Leachability of Arsenic (As) Contaminated Landfill Soil Stabilised by Cement and Bagasse Ash

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Abstract. Contaminated soil with heavy metals, especially Arsenic (As) has become a major issue worldwide. As is reported to be a metal that affects human health and is related to have caused serious diseases that interrupts the nervous system, blood vessels and kidneys. However, proper treatment techniques such as Stabilization/Solidification (S/S) method can be employed and is capable of controlling these heavy metals from contaminating the soil strata and groundwater resources. This study is to investigate the leachability of Arsenic (As) in S/S method when bagasse ash (BA) is added to remedy contaminated Landfill soil. Cement is added at a proportion of 5%, 10%, 15% and 20% in sample weights without BA while in another sample; the cement replaces BA at a proportion of 2.5%, 5%, 7.5%, and 10%. All samples were allowed to harden and cured at room temperature for 7, 14 and 28 days. The effectiveness of the treatment was assessed by conducting Synthetic Precipitation Leaching Procedure (SPLP). Results indicate that pH and leachability are found to have major influence on metal release. The final pH after leaching tests showed improvements especially samples containing BA. In addition, the concentration of As in the SPLP test after the curing period of 28 days were detected to be below the leachability limit as regulated by WHO's Guidelines for Drinking-water Quality. As a whole, the results obtained from testing showed that sample containing 10% cement with 10% BA is the most effective and is the optimum mix since this proportion succeeded in minimising the leachability of As at total reduction by 100%. In conclusion, partial replacement of cement with BA in the binder system has been successful in reducing the leachability.

Keywords: Soil Remediation, Stabilisation/Solidification, Geo-environmental Engineering

1. Introduction

Malaysia is a South East Asian country which is undergoing rapid development and excessive population growth. Issues like the demand for a major infrastructure projects, new houses and commercial building has been occur in a developing country like ours. These issues somehow produce a large amount of waste in a construction industry and the collected waste will be unloaded at the selected landfill [1]. Landfill is the most common disposal alternative for many countries by burial the municipal waste. Hodgson et. al., (1992) stated that in 1990s the most waste management that had been used is landfilling. Landfilling also is the cheapest way in term of exploitation and capital cost to dispose all of the waste.

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Unfortunately, the landfilled waste normally contain chemical particles and bacteria such as heavy metal and organic pollutant which produce negative impact to environment, groundwater qualities and human health [3]. Mohamed et al. [4] has reported that toxic from heavy metal can be exposed to environment and may cause kidney damage, decrease mental capacity and neurological damage to human. Pollution groundwater and surface water at the landfill site is due to the landfill leachate that contains major type of pollutant especially heavy metals. Heavy metals constitute is one of the pollutant group that are kept under surveillance because they can be particularly dangerous when they are able to contaminate drinking water. Heavy metal particle such as lead, cadmium, copper, mercury, nickel, arsenic and zinc are released to the environment rapidly and it will settle in bottom sediments of water bodies either direct discharge or surface run-off [5].

From the toxic heavy metals mentioned, Arsenic (As) is the most dangerous because it is inorganic compounds that are extremely toxic and may cause gastrointestinal symptoms, cardiovascular, and nervous system function disturbances and eventually death. Arsenic (As) occurs naturally in the environment as a result of the weathering of parent rocks even though rarely in its elemental form. Arsenic (As) has been used as a decolorizer in the manufacture of glass, in various metallurgical processes such as the production of alloys, veterinary and human medicines, and lead-acid batteries [8]. Furthermore, arsenic (As) also give harmful to the human health due to the increased awareness of the health risks associated with consumption of arsenic containing in water and it also is use for agricultural applications such as herbicides and insecticides [9].

