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MOHD IKHMAL HAQEEM HASSAN<sup>©1</sup>, AESLINA ABDUL KADIR<sup>©1,4\*</sup>, NOR AMANI FILZAH MOHD KAMIL<sup>©1</sup>, NURUL NABILA HUDA HASHAR<sup>©1</sup>, NOOR AMIRA SARANI<sup>©1</sup>, BADARUDDIN IBRAHIM<sup>©2</sup>, KAHIROL MOHD SALLEH<sup>©2</sup>, MOHD MUSTAFA AL BAKRI ABDULLAH<sup>©3,4</sup>

# MECHANICAL PROPERTIES AND TOXICITY CHARACTERISTIC OF PETROLEUM SLUDGE INCORPORATED WITH PALM OIL FUEL ASH AND QUARRY DUST IN SOLIDIFICATION/STABILIZATION MATRICES

This paper discussed the treatment of Malaysian petroleum sludge by incorporating palm oil fuel ash (POFA) to replace Portland cement and quarry dust (QD) replaces sand in the solidification /stabilization (S/S) method. Preliminary studies, including chemical composition, heavy metal characterization, density test, compressive strength test, and toxicity characteristic leaching procedure (TCLP), were done to evaluate POFA and QD suitability in S/S matrices. The 10% replacement of POFA recorded a considerable density value ranging from 1500 kg/m<sup>3</sup> to 1660 kg/m<sup>3</sup>. As for S/S matrices containing petroleum sludge, the results indicate the possibility to of encapsulating the sludge in the matrices up to 10%. The highest strength of S/S matrices with petroleum is from PS5% samples with 15.61 MPa at 28 days. The toxicity characteristic of heavy metals from the S/S matrices was below the permissible limit set by USEPA. This investigation could be an alternative solution for petroleum sludge, POFA, and QD disposal and has excellent potential for replacing other treatment approaches employed at the advanced treatment stage for petroleum refinery effluents. *Keywords:* petroleum sludge; solidification/stabilization; palm oil fuel ash; quarry dust; waste utilization

#### 1. Introduction

Heavy metals waste usually needs solidification/stabilization (S/S) before landfill to lower the leaching rate. The main reason for the acceptance of S/S treatment used as it immobilizes and stabilizes hazardous substances [1]. There are two significant ways to deal with heavy metals contamination: a chemical transformation into a compound that reduces the potential hazard to tolerable human health levels and the environment (stabilization). The other way is to reduce the metal mobility through physical encapsulation of low-permeability material in the solidification process. Many contaminants can be trapped in an alkaline environment created by cement-based processes. The leaching from soils treated with these methods depends on the waste that remains in the cement pores or is chemically immobilized by the cement [2].

The compressive strength of cement paste containing pozzolan materials is contributed by hydration reaction, packing effect and pozzolanic reaction. Hydration reaction is the chemical between Portland cement and water as pozzolanic reaction silica compound and calcium hydroxide. The packing result is a proper arrangement of tiny particles that fill the voids and contribute to compressive strength increment [3,4].

Compressive strength is the main factor in deciding whether the solidified waste is put into a landfill or used as a construction material [5]. Additionally, [6] suggested that the minimum strength for waste disposal using S/S was above 0.34 MPa to meet landfill disposal limit regulatory.

On the other hand, the purpose of S/S is to achieve and maintain the desired physical properties and chemically stabilize or permanently bind contaminants [7,8]. S/S method is used to prevent or minimize hazardous compounds' release into the environment by producing a solid mixture, improving handling characteristics, decreasing surface area for contaminant transport, and reducing mobility contaminant transport and bonding the contaminants into a non-toxic form [9].

Due to the high concentration of toxic substances in the petroleum sludge, this sludge's improper disposal can pose severe threats to the receiving human health and environment. The petroleum sludge can disturb the soil's physical and chemi-

Corresponding author: aeslina.ak@gmail.com



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<sup>1</sup> UNIVERSITI TUN HUSSEIN ONN MALAYSIA, FACULTY OF CIVIL AND ENVIRONMENTAL ENGINEERING, 86400 PARIT RAJA, BATU PAHAT JOHOR, MALAYSIA

<sup>&</sup>lt;sup>2</sup> UNIVERSITI TUN HUSSEIN ONN MALAYSIA, FACULTY OF TECHNICAL AND VOCATIONAL EDUCATION, 86400 PARIT RAJA, BATU PAHAT JOHOR, MALAYSIA

