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Long-Term Leaching Behavior of Petroleum Sludge Waste Containing Palm Oil Fuel Ash and Quarry Dust by Using Solidification/Stabilization Method

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ABSTRACT---In recent years, petroleum industry has generated huge amount of petroleum sludge. There are many types of treatment method in petroleum sludge (PS) and one of the treatments that arise the attention to treat the sludge is solidifica-tion/stabilization (S/S) method as it immobilizes and stabilizes the hazardous substances within a solid matrices. The main objectives of this research are to formulate the correct mixture of PS in S/S matrices and to stimulate the long-term leaching behaviour of S/S matrices in PS treatment with palm oil fuel ash (POFA) and quarry dust (QD) as partial cement and sand replacement respectively. The characterization of PS, POFA and QD were done by using X-Ray Fluorescence (XRF). Furthermore, different percentages of petroleum sludge (control, 0%, 5%, 10%, 20%, 30%) were incorporated with fixed 10% of POFA and 20% of QD replacement into S/S matrices. This research also includes a comparison of two different leaching test parameters such as liquid to surface area ratio, pH, and type of leaching solution on the samples. The leaching test conducted namely are Semi-Dynamic Leaching procedure (SDLP) and Static Leaching Test (SLT). The leachability results from SDLP and SLT indicated that all concentration for nine (9) heavy metals were below the permissible limit set by United States Environmental Protection Agency (USEPA, 1996) standards. In addition, the S/S effectiveness was evaluated by measuring effective diffusion coefficients (De) and leachability index (Lx). Results of S/S treatment shown that De values were lower and Lx values were higher than 8 thus the S/S can be disposed in sanitary landfill.

As a conclusion, the results for the leaching behaviour of PS up to 10% incorporated with 10% of POFA replacement in cement and 20% of QD replacement in sand demonstrated minimum leaching of the heavy metals from S/S matrices.
This indicated that S/S method could be an alternative disposal method for PS and has potential for replacing other treatment methods that applied at treatment stage for petroleum refinery effluents.

Keywords: petroleum sludge, solidification/stabilization, palm oil fuel ash, quarry dust, waste utilization

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1. INTRODUCTION

Petroleum has become the world's important source of energy due to its high energy density, easy transportability and relative abundance. The petroleum industry is one of the industries that contribute to sludge waste in Malaysia due to high demand from the consumer (Islam, 2015). According to Department of Environment (DOE) Malaysia, petroleum sludge is classified as scheduled waste due to the oil content and in most cases, it is also classified as a liquid which means that is must be landfilled at a secure landfill to meet the guidelines as proposed by DOE of Malaysia (Murshid, 2018). During oil processing at the refineries, a great amount of petroleum sludge was formed. Then the sludge is stored in special storage ponds of the open type (Vdovenko *et al.*, 2015). The disposal of petroleum sludge to the environment will lead to various toxic caused by heavy metals (Talib *et al.*, 2008). For example, the heavy metals concentration in raw petroleum sludge from refineries was reported in recent literatures as 1299 mg/kg for Zinc, 60200 mg/kg for Iron, 500 mg/kg for Copper, 480 mg/kg for Chromium, 480 mg/kg for Nickel and 565 mg/kg for Lead (Islam 2015). This problem happened due to the nature of the crude oil, the processing capacity, the downstream capacities and the design of the effluent treatment plant. Petroleum sludge usually located in refineries due to pump failures, desalter failure, oil draining from tanks and operation units (Elektorowicz *et al.*, 2005). In the petroleum refining process, it has generated wastewater of about 1.6 times the volume of crude oil processed which is a great amount of petroleum sludge is formed during oil processing at the refineries (Coelho *et al.*, 2006). Uncontrolled handling of this sludge can lead to environmental pollution and affects the aesthetic quality.

Due to the harmful side effects of petroleum sludge, it has attracted many types of treatment methods such as centrifugation, solidification/stabilization, solvent extraction, land farming, pyrolysis, biodegradation, photocatalysis, ultrasonic treatment, incineration and others (Benlamoudi *et al.*, 2017; Hamada *et al.*, 2018; Hua *et al.*, 2013). Solidification/Stabilization (S/S) method defines as a process that used to improve the geotechnical properties of unsuitable soils or to remediate contaminated waste materials. Basic application of S/S involves by mix the contaminated material with a hydraulic binder such as cement or secondary additive. This method will reduce the hazardous aspect of the waste (Shirani, 2011).

Even though solidification/stabilization (S/S) method has widely been used for a long period of time, the chemical specification and binding mechanism of metal ions with cement and other reagents have not been fully characterized. Currently, researchers are investigating on cement replacement used in S/S matrices (Li, 2001). Various researchers have been carried out using palm oil fuel ash and quarry dust in partial cement and sand replacement in S/S method which resulted as a good binder due to its abundance and high pozzolanic characteristics (Thomas, 2017).

The use of palm oil fuel ash and quarry dust could also reduce the amount of wastes generated from the industry (Hamid *et al.*, 2018; Chen *et al.*, 2009; Chun *et al.*, 2008). From the previous study, Xue *et al.*, (2017) investigate on leaching behaviour of lead in solidified/stabilized waste using a two-year semi-dynamic leaching test. From the result, it concluded that S/S technology provides a convenient and economical method for treating heavy metal contaminated soils. According to Azhar *et al.*, (2016), ordinary Portland cement (OPC) are the most frequently studied for the remediation of heavy metals in soil by using S/S method. This study also has revealed that, by using OPC incorporated with other potential additive such as fly ash, bottom ash and others have showed satisfactory results as well as help on reduced the remediation cost. The long-term leaching characteristics of S/S matrices are controlled by diffusion. According to Dermatas *et al.*, (2004), the leaching of contaminants out of cement-based waste form is mostly a diffusion controlled process. Thus, the long-term leachability of heavy metals from S/S matrices was evaluated by using the method of American Nuclear Society (ANSI/ANS-16.1,2003). Meanwhile, the S/S effectiveness in this research was evaluated by measuring effective diffusion coefficients (De) and leachability index (Lx). Therefore, this research focuses on investigation of the long-term leaching of heavy metals from petroleum sludge by using S/S matrices incorporated with palm oil fuel ash as replacement of cement and quarry dust as replacement of sand respectively. In addition, the S/S is a method that was used to encapsulate the pollutants within matrices using the cement as a hydraulic binder. Cement based S/S method is a process in which petroleum sludge is mixed with cement, sand and water. In this research, two types of leaching method are used to give the result on composition of heavy metals from S/S matrices and effect from the long-term leaching of heavy metals by using Semi-Dynamic Leaching Procedure (SDLP) and Static Leaching Test (SLT).

