



PALM OIL FUEL ASH (POFA) RECYCLING IN PORCELAIN MANUFACTURE: EFFECTS ON PHYSICAL AND MICROSTRUCTURAL PROPERTIES AT DIFFERENT TEMPERATURES

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Abstract

Silica from palm oil fuel ash (POFA) was incorporated in a porcelain composition in substitution of quartz. The influence of the substitution on the physical behavior and microstructure has been investigated. The POFA was grinded in a ball mill until the particle size was reduced to about 50 μm . Then it was heated at a temperature of 600 $^{\circ}\text{C}$ for 1.5 hr in an electric furnace. About 5 wt% to 25 wt% of POFA was used to substitute the quartz in the porcelain composition. The mixed powder was then pressed into pellets at pressure of 91 MPa. All the pellets were sintered at a temperature of 1000 $^{\circ}\text{C}$, 1100 $^{\circ}\text{C}$, 1200 $^{\circ}\text{C}$ and 1280 $^{\circ}\text{C}$ for 2.0 hr soaking times each. It was found that the volume shrinkage increases with the increase in substitution of quartz by POFA. However, percentage porosity decreases as the substitution increases. With a value of 2.4 % the least porosity was achieved at the temperature of 1100 $^{\circ}\text{C}$. Similarly microstructure study reveals that densification took place at the temperature of 1100 $^{\circ}\text{C}$. This is because of glassy phase that are formed as a result of increase in temperature and the substitution of quartz by POFA.

Key words: Percentage porosity; POFA; Porcelain; Quartz; Temperature

Introduction

Porcelains consists of approximately 50 % kaolin ($\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2 \cdot 2\text{H}_2\text{O}$), 25 % silica (SiO_2), and 25 % feldspar ($\text{K}_2\text{O} \cdot \text{Al}_2\text{O}_3 \cdot 6\text{SiO}_2$) (Prasad *et al.*, 2001a). This composition makes a material body with plasticity and a wide firing temperature range at a relatively low cost (Jamo *et al.*, 2015). Porcelain is a type of ceramic highly valued for its beauty and strength. It is known as a material for high-quality vases, table ware, blenders, reactor chambers, and condensers, pipes, cooling coils, pumps, figures and decorative objects. Porcelain is a type of the ceramic materials which have the vitreous characteristics. Vitrification indicates a high degree of melting on firing which confer low porosity and high glass content on fired porcelain (Hassan *et al.*, 2014a) As Porcelains have high hardness, low electrical and thermal conductivities, and brittle fracture (Noh *et al.*, 2014).

Some improvements in the mechanical properties have been observed by several authors (Hassan *et al.*, 2014b), through the reduction of the particle size of quartz and non-plastic materials. Hassan *et al.*, (2014b) reported that dissolved quartz in the glassy phase and cristobalite phase precipitation has helped in improving the mechanical properties of the Porcelain.



However, quartz grains embedded in the porcelain glassy matrix have a deleterious effect on the mechanical strength mainly because of its transformation during cooling (Prasad *et al.*, 2001a) which results in the development of stresses which initiate fracture (Jamo *et al.*, 2015). The thermo-mechanical properties of whiteware bodies change greatly during the reconstructive and the displacive transformation of free silica due to change in volume, which was reviewed in detail by Jamo *et al.*, (2014).

Several investigators (Maity and Sarkar, 1996; Prasad *et al.*, 2001b; Prasad *et al.*, 2002; Derevyagina *et al.*, 1980, Das and Dana, 2004) tried to improve the mechanical properties of whiteware bodies by replacing quartz with other materials viz; sericiticpyrophyllite, kyanite, bauxite, sillimanite sand alumina, fly ash and rice husk ash (RHA). Although the alumina in different forms has a favourable influence on the mechanical properties of whiteware due to the formation of primary mullite, it lowers the recrystallisation of secondary mullite due to an increase in the viscosity of the glassy phase. On the other hand, researchers such as Prasad *et al.*, 2001a; Jamo *et al.*, 2015; Noh *et al.*, 2014; Tasnim *et al.*, 2018 reported the use RHA as a substitute for quartz in the Porcelain ceramic tile. POFA when incorporated into the porcelain body is expected to add value to the properties of porcelain.

