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PHYSICO-CHEMICAL PROPERTIES OF PALM OIL FUEL ASH AS COMPOSITE SORBENT IN KAOLIN LANDFILL LINER SYSTEM

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ABSTRACT
This paper presents information on the physico-chemical properties of palm oil fuel ash (POFA) a biomass residue, while justifying its use as composite sorbent when pre-mixed with kaolin, for the purpose of designing sanitary landfill liner. Physical analysis conducted includes particle size distribution and density tests for the ground ash. Chemical tests include Energy Dispersive X-Ray diffractometer (XRD), Energy Dispersive X-ray Fluorescence (EDXRF). Other chemical tests conducted are the specific surface area (SSA); using Particle size analyzer (PSA), scanned electron microscope (SEM), mass loss on ignition (LOI), at 440 $\pm$ 25 ^{0}C, for a period of 4 h. The pH-value for POFA- aqueous environment was also determined. Quantity of Silica oxide (SiO_2) present in the ground ash was found to be 52.35 %. The summation of SiO_2, Al_2O_3, and Fe_2O_3 present in the POFA was approximated to 72 %, while the inherited calcium oxide (CaO-lime) was approximated to 12 %. Both percentages classify the POFA as a Class C fly ash. Exchangeable cations detected include Na^{2+}, K^{2+}, Ca^{2+} and Mg^{2+}. Morphological analysis conducted also shows that the ground POFA was amorphous in structure. From these properties inferences were anticipated on the expected chemical reactions of the kaolin-POFA mixture for the design of composite sanitary landfill liner. The novel material is hoped to provide a more efficient chemical trapping mechanism of toxic heavy metal ions released from solid wastes that are been dumped in landfills. Conclusively, it is anticipated that POFA a bio-sorbent may be suitably used as composite sorbent for improvement on kaolin designed as lower component of engineered sanitary landfill liner.

***Keywords*:** Physico-chemical properties; kaolin; POFA; landfill liner; leachate

1. INTRODUCTION

1.1 Background

Colossal quantities of solid waste generation during palm oil production include pressed palm mesocarp fiber (PMF) and palm kernel shell (PKS). Financial and environmental impact of these bio-residues include high cost of disposal in landfill or direct open dump and socio-ecologically unaesthetic respectively. Highlighted detrimental impacts and many more call for an urgent and alternative use of the wastes. It has been confirmed that PMF and PKS are recycled as sources of renewable energy, to produce boiler steam use in palm oil processing firms, and for generating electricity in thermal power plants (Abdul et al., 2011; Chai et al., 2011; Sulaiman et al., 2011; Wunchock et al., 2011a, b; Shuit et al., 2009, Weerachart et al., 2009; Chun et al., 2008). The by-product from the combustion of PMF and PKS at elevated temperature in the range of 800 to 1000 ^{0}C is often referred to as POFA. Large quantity of POFA generation are witnessed in transition countries like Malaysia, Indonesia, and Thailand, where palm oil exportation contributes substantially to the country's revenue generation (Faisal et al., 2011; Chai et al., 2011; Sulaiman et al., 2011; Wunchock et al., 2011a, b; Yoon Lin et al., 2011; Siti Shawalliah et at., 2010; Weerachart et al., 2010; Abdullah et al., 2009; Chun et al., 2009). Hence, POFA as solid waste generation call for urgent adaptive technological approaches as panacea to alternative use the ash may be re-use, at the same time benefiting the nations. Else, POFA may constitute potential source of environmental pollution and endangering human health. Another problem that may arise is that the waste may serve as impedance to the global yarning for "zero waste" generation. More recently, studies have shown that POFA may be used as either binary or ternary supplementary cementitious material in concrete production (Abdul et al., 2011; Mohammad et al, et al., 2011; Wunchock et al., 2011; Weerachart et al., 2009; Chai et al., 2007; Chindaprasirt et al., 2007; Weerachart et al., 2007). In addition, Mei et al. (2009) have established the potential benefit of using POFA as bio-sorbent in waste water treatment. In addition, Chun et al. (2008) have partially replaced cement with POFA and used the binary cementitious matrix as sludge stabilizing agent. Taking advantage of its porous structure, Nor Fatiha et al. (2005) have also used POFA as an active adsorbent for flue gas desulfurization. Hitherto, studies documented specifically on the incorporation of POFA as mineral additive in the design of landfill geo-lining system are obviously lacking. The authors are of the opinion that a strong reason might be due natural restriction in suitable climatic and vegetation conditions for palm oil plantation, thus impeding

