Recent Trends in Civil Engineering and Built Environment Vol. 1 No. 1 (2020) 124-132


**RTCEBE**

Homepage: http://penerbit.uthm.edu.my/periodicals/index.php/rtcebe
e-ISSN : 2773-5184

# Potential of Sugarcane Bagasse as Binder in Treatment of Petroleum Sludge by Using Stabilization/Solidification Method

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DOI: https://doi.org/10.30880/rtcebe.2020.01.01.011
Received 29 September 2020; Accepted 07 December 2020; Available online 13 December 2020

**Abstract**: Petroleum sludge is one of the main hazardous wastes generated in the petroleum industry. It is a complex emulsion of various petroleum hydrocarbons and heavy metals. Due to its hazardous characteristics and quantities continuously increased around the world, the effective treatment of petroleum sludge has attracted widespread attention. Proper treatment techniques such as the Stabilization/Solidification (S/S) method have been employed and capable of immobilization of contaminants in leachate. In this study, sugarcane bagasse (SCB) was used partially to reduce the amount of cement in the S/S technique. The objectives were determination the leaching of heavy metals and the strength of the mixture. In the leaching test, concentration for all metals, namely, copper, zinc, chromium, nickel, lead, arsenic, and barium were far below the permissible limit in the USEPA standard. Based on the result, 10% SCB shows the lowest concentration of heavy metals in leachate. In strength performance, the control sample (0% SCB) shows the highest strength. The incorporation of SCB in mixture reduces the strength of the mixture. Only a 5% SCB mixture shows the strength is suitable for landfill disposal. In a conclusion, the SCB was very effective in terms of reducing the leaching concentration. However, in terms of strength SCB was not effective. Thus, it is recommended to include sand, quarry dust or any material which can increase the strength of the mixture in a future study.

**Keywords**: Sugarcane Bagasse, Petroleum Sludge, Stabilization/Solidification, Heavy Metal

## 1. Introduction

Petroleum industries generates enormous volumes of hazardous waste called petroleum sludge. One ton of petroleum sludge waste is generated for every 500 tons of crude oil processed [1]. Petroleum

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sludge is referring to the waste generated due to storage of crude and products that containing mixture of high concentration of hydrocarbon and accumulated heavy metals such as copper, zinc, chromium, nickel, lead, arsenic and barium. It could be produced essentially through the refineries industries during the exploration, production, transportation, storage and refining process. As being recognized as a hazardous waste under the Environment Protection Act and Hazardous Wastes Handling Rules, the improper disposal or insufficient treatment if petroleum sludge can pose serious threats to the environment and human health [1]. This sludge cannot be disposed to the landfill unless they are totally remediated.

The impacts of petroleum sludge are not only related to human risk but also to the degradation of the environment. Potential health effects of this waste, either acute or chronic are subjected to duration of exposure and dosage of receiver [2]. Improper disposal and handling of petroleum sludge may pose a serious threat to groundwater. The toxic substances in the waste may migrate to the receiving stream and contaminate drinking water if not properly treated and disposed [2]. Petroleum sludge is one of the listed in scheduled waster under Environmental Quality (Scheduled Waste) 2005, and must be disposed off at a licensed landfill. Petroleum sludge consisting of hydrophobic substances and substances resistant to biodegradation [2]. The disposal of petroleum sludge to the environment could lead to various toxic effects caused by heavy metals. This toxic pollutant may appear to resist desorption as well degradation.

Towards these problems, a variety of methods have been developed for the treatment of petroleum sludge such as incineration, oxidation treatment, land treatment and bioremediation [1, 3, 4, 5]. Even if the treatment of the sludge using these methods is somehow effective, but the problems of contamination, odor, leaching of heavy metals and fire hazard would still be created. There are some disadvantages for this method by using clean-up technologies such as incineration and burial of sludge insecure landfills are expensive [6].

Due to this matter, stabilization/solidification was implemented to minimize the petroleum sludge toxicity based portland cement (PC) as a main binder with selected additives that is sugarcane bagasse (SCB) for immobilization of contaminant in the sludge. Stabilization/Solidification is quick and effective waste treatment technique [7]. It is one of the encapsulation methods that have been mainly used for hazardous wastes treatment [8]. It provides high strength, low permeability and high durability that ensure the immobilization of heavy metals and organic contaminants within the mixture [9]. In Malaysia, sugarcane bagasse is abundantly produced waste materials, which widely produced by the local industry. There are many ways to utilizing this type of waste as construction material has significant environmental benefits. Therefore, the utilization of sugarcane bagasse may provide a sustainable solution towards the environment rather than being disposed to the landfill. In addition, this solution is decrease the sludge treatment cost by utilizing the binder to replace proportion of cement as well providing costless materials for S/S method that may lead to construction applications.