POFA is a by-product of palm oil industry. It is as a result of the combustion of palm oil plant (*ElaeisGuineensis* tree) residues. In total, about 90 million metric tons of trunks, shells, husks, palm press fibers, and empty fruit bunches are produced every year (Tasnim *et al.*, 2018). After the extraction of the oil from the fresh palm fruit, both husk and shell are burnt in palm oil mill plants at a temperature of 800 °C -1000 °C as boiler fuel to produce steam needed for electricity generation and palm oil extraction (Jamo *et al.*, 2015; Alsubari *et al.*, 2016). The burning process results in an ash, which is referred to as palm oil fuel ash (POFA). After combustion in the steam boiler, about 5% POFA by weight of solid wastes is produced (Hassan *et al.*, 2015). Since the tropical countries are continuously increasing the production of palm oil, the quantity of POFA is also increasing and thus creating a huge environmental load (Chavalparit, 2006; Safiuddin *et al.*, 2011; Noh *et al.*, 2016). In Malaysia, an investigation was carried out to examine the potential of POFA to be used as a fertilizer for the agricultural purpose (Chandara *et al.*, 2010). However, due to the absence of sufficient nutrients to be used as a fertilizer, POFA is mostly dumped in open field near palm oil mills without any profitable return, thus causing environmental pollution and human health hazard (Prasad *et al.*, 2001b). As an effort to find a solution to these problems, several studies were conducted to examine the feasibility of using POFA in concrete. It has been found that the properly processed POFA can be used successfully as a supplementary cementing material for the production of concrete (Prasad *et al.*, 2001a).



However, this is not enough, an alternative means has to be looked upon in order to further reduce the problem. This will help towards having cleaner environment, reduction in cost of production and add value to the some of the properties of the porcelain. The present study therefore seeks to find a means of utilizing POFA as a substitute for quartz in order to protect environmental pollution and possibly to values to add some of the properties of porcelain. The present study wishes to investigate the palm oil fuel ash (POFA) recycling in porcelain manufacture: effects on physical and microstructural properties at different temperature.

Methodology

The removal of excess carbon and other unburned organic materials contained in POFA is important to avoid their potential negative effect on finished product. Thus, the POFA was dried in an oven at 100 °C for 24 hr and then sieved using a set of sieves (50 µm) to remove the particles coarser than 50 µm. The untreated POFA was then grinded in a ball mill to reduce the particle size to improve reactivity. The milling time was set at approximately 1.5 hr at the rate of 200 rev/min. The untreated POFA was heated at a temperature of 600 °C for 1.5 hr in an electric furnace.

Porcelain raw materials were grinded separately in a ball mill. The powders were sieved using sieve shaker and dried in an oven. The POFA was gradually poured into the porcelain powder from 5 wt% to 25 wt% (Table 1). These compositions were mixed using a ball mill for 1.5 hr. The mixed powder was pressed into pellets at mould pressure of 91 MPa. All the pellets were sintered at a temperature of 1000 °C, 1100 °C, 1200 °C and 1280 °C for 2 h soaking time each, at a heating rate of 5 °C per minute. The compressive strength was determined using Universal Testing Machine (UTM). The chemical composition of the POFA was studied using X-Ray Fluorescence (XRF) machine while the amorphous structure of POFA was identified through XRD and the microstructural features were studied by Scanning Electron Microscope SEM.

