STABILISATION AND SOLIDIFICATION OF CONTAMINATED SOIL USING CEMENT AND SUGARCANE BAGASSE ASH (SCBA)

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Dedicated to my beloved father and my late mother,
Hamidah Abdul Aziz, May Allah (SWT) forgive all her sins and
May He make Jannatul Firdaus to be her final abode
(Al-Fatihah)

And

Beloved family Kakak, Abangtek, Abang Nazreeq, Sara,
Teachers right from chilhood up to now and friends

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ABSTRACT

Soil that is contaminated with heavy metals has become a major issue worldwide. However, proper remediation techniques such as stabilisation/solidification (S/S) method can be employed and is capable of controlling these heavy metals. Conventionally, the common S/S method used cement as binder on remediating the contaminated soil particularly heavy metals. This research is to investigate the effect of physical and leachability of contaminated soil in S/S method when Sugarcane Bagasse Ash (SCBA) is added to remedy contaminated soil. Landfill contaminated soil was used to test the effectiveness of those binder. Cement was added at a proportion of 5%, 10%, 15% and 20% in sample weights without SCBA while in another sample; the cement was replaced by SCBA at a proportion of 2.5%, 5%, 7.5% and 10%. All samples are to be allowed to harden and cured at room temperature for 7, 14 and 28 days. The effectiveness of the treatment was assessing by conducting physical testing such as Unconfined Compression Strength, Water Absorption and Permeability test. In addition, leaching tests were performed to identify the leachate behavior of heavy metals during treatment. Three leaching tests were conducted and they were the Toxicity Characteristic Leaching Procedure (TCLP), Synthetic Precipitation Leaching Procedure (SPLP) and Dynamic Leaching Test (DLT). Through the physical testing, samples containing 10% OPC mixed with 10% SCBA were found to improve the compressive strength, reduced the water absorption and water permeability measuring 1550 MPa, 17.94% and 4.41×10^{-10} m/s respectively. In the same way, through the statistical analysis, the R-squared for UCS with respect to mixed design is high at 98%. However, the value for both water absorption and permeability recorded to be marginally low, compared to the value for strength at 89% and 88% respectively. Through the TCLP and SPLP test, results indicated that when SCBA added to OPC content in soil samples, less heavy metal been leached out from the S/S sample. In average, the satisfying result was shown by samples containing 10% OPC + 10% SCBA where reduction of heavy metals in final leachate is more than 90% for As, Cd, Cr, Pb and Zn. Through the Dynamic Leaching Test, sample containing 10% OPC +10% SCBA showed the satisfactory leachability index (*Lx*) at 9.17, 9.17, 8.81, 8.17 and 6.97 for As, Cd, Cr, Pb and Zn respectively. This indicates that the use of cement and SCBA as a binder was successful in remediating the contaminated soils through the S/S method.

ABSTRAK

Tanah yang dicemari dengan logam berat merupakan isu utama di seluruh dunia. Walau bagaimanapun, teknik rawatan tanah yang betul seperti teknik penstabilan/pemejalan (P/P) dilihat mampu mengawal pencemaran tanah. Objektif utama kajian ini adalah untuk mengkaji kesan penambahan abu hampas tebu terhadap kekuatan dan larut resap logam berat dari tanah yang distabilkan mengunakan teknik (P/P). Tanah tercemar dari kawasan tapak pelupusan sampah digunakan bagi mengkaji keberkesanan bahan pengikat tersebut. Simen ditambah pada kadar 5%, 10%, 15% and 20% manakala abu hampas tebu diganti sebahagian dari peratusan simen pada kadar 2.5%, 5%, 7.5% and 10%. Kesemua sampel dibiar mengeras dan diawet pada suhu bilik selama 7, 14 dan 28 hari. Keberkesanan rawatan dinilai melalui ujian fizikal seperti ujian mampatan tak terkurung, ujian penyerapan air dan ujian kebolehtelapan air. Selain itu, ujian pengurasan juga dijalankan bagi mengenalpasti kriteria larut resap logam berat semasa rawatan. Tiga ujian pengurasan yang telah dijalankan iaitu Prosedur Pengurasan Ciri Ketoksikan (PPCK), Prosedur Pengurasan Hujan Tiruan (PPHT) dan dan Ujian Pengurasan Dinamik (UPD). Melalui ujian fizikal, campuran sampel yang mengandungi 10% OPC dengan 10% abu hampas tebu menunjukkan peningkatan ketara terhadap kekuatan mampatan, mengurangkan peratusan kadar penyerapan air serta kebolehtelapan air dengan nilai masing-masing sebanyak 1550 kPa, 17.94% and 4.41 x 10^{-10} m/s. Pada masa yang sama, melalui kajian statistik, nilai R^2 bagai kekuatan mampatan adalah tinggi sebanyak 98%. Walau bagaimanapun, nilai R^2 bagi penyerapan air dan kebolehtelapan menunjukkan purata nilai yang rendah dengan masing-masing 89% dan 88%. Melalui ujian PPCK dan PPHT, keputusan menunjukkan pengurangan logam berat di dalam larut resap dengan penambahan abu hampas tebu terhadap OPC di dalam sampel tanah. Secara purata, keputusan yang memuaskan telah ditunjukkan oleh sampel yang mengandungi 10% OPC +10% abu hampas tebu dengan pengurangan kepekatan logam berat melebihi 90% bagi As, Cd, Cr, Pb dan Zn. Melalui Ujian Pengurasan Dinamik, sampel yang mengadungi 10% OPC + 10% abu hampas tebu menujukkan keputusan yang memuaskan dengan indek pengurasan (*Lx*) sebanyak 9.17, 9.17, 8.81, 8.17 dan 6.97 masing-masing bagi As, Cd, Cr, Pb dan Zn. Ini menunjukkan penggunaan simen dan abu hampas tebu sebagai pengikat berjaya merawat tanah tercemar melalui kaedah rawatan P/P.

