011), Nor Amani Filzah and Suhaimi (2010), Rahim et al. (2010), Sakawi et al. (2013), Samuding et al. (2012), Siti Nur Syahirah et al. (2013), Suratman et al. (2011), Syafalni et al. (2014) and Yusoff et al. (2013) have revealed the impacts of heavy metal to the groundwater and soil in vicinity of operating and even in closed landfill sites. Nahrim Malaysia also reported that mercury and arsenic contamination from landfills into groundwater were 9% and 27% respectively (Tawnie et al., 2016) and these numbers will continuously grow if immediate remedial and protection are not taken.

Thus, this study is aimed to elucidate the degree of heavy metal pollution in the landfills especially when it was not properly installed with effective protection system, and thus can raise awareness to the local authorities to take immediate action. Based on the identified significant heavy metal pollutant in landfills leachate, this study will also aim to select the most suitable soil for the removal of significant heavy metal pollutant. This locally available natural soil will be used as reactive media in the PRB system proposed to be installed as remediation technique and pollution control in landfill (especially non-sanitary landfill) in Malaysia.

1.3 Aim and objectives

The goal of this research is to determine the treatability of heavy metals in landfill leachate by verifying the remediation techniques for the most significant heavy metals pollutant in landfill leachate in Malaysia. The specific objectives are:

1. To investigate the degree of heavy metals pollution in leachate and leachate/wastes impacted soil and to identify the most significant heavy metal pollutant in different types of landfills in Malaysia.
2. To determine the most effective soil as adsorbent (reactive medium) in PRB system for heavy metal removal.
3. To evaluate soil performance as reactive media in PRB system for heavy metal removal in soil column system (laboratory scale).

1.4 Scopes and limitation of the study

This study focused on investigating the degree of heavy metals pollution in leachate and impacted soil and determine the treatability of the pollutant using natural soil. The main factors that were taken into consideration to conduct this study include the selection of sampling sites, the research design, and the instruments to be used.

Samples of leachate and soils were taken from different types of landfills (sanitary landfills, safely closed landfills and open dumping sites), with permission from Jabatan Pengurusan Sisa Pepejal Negara (JPSPN) and the helps from the Southern Waste Corporation (SWCorp) and Landfill Managers. Selections of landfills were based on the list of landfills under the management of SWCorp. Sungai Udang and Ladang Tanah Merah landfills were chosen for the category of sanitary landfills; Bukit Palong, Pajam and Ulu Maasop for closed landfills; while Kampung Keru and Ulu Maasop (active site) were chosen for operating dumpsites.

Samples points from both sanitary landfills (Sungai Udang (6 sampling points), and Ladang Tanah Merah (3 sampling points) as well as two of the closed landfills, Bukit Palong (3 sampling points) and Pajam landfills (3 sampling points) were collected from raw leachate collection ponds, treatment processes as well as treated leachate discharge points. Whereas leachate samples from Ulu Maasop (closed landfill – 2 sampling points) were collected from leachate collection well installed in the landfill. As for dumpsites (Kampung Keru and Ulu Maasop (active site)), leachate samples were collected from leachate stream/runoff within the landfills. Soil from sanitary and closed landfills were collected at the landfills waste retaining soil-bund, while soil samples from dumpsites were collected from few locations which are affected by wastes and leachate. For comparison, natural soil samples were also taken from unaffected areas (by leachate and wastes) but still within the landfills. These soil samples were referred as background samples to epitomize the quality of the native soils. The natural soil samples were also collected to be use as potential adsorbent of heavy metal from landfill leachate.

After samplings, characteristics of leachate and leachate/waste impacted soil (in particular of heavy metals pollutants) were identified. Since the samples were taken from different types of landfills in Malaysia in different locations, the characteristics of leachate and leachate impacted soil obtained were anticipated to closely resemble the characteristics (in particular for heavy metals) in this country. Nevertheless, it should be noted that the characteristics of leachate would vary, depends on the age, sizes and capacity of the landfills, and also influenced by the physical, chemical, and biological activity of the wastes, soils and water occurring within the landfill. Soil used as adsorption media in the proposed permeable reactive barrier system were also considered as locally available soil as it were obtained within the landfills area.

The second stage in this study is divided into two (2) main phases which are identification of the most effective soil for heavy metal removal and evaluation of the performance of the soil by using column test study. For the first phase, soil characterizations, heavy metal adsorption capacity of soil using batch test, and adsorption mechanisms analysis were conducted. In phase 2, performance of the selected soil for heavy metal adsorption as well as retardardation factor and dispersion coefficient of heavy metal in soil media were evaluated using column test study.

All of the experiments were conducted in Shizen iKohza, Malaysia Japan International Institute of Technology, UTM and BIOREC Laboratory, Civil Engineering Faculty, UiTM. Methods of experiments that were used are by using portable multi parameter (HORIBA) to measure pH, temperature, electrical conductivity (EC), total dissolved solids (TDS), turbidity, and dissolved oxygen (DO) on landfill leachate by onsite measurements; ICP-OES for heavy metals analysis in leachate, soil and heavy metal solutions; microwave digestor for digestion of soil for heavy metal analysis; X-Ray Diffractometer (XRD), Electron dispersive X-Ray spectroscopy (SEM-EDX), specific surface area by Brunauer-Elmer-Teller (BET) analysis, cation exchange capacity (CEC) and pH, X-Ray Photoelectron spectroscopy (XPS) analysis for characterizations of soil and mechanism of adsorption; batch equilibrium test for heavy metal adsorption capacity of soils, and for the selection of the most suitable soils for heavy metal removal; and column studies to evaluate selected soil performance and to determine retardation factor and dispersion coefficients of heavy metal in soil to be use in permeable reactive barrier system.