<sup>&</sup>lt;sup>3</sup> UNIVERSITI MALAYSIA PERLIS, FACULTY OF ENGINEERING TECHNOLOGY (FETECH), 01000 KANGAR, PERLIS, MALAYSIA

<sup>&</sup>lt;sup>4</sup> UNIVERSITI MALAYSIA PERLIS (UNIMAP), CENTER OF EXCELLENT GEOPOLYMER AND GREEN TECHNOLOGY (CEGEOGTECH), MALAYSIA

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cal properties, leading to soil morphological change [10]. The attendant hazards may trigger processes that may have adverse effects on the ecosystem of such areas. Water used during crude oil extraction is contaminated and may contain varying organic matter, heavy metals, volatile hydrocarbons, and many other potentially toxic compounds [11].

Also, a study has proved that heavy metals exist in petroleum sludge [12]. Also, the results show the leachability and toxicity of this hazardous petroleum sludge encapsulated in building blocks. The heavy metal concentration in the block's leachate is within the allowable level recommended by the standard. In particular, waste materials used in building applications contribute to long-term sustainability while addressing waste management concerns [13].

Therefore, this study focuses on the S/S of Malaysian petroleum sludge by incorporating palm oil ash to replace cement in the S/S method. In addition, this study provides a sustainable solution by decreasing the sludge treatment cost by utilizing palm oil ash to replace cement and providing costless materials for the S/S method that may lead to construction applications. Treatment methods of S/S often give potential products with economic benefits, but also preserve resources and the atmosphere [14,15].

# 2. Materials and methods

The experimental program 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 (Fig. 1) has been chosen to be treated and was collected from an 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



Fig. 1. Petroleum sludge

total refining capacity of more than 440,000 barrels per day. 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  $110^{\circ}$ C [12].

Meanwhile, POFA was obtained from Kian Hoe Plantation Company at Kluang, Johor (Fig. 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  $800^{\circ}$ C to  $1000^{\circ}$ C. After procuring the ash, it was oven-dried at a temperature of  $110^{\circ}$ C to remove moisture. Further, it was sieved by using a 90 µm sieve to eliminate unburned fibres and improve its fineness as particle size plays a crucial role in pozzolanic reactivity. Hence, to enhance the pozzolanic reactivity, POFA must be ground to produce a smaller particle size, as suggested by [16].



Fig. 2. Palm oil fuel ash (POFA)

QD was received from Bina Kuari (K) Sdn. Bhd located in Kedah (Fig. 3). The quarry dust waste was dried and placed in the oven at 105°C. After drying, quarry dust waste was ground and crushed to make it easier in the sieving process and eliminate impurities. The waste criteria are dust (75  $\mu$ m) that complies with Malaysian Standard MS 522: Part 1: 1989.



Fig. 3. Quarry dust (QD)

#### 2.2. Raw materials characterization

The characterization of the main composition and heavy metals in raw materials such as petroleum sludge, palm oil ash, quarry dust, and cement is essential for determining the materials' components. Therefore, the main composition of raw materials of cement, sand, POFA, and QD was examined using X-ray fluorescence (XRF) and to get the result. Sample preparation needs to be done by preparing the pressed pellet.

# 2.3. Density and compressive strength

The density of all experimental samples was conducted according to BS EN 12390-7:2009. Cube samples with size  $100 \times 100 \times 100$  mm that has been cured for 7, 14 and 28 days were used in this test. Meanwhile, the compressive strength test in this research was according to ASTM-C109, a standard test method for compressive strength of hydraulic cement mortars. The cube samples with an average of three samples for each proportion were tested using a compression testing machine of 2000 kN capacity with 0.75 kN/s of loading rates.

# 2.4. Toxicity characteristic leaching procedure (TCLP)

Specifically, this method (USEPA, TCLP 1311) is designed to determine the mobility of organic and inorganic contaminants in the liquid, solid and multiphase wastes. In this research, the S/S matrices sample were crushed into four portions finer than 9.5 mm for the analysis. The extraction fluid used for the extraction depends on the alkalinity of the waste material. In this research, 5.7 mL of glacial acetic acid was diluted with reagent water to a 1 L. The pH of this fluid is  $2.88 \pm 0.5$  when correctly prepared. After determining the extraction fluid, the samples were prepared in screw-capped polyethylene bottles filled with crushed concrete samples and leaching fluid at the ratio of 1:20. The bottles were agitated at 30 rpm for 18 hours in an end-overend manner. The leachate collected was then filtered using 0.7 µm glass fibre filters preserved the sample before being analyzed using Inductively Coupled Plasma Mass Spectrometry (ICP-MS) to determine the dissolved metals.

