EFFECT OF LEACHING ON LIME STABILISED CONTAMINATED SOIL AND MINING WASTE

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Dedication to my beloved wife Suraya Sintang
My children Hilal Hakimy, Bahrul Ilmy and Ain Nazyfah
To my parents, sisters and brother

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ABSTRACT

Degradation of environment due to industrial and mining related activities is identified as one of the major problem facing by most industrial and developing countries. Therefore, scientific research are needed to acquire knowledge to manage such toxic waste, which has a potential to degrade on the environment as well as on human health. Leaching column tests were conducted on two soil samples and one mining waste sample in order to investigate the potential of lime stabilisation technique to immobilise heavy metals. The soil samples were obtained from Kg. Kayumadang, Telipok, Sabah (KMT) and Kg. Bongkud, Ranau, Sabah (KGB), whereas the mining waste was obtained from Lohan Dam, Ranau, Sabah (LDA). The amounts of lime used for the stabilisation were 2%, 4%, 6%, and 8% of dry weight. Five selected heavy metals namely Cu, Cr, Ni, Pb, and Zn were chosen to understand their behavioural changes with the long term leaching process. These heavy metals were spiked into the unstabilised and lime stabilised samples in the leaching columns. The pH values of leachate from all stabilised KMT, KGB, and LDA samples were found to be alkaline, indicating that the heavy metals were adsorbed on the surface of the cementitious minerals, whilst the Ca ions were released from the cementitious compound. The trend of migration profiles showed that the concentrations of the heavy metals decreased with depth for all samples. On the other hand, very low concentrations of heavy metals were detected in the leachate of the lime stabilised samples. Mass balance calculations showed that Zn had the highest mobility in KMT sample, whereas Ni was the highest in KGB and LDA samples. Calculations also showed that Pb was the least mobile in KMT, whereas Cr was the slowest in both KGB and LDA samples. The permeability of all stabilised samples decreased significantly immediately after leaching processes due to the clogging of pore by fine materials and the development of cementitious mineral. Scanning electron microscope (SEM) study showed that the bottom part of the leached columns had tights structure with low pore spaces, which were clogged-up by fine materials. Using X-ray diffraction (XRD) technique, quartz, kaolinite, feldspar and cementitious minerals were detected in the stabilised samples. Nevertheless, SEM method illustrated better image of the cementitious mineral. Soil classification tests, chemical characterisations, mineralogical identifications, and engineering properties verification were also performed on both unstabilised and stabilised samples to examine the liming effect on their properties. Maximum unconfined compressive strength (UCS) was achieved by adding 6% lime to the KMT, KGB, and LDA samples, and this strength increased with age. Based on the leaching column and UCS tests, 6% lime is suggested as the optimum value for stabilisation of KMT and KGB samples. Hence, lime stabilised KMT and KGB could be used as a clay liner due to their low permeability and high strength. It is also possible to stabilise LDA with 6% lime with the addition of fine material to facilitate stabilisation process, thus immobilising heavy metals within the stabilised mining waste.

