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UTILIZATION OF SUGARCANE BAGASSE ASH FOR STABILIZATION / SOLIDIFICATION OF LEAD-CONTAMINATED SOILS

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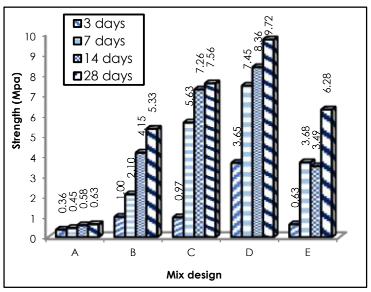
Graphical abstract



Abstract

Recently, many researchers are interested in using agricultural waste as an additive to remediate the contaminated soils. In this study, the effectiveness of sugarcane bagasse ash (SCBA) as the substitution binder to Ordinary Portlant Cement (OPC) content in Stabilization/Solidification (S/S) method was investigated through the physical and chemical characteristics namely the Unconfined Compressive Strength (UCS) and Toxicity Characteristic Leaching Procedure (TCLP). Synthetic contaminated soil was prepared in bulk by mixing soil samples with lead nitrate to achieve the concentration of 500 ppm. The OPC and SCBA varying from 5 % to 20 % were added to stabilize and solidify the contaminated soils. The cylindrical specimens (D = 38 mm, H = 76 mm), was compacted in five layers with 50 blows each. A further 3, 7, 14 and 28 days were allowed for curing in the temperature 25±2 ◦C and humidity > 80%. Results indicate that all samples containing OPC and SCBA satisfy the US EPA strength requirement of 0.35 MPa for S/S sample. The TCLP testing shows that sample containing OPC with SCBA has been successful treated which produced the leachability below US EPA limit for lead of 5 mg/L. In conclusion, the use of SCBA as part of replacement of OPC has been successful in increasing the strength and reducing the leachability compared to untreated sample.

Keywords: Ordinary Portland cement, sugarcane bagasse ash, lead

Abstrak

Kebelakangan ini, ramai penyelidik berminat untuk menggunakan sisa pertanian sebagai bahan tambahan bagi merawat tanah yang tercemar. Dalam kajian ini, keberkesanan Abu Hampas Tebu (SCBA) sebagai pengikat bagi menggantikan kandungan Simen Portland Biasa (OPC) dalam campuran Penstabilan/Pemejalan (S/S) ke atas sampel yang dirawat telah disiasat melalui ciri-ciri fizikal dan kimia seperti Ujian Kekuatan Mampatan Tak Terkurung (UCS) dan Prosedur Pengurasan Ciri Ketoksikan (TCLP). Tanah sintetik yang tercemar telah disediakan secara pukal dengan mencampurkan sampel tanah dengan plumbum nitrat, untuk mencapai kepekatan 500 ppm. OPC dan SCBA dengan nisbah antara 5 % kepada 20 % telah ditambah untuk menstabil dan mengukuhkan tanah yang tercemar. Spesimen silinder (D = 38 mm, H = 76 mm) telah dipadatkan sebanyak lima lapisan dengan 50 pukulan setiap satu. Sebelum ujian dijalankan, spesimen dibiarkan selama 3, 7, 14 dan 28 hari untuk mengeras pada suhu 25 ± 2 ◦C dan kelembapan> 80%. Keputusan menunjukkan bahawa semua sampel yang mengandungi OPC dan SCBA memenuhi kekuatan yang ditentukan US EPA iaitu 0.35 MPa. Ujian TCCP menunjukkan sampel yang mengandungi OPC dan SCBA berjaya dirawat iaitu menghasilkan larut resap di bawah had US EPA dengan kepekatan 5 mg/L. Secara kesimpulannya, penggunaan SCBA sebagai bahan pengganti separa kepada kandungan OPC berjaya meningkatkan kekuatan tanah serta mengurangkan larut resap berbanding specimen yang tidak dirawat.

Kata kunci: Simen Portland biasa, abu hampas tebu, plumbum

1.0 INTRODUCTION

Soil is a basic environmental element that constitutes the ecosystem and is an important basic material for the survival and development for human beings [1]. Thus, this medium is considered a highly potential medium that is easily exposed to contamination. John *et al.* [2], describes land contamination as areas with high concentration (above normal background level) of substance, which may have arised from previous land use.