Table 1: The composition with substitution of quartz by POFA (wt %)

Sample name	Kaolin	Feldspar	Quartz	POFA
AQ1	50	25	25	0
AQ2	50	25	20	5
AQ3	50	25	15	10
AQ4	50	25	10	15
AQ5	50	25	5	20
AQ6	50	25	0	25

MettlerTeldo XS-64 device available at Ceramic Laboratory Universiti Tun Hussein Malaysia was used for this purpose; the experiment was carried out according to American Society for Testing Materials (ASTM) C373 (Phonphuaket *et al.*, 2016). The samples were dried in an oven at



150 °C for 2 hours, followed by cooling in a desiccator. The dry specimen was weighed and labelled as (D), and after that the sample was put inside distil water and boiled for 5 hours. Then the samples were soaked at room temperature for 24 hours and weighed as S. Then it was taken out of water and the filter paper was used to remove excess water and then it weighed again as M. The volume of the sample is given by;

$$V = M - S \text{ (cm}^3\text{)} \text{ (Phonphuak et al., 2016)} \quad 1$$

Where;

V = Exterior volume (cm^3)

M = Saturated mass (wet) (g)

S = Floating in water mass (g)

The bulk density, B , expresses in g/cm^3 , was calculated as follows:

$$B = \frac{D}{V} \left(\frac{\text{g}}{\text{cm}^3} \right) \text{ (Phonphuak et al., 2016)} \quad 2$$

Where;

B = Density (g/cm^3)

D = Dry mass (g)

V = Exterior volume (cm^3)

The porosity P , is expressed as a percent, the ratio of the volume of the pores of the specimen to its exterior volume. Calculate the porosity as follows:

$$P = \left(\frac{M-D}{V} \right) \times 100\% \text{ (Phonphuak et al., 2016)} \quad 3$$

Where;

P = Porosity (%)

D = Dry mass (g)

M = Saturated mass (wet) (g)

V = Exterior volume (cm^3)

Results and Discussions

Figure 1 shows the particle size of POFA. The figure shows the percentages which are (d_{10}) and (d_{60}) of the POFA particle size are $1.31 \mu\text{m}$ and $15.36 \mu\text{m}$ respectively. The average particle (d_{30}) size is $5.40 \mu\text{m}$. Jamo et al., (2014) asserts that particle size within this range contributes significantly to the increase in the physical and microstructural properties porcelain body.