POFA generation at wider spread as agro-based solid waste material. Chemical compositions of the agro-waste ash have shown that it contains high amounts of aluminosilicate compounds; in fact, reasonable quantity of calcium oxide are present in the ash as inherited lime contents (Abdul et al., 2011; Chai et al., 2011; Wunchock et al., 2011a,b). In addition, amorphous silica from POFA is of benefit in various environmental engineering endeavours; it may assist in providing adequate negatively charged sites required for an enabling environment during cation exchange reaction with ions of toxic heavy metals present in waste waters (Foo et al., 2009; Rejini et al., 2009).

Against this background, the main objective of the study is to critically examine the physico-chemical properties of POFA, and examine how it can be applicable as co-sorbent in kaolinitic clay landfill liner system. The dysfunctional attenuation properties of kaolin have been established elsewhere (Cynthia and Raymond, 2002; Daniel, 1981; O'Flaherty, 1976). It is hoped that the use of POFA in complementing kaolin for the design of composite geo-liner system will not only improve the geotechnical properties, but also enhances its chemical occlusion efficiency.

2. EXPERIMENTAL PROGRAM

2.1. Palm oil fuel ash

POFA used for the study was collected via two shipments from Tai Tak oil milling firm in Kota Tinggi, Johor State of Malaysia Peninsula. The factory utilizes PMF and PKS for the co-firing process of the two installed turbine operating boiler systems. The PMF and PKS serves as means of generating clean and eco-friendly energy for the oil processing operations. The oil mill co-fire both wastes materials at an average temperature of 625 ℃ . However, typical inlet temperature utilized in biomass boiler mills and thermal power plants ranges from 800 to 1000 ^{0}C (Wunchock et al., 2011a). Although, Mohd Warid & Khairunisa (2009), reported that high-grade amorphous silica may be produced when bio-wastes are burnt at control temperatures lower than 700 ^{0}C under oxidizing conditions. The physical properties of the as-received and ground POFA are presented in Table 1.

2.2. Particle size analysis and specific surface area of fine ground POFA

Prior to particle size gradation test, two stage pulverization processes were performed on the sieved ash via modified Los Angeles abrasion test machine containing 13 stainless steel rods (479 mm and 12.57 mm average length and dia. respectively), and a bench-top Pulverisette 6 grinder respectively. After grinding, the range of particle sizes and specific surface area (SSA) were analyzed via bench-top Particle size analyzer (PSA-CILAS 1180). The particle density analysis of the ground POFA was determined as specified in the BS 1377: Part 4 (BSI, 1990). Results obtained as average value on three replicate specimens are presented in Table2.

Table 1 Physical characteristics of the raw and processed POFA

Physical properties	Appearance before ignition	Appearance after ignition	Texture	shape
Raw POFA	dark spongy	porous, grayish	hard, gritty, light, cellular	irregular
Ground POFA	grayish, powdery	brownish	Powdery	round

Table 2 Particle size distribution, specific surface area and average density of POFA subjected to 2 stage grinding

Mode of grinding	Grinding stage	Particle dia. when 10 % of total particles were captured(μm)	Cumulative Particle dia. when 30 % of total particles were captured(μm)	Cumulative Particle dia. when 60 % of total particles were captured(μm)	Specific surface area (cm^2/g)	Average particle density, ρ_s (Mg/m^3)
Los Angeles Abrasion	1	2.99	14.34	39.57	4288.94	2.48
Pulverisette 6	2	2.30	8.56	26.94	5405.40	

2.3. Surface morphological structure

Scanned electron morphological (SEM) structures for the raw and powdered POFA were examined via Energy Dispersive X-Ray Analyzer (EDX-JEOL JSM_6380LA). In the set up, aliquot of each test coupon was mounted on stubs and gold-coated to make them electrically conductive. Fig.3a-c shows morphological structures revealed for raw ash passing through 150μm, 75μm, 38μm test sieves; Fig.3d shows structure when raw ash was blended and subjected to two stage pulverization.