2. MATERIALS AND METHODS

The experimental program presented in this paper was done at Universiti Tun Hussein Onn Malaysia and Kolej Kemahiran Tinggi Mara located in Batu Pahat, Johor, Malaysia. The details of the experimental works used are described in the following sections.

2.1. Raw materials preparation

Petroleum sludge (Figure 1) has been chosen to be treated and was collected from oil refinery plant located in Melaka. The Petronas Melaka Refinery Complex located in the state of Melaka in Malaysia houses two refining trains which are known as PSR-1 and PSR-2. Specifically, this refinery plant has a net total refining capacity of more than 440,000 barrels per day and their refinery operations allow them to meet the strong demand for refined petroleum products both domestically and overseas. The petroleum sludge was dried in the oven for 48 hours at a temperature of 110˚C (Johnson *et al.*, 2016).

Fig. 1: Petroleum sludge

Meanwhile, POFA was obtained from Kian Hoe Plantation Company at Kluang, Johor (Figure 2). POFA for this research procured from industry is a waste of oil palm dry biomass which was burnt as a fuel at a temperature of 800 ºC to 1000 ºC. After procuring the ash, it was oven dried at a temperature of 110 ºC to remove moisture and further, it was sieved by using 90 µm sieve to eliminate unburned fibers and to improve its fineness, as particle size plays a crucial role in pozzolanic reactivity. Hence, in order to enhance the pozzolanic reactivity POFA must be ground to produce lower particle size as suggested by Kroehong *et al.*, (2011).

Fig. 2: Palm oil fuel ash (POFA)

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QD was received from Bina Kuari (K) Sdn. Bhd. located at Kedah (Figure 3). The quarry dust waste was dried and placed in the oven at 105^oC. After drying, quarry dust waste was grinded and crushed to make it easier in the sieve process as well to eliminate impurities. The criteria of the waste are very fine dust (75 µm). Beside that, Portland cement was supplied by Lafarge Malaysia that comply with Malaysian Standard MS 522: Part 1: 1989.

Fig. 3: Quarry dust (QD)

2.2. Raw materials characterization

Characterization of main composition and heavy metals in raw materials such as petroleum sludge, palm oil ash, quarry dust and cement are an important parameter to determine the components that are exist in the materials. Therefore, the main composition of raw materials of cement, sand, POFA, and QD were examined using X-ray fluorescence (XRF). This method is fast, accurate and non-destructive with a minimum of sample preparation. To get the result, sample preparation needs to be done by preparing the pressed pellet.

2.3 Semi-dynamic leaching procedure (SDLP)

This method is suitable for many types of solid materials in monolithic form for example cement and solidified wastes. Other than that, it may be suitable for compacted granular materials for example soils, sediments and stacked granular wastes. Furthermore, this method is running under diffusion-controlled release conditions, as a function of leaching time (USEPA, Method 1315, 2013). In this research, deionised water was used as a leaching fluid and was tested until pH reached 7. Then, the S/S matrices samples were immersed uniformly for sixty-three days (63 days) with deionised water. The samples were suspended or held in the leaching fluid. The leaching fluid were renewed and the samples was test out periodically and analyzed on the specified days. The leachate collected was measured with pH meter and preserved with nitric acid until pH below than 2 before being analyzed using Inductively Coupled Plasma Mass Spectrometry (ICP-MS) to determine the dissolved metals. Figure 4 to Figure 7 shows the SDLP process.

Fig. 4: Setting-up container

Fig. 5: S/S matrices were suspended in the leaching fluid

Fig. 6: Leachate was collected

Fig. 7: Leachate collected was measured using pH meter

2.4. Static leaching test (SLT)

SLT method, (ANSI/ANS-16.1) was used to evaluate the release of an element from a material that exists and thus provides characteristics of pollutants that release from waste materials. Static Leaching test procedure was conducted similar with SDLP method. Instead of using deionised water as a leaching fluid, it used 60% of sulphuric acid and 40% of nitric acid in deionised water. The leaching fluid that were prepared was measured until reached pH 4.2 before the S/S matrices were immersed in the solution (Figure 8).

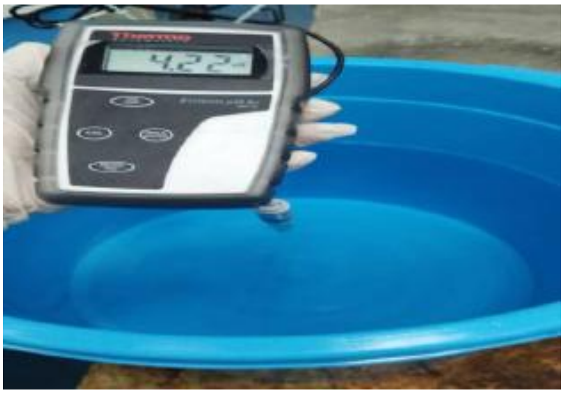

Fig. 8: Leaching fluid were measured until reached pH 4.2

The leaching fluid was renewed, and the samples was test out periodically and analysed on the specified days as tabulated in Table 1 which is similar to SDLP. Then the leachate collected was measured with pH meter and preserved before being analysed using Inductively Coupled Plasma Mass Spectrometry (ICP-MS).