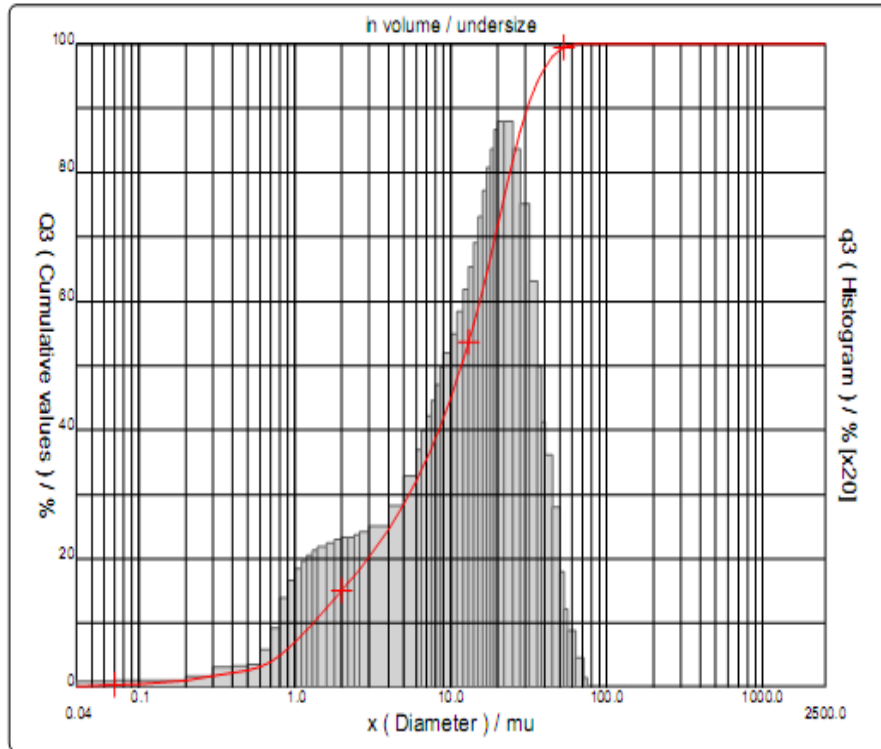


Figure 1: Result of particle size analysis of POFA

It is apparent from Figure 2 that porosity decreases with increase in replacement and temperature. This graph can be grouped into two. The first group is between the temperatures of 1000°C and 1100°C. In this group the porosity decreases with increase in replacement of quartz by POFA. The minimum porosity was achieved on 15 wt% with values of 6.0% and 2.4%. The porosity increases after reaching minimum. The decrease of porosity attributed to excess glassy formation.

According to the models of sintering by viscous flow of glass, such as Frenkel's and Mackenzie-Shuttleworth's models, the sintering rate depends directly on the surface tension and inversely on the viscosity of the liquid phase at the sintering temperature (Zanelli *et al.* 2011). According to Alves *et al.* (2011), when there is a suitable amount of glassy phase content it is quite advantageous for the porcelain tile as it only fills up the pores. During sintering process, samples will experience shrinkage due to the temperature and the formation of a glassy phase that is mainly originated from the feldspar. Increasing POFA causes both an increase in liquid phase amount and a decrease in liquid phase viscosity. Under the surface energy forces created



by the fine pores contained in the ceramic body, the liquid phase tends to approach the particles and, therefore, open porosity decreases.

For the second group which is for the temperatures of 1200°C and 1280°C. For the temperature of 1200°C the minimum porosity was recorded with a value of 4.7% 15wt% of POFA. Increasing POFA content into the porcelain results in increase in cristobalite which brings about decrease in porosity. And for the temperature of 1280°C the minimum porosity was achieved with a value of 6.5% 0 wt% of POFA respectively. This could be related to the amount of oxides such as Fe_2O_3 , P_2O_5 among others (Santos *et al.*, 2012).

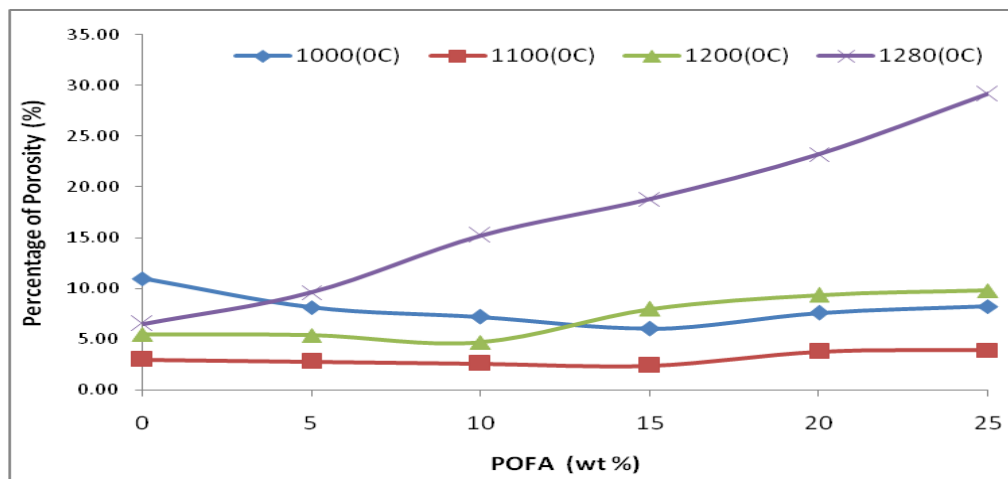


Figure 2: Effect of temperature on percentage of porosity of the samples with different percentage of POFA wt%

