

ASSESSMENT OF LEACHING BEHAVIOR
OF LATERITE SOIL AMENDED WITH
MAGNESIUM RICH SYNTHETIC GYPSUM
(MRSG)

AMIRA AINNA BINTI ALIAS

B. ENG(HONS.) CIVIL ENGINEERING

UNIVERSITI MALAYSIA PAHANG

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To mom and dad
Who always picked me up on time
And encouraged me to go on every adventure
Especially this one

If anyone travels on a road in search of knowledge,
Allah will cause him to travel on one of the roads of paradise...
(Abu Dawood)

ASSESSMENT OF LEACHING BEHAVIOR OF LATERITE SOIL AMENDED
WITH MAGNESIUM RICH SYNTHETIC GYPSUM (MRSO)

AMIRA AINNA BINTI ALIAS

Thesis submitted in fulfillment of the requirements
for the award of the
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ABSTRAK

Risiko kebolehcapaian dan risiko alam sekitar unsur nadir bumi (REE) dan logam berat dalam sedimen akuifer daripada sisa buangan industri baru-baru ini, Sisa-sisa Pelupusan Bahan-Bahan Lynas (LAMP), Gebeng, Pahang, Penyucian Bersih Air (WLP) dan Pengaliran Bersih Residu (NUF) yang dikenali sebagai Magnesium Rich Synthetic Gypsum (MRSG) disiasat sebagai penilaian terhadap kelakuan larut. Tujuan utama kajian ini adalah untuk mengenalpasti sifat pelarut tanah laterit dan MRSG yang dipinda tanah di bawah ujian lajur dan ujian penyatuan larutan dan membandingkan kepekatan unsur dengan Prosedur Penyulingan Ciri Ketoksikan (TCLP) dan Standard Kualiti Air Minuman Negara (NWQS). MRSG sangat mendapat permintaan dalam industri yang digunakan untuk tujuan aplikasi tanah. Penggunaan bahan-bahan ini berlebihan di tanah menyebabkan pemobilisasi logam dari matriks tanah dan penyusupan selanjutnya ke air bawah tanah. Pelepasan cecair yang tercemar yang mengandungi kepekatan logam berat yang tinggi yang dilepaskan dari petak tanah yang mendasari akan menyebabkan kerosakan alam sekitar yang serius. Ujian larut resap biasanya digunakan untuk menilai potensi pelupusan bahan toksik dan memahami kelakuan larut. Kajian-kajian ini dilakukan dengan menganalisis kepekatan unsur oleh Spektrometri Massa Plasma (ICP-MS). Keputusan eksperimen menunjukkan bahawa kepekatan magnesium menjadi lebih tinggi apabila laterit bercampur dengan WLP dan NUF. Diikuti oleh 524.9 mg / l untuk condisoil 1: 2: 7, 911.9 mg / l untuk condisoil 1: 3: 6 di bawah 6.2 kPa manakala di bawah 1589.8 kPa kepekatan meningkat sebanyak 508.3 mg / l kepada 1011 mg / l dalam kondisoil. Keputusan yang direkodkan kepekatan unsur dalam ujian Penyatuan Larutan lebih tinggi berbanding ujian larutan Lajur. Dari keseluruhan kajian, semua unsur kepekatan pencemaran berada di bawah garis panduan had TCLP. Walau bagaimanapun, kepekatan unsur magnesium, aluminium, kromium dan nikel didapati melebihi had yang ditetapkan oleh NDWQS.

ABSTRACT

The leachability and potential environmental risks of rare earth element (REE) and heavy metal in aquifer sediments from the recent industrial by-product waste of Lynas Advanced Materials Plan (LAMP), Gebeng, Pahang, Water Leached Purification (WLP) residue and Neutralization Underflow (NUF) residue as known as Magnesium Rich Synthetic Gypsum (MRSG) were investigated as assessment of leaching behavior. The main purpose of this study is to determine the leaching characteristic of laterite soil and MRSG amended soil under column test and oedometer test and comparing elements concentration with Toxicity Characteristic Leaching Procedure (TCLP) and National Drinking Water Quality Standard (NDWQS). The demand for MRSG is highly increased for land application purposes. Excessive usage of this materials on land may cause metal mobilization from the waste and subsequently infiltrated and polluted groundwater. Leaching of contaminated liquid containing high concentrations of heavy metals release from underlying groundwater will causing serious environmental damage. Leaching test are commonly used to assess the leaching potential of toxic substances and understand leaching behavior. Tests were carried out to analyse the element concentration using Inductively Coupled Plasma Mass Spectrometry (ICP-MS). Experimental result demonstrated that magnesium concentration increased when laterite was mixed with WLP and NUF, followed by 524.9 mg/l for 1:2:7, 911.9 mg/l for 1:3:6 mixture under 6.2 kPa while under 1589.8 kPa concentration increased by 508.3 mg/l to 1011 mg/l, respectively. The result recorded element concentration in oedometer leaching test were greater compared to columns leaching test. Overall, all element of concentration of contaminants were below the guideline of TCLP limit. However, element concentration of magnesium, aluminium, chromium and nickel were found exceed the limits set by NDWQS.

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LIST OF SYMBOLS

PG	Phosphogypsum
Mg	Magnesium
Sc	Scandium
Y	Yttrium
La	Lanthanum
Lu	Lutetium
Th	Thorium
Ce	Cerium
Pr	Praseodymium
Nd	Neodymium
Pm	Promethium
Sm	Samarium
Eu	Europium
Gd	Gadolinium
Tb	Terbium
Dy	Dysprosium
Ho	Holmium
Er	Erbium
Tm	Thulium
Yb	Ytterbium
Al	Aluminium
K	Potassium
Ca	Calcium
Si	Silicon
P	Phosphorus
Cr	Chromium
Ni	Nickel
Cu	Copper
Ba	Barium
Pb	Lead
Ce	Cerium

Nd	Neodymium
kPa	kilo Pascal/pressure
mg/l	milli gram per liter
TiO ₂	Titanium dioxide
CaSO ₄ .2H ₂ O	Calcium Sulphate dihydrate
CaCO ₃	Calcium carbonate
SiO ₂	Silicon dioxide
Al ₂ O ₃	Aluminium oxide
Fe ₂ O ₃	Iron oxide

LIST OF ABBREVIATIONS

MRS	Magnesium Rich Synthetic Gypsum
LAMP	Lynas Advanced Material Plant
WLP	Water Leach Purification
NUF	Neutralization Underflow Residue
SW205	Schedule Waste
DoE	Department of Environment
AELB	Atomic Energy Licensing Board
TCLP	Toxicity Characteristic Leaching Procedure
NDWQS	National Drinking Water Quality Standard
ICP-MS	Inductively Couple Plasma Spectrometry
IAEA	International Atomic Energy Agency
FAO	Food and Agriculture Organization
REE	Rare Earth Element
LREEs	Light rare earth elements
MREEs	Medium rare earth elements
HREEs	Heavy rare earth elements
TENORM	Technologically Enhanced Naturally Occurring Radioactive Material
USEPA	United States Environmental Protection Agency
ISO	International Organization for Standardization
ASTM	American Society for Testing and Materials
CEC	Cation exchange capacity

CHAPTER 1

INTRODUCTION

1.1 Background of Study

Lynas Advanced Material Plant (LAMP) is the world's largest rare earth processing plant. LAMP is located in Gebeng Industrial Center, Kuantan Pahang. The extraction process of rare earth materials has resulted in two major residues: Water Leach Purification (WLP) and Neutralization Underflow Residue (NUF), each at 64000 tonnes and 135000 tonnes per year (Lynas, 2018). This material, also called Mg Rich Synthetic Gypsum (MRSG). In addition, the WLP and NUF are also rich in other elements such as gypsum and magnesium that can be used in agriculture for the improvement of soil quality. In order to leverage this material as soil conditioner, a study has been conducted to explore the potential use of this ancillary waste in agriculture. A new substance named Condisoil was produced through a mixture of WLP, NUF and organic filler. Condisoil is suitable for soil trimming and soil improvement in the agricultural industry because it is rich in nutrients needed in plant growth. The Condisoil composition consists of 10% WLP, 20% NUF and 70% other laterite matter. When these components are mixed, Condisoil should contain a natural radionuclide concentration to the background level or lower than the 1000 Bq kg⁻¹ limit value allowed by the Atomic Energy Licensing Board (AELB). This allows Condisoil to be categorized as non-radioactive material (Aznan et al., 2018).

At that time, the product was called Phosphogypsum (PG) derived from lynas residues because the name Condisoil was not yet used and had yet been trademarked. The amount of PG recommended for amending agriculture soils varies between 500 and 1000kgha⁻¹ (Mays et al., 1986). Therefore, and based on the announced benefits of PG to increase crop yields more recently, and only based on 226Ra limits (Borges et al., 2013) or on total metal concentrations considered individually (Hwaiti et al., 2010), PG

from Brazil and Jordan, respectively, were recommended for fertilization purposes and as a soil conditioner. Excessive usage of agrochemical fertilizers and soil amendments were randomly applied on agricultural fields causing metal remobilization from the soil matrix and subsequent infiltration to groundwater (Atafar et al., 2010; Zang and Shan, 2008). Furthermore, the demand for more landfills and industrial waste disposal sites also increased (Leao et al., 2004; Younes et al., 2015). Subsequently, such studied can allow the assessment of the effect of the various elements on fauna and flora, and their possible leachability to groundwater (Zhao et al., 2009). The radioactivity and the occurrence of leachable metals represent a potential danger for its application, particularly in agriculture (Al- Hwaiti and Al- Khashman, 2015; Ammar et al., 2016; Elloumi et al., 2015).

Bauxite is the most important raw material for the production of aluminium and is a natural weathering product of alumina-rich rocks, consisting mainly of one or more aluminium hydroxide minerals and depending on the precursor rock, a mixture of quartz, iron oxides, TiO₂- polymorphs and aluminosilicates (Hill and Sehnke, 2006). Commonly, bauxite is divided into lateritic and karst bauxite, where lateritic bauxite is underlain by aluminosilicate rocks and comprise about 90% of the world's bauxite reserves (Freyssinet et al., 2005). Lateritic soils are one of important soils and widespread in tropical areas and subtropical climates. The significant features of the lateritic soils are their unique color, poor fertility, and high clay content and lower cation exchange capacity. In addition, lateritic soils possess a great amount of iron and aluminium oxides (Shaw, 2001). Iron oxides, existing mainly in the amorphous and crystalline inorganic forms, are one of major components in many soil orders.

Assessment of the leachability of toxic substances from contaminated soils is crucial for evaluating their environmental impact on soil and groundwater. Leaching test are commonly used to assess the leaching potential of toxic substances and understand leaching behavior. Several standard column leaching tests (ISO/CD 21268 – 3, 2017; DIN 19528, 2009; US EPA Method 1314, 2013) enable contact between contaminated soil and flowing leachant and are used to indicate the leaching behavior of target substance under simulated field conditions. Soil column leaching studies, performed under controlled conditions, can help in understanding the behavior of certain elements or compounds in a defined area (Alvarez, 2007; Arfania and Asadzadeh, 2015; Maszkowska et al., 2013).

In this study, the leaching behaviour includes the analyses of chemical contaminants of core samples and samples collected from waste process within a process of lanthanide concentrate that rich with Rare Earth Element (REE). The leaching rate as well as the overall REE and elemental recovery values were used as response variable to assess the potential for land application. The contributes to environmental policy design by Toxicity Characteristic Leaching Procedure (TCLP) and National Drinking Water Quality Standard (NDWQS), the examining issues pertinent to laterite and waste residue and its environmental impact toward soil and groundwater.

1.2 Problem Statement

MRSG is waste by-product that is highly increased demand in industrial used for land application purposes. The components of MRSG including rare earth elements, heavy metal and radionuclides were included in chemicals of concern list as toxicity. Several other elements were present as impurities. This metal needs to be monitored when MRSG is on land applied. Excessive usage of this materials on land may cause metal remobilization from the soil matrix and subsequent infiltration to groundwater. The radioactivity and the occurrence of leachable metals represent a potential danger for its application, particularly in agriculture. Leaching of contaminated liquid containing high concentration of heavy metals release from underlying soil compartments will causing serious environmental damage. Leaching test are commonly used to assess the leaching potential of toxic substances and understand leaching behavior. Assessment of the leachability of toxic substances from contaminated soils is crucial for evaluating their environmental impact on soil and groundwater. Thus, the leaching characteristic of laterite soil and its mixture with MRSG were determined by oedometer leaching test and columns test. Then the elements of contaminants content were analyzed by Inductively Couple Plasma Spectrometry (ICP-MS).

1.3 Research Objective

The objective of this research were as follows:

- (i) to determine the leaching characteristic of laterite soil and its mixed with Magnesium Rich Synthetic Gypsum (MRSG) using soil residue from Lynas Advanced Material Plant (LAMP), Neutralization Underflow residue (NUF) and Water Leached Purification residue (WLP)

- (ii) to compare the leaching concentration of laterite and MRSG amended soil under column test and oedometer test
- (iii) to compare the chemical properties of leaching test with Toxicity Characteristic Leaching procedure (TCLP) Limit and National Drinking Water Quality Standard (NDWQS).

1.4 Scope of Study

The study focuses on the use of two type of residue from Lynas Advanced Material Plant (LAMP) at Gebeng, Pahang. Residual soil which is Neutralization Underflow residue (NUF) and Water Leached Purification residue (WLP) in the form of condisoil. Laterite soil was mixed to study the leaching characteristic of the soil. Several laboratory tests were conducted to obtain the geotechnical soil properties and soil parameters required to determine the leaching characteristic of soil. Two major laboratory tests need to consider to obtain the leaching water of each sample by conducted column test while oedometer test should applied some different loading for each sample. The leaching water sample, then will be tested in Central Laboratory of University Malaysia Pahang using Inductively Couple Plasma Spectrometry (ICP-MS) testing to obtain the soil parameter of each samples. From the result of soil parameters by ICP-MS testing, then compered the leaching characteristic of MRSG amended soil under column testing and oedometer testing. The chemical composition of leaching of all samples also need to be compared with Toxicity Characteristic Leaching Procedure (TCLP) limit and National Drinking Water Quality Standard (NDWQS) limit to consider the leaching contamination and environment hazard.

