Leachate Characteristics of Contaminated Soil Containing Lead by Stabilisation/Solidification Technique.

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Abstract. Contaminated soil is often a problem and typical concern in the developing countries due to the lack of clean soil for development, such as in Malaysia. The objective of this study is to investigate the performance of the available techniques for soil remediation contaminated with lead (Pb) through Stabilisation/Solidification (S/S) technique. In this study, cockle shell powders at different percentages (2.5%, 5% and 7.5%) were added as a partial replacement of cement. Toxicity Characteristics Leaching Procedure (TCLP) was conducted to determine the effectiveness of the S/S technique to treat the contaminated soil. The results showed a 99% reduction of Pb concentration after the contaminated clay soil was treated with S/S technique. The combination of cement and cockle shell powders is very effectively used in the remediation of lead contaminated soil and reducing the amount of cement usage for sustainability approaches.

Introduction

The rapid growths of development somehow lead to the increasing of contamination in Malaysia. According to Azzahra [1], the rise of contamination rate, especially the soil contamination, has affected the equivalence between human and the ecosystem on earth. Due to the increasingly serious hazards caused by soil pollution, especially problems arising from heavy metals that act as contaminants, soil pollution remediation has recently attracted more attention.

Azmi et al. [2] stated that there are several techniques to remediate the contaminated soils such as phytoremediation, electrokinetics stabilisation and others. However, stabilisation/solidification (S/S) technique is normally employed because of its simplicity of usage and cost effectiveness. Furthermore, S/S technique is a widely used remediation technique characterised by a broad range of applications, a short process cycle and has been systematically studied in developing nations [3].

Heavy metals are widespread in urban/rural and industrial areas due to the industrial and agricultural activities such as smelting, metal mining, gasoline processing, as well as the application of fertilisers and agricultural chemical [4]. Heavy metals residue in soils can penetrate into the groundwater and get into the food chain through various activities. Inhalation and ingestion of heavy metal contaminated soils have been identified as a serious threat to human health, especially to the children's [5].

Contaminated soil with heavy metal pollution poses a risk to the environment and human health due to the rapid development [6]. Sources of pollutants into soil occur via many pathways such as dumping of solid and liquid wastes in ponds, mine waste and others. These factors can be dangerous for living organisms. Liquid substances in particular, can easily pollute large volume of soil, as they without doubt easily penetrate and disperse in soil [7]. Contaminated soils also have the potential to contaminate the ground water and surface water supplies through leachates that are produced from the heavy metal contaminated sites.

Leachates are the products of the mobilisation of soluble heavy metals, which can be transported to contaminated surfaces or groundwater. Therefore, it is essential to remove or reduce the presence of these inorganic contaminants (heavy metals) in order to diminish the possibility of uptakes by

living organisms and also to prevent them from contaminating surfaces and groundwater by dissolution or dispersion [8]. Due to this problem, the S/S technique was used to remediate the contaminated land. Previous research has been conducted on various additives such as cement, lime and fly ash [9].

In this study, cockle shell was used as an additive to reduce the concentration of contaminant in soil. Yao et al. [10] mentioned that bivalve shells are available in abundance and are commonly regarded as a waste. Bivalve shell has the potential to be used as raw material since it consists mainly of CaCO_3 with a small amount of the organic matrix. Recycling shell waste could potentially reduce the disposal problems, and also turn an otherwise useless waste into highly value-added products [10]. Therefore the performance of cockle shell in replacing cement by using S/S technique was investigated.

Materials and Methods

Materials and Samples Preparation The clay soil was collected at the RECESS, UTHM. The clay soil was first dried to remove free water; then it was ground and sieved with 2 mm of aperture size [11]. After that, the clay soil was spiked with lead to produce an artificial contaminated clay soil sample with a lead concentration of 2500 ppm. Therefore, 0.270 gram of PbO was diluted in 100 ml of pure water to form a dilution stock. The solution was stirred until fully soluble and mixed with the clay soil. In order to achieve a more homogeneous distribution of lead in the clay soil, the artificial contaminated clay soil sample was allowed to sit for two days before any solidification process were conducted [11]. Ordinary Portland Cement (Crocodile, Ms 522) and cockle shell powder were used as a binder to solidify/stabilise the contaminated clay soil. Approximately 250 gram of cockle shells was washed, scrubbed to remove dirt, boiled for 10 minutes and then cooled at room temperature. The shell were then washed thoroughly with distilled water and dried in an oven for 7 days at 50°C [12]. The cockle shells were finely ground using a grinder. The powders were sieved using a sieve with an aperture size of $90\text{ }\mu\text{m}$ to obtain micron-sized ($10\text{--}90\text{ }\mu\text{m}$ in diameter) powders [13].

All of the materials were mixed uniformly as possible using different percentages as shown in Table 1. Then, the mixture was compacted to achieve predetermined maximum density in a few cylindrical molds having a height of 100 mm and diameter of 50 mm, each. After the compacted process was done, the samples were removed from the molds, wrapped with plastic and placed in a 95% humidity room for 7, 14 and 28 days curing time. The samples were prepared according to Chang et. al [14], where the volume of the mold should consists of 40% of waste which is referred to the contaminated clay soil added with 35% of cement and 25% of water.

Table 1 Percentages of cement and cockle shell powders.