To overcome this problem, various remediation methods have been develop such as stabilization/solidification (S/S) techniques, electro-kinetic techniques, phytoremediation technique and in-situ immobilization technique. Among the methods, S/S technique is an effective, yet economic remediation technology to immobilize heavy metal in contaminated soils and water. Stabilization/solidification (S/S) of heavy metals has been done through the additional of cementations binder like cement incorporated with some potential waste such as agricultural by product [10][11]. Therefore, this study is conducted to investigate the chemical behavior of Arsenic contaminated soil stabilised by cement and bagasse ash (BA).

2. Materials and Methods

2.1 Collection of Soil Sample
The contaminated soil sample has been collected at Bukit Bakri Landfill Site (BBLS), Muar Johor, The top of the soil to a depth of 1 meters has been removed in order to avoid taking the humus, waste and plant roots. Then, the soil was placed in polystyrene containers. After that, the soil sample has been taken to the laboratory to be dried in the oven at 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. Figure 1 shows the location of soil sample collection at BBLS, Muar Johor.
2.2 Cement and Bagasse Ash (BA)
Cement was used as a primary binder reagent in stabilization/solidification (S/S) method. Type of cement that involved in this study is Ordinary Portland Cement (OPC) type 1. Normally, when the soft soil mixed with cement, it can stabilized the soil because cement and water react to form cementitious calcium silicate and aluminate hydrates, which bind the soil particles together [12].

The agricultural waste of sugarcane bagasse ash has been chosen to incorporate with Ordinary Portland Cement (OPC) in S/S technique. Tajudin et. al., [13] stated that sugarcane bagasse ash was known as a mineral additive in cementitious materials. Besides that, sugarcane bagasse ash also one of low cost material, can reduced the remediation cost normally incurred by cement usage and the main advantages of sugarcane bagasse ash was environmental friendly [14]. The raw form of sugarcane bagasse has been dried under natural environment and burnt until change to ash form. These materials are considered as uncontrolled burned bagasse because it is burnt under uncontrolled temperature. The bagasse ash (BA) was further burnt under controlled temperature at 650°C for one hour. This burning process brought down the carbon content at least 4.9% [14]. After cooling, the ash has been ground to 90 μm using grinder machines.

2.3 Production of Stabilization/Solidification (S/S) Samples
In this study, the percentage of cement was partially replaced with bagasse ash (BA) as additive to the soil. The weight of each sample was measured to avoid any material waste, and the quantity of each component was calculated through its percentage in the sample according to Table 1. Sample batches was be triplicated for 3 hydration durations, which is 7, 14 and 28 days. Sample was mixed in bulk for relative homogeneity prior to packing and storage. The water cement ratios used in this research has been determined from the optimum moisture content (OMC) from the compaction test which ranges from 0.20 to 0.40 (w/c) depending on the quantity of bagasse ash (BA) added. Next, the raw materials were mixed in by using a small mixer to ensure the homogeneity of the S/S sample. Then, the S/S sample is to be compacted in a split mould to form a sample of 38 mm in diameter and 76 mm in height. A specially designed miniature hand compacting tool was used to compact the mixture into 4 layers with 50 blows each [15]. The extruded specimens was wrapped and stored for 7, 14 and 28 days prior to testing and before being passed into the chemical testing. Table 1 shows the mix design of S/S sample.
Table 1. Mix Design of S/S sample

| Name of Sample | Label | Percentage of Binder (%) |
|----------------|-------|--------------------------|
|                |       | Soil | OPC | BA |
| Soil           | A     | 100  | 0   | 0  |
| Soil + 5% cement | B   | 95   | 5   | 0  |
| Soil + 10% cement | C   | 90   | 10  | 0  |
| Soil + 15% cement | D   | 85   | 15  | 0  |
| Soil + 20% cement | E   | 80   | 20  | 0  |
| Soil + 2.5% cement + 2.5% SCBA | F   | 95   | 2.5 | 2.5 |
| Soil + 5% cement + 5% SCBA | G   | 90   | 5   | 5  |
| Soil + 7.5% cement + 7.5% SCBA | H   | 85   | 7.5 | 7.5 |
| Soil + 10% cement + 10% SCBA | I   | 80   | 10  | 10 |
| Soil + 5% SCBA | J    | 95   | 0   | 5  |