1.5 Significance of Study

The better understanding in leaching behavior of laterite soil and its mixture with MRSG. Evaluating the leaching behavior of waste product from Lynas was able to determine the potential impact toward it used on land application. This study is hope to offer the suitable analysis to counter the environment hazard and the consequent impact upon the community. Besides, it is hope to benefit not only to the Government but also elevated the livelihood of people associated with the industry. Nevertheless, it has also raised concern of wanton environmental damages and consequent health effects.

Therefore, this study was hope to preserve the environment from the occurrence of any contaminants to ensure the health and well- being of the community.

1.6 Thesis Outline

The thesis is divided into five specific chapter to completing of this research study:

Chapter 2 describes comprehensively on leaching behavior and the effects toward environment and its effect to groundwater. In this chapter also will describes the characteristic, effect, and application of Water Leached Purification residue (WLP), Neutralization Underflow residue (NUF) and laterite soil.

Chapter 3 present the research methodology that needs to be carried out as to achieve the objective set out in physical properties, the water content, geotechnical test for obtaining soil leaching water and parameter of soil leaching by laboratory testing under Inductively Couple Plasma Mass Spectrometry (ICP-MS) method. In this chapter, it has a detailed description of methodology of each test.

Chapter 4 present the results for each test on physical and chemical properties, and parameter of soil leaching. Comparing the leaching parameter under column test and oedometer test and also comparing with Toxicity Characteristic Leaching Procedure (TCLP) and National Drinking Water Quality Standard (NDWQS).

Chapter 5 describe the conclusion of the effect of leaching behavior of soil considering the environmental contamination and environment hazards.

CHAPTER 2

LITERATURE REVIEW

2.1 Introduction

This chapter were discussed the previous research related to Mg Rich Synthetic Gypsum (MRSg). In particular, the effect of used this material to land application has been focused. The leachability of soil was also review in this chapter.

2.2 Rare Earth Element

Rare earth elements (REEs) or RE metals are technically defined as the 15 elements in the lanthanide (La) series, yttrium (Y), and scandium (Sc). A precise classification often used in extraction is as follows (Koltun and Tharumarajah, 2014):

- (i) Light rare earth elements (LREEs): lanthanum (La), cerium (Ce), praseodymium (Pr), neodymium (Nd), and promethium (Pm).
- (ii) Medium rare earth elements (MREEs): samarium (Sm), europium (Eu), and gadolinium (Gd).
- (iii) Heavy rare earth elements (HREEs): terbium (Tb), dysprosium (Dy), holmium (Ho), erbium (Er), thulium (Tm), ytterbium (Yb), lutetium (Lu), scandium (Sc), and yttrium(Y).

The MREEs and HREEs, which are of vital importance for the hi-tech industry because of their unique properties, are high-priced and in low-supply (Chen, 2011). Despite the term “rare earth”, REE are relatively plentiful in the earth crust. However, REE scarcely occur in the concentrated and economically exploitable mineral deposits. Their abundance in the earth crust is even higher than that of other commonly used elements such as platinum group elements (Farzaneh et al., 2017). Half of the world’s 110 million tons of reserves exist in China while other sizable deposits are found in

Russia, United States, Brazil, India, Australia, Canada, and Greenland. The countries known to be actively mining REE are China, Russia, India, Brazil and Malaysia (Hao et al., 2015). Among these countries, China has the largest REE production, accounting for more than 93% of global supply (Yap, 2015).

2.2.1 Characteristic of REE

REEs are commonly found in mineral compounds such as carbonates, oxides, phosphates and silicates (Yang et al., 2018). Most of the RE bearing minerals, especially carbonates and phosphates, are rich in light REEs (LREEs), i.e., cerium, lanthanum, neodymium, promethium, samarium and scandium (Jha et al., 2016). Although few REE-containing minerals could be economically mined and processed (Kanazawa et al., 2006), REE have been identified in over 250 mineral classes, such as halides, carbonates, oxides, phosphates, silicates. Bastnaesite, monazite and xenotime are the main REE-bearing hard rock minerals which have been successfully used as the commercial minerals for REE extraction. The REEs in nature can also be transported in fluids due to naturally acidic, high temperature conditions, especially the heavy REEs (HREEs) (Bao et al., 2008).

The ion-adsorption REE mining regions of southern China, REEs have been found to accumulate in water, soil, and flora in mining areas (Hao et al., 2015; Meryem et al., 2016) with REE concentrations reported ranging from 396 to 2314 $\mu\text{g/g}$ and the lowest detected soil concentrations in the region being double the national average (Liang et al., 2014). The ion-adsorption type rare earth ore, first discovered in 1969, is mainly located in the seven provinces of Southern China (Xu, 2002). This kind of ore has many advantages, simple leaching process and rich in middle and heavy rare earth elements, which make it a valuable strategic mineral resource (Xiao et al., 2015). It is generally accepted that rare earth in the ion-adsorption type rare earth ore presents in four phases (Chi et al., 2005): water soluble phase, ion-exchangeable phase, colloidal sediment phase and mineral phase. The ion-exchangeable phase rare earth accounts for more than 80% of the whole-phase rare earth in general. It can be easily exchanged and released when encountering the cations such as NH_4^+ , Mg^{2+} . According to the ion-exchange property, a series of hydrometallurgy methods for this special ore have been developed along the green chemical leaching approach (Huang et al., 2015).

The REEs have a high ionic charge due to their 3+ valence and small radius. Due to a decrease in the ionic radii light to heavy elements, the HREEs tend to have the least mobility as a result of adsorption onto an oppositely charged mineral surface such as the negatively charged basal plane of clays. REE ions are adsorbed onto negatively charged clay surfaces or interlayers as either cation exchange clay-REE physisorption, or hydrolyzed clay-O-REE²⁺ species chemisorption (Moldoveanu et al., 2012). The cation exchange reactions are pH and temperature independent, whereas the chemisorption processes are endothermic and occur at high pH (Bradbury et al., 2002). The ion exchangeable REEs associated with clay minerals can be exchanged using salt cationic solution ammonium sulfate, sodium chloride, magnesium chloride at pH values between 4 and 8 (Tian et al., 2010; Papangelakis et al., 2014; Yang et al., 2013).

2.2.2 Application of REE

REEs are often labelled as the “material of the future” (Hao and Liu, 2011) because they serve as important ingredients in everyday products (Massari and Ruberti, 2013; Schüller et al., 2011). The global demand for REE and their compounds has noticeably increased in many high-tech applications owing to their chemical, catalytic, electrical, magnetic, and optical properties. For examples, REE are used in hybrid cars, wind turbines, compact fluorescent lights, rechargeable batteries, catalytic convertors, flat screen televisions, mobile phones, and disc driver (Borai et al., 2002; Savel’eva, 2011; Abreu and Morais, 2010; Parhi et al., 2015; Jorjani et al., 2011). According to Husna and Kuntala (2017) REE is used in electronic equipment, medical devices, communication devices, green technologies and agricultural application such as fertilizers are dependent on the use of REEs. There are about 200 known rare-earth containing mineral deposits, mostly as carbonatites spread around the world. Contrary to a lay persons’ understanding, REs is not rare in natural occurrence (cerium is more abundant than tin and yttrium is more abundant than lead), though REEs have much less tendency to become concentrated in exploitable ore deposits (U.S. Geological Survey, 2002), in particular HREEs. This being so, only a few mineral species, such as bastnaesite, monazite and RE bearing clay, have been recovered for commercial production.

Bastnaesite deposits in China and the United States constitute the largest percentage of the world's rare -earth economic resources (U.S. Geological Survey, 2008). China has a monopoly¹ over global production of REEs. However, in 2009, China implemented export quota restrictions on REEs (Jepson, 2012). According to Zhang et al. (2015), the export quota was issued in 2005 (51,000 t) and decreased gradually to 46,000 t, coupled with 10 per cent export tariffs on rare earth minerals. In the following year, the quota decreased to 45,370 t, declined further to 34,156 t in 2008. In 2009, China issued an export quota of 31,310 t, and reduced it to 24,280 t in 2010.

2.3 Lynas's Residues

Lynas Corporation, an Australian based mining company are constructing a rare earth processing plant, known as Lynas as shown in Figure 2.1. Advanced Materials Plant (LAMP) in Gebeng industrial estate in Kuantan, Malaysia. The LAMP will process lanthanide concentrate which will be trucked from the mine site in Mt Weld Western Australia to the Port of Fremantle where it will be shipped to Malaysia. The lanthanide concentrate is rich with Rare Earth Element (REE). The REE are a group of 17 elements, including scandium (Sc), yttrium (Y) and 15 lanthanides from lanthanum (La, 57) to lutetium (Lu, 71). They show the similar chemical properties, and tend to occur in the same mineral deposits (Tyler G., 2004). This LAMP plant extracts the rare earth elements produces a by-product called Magnesium Rich Synthetic Gypsum (MRSG) (Sahibin et al., 2018). The residues are commonly produced, namely WLP and NUF.



Figure 2.1 Lyans Advanced Materials Plant (LAMP) in Gebeng, Kuantan, Malaysia

Source: (National Toxics Network, 2012)

2.3.1 Water Leached Purification residue (WLP)

The WLP residues contain relatively high levels of the nutrients phosphorus and magnesium which have potential agricultural markets. Phosphorus is widely recognized as a rate limiting nutrient in soils throughout the world, including Australia, Europe and America. However, as it competes with fertilizers carrying from 8% to 21% by weight of phosphorus, it was conceded that proximate markets would be essential to product marketability (Lynas, 2008).

The WLP residues resulting from the leaching and purification of the water-soluble lanthanide components from the calcined, cracked concentrate (Lynas, 2008). It is primary iron phosphate, with minor iron hydroxide, aluminium hydroxide and gypsum. The WLP residue however contains thorium (Th) at activity concentration of about 6 Bq/g and classified as radioactive material by the Atomic Energy Licensing Board (AELB) but defined by International Atomic Energy Agency (IAEA) as a very low-level radioactive waste.

2.3.2 Neutralization Underflow residue (NUF)

The NUF residues carry high levels of magnesium, aluminium and calcium sulphate. In particular, magnesium sulphate present as the mineral Kieserite is beneficially used throughout the acid soils of the humid tropics, the dominant soil type in Malaysia, at rates of up to 100 kg/hectare as a palm oil nutrient (Kali & Stauff Corporation, 2016). This is because magnesium and potassium are considered more critical to nutrition than the traditional fertilizer components nitrogen and phosphorus Preferred Fertilizer Materials for Acid Soils of the Upland Humid Tropics, Food and Agriculture Organization (FAO) of the United Nations, 1987). Although the content of aluminium is considered less desirable than the magnesium content desired by FAO, the calcium sulphate is also considered to complement the materials value since it acts as a source of both calcium and sulphur for these soils.

The NUF residues consisting principally of the reaction product of an acidic sulphuric acid derived liquor with calcium, magnesium and aluminium based minerals to produce calcium, magnesium and aluminium sulphates. It is a solvent extraction and product finishing raffinate neutralized with lime (Lynas, 2008). NUF is also known as Magnesium rich synthetic gypsum as it is primary gypsum and regulates as SW205 by the Department of Environment (DoE), Malaysia.

2.3.3 Chemical compositions of WLP & NUF

Table 2.1 shows the content of NUF and WLP residues. NUF highly containing about 73-74 % of gypsum. Other minor percent of element are magnesium hydroxide, calcium hydroxide, calcium carbonate and silica. Meanwhile, WLP containing 25-25% of iron phosphate, 25 % of calcium carbonate, 10% of phosphogypsum and also containing in minor percent of radionuclide which is uranium oxide and thorium hydroxide.

Table 2.1 Chemical composition of WLP and NUF

NUF		WLP	
Content	% wet wt	Content	% wet wt
Gypsum $\text{Ca}_2\text{SO}_4 \cdot 2\text{H}_2\text{O}$	73 -74	Iron phosphate $\text{FePo}_4 \cdot \text{Nh}_2\text{o}$	25-50
Magnesium hydroxide $\text{Mg}(\text{OH})_2$	17.1	Calcium carbonate CaCO_3	25
Calcium hydroxide $\text{Ca}(\text{OH})_2$	4.2	Phosphogypsum $(\text{Ca},\text{P})\text{SO}_4 (\text{H}_2\text{O})\text{X}$	10
Calcium carbonate CaCO_3	2.3	Uranium oxide	<0.01
Silica (amorphous) Si	0.4	Thorium hydroxide	<0.2

Source: (Lynas, 2018)

2.3.4 MRSF in land application

Magnesium Rich Synthetic Gypsum can be applied in oil palm plantations as a source of Magnesium fertilizer (Sahibin et al., 2018). In the last few decades, kieserite from Germany and ground magnesium limestone (GML) is the main sources of magnesium fertilizer for oil palm plantation in Malaysia. Since, the high price of Germany kieserite and low reactivity of GML make it feasible for the plantation industry to seek for an alternative sources of magnesium fertilizer. Besides, the Atomic Energy Licensing Board (AELB, 2008) of Malaysia had confirmed that MRSF was non-radioactive. It means this material has the potential to be utilized in agriculture.