## 2. Methodology

### 2.1 Raw Materials Preparation

There are three materials that were used in this study, namely Portland Cement (PC), Sugarcane Bagasse (SCB), and Petroleum Sludge (PS). The portland cement was purchased from local supplier and kept in dry location. SCB was collected as a waste from a local supplier that sell the sugarcane juice. SCB was collected after the juice was extracted and was undergo the segregation process. In laboratory, SCB was washed to remove other impurities. Then, SCB was dried to eliminate the moisture content and was grind to 300 µm. Sludge was collected from Petronas Refinery, Melaka. The sludge was dried in an oven for 24 hours at a temperature of 110ºC and the sample was kept in a cool room.

2.2 Preparation of S/S mixtures

The PS was added together with PC and SCB then allowed it to homogenize for approximately 5 minutes in order to eliminate any lumps that may have formed. Then, water was added gradually at water-to-mixture (W/M) ratio of 0.5. After mixing, the mixture was casted into 100 mm x 100 mm x 100 mm cube molds in 3 layers; with each layer was compacted by manual compactor to yield good packing of the mixture. All the samples were triplicated for two hydration durations which were 7th and 28th days for air drying of the solidified samples in a cabinet at a controlled condition (Temperature= 25±2˚C, Humidity> 90%). Table 1 shows the mixtures of SCB, PS and PC by using 100 $mm^3$ mould.

**Table 1: Ratio of SCB, PS and PC use in this study**

| Sample | Binder | | Sludge (%) | Water – Cement ratio |
|---|---|---|---|---|
| | **OPC (%)** | **SCB (%)** | | |
| A (control) | 70 | 0 | 30 | 0.50 |
| B | 65 | 5 | 30 | 0.50 |
| C | 60 | 10 | 30 | 0.50 |
| D | 55 | 15 | 30 | 0.50 |
| E | 50 | 20 | 30 | 0.50 |

2.2 Leaching Test

Crushed block leaching test, namely Toxicity Characteristic Leaching Procedure (TCLP) as described in SW-846 Method 1311[10] was conducted on the block samples subsequent to 7 days and 28 days of air drying. Before TCLP was commenced, the cube samples were first roughly crushed using a steel hammer. Then, samples of the roughly crushed were collected and crusher further using the grinder machine equipment. When they were reduced to fine grains and tiny rocks, they were sieved to obtain samples that passing through 9.5mm sieve. The sieve samples were then used for extraction in TCLP. After cube has been sieved, process of extraction fluid was done that was dilute 5.7 mL glacial acetic acid with reagent water (distilled water) to a volume of 1 litre. Then check the pH of fluid to make sure it reaching the 2.88 + 0.05. Analytical Testing Model DC-20 Rotary Agitators was used for evaluating the mobility of regulated compounds. Finally, the samples were analyzed using Inductive coupled plasma mass spectrometry (ICP-MS) to determine the concentration of dissolved metals [13].

2.3 Unconfined Comprehensive Strength (UCS)

The UCS test is one the main important test for the S/S method to determine the safe disposal which must above than 0.35 MPa. This test was conducted in order to evaluate the strength of the solidified samples according to BS 1881 – 116: 1983 [13].