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LIST OF SYMBOL AND ABBREVIATION

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

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

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CHAPTER 1

INTRODUCTION

1.1 Research Background

Soil is a basic environmental element constituting the ecosystem. It is also an important material for the survival and development of human beings. With advancements in industrialisation and urbanisation, the environmental safety of soil has deteriorated in developing countries due to industrialisation and urbanisation. These factors have contributed to the problem of unsustainable soil contamination. The main factor which contributes to soil contamination is man-made waste. Generally, waste produced naturally such as dead plants, carcasses of animals and rotten fruits and vegetables will contribute to soil fertility (Agamuthu *et al.*, 2013). However, waste generated from human activities are full of chemicals that that can lead to soil contamination.

Debates have been revolving around industrialisation and its association with environmental issues such as industrial activity, agricultural activities, waste disposal, accidental oil spills and acid rain (Napia, 2012). Industrial activity has been the biggest contributor to environmental issues in the last century, especially with the increasing mining and manufacturing activities. Most industries are dependent on extracting minerals from the earth. Whether it is iron ore or coal, the byproducts are often contaminated and not disposed off in a manner that can be considered safe (Li *et al.,* 2017). As a result, industrial waste lingers on the soil surface for a long time and makes it unsuitable for use.

Subsequently, agricultural activities have also contributed to soil contamination. Chemical utilisation has gone up tremendously since the invention of modern pesticides and fertilisers (Li *et al.,* 2012). The chemicals from pesticides and chemicals normally remain in soil even after extended periods of time. The contaminants seep into the ground along with water and this gradually reduces the fertility of soil. These chemical products affects the composition of soil and makes it easier to be eroded by water and air. As a result, plants will absorb many of these pesticides. When decomposition of the plants occurs, it contributes to soil contamination (Kamari *et al.,* 2011).

On the other hand, soil contamination can also be caused accidental oil leakages that normally occur during storage and transport of chemicals. This significant problem can be seen at most fuel stations. The chemicals present in fuel will cause the quality of soil to deteriorate, making them unsuitable for cultivation. These chemicals may enter groundwater through soil and make it undrinkable (Halim *et. al.,* 2005). Moreover, acidic rain has been identified as one of the major factors that contribute to soil contamination. It is caused when pollutants present in the air mixes up with the rain and fall back on the ground. Acidic rain could dissolve some of the important nutrients found in soil and change the structure of soil (Covelo *et al.,* 2007a; Covelo *et al.,* 2007b).

In order to solve problems related to soil contamination, particularly by heavy metals, soil remediation is very much needed. The EU and the UK legislation have recently encouraged the use of remediation techniques in order to ensure human safety (Harbottle *et al.,* 2007). Contaminated soils can be remediated through leaching, venting or vapour extraction, microbial decomposition, composting, vegetative uptake, removal and stabilisation/solidification using binders (Fauziah *et al.,* 2013). Leaching of soluble contaminants is done by flushing soil with water and safely draining away the diluted leachate. Microbial decomposition is carried out by organisms in the soil that are capable of decomposing organic contaminants by rendering them harmless. Microbial activity can often be stimulated by adding nutrients, aerating the soil if is waterlogged, or irrigating the soil if it is dry (Liu *et al.,* 2013).

Soil composting consists of mixing contaminated soil with an admixture of readily decomposable organic matter to stimulate microbial activity and placing the mixture in piles to remediate contaminated soil (Dermont *et al.,* 2008). The vapour extraction technique is normally applied to soils contaminated by volatile organic chemicals such as trichloroethylene from spilled solvent or benzene from petroleum storage tanks. The soil to be purified is placed on an impervious surface and covered with an impervious cover (generally plastic sheeting). Then, the air is drawn through the soil via perforated pipes and finally vents the soil to the atmosphere or to a carbon trap (Dermont *et al.*, 2008).

The phytoremediation technique utilises plants to absorb and remove contaminants from soil. Plants used in this particular technique tend to concentrate a specific element such as heavy metals, and allow its removal and safe disposal at the time of harvest (Oosten & Maggio, 2014). Alternatively, a common practice in the effort to remediate soils charged with inorganic pollutants is to apply a binder to counter soil acidity and suppress the solubility of the contaminants through the stabilisation/solidification method.