Samples	Percentages of Binders (%)
clay soil (contaminated) + cement	5% cement
	10% cement
	15% cement
clay soil (contaminated) + cement + cockle shell powders	2.5% cement + 2.5% cockle shell powders
	5% cement + 5% cockle shell powders
	7.5% cement + 7.5% cockle shell powders

X-ray Fluorescence Testing (XRF) XRF was used to perform elemental analysis of the pure clay soil before and after the clay soil was contaminated by lead. Both samples were ground to particle size less than $50\text{ }\mu\text{m}$. Then the samples were heated at 105°C for 2 hours. The samples then were fused with flux for 10 minutes at 1050°C to make sheet glass. After that, the sheet glass was put into an instrument (SRS3400) for quantitative XRF analysis [15].

Chemical Testing Leaching test was carried out after the samples were cured for 7, 14 and 28 days. The samples were crushed and ground to reduce the particle size to less than 2.0 mm and agitated in 20 mL of 5.7 mL acetic acid with reagent water of 1 L (1: 20 ratio) with a pH of 2.88 ± 0.005 for 18 hours at 30 rpm. The leachate was filtered through 0.45 μm membrane filter to remove suspended solid. The final extraction was analysed using atomic absorption spectroscopy (AAS) for lead concentration. TCLP test was commonly used in comparing the effectiveness of one S/S process with another. Therefore, this technique was employed in this study to quantify the relative effectiveness of stabilising lead contaminants by using cockle shell powders and cement binder.

Result and Discussion

X-ray Fluorescence Analysis (XRF) Comparison was made between the pure and artificial contaminated clay soil as shown in Table 2. The pure clay soil did not indicate any presence of Pb element while artificial contaminated clay soil showed the presence of Pb element which was equivalent to 2500 ppm. The mixing of samples prepared and the concentration of contaminant obtained was the same. Therefore, the mixing procedure of the clay soil and Pb was successful.

Table 2 XRF data obtained from the pure clay soil and contaminated clay soil

Clay soil		Contaminated clay soil	
Formula	Concentration	Formula	Concentration
orig-g	9	orig-g	9
added-g	3	added-g	3
CO ₂	0.1	CO ₂	0.1
SiO ₂	47.20%	SiO ₂	43.40%
Al ₂ O ₃	17.20%	Al ₂ O ₃	15.70%
Fe ₂ O ₃	4.87%	Fe ₂ O ₃	5.09%
SO ₃	3.12%	SO ₃	3.48%
CaO	0.25%	CaO	0.83%
		Pb	0.25%

Leachability of Lead Concentration Table 3 shows the values of lead concentration for all samples were reduced. The samples were cured for 7 days, 14 days and 28 days. There are 3 samples for each type. The mean concentration was calculated and used as the main concentration for the analysis.

Table 3 The values of Pb concentration (ppm)

	7 days	14 days	28 days
Clay Soil (contaminated) + 5% cement	7.126	6.852	6.345
Clay Soil (contaminated) + 10% cement	2.847	1.945	1.147
Clay Soil (contaminated) + 15% cement	0.656	0.488	0.364
Clay Soil (contaminated) + 2.5% cement + 2.5% cockle shell powder	4.742	3.416	2.413
Clay Soil (contaminated) + 5% cement + 5% cockle shell powder	1.860	1.480	1.467
Clay Soil (contaminated) + 7.5% cement + 7.5% cockle shell powder	0.381	0.051	0.047

Fig. 1 Concentration of Lead after contaminated soil was treated using S/S technique

Cement (15%) was used as a binder to treat the contaminated clay soil. Some adjustment was created by replacing half of the cement amount used with cockle shell powders; combination of 7.5% cement and 7.5% cockle shell powders. As seen in Fig. 1, the results show a decreasing in concentration of Pb from 0.488 ppm to 0.051 ppm with the percentage reduction of 99% for 14 days of curing. Previous study conducted by Azmi et al. [4] showed that the percentage of reduction in the 14 curing days with 15% cement was 98.96%, while the percentage reduction for the combination of cement and cockle shell powders was 99.9%, which is more efficient.

Different percentages of cement (5% and 10%) were used as a binder for the treatment. As the results, lead concentrations recorded were 6.852 ppm and 1.945 ppm, respectively. The same change was made by substituting half of the quantity of cement used with cockle shell powders given a 99% of lead concentration reduction in 14 days of curing.

Previous research on the solidification/stabilisation (S/S) of lead contaminated soil using Ordinary Portland Cement and rice hush ash (RHA) showed a reduced in the leachability of lead from the treated sample [11]. Lead concentrations were reduced from 0.66 ppm to 0.08 ppm and gave a percentage of 87.9 % in the reduction of lead concentration after the treatment process was conducted.

The combination of cement and cockle shell powder also shows a decrease in the concentration of lead by 99.9% and it is more efficient than the combination of cement and RHA. The usage of the combination of cement with cockle shell powders to treat contaminated soil is proven suitable and effective. Jung et al. [12] past researches used waste oyster shell as amendments to improve the soil quality and stabilised Pb and Cd in contaminated soil.

Conclusion

As a conclusion, cockle shell powders could be an alternatively low cost material in the S/S technique. The suitability of the additive (cockles shell powders) used for the combination with cement is proven very effective. It has given a tremendous result in reducing to 99% of the lead concentration in contaminated clay soil. It also provides an inventory of some waste products that is highly potential to be used as cement for the S/S technique in the developing countries as a good way of a better nation towards sustainability.

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