Table 1: Schedule of eluate renewals

Label	Duration hour (h)	Duration day (d)	Cumulative Leaching Time (d)
T01	2.0 ± 0.25	-	0.08
T02	23.0 ± 0.5	-	1.0
T03	23.0 ± 0.5	-	2.0
T04	-	5.0 ± 0.1	7.0
T05	-	7.0 ± 0.1	14.0
T06	-	14.0 ± 0.1	28.0
T07	-	14.0 ± 0.1	42.0
T08	-	7.0 ± 0.1	49.0
T09	-	14.0 ± 0.1	63.0

Meanwhile, Table 2 shows the comparison between SDLP and SLT in this research.

Table 2: Comparison of SDLP and SLT

Test Method	SDLP	SLT
Leaching Fluid	Deionised Water	60% Sulphuric Acid and 40% Nitric Acid in deionised water
Leaching Fluid pH	7	As low as 4.2
Time in Acid bath	(1,5,7 and 14 days interval)	(1,5,7 and 14 days interval)
Liquid to surface area ratio	9± ml/cm^2	9± ml/cm^2

2.4. Effective diffusion coefficient (de) and leachability index (lx)

The long-term leachability of heavy metals from S/S matrices was evaluated using the method of American Nuclear Society (ANSI/ANS-16.1, 2003). Based on the method, it has standardized a Ficks's law-based mathematical diffusion model using Eqs. (1) and (2) respectively to evaluate the leaching rate with respect to time:

$$De = \pi \underline{(a_n / A_o)}^2 \times \underline{(V)}^2 \times T\,(1)\ (At)_n\,S \qquad (1)$$

where an is the contaminant loss (mg) during the particular leaching period with subscript n, Ao is the initial amount of contaminant that exists in the specimen (mg), V is the specimen volume (cm^3), (At)n is the duration of the leaching period in seconds, S is the surface area of the specimen and T is the time that elapsed to the middle of the leaching period, n (sec). From the formula, ANS 16.1, (2003) also provides the other parameters to evaluate the effectiveness of S/S matrices which is Lx. The Lx is defined as follows with B = 1 cm^2/s:

$$Lx = \sum_1^n (\log(\frac{B}{De})) \qquad (2)$$

According to Environment Canada, (1991) Lx can be used as a performance criterion for S/S matrices utilization and disposal. Xue *et al.*, (2016) discussed that when the Lx value is larger than 9, the S/S matrices can be acceptable to use for specific utilization for example road base material, quarry rehabilitation, lagoon closure and others. Another researcher Dermatas *et al.*, (2004) also discussed that when the Lx value is between 8 and 9, the S/S matrices could be disposed to landfill. However, if the Lx has value lower than 8, the S/S matrices cannot be disposed of (Dermatas *et al.*, 2004).

2.5 Mix proportion of the Solidification and Stabilization (S/S) matrices

Preliminary stage is designed to investigate the optimum value for palm oil ash and quarry dust replacement in S/S matrices. This stage is important to find the optimum percentage of POFA and QD replacement to be incorporated with petroleum sludge in S/S matrices. Two types of S/S matrices were produced in the preliminary stage of this research, each of matrices were used to study both the effect of palm oil fuel ash replacement in cement and to investigate the effect of quarry dust replacement in sand. As such, the investigation begins with replacement of palm oil fuel ash (POFA) to cement with 10%, 20%, 30%, and 40% respectively. The S/S matrices was tested in comparison to the control mix that had a ratio of 1:2.75 of cement to fine aggregates. Meanwhile w/c ratio of 0.485 which is in accordance with the ASTM C109 standard for compressive strength testing of hydraulic cement mortars. Next, the same percentages will be used as a sand replacement by using quarry dust which is 10%,20%, 30% and 40% respectively. Mix proportions are shown in Table 3 and Table 4. The actual mix proportion of petroleum sludge incorporated with POFA and QD by using S/S method were developed after the optimum percentages of POFA and QD replacement has been achieved. From the preliminary stage, the results show that 10% POFA and 20% QD were the optimum replacement of cement and sand respectively.

Table 5 shows the mix proportion of the S/S matrices.

Table 3: Details of mix proportion for palm oil fuel ash replacement

Samples	Percentage of POFA replacement (%)	Binder (kg)		Fine Aggregates (1:2.75 wt binder) (kg)		Water to binder ratio (%)
		Cement	POFA	Sand	Quarry dust	
A (Control)	0	7.893	0	21.706	0	0.485
POFA10%	10	7.104	0.789	21.706	0	0.485
POFA20%	20	6.314	1.579	21.706	0	0.485
POFA30%	30	5.525	2.367	21.706	0	0.485
POFA40%	40	4.736	3.157	21.706	0	0.485

Table 4: Details of mix proportion for quarry dust replacement

Samples	Percentage of POFA replacement (%)	Binder (kg)		Fine Aggregates (1:2.75 wt binder) (kg)		Water to binder ratio (%)
		Cement	POFA	Sand	Quarry dust	
A (Control)	0	7.893	0	21.706	0	0.485
QD10%	10	7.893	0	19.535	2.171	0.485
QD20%	20	7.893	0	17.365	4.341	0.485
QD30%	30	7.893	0	15.194	6.512	0.485
QD40%	40	7.893	0	13.024	8.682	0.485

Table 5: Details of mix proportion for S/S matrices of petroleum sludge incorporated with POFA and QD