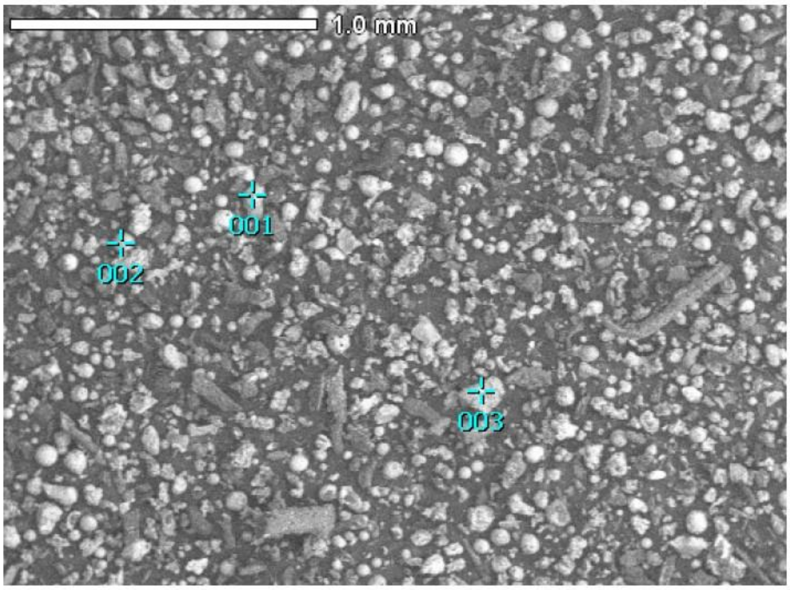


Figure 1a Morphological structure for raw ash passing + 150μm test sieve

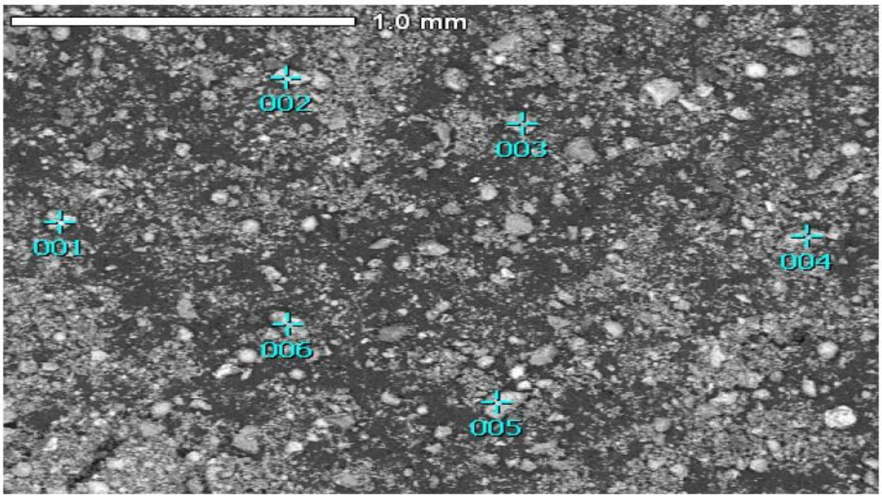


Figure 1b Morphological structure for raw ash passing + 75μm test sieve

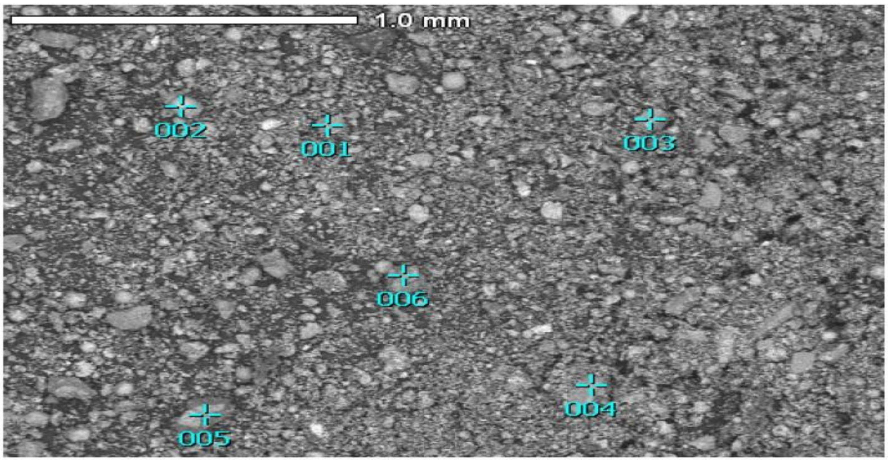


Figure 1c Morphological structure for raw ash passing +38μm test sieve

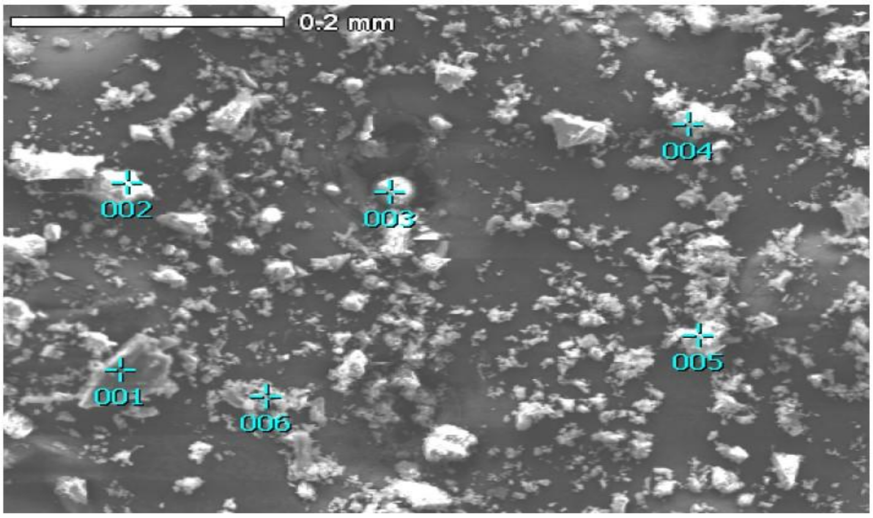


Figure 1d Morphological structure for ground ash

2.4. Chemical and elemental compositions

Chemical compounds and elements present in the ground POFA were analyzed using Energy Dispersive X-ray Fluorescence (EDXRF-Minipal 4 X-ray spectrometer, model PW4030) analyzer, operated at standardless. Interested chemical compound was centered on quantity of SiO_2, responsible for releasing Si^{3+} ions during isomorphous reaction with Al^{3+} ions; CaO, responsible for releasing Ca^{2+} ions, during cation exchange reaction and crowdingout effect, and for the initiation of pozzolanic reaction. Particular interest was centered on the presence and background concentrations of any trace toxic heavy metals. **Table 3** presents chemical compounds detected from the analysis.

2.5. Mineralogical analysis

The mineral compositions of the ground POFA were detected via Brucker D8 Advanced X-Ray diffractometer (XRD). Major mineral contents picked by the instrument were then identified using ICDD 2006 mineral identification approach, as shown in Figure 2.

Table 3 Chemical and elemental composition of ground POFA