2.4 Synthetic Precipitation Leaching Procedure (SPLP)

The Synthetic Precipitation Leaching Procedure (SPLP) is used to determine the concentration of contaminants that been leach from the soil (SW-846 Method 1312 US EPA, 1998). The function of SPLP is to exactly evaluate the effects of the acid rain on the contaminated solid as well. It is considered that in a natural environment, the acid rain would apply a worst-case scenario to the waste during the practice of disposal. Two fluids (aqueous solution of sulfuric and nitric acids) were used in this test. Primarily, extraction fluid is slightly acidic at pH 4.20 reflecting the air pollution impacts of heavy industrialization and coal utilization. The other extraction fluid with pH 5.00 was also used for locations with less industrialization and smaller population densities [15]. Therefore, the SPLP extraction fluid used in this research will be a pH of 4.20 to investigate the effectiveness of using bagasse ash (BA) as partial replacement to cement in the worst-case conditions.

In this study, 100 g of crushed samples which passed a 9 mm sieve was placed in a bottle prior to the addition of leaching solution of nitric/sulfuric acid (pH 4.20) to provide ratio of 20:1 mass ratio of leachant to solidified samples. The containers were then agitated using a rotating machine at 30 rpm for 18 hours. Leachate pH was measured at the end of the extraction period. After that, the solid and liquid phases are to be separate by filtration using Grade GF/F 0.7 μm glass fiber filter paper filters. The filtrate were acidified with nitric acid to a pH < 2 and refrigerated (<4°C) prior to heavy metal analysis.

3. Results and Discussions

3.1 pH of Sample at Different Extraction Period and Curing Day

The Figure 2(a) shows pH value of S/S sample at different extraction period with 0 hours and 18 hours at 7 days. From the figure above, it shows that there are a group of pH value which is higher (8 to 14), moderate and lowest pH value. Sample C, D, E, G, H and I shows the higher pH value with 8.94, 10.24, 11.34, 8.70, 10.59 and 11.69 respectively. This is due to the natural alkali behavior of cement and bagasse ash that help to increase the pH value of S/S samples. Sample B, F and J showed some neutral pH value (near pH 7 to 8) with 7.37, 7.86 and 6.81 respectively. While, sample A showed the lowest pH value of 5.62. This significant result occur when the percentage of OPC and BA used are in small percentage and affected by the hydration time [16]. The significant finding has been reported by Malviya & Chaudhary [17] which stated the additional of binder (normally OPC) and curing period always becomes the medium to help the development of pH towards S/S samples for heavy metals remediation.

In the same way, figure 2(b) and 2(c) also showed the significant result as 7 days of curing periods. However, pH value of S/S sample at 14 and 28 days was slightly highest than the pH value at 7 days. The figure indicates that pH value for sample containing OPC standalone (B, C, D, E) and sample OPC incorporated with BA (F, G, H, I) was increase parallel. While sample BA standalone (sample J) was
slightly decreased and control sample (sample A) shows the lowest pH value as it is not containing any binder. From the result, it is worth to conclude that when the percentage of OPC was higher; the pH value will increased to alkali condition. As a proved, sample E containing 20% OPC shows the highest pH value of 11.88 while sample B containing 5% OPC increased the pH at only 8.56. However, this study has revealed a new significant result when additional of BA to the OPC content produce highest pH compare to OPC standalone (Sample I with pH 11.96). This significant result shows that utilization of BA as a partial replacement to OPC content produce better result on helping the S/S sample to increase the pH to the alkali condition.