Samples	Percentage of Petroleum sludge (%)/(kg)	Binder (kg)		Fine Aggregates (1:2.75 wt binder) (kg)		Water to binder ratio (%)
		Cement	POFA	Sand	Quarry dust	
A (Control)	0	7.893	0	21.706	0	0.485
PS 0%	0/0	7.104	0.789	17.365	4.341	0.485
PS 5%	5/1.480	7.104	0.789	17.365	4.341	0.485
PS 10%	10/2.960	7.104	0.789	17.365	4.341	0.485
PS 20%	20/5.920	7.104	0.789	17.365	4.341	0.485
PS 30%	30/8.880	7.104	0.789	17.365	4.341	0.485

3. RESULTS AND DISCUSSION

3.1 Petroleum sludge characterization

Petroleum sludge waste is characterized for organic and metal analysis. The properties of petroleum sludge obtained from Petronas Refinery are tabulated in Table 6. The sludge has moisture content of 34.6 % and specific gravity of 0.994. The loss on ignition at 900° is 45.9%. pH for this petroleum sludge was recorded in acidic condition which is at pH 6. Other than that, the organic fraction of petroleum typically consisted of aromatic and semi volatile compounds. These compounds are detected in form of TPH of organic fraction.

Table 6: Petroleum sludge characterization

Properties	Sludge Characterization		
	Parameter	Value	Method
Chemical/ Physical Properties	Moisture Content	34.6%	TC WI :2003 (E)
	Specific Gravity	0.994	BS 1377: Part 1: 1990
	Loss of Ignition @900□c	45.9%	TC WI: 2003 (E)
	pH	6	USEPA SW-846:9045D.
Organic Fraction	TPH C6-C9	984 mg/kg	USEPA 8260B
	TPH C10-C14	56300 mg/kg	USEPA 8015B
	TPH C15-C28	188000 mg/kg	USEPA 8015B
	TPH C29-C36	47100 mg/kg	USEPA 8015B

3.2 Heavy metals and elemental composition

Based on Table 7, it shows that there were nine heavy metals exist in petroleum sludge, POFA, Portland cement, QD and sand. There elements of heavy metals studied in this research which are Cu, Pb, Zn, Ni, Ba, Cr, Fe, As, and V. Based on the table, the result indicated that there are several elements of heavy metals concentration that exceeded the permissible limit set by USEPA (1996) such as Pb (7 mg/L to 62 mg/L), Ni (4 mg/L to 22.2 mg/L), Cr (9 mg/L to 120 mg/L) and As (6 mg/L to 39.7 mg/L) respectively. Another heavy metals such as Cu (6 mg/L to 52 mg/L), Zn (11 mg/L to 189 mg/L) and V (40.6 mg/L to 84 mg/L) shows that the result is below the regulatory limit. Other than that, the highest heavy metals concentration was Fe with 6116 mg/L, 296 mg/L and 416 mg/L for PS waste, Portland cement and QD waste respectively, followed by Ba with 360 mg/L for POFA and 24 mg/L for sand. Besides that, there are three (3) elements are not detected which is Ba in Portland cement, Fe in POFA and V in sand.

Table 7: Heavy metal concentration in petroleum sludge and raw materials

Heavy metals concentration	Formula	Concentration (mg/L)					
		Materials					Permissible limits USEPA (1996)
		PS	POFA	Portland cement	QD	Sand	
Copper	Cu	18	6	20	12	52	100
Lead	Pb	25.1	19	60	62	7	5
Zinc	Zn	189	34	132	151	11	500
Nickel	Ni	22.2	8	17	14	4	1.3
Barium	Ba	50.4	360	-	389	24	100
Chromium	Cr	120	12	69	16	9	5
Iron	Fe	6116	-	296	416	-	-
Arsenic	As	39.7	16	37	6	6	5
Vanadium	V	40.6	60	84	43	-	-
Highest		Fe (6116mg/L)	Ba (360mg/L)	Fe (296mg/L)	Fe (416mg/L)	Cu (52mg/L)	
Lowest		Cu (18mg/L)	Cu (6mg/L)	Ni (17mg/L)	As (6mg/L)	Ni (4mg/L)	

Meanwhile, Figure 9 shows the highest percentage of chemical composition for POFA, QD and sand are silica (SiO_2) which is 46.31%, 53.00% and 51.80% respectively. As for portland cement, the highest composition is the calcium oxide with 53.30%. Meanwhile, the lowest concentration results for portland cement and QD are titanium oxide with 0.23% and 0.51% respectively. Other than that, the lowest percentage for natural sand with 0.3% is potassium oxide (K_2O) whilst concentration of magnesium oxide (MgO) with 0.2% shows the lowest value in POFA.

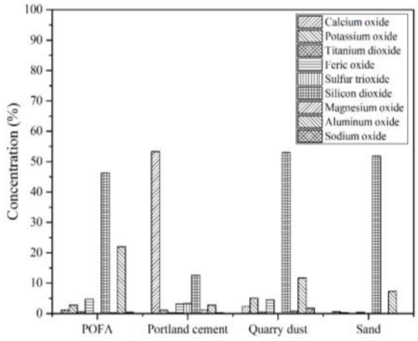

Fig. 9: Elemental composition of raw materials

From the results, it has clearly been seen that the most dominant of concentration in raw materials is calcium, silica and alumina. These elemental compositions that also known as C-S-H gel are the main materials that could enhance performance of the S/S matrices due to its pozzolanic reaction. In ASTM C 618 (2001) pozzolonic material defined as a material that contains siliceous and aluminous material by composition. Basically, a pozzolonic material has little or no cementing property, despite of the characteristic, when it has a fine particle size in the presence of moisture, it can react with calcium hydroxide at ordinary temperatures to provide the cementing property.The abundance of silica in POFA will generate a viable combination to create a good performance of S/S matrices. In depth, utilization of POFA improves resistance to chloride ion penetration (Chindaprasirt *et al.*, 2008; Awal *et al.*, 1998) enhances resistance to acidic environment (Tay, 1990) and sulphate attack (Jaturapitakkul *et al.*, 2007).