S/No.	Chemical nomenclature	Metal Oxides		Metals		
		Chemical Symbol	Mean chemical composition (EDXRF) (%)	Elemental constituents	Multiplier for Oxide-to-Element conversion	Mean elemental composition (EDXRF) (%)
1	Silicon oxide	SiO_2	52.35	Si	0.4671	24.45
2	Aluminum oxide	Al_2O_3	6.27	Al	0.5291	3.32
3	Iron oxide	Fe_2O_3	13.36	Fe	0.6988	9.34
4	Calcium oxide	CaO	11.72	Ca	0.7148	8.38
5	Magnesium oxide	MgO	-	Mg	0.6031	-
6	Potassium oxide	K_2O	15.52	K	0.8299	12.88
7	Sulphur trioxide	SO_3	1.50	S	0.4000	0.60
8	Manganese oxide	MnO	0.11	Mn	0.7740	0.09
9	Combination	$SiO_2 + Al_2O_3 + Fe_2O_3$	71.98	-	-	-
10	Mass Loss on ignition at 440^0C	LOI(raw/ground)	22.64/9.68	-	-	-
11	H^+ concentration	pH	10.35			

2.6. Mass loss on ignition

To ascertain the quantity of unburnt carbon present in the POFA, mass loss on ignition (LOI) test was performed in accordance with test procedure specified in the BS 1377: Part 3 (BSI, 1990). The test was carried out on triplicate test coupons for both the as received and ground POFA samples, via ELE-electric muffle furnace SCALTEC. Calibrated SPB32 120g analytical bench-top balance was used for weighing all test coupons. The furnace was maintained at temperature of $440\pm\ 25\ ^{0}$C for 4 h. Specimens were later transferred to a desiccator for cooling to constant weight not exceed 0.1*%* of the previous measured mass. For each specimen, LOI value was determined by calculating mass after ignition as *%* of initial mass. Average values were recorded and results presented in Table 3.