ABSTRAK

Penurunan kualiti alam sekitar akibat aktiviti-aktiviti berkaitan perindustrian dan perlombongan menjadi masalah utama yang dihadapi oleh kebanyakan negara industri dan negara membangun. Oleh yang demikian kajian saintifik adalah perlu untuk memperolehi pengetahuan bagi mengurus bahan buangan bertoksik yang berupaya menurunkan kualiti alam sekitar dan kesihatan manusia. Ujian kolum larutlesapan telah dijalankan ke atas dua sampel tanah dan satu sampel sisa lombong bertujuan mengkaji keupayaan kaedah penstabilan kapur untuk mentakmobilkan logam-logam berat. Sampel-sampel tanah diperolehi daripada Kg. Kayumadang, Telipok, Sabah (KMT) dan Kg. Bongkud, Ranau, Sabah (KGB), manakala sisa lombong diperolehi daripada Empangan Lohan, Ranau, Sabah (LDA). Peratusan kapur yang digunakan untuk penstabilan adalah 2%, 4%, 6%, dan 8% dengan berat kering sampel. Lima logam berat terpilih iaitu Cu, Cr, Ni, Pb, and Zn telah dipilih untuk kajian perubahan perlakuannya dengan proses larutlesapan jangka panjang. Logam-logam berat ini digaul kedalam sampel-sampel tidak distabilkan dan distabilkan dalam kolum larutlesapan. Nilai pH cecair larutlesapan daripada kesemua sampel-sampel KMT, KGB, dan LDA yang distabilkan didapati beralkali, menandakan logam-logam berat telah dijerap di permukaan mineral bersimen, manakala ion Ca telah dilepaskan daripada sebatian bersimen. Kajian tren profil-profil migrasi menunjukkan nilai kepekatan logam-logam berat berkurangan dengan kedalaman pada semua sampel. Sebaliknya, logam-logam berat dalam cecair larutlesapan daripada tanah yang distabilkan didapati amat rendah. Kiraan keseimbangan jisim menunjukkan Zn mempunyai mobiliti yang tinggi dalam sampel KMT, manakala Ni adalah tertinggi dalam sampel-sampel KGB dan LDA. Kiraan juga menunjukkan bahawa Pb mempunyai mobiliti terendah dalam sampel KMT, manakala Cr adalah terendah dalam sampel-sampel KGB dan LDA. Kebolehtelapan kesemua sampel yang distabilkan berkurang secara mendadak selepas proses larutlesapan di sebabkan oleh penyumbatan bahan-bahan halus dan pembentukan mineral bersimen. Kajian mikroskop elektron pengimbas (SEM) menunjukkan struktur padat dengan ruang pori yang rendah dan penyumbatan bahan-bahan halus pada bahagian bawah kolum. Kuarza, kaolinit, feldspar dan mineral bersimen dalam sampel-sampel yang telah distabilkan dikesan dengan menggunakan pembelauan sinar-X (XRD). Walaubagaimanapun, mineral bersimen dilihat lebih jelas dalam SEM. Ujian-ujian pengelasan tanah, pencirian kimia, pengenalan mineralogi, dan penentuan ciri-ciri kejuruteraan juga dijalankan keatas sampel-sampel yang tidak distabilkan dan yang distabilkan. Sampel-sampel tanah dan sisa lombong yang ditambah dengan 6% kapur menyebabkan kekuatan mampatan takterkurung (UCS) yang maksima dicapai dan ia juga bertambah dengan masa. Berdasarkan ujian-ujian kolum larutlesapan dan UCS di atas, 6% kapur dicadangkan sebagai nilai optimum untuk penstabilan sampel KMT dan KGB. Penstabilan keatas KMT dan KGB dicadangkan sebagai pelapik lempung disebabkan kebolehtelapan yang rendah dan kekuatan yang tinggi. Penstabilan LDA dengan 6% kapur di samping tambahan bahan halus juga boleh dilakukan bagi mempermudahkan proses penstabilan, seterusnya **mentakgerakan** logam berat di dalam sisa lombong yang distabilkan.

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AAS	-	Atomic Absorption Spectrophotometry
ASTM	-	American Society of Testing and Material.
BS	-	British Standard.
Ca	-	Calcium.
CASH	-	Calcium Aluminate Silicate Hydrate.
CEC	-	Cation Exchange Capacity.
Ce/Co		Relative Concentration
Cr	-	Chromium.
CSH	-	Calcium Silicate Hydrate.
Cu	-	Copper.
I_P	-	Plasticity Index.
k	-	Permeability.
L_L	-	Liquid Limit.
Meq	-	Milliequivalents.
Ni	-	Nickel.
OMC	-	Optimum Moisture Content.
Pb	-	Lead.
ρ_D	-	Dry density.
P_L	-	Plastic Limit.
PV	-	Pore Volume.

ppm	-	Parts Per Million.
SEM	-	Scanning Electron Microscope.
SG	-	Specific Gravity.
S_L	-	Shrinkage Limit.
SSA	-	Specific Surface Area.
Wo	-	Moisture content.
XRD	-	X-Ray Diffraction.
XRF	-	X-ray Fluorescence.
Zn	-	Zinc.