The WLP and NUF are also rich in other nutrient content such as gypsum and magnesium that can be used in agriculture for the improvement of soil quality to increase crop production (Aznan et al., 2018). Ca, Mg and S are needed by oil palm under production in high amount for its healthy growth and production, and it is believed that these macronutrients can be added into soil in the plantations by continuous application of MRSF. Besides, by virtue of being alkaline, the pH of MRSF is 8.8, due to the presence of magnesium hydroxide, calcium hydroxide and some calcium carbonate, soil pH can be raised to the desired level for oil palm cultivation. Furthermore, MRSF contained some Si that may help prevent certain oil palm disease prevailing in the plantations throughout the country (Najihah et al., 2014). A new substance named Condisoil was produced through a mixture of WLP, NUF and organic filler and these Condisoil is suitable for soil trimming and soil improvement in the agricultural industry because it is rich in nutrients needed in plant growth (Aznan et al., 2018).

2.4 Phosphogypsum

Phosphogypsum (PG) is a by-product produced during the production of phosphoric acid by the wet process. It is estimated that approximately 5 t of PG is produced per 1 t of phosphoric acid. The annual production of PG is in the range of 200-280 Mt worldwide (Lopez et al., 2011). PG is highly acidic (pH 1) when initially stored, due to residual sulfuric acid, and consists mainly of calcium sulphate dehydrated ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$ or gypsum or hemi-hydrated ($\text{CaSO}_4 \cdot 1/2\text{H}_2\text{O}$ or bassanite), but also contains a high level of impurities including fluorides, sulphates, natural radionuclides, metals, and other trace elements (Rutherford et al., 1994; Arocena et al., 1995; Al-Masri et al., 2004). PG is a by-product of the fertilizer industry, specially the production of phosphoric acid from phosphate rock (Nurhayat et al., 2007) and the main producers of phosphate rock and phosphate fertilizer are located in the USA, the former USSR, China, Africa and the Middle East. The phosphate industry is also an important contributor to national economies in many developing countries. The mineralogical composition of phosphate ore, as described by various researchers (Carbonell et al., 2002; Oliveira and Imbernon, 1998), is dominated by fluorapatite [$\text{Ca}_{10}\text{F}_2(\text{PO}_4)_6 \cdot \text{CaCO}_3$], goethite and quartz, with minor amounts of Al-phosphates, anatase, magnetite, monazite and barite. Heavy metal and trace elements such as cadmium (Cd) and nickel (Ni) are also detected. Phosphate ores are naturally highly radioactive and their radioactivity originates mainly from uranium (U) and thorium (Th).

The geochemical characteristics of phosphogypsum are strongly influenced by the ore phosphate rock composition and by the chemical behavior of impurities released during manufacturing process (Lottermoser, 2010). Thus, although phosphogypsum is mainly composed by gypsum, this waste also contains some impurities such as phosphate, sulfate and fluoride, mainly in form of residual acids, toxic trace elements (e.g. As, Cd, Cr, Cu or Zn) and radionuclides from uranium decay series. Furthermore, wet processing causes the selective separation and concentration of naturally occurring radium (Ra), uranium (U) and thorium (Th): about 80% of Ra is concentrated in PG while nearly 86% of U and 70% of Th end up in the phosphoric acid. Determining the types of impurities present can be very important when defining waste management processes and environmental policies. Moreover, approximately 11 million tons of PG was accumulated for ore than 30 years of double superphosphate production at the Sredneuralsky Copper

Smelting Plant. It was found that the average content REE oxides in this PG is 0.43 – 0.52 %. Therefore, total amount of REE in the PG is estimated to be approximately 50 000 tons (Rychkov et al., 2018).

From all PG generated in the world, only the 15% is recycled, mainly as building materials (Tayibi et al., 2009). The remaining 85% is disposed of in large stacks, commonly in coastal areas, without any treatment and exposed to weathering processes. During operation, PG is commonly stockpile near the fertilizer plant over a composite liner to avoid infiltration. Upon closure, the stack is often capped with an impermeable layer to avoid leaching of contaminants. The USEPA has classifies PG as a Technologically Enhanced Naturally Occurring Radioactive Material (TENORM).

2.4.1 Characteristic of Phosphogypsum

PG properties are dependent upon the nature of the phosphate ore used, the type of wet process employed, the plant operation efficiency, the disposal method, and the age, location and depth of the landfill or stack where the PG is dumped (Arman and Seals, 1990). PG is a powdery material that has little or no plasticity and is composed mainly of calcium sulphate dihydrate (>90% gypsum) and sodium fluorosilicate (Na_2SiF_6) (Berish, 1990; Kamici et al., 2006). Due to the residual phosphoric, sulphuric and hydrofluoric acids contained within the porous PG, it is considered an acidic by-product ($\text{pH} < 3$). PG from filter cake usually has a free moisture content of 25-30%. The vertical hydraulic conductivity of PG has been reported to range between 1×10^{-3} and 2×10^{-5} cm/s (SENES,1987). The free water content may vary greatly, depending upon its pH, and it is highly soluble in saltwater (≈ 4.1 g/l) (Guo et al., 2001). Its particle density ranges between 0.9 and 1.7 g/cm³ (SENES, 1987) and its bulk density between 0.9 and 1.7 g/cm³ (Vick, 1977; Keren and Shainberg, 1981; May and Sweeney, 1984).

As PG is dewatered and weathered in storage piles, the acidity is progressively reduced. Phosphogypsum stacks may also emit radon gas and fluorine compounds (SiF_4 , HF) in significant amounts into the atmosphere. It has been reported that one of the main problems of PG piles is the emanation of ^{222}Rn from the alpha-decay of ^{226}Ra (Rutherford et al., 1994; USEPA 1992). In more windy areas the spread of fine PG particles cannot be disregarded as well.

With regard to radioactivity, as mentioned above PG contains relatively high levels of U-series radionuclides naturally present in the phosphate rock. Depending on the quality of the rock source, PG can contain as much as 60 times the levels normally found prior to processing. The most important source of PG radioactivity is reported to be Ra (Rutherford et al., 1994). Some author has also reported high U and Po activity. Ra produces radon gas (Rn), which has a short half-life of 3.8 days, an intense radiation capacity, and causes significant damage to internal organs (USEPA, 2002). For this reason, the USEPA has classified PG as a “Technologically Enhanced Naturally Occurring Radioactive Material” (TENORM) (USEPA, 2002) and PG exceeding 370 Bq/kg of radioactivity has been banned from all uses by the EPA since 1992.

2.4.2 Land Application of Phosphogypsum

An industrial plant of phosphate fertilizers originates about 4–6t of PG per ton of phosphoric acid produced (Olfa et al., 2015). Today the biggest issues facing the Tunisian phosphate industry is the pollution of the gulf of Gabes by PG. The Sfax chemical fertilizer company, located on the south coast near Sfax city, produces a considerable and increasing amount approximately 10 million tons of PG per year which is stored in piles in the vicinity of the industrial plant. These piles pose serious risks to the surrounding, partly urban area, by changing air quality, as previously mentioned by (USEPA, 1992; Kuryatnyk et al., 2008). In addition, soils and groundwater are affected by acidic and metal-rich infiltrations. Therefore, alternatives to PG stacking are urgently needed (Olfa et al., 2015). An industrial plant of phosphate fertilizers originates about 4–6t of PG per ton of phosphoric acid produced. Presently, the approaches used by the phosphate industry to deal with PG are: (i) discharging to water bodies; (ii) backfilling of mine pits; (iii) dry stacking and (iv) wet stacking. All the solutions have serious environmental impacts stated by International Atomic Energy Agency (IAEA, 2013).

The use of PG as an agriculture fertilizer has been practiced in many parts of the world for decades (USEPA, 1992; Mullins et al., 1990). It has been applied in agricultural soils as a calcium, phosphorus and sulfate supplement to enhance crop production and to recover acidic soils reducing Al-toxicity (Toma et al., 1997; Alva and Summer, 1990; Garrido et al., 2003) and sodic soils (Nayal et al., 2013; Enamorado et al., 2014). It was also used alone or in combination with other synthetic organic polymers for preventing runoffs and erosion in agricultural soils exposed to heavy rainstorms (Tang et al., 2006).

The amount of PG recommended for amending agriculture soils varies between 500 and 1000kg ha^{-1} (Mays et al., 1986). Therefore, and based on the announced benefits of PG to increase crop yields more recently, and only based on Ra limits (Boorges et al., 2013) or on total metal concentrations considered individually (Al-Hwaiti et al., 2010), PG from Brazil and Jordan, respectively, were recommended for fertilization purposes and as a soil.

2.5 Bauxite

Bauxites are residual deposits that formed from intense continental subaerial weathering and alteration, generally under humid tropical to subtropical climates (Bárdossy, 1982). Three main genetic types were proposed: 1) Lateritic bauxites, predominantly gibbsitic and derived by in situ lateritization of underlying aluminosilicate rocks; 2) Tikhvintype bauxites, overlying the eroded surface of aluminosilicate rocks, and being the erosional products of pre-existing lateritic bauxites; 3) Karst bauxites, containing boehmite and/or diasporite as the main Al minerals, developed on exhumed carbonate platforms (Bárdossy, 1982; Bárdossy and Combes, 1999; Ahmadnejad et al., 2017). Bauxite resources are widely distributed in the world and are the main resources for aluminum production (Ma et al., 2009, 2011). The bauxite ores which have high alumina content and high $Al_2O_3:SiO_2$ ratio are needed in the Bayer process for aluminum production (Li et al., 2016). Besides aluminum, bauxite is also mined for Fe, Ti, Ca, Si and Na and kaolin (Jusop, 2016). Bauxites are characterized by high Al and Si and low $Al_2O_3:SiO_2$ ratio (5–8) for most (80%) of bauxites in China (Zhao et al., 2010). Almost all of bauxite ores in China are diasporic-characterized and do not meet the demands in the Bayer process for aluminum production due to their high silica content and low $Al_2O_3:SiO_2$ ratio (Ma et al., 2009). Silicon is not only the main impurity in bauxite but also one of the most harmful impurities in the process of aluminum production using the Bayer process (Chen et al., 2016a).

The main composition in bauxite are mineral gibbsite [$Al(OH)_3$], goethite ($FeOOH$), hematite (Fe_2O_3) and kaolin (Tessens and Shamshuddin, 1983; Shamshuddin and Fauziah, 2010). Bauxite is reddish brown in color which is due to the presence of mineral hematite. Specifically, for bauxite, occurrence of this mineral in Peninsular Malaysia has been mostly found in Pengerang, Johor (southern Malaysia) and Kuantan, Pahang (new hot spot for bauxite mines in east-coast Malaysia) (Kusin et al., 2017).

These two areas are found to be suitable for bauxite formation due to high soil temperature and annual rainfall throughout the year (Jusop, 2016). Bauxite is usually strip-mined because it is found one or two meters below the surface of the soil layer (Rajah, 1986; Paramanathan, 1977). This is typically found in open pit mining, i.e. a type of strip mining, where the material is excavated from an open pit. During every step of the process, there is always a hazard to the environment. Crushing of rock will generate large quantity of dust. During the separation process, rock slurries will produce free metals and if not properly handled, the liquids can leak into bedrocks (Saxena and Singh, 2000). Bauxite mines are developed preferentially on flat-topped plateaus and occur on large continental-scale plan surfaces exposed to a tropical monsoon climate, whereby optimal hydraulic conditions are controlled by the balance between precipitation and evaporation (Herrington et al., 2016; Thorpe and Watve, 2016).

Of all known bauxite deposits, approximately 88% belong to the laterite-type, 11.5% are of the karst-type, and the remaining 0.5% are of the Tikhvin-type (Meyer, 2004). The most widespread ore deposits are lateritic bauxites, representing the 6th reserve in the world. Lateritic bauxites of economic interest have formed on a range of different sedimentary, igneous and metamorphic parent rocks worldwide. A sufficiently high aluminium content, together with favorable physical rock properties and weathering conditions determines the potential of a bauxite deposit as raw material for the aluminium industry (Valeton, 1972; Schellmann, 1983; Bárdossy and Aleva, 1990; Patterson et al., 1986; Aleva, 1994; Tardy, 1997). Fundamentally, laterite is formed from the residual concentration of the less soluble constituents (mainly Fe and Al) contained in the parent rock. Bauxite will be used for a laterite enriched in free aluminium hydroxide minerals such as gibbsite and boehmite (cf., Bárdossy and Aleva, 1990). Bauxite deposits in South America are all of lateritic affinity but developed on a variety of parent rocks (precursors) ranging from Phanerozoic siliciclastic sediments to Precambrian igneous and metamorphic lithologies (Bárdossy and Aleva, 1990; Carvalho et al., 1997; Bogatyrev et al., 2009).

2.6 Laterite Soil

Laterites are soil types rich in iron and aluminum that are formed in tropical areas. Most laterites are rusty-red because of the presence of iron oxides. Iron oxides, existing

mainly in the amorphous and crystalline inorganic forms, are one of major components in many soil orders (Tzu, 2014). They develop by intensive and long- lasting weathering of the underlying parent rock. Tropical weathering is a prolonged process of chemical weathering which produces a wide variety in the thickness, grade, chemistry and ore mineralogy of the resulting soils. The initial products of weathering are essentially kaolinized rocks called saprolites (Emeka and Olufikayo, 2004). These are soils rich in hematite kaolinite-goethite and formed by progressive enrichment of iron and silica, from the parent rock to the top of the profile (Giorgis et al., 2014). Parent material is a key factor affecting the iron and mineral composition and distribution for lateritic soils. (Anda et al., 2008) reported a series of oxisols derived from serpentinite, basalt, and andesite and found that the content of iron oxides has an obvious different distribution. Approximately 19% of iron oxide was determined for the lateritic soils derived from serpentinite. Different parent materials also bring the different physical and chemical properties.