3.3 Results of Semi-Dynamic Leaching Procedure (SDLP)

Based on nine (9) heavy metals in this research, the highest concentration for heavy metals using SDLP was Fe with 0.229 mg/L for PS30% at T05, followed by zinc and lead with 0.0897 mg/L at T06 (PS30%) and 0.0868 mg/L at T06 (PS30%) respectively. In addition, most of the highest concentrations of heavy metals leach out from sample PS30% for example chromium (0.0304 mg/L), vanadium (0.0203 mg/L), arsenic (0.00442 mg/L) and nickel (0.000209 mg/L). Meanwhile, copper and barium concentration also shows higher at PS30% with 0.0569 mg/L and 0.0135 mg/L respectively. However, all concentrations for nine (9) heavy metals in this research are not passing the limit of USEPA. Thus, the elements satisfied the standard requirements. In this research, at the beginning stage of heavy metals characterization the result for PS raw materials showed concentration of Pb, Ni, Cr and As has exceeded the permissible limit that set by USEPA. Therefore, it shows that petroleum sludge can be treated using S/S matrices as the concentration of heavy metals by S/S matrices in SDLP leaching test are within the acceptable range limits. Figure 10 to Figure 18 shows that at the beginning of the leaching test, the concentration of heavy metals was in somewhat lower levels of release. Apart from that, Dermatas et al., (2004) mention that during the first 5 days of leaching testing by using deionised water would probably showed the lower result. Xue et al., (2016) discussed that the lower result in the beginning time is due to frequent replacement of the leachant. The value concentration of heavy metals shows rise up from T01 to T04 while peak value are shown in T05 and T06. Then at T07 the value of concentration start to drop due to the decreased of pH. According to Rachana et al., (2006), pH value affect the leaching, fixation and speciation of metals in S/S matrices. All data were interpreted in details in bar graphs as below. value for S/S matrices mix is slightly lower than the control matrices.

Figure 11 to Figure 18 shows the result of SDLP for various heavy metals.