2.7. pH value

In earthen sanitary landfill liner, the pH controls most chemical and biological processes (Chun et al., 2008). The test was performed on the ground POFA-aqueous solution in accordance with the test procedure specified in the BS 1377: Part 3 (BSI, 1990). In the electronic method, HACH Sension 1 bench-top pH meter and CORNING-PC-420D stirrer was operated at an ambient temperature of $26 \pm 0.1\ ^{0}C$. Triplicate aliquot of the ground ash specimens were prepared and mixed with distilled water at a liquid-to-solid ratio of 2.5.

That is, 30 ± 0.1 g of the POFA in 75 mL of distilled water, using 100 mL beaker. Each of the POFA-distilled water aqueous solutions was then stirred for few minutes via Thermolyne Cimarec Stirring Hotplate, with its temperature knob turned off to prevent specimen heating. The suspension was later covered with aluminum foil and allowed 8 h. contact time before measuring the pH. The pH values obtained for all tested coupons were then averaged and final result is presented in Table 3.

3. DISCUSSION

3.1. Physico-chemical properties of POFA

3.1.1. Beneficiation of POFA

As can be seen in Table 1, physical characteristics of the as-received and ground POFA shows that visual appearance of the raw ash was characterized by dark spongy and porous structure. Main constituents of the raw ash include hard but light weight honeycombed impurities, and irregular shape and cellular textures. The descriptions were in agreement with the results of Abdul et al. (2011) and Weerachart et al. (2009).

However, POFA beneficiation through sieving and the two-stage pulverization processes revealed that the ash was having grayish appearance, after sieving through the 150 μm test sieve. In addition, the matrix of the ground POFA possess very fine and powdery airborne textured. The gray colour indicates significant amount of carbon contents have been removed through the sieving exercise.

3.1.2. Effect of beneficiation processes on particle size distribution and surface area

Table 2 presents the particle size distribution and corresponding SSA obtained from the powdered POFA. The table also shows the average particle density of the ash. It can be seen that from the first stage mechanical activation, the mean particle diameter when 60 % from the tested particles were captured was 40 μm; the corresponding SSA value was 4288.94 cm2/g. Second stage pulverization substantially reduced the mean particle diameter to 27μm. Corresponding SSA for the second stage grinding significantly increased to 5405.40 *cm2/g*. The value indicates about 32% reduction in the mean diameter, with a corresponding increase in SSA by 26%. Also, the values represent a remarkable improvement in exposure of negative charged reactive surface of the ground POFA (Gambhir, 2004). Also it is anticipated that an effective filler effect and pozzolanic reaction will be achieved from the beneficiated particle sizes recorded (Chai et al., 2011).