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

The preliminary investigates the optimum value for palm oil ash and quarry dust replacement in S/S matrices. This stage is vital to finding the optimum percentage of POFA and QD replacement incorporated with petroleum sludge in S/S matrices. Two types of S/S matrices were produced in the preliminary stage of this research. The matrices were used to study the effect of POFA replacement in cement and investigate the effect of quarry dust replacement in the sand. As such, the investigation begins with the replacement of palm oil fuel ash (POFA) to cement with 10%, 20%, 30%, and 40%, respectively. The S/S mortar was tested compared to the control mix with a ratio of 1:2.75 of cement to fine aggregates.

The standard w/c ratio for compressive strength is 0.485 by following the ASTM C109. Next, the exact percentages were used as a sand replacement using quarry dust which is 10%, 20%, 30% and 40%. Mix proportions are shown in TABLE 1 and TABLE 2.

TABLE	1
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Details of mix proportion for palm oil fuel ash replacement

Samples	Percentage of POFA replace-	Binder (kg)		Fine Aggregates (1:2.75 wt binder) (kg)		Water to binder	
	ment (%)	Cement	POFA	Sand	Quarry dust	ratio (%)	
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 2

Details of mix proportion for quarry dust replacement

Samples	Percentage of POFA	Binder (kg)		(1:2.	gregates 75 wt r) (kg)	Water to binder
	replace- ment (%) Cement POFA		Sand	Quarry dust	ratio (%)	
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

The actual mix proportion of petroleum sludge incorporated with POFA and QD using the S/S method was developed after the optimum percentages of POFA and QD replacement have 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 3 shows the mixed proportion of the S/S matrices.

#### TABLE 3

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

Samples	Percentage of Petroleum	Binder (kg)		(kg) Fine Aggregates (1:2.75 wt binder) (kg)		Water to binder
	sludge (%)/(kg)	Cement	POFA	Sand	Quarry dust	ratio (%)
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 by organic and metal analysis. The properties of petroleum sludge obtained from Petronas Refinery is tabulated in TABLE 4. The sludge has a moisture content of 34.6 % and a specific gravity of 0.994. The loss on ignition at 900°C is 45.9%. pH for this petroleum sludge was recorded in an acidic condition at pH 6. Other than that, the organic fraction of petroleum typically consisted of aromatic and semi-volatile compounds. These compounds are detected in the form of TPH of the organic fraction. with 389 ppm and 360 ppm, respectively. Meanwhile, the elements such as Cu (12 to 52 ppm), Pb (7 to 62 ppm), Zn (11 to 151 ppm), V (43 to 84 ppm), Ni (4 to 17 ppm), Cr (9 to 69 ppm) and As (6 to 37 ppm) are recorded from the matrices.

TABLE 6 shows the highest percentage of chemical composition for POFA, QD, and sand is silica  $(SiO^2)$  which is 46.30%, 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<sub>2</sub>O), whilst the concentration of magnesium oxide (MgO) with 0.2% shows the lowest value in POFA.

composition of raw materials

TABLE 6

Duonoution	Sludge Characterization					
Properties	Parameter	Value	Method			
	Moisture Content	34.6%	TC WI :2003 (E)			
Chemical/	Specific Gravity	0.994	BS 1377: Part 1: 1990			
Physical Properties	Loss of Ignition @900°c	45.9%	TC WI: 2003 (E)			
	рН	6	USEPA SW-846:9045D			
	ТРН С6-С9	984 mg/kg	USEPA 8260B			
Organic	TPH C10-C14	56300 mg/kg	USEPA 8015B			
Fraction	TPH C15-C28	188000 mg/kg	USEPA 8015B			
	ТРН С29-С36	47100 mg/kg	USEPA 8015B			