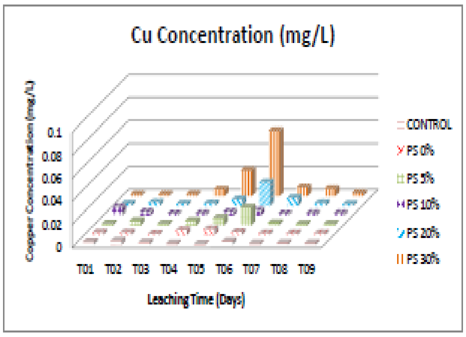

Fig 10: Cu concentration in SS matrices by using SDLP

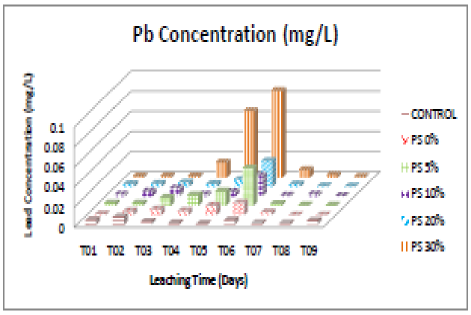

Fig 11: Pb concentration in SS matrices by using SDLP

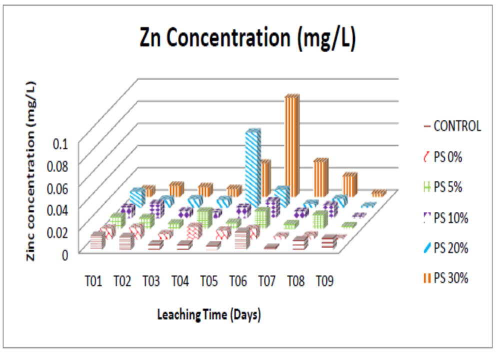

Fig 12: Zn concentration in SS matrices by using SDLP

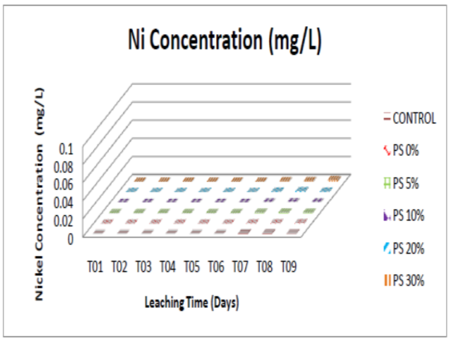

Fig 13: Ni concentration in SS matrices by using SDLP

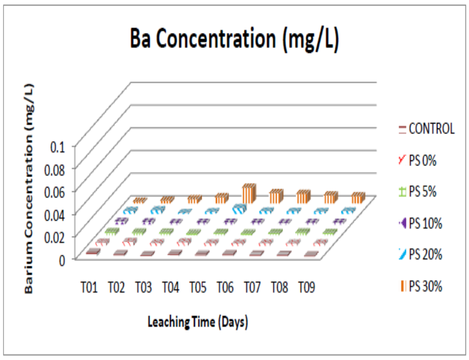

Fig 14: Ba concentration in SS matrices by using SDLP

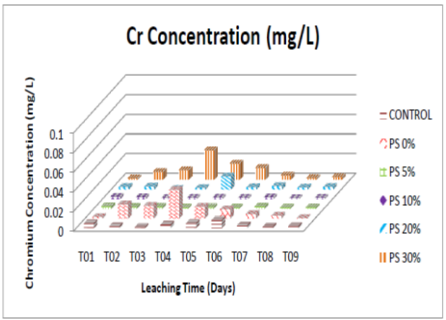

Fig 15: Cr concentration in SS matrices by using SDLP

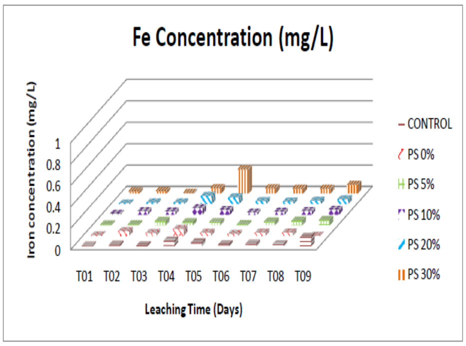

Fig 16: Fe concentration in SS matrices by using SDLP

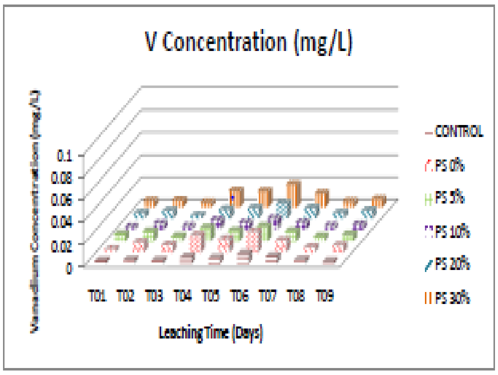

Fig 17: As concentration in SS matrices by using SDLP

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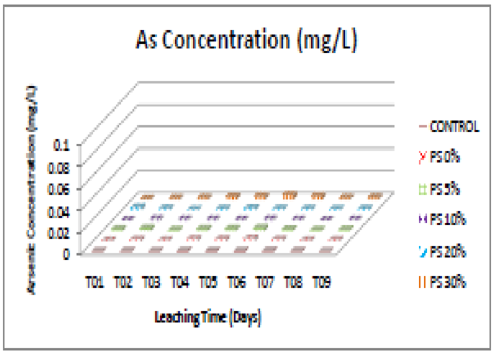

Fig 18: V concentration in SS matrices by using SDLP

3.4 Results of Static Leaching Test (SLT)

The result from Figure 19 to Figure 27 depicts that the highest concentration of heavy metals in S/S matrices detected from SLT was Fe on day 1 (T01) at PS30% with 0.273 mg/L, followed by Zn on day 1 (T01) at PS30% with 0.0533 mg/L. Other than that, the bar graphs below (Figure 19 to Figure 27) shows that all the concentration of heavy metals was higher on day 1 (T01) for example copper, lead, nickel, barium, chromium and arsenic with 0.0254 mg/L, 0.0200 mg/L, 0.00362 mg/L, 0.0236 mg/L, 0.0251 mg/L and 0.0360 mg/L respectively. In contrast with vanadium, the higher concentration was shown in T06 at PS30% with 0.0134 mg/L. However, all the results obtained showed that the values of concentrations for nine (9) heavy metals in this research are not passing the limit of USEPA and EPAV. Thus, the elements for SLT leaching test also satisfied the standard requirements. According to Kadir *et al.*, (2015), the lower concentration of heavy metals is due to the S/S process takes place when apply cement binders.