Soil contaminations especially by heavy metals lead to environmental and human health issues that need effective and affordable solutions. The main causes for these problems are from a large number of industrial activities which produce wastes and contaminants that reach the soil through direct disposal, emissions and other pathways [3]. Moreover, soil properties are affected by previous land use as well as current activities on lands. Human activities have intentionally added substances such as pesticides, fertilizers and other amendments to soil [4]. Additionally, leaks of chemicals used for commercial or industrial purposes and accidental spills are also sources of contamination.

Furthermore, improper waste disposal and mismanagement of soil is the main contributors to environmental pollution [5]. In Asia, sustainable waste management are being practiced especially in developed countries such as Korea and Japan, while in developing countries such as India, Indonesia and Malaysia, these issues seem hard to be solved. In Malaysia, the current municipal solid waste (MSW) generation has exceeded 19,000 tonnes daily. With the lack in waste recycling has made the matters worse [6].

Ordinary Portland cement (OPC) is the most used binder to immobilize the heavy metals in soils as a remediation technology named as the Stabilization/ Solidification (S/S) method [7]. The U.S Environmental Protection Agency (EPA) has identified the S/S method as the Best Demonstrated Available Technology for many hazardous wastes.

Towards sustainablity, there is much interest using other binders to be partial OPC replacement as cement has been pointed as one of the major contributors of anthropogenic CO_2 emissions towards the environment during its productions (about 5 % globally). In the recent years, researcher has often substituted portion of OPC in Stabilization/Solidification S/S method with agricultural wastes such as incinerator bottom ash and coal fly ash [8]. Concrete technologies are beginning to find application using sugarcane bagasse ash (SCBA) as partial substitute of cement to improve the properties of cement mortar, either in the mechanical or physical properties [9], improve the normal concrete [10], self-compacting concrete [11] and to improve the durability of concrete under aggressive environment [12]. In general, Bagasse ash is the waste generated by the combustion of sugarcane bagasse. Apart from silica which is the major component, bagasse ash contains other oxides as well as unburned carbon [13]. Large amounts of bagasse ash have been produced annually in developing countries but there is very limited used in the remediation of of contaminated soil. These significant problems lead to the disposal of bagasse ash to the landfills and causing environmental problems such as soil pollution. Therefore, a study has been conducted to investigate the effectiveness of SCBA as a partial replacement to OPC content in S/S method of lead-contaminated soil.

1.1 Impact of Contaminants in Soil Composition

Contaminants occur naturally in soil, the amount of the substances may exceed the allowable limit for the health of humans, animals and plants. Some organic contaminants can undergo chemical modification or degrade into substances that may be more or less toxic than the original compound [14]. In the same way, chemical elements such as metals cannot be destroyed but the characteristics will convert either more or less easily to be taken up by plants or animals [15]. In addition, wide range of contaminants varies in their tendency to end up in water held in soil or in the underlying groundwater from leaching through the soil. There are certain characteristics of the soil that shows it has been affected by contaminants [16]. The important characteristics that may be affected by contaminants include soil mineralogy and clay content (soil texture), pH (acidity) of the soil, amount of organic matter in the soil, moisture levels, temperature and presence of other chemical.

Heavy metal contaminated soil is a worldwide problem that urgently needs to be solved. On the other hand, heavy metal contaminated soil is mainly due to the subsequent migration of leachate form and within the waste landfill cells. According to Agamuthu *et al.* [6], the natural processes such as infiltration within the boundaries of the waste cells can accelerate the process of heavy metal leaching from the waste components that are sources of heavy metals within the landfill system. The release of heavy metals into the adjacent environment is a serious environmental concern and a threat to public health and safety. Kamari *et al.* [17], stated that the processes of metal released from the solid phase into soil solution were effected by their presence in elevated concentrations and biological behaviour.