## 3. Result and Discussion

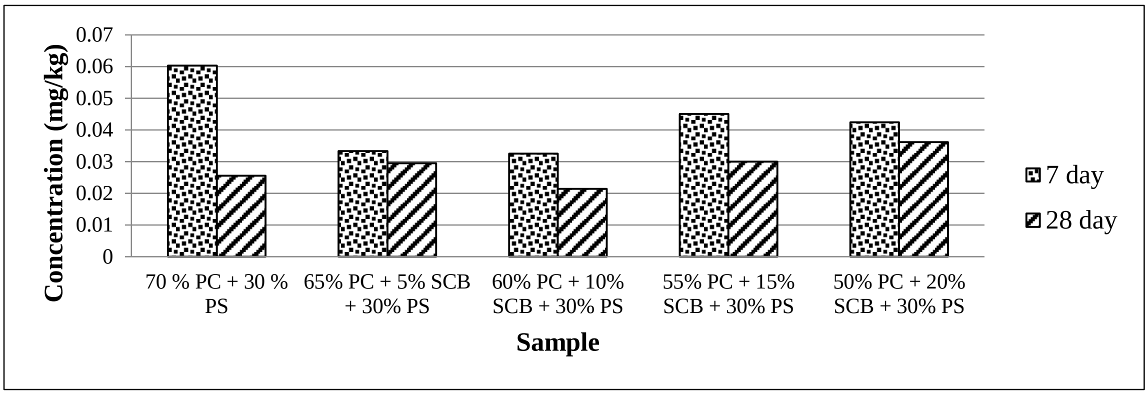
These results are divided into two parts according to the objectives of this study. The first part is analysis of the concentration of heavy metal in leachate and compare with standard. The second parts represent the analysis of the result of the compressive strength that based on days of cured, namely, 7 days and 28 days. According to U.S. EPA, 0.35 MPa is the minimum compressive strength required for the safe disposal of stabilized waste [12].

3.1 Leaching test

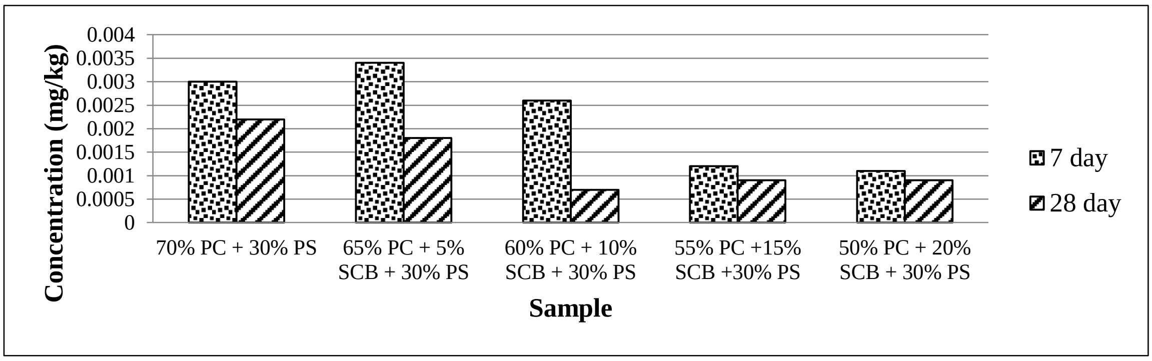
In leaching test, the result can be divided into two categories, namely, positive result and negative result. In positive result, concentrations of Zn, Pb and Ba were reduced when SCB was replaced PC in mixtures. Whereas, concentration of Cu, Cr, Ni and As were increased compared to control sample, which no SCB in the mixtures.

Figure 1 shows the concentration of Zn, Pb and Ba for all samples. Based on the figure, the highest leaching concentration of Zn that undergo 7 days curing process was 0.0603 mg/kg, whereas the lowest was 0.0325 mg/kg. For 28 days curing the Zn highest concentration was 0.0362 mg/kg, whereas the lowest was 0.0214 mg/kg. The highest leaching of Pb concentration that undergo 7 days curing process was 0.0030 mg/kg, whereas the lowest was 0.0011 mg/kg. For 28 days curing the highest Pbconcentration was 0.0022 mg/kg, whereas the lowest was 0.0007 mg/kg. For Ba, the highest leaching concentration that undergo 7 days curing process was 0.653 mg/kg, whereas the lowest was 0.2705 mg/kg. For 28 days curing the highest concentration was 0.6505 mg/kg, whereas the lowest was 0.1738 mg/kg.