Lateritic soils are products of tropical weathering with red, reddish- brown or dark brown color, with or without nodules or concretions and generally but not exclusively found below hardened ferruginous crusts (Ola, 1978). Laterite formation factors include climate precipitation, leaching, capillary rise and temperature, topography drainage, vegetation, parent rock iron rich rocks and time of these primary factors. However, climate is considered to be the most important factor.

As a soil stabilization, aims at improving soil strength, controlling dust and increasing resistance to softening by water through bonding of the soil particles together thereby water proofing the particles or a combination of the two (Amu and Adetuberu, 2010; Sherwood P., 1993). The simplest stabilization processes are compaction and drainage (if water drains out of wet soil, it becomes stronger). The other process is by improving the gradation of particle size and further improvement can be achieved by adding binders to weak soils (Rogers and Glendinnings., 1993). Soil stabilization can be accomplished by several methods, all these methods fall into two broad categories namely mechanical and chemical stabilization. Mechanical Stabilization is a physical process that involves altering the physical nature of native soil particles by either induced vibrations or compaction or by incorporating other physical properties such as barriers and nailing. Chemical Stabilization involves initiating chemical reactions between stabilizers

(cementitious material) and soil minerals (pozzolanic materials) to achieve the desired effect of improving the chief properties of a soil that are of interest to engineers namely volume stability, strength, compressibility, permeability and durability (Emeka and Olufikayo, 2016; Ingles., Metacalf., 1972).

Lateritic soils are generally used for road construction in Nigeria. Lateritic soils in its natural state generally have low bearing capacity and low strength due to high clay content. The strength and stability of lateritic soil containing large amounts of clay cannot be guaranteed under load in the presence of moisture (Alhassan, 2008). The use of lateritic soils consisting of high plastic clay content results in cracks in and damage to pavement, roadways, foundations or any civil engineering construction. The need to improve the strength and durability of lateritic soil in recent times has become imperative, this has geared researchers towards using stabilizing materials that can be sourced locally at a very low cost (Bello et al., 2015). These local materials can be classified as either agricultural or industrial wastes (Amu et al., 2011). In cases where sourcing for durable soil may prove economically unwise, the viable option is to stabilize the available soil to meet the specified requirements of construction (Mustapha, 2005; Osinubi, 1999).

2.7 Leaching

Leaching is the process by which soluble constituents are dissolved from a solid material such as rock, soil, or waste into a fluid by percolation or diffusion. Thus, when fill materials come into contact with liquid including percolating rainwater, surface water, groundwater, and liquids present in the fill material, constituents in the solid phase will dissolve into the liquid forming a leachate. The extent to which the constituents dissolve into the contact liquid will depend upon site- and material-specific conditions chemical, physical, and biological factors and the length of time involved. The composition of the leachate generated from the material and its potential to impact water quality are key factors in evaluating the suitability of the material for use as fill. Mobility, transport and leaching behavior of metals and other contaminants in the environment have been of great interest to many researchers (Du et al., 2009; Eggleton and Thomas, 2004; Kalnejais et al., 2010; Lesven et al., 2010; Syrovetsnik et al., 2007; Thevenon et al., 2013). There are diverse factors and processes impacting metal behavior in soil and sediment columns, which in turn rely on metal characteristics and their encompassing matrices.

The mobility of metals depends on the speciation, sorption behavior, specific electronegativity, ionic radius and reaction time, where mobility can further be impacted by the physicochemical properties of soils and sediments, such as pH, redox potential, cation exchange capacity (CEC), mineral composition, grain size, surface charge, soil structure and texture, permeability, organic content, and vegetation cover (Alvarez, 2007; Balkis et al., 2009; Gonzalez et al., 2006; Herndon and Brantley, 2011; Hiller et al., 2010; Maszkowska et al., 2013). Industry by-product phosphogypsum (PG) has high potential to release hazardous metals that consequently cause toxicity to soil microflora (Hentati et al., 2015; Jalali et al., 2016). The radioactivity and the occurrence of leachable metals represent a potential danger for its application, particularly in agriculture (Al-Hwaiti and Al-Khashman, 2015; Ammar et al., 2016; Elloumi et al., 2015).

Excessive usage of agrochemical fertilizers and soil amendments were randomly applied on agricultural fields causing metal remobilization from the soil matrix and subsequent infiltration to groundwater (Atafar et al., 2010; Zhang and Shan, 2008). Widespread use of pesticides in agricultural areas promote the deterioration of groundwater quality through leaching from soil (Hildebrandt et al., 2008; Wadhwa et al., 2010; Jurado et al., 2012; Labiteetal, 2013). Pesticides, when released into the environment can follow many pathways. They can volatilize and pass into the air. They can be degraded by sunlight, microorganisms via biochemical reactions, or chemical reactions with water. They can also be adsorbed y soil particles and move with eroding soil or move with surface runoff.

Moreover, they can stay in liquid phase in dissolved form and move through the soil by either in filtration or through in corporation, possibly getting into groundwater. Many factors affect the possibility of a pesticide reaching groundwater, such as pesticide properties i.e. solubility, adsorption, degradation and volatility (Damalas and Eleftherohorinos, 2011; Brown and Ingianni, 2012; Trautmann et al., 2012; Grodner et al., 2014); soil properties, i.e. organic matter content, soil texture and soil permeability (Damalas and Eleftherohorinos, 2011; Brown and Ingianni, 2012; Grodner et al., 2014); site conditions, i.e. depth to groundwater, rainfall, climate and geologic conditions (Brown and Ingianni, 2012; Grodneretal, 2014); and lastly management practices, i.e. pesticide application methods, rate and timing (Arias-Estévezetal, 2008; Grodneretal, 2014).

From Qin et al. (2010) and Li et al. (2015) runoff can be considered a major way of phosphorus (P) losses compared with P leaching, but recent investigation indicated that P leaching from soils is a matter of concern. Several researchers (Eghball et al., 1996; McDowell and Sharpley, 2011; Maguire and Sims, 2002; Dao et al., 2005; Abdala et al., 2015) indicated that accumulation of P in the surface soil may intensify risk of P losses and increase movement of P below the plough layer. Many factors such as soil P sorption saturation and soil texture can affect leaching of P from soils. Significant leaching of P have been reported from P-saturated soils (Breeuwsma and Silva, 1992), sandy soils (Sims et al., 2004) and where soils have received large amounts of manure or compost (Jalali and Ostovarzadeh, 2009). In Hamedan, western Iran, about 65% of the cultivates soils are light texture soils (Jalali and Jalali, 2016) have received P fertilization for more than 40 years. Intensive fertilization leads to accumulation of P in soils, increasing the risk of pollution of the aquatic environment by dissolved P losses in surface runoff (Sharpley and Withers, 1994).

2.7.1 Leaching Method

Leaching test are commonly used to assess the leaching potential of toxic substances and understand leaching behavior. Several standard leaching tests have been developed. Liu et al. (2008); Delay et al. (2007); Grathwohl et al. (2009) stated that the possible leaching potential of contaminated materials on the soil-groundwater pathway can be determined by laboratory leaching tests such as column and batch tests. Standard batch leaching tests (Ministry of the Environment (Japan), 1991; ISO/TS 21268-1 2007 and ISO/TS 21268-2, 2007; US EPA Method 1313, 2012; DIN 19529, 2015) facilitate contaminant leaching using agitation and are used as compliance tests due to their simplicity. Standard column leaching tests (ISO/CD 21268-3, 2017; DIN 19528, 2009; US EPA Method 1314, 2013) enable contact between contaminated soil and flowing leachant and are used to indicate the leaching behavior of target substance under simulated field conditions.

The column test (CEN/TS 14405:2004) is a percolation leaching test for soils and granular waste materials on a laboratory scale. It is a standard test used for the risk assessment of pollutants release from solid materials into seepage water, groundwater or surface waters. The test results can be used to predict the release of constituents from a waste in a specific scenario, as a function of time, by means of modelling under the

hydraulic, geotechnical, hydrological, chemical, physical and biological conditions of the scenario. It simulates the leaching behavior of a material in the short and medium term by relating the release of inorganic parameters to the liquid to solid ratio (L/S) (Roeland et al., 2016).

The column leaching test is used in Flanders northern region of Belgium as a legal instrument to acknowledge compliancy of waste materials that are to be used as or in building materials with environmental legislation. In the test, water is forced to flow upstream a column filled with sample materials, allowing constituents to leach into the water. At 7 specific cumulative L/S ratios (0.1, 0.2, 0.5, 1, 2, 5, 10 l/kg), the leachate is collected – these are called fractions and sent for chemical analysis. The exact procedure used in Flanders is defined in CMA/II/A.9.1 (Flemish Environmental Legislation, 2016) that, as well as the CEN/TS 14405:2004, is based on the Dutch NEN 7343. The main difference with CEN/TS 14405:2004 is the limitation of particle size (<4 mm) and a fixed column diameter (5 cm) of the column in which the material is tested. In CEN/TS 14405:2004, a column diameter of 10 cm is allowed as well, with a particle size < 10 mm. Alternative standards for percolation column leaching tests exist worldwide, e.g. US EPA Method 1314, ASTM D4878, DIN 19528, which typically differ in the L/S at which fractions are collected, water flow rate and, in some of these methods, the upper limit of the grain size. The Flemish legal limit values for the cumulative release (at L/S = 10) are defined in VLAREMA (Roeland et al., 2016).

Batch test are used as compliance tests for contaminated soil and waste in many countries, e.g. Japan (Ministry of Environment, 1991), Germany (German Standardization Organization, Deutsches Institute fur Normung, 2015) and the United States (United States Environmental Protection Agency, 2013), and in international standards (International Standardization Organization, 2007) because such tests are simpler than other leaching tests, such as up-flow column percolation tests, and they can be performed quickly (Tetsuo et al., 2017).

The basic procedure for a batch test performed on inorganic substances is quite simple. Firstly, a solid sample is placed in a suitable bottle and a leachant is added at a specified liquid-to-solid ratio. Secondly, the mixture is agitated for a specified period. Next, the solid materials are allowed to settle and the mixture is centrifuged. Then, the liquid is decanted and determined.

There is some important requirement for successful batch test methods. The first requirement is equilibrium; allowing equilibrium or semi-equilibrium conditions to be reached is the most important way of obtaining comparable batch leaching test results (International Standardization Organization, 2007). Every test requires the assumption that equilibrium or semi-equilibrium is reached at the specified contact time. However, the equilibrium status that is reached will depend on the soil type and the species being investigated. In the ISO TS21268-1 procedure, it is stated that contact for 24h is sufficient for equilibrium to be reached and that 6 h may be sufficient in the following specific cases when “it can be demonstrated that equilibrium or semi-equilibrium is reached or, for quality control purpose, a quick turn-around time is required” (International Standardization Organization, 2007).

The second requirement is the removal of suspended matter and colloidal particles. It is well known that organic contaminants, such as polycyclic aromatic hydrocarbons (PAHs) and PCBs, are strongly adsorbed into colloidal particles, such as organic matter and clay particles (Jacobsen et al., 1998; Prechtel et al., 2002; Massoudieh and Ginn, 2007). To avoid or reduce the effect of organic contaminants adsorbed into colloidal particles, the removal of the colloidal particles is important. In the procedure used in Germany and the ISO TS 21268-2 methods, the supernatant after agitation was centrifuged at 20,000-30,000g for 30 min to remove the colloidal particles without 0.45- μm MF filtration for organic substances. In the German standard, it is specified that the turbidity of the leachate must remain below 20 formazin nephelometric units (International Standardization Organization, 2007) German Standardization Organization, Deutsches Institut für Normung, 2015).

The above mentioned studied of leaching tests procedure, column leaching is the best testing for hazardous contaminated soil. This method suitable for the assessment of leaching of laterite soil amended with magnesium rich synthetic gypsum to finalize the analysis of leaching contaminants soil. Besides, another rare used method of leaching such as oedometer test are used to conduct the leaching behavior. The test needs some load applied into the soil to get the leaching characteristic. It simulates the leaching behavior as waste material is dump in stockpile.

2.7.2 Groundwater contaminants

Groundwater quality has been a focus of research because hazardous substances such as heavy metals presented in groundwater can enter the food chain and ultimately harm aquatic organisms and human beings (Ming Chen et al., 2016; Jarup, 2003; Nouri et al., 2008; Zeng et al., 2013). For hydrosphere, about 13-30% of the total volume of freshwater is groundwater (Dragoni and Sukhija, 2008), which is the source of drinking water for over 50% of the world's population. Thus, the increase of heavy metal contents in groundwater would pose potential threats to human health and survival (Hofmann et al., 2015; Zhang et al., 2015). Also, groundwater is being influenced by climate change, although it was previously suggested that groundwater quality was related to climate change (Alley, 2011). Dragoni and Sukhija (2008) pointed out that we should not overlook the effect of climate change on groundwater quality. Groundwater management and protection requires sufficient information on the response of groundwater to human activities and climate change.

Heavy metals may percolate into the underlying groundwater resources (Ghosh and Singh, 2005) thereby creating further challenges to water quality and sustainable water supply. Accordingly, Lin et al. (2012) noted that heavy metal pollution is a crucial global problem and therefore, merits continuous investigation in order to provide an up-to-date information required for successful risk mitigation. Heavy metals emanate from myriad of sources in the environment including industrial waste, spillage of petrochemicals, fertilizer application, atmospheric deposition and mine tailings (Wuana and Okieimen, 2011). Areas associated with significant oil and gas exploration and drilling activities are therefore, most likely to experience challenges with heavy metals due to the generation and release of produced water and solid wastes contaminated with heavy metals (Christie, 2012; Namdari et al., 2017; Doyi et al., 2018).