1.2 Toxicity of Heavy Metals

Heavy metals such as Lead (Pb), Cadmium (Cd), Mercury (Hg) and Arsenic (As) can threaten people's health either accidental soil ingestion or from polluted drinking water [18]. In small quantities, there are some heavy metals that nutritionally essential for human body for healthy purposes. However, it can become dangerous when the heavy metals are not digested by the body and accumulate in the soft tissues. In the same way, heavy metals may be absorbed into the human body through various mediums including absorption through the skin, food, water and air [18].

Lead is one of the heavy metals that have both advantage and disadvantages behaviour generated from many industrial processes such as an unnatural lead-cycle, application in gasoline and extensively used as a constituent of the lead-acid battery in car batteries. Additionally, lead becomes an important metal for pipes industry and widely used as electrodes in the electrolysis process. In the same way, lead has been used as a coloring materials for ceramic glazes, as projectiles and in some candles to treat the wick [19]. Due to its malleability, lead is considered as one of the oldest metals used by humans for different purposes like in the manufacture of lead-acid storage batteries, alloys, plumbing, cable covering, heavy machinery and recently, it is used as an important source in gasoline combustion [20].

In the environment, lead accumulates and remains in the soil through the activities carried out by humans such as mining and smelting causing environmental problems such as soil erosion [21]. The adverse effects such as reproductive level in plants and animals, changes in community composition, losses in biodiversity and neurological effects in vertebrates, stem from lead sources near the ecosystem.

1.3 Lead in Environment

Lead is persistent in the environment and accumulates in soils and sediments through deposition from air sources, direct discharge of waste streams to water bodies, mining, and erosion [22]. Ecosystems that are near point sources of lead demonstrate a wide range of adverse effects including losses in biodiversity, changes in community composition, decreased growth and reproductive rates in plants and animals, and neurological effects in vertebrates.

Lead is found in ore with zinc, silver and copper and has been extracted together. According to Yan et. al., [23], lead occurs naturally in the environment. However, most of the lead concentration that is found in the environment is the result of human activities such as application of lead in gasoline and an unnatural lead-cycle. Furthermore, in car engines, the lead that was burned, produces lead oxide. This oxide will enter the environment from the car exhaust [24]. The process suggests that the largest particles will drop to the ground immediately and pollute the soil or water surface, while the smaller particles will travel long distance through air and remain in the atmosphere. Finally, parts of this lead, either small or large particles, will return back to earth when it rains. Furthermore, this cycle caused by human activities and production is considered more detrimental than the natural lead cycle and becomes the biggest worldwide issue [25]. Kamari et. al., [26] studied lead contamination in mango, guava and papaya grown on ex-mining land in Malaysia and found that the concentration of lead in the fruits exceeded the Malaysian Food Act permissible limits.

1.4 Lead Impact to the Human Health

Humans may be exposed to lead and chemicals that contain lead via air, drinking water and food. Once taken into the body, lead distributes throughout the body through the blood and is accumulated in the bones [25]. Depending on the level of exposure, lead can adversely affect the nervous system, kidney function, immune system, reproductive and developmental systems and the cardiovascular system. Lead exposure also affects the oxygen carrying capacity of the blood. Yap et al. [27] mentioned the lead effects that are most commonly encountered in current populations are neurological effects in children and cardiovascular effects such as high blood pressure and heart disease in adults. Infants and young children are especially sensitive to even low levels of lead, which may contribute to behavioral problems, learning deficits and lowered IQ.

Moreover, lead is distributed to many tissues and organ systems of the body. It is important to note that lead cannot be destroyed or changed to something else in the body. The amount of lead stored in the body has been described as a "body burden" by lead. Among adults, over 95% of lead is stored in bones. Meanwhile, for children, about 70% of lead is stored in bones [28]. This lead is not simply stored away in bones forever, but moves in and out as the body functions normally. For example, as children grow their bones restructure to permit normal shapes as they develop.

1.5 Type of Available Remediation Technique

According to Yao et al. [29], there are three types of remediation techniques that are suitable for heavy metal contaminated soil and they are physical remediation, chemical remediation and biological remediation. Physical remediation mainly includes soil replacement method and thermal desorption [30]. Soil replacement means using clean soil to replace or partly replace the contaminated soil with the aim of diluting the concentration of pollutants, increase the soil environment capacity and thus, remediate the soil. While, Shi et. al., [31] stated that thermal desorption is based on pollutant volatility where and the contaminated soil is heated using steam, microwave, or infrared radiation until the pollutant is volatile. The volatile heavy metals are then collected using vacuum with negative pressure to remove the heavy metals.