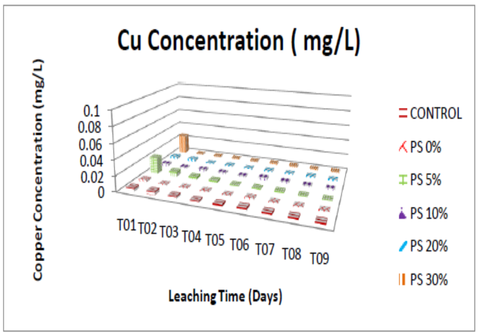

Fig 19: Cu concentration in SS matrices by using SLT

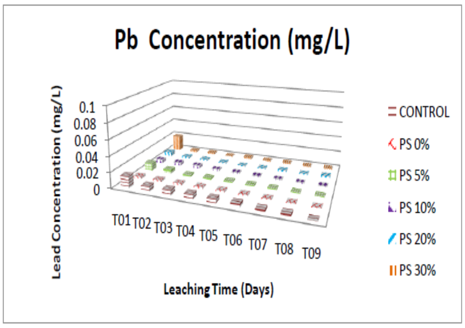

Fig 20: Pb concentration in SS matrices by using SLT

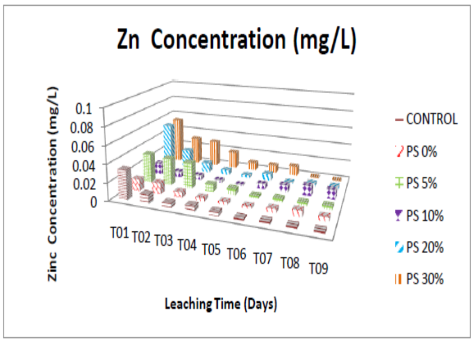

Fig 21: Zn concentration in SS matrices by using SLT

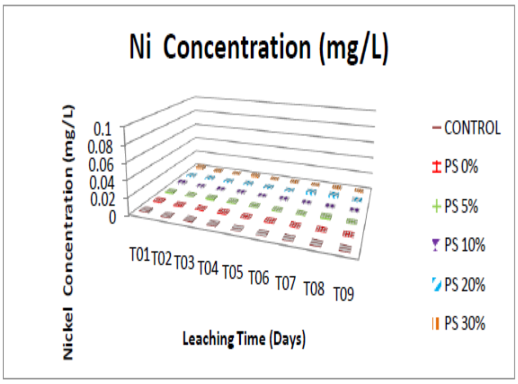

Fig 22: Ni concentration in SS matrices by using SLT

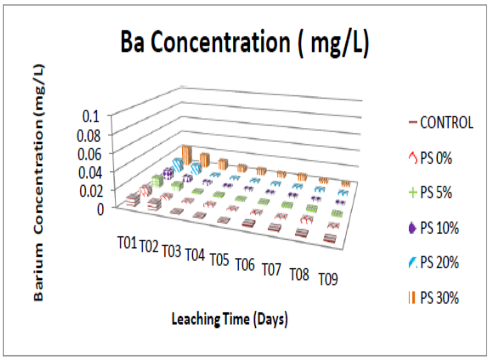

Fig 23: Ba concentration in SS matrices by using SLT

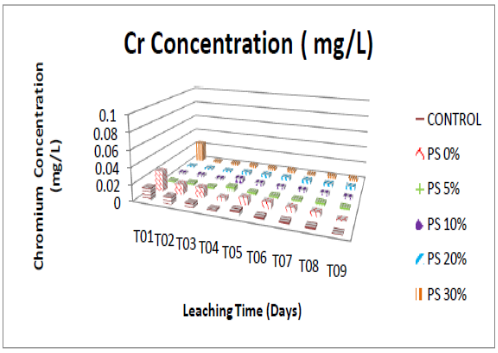

Fig 24: Cr concentration in SS matrices by using SLT

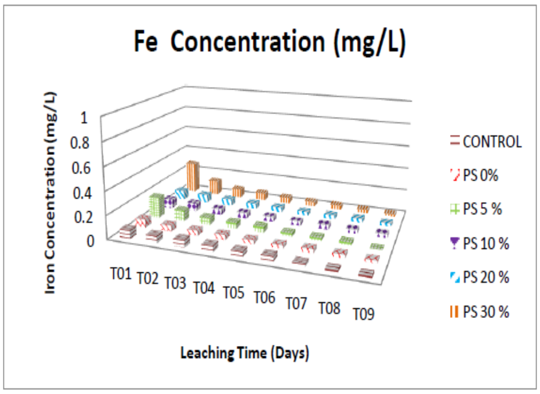

Fig 25: Fe concentration in SS matrices by using SLT

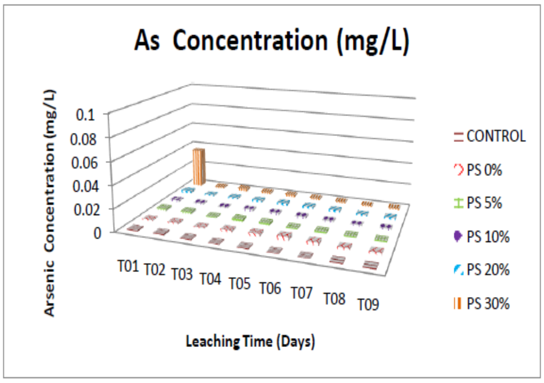

Fig 26: As concentration in SS matrices by using SLT

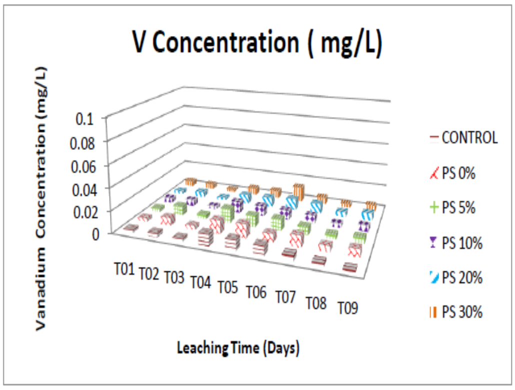

Fig 27: V concentration in SS matrices by using SLT

3.5 Effective diffusion coefficient (De) and leachability index (Lx)

The calculated results of the two parameters for SDLP and SLT leaching test are shown in Table 4.8. From the table, it shows the value of De and Lx for the different type of sample percentage and heavy metals. In Table 8, De was calculated from the first day of immersion until 63 days of immersion leaching times. The results indicated that the overall value of Lx for both leaching test are in a range larger than 8 which means that the S/S matrices in this study can be disposed of in the landfill.

The value of Lx for SDLP and SLT were in range of 8.0 to 11.38 and 9.90 to 11.67 respectively. The highest value of Lx for SDLP test were 11.38 at PS10% for copper with De 4.18E-12 whilst the lowest value was 8.00 with De value 1.11E-08 at PS 30% (Pb). Meanwhile, the highest value of Lx for SLT test were 11.67 at PS5% (As) and PS30% (Cu) respectively. The lowest value of Lx for SLT was 9.90 with 1.27E-10 (V) at PS0%. For comparison between SDLP and SLT, De for SLT are lower than SDLP and thus give a higher result in Lx for SLT. Xue *et al.*, (2016) investigated that the decrease in De under-acid conditions was due to a large amount of heavy metals being leached out thus resulting in a decrease in the leaching rate of heavy metals. In addition, most of the concentrations of heavy metals are leached out at first day of immersion as the pH initial for SLT test was acidic (pH 4.2). Meanwhile, the SDLP showed concentration result was higher after 3 days of immersion.