Another matter of concern is the leachability of hazardous elements from PG and thus the contamination of groundwater underlying PG stacks (May and Sweeney, 1982; May and Sweeney, 1983; Carter and Scheiner, 1992; Berish, 1990). Since PG waste is generally transported and disposed as an aqueous slurry, PG piles can be affected by tidal variations and dissolution/leaching of the elements naturally present in the PG can occur. Dissolved elements may be deposited in nearby soils or transferred to waters and finally to living beings (Reijnders, 2007). The international limit prescribed by the European

Atomic Commission (EURATOM) is 500 Bq/kg (Sunil, 2002; EURATOM Council Directive, 1996). ²²⁶Ra leachability from PG stockpiles has been evaluated by several researchers.

Haridasan et al. (2002) conducted leaching studies on PG samples from Kochi, India, using distilled water (pH = 6.0) and rainwater (pH = 5.0-5.8) as leachants in different contact time and solid/liquid ratio conditions. The laboratory results indicated that rainwater leached less ²²⁶Ra (0.09-0.28 Bq/l) than distilled water (0.08-0.38 Bq/l). When PG was exposed to natural weather conditions (rain) the maximum ²²⁶Ra activity in the leachate was 0.53 Bq/l while the minimum was 0.07 Bq/l. Most ²²⁶Ra values determined in the leachates exceeded the limit value of 0.1 Bq/l prescribed by the Bureau of Indian Drinking Water Standards.

Azouazi et al. (2001) investigated ²²⁶Ra solubility from Moroccan PG in aqueous solution. The initial ²²⁶Ra activity in phosphate rocks and PG was found to be around 1700 and 1420 Bq/kg, respectively. The leaching study involved mixing 20 g of PG with 50 ml of distilled water for 20 h at various pH values (2.10-8.84). The results showed an average ²²⁶Ra leaching rate of 26.4%, which could be cut to 6% when the leached PG was previously calcined at 800°C. When the calcined PG was leached with acidified aqueous solutions, HCl (4 N) or H₃PO₄ (22.1 N), no ²²⁶Ra was detected. Analysis of a water sample from an area close to the phosphate mine revealed the presence of 0.2 Bq/l of ²²⁶Ra, which is below the radium chemical safety limit for water (1 Bq/l).

CHAPTER 3

METHODOLOGY

3.1 Introduction

Leaching characteristic of soil investigated through laboratory test. From the various laboratory tests, the physical properties of soil laterite soil and magnesium rich synthetic gypsum amended soil were determined. In order to determine the parameter of soil leachate, column test and oedometer test are performed to get the soil leaching of the soil specimen. After getting the soil leaching, the water of soil leaching is tested in Centre Laboratory of University Malaysia Pahang. Inductively coupled plasma mass spectrometry (ICP-MS) laboratory testing was conducted to analyze the parameter of leaching characteristic. From the result, the parameter then compared with Toxicity characteristic leaching procedure (TCLP) limit standard, and National Water Quality Standard (NWQS) to determine the leaching characteristic of soil sample whether it safe for groundwater and land application. In this chapter, all the tests used were discussed to achieve the objective of this study.

3.2 Site Description

In this study, the residues were collected from Lynas Advanced Materials Plants. WLP and NUF are industrial by-product by LAMP also known as MRSG. LAMP located at Gebeng, Kuantan, Pahang. Laterite soil was obtained from bauxite mining area in Bukit Goh, Kuantan, Pahang. LAMP distance approximately 44 km from Gebeng to UMP, Gambang.

3.3 Selection of Material

The soil specimen for both MRSG and laterite soil were obtained at Kuantan, Pahang. Then, the soil specimens were brought to the laboratory in sealed bags.

3.4 Specimen Preparation

The soil specimens were crushed and kept in sealed bags. In this study, soil sample were divided into three categories which is laterite soil, Condisoil 1:2:7 and Condisoil 1:3:6. Condisoil 1:2:7 is mixture ratio of soil 100 g of WLP, 200 g of NUF and 700 g of laterite soil. Condi soil 1:3:6 is mixture ratio of soil 100 g of WLP, 300 g of NUF and 600g of laterite soil. The third condition is laterite soil with mass of 1000 g. Every soil samples were sieved passing sieve no 425 μ m using sieve shaker as shown in Figure 3.1. Then all specimens were kept in sealed bags before being tested.



Figure 3.1 Soil samples were sieved passing sieve no 425 μ m using sieve shaker

3.5 Properties of soil

Laboratory tests to determine physical properties of soil Atterberg limits was referred using different standard as shown in Table 3.1.

Table 3.1 Standard used for the physical properties test

Physical Properties	Method
Liquid Limit, LL	Cone penetrometer method (BS 1377: Part 2:1990: 4.3)
Plastic Limit, PL	(BS1337: Part 2: 1990: 53)
Shrinkage Limit, SL	Standard test method for shrinkage factors of soil by Wax Method (ASTMD 4943 - 08)

3.5.1 Liquid Limit Test

This liquid limit test was referred to the BS1337: Part 2: 1990:43. About 250 g oven dried laterite soil passing 2 mm were left air dried within about 20 minutes. The distilled water was added to form paste for sample and then transferred to the cylindrical cup of cone penetrometer. The soil paste was pressed against the side of the cup to avoid trapping air and also was pressed into the bottom of the cylindrical cup without creating an air pocket. The penetrometer was adjusted by ensuring the cone point touches or contact with the top of surface of the soil paste. Then the vertical clamp was released to penetrate into the soil paste after 5 seconds under its own weight. The stage of test was repeated for three times of values of penetration in expectation of 13.5 to 27.5 mm. Then, about 10 g from the area penetrated by the cone penetration of 20 mm was taken using small spatula for moisture content measurement. Same step and method were repeated for Condisoil 1:2:7 and Condisoil 1:3:6.

3.5.2 Plastic Limit Test

This test was conducted followed BS1337: Part 2: 1990:53. In this test, the soil sample was rolled, gathering together and kneading under the required pressure for rolling until the tread crumbles can no longer into a 3.2 mm diameter thread. Same step and method were repeated for Condisoil 1:2:7 and Condisoil 1:3:6.

3.5.3 Shrinkage Limit Test

This shrinkage limit test is based on ASTM D4943-08 standards test method. At first, the soil sample was added with distilled water until it reaches 1.2 times the liquid limit of soil sample. The empty container was weighed first and grease is applied into the internal of the container and weighed again. Then, the soil sample is transferred into the

cylindrical cup of cone penetrometer apparatus, and need to ensure that no air is trapped in the soil sample and the weight was recorded. The soil samples were left dried up until there was no changes occurred in terms of soil weight. Then, the weight of metal cup including soil samples after oven dried were recorded. The dry soil sample was tied with thread and then coated with wax. The weight of dry soil with thread and weight of dry soil with thread and wax shall be recorded. The shrinkage limit of the soil was taken by the corresponding of the moisture content to void ratio of the soil and can be calculated through Eq. (3.1) and the apparatus set up shown in Figure 3.2. The same step and method were repeated for Condisoil 1:2:7 and Condisoil 1:3:6.



Figure 3.2 Apparatus set up for shrinkage limit test

$$\text{Shrinkage Limit} = \frac{W - ((V - V_s) / W_s) \times 100\%}{1} \quad (3.1)$$

Where W = Moisture content of wet soil sample

V = Volume of wet soil sample

V_s = Volume of dry soil sample

W_s = Weight of oven dried soil sample

3.6 Column Leaching Test

This test is referred to ASTM D4874 standard test method for column leaching test. This column test was performed to collect the water leachate of laterite soil, Condisoil 1:2:7 and Condisoil 1:3:6. For the purpose of this study, a total of 3 specimen of soil were prepared in 1000 g for each soil. The apparatus of glass column is shown in Figure 3.3.



Figure 3.3 Apparatus of glass column leaching test

Approximately 1000g of soil sample used and placed in a 30.40 cm diameter and 26 cm height glass column. The soil column was installed onto a stand, and distilled water was continuously supplied from the top in keep a constant hydraulic head of 30 mm. The leachate was late collected by a container under the soil column and as volume was recorded. One sample of leachates were collected for each soil column and each sample were kept in pill box bottle as shown in Figure 3.4. Then, experiment was repeated for other 2 soil sample using the same step and method. The leaching experiment was conducted in the laboratory at 25.12 °c. Lastly, the leachate was analysed using Inductively Coupled Plasma Mass Spectrometry (ICP-MS) at Centre Laboratory of University Malaysia Pahang.

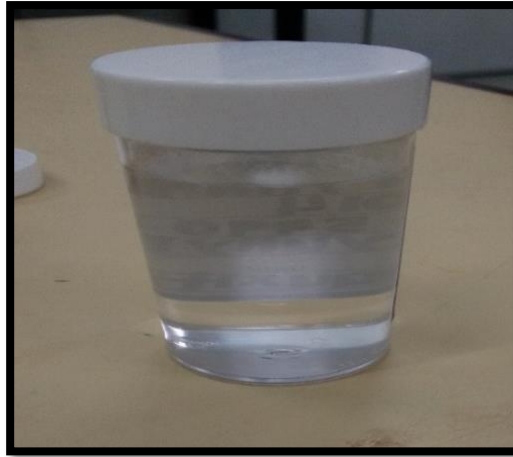


Figure 3.4 Sample of leachate collected from soil column

3.7 Oedometer Leaching Test

Similar to the oedometer test carried out for the determinators of consolidation and settlement behavior, the oedometer test was again employed to determine the leaching potential of laterite soil and its mixture with WLP and NUF. In this test, the sample were subjected to high vertical applied stress force squeezing so that the pore water is expelled out from the specimen. It is commonly expected that cations and anions move out of soil along with expelled water. This test also represents actual on-site conditions when the material is subjected to over-buried press soil by vertical loading from structure. The apparatus of oedometer is shown in Figure 3.5.



Figure 3.5 Apparatus of oedometer having 20 mm height and 50 mm diameter

Oedometer apparatus:

- i Ring, 50 mm diameter and 20 mm high of stainless steel, rigid with polished internal surface and cutting edge.
- ii Cell body and base (watertight)
- iii Ring retainer and fixing screws or nut
- iv Loading cap (pressure pad)
- v Two porous discs of sintered bronze and free draining with plane upper and lower surface.

For this study, a total 6 specimen of laterite soil, Condisoil 1:2:7 and Considosil 1:3:7 were prepared in 100 g of each sample. 3 specimens of different soil were used for vertical applied pressure 6.2 kPa and another for pressure 1598.9 kPa. Each of specimen need for moisture content determination and distilled water were used to add to form paste as shown in Figure 3.6. Then, the specimen sealed in plastic bags and kept in one day at room temperature.

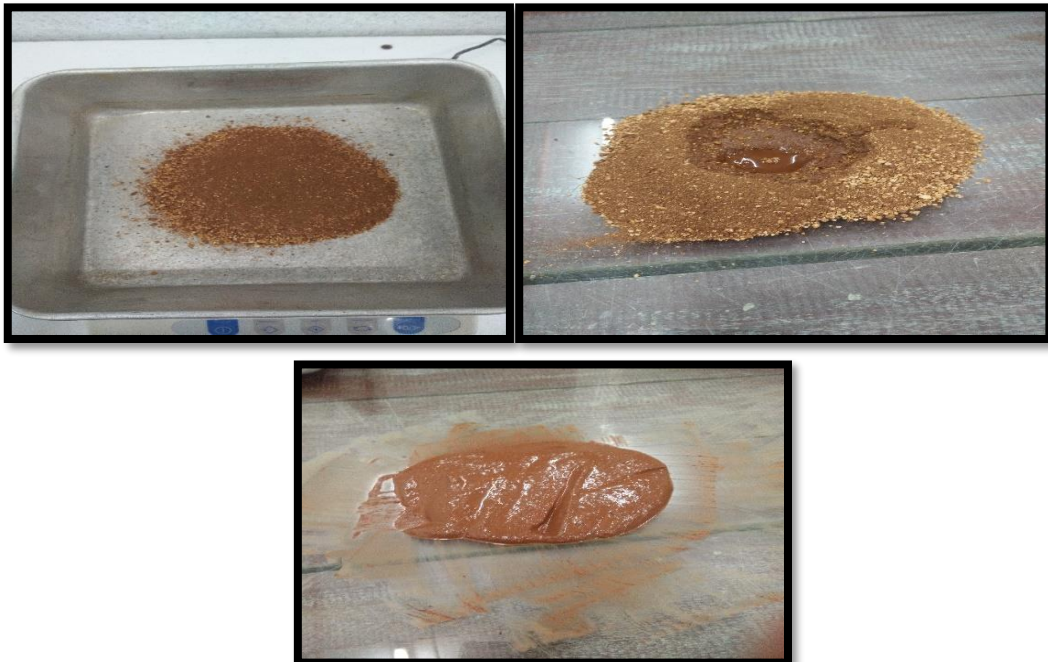


Figure 3.6 Distilled water was added to form soil paste

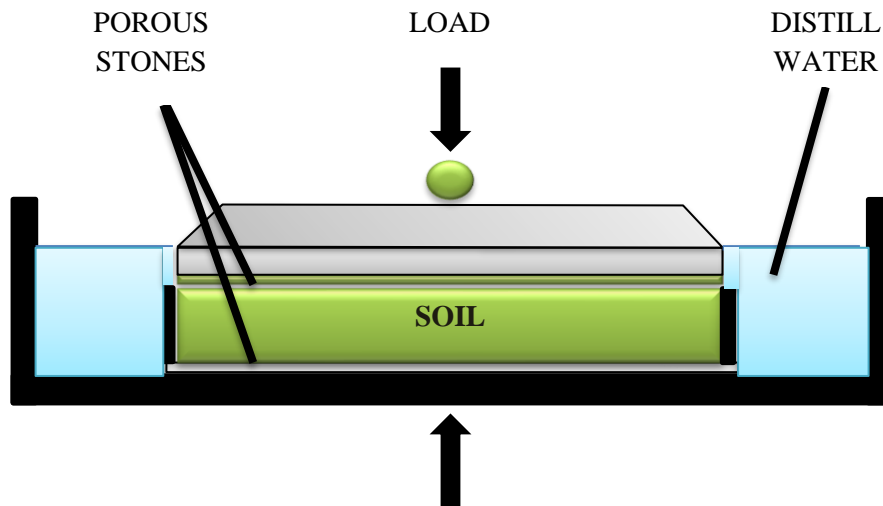


Figure 3.7 Set-up apparatus of soil sample in the ring

After one day, the specimen takes out from the plastic bag and filled the soil in the ring. Ensure that the mass of ring need to measure first. The soil paste was pressed against the side of the ring to avoid trapping air. Filter paper Whatman no 54 to be placed one on top of the test specimen between soil and porous stones and shall have a diameter equal to the inside diameter of the ring and one filter paper to be placed at the bottom of the specimen shall a diameter which need not less than outside diameter of the ring. The set-up apparatus of soil pastes in the ring as shown in Figure 3.7.