Chemical remediation is divided into 3 types which are chemical leaching, chemical fixation and

Electrokinetic remediation. Firstly, chemical leaching is a process of washing the contaminated soil using fresh water, reagents and other fluids or gases that can leach the pollutant from the soil. According to Khan et. al., [32], heavy metals in soil are transferred from soil to liquid phase through ions exchange, precipitation and adsorption process in chemical leaching remediation. Secondly, chemical fixation is a process of adding reagents or materials into the contaminated soil and using them with heavy metals to form insoluble or hardly movable, low toxic matters, thus decreasing the migration of heavy metals into water, plants and other environmental media and achieving the remediation of soil [29].

Besides that, the electrokinetic remediation technique or EK is a new remediation technique which is mainly applying voltage at the two sides of the soil and then forming electric field gradient. According to Syakeera et al. [33], basically, this technique is to improve the volume stability of the soil around and beneath the foundation. This technique involves applying an electrical current across the soil mass to boost the chemical migration from the injection point with the purpose of reacting beneficially with the soil to bring about an improvement in its properties.

Another promising technology in soil remediation is biological remediation. The biological remediation is a process of changing the physical and chemical characteristics through migration and transformation process of heavy metals by microorganisms [34]. The remediation mechanisms include extracellular complexation, oxidation-reduction and intracellular accumulation. Additionally, the microbial leaching by microorganism is a simple and effective technology for extracting valuable metals from low-grade ores and mineral concentrates. In the same way, Yao et al. [29] argued that the microbial leaching has some potential in remediation of mining sites, treatment of mineral industrial waste products, detoxification of sewage sludge and for remediation of soils and sediments contaminated with heavy metals.

Besides that, phytoremediation is also a part of biological remediation. According to Oosten & Maggio [35], phytoremediation is a remediation technique that uses living green plants to fix or absorb and clean the contaminants or reduce the risk provide by heavy metals. The phytostabilization, phytovolatilization and phytoextraction are the main three types of phytoremediation [35]. Phytostabilization is referred to a fixing of metals using plants through adsorption, precipitation and reduction of roots, and thus reducing their migration into the groundwater and food chain. Despite of phytostabilization, phytovolatilization involves transferring heavy metals into a volatile state or adsorption of the metals in gaseous matter by using special agents secreted by the roots of the plants. While, phytoextraction involves adsorption of the heavy metals using tolerant and accumulating plants by transferring and storing at ground parts.

Among the methodsmentioned, the physical method particularly S/S method has been utilized effectively, and is extensively used in developed countries for the past decade for treatment of heavy metal wastes and contaminated soils [36].Therefore in this research, the agricultural waste of sugarcane bagasse ash has been investigated for its suitability to replace cement in S/S method.

2.0 MATERIALS AND METHODS

2.1 Materials

The natural clay was collected at the Research Centre for Soft Clay (RECESS), Universiti Tun Hussein Onn Malaysia (UTHM). The soil were placed in polystyrene containers and taken to the laboratory to be dried in the oven at about 105°C for 24 hours. After drying for 24 hours, the soil was crushed using a rubber hammer before being decimated into 2 mm in size using a grinder machine. The soil which passes the 2 mm sieve size was stored in polyethylene plastic. Remediation of lead-contaminated soil was conducted using OPC in accordance to MS 522 (1989) specification. The SCBA was used as a substitute material to cement content by various ratio as presented in Table 1.