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CHAPTER I

INTRODUCTION

1.1 Related Environmental Issues

The global environmental issues today focused on the problems of contamination of land, water and air. Disposal of municipal solid waste and chemical contamination have become a major problem currently faced by most developing and industrial countries. This is due to the increasing population (consumer) and rapid increase in the industrial by-products. In conjunction with the continuing environmental problems, the United Nations held a Conference on Environment and Development (UNCED), in Rio de Janeiro in 1992. As a result, the conference declared the fundamental principles and the programmed action for achieving sustainable development, which is called 'Rio Declaration' (UNCED, 1992). The Principle 15 in the Rio Declaration mentioned, that "*In order to protect the environment, the precautionary approach shall be widely applied by states according to their capabilities. Where there are threats of serious or irreversible damage, lack of full scientific certainty shall not be used as a reason for postponing cost-effective measures to prevent environmental degradation*". For that reason, part of declaration has urged the researchers to focus their research to find the implementation of the best technique to manage the toxic chemical. The scientific and technological activity in chapter 19 of Agenda 21 in Rio Declarations aims to strengthen research on safer alternatives to toxic chemicals that pose an unreasonable and otherwise unmanageable risk to the environment or human health and to those that are toxic, persistent and bio-accumulative and that cannot be adequately controlled. This indicates that scientific research is needed to prevent and to manage the toxic chemical, which has potential to degrade our environment as well as human health. Therefore, one of the major task faced by the geoenvironmental engineering researchers today is to develop methods to evaluate the release, transport, and long term changes of contaminants from the waste disposal sites such as landfill, containment barrier and, contaminated land.

In conjunction with the Rio Declarations, the world summit on sustainable development that was held in Johannesburg in 2002 has renewed the Declarations to manage chemicals throughout their life cycle and of hazardous wastes for sustainable development and for the protections of human health and the environment (UNCED, 2002). Amongst the actions were to encourage partnership to promote activities aimed at enhancing environmentally sound management of chemicals and hazardous wastes and to promote reduction of the risks posed by heavy metals that are harmful to human health and the environment.

In Malaysia, the phenomenon of environmental degradation showed similar pattern with most other countries where contamination is a major issue. For example the trend of water quality status of river basins over the past 10 years (trend between 1990 – 1999) show the reduction in the number of clean river (DOE, 1999). However, it was also reported that in terms of heavy metal contamination out of the 5,274 river water samples analysed for heavy metals only 7% of the samples exceeded Class III (Extensive Treatment Required). It is noticed that two main sources of water pollution were manufacturing industries and sewage effluent, where the percentage is 43.7% and 45.5% respectively. In Malaysia, generally land contamination is still under control and low, nevertheless, certain areas such as mining waste containment and landfill area should be concerned due to the high risk of environmental degradation. In the long term processes the accumulation of chemicals such as heavy metals in that particular area would cause enrichment of the chemical contaminants. The heavy metals also could degrade the land and water systems by leaching processes. It could bring damage to human health through drinking water or food from plants and animals, which took the chemicals from the contaminated land and contaminated water.

Normally a clay liner is used at the bottom part of a landfill to provide protection for the surrounding soil and ground water from contamination through leaching process. In general, a liner system consists of both a geomembrane barrier and a natural clay liner. Although the geomembrane is an absolute barrier nevertheless, heavy metals pollutants can permeate it by permeation due to different pressure and diffusion (Sharma & Lewis, 1994). In other words the leaching process also could occur through the geomembrane in the landfill area. In an improperly managed or old landfill area and mining waste containment, the liquid, which contained heavy metal is derived from the interaction of rainwater with solid waste or contaminated soil, contact with surface water bodies or groundwater. Hence, the heavy metals such as copper, chromium, nickel, lead, and zinc from the hazardous waste can diffuse into the groundwater or surface water systems. To prevent the contaminant from passing into the surrounding environment, the first step in landfill and containment design is to site them far from the groundwater table and far from groundwater abstraction wells. The second step is to modify or stabilise the clay liner to increase the capability of soil to adsorb and immobilise the heavy metals. Furthermore, the waste itself can be stabilised with suitable chemical such as lime and other active material. Stabilisation or solidification using lime could also improve the physical properties of the soil and capability to increase its durability and strength. Therefore, understanding the physical characteristic, chemical properties, mineralogical identification, microstructural study of both soil and waste are important for the stabilisation purposes. It is also necessary to understand the nature of the environment and the transportation processes of heavy metals in soil and water systems by means of leaching test. Leaching test is essential to evaluate the performance of lime and additive chemical as a stabilisation agent to immobilise the heavy metals in the landfill area and in the mining waste containment. Design and development of physical laboratory testing is essential to assess the potential application before suggestion can be made. This research will look into two major applications namely the stabilisation of contaminated soil and the improvement of landfill system through stabilised clay liner.