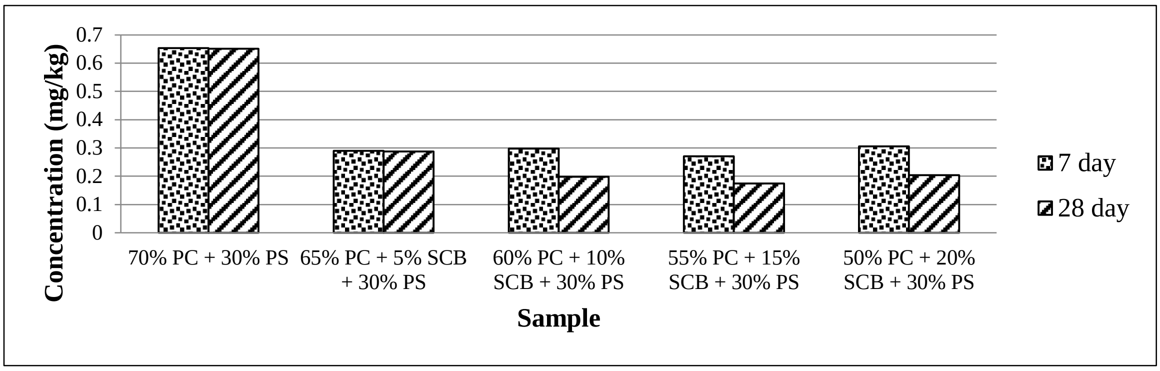
There were drops of concentration of Zn and Ba based on control sample that is 70 % PC and 30 % PS to mixtures that were replaced with SCB. Whereas, Pb concentration was increased when replaced with 5% SCB on 7 days curing. However, when replaced 10% SCB, Pb concentrations were reduced for both 7 days and 28 days curing. The lowest leaching concentration and also the most effective to reduce the leaching concentrations of Zn, Pb and Ba were the mixtures of 60 % PC, 10 % SCB, and 30 % PS. Thus, using 10 % SCB to replace PC was appropriate to minimize the leaching concentration of Zn, Pb and Ba.




(a)




(b)

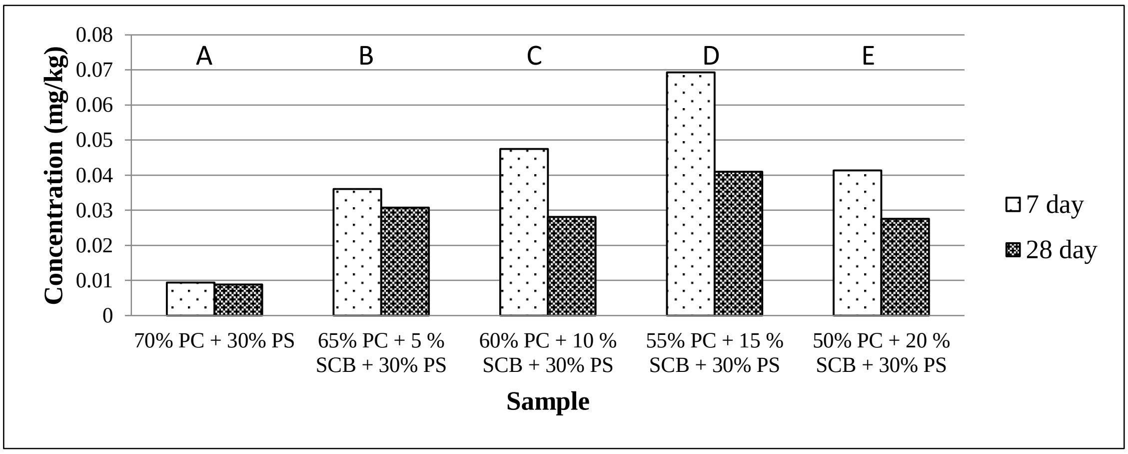



(c)

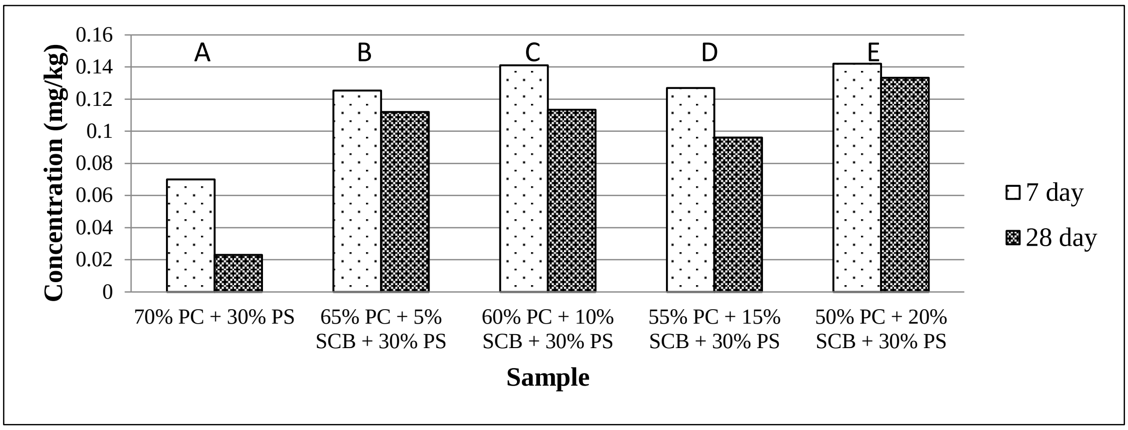
**Figure 1: Concentration of Zn (a), Pb (b) and Ba(c) in different ratio of PC, SCB and PS**