Several limitations have been identified that narrow the range of scope in this study. The research works were narrowed by the types of landfills selected, sampling periods, types of heavy metal (arsenic) to be removed, and soil selection for arsenic adsorption methods. The types of landfill selected were sanitary landfills, safely closed landfills and active dumpsites. For each type of landfill, two to three landfills were selected. The sampling periods is limited based on the landfills selected. As for sanitary and closed (Sg. Udang, Ladang Tanah Merah, Bukit Palong and Pajam) landfills, sampling were conducted only once due to limited access granted from the landfill operators. Nevertheless, three cycles of samplings were conducted in Ulu Maasop (closed and actives sites) and Kampung Keru landfills. Both of these landfills were non-sanitary landfills and has been a major focus in this study since both of it are non-sanitary landfills.

In this study, among all metals, arsenic was identified as the most significant heavy metal pollutant in landfills. There is no single way which can remove total arsenic (arsenic (III) and arsenic (V)) at the same time. The changes of arsenic into different oxidization forms with the changing of pH and the redox potential increase the challenge and complexity of arsenic removal in aqueous solution (Mohanty, 2017; Weerasundara et al., 2021). Thus, arsenic (III) was selected for removal due to the level of toxicity, high solubility and more mobile compared to arsenic (V), and more reasons as specified in Chapter 2 (Literature Review).

Natural soil used for arsenic adsorption were collected from different landfills, characterized and screened by using batch equilibrium test. Since this study used natural soil as adsorbent, there will be several limitations on the reaction mechanisms as compared with the usage of synthetic/engineered adsorbents.

1.5 Significance of study

Leachates are a potential hazardous waste from landfill sites. Heavy metals were present at relatively high concentrations in the landfill in Malaysia. The exposure of heavy metals into the environment is great concern due to their serious effects on food chain, natural surface water and groundwater systems and furthermore on animal and human health. Therefore, this study initiated to:

1. Identify the most ‘critical heavy metals pollutants’ generated from landfills leachate in Malaysia. This study also illustrated the efficacy of treatment available in sanitary landfills and some of the closed landfills, in focussing for heavy metal parameters. Besides, this study provides the extent of metals pollution to the soil in vicinity of the landfills, especially at the unlined landfills. The results of this study provides helpful information for regulating and managing future waste disposal approaches at preventing environmental impacts of heavy metals pollution. Studies on the heavy metal characteristics, leachate pollution potentials and impacts to the soil proved that the problems in the non-sanitary landfills need to be prioritized and solved.

2. Natural soil with the highest adsorption capacity for heavy metal will be propose to be use in the PRB system to treat leachate generated from landfills in Malaysia. The use of soil as reactive material promise not only low cost materials and environmental benignity, but the soil is also locally available in Malaysia. Besides, the concept of adsorption used in the removal of pollutant also considered to be one of the cheapest and simple treatment method to date.

3. PRB system proposed are one innovative ‘onsite treatment’ technology, which is simple, inexpensive solution and involved very little maintenance and operation. It is a passive treatment systems, and able to treat a wide range of contaminants, especially in open dumpsites to reduce the risk of polluting soil and groundwater by leachate flow. The determination of retardation factors and dispersion coefficient from the column studies may contribute to the design and estimation of longevity of PRB system.

1.6 Thesis outline

This thesis is divided into five chapters. The first chapter is the introduction which comprise of the background of the study, problems statement, objectives, scope and limitations as well as significances of the study. The problems which led to the research being conducted, the aims as well as the beneficial contributions of this research were clearly stated in this chapter.

Chapter 2 covers the Literature Review part, which is essential for better understanding of the research conducted. In this chapter, overview of waste generation, land disposals, types and issues regarding landfilling methods were briefly discussed. Characterizations and the impacts of landfill leachate on the surface soils and groundwater were thoroughly discussed in this chapter. As for the characterizations of landfill leachate, more focus were given to heavy metal parameters. Furthermore, treatment methods for heavy metal removal from landfill leachate were reviewed, especially those applicable to be installed onsite / at the dumpsites. The use of locally available materials and low cost methods for heavy metal removal were also properly discussed.

Chapter 3 deals with the research and experimental methods for this study. It covers the selections of landfills in Malaysia, the sampling of landfill leachate, leachate impacted soils, and natural soil (collected in vicinity of landfills) for potential used as reactive media in PRB system for heavy metal removal. Experimental procedures for leachate and soil characterizations, as well as analysis of impacts of leachate were discussed. Furthermore, procedures for screening and selection of soil were illustrated and explained.

Chapter 4 deals with the chacterizations of leachate in landfills, leachate contamination potentials, and impacts of landfill leachate on soil in vicinity of the landfills in focussing for heavy metals parameters. This chapter also covers the screening of locally available soil from landfills for the potential used as reactive media in proposed PRB system for dumpsites in Malaysia. After identifying the most significant heavy metal pollutant in landfills and screening of the soil as adsorbent, the most efficient soil for heavy metal removal was selected and discussed. Besides, possible reaction mechanism of heavy metal adsorption in soil were also briefly discussed in this chapter. This chapter also illustrated the breakthrough behaviour of heavy metal in packed bed columns as to investigate the performance of soil for heavy metal removal in a larger scale.

Chapter 5 summarises the findings from the overall studies and conclusions from the analysis of the data collected. It also gives recommendations for the improvement of landfills in terms of monitoring and operations and some suggestions for future research works.

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