Petroleum sludge characterization

#### 3.2. Heavy metals and elemental composition

TABLE 5 shows eight heavy metals in petroleum sludge, POFA, Portland cement, QD, and sand. The highest element recorded in petroleum sludge is Zn with 189 mg/L, followed by Cr with 120 mg/L, and Ba with 50.4 mg/L. Other petroleum sludge elements such as Cu, Pb, V, Ni and As were recorded in the range of 18 mg/L to 40.6 mg/L. Meanwhile, for the raw materials, the highest element recorded is Ba in QD and POFA

TABLE 5

TABLE 4

Heavy metal concentration in petroleum sludge and raw materials

	Concentration (mg/L)							
Heavy metals	Materials							
concentration	Petroleum sludge	POFA	Portland cement	QD	Sand			
Copper	18	6	20	12	52			
Lead	25.1	19	60	62	7			
Zinc	189	34	132	151	11			
Vanadium	40.6	60	84	43				
Nickel	22.2	8	17	14	4			
Barium	50.4	360		389	24			
Chromium	120	12	69	16	9			
Arsenic	39.7	16	37	6	6			

Chaminal		Concentration (%)				
Chemical composition	Formula	POFA	Portland Cement	Quarry Dust	Sand	
Calcium oxide	CaO	1.09	53.30	2.35	0.58	
Potassium oxide	K <sub>2</sub> O	2.81	1.07	5.05	0.30	
Titanium dioxide	TiO <sub>2</sub>	0.66	0.23	0.51		
Ferric oxide	Fe <sub>2</sub> O <sub>3</sub>	4.76	3.14	4.52	0.39	
Sulfur trioxide	SO3		3.26			
Silicon dioxide	SiO <sub>2</sub>	46.30	12.60	53.00	51.80	
Magnesium oxide	MgO	0.20	1.16	0.80		
Aluminum oxide	Al <sub>2</sub> O <sub>3</sub>	22.00	2.79	11.70	7.30	
Sodium oxide	Na <sub>2</sub> O	0.45	0.19	1.75		
Highest		SiO <sub>2</sub> (46.30)	CaO (53.30)	SiO <sub>2</sub> (53.00)	SiO <sub>2</sub> (51.80)	
Lowest		MgO (0.20)	TiO <sub>2</sub> (0.23)	TiO <sub>2</sub> (0.51)	(0.30) (0.30)	

From the results, it has been seen that the most dominant concentration in raw materials is calcium, silica, and alumina. These elemental compositions, known as C-S-H gel are the primary materials that could enhance the performance of the S/S matrices due to their pozzolanic reaction. Referencing ASTM C 618, pozzolanic material is containing siliceous and aluminous by composition. A pozzolanic material has little or no cementing property. Despite 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 generates a possible combination to create a good performance of S/S matrices. In-depth utilization of POFA improves resistance to chloride ion penetration, enhances resistance to an acidic environment, and sulphate attack [17-19].

#### 3.3. Density and compressive strength

# 3.3.1. Density

Fig. 4 and Fig. 5 shows the density of different percentages of POFA (replaced cement) and QD (replaced sand) with 0% (control), 10%, 20%, 30%, and 40% at different ages (7, 14 and

28 days). The values of density for control samples range from 2100 kg/m<sup>3</sup> to 2260 kg/m<sup>3</sup>. Fig. 5 shows that matrices' density with POFA replacement by different percentages of 10%, 20%, 30% and 40% resulted in much lower than the control samples. The replacement of POFA by 10% gave a considerable value of density value ranging from 1500 kg/m<sup>3</sup> to 1660 kg/m<sup>3</sup>. The results show that a higher percentage of POFA replacement decreases the density of the solidified matrices. This hypothesis is supported by [20], which used oil palm fibre reinforced mortar utilizing POFA as a complimentary binder. Due to POFA having a lower density, the mortar mix's density value is slightly lower than the control mix.



Fig. 4. Density of matrices with different percentages of POFA replacement in cement at 7, 14 and 28 days

For quarry dust as a sand replacement, similar trends of density values are shown in Fig. 5. The density value of the QD matrices was recorded slightly lower than the control samples. The replacement of QD in natural sand by 20% gave a value of density ranging from 2120 kg/m3 to 2167 kg/m<sup>3</sup>. Besides, 40% of QD replacement recorded the lowest density value with an average of 2073 kg/m<sup>3</sup> to 2087 kg/m<sup>3</sup>.