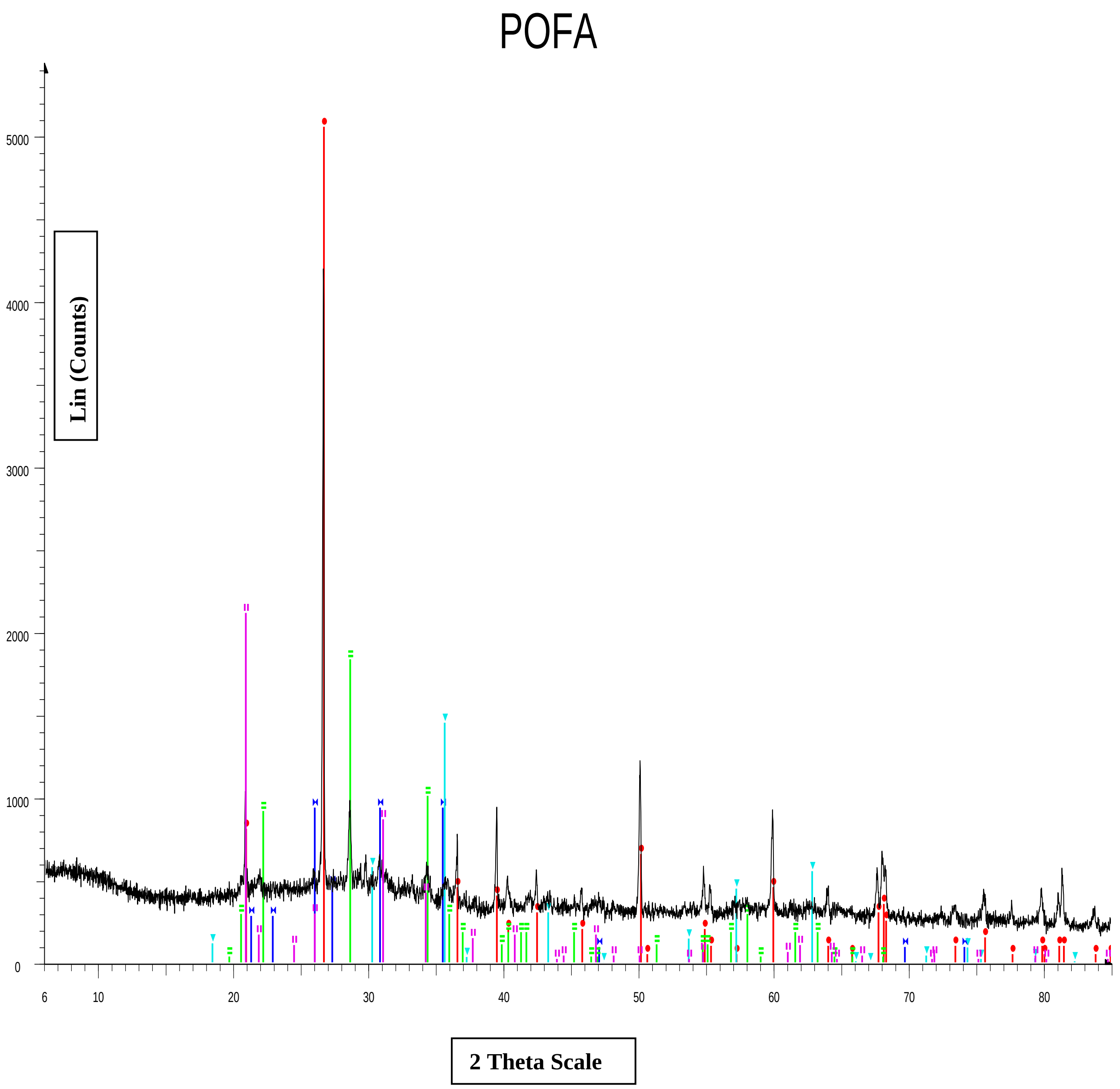
3.1.3. Effect of beneficiation processes on surface morphology

Microstructure of sieved POFA retained on 75 and 38 μm test sieves, and receiving pan is presented in Figs. 1a, b and c respectively. On the order hand, Fig.1d shows the scanned morphological structure of the ash after mechanical activation. Generally, the first three SEM analyses reveal distinct grain size reduction, as finess progresses. Finally, SEM analysis for the homogenized POFA followed by the 2-stage mechanical grinding is as shown in Fig.1d. From the figure, it can be visualized that there is strong possibility that the overwhelming dark background appearance are the carbon content present in the ash as impurities and other foreign materials. However, SEM structure of raw and ground POFA resembles the presented in Weerachart et al. (2009).

3.2. Potential use of POFA as co-sorbent in kaolin landfill liner system

3.2.1. Effect of ground POFA on isomorphous reaction in kaolin liner

As illustrated in Table 3, the main chemical composition of the ground POFA was SiO_2 (52.35%). Possibility abound that in the presence of moisture and suitable pH environment, the dissociation of the SiO_2 from the ash to release Si^{4+} ions may take place (Chun et al., 2008; Prashant, 2005). In addition, under same condition, the Al_2O_3 (6.27%), may as well dissociates to yield Al^{3+} ions. In particular, the dissociated Al^{3+} ions from the ground POFA may undergo some isomorphous substitution by replacing the Si^{4+} present in the silica tetrahedral sheets without altering the silica structure. The substitution reactions may then results in a deficit of positive charge, and a corresponding development of a negative charge on the kaolinite mineral (O'Flarhearty, 1976). The reaction is anticipated to complement the weak isomorphism in the kailinitic clay when used as co-sorbent in the design of earthen sanitary landfill liner.