3.2 Effect of pH on the Leachability of Sample at 7, 14 and 28 Days
The Figure 3 shows that the leachability of Arsenic (As) of S/S sample using Synthetic Precipitation Leaching Procedure (SPLP) at 7, 14 and 28 curing days. From the figure, it is shows that the leachability of As was decrease constantly for all sample. Sample A (control sample) shows the highest value of As concentration at 7, 14 and 28 days with 1911, 1898 and 1309 ppb respectively. This is due to the no binder used to help the C-S-H process which is normally occur when cement or binder added to S/S samples. Contradict to the sample with BA stand-alone (sample J), the concentration of As was
dramatically decreased at 1.898, 7.001 and 1.921 ppb for 7, 14 and 28 days respectively. This significant result shows the advantages of adding the BA to S/S sample at about 99% reduction of As concentration. In the same way, sample G, H and I shows the better result at 100% reduction of As. This result has been expected through the previous study by Janusa et. al., [18] which reported that additional of alkali product helped to reduce the heavy metals concentration at certain curing period as well as type of binder added to the S/S sample.

As a conclusion, from the SPLP testing, the concentration of As are depending on the type of binder used. The utilizing of BA to the OPC content shows a better result on leachability of As. It is proved by sample G, H and I which is produce as low as zero As concentration even at 7 days of curing period. In addition, towards sustainable, sample G can be describe as the optimum mix design for As contaminated soil with only use 5% of OPC to provide zero As concentration in final leachate.

### Figure 3. Arsenic Leachability of S/S sample at 7, 14 and 28 days

#### 3.3 Relationship of pH and As Concentration of S/S Sample at Different Curing Period

The Figure 4(a) shows the pH on As leachability by using the SPLP method at 7 days. The pH values of leachate samples (D, E, H and I) are essentially alkaline at 8.9, 11, 8.7 and 11 respectively while leachate pH of sample (A, B, F and J) are slightly acidic at 4, 4.7, 5 and 4.2 respectively. The addition of BA to cement content has a positive effect on As immobilization. More specifically, the addition of BA results in further widening the pH range 8 to 11 for 7 days of curing periods.

The Figure 4(b) shows the pH on As leachability due to SPLP method at 14 days. The pH values of leachate samples (D, E, H and I) are essentially alkaline at 9.6, 12, 9.1 and 11 respectively while leachate pH of sample (A, B, F and J) are slightly acidic at 4.1, 5.2, 5.5 and 4.5 respectively. Sample C and G are neutral at 7.3 and 6.8. From the figure, result indicate that at 14 days of curing period, the sample with 10% OPC 10% BA leached the least amount of As from soil. This could be since bagasse was determined as an adsorbent material to reduce the leachability of Arsenic.

The Figure 4(c) shows the pH and As leachability of SPLP leachate. The trend of leachability of As in S/S sample at 28 days same as 7 and 14 days. These significant results happen due to the effect of binder with the hydration day. The intensity of pH that often leached Arsenic ranged from 9 to 10 [19]. However, at a lower pH, the concentration will be higher as shown in figure 4(c) where the overall pH ranged from 6.7 to 13 and Arsenic has been leached out up to 99%. This suggests that leachability may be the main mechanism in this study as is proven to be very pH dependent as highlighted by Yin et. al., [20].
Conclusion
The study findings highlight the leachability of Arsenic stabilised with cement and bagasse ash are successful and possible to be partial replacement binder of OPC content in S/S method to treat the contaminated soil, especially by Arsenic. Generally, the use of BA has successfully obtained the desired leachability of Arsenic compared to the sample containing OPC alone. To be better, samples stabilised by OPC with BA are observed below the landfill disposal limit for Arsenic of 0.01 mg/L refer to WHO's Guidelines for Drinking-water Quality. On the other hands, this study has proved that the additional of alkali product helped to reduce the heavy metals concentration at certain curing period as well as type of binder added to the S/S sample. In the same way, this study also revealed that leachability may be the main mechanism as is proven to be very pH dependent. As a conclusion, sample containing lower percentage of OPC are required a longer curing times to retain the heavy metals in soil. Apart from that, sample with 10% OPC and 10% BA has been identified as the optimum mix design in obtaining the desired leachability as well as reducing the usage of OPC. The partial replacement of OPC with BA has successfully reducing the leachability compared to the control sample.

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