Fig. 5. Density of matrices with different percentages of QD replacement in sand at 7, 14 and 28 days

Fig. 6 shows the density of the S/S of different petroleum sludge percentages (0%, 5%, 10%, 20% and 30%). The results showed that the densities slightly decreased with the increment

of petroleum sludge. The highest density of S/S matrices of petroleum sludge was recorded from PS 5% with 2037 kg/m<sup>3</sup> at 7 days, and the lowest density was obtained from PS 30% with 1793 kg/m<sup>3</sup>. One of the factors that lead to the lower density is the replacement of the POFA. It shows that 10 % of POFA replacement decreased the solidified matrices' density [20].



Fig. 6. Density of SS matrices of petroleum sludge at 7, 14 and 28 days

### 3.3.2. Compressive strength

The solidified matrices were tested at 7 days, 14 days and 28 days. From the observation based on the control sample, the compressive strength develops over curing time. The compressive strength results for different percentages of POFA with 0% (control), 10%, 20%, 30%, and 40% are shown in Fig. 7. The control samples without POFA and QD replacement showed the highest compressive strength value for 7, 14 and 28 days.



Fig. 7. Compressive strength with different percentages of POFA replacement in cement at 7, 14 and 28 days

On 28 days, 27.75 MPa was recorded as the highest value of compressive strength achieved by the control sample while followed by 10% of POFA replacement (22.60 MPa), 20% (18.13 MPa), 30% (11.62 MPa) and 40% (9.35 MPa) respectively. Compared to control samples, compressive strength development for POFA replacements was slightly slower. The

difference in strength recorded at 7 days to 28 days can be distinctly proved. In comparison to the control sample, the POFA replacement by 10%, 20%, 30% and 40% showed a decrease in compressive strength value with 21.4%, 36.93%, 59.58% and 67.48% respectively.

Additionally, as suggested by [6], all the samples were above the landfill disposal limit regulatory, higher than 0.34 MPa at 28 days. Nevertheless, only 10% of POFA replacement in solidified matrices is above the comparative mortar limit of 20 MPa on 28 days.

On the other hand, Fig. 8 shows the compressive strength results for different percentages of QD as a partial sand replacement in solidified matrices. The strength for 20% of QD replacement recorded is at its peak on day 28, with 24.91 MPa followed by 10% (23.75 MPa), 30% (22.84 MPa) and lastly 40% (13.40 MPa) respectively. The 20% of QD replacement is the most viable to imitate the control sample's compressive strength. However, compared with the control sample, the compressive strength for 20%, 10%, 30% and 40% of QD replacement is decreased by 10.23%, 14.44%, 17.69%, and 50.28%, respectively. These fluctuated values of compressive strength is quite significant as fine particles may seal voids in the paste and act as precipitation sites for the hydration products, resulting in increased matrices' strength. This phenomenon is known as a filler effect. However, if high amounts of fines are added, the bigger aggregate grains are further separated by the smaller particles and reduce the filler effect [21].



Fig. 8. Compressive strength with different percentages of QD replacement in cement at 7, 14 and 28 days

Meanwhile, Fig. 9 depicts the compressive strength for S/S of petroleum sludge (0%, 5%, 10%, 20%, 30%) at different ages. respectively. The figure shows that the compressive strength of S/S matrices decreased with the increase of petroleum sludge. It also shows that the strength of S/S matrices was increasing day by day. The highest strength of S/S matrices with petroleum is from PS5% samples with 15.61 MPa at 28 days. It shows that 5% of petroleum sludge is suitable to be treated in S/S matrices. Besides that, PS30% demonstrated the lowest compressive strength with 1.45 MPa at 28 days. Hence, the samples met the requirement for the S/S matrices to be disposed of at the landfill, which is above 0.35MPa.



Fig. 9. Compressive strength SS matrices of petroleum sludge at 7, 14 and 28 days

# 3.4. Toxicity characteristic leaching procedure (TCLP)

Fig. 10 compares heavy metal concentrations for the control sample with S/S matrices replaced with different percentages of POFA(10%, 20%, 30% and 40%) by using the TCLP method. From the observation, the Ba concentration seems to have the highest reading of leaching for all samples. The highest peak of Ba leaching was from POFA40% at 7 days (0.8410 mg/L), followed by POFA40% at 14 days (0.7420 mg/L) and the lowest from the control sample at 28 days (0.1060 mg/L). The highest value of Zn concentration was recorded in POFA30% (0.2500 mg/L, 0.2440 mg/L and 0.2040 mg/L at 7, 14 and 28 days respectively) and POFA40% (0.3270 mg/L, 0.3100 mg/L and 0.2930 mg/L at 7, 14 and 28 days respectively).