Quatz (SiO_2, Y: 93.05 %); Magnesium Oxide (MgO_4, Y: 17.26 %); Potassium Magnesium Silicate ($K_2MgSi_3O_8$, Y 33.75 %); Phosphorus Oxide (P_2O_5, Y: 38.92 %) and Magnesium Aluminum Iron Oxide ($MgAl_2Fe_{1.8}O_4$, Y: 26.69 %)

Figure 2 Mineralogical composition of ground POFA

3.2.2. Effect of ground POFA on cationic exchange reactions in kaolin liner

From Table 3, it is also noteworthy that the calcium oxide; CaO (≈ 12 %) inherited by the POFA may as well dissociates under conducive hydrated environment to release Ca^{2+} ions. The Ca^{2+} ion is known to possess strong ionic potential (O'Flarhearty, 1976). Although having weak ionic potential, the potassium oxide, K_2O (≈ 16 %%) may as well dissociates and release its K^{+} ions. Both of the metal ions may readily undergo ionic exchange with ions of toxic heavy metallic such as Cu^{2+}, Zn^{2+}, Cd^{2+}, and Pb^{2+} present in landfill leachate. Hence, strong anticipation in the processed POFA complementing the weak kaolin as co-sorbent in the design of earthen sanitary landfill liner may result.

3.2.3. Effect of ground POFA on crowding of calcium ions in kaolin liner co-sorbent in kaolin landfill liner

In addition, it may be anticipated that the released Ca^{2+} ions from the ground POFA into the leachate environment may be in excess of cationic exchange capacity of the co-sorbent system. Hence, in this scenario, it is also anticipated that the kaolin-POFA liner saturated with Ca^{2+} ions may experience particle flocculation due to Ca^{2+} ions crowding effect (O'Flarhearty, 1976). Anticipated long term effect of crowding of Ca^{2+} ions may result to the reduction in hydraulic conductivity of the kaolin-POFA matrix as sanitary landfill liner system.

3.2.4. POFA as a complementary pozzolanic material in kaolin landfill liner system

The summation of SiO_2 , Al_2O_3 and Fe_2O_3 plays important role in the expected efficacy of mineral or natural admixtures. As can be seen in Table 3, the chemical analysis reveals that in accordance with the ASTM C618-01 (2001) specification, the summation of SiO_2 , Al_2O_3 and Fe_2O_3 present in the ground POFA satisfies requirements to be classified as Class C supplementary cementitious material (SCM). It is anticipated that in a hydrated environment, the CaO in the Kaoin-POFA mixture may react with the SCM. In long term, expected overall effect of the reaction is that the composite landfill liner system may chemically change into a natural cement structure of calcium silicates/aluminates under suitable pH environment (Chun et al., 2008; Prashant, 2005). In addition, from Figure 2, the present of magnesium oxide (MgO4, $\approx 17\%$)), potassium magnesium silicate ($K_2MgSi_3O_8$, $\approx 34\%$), and magnesium aluminum oxide ($MgAl_2Fe1.8O_4$, $\approx 37\%$)) in the POFA also implies its suitability as cementing agent in the kaolin-POFA composite system (Chun et al., 2008). It is anticipated that the overall effect of the minerals may substantial increase the attenuation efficiency of the kaolin-POFA matrix, through the occlusion of injurious heavy metal ions present in landfill leachate. Significant quantity of quartz (SiO_2,$\approx 93\%$) mineral detected was in agreement with results of Foo & Hameed (2008).

4. CONCLUSIONS

Based on the physico-chemical test carried out on the POFA as discussed above, the following conclusions are drawn:

1) An effective filler effect and pozzolanic reaction may be achieved from the beneficiated POFA and kaolin mixture.
2) Admixing kaolin with POFA may complement the weak isomorphism in kailinitic clay when used as co-sorbent in the design of earthen sanitary landfill liner.
3) Long term effect of crowding of Ca^{2+} ions may result to the reduction in hydraulic conductivity of the kaolin-POFA matrix as sanitary landfill liner system.
4) The overall effect of the various mineral constituents of the POFA may substantial increase the attenuation efficiency of the kaolin-POFA matrix, through the occlusion of injurious heavy metal ions present in landfill leachate.

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