Figure 2 shows the concentration of Cu, Cr, Ni and As for all mixtures. Based on the figure, sample that using ratio of 55 % cement, 15 % SCB and 30 % PS shows the highest Cu and As concentration in leaching test. The highest Cu concentration that undergo 7 days curing process was 0.0693 mg/kg, whereas the lowest was 0.0094 mg/kg. For 28 days curing the highest Cu concentration was 0.0410 mg/kg, whereas the lowest was 0.0088 mg/kg. The highest Cr concentration that undergo 7 days curing process was 0.1420 mg/kg, whereas the lowest was 0.0700 mg/kg. For 28 days curing the highest Cr concentration was 0.1333 mg/kg, whereas the lowest was 0.0230 mg/kg. The highest Ni concentration that undergo 7 days curing process was 0.1540 mg/kg, whereas the lowest was 0.1190 mg/kg. For 28 days curing the highest Ni concentration was 0.1360 mg/kg, whereas the lowest was 0.0934 mg/kg. The highest leaching As concentration that undergo 7 days curing process was 0.0023 mg/kg, whereas the lowest was 0.0007 mg/kg. For 28 days curing the highest As concentration was 0.0019 mg/kg, whereas the lowest was 0.0006 mg/kg.

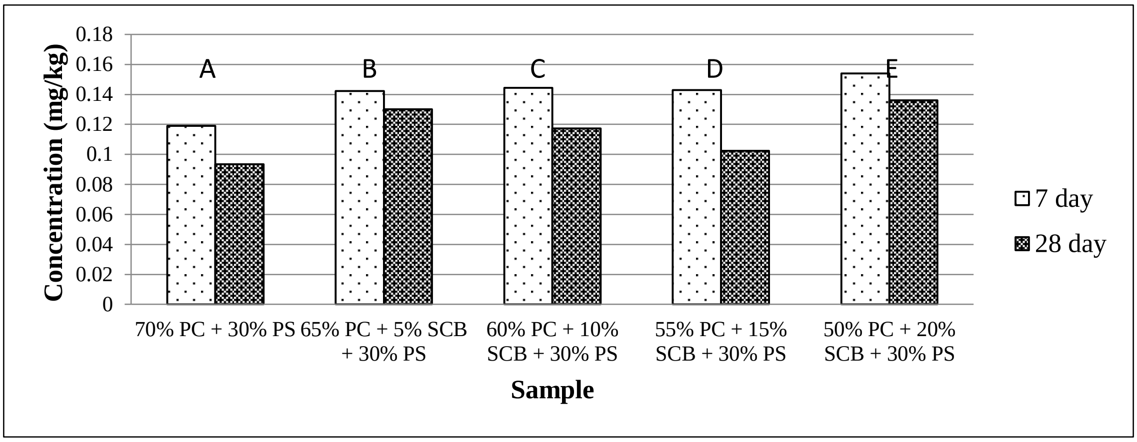
Control sample that is 70 % PC and 30 % PS shows the lowest leaching concentration. Based on this result, replacing SCB to reduce the leaching concentration of Cu, Cr, Ni and As were less suitable due to of the control sample was more effective to reduce the concentration of these metals. Sample that using ratio of 55 % PC, 15 % SCB and 30 % PS shows the highest value of leaching Cu and As concentrations. Whereas, for Cr and Ni, ratio of 50% PC, 20% SCB and 30% PS shows the highest concentration of this metals in leaching test. Replacing SCB in the mixtures was increased these metals concentration in leachate may be due to these metals are in high concentrations in SCB. This can be proved by conducting digestion test on SCB and then the characteristics of SCB will be known. Due this is preliminary study, it is highly recommended to conduct characteristics study of SCB and other raw materials.