1.2 Study Area

The study areas are located in Kayumadang Telipok, Kg. Bongkud Ranau and Lohan Dam Ranau, Sabah. Kayumadang is one of the villages in Telipok, Sabah and located about 20 km from the northern part of Kota Kinabalu City Centre. There is a new landfill area in this village, which commenced operation in 1995. The waste disposal from domestic consumer and waste from the industrial area in the Kota Kinabalu area were disposed of in this landfill. The soil samples were collected from the road cuts near the landfill area and named as KMT samples (Kayu Madang, Telipok) i.e situated at the longitude of 116°10.722"E and latitude 06°06.441"N.

Kg Bongkud is situated in the Ranau district that is in the northern part of Lohan valley, Ranau is located at the longitude of 116°44.988"E and latitude 06°01.666"N. This village is bordered by intrusion of igneous rock in the northern part. The igneous rocks intruded the sedimentary rock of Crocker Formation. The intrusion of igneous is believed to be the factor of mineral accumulation in Ranau area and has good opportunity for the mining activity. The soil sample was collected from the hilly area. This residual soil derived from the weathering of igneous and sedimentary rocks are named as KGB samples (Kg. Bongkud, Ranau).

Mamut Copper Mining is located in Ranau district at the western coast of Sabah, Malaysia about 68 kilometers to the eastern part of Kota Kinabalu City Center, that is at the longitude of 116°44.166"E and latitude 06°00.691"N. It is the largest copper mine, and the largest producer of gold and copper in Malaysia. But, there are not much mineable reserve left when the mines end its production in the year 2000. The production of waste and haul ore average rate is 10 million tonnes and 6 million tonnes a year respectively. The mining waste is dumped in the eastern part of the mining area located at Lohan Dam, Ranau, Sabah. The mining waste sample obtained is called LDA samples (Lohan Dam).

1.3 Statement of Problem

Chemical and physical processes could alter the hazardous waste in a landfill system. One of the alteration or degradation products is leachate. The leachate contain heavy metal which are some of the most harmful wastes, hence will produce contamination into the environment. Abandoned and improperly managed landfill could be also the other sources of heavy metals contamination. In the mining waste containment the weathering of sulphide minerals such as pyrite and galena, which produce acid drainage is the main sources of the heavy metals. The migration of heavy metals from the hazardous waste or mining waste into the clay liner in landfill system occurred in an acidic environment. Therefore, two sources of contaminant are identified namely:

i. Contaminant from landfill area through clay liner material represented by KMT and KGB samples.
ii. Contaminant from mining waste containment represented by LDA sample.

The purpose of clay liner is to enhance environmental protection. They serve as a hydraulic barrier to the flow of contaminants. Therefore, they must have a low hydraulic conductivity for long periods of time. They act mainly to retain the contaminants. Understanding the leaching behaviour of heavy metals in the contaminated soil and mining waste is important in order to develop a method to restrain their mobility.

As a summary, the sources of heavy metals in the studied area were originated from the leaching of landfill leachate, and leaching from the mining waste. Therefore, understanding the leaching behaviour of heavy metals in the landfill liner and mining waste is important in order to develop a method to manage the contaminated soil and waste in this area. One of the possible solutions for the treatment of leachate generating hazardous waste or mining waste would be employing lime stabilisation. Clay liner could be stabilised with lime to increase its capacity to retain the heavy metals. Lime could react with clay and produce cementitious minerals, which crystallized with age hence increased the retention of heavy metals in the clay liner. In the long term, the development of cementitious minerals and clogging of fine materials could decreased the permeability of the clay liner, therefore increase the capability of the soil to protect the migration of heavy metals from degrading the environment.