Moreover, the Cr concentrations in both control sample (0.0497 mg/L, 0.0359 mg/L and 0.0248 mg/L at 7, 14 and 28 days respectively) and POFA40% (0.0421mg/L, 0.0304 mg/L and 0.0282 mg/L at 7, 14 and 28 days respectively) were recorded the higher value compared to the other heavy metals. Meanwhile, the similar patterns showed in V concentration for both samples with value (0.0247 mg/L, 0.0231 mg/L and 0.189 mg/L at 7, 14 and 28 days respectively) for control sample and (0.0463 mg/L, 0.0341 mg/L and 0.0189 mg/L at 7, 14 and 28 days respectively) for POFA40%. On the other hand, Ni concentration was recorded highest from POFA40% at 7 days (0.0632 mg/L) and the lowest from POFA10% at 28 days (0.0103 mg/L). For Cu, As and Pb concentrations, the value was marginally low throughout the TCLP test. According to [22], it is suggested that As containing soils posed very low environmental risks and qualified as non-hazardous materials for landfill disposal. An interlocking framework of cement hydration products surrounding the S/S solids could reduce the mobility of As to the solidified matrices' encapsulation effect.

Referring to Fig. 11 shows the comparison of heavy metal concentrations for the control sample with samples replaced with different percentages of QD (QD10%, QD20%, QD30%, and QD40%) using TCLP. The results show that the Ba concentration was the major heavy metal leached for all samples with QD40%



Fig. 10. Comparison of heavy metal concentrations for different percentages of POFA replacement at 7, 14 and 28 days



Fig. 11. Comparison of heavy metal concentrations for different percentages of QD replacement at 7, 14 and 28 days

at 7 days recorded with 0.6850 mg/L as the highest value while QD10% at 28 days was the lowest value at 0.1030 mg/L. The concentration may be high as the QD replacement is the highest compared to the other samples. The Zn concentrations for QD10%, QD20%, QD30% and QD40% at 7 days (0.0397 mg/L, 0.0389 mg/L, 0.0568 mg/L and 0.0890 mg/L respectively) were slightly higher than the control sample (0.0188 mg/L). Meanwhile, the lowest heavy metals concentration recorded at QD10% at 28 days and QD40% at 28 days with the same value, 0.0101 mg/L. The same trends were observed for V concentrations from all samples with QD40% the highest value (0.0338 mg/L). The Pb concentration was recorded highest from the control sample with 0.0019 mg/L and the lowest from QD10%, QD20% and QD40% with 0.0004 mg/L. Other heavy metals such as Cu and As were slightly low in all the matrices and still permissible compared to USEPA. The S/S processes reduce heavy metal mobility due to the adsorption, encapsulation or precipitation effect in the solidified matrix. The effect of the leachate's final pH can be partly attributed to the stability of heavy metal [22].

Fig. 12 depicts the leaching of heavy metals for S/S of petroleum sludge with different percentages (0%, 5%, 10%, 20% and 30% of petroleum sludge) using TCLP. The results show the Zn and Ba concentrations were the most abundant metals leached from the samples. The highest amount of Zn was recorded from PS30% with 1.9204 mg/L at 7 days, followed by PS20% with 1.8990 mg/L. However, these values are considered low concentration compared to permissible limits set by USEPA, which is 500 mg/L of Zn concentration. Besides that, Ba recorded the second-highest heavy metals that leached out from all the samples ranging from 1.6650 mg/L (PS30%) to 0.1060 mg/L (control). Meanwhile, the lowest heavy metals concentration, which is Pb recorded from a control sample at 28 days with 0.0005 mg/L. Cu also demonstrates the lowest value with 0.0009 mg/L from the control sample at 28 days. Pb and Cu were less leached out from all the samples due to the minor abundance in the petroleum sludge and raw materials themselves. From the results, the heavy metals concentrations for all S/S samples were within the limit of USEPA.