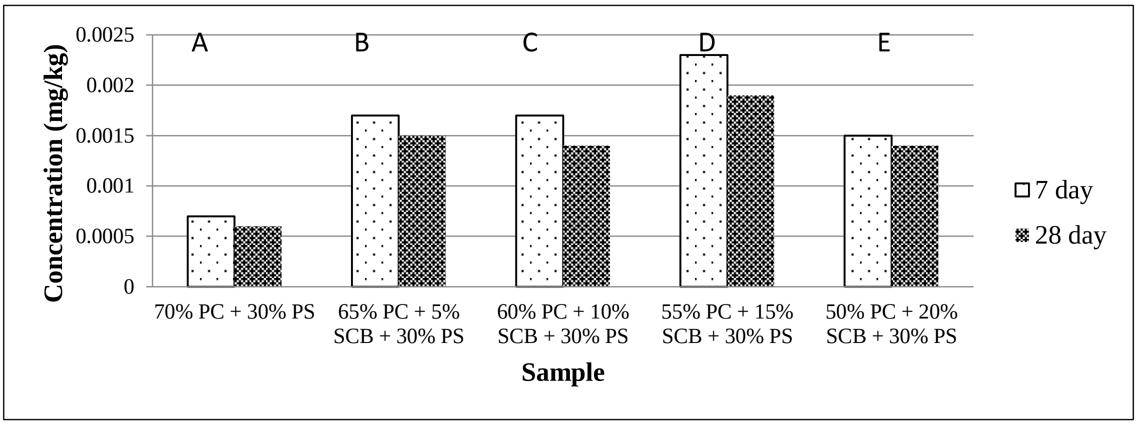

(a) Cu




(b) Cr




(c) Ni




(d) As

**Figure 2: Concentration of Cu (a), Cr (b), Ni (c) and As (d) in different ratio of PC, SCB and PS**

The main aim of this study is to remediate the sludge by immobilization the heavy metals. Therefore, the comparison of metals concentration in sludge and in leachate was made. As for leaching test that were conducted which was TCLP, the concentrations of Zn that leached out were far below than the initial concentration of raw petroleum sludge, 189 mg/kg [13]. By using S/S method, Zn concentration was dropped to average for all samples that was 0.0356 mg/kg, which only 0.02% by comparing to concentration in raw sludge. Average Ba concentration for all samples in leachate was 0.3632 mg/kg, which was 0.72% by using S/S method. The concentrations of Ba that leached out from all samples were far below by comparing to the initial concentration of raw petroleum sludge, 50.4 mg/kg [13].

For Pb, the concentrations that leached out were far below compared to initial concentration in raw petroleum sludge, 25.1 mg/kg [13]. By using S/S method, Pb concentration was dropped to average for all samples was 0.0023 mg/kg, which was only 0.01% by comparing to concentration in raw sludge. Based on USEPA standard related to TCLP testing, concentration below than 5 mg/kg is suitable for safe disposal into landfill [8]. All samples show the Pb concentration in leachate were under the permissible limit. These results indicate that the maximum leaching of Pb by replacement of SCB in S/S matrices was effectively immobilization these metals from the sludge.

Average Cu concentrations in leachate for all samples was 0.0407 mg/kg. This concentration was much lower compared to the initial concentration of raw petroleum sludge, 18 mg/kg [13]. The different concentration was 0.2% by comparing concentration from the raw sludge and leachate. Whereas, Cr concentration in sludge was 120 mg/kg and by using the S/S method, Cr concentration was measured at average for all samples was 0.1211 mg/kg. The different was 0.1% by comparing Cr concentration in raw sludge and leachate. In addition, this average concentration was below the permissible limit, which the concentration is 5 mg/kg by referring to USEPA standard in TCLP testing. For Ni, 0.141 mg/kg was the average concentration for all samples. This concentration was much lower compared to initial Ni concentration in sludge, which was detected at 22 mg/kg [10]. The difference of Ni concentration in sludge and leachate was 0.64%. By referring to USEPA standard, 1.3 mg/kg is the permissible limit for safe disposal. Thus, S/S method was successfully immobilization of Ni in mixtures. In raw sludge, 14.1 mg/kg of As concentration was measured and this concentration was above the permissible limit which is 5 mg/kg by referring to USEPA standard. By using S/S method, the As concentration was detected at 0.0016 mg/kg, which the difference is 0.01%.