Table 1 Sample mix proportions (by weight)

Binder			Total mass of the sample (g)	w/b*	Symbol
Clay soil (%)	OPC (%)	SCBA (%)			
100	-	-	160	0.4	A
80	20	0	160	0.4	B
80	10	10	160	0.4	C
80	5	15	160	0.4	D
80	15	5	160	0.4	E

*water/solid (w/b)

2.2 Sample Preparation

The soil samples were prepared by mixing the clay soil with a precise quantity of Pb in order to imitate the real contaminated soil [23]. The Pb was generated from the lead nitrates ($Pb(NO_3)_2$) at the target concentration of 500 ppm. This high concentration is intentionally used to measure the effectiveness of sugarcane bagasse as a partial additive to cement. On the other hand, the raw form of sugarcane bagasse has been dried under natural environment before being dried in oven at the temperature of about 105°C for 24 hours. The sugarcane bagasse was then burned in a furnace at temperature 700°C for 4-8 hours to produce sugarcane bagasse ash, called as activation of carbon as shown in Figure 1. The purpose of activation process is to enhance the pore surface area to absorb more contaminants. After that, the SCBA was cooled in the room temperature before being ground to 20 µm in size.

2.3 Testing Procedure

The Unconfined Compressive Strength (UCS) test and Toxicity Characteristic Leaching Procedure (TCLP) has been conducted to determine the strength and leachability of S/S sample respectively. The UCS test was carried out using the GeocompLoadTrac II system available in RECESS, UTHM. Testing was conducted according to BS1377-7:1990. The S/S sample was compacted in a mould with 38 mm in diameter and 76 mm in height. A specially designed miniature hand compacting tool was used to compact the mixture into five layers with 50 blows each. The extruded specimens were wrapped and stored for 7, 14 and 28 days prior to testing and before being passed into UCS tests. On the other hands, TCLP testing were conducted by adding 100g of the solidified/stabilized sample, crushed prior to the leaching test to pass a 9 mm sieve. The extractions were carried out using 20:1 ratio of liquid to solid in an acetic acid solution at pH 2.88 ± 0.05 depending on the sample pH in 500 mL bottles, rotated using a rotary agitating machine at 30 rpm for 18 ± 2 hours, as shown in Figure 2. After the sample was extracted in the acetic fluid, the solid and liquid phases were separated by filtration using Grade GF/F 0.7 µm glass fiber filter paper. The sample was filtered within 2 hours period after extraction to ensure method accuracy [37]. This procedure differs from The U.S Environmental Protection Agency (EPA) for TCLP [11] method which is normally use 1:10 ratio. After filtration, the heavy metal concentrations in the final leachate were determined using Perkin-Elmer AA800 Atomic Absorption Spectrometer (AAS).

Figure 1 Sugarcane Bagasse burning process in the furnace

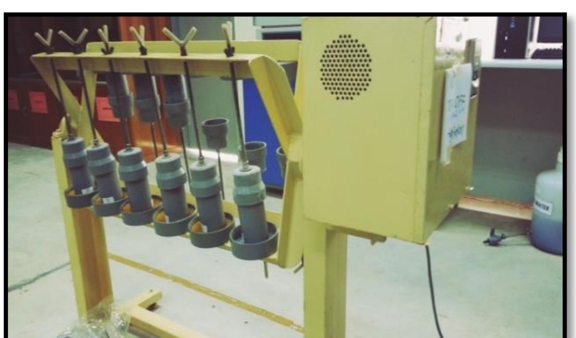


Figure 2 Leachate sample and rotary agitated machine

3.0 RESULTS AND DISCUSSIONS

3.1 Unconfined Compressive Strength

After the contaminated soil has been stabilized and solidified with OPC and SCBA, samples were subjected to UCS test. This research shows that by adding high percentage of SBA and OPC increased the strength up to 93% at 28 days of curing period. It was observed that UCS of all S/S samples exceeded the minimum US EPA limit for landfill disposal of 0.34 MPa. From Figure 3, the additional of SCBA into the sample exhibited the distinct effect on the UCS value. Sample D containing 5% OPC and 15% SCBA produces the highest UCS value with 9.72 MPa at 28 days of curing.

In the same ways, the UCS value of sample containing SCBA (sample B, C and E) also produced a satisfy UCS value with 5.33, 7.56 and 6.28 MPa respectively at 28 days of curing. Contradict to the sample B, C and D, sample A (control sample) produced the lowest UCS values with only 0.63 at 28 days of curing.This is expected as a presence of highest CaO in SCBA to sustain the strength development throughout the curing periods. However, the UCS value of control samples were found to be increased along the curing period even without additional of binders. This significant increment is assumed to be the results of physical changes induces by the loss of moisture content during the hydration periods.