Fig. 12. Comparison of heavy metal concentrations for different percentages of petroleum sludge matrices at 7 14 and 28 days

# 4. Conclusion

From all the results obtained, all the values are comparable with control samples (without any replacement materials) and under the permissible limits set by USEPA. Thus, the utilization of by-product waste from industry in the S/S method could be a viable solution for waste disposal problems and become an innovation in the construction industry's sustainable development approach.TCLP was carried out to measure the leachability of heavy metals concentrations in s/s matrices. The heavy metals selected in this study are Copper (Cu), Arsenic (As), Nickel (Ni), Lead (Pb), Vanadium (V), Chromium (Cr), Zinc (Zn) and Barium (Ba). The highest leaching of Zn and Ba were recorded compared to the other heavy metals as this heavy metal concentration reading is already high in the raw materials itself.

On the other hand, the toxicity characteristic of the heavy metal concentrations for all S/S matrices in TCLP was below the permissible limit set by USEPA. These findings have been approved according to their characterization, properties, such as the comparable compressive strength during the curing period and acceptable density and toxicity characteristics. Replacement of the raw materials not only could decrease the enormous quantities of palm oil fuel ash and quarry dust disposal towards the environment but mainly could treat hazardous waste such as petroleum sludge.

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# REFERENCES

- O. Malliou, M. Katsioti, A. Georgiadis, A. Katsiri, Cement Concrete Comp. 29 (1), 55-61 (2007).
- [2] K. Rahele, M. Mehrdad, R. Somayeh A. Ehsan, G. Farshid M. Mostafa, Environ. Pollut. 263, (2020).
- [3] I. Navarro-Blasco, A. Duran, R. Sirera, J.M. Fernández, J.I. Alvarez, J. Hazard Mater. 260, 89-103 (2013).
- [4] N. Jain, ISRN Civil Eng. 2011, 1-6 (2011).
  DOI: https://doi.org/10.5402/2011/183158
- [5] G. Salihoglu, V. Pinarli, N.K. Salihoglu, G. Karaca, Journal of Environmental Management, 85 (1), 190-197 (2007).
- [6] C.Y. Yin, W.S.W. Ali, Y.P. Lim, J Hazard Mater. 150, 413-418 (2008).
- [7] Q.Y. Chen, M. Tyrer, C.D. Hills, X.M. Yang, P. Carey, Waste Manage. 29 (1), 390-403 (2009).
- [8] S.P. Dunuweera, R.M.G. Rajapakse, Adv. Mater. Sci. Eng. (2018).
- [9] A. Tuncan, M. Tuncan, H. Koyuncu, Waste Manage Res. 18, 489-505 (2000).
- [10] S.J. Robertson W.B. McGill, H.B Massicotte, P.M. Rutherford, Biol. Rev. 82, 213-240 (2007).
- [11] G. Hua, J. Li, G. Zeng, J. Hazard Mater. 261, 470-490 (2013).
- [12] O.A. Johnson, Int. J. Appl. Eng. Res. 10 (24), 45479-45481, (2015).
- [13] R.M. Abousnina, A. Manalo, W. Lokuge, Procedia Eng. 145, (2016).
- [14] N.Asim, M.Badiei, T. Marzieh, M. Mohammad, A. Masita, A.G. Mohammad, S. Gasaymeh, K. Sopian, J. Clean. Prod. 284, (2021).
- [15] G. Hu, H. Feng, P. He, J. Li, K. Hewage, R. Sadiq, J. Clean. Prod. 251, (2020).
- [16] W. Kroehong, S. Theerawat, Proc Eng. 14, 361-369 (2011).
- [17] A.S.M.A. Awal, ASM, M.W. Hussin, Proc. Eng. 14, 2650-7 (2011).
- [18] J.H. Tay, K.Y. Show, Resour, Conserv. and Recy. 13, 27-36 (2005).
- [19] J.C Jaturapitakkul, K. Kiattikomol, W. Tangchirapat T. Saeting, Constr. Build. Mater. 21 (7) 1399-1405 (2007).
- [20] A.N. Raut, C.P. Gomez, Constr. Build. Mater. 126, pp. 476-483.
- [17] Q.Tang, Y. Liu, F. Gu, T. Zhou, Adv. Mater. Sci. Eng. 2016, 1-10 (2016).
- [22] J. Li, J. C.S Poon. Chemosphere, 173, (2017).
  DOI: https://doi.org/10.1016/j.chemosphere.2017.01.065