### 3.2 Unconfined Compressive Strength

Strength is one of the main testing in S/S method. For safe disposal of stabilize hazardous waste, the strength must above than 0.35 MPa [8]. Beside that, the strength is an important factor because it determines the maximum load that can sample bear, which means the mixtures may be potential to be use in construction material. The strength of the mixture depends on the partial replacement of cement and also the hydration day. The result of the strength in this study is presented in Figure 3. The results clearly indicated that the strength of sample increasing with extended period of curing, which from 7 days to 28 days for all samples. This is due to the availability of sufficient moisture content for the hydration process to proceed and bonding of all material inside the cube. The strength for replace by using SCB between 7 and 28 days show the percentage of average increment of strength, which was 12 %, but still not surpassing the requirement for safe disposal into landfill.

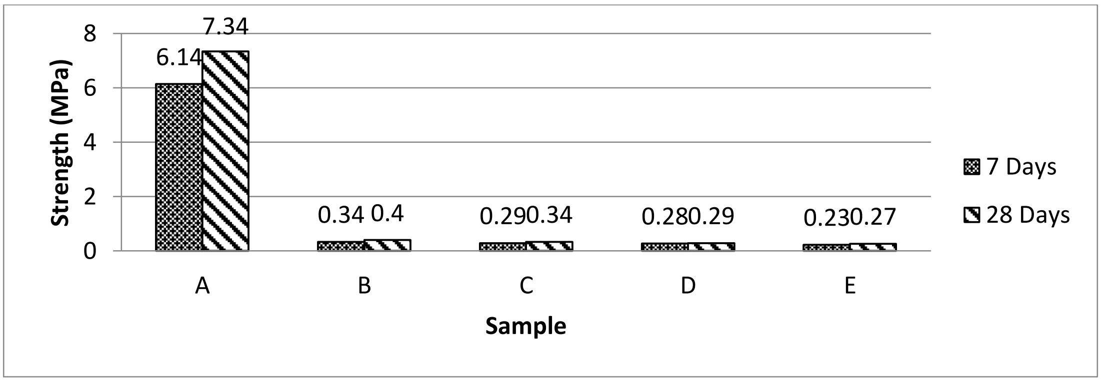



**Figure 3: Strength of the mixtures at 7 and 28 days**

It clearly shows that the sample without replacement of SCB shows the highest strength. Strength for this sample was measured at 6.14 MPa for 7 days and 7.34 MPa for 28 days. Then, the sudden drop of strength was measured when mixtures were replaced with SCB at 5% with the strength were 0.34 MPa and 0.4 MPa for 7 days and 28 days curing, respectively. The strength was gradually decreased with increasing of SCB at 10%, 15% and 20%, which was measured at 0.29 and 0.34, 0.28 and 0.29, 0.23 and 0.27 MPa for 7 days and 28 days, respectively. Based on this result, it shows the greater different in term of strength between the control sample and samples with replacement using SCB.

The control sample was surpassed the requirement for safe disposal at landfill, which was higher than 0.35 MPa. Whereas, replacement with SCB, only 5 % SCB at 28 days of curing was the only surpassing the limit for safe disposal. During the experiment, SCB was quickly adsorbed the water compared to cement. Thus, this process was effect the hardening process of S/S samples. This process also causes the low strength when samples with SCB replacement. In future studies, it is highly recommended that SCB need to modify to reduce the absorption capacity.

## 4. Conclusion

This study shows that by using TCLP leaching test, the heavy metals concentrations in leachate were far the permissible limit based on USEPA standard. In addition, there is significant reduction of heavy metals by comparing concentrations in leachate and initial concentration of raw petroleum sludge. Replacement with SCB shows the reduction of Zn, Pb and Ba concentrations in leachate compared to control sample. Whereas, Cu, Cr, Ni and As show the opposite result. Based on Zn, Pb and Ba leaching test, the highest potential ratio was 60 % PC + 10 % SCB + 30 % PS, which detect the concentrations of Zn, Pb and Ba were 0.0214 mg/kg, 0.0007 mg/kg and 0.1978 mg/kg, respectively. This significant reduction indicated that the use of SCB as partial cement replacement is possible due to reduction of Zn, Pb and Ba concentrations during leaching test.

Through unconfined compression strength, the result shows that using SCB as partial cement replacement in treatment of petroleum sludge was not suitable for disposal because the strength was not achieve the requirement for safe disposal of stabilized waste that is according to U.S EPA, which is 0.35 MPa. In this case, only the ratio of 5 % SCB was achieved the safe disposal standard, which was 0.40 MPa at 28 days of curing. By comparing days of curing, 28 days' result shows the increment of strength by comparing to 7 days, but still not surpassed the requirement standard.

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