1.4 Objectives

The objectives of this study are:

i. To determine the chemical properties, physical properties, mineralogical identification, microstructural, and geotechnical properties of KMT, KGB, and LDA samples.

ii. To establish the effect of lime on KMT, KGB, and LDA samples properties as follows:

   a. Chemical properties: pH, cation exchange capacity (CEC), specific surface area (SSA), and bulk chemistry.
   b. Physical properties: Atterberg limits specific gravity (SG), and particle size distribution.
   c. Mineralogical identification for both of clayey soil and mining waste using X-ray diffraction (XRD), and scanning electron microscope (SEM).
   d. Geotechnical properties such as compaction, unconfined compressive strength (UCS) and permeability.

iii. To examine the effect of lime on leaching process of heavy metals in KMT, KGB, and LDA samples.

iv. To produce an application design for controlling contamination from landfills systems, and mining waste.

1.5 Scope of the Study

i. All of the samples were collected from Sabah i.e soil from Kg. Kayu Madang Telipok (KMT), soil from Kg. Bongkud, Ranau (KGB) and mining waste from Lohan Dam, Ranau (LDA).

ii. Hydrated lime [$Ca(OH)_2$] was used as a major stabilisation agent.

iii. Supplement pozzolana for the mining waste was collected from Kg. Melawa, Sepanggar, Kota Kinabalu (FM).

iv. Laboratory works include of physical, chemical, mineralogical, microstructural and, geotechnical properties of the material before leaching and after leaching for both stabilised and unstabilises condition.

v. Leaching column tests were conducted mostly until 7 pore volumes (PV) of contaminant in order to assess the suitability of lime as a stabilising agent and immobiliser for the selected heavy metals in KMT, KGB, and LDA samples.

vi. The category of contaminant comprises of selected heavy metals (chromium, copper, nickel, lead and zinc) are manually spiked into the leaching columns to represent the contaminant from the landfill site, and mining waste.

1.6 Significance of Research

This study is important for the geotechnical engineers or land development agencies to plan any construction involving landfill site, contaminated soils and mining waste containment. Special measurements, treatment and design procedures are important and need to be fully understood. Huge distribution of soil from the weathering of sedimentary rock (Crocker Formation) in Kota Kinabalu area, the occurrence of contaminated soil in Kg. Bongkud area and, million of tonnes of mining waste containment in almost 1000 acres have contributed to a significant effect on land use activity as well as the health of the people within the vicinity. Therefore, it is vital to study the suitability and effect of lime to stabilise the heavy metals in mining waste and contaminated soil.

1.7 Thesis Organisation

This thesis consists of eight chapters, and the contents of each chapter is explained as follows:

CHAPTER I: This introductory chapter presents the current issues of environment, which relates to the statement problem in the study area. It also explains the scope and the significance of this research.

CHAPTER II: This chapter reviews the source of heavy metals in soil and their concentration in contaminated soil. The remediation of contaminated soil will be discussed. The lime stabilisation will also be discussed in detail, where it consists of the mineralogy of lime-soil reactions, microstructural development, physical characteristics and engineering properties.

CHAPTER III: Chapter III presents the sampling site of KMT, KGB, and LDA samples in Sabah. In addition to the description of the properties this chapter also will highlight the general test procedures of the physical and chemical test and also the concept of SSA, XRD, XRF, and SEM studies.

CHAPTER IV: Chapter IV explained the effect of lime on KMT, KGB, and LDA samples properties. The effect of lime on the physical characteristics consists of Attterberg limits, specific gravity, and particle size distributions. The analysis of chemical properties includes, cation exchange capacity, pH, specific surface area, and X-ray fluorescence will analysis for the chemical properties. Finally, the geotechnical tests such as compaction, unconfined compressive strength and, permeability will be presented.

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