Then, add distilled water on top of ring as shown in Figure 3.8 and the sample will be left at room temperature in one day before the oedometer test carried out next.

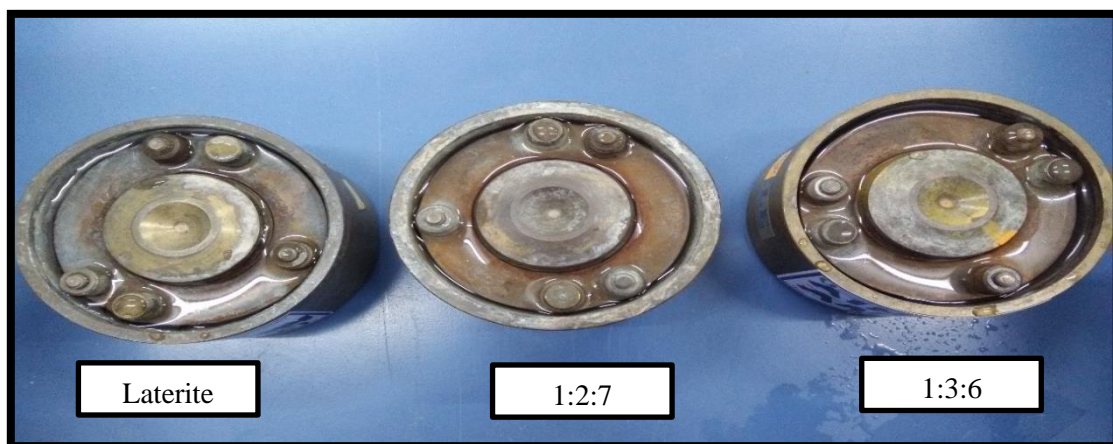


Figure 3.8 Distilled water added at top of ring

The next one day, set-up apparatus for oedometer test to be carried out as shown Figure 3.9. Vertical load pressure of 6.2 kPa was applied for soil laterite, Condisoil 1:2:7 and Condisoil 1:3:6. The specimens were left there about one week for the applied stress to the soil, so that the pore water is expelled out form the specimen.



Figure 3.9 Set-up apparatus of soil sample in the ring a) placed the ring at the top of oedometer apparatus b) applied load pressure c) overview of oedometer set-up apparatus

The soil leachate was collected by using syringe that modified to have long tube for easier to collect the leachate from the ring. The syringe shown in Figure 3.10. Then the soil leachate was kept in the pill box bottle as shown in Figure 3.11 and labelled in sticker. The step and method were repeated for each soil sample for applied pressure 1598.8 kPa. Lastly, the leachate was analysed using Inductively Coupled Plasma Mass Spectrometry (ICP-MS) at Centre Laboratory of University Malaysia Pahang.



Figure 3.10 Syringe used to collect the leachate from the ring

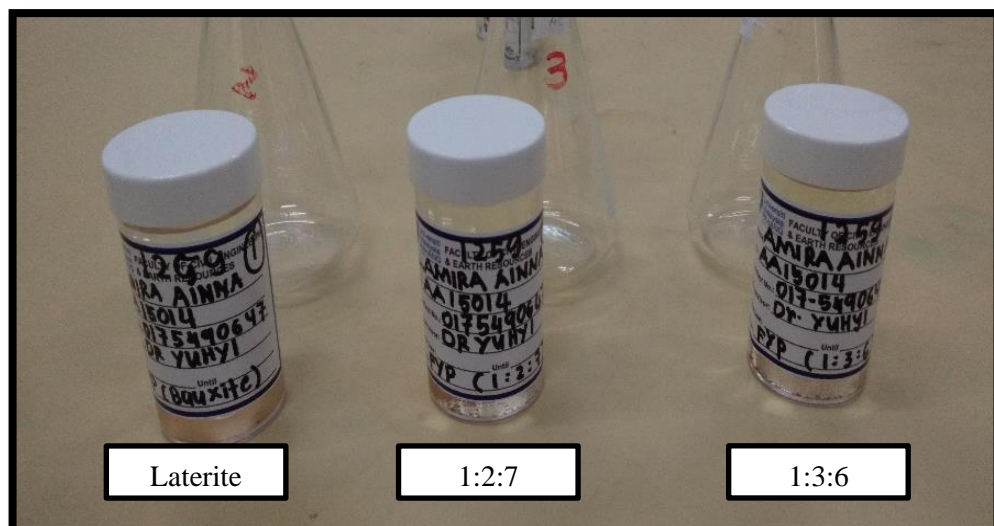


Figure 3.11 The soil leachate were keep in pill box bottle and labelled

3.8 Leaching Analysis using ICP-MS Laboratory Testing

The soil leachate sample were analyzed using Inductively Coupled Plasma Mass Spectrometry (ICP-MS) as shown in Figure 3.12 at Centre Laboratory of University Malaysia Pahang. This machine available to identify concentration such as heavy metal including rare earth elements (REEs) in solid and liquid form.



Figure 3.12 Inductively Coupled Plasma Mass Spectrometry (ICP-MS)

Brand: Agilent. Model: 7500a. Year: 2009

Source: Centre Laboratory of UMP

CHAPTER 4

RESULTS AND DISCUSSION

4.1 Physical Properties

In this chapter, all result was discussed to identify the objective are achieved. The result of soil properties for laterite and its mixed of MRSG which is Atterberg limit also identified.

4.1.1 Effect of MRSG amended soil on Liquid Limit

Table 4.1 Effect of soil properties of laterite soil amended with MRSG

Properties	Laterite	Condisoil 1:2:7	Condisoil 1:3:6
Liquid limit, LL (%)	34.00	40.50	45.00
Plastic limit, PL (%)	21.50	26.55	34.00
Shrinkage limit, SL (%)	14.80	18.40	20.00

Table 4.1 shows liquid limit of laterite soil is 34%. After MRSG was mixed the value was increased to 40.50% for Condisoil 1:2:7. Then, the value was continuously increased to 45% for Condisoil 1:3:6. Through the result of liquid limit, the water content of soil was determined and used as the measurement of soil mixing into paste form for Oedometer test. The moisture content of different soil condition was shown in Table 4.2.

4.1.2 Effect of MRSG amended soil on Plastic Limit

Plastic limit shows an increment from 21.50% for laterite soil to 26.55% for Condisoil 1:2:7. It continuously increased to 34% for Condisoil 1:3:6.

4.1.3 Effect of MRSG amended soil on Shrinkage Limit

Shrinkage limit of Condisoil 1:2:7 increased to 18.40% compared to 14.80% for laterite soil. Meanwhile, for Condisoil 1:3:6 the value still increased to 20%.

Table 4.1 Moisture content of different soil condition

Soil Specimen	Moisture Content (%)
Laterite	32
Condisoil 1:2:7	34
Condisoil 1:3:6	35

4.2 Chemical Characterization of MRSG amended soil in Soil

The MRSG amended soil used in the current study was grounded to powder form. The elements presents were identified using X-Ray Fluorescence (XRF). The elements determined were shown in Table 4.3.

Test result are presented in Table 4.3. It can be noticed that some elements concentration was increased with the MRSG mixing in 1:2:7 and 1:3:6. The elements including P, S, Ca, Mn, Zn, K and Nb. However, elements such as Al, Ti, Si, Ni, Sr, Cr, V, Zr, decreased the concentration when MRSG was amended. These elements show the concentration in laterite soil higher compare to MRSG amended soil. Meanwhile, Ce, Mg, La, Nd, Cl, Th, Pb, Y and Ga were present only in MRSG amended soil. But Cu does not shows change 1 mg/l for laterite soil, Condisoil 1:2:7 and Condisoil 1:3:7.

4.3 Chemical Characteristic of MRSG amended soil in Leaching test

Leaching characteristic of soil were categorized into two type of element which is major element and trace element. The soil elements categories were shown in Table 4.4.

Table 4.3 Element characterization of laterite soil and MRSG amended soil

Element	Laterite	Condisoil 1:2:7	Condisoil 1:3:6
	mg/l		
Fe	2381	2245	1910
Al	987	618	449
Ti	346	295	258
Si	82	59	48
Ni	63	55	45
Sr	43	6	6
P	24	89	119
S	11	208	250
Ca	9	342	481
Cr	7	4	3
Mn	5	10	12
V	5	4	4
Zr	4	3	3
Zn	2	3	3
K	2	3	4
Ce	-	74	82
Mg	-	44	57
La	-	42	46
Nd	-	20	22
Cl	-	8	13
Th	-	4	4
Pb	-	1	1
Y	-	64	73
Ga	-	50	36
As	-	-	-
Ba	-	-	-
Cd	-	-	-
Co	-	-	-
Mo	-	-	-
Sn	-	-	-
Hg	-	-	-

Table 4.4 Elements characterization of MRSG amended soil in leaching test

Major Element	Trace Element
Mg	Cr
Al	Ni
K	Cu
Ca	Ba
Si	Pb
P	La
	Ce
	Nd
	Th

Table 4.4 shows the elements characterization under leaching test. Elements includes Mg, Al, K, Ca, Si, P were categories as major elements. Meanwhile, elements of Cr, Ni, Cu, Ba, Pb, La, Ce, Nd and Th were categories as trace element. The elements classification has been stated in previous study by (Moreno et al., 2018) in the phosphate ore of PG contains impurities of major elements (Si, Al, Fe and Ti), and trace elements (Sr, Cr, V, Zn, Y, Ni, Ba and Th).

4.4 Leaching Characteristic of MRSG amended soil

Oedometeric test and columns test were conducted to collect the leaching of the soil. The result in Figure 4.1, 4.2, 4.3, 4.4, 4.5, and 4.6 shows the leaching characteristic of oedometeric test. Meanwhile, Table 4.5 is the result for the leaching characteristic of columns test.

4.4.1 Oedometric Test

The result from the Oedometer Leaching test analysis using ICP-MS are presented in figure below. Similar to the oedometer test carried out for the determinate of consolidation and settlement behavior, the oedometer test was again employed to determine the leaching potential of laterite and its mixture with WLP and NUF. In this this, the samples were subjected to high vertical applied stress, so that the pore water is expelled out from the specimen. It is commonly expected that cations and anions move out of soil along with expelled water. This test also represents actual on-site conditions when the material is subjected to over-buried press on vertical loading from structures.

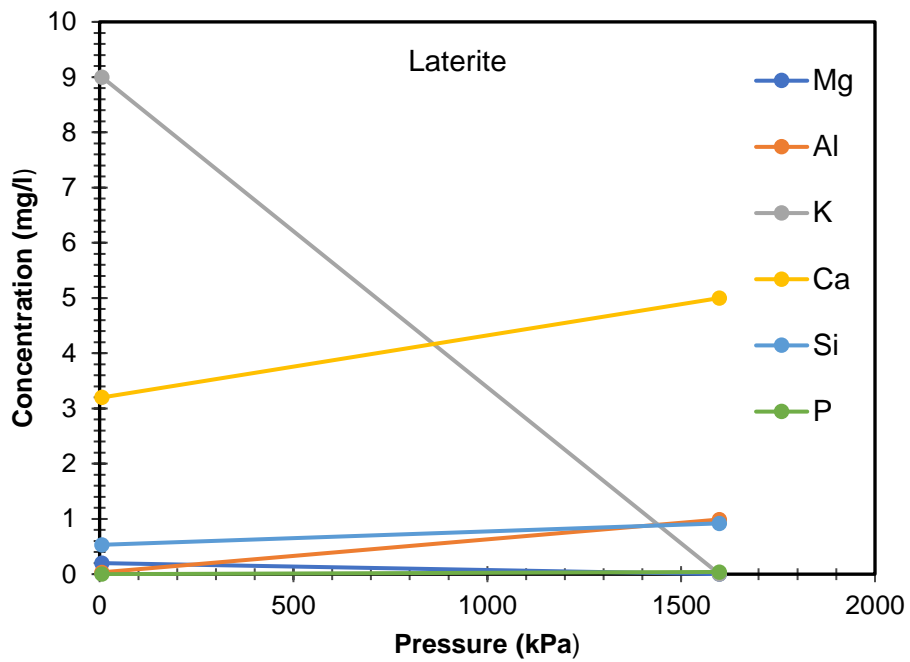


Figure 4.1 Major element Mg, AL, K, Ca, Si, P of laterite soil under Oedometer Leaching Test for load pressure of 6.2 kPa and 1598.8 kPa