Table 8: De and Lx for SDLP and SLT

Samples	Heavy metals	Effective Diffusivity, De (cm2/s)		Leachability Index (Lx)	
		SDLP	SLT	SDLP	SLT
CONTROL	Cu	1.26E-11	1.17E-11	10.90	10.93
PS 0%		3.00E-11	4.53E-12	10.52	11.34
PS 5 %		3.02E-10	2.41E-12	9.52	11.62
PS 10 %		4.18E-12	1.06E-11	11.38	10.98
PS 20 %		3.35E-10	6.86E-12	9.48	11.16
PS 30 %		3.94E-09	2.15E-12	8.40	11.67
CONTROL	Pb	1.41E-11	3.79E-12	10.85	11.42
PS 0%		4.12E-10	9.19E-12	9.38	11.04
PS 5 %		5.33E-10	2.90E-12	9.27	11.54
PS 10 %		3.38E-10	6.05E-12	9.47	11.22
PS 20 %		1.90E-10	3.14E-12	9.72	11.50
PS 30 %		1.11E-08	2.47E-12	8.00	11.66
CONTROL	Zn	1.04E-11	2.53E-12	10.98	11.60
PS 0%		1.21E-11	5.78E-12	10.92	11.24
PS 5 %		1.71E-11	5.93E-12	10.77	11.23
PS 10 %		1.87E-11	7.06E-12	10.73	11.15
PS 20 %		5.42E-11	2.72E-12	10.27	11.57
PS 30 %		4.37E-10	3.96E-12	9.36	11.40
CONTROL	Ni	1.57E-11	4.18E-12	10.80	11.38

PS 0%		4.40E-11	6.27E-12	10.36	11.20
PS 5 %		2.78E-11	1.03E-10	10.56	9.99
PS 10 %		2.08E-11	5.76E-12	10.68	11.24
PS 20 %		4.91E-11	5.97E-12	10.31	11.22
PS 30 %		1.10E-10	4.13E-12	9.96	11.38
CONTROL	Ba	6.57E-12	4.73E-12	11.18	11.33
PS 0%		1.55E-11	3.28E-12	10.81	11.48
PS 5 %		1.09E-11	3.41E-12	10.96	11.47
PS 10 %		8.53E-12	3.33E-12	11.07	11.48
PS 20 %		2.16E-11	3.68E-12	10.67	11.43
PS 30 %		5.66E-10	4.10E-12	9.25	11.39
CONTROL	Cr	1.31E-11	3.72E-12	10.88	11.43
PS 0%		1.08E-09	3.87E-12	8.97	11.41
PS 5 %		9.98E-12	8.54E-11	11.00	10.07
PS 10 %		9.07E-12	6.02E-11	11.04	10.22
PS 20 %		7.82E-11	1.19E-10	10.11	9.93
PS 30 %		1.61E-09	2.33E-12	8.79	11.63
CONTROL	Fe	6.48E-10	8.46E-12	9.19	11.07
PS 0%		5.43E-10	6.50E-12	9.27	11.19
PS 5 %		3.94E-10	3.35E-12	9.40	11.48
PS 10 %		2.56E-10	7.77E-12	9.59	11.11
PS 20 %		2.64E-08	4.76E-10	9.00	11.13
PS 30 %		9.00E-11	2.93E-10	9.53	11.52

Table 8: (continued)

Samples	Heavy metals	Effective Diffusivity, De (cm2/s)		Leachability Index (Lx)	
		SDLP	SLT	SDLP	SLT
CONTROL	As	2.90E-11	1.28E-11	10.54	10.89
PS 0%		8.26E-11	1.14E-10	10.08	9.94
PS 5 %		2.09E-11	1.46E-11	10.68	10.84
PS 10 %		5.27E-11	2.98E-11	10.28	10.53
PS 20 %		6.85E-12	9.93E-12	11.16	11.00
PS 30 %		3.65E-10	2.14E-12	9.44	11.67
CONTROL	V	7.99E-11	9.84E-11	10.10	10.01
PS 0%		2.63E-10	1.27E-10	9.58	9.90
PS 5 %		3.49E-11	1.31E-10	10.46	9.88
PS 10 %		1.11E-10	5.42E-11	9.96	10.27
PS 20 %		4.76E-11	1.20E-10	10.32	9.92
PS 30 %		8.28E-11	3.34E-11	10.08	10.48

CONCLUSION

In conclusion, all the characteristics, sludge percentages and leachability of S/S matrices incorporated with (PS, POFA and QD) waste were determined. Based on the results obtained by XRF showed that the chemical composition of raw material for POFA and QD was high in silicon dioxide. The results revealed that the highest percentage of composition for POFA and QD was recorded with 46.30% and 53.0% respectively. Moreover, by using XRF, it was found that the concentrations of heavy metals for PS and QD were high in iron (Fe) with value 6116 mg/L and 416 mg/L respectively. In contrast with POFA, the highest heavy metals was in barium (Ba) with 360 mg/L. Meanwhile, all leachability result for SDLP and SLT satisfied the permissible limit that set by United State Environment Protection Agency (USEPA, 1996) and Environment Protection Agency Victoria (EPAV, 2009). Raw materials result by using XRF test showed concentrations of heavy metals are passing the permissible limit. However, by using S/S matrices in this research showed that all heavy metals concentration for Copper (Cu), Arsenic (As), Nickel (Ni), Lead (Pb), Vanadium (V), Iron (Fe), Zinc (Zn), Barium (Ba) and Chromium (Cr) were below 1 mg/L. As the highest concentration for heavy metals for both test were Fe, SLT recorded the highest value compared to SDLP. The SLT test used acid solution (pH 4.2) causing the heavy metals to leach out greater on the day 1 compared to SDLP which shows result higher on day 5 of immersion time. In this research, concentrations of heavy metals for SDLP were begun to decline from T07 while SLT showed the concentrations of heavy metals continuously decrease. Other than that, the overall result of Lx value for SDLP and SLT is between 8 to 11.38 and 9.90 to 11.67 respectively. From the result, it showed that all percentages of S/S matrices (Control, PS0%, PS5%, PS10%, PS20% and PS30%) could be disposed off to landfill. In addition, it proved that S/S matrices can be used in controlled utilization for example road base material, quarry rehabilitation and others as there are value of Lx has greater than 9.

As the Lx value is greater than 8, the petroleum sludge could be treated by using S/S matrices with designated a replacement thus enhancing the method for disposal of this hazardous waste.

Acknowledgement

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