It can be seen from Figure (3a) that the grains are big due to undissolved minerals at the temperature of 1000 °C, but as the temperature increases to 1100 °C the grains becomes smaller (Figure 3b). Least porosity can be seen from Figure 3c. More mullite and cristobalite are also formed as the temperature increases, the grain size become smaller, densification took place because of glassy phase that are formed as a result of increase in temperature and substitution of quartz by POFA. As the temperature reaches 1300 °C (Figure 3d) pores re-emerged, this could be as result of bloating (i.e., pore volume expansion), which arises from higher pressures (at high temperatures) of gases such as nitrogen, carbon monoxide and carbon dioxide entrapped within closed pores, similar result were obtained by Prasad *et al.* (2001).

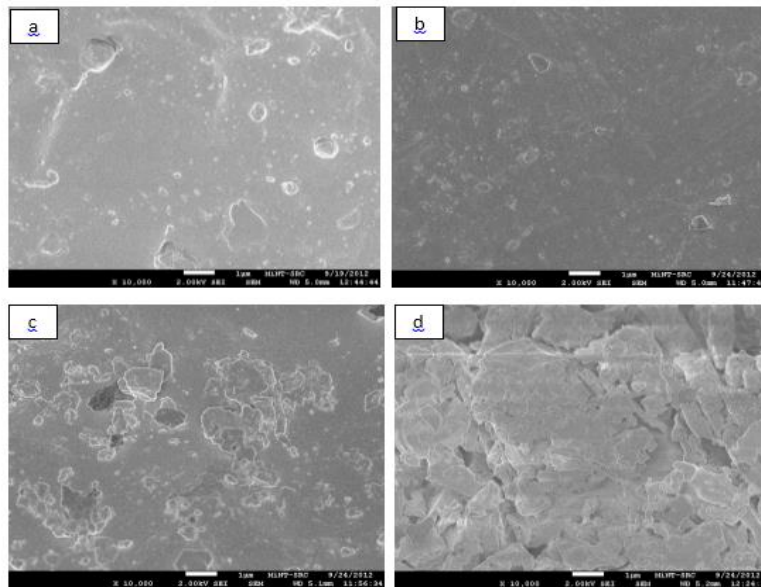


Figure 3: SEM of the sample containing 15 wt% POFA sintered at a temperature of (a) 1000 °C (b) 1100 °C (c) 1200 °C (d) 1280 °C. All micrograph were taken with 1000X magnification

Table 2 shows the quantitative analysis of the XRD analysis. The percentage of mullite and cristobalite increases as the temperature increases from 1000 °C to 1100 °C while that of quartz decreases with temperature. However, as the temperature increases to 1280 °C the values of the three minerals decreases. It could be deduced that the temperature of 1100 °C is good for the porcelain production instead of the conventional 1280 °C.

Table 2: XRD quantitative analysis of the samples containing 15 wt% of POFA sintered at different temperatures (mould pressure = 91 MPa, soaking time = 2 hours)

Temperature (°C)	Quartz (%)	Mullite (%)	Cristobalite (%)	Glassy phase
1000	45.0	25.1	10.0	19.9
1100	33.7	40.5	24.0	1.8
1200	30.4	35.1	20.8	13.7
1280	21.3	27.4	20.9	30.4

Conclusion

Recycling of POFA in porcelain structure was found to be beneficial towards the improvement of physical and microstructural properties. The porosity decreases as the substitution of POFA increases from 0wt% to 15 wt%. The minimum porosity was recorded at the temperature of 1100 °C with a value of 4.7%. Microstructural results reveals that the porosity decreases as the



temperature increases. The least porosity was observed at the temperature of 1100 °C. The XRD result also reveals that mullite and cristobalite increases with the increase in temperature. Increase in mullite and cristobalite contributes significantly to the physical properties of the porcelain. It could be concluded that the 1100°C and 15 wt% of POFA is recommended for the manufacturing of porcelain, instead of the conventional 1280°C. This helps to reduce the energy of the production and consequently the cost of production.

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