In addition, the figure also indicated that the UCS values for each sample at 28 day are constantly higher than at 3 day of curing. This observation is also expected as hydration of the samples requires a certain periods to help water molecules permeate into pores of cementitious product to form C-S-H which is important for strength development in S/S method.

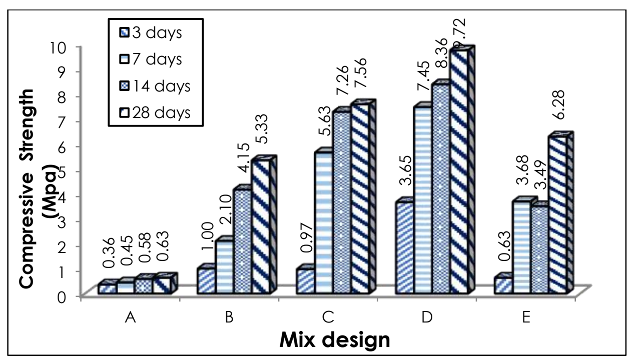


Figure 3 Strength development of S/S sample

3.2 Leachability

Figure 4 shows the leachability of Pb in TCLP extraction at different curing days. From the figure, there are great different between Pb concentration at 3, 7, 14 and 28 days. Concentration of Pb at 3 day is constantly higher than concentration at 28 days. This result clearly shows that hydration days are an important factor that leads to the leachability development. This scenario has been study by other researchers. As an example, Halim *et al.*

[38] mentioned that at 28 days of hydration, the S/S sample was in mature phase to stabilize and solidify the contaminants in soil.

Apart from that, Figure 4 shows a positive effect towards leachability of lead in S/S sample containing SCBA. The concentrations of lead were decreased from 500 mg/L (before remediation) to 0 mg/l (after remediation) at 28 days. This 100% reduction is showed by sample C, D and E that contain10% to 15% of SCBA. These significant results proved that the used of SBCA in soil remediation can be accepted as mentioned by Ghazali *et al.* [39] the appropriate modifications of SCBA in soil remediation shows some improvement of heavy metals leachability.

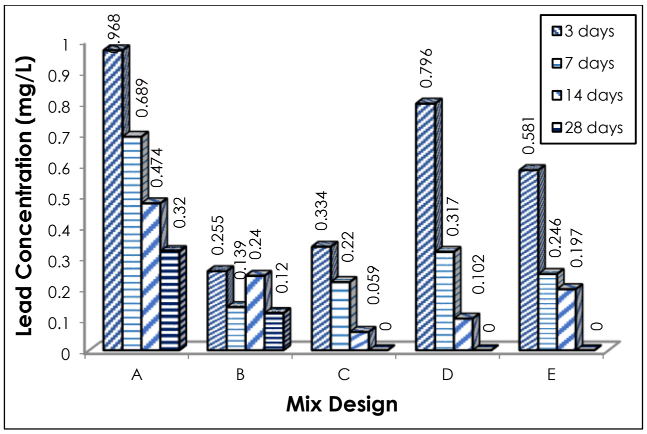


Figure 4 Leachability of S/S sample

4.0 CONCLUSION

From the study, it can be highlighted that the utilization of SCBA is possible partial replacement material of OPC content in S/S method to treat the contaminated soil, especially by heavy metals. Overall, the use of SCBA has successfully obtained the highest strength compared to the sample containing OPC alone. However, all samples are observed to exceed the landfill disposal limit of 340 kPa for waste except control samples that are slightly below the regulatory limit. On the other hands, TCLP extraction has successfully reduced the concentration of leadbelow the standard limit by US EPA where the concentration of lead in soil sample containing OPC and SCBA are constantly low. As a conclusion, sample containing lower percentage of OPC are required a longer curing times to obtain the desired strength. Apart from that, sample with 5% OPC and 15% SCBA has been identified as the optimum mix ratio in obtaining the desired strength as well as reducing the usage of OPC. In additional, the same mix design has leached out 0 mg/L lead or 100% lead reduction in final leachate. In conclusion, the partial replacement of OPC with SCBA has successfully in increased the strength and reduced the leachability compared to the untreated sample.

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