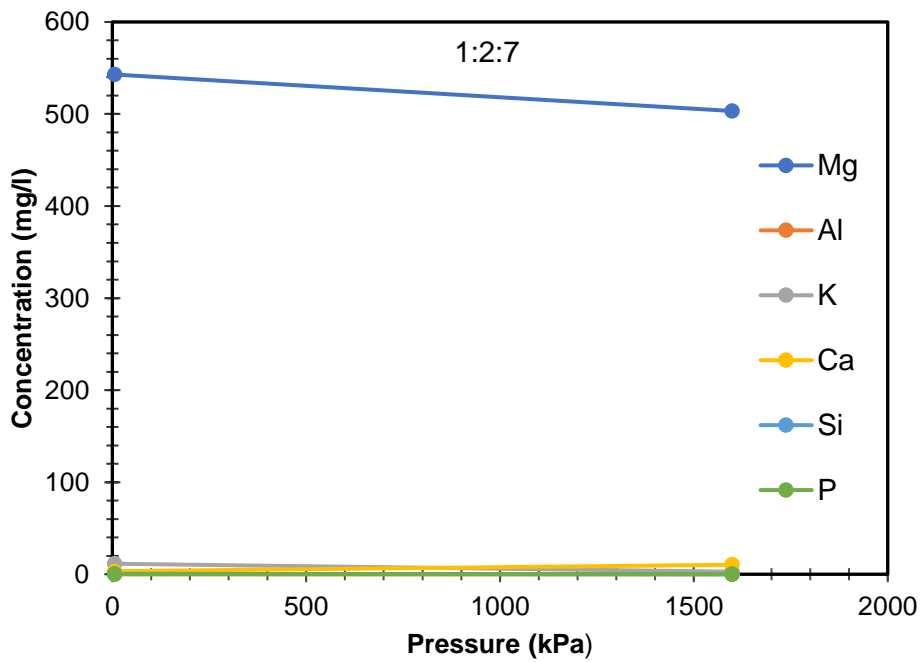


Figure 4.2 Major element Mg, AL, K, Ca, Si, P of MRSG amended soil under Oedometer Leaching Test for load pressure of 6.2 kPa and 1598.8 kPa

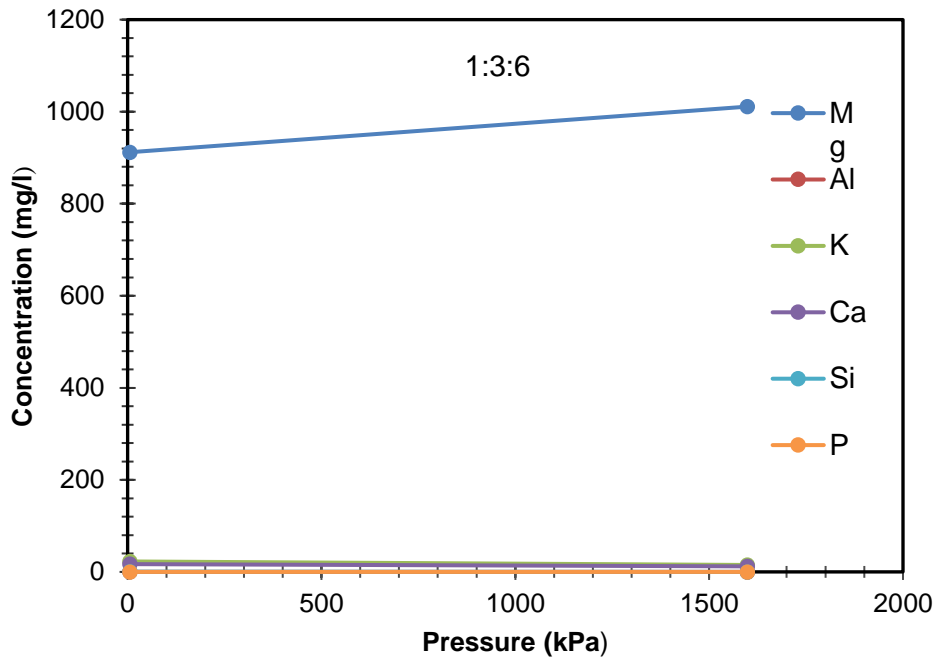


Figure 4.3 Major element Mg, AL, K, Ca, Si, P of MRSG amended soil under Oedometer Leaching Test for load pressure of 6.2 kPa and 1598.8 kPa

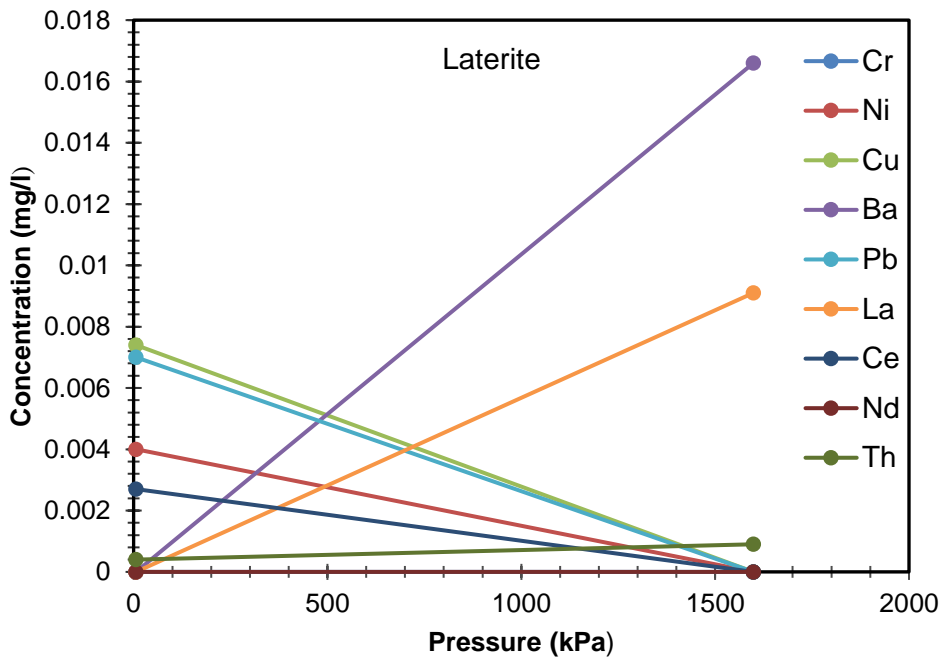


Figure 4.4 Trace element Cr, Ni, Cu, Ba, Pb, La, Ce, Nd, Th of laterite soil under Oedometer Leaching Test for load pressure of 6.2 kPa and 1598.8 kPa

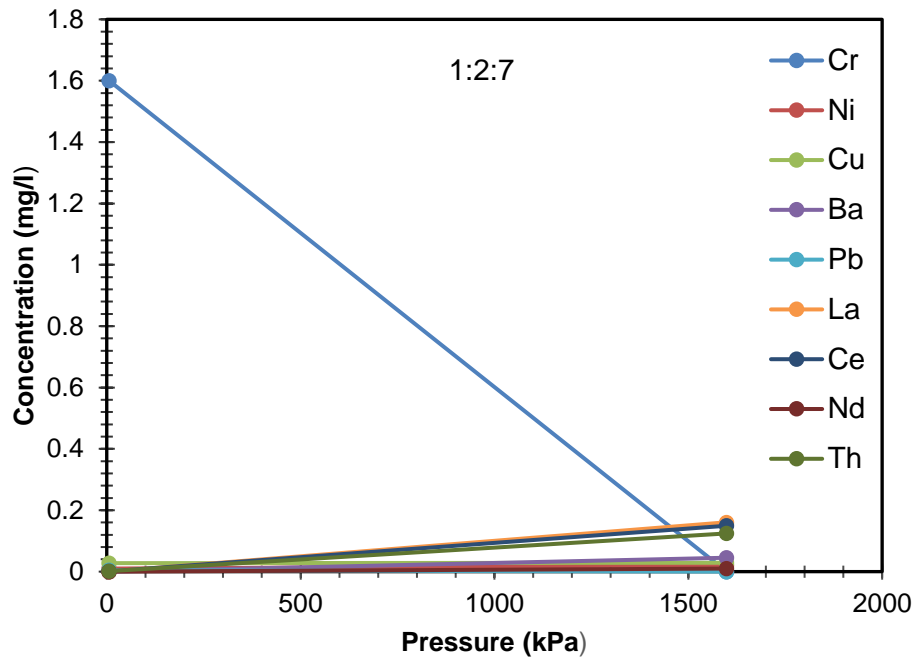


Figure 4.5 Trace element Cr, Ni, Cu, Ba, Pb, La, Ce, Nd, Th of MRSg amended soil under Oedometer Leaching Test for load pressure of 6.2 kPa and 1598.8 kPa

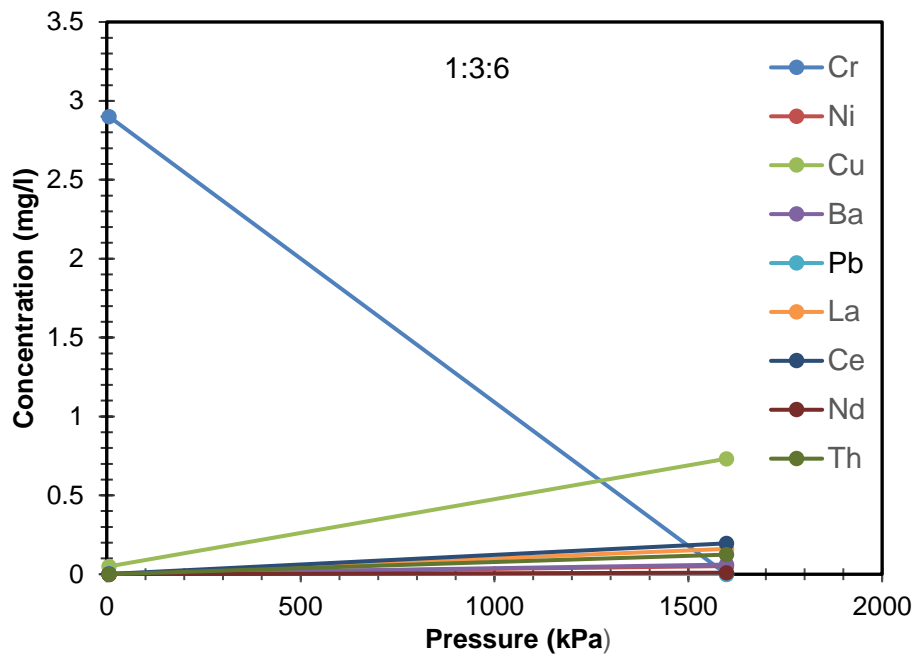


Figure 4.6 Trace element Cr, Ni, Cu, Ba, Pb, La, Ce, Nd, Th of MRSg amended soil under Oedometer Leaching Test for load pressure of 6.2 kPa and 1598.8 kPa

Figure 4.1 shows the concentration of aluminium, calcium, phosphorus and silicon increased linearly with increasing of pressure load on soil sample. However, there were some elements concentration diminished with the increment of load such as magnesium and potassium. It is contrast with the expected development as the samples were subjected to high vertical applied stress, so more pore water is expelled out from the specimen. Figure 4.2 shows the leaching of magnesium increased highly to 542.9 mg/l after the laterite was mixed with MRSG. But the concentration marked drop was recorded at 503.3 mg/l when load pressure increased. According to subjected to high vertical applied stress, the concentration increased when pressure increased. Due to increasing pressure load the concentration of magnesium is expected to increased. The leaching of magnesium in Figure 4.3 shows it rise highly in Condisoil 1:3:6 about 911.9 mg/l for 6.2 kPa and go up to 1011 mg/l when applied load increased. A study conducted by (Idris, 2012) said that high amount of Mg content in the soils has an adverse effect on soil structure, flocculation and infiltration ratio which is a rather sensitive indicator of soil physical conditions.

Meanwhile, Figure 4.4 shows the trace element Cr, Ni, Cu, Ba, Pb, La, Ce, Nd, Th of laterite soil under Oedometer Leaching Test for load pressure of 6.2 kPa and 1598.8 kPa. The concentration of barium, lanthanum and thorium were rose with the increment of applied load. But, the element concentration of nickel, copper, lead and cerium reduced with the increment of load. The result contrast with the study in engineering perspective, an increase in the applied stress on sample would lead to the increment of cations and anions move out of soil along with expelled water. Besides, chromium and neodymium were not detected in the laterite soil. However, in Figure 4.5 and Figure 4.6 shows increment of chromium concentration to 1.6 mg/l for Condisoil 1:2:7 and rise to 2.9 mg/l for Condisoil 1:3:7 after laterite mixed with MRSG for pressure load 6.2 kPa but it decreased in the load increased. Element concentration of lanthanum, thorium, nickel, copper and thorium shows an increment when soil amended with MRSG and grow up when applied pressure increased for both 1:2:7 and 1:3:6. The finding of this result was found to be similar to that when the samples were subjected to high vertical applied stress, so that more pore water is expelled out from the specimen.

4.4.2 Columns Test

The concentration of leached contaminants shown in Table 4.5. Based on the test results, the concentration is high in laterite leachate out. This is expected due to the fact that laterite is highly weathered material. Thus, the reading obtained from laterite may be considered as background concentrations for determining the leaching characteristics of WLP and NUF mixtures.

Table 4.5 Element concentration of laterite and MRSG amended soil under column test

Parameter	Column		
	Laterite	Condisoil 1:2:7	Condisoil 1:3:6
	mg/l		
Mg	-	-	-
Al	0.323	0.0447	0.0306
K	-	-	-
Ca	4.7	3.6	2.7
Cr	-	-	-
Ni	-	-	-
Cu	-	-	-
Ba	-	-	-
Pb	-	-	-
Si	-	-	-
P	-	-	-
La	-	-	-
Ce	-	-	-
Nd	-	-	-
Th	-	-	-

Table 4.5 shows there is 2 chemical species were detected in minor concentration. These is including aluminium and calcium. Aluminum and calcium concentration reduced in Condisoil compared to laterite. Aluminium reduced by 86% and 91% while calcium concentration reduced by 23% and 43% in Condisoil 1:2:7 and 1:3:6, respectively.