Stabilisation/Solidification (S/S) is a term used to describe the technology that involves mixing contaminated medium and binding reagents to reduce hazardous substances into non-hazardous substances which are environmentally acceptable for current land disposal (Kumpiene *et al.,* 2008). Even though stabilisation and solidification are similar terms, the effect of the binding reagent on waste is different. Stabilisation refers to a process that reduces the chemical reaction by converting waste into a less hazardous substance. Meanwhile, solidification is a more specific process that treats material to increase its solidity and structural integrity (Erdem & Ozverdi, 2011). Additionally, solidification does not remove nor degrade contaminants, but prevents or eliminates their mobility.

The S/S method mainly consists of mixing contaminated material with suitable stabilisers. Lime, cement, and other cementitious industrial waste materials are commonly used in S/S treatments. Among the types of binders mentioned, cement-based systems are the most widely used due to its relatively low cost, wide availability and versatility (Gollmann *et al.,* 2010). However, the manufacture of cement often leads to environmental pollution. The CO_2 emitted from the manufacturing process has a major influence on climate change due to the greenhouse effect. At present, the use of cement is slowly being replaced by renewable binders such as agricultural byproducts which are more sustainable, cost effective and have the potential to improve the leaching characteristics of contaminated soils. In addition, the need for safe and environmental friendly methods for the elimination of heavy metals from contaminated soil has necessitated research on agricultural waste byproducts such as sugarcane bagasse ash, rice husk ash, sawdust, coconut husk ash, oil palm shells and so on.

The utilisation of agricultural byproducts in the production of cement-bonded materials offers an attractive alternative. Hence, in this research, sugarcane bagasse ash (SCBA) has been investigated for its suitability as a cement replacement in the S/S remediation method. The usage of sugarcane bagasse ash (SCBA) may potentially help to solve disposal problems and provide a cost-effective cement replacement material. On the other hand, sugarcane production was recorded at 1.8 billion tonnes in 2012 and is expected to increase every year. Malaysia possesses nearly 37,000 acres of sugarcane plantations. Therefore, it is fairly easy to collect sugarcane bagasse with the establishment of sugarcane collection centres. For instance, the Federal Agriculture Marketing Authority (FAMA) in Malaysia has set up a Sugarcane Collection Center or Pusat Pengumpulan Tebu (PPT) in Batu Pahat, Johor, for export purposes. Therefore, the use of agricultural wastes particularly SCBA would help solve disposal problems of agricultural waste and provide a sustainable cement replacement material.

1.2 Problem Statement

The use of the stabilisation/solidification (S/S) remediation method to treat polluted soil where cement is employed as a binder has existed for decades (Gonzalez *et al.*, 2012). Nevertheless, there are a number of challenges associated with this method where 5% to 8% of global CO_2 emission to the atmosphere is caused by the production of cement. In the same way, most contaminants like Cd, Cr, As, Pb and Zn will interfere with the hydration of cement during the remediation process (Spence & Shi, 2004). This is because cement will undergo the hardening process at a pH value of over 12.5. The optimum pH range for precipitating amphoteric metals is about 10 and the use of cement alone cannot help control the oxidation state of metals. However, these disadvantages can be solved by adding various additives such as agricultural waste into cement to reduce the high pH value. The use of agricultural waste helps to alleviate serious issues such as handling and disposal processes after their end-production. Due to the difficulty in disposing huge quantities of agricultural waste, researchers all over the world have attempted to investigate the potential uses of agricultural waste such as sugarcane bagasse ash (SCBA). As a pozzolanic material, SCBA is increasingly being tested and applied in the construction industry and the mix design of concrete. In spite of the increasing interest in the potential uses of SCBA in concrete, its use as a soil stabiliser to treat heavy metal contaminated soil has yet to be explored in the current literature. Therefore, this study was conducted to investigate the potential of SCBA as a cement replacement in the remediation of heavy metal contaminated soil. This significant study is also expected to provide better information for future researchers to conduct research in the same field. Furthermore, a comprehensive study in the laboratory will help to improve the quality of soil remediation.

1.3 Research Objectives

The aim of this study is to evaluate the performance of sugarcane bagasse ash (SCBA) as a partial replacement of cement (OPC). The research objectives to be achieved in this study are:

1. To determine the physical and chemical properties of contaminated soil, ordinary Portland cement (OPC) and bagasse ash (BA).
2. To examine the leaching behaviour of heavy metals from the contaminated soil stabilised and solidified by cement (OPC) and bagasse ash (BA) using the Toxicity Characteristic Leaching Procedure (TCLP), Synthetic Precipitation Leaching Procedure (SPLP) and Dynamic Leaching Test (DLT).
3. To evaluate the effects of cement (OPC) and bagasse ash (BA) addition on the physical characteristics of S/S samples.
4. To model and optimise the parameters over physical characteristic responses by applying the Response Surface Methodology (RSM).

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