4.5 Comparing Oedometer and Columns Leaching Test

This table section describes the comparison analysis result between oedometer and column leaching test

Table 4.6 Comparison of oedometer and columns leaching test at laterite soil and condisoil

Element	Laterite		
	6.2 kPa	1598.8 kPa	Column
Al	0.034	0.99	0.323
Ca	3.2	5	4.7

Element	Condisoil 1:2:7		
	6.2 kPa	1598.8 kPa	Column
Al	0.058	-	0.0447
Ca	3.3	10.3	3.5

Element	Condisoil 1:3:6		
	6.2 kPa	1598.8 kPa	Column
Al	0.15	-	0.0306
Ca	16.9	12	2.3

The Table 4.6 shows the result of the leaching concentration of two different method of test. Clearly, shows the concentration of both element in oedometer leaching test higher compared to columns. A possible reason for such occurrence could be that due to high vertical applied stress on sample in oedometer leaching test. Cations and anions come out by the water in the pores out from the specimen. The increase in pressure within the pore water causes more chemical properties will move out. From previous study by Lin et al. (2017) said the element showed different amount of release between batch and column tests. Grathwohl and Susset (2009) reported that for the elements that leached more from batch test, these elements might not have achieved equilibrium condition in the column test.

4.6 Comparing Chemical Properties with TCLP and NDWQS

Table 4.6 shows the allowable concentration at contamination for TCLP and NDWQS. According to Environment, Health and Safety Online (EHSO) stated Toxicity Characteristic Leaching Procedure (TCLP) is designed to determine the mobility of both organic and inorganic analytes present in liquid, solid and multiphase waste. This is usually used to determine if waste may meet the definition of EP toxicity, that is, carrying a hazardous waste. Toxicity wastes are harmful or fatal when ingested or absorbed. When toxic wastes are land disposed, contaminated liquid may leach from the waste and pollute groundwater. The TCLP helps identify wastes likely to leach concentrations of contaminants that may be harmful to human health or the environment. Meanwhile, National Drinking Water Quality Standard (NDWQS) serves as a benchmark to provide recommended water quality criteria for respective parameters. A Slightly polluted water body would require treatment prior to use for drinking or domestic purpose and much polluted water would only be suitable for limited purpose.

The leaching characteristic of trace element and major element in soil residues were investigated using the standard TCLP and NDWQS as shown in Table 4.6. The concentration of major element and trace element both in oedometer and columns leaching test were met the TCLP limit. The level of their concentration was below the critical limit set by the TCLP. Based on result studies by Sahibin et al. (2018) the level concentration of heavy metal and other elements of concern in the soil was below the critical limit set by the Standard and Industrial Research Institute of Malaysia (SIRIM) and the Department of Agriculture (DOA), which are the relevant authorities and agencies in Malaysia responsible for introducing guidelines for the usage of schedule wastes on agriculture land. It means that MRSG used in the current study has the potential to be safely applied on land application.

But element concentration of magnesium, chromium, nickel for oedometer test, aluminium for both leaching tests were exceeding the limit of NDWQS. It is contrast with previous study by Sahibin et al. (2018) stated that heavy metal and other elements of concern in the surface water before and after MRSG application were below the benchmark value of Ministry of Health Malaysia (2000). Water quality indexing is a common method used by many countries to assess the overall status of rivers. Although these indices differ from country to country, it is based on a few pre-determined physico-

chemical parameters and compounded to numerical rating for evaluation of river water quality (Bhargava, 1983; Pesce & Wunderlin, 2000).

Table 4.7 Chemical properties of oedometeric and columns test compare to TCLP and NSWQS

Element	Oedometer						Column			TCLP	NDWQS
	6.2 kPa			1589.8 kPa			Laterite	1:2:7	1:3:6		
	Laterite	1:2:7	1:3:6	Laterite	1:2:7	1:3:6					
	mg/l										
Mg	0.2	542.9	911.9	-	503.3	1011	-	-	-	-	150
Al	0.034	0.058	0.15	0.99	-	-	0.323	0.0447	0.0306	-	0.2
K	9	11.4	22.7	-	2.8	15.1	-	-	-	-	-
Ca	3.2	3.3	16.9	5	10.3	12	4.7	3.5	2.3	-	-
Cr	-	1.6	2.9	-	-	-	-	-	-	5	0.05
Ni	0.004	0.01	0.004	-	0.021	0.052	-	-	-	-	0.02
Cu	0.0074	0.0277	0.05	-	0.029	0.7322	-	-	-	-	1
Ba	-	-	-	0.0166	0.0453	0.0613	-	-	-	100	-
Pb	0.007	0.0047	0.0046	-	-	-	-	-	-	5	0.01
Si	0.532	0.451	0.522	0.92	-	-	-	-	-	-	-
P	-	-	-	0.0401	-	-	-	-	-	-	-
La	-	0.0002	0.0001	0.0091	0.1608	0.1608	-	-	-	-	-
Ce	0.0027	0.001	0.0015	-	0.1496	0.196	-	-	-	-	-
Nd	-	-	-	-	0.01	0.01	-	-	-	-	-
Th	0.0004	0.0016	-	0.0009	0.1245	0.1245	-	-	-	-	-

CHAPTER 5

CONCLUSION

Based on the findings in the thesis, the following conclusion were draw.

1. Based on the result of leaching characteristic of laterite soil amended with MRSG, the chemical properties of soil can be categorized into two type of elements which is major element and trace element. In major element shows magnesium concentration become higher when laterite was mixed with WLP and NUF. Followed by 524.9 mg/l for condisoil 1:2:7, 911.9 mg/l for condisoil 1:3:6 under 6.2 kPa while under 1589.8 kPa concentration increased by 508.3 mg/l to 1011 mg/l in condisoil, respectively. Meanwhile, for trace element concentration of lanthanum increased to 0.1608 mg/l in condisoil, thorium concentration increased to 0.1245 mg/l, nickel concentration increased to 0.021 mg/l and 0.052 mg/l in condisoil, copper concentration increased to 0.029 mg/l and 0.7322 mg/l and thorium concentration increased to 0.1245 mg/l under 1589.8 kPa.
2. Experimental results demonstrated that element concentration recoded in Oedometer leaching test were higher compared to Columns leaching test. It is because the sample in Oedometer test were subjected to high vertical applied stress. So that the pore water is expelled out from the specimen. It is commonly expected that cations and anions move out of soil along with expelled water.
3. From the results according to leaching behavior, comparison of elements concentration in Oedometer leaching test and Columns leaching test to TCLP limit shows the level of their concentration was below the critical limit set by the TCLP. This is relevant stated by authorities in Malaysia responsible for introducing guidelines for usage of schedule waste on land application. However,

result of comparison to NDWQS shows element concentration of magnesium, chromium, nickel and aluminium exceed the limit but from previous studies stated heavy metals and other elements of concern in the surface water before and after MRSG application were below the bench mark value of Ministry of Health Malaysia. The drinking water standard of Malaysia is adopted from World Health Organizations standard. It means that the MRSG used in the current study has the potential to be safely applied on land application. Moreover, (SIRIM, 2017) confirms that both formulations of Condisoil are non-toxic, non-carcinogenic and compliant with all test parameters including heavy metal.

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APPENDIX A

A Toxicity Characteristic Leaching Procedure (TCLP)

Maximum Concentration of Contaminants for the Toxicity Characteristic Leaching Procedure (TCLP).

DOE CW No. ¹	Contaminant	CAS No. ²	Maximum Level (mg/L)
C004	Arsenic	7440-38-2	5.0
C005	Barium	7440-39-3	100.0
C018	Benzene	71-43-2	0.5
C006	Cadmium	7440-43-9	1.0
C019	Carbon tetrachloride	56-23-5	0.5
C020	Chlordane	57-74-9	0.03
C021	Chlorobenzene	108-90-7	100.0
C022	Chloroform	67-66-3	6.0
C007	Chromium	7440-47-3	5.0
C023	o-Cresol	95-48-7	200.0 ³
C024	m-Cresol	108-39-4	200.0 ³
C025	p-Cresol	106-44-5	200.0 ³

C026	Cresol		200.0 ³
C016	2,4-D	94-75-7	10.0
C027	1,4-Dichlorobenzene	106-46-7	7.5
C028	1,2-Dichloroethane	107-06-2	0.5
C029	1,1-Dichloroethylene	75-35-4	0.7
C030	2,4-Dinitrotoluene	121-14-2	0.13
C012	Endrin	72-20-8	0.02
C031	Heptachlor (and its epoxide)	76-44-8	0.008
C032	Hexachlorobenzene	118-74-1	0.13
C033	Hexachlorobutadiene	87-68-3	0.5
C034	Hexachloroethane	67-72-1	3.0
C008	Lead	7439-92-1	5.0
C013	Lindane	58-89-9	0.4
C009	Mercury	7439-97-6	0.2
C014	Methoxychlor	72-43-5	10.0
C035	Methyl ethyl ketone	78-93-3	200.0
C036	Nitrobenzene	98-95-3	2.0
C037	Pentachlorophenol	87-86-5	100.0
C038	Pyridine	110-86-1	5.0
C010	Selenium	7782-49-2	1.0
C011	Silver	7440-22-4	5.0
C039	Tetrachloroethylene	127-18-4	0.7
C015	Toxaphene	8001-35-2	0.5
C040	Trichloroethylene	79-01-6	0.5
C041	2,4,5-Trichlorophenol	95-95-4	400.0
C042	2,4,6-Trichlorophenol	88-06-2	2.0
C017	2,4,5-TP (Silvex)	93-72-1	1.0
C043	Vinyl chloride	75-01-4	0.2

APPENDIX B

B National Drinking Water Quality Standard (NDWQS)

Drinking water quality standard

Parameter	Group	RECOMMENDED RAW WATER QUALITY	DRINKING WATER QUALITY STANDARDS
		Acceptable Value (mg/litre (unless otherwise stated))	Maximum Acceptable Value (mg/litre (unless otherwise stated))
Total Coliform	1	5000 MPN / 100 ml	0 in 100 ml
<i>E.coli</i>	1	5000 MPN / 100 m	0 in 100 m
Turbidity	1	1000 NTU	5 NTU
Color	1	300 TCU	15 TCU
pH	1	5.5 - 9.0	6.5 - 9.0
Free Residual Chlorine	1	-	0.2 - 5.0
Combined Chlorine	1	-	Not Less Than 1.0
Temperature	1	-	-
<i>Clostridium perfringens</i> (including spores)	1	-	Absent
Coliform bacteria	1	-	-
Colony count 22°	1	-	-
Conductivity	1	-	-
Enterococci	1	-	-
Odour	1	-	-
Taste	1	-	-
Oxidisability	1	-	-
Total Dissolved Solids	2	1500	1000
Chloride	2	250	250
Ammonia	2	1.5	1.5
Nitrat	2	10	10
Ferum/Iron	2	1.0	0.3
Fluoride	2	1.5	0.4 - 0.6
Hardness	2	500	500
Aluminium	2	-	0.2
Manganese	2	0.2	0.1
Chemical Oxygen Demand	2	10	-
Anionic Detergent MBAS	2	1.0	1.0
Biological Oxygen Demand	2	6	-

Nitrite	2	-	-
Total organic carbon (TOC)	2	-	-
Mercury	3	0.001	0.001
Cadmium	3	0.003	0.003
Arsenic	3	0.01	0.01
Cyanide	3	0.07	0.07
Plumbum/Lead	3	0.05	0.01
Chromium	3	0.05	0.05
Cuprum/Copper	3	1.0	1.0
Zinc	3	3	3
Natrium/Sodium	3	200	200
Sulphate	3	250	250
Selenium	3	0.01	0.01
Argentum	3	0.05	0.05
Magnesium	3	150	150
Mineral Oil	3	0.3	0.3
Chloroform	3	-	0.2
Bromoform	3	-	0.1
Dibromoklorometana	3	-	0.1
Bromodiklorometana	3	-	0.06
Fenol/Phenol	3	0.002	0.002
Antimony	3	-	0.005
Nickel	3	-	0.02
Dibromoacetonitrile	3	-	0.1
Dichloroacetic acid	3	-	0.05
Dichloroacetonitrile	3	-	0.09
Trichloroacetic acid	3	-	0.1
Trichloroacetonitrile	3	-	0.001
Trihalomethanes - Total	3	-	1.00
Aldrin / Dealdrin	4	0.00003	0.00003
DDT	4	0.002	0.002
Heptachlor & Heptachlor Epoxide	4	0.00003	0.00003
Methoxychlor	4	0.02	0.02
Lindane	4	0.002	0.002
Chlordane	4	0.0002	0.0002
Endosulfan	4	0.03	0.03
Hexachlorobenzena	4	0.001	0.001
1,2-dichloroethane	4	-	0.03
2,4,5-T	4	-	0.009
2,4,6-trichlorophenol	4	-	0.2
2,4-D	4	0.03	0.03
2,4-DB	4	-	0.09
2,4-dichlorophenol	4	-	0.09
Acrylamide	4	-	0.0005

Alachlor	4	-	0.02
Aldicarb	4	-	0.01
Benzene	4	-	0.01
Carbofuran	4	-	0.007
MCPA	4	-	0.002
Pendimethalin	4	-	0.02
Pentachlorophenol	4	-	0.009
Permethrin	4	-	0.02
Pesticides	4	-	-
Pesticides - Total	4	-	-
Polycyclic aromatic hydrocarbons	4	-	-
Propanil	4	-	0.02
Tetrachloroethene and Trichloroethene	4	-	-
Vinyl chloride	4	-	0.005
Gross alpha (α)	5	0.1Bq/l	0.1Bq/l
Gross beta (β)	5	1.0 Bq/l	1.0 Bq/l
Tritium	5	-	-
Total indicative dose	5	-	-