INFLUENCE OF MOULD PRESSURE AND SUBSTITUTION OF QUARTZ BY PALM OIL FUEL ASH ON THE HARDNESS OF PORCELAIN BODY

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ABSTRACT
This paper presents use of treated palm oil fuel ash (POFA) as a substitute material for quartz in fabricating an improved porcelain tile. Untreated POFA was dried in an oven at 100 ºC for 24 h. The untreated POFA was then ground in a ball mill to reduce the particle size to improve reactivity. It was then sieved using a set of sieves (50 µm) to remove the particles coarser than 50 µm. The untreated POFA was heated at a temperature of 600 ºC for 1.5 h. The mixed powder was pressed into pellets at pressure of 31 MPa, 61 MPa, 91 MPa and 121 MPa. All the pellets were sintered at a temperature of 1100 ºC for 2 h soaking time. It was found that the bulk density and Vickers hardness of porcelain increases with increase in substitution of quartz by POFA. The highest bulk density and Vickers hardness of the porcelain was achieved on 15 wt% substitution of quartz by POFA at a Mould pressure of 91 MPa.

Keywords: Bulk density; POFA; Porcelain; Porosity; Volume shrinkage; Quartz

INTRODUCTION
POFA is a by-product of biomass thermal power plants where oil palm residues are burned to generate electricity. Malaysia is one of the largest producer of palm oil with around 41 % of the total world supply in years 2009–2010 (Altwair et al, 2013; Sata et al, 2012; Chandra et al, 2010; Chindaprasirt et al, 2009). Thailand for example, it had been estimated that 2.1 million tons of biomass was used as fuel in 2004, producing about 100,000 tons (5 %) of biomass ash (Tangchirapat and Chai, 2010; Chindaprasit and Sumrerng, 2008). Since palm oil is one of the major raw materials used to produce bio-diesel, it is likely that the production of POFA will increase every year. Very little of the POFA produced is actually used. While some of it serves as low-value material for backfill or fertilizers, most of the POFA is posed as waste in landfills, causing environmental sand other problems (Kroehong et al, 2011).

To solve this potential environmental problem, many researchers have studied the use of POFA in blended cement. It has been reported that POFA has low pozzolanic reactivity and should not be used as a cement replacement in quantities greater than 10% by mass of binder (Tay, 1990). In order to improve the pozzolanic reactivity POFA, ground POFA (GPOFA) has been investigated. GPOFA is a good pozzolanic material and can be used as a replacement in Portland cement up to levels of 20 % and 30% for medium particle size and small particle size, respectively (Chindaprasit et al, 2007). In addition, the dosage of superplasticizer (SP) needs to be increased to maintain the same fluidity of concrete without GPOFA (Chindaprasit et al, 2008; Rukzon et al, 2009; Chandra et al, 2010). In previous work, the removal of unburned carbon from GPOFA by heat treatment at 500 ºC for 1hour which produced treated ground palm oil fuel ash (TGPOFA) was investigated to improve the fluidity of blended cement.

The results showed that blended cement pastes containing TGPOFA has better fluidity than those containing GPOFA. This is due to the SP adsorption of blended cement pastes containing TGPOFA which was lower than those containing GPOFA. Besides that, semi quantitative XRD analysis was used to determine mineralogical compounds of GPOFA and TGPOFA. It showed that GPOFA and TGPOFA contain large amounts of glassy phase (Chandra et al, 2011).

Porcelain is a highly vitrified ceramic material produced from a body formulated by mixtures of clay, quartz and feldspar. The clay [Al₂Si₂O₅(OH)₄], gives plasticity to the ceramic mixture, flint or quartz (SiO₂), maintains the shape of the formed article during firing; and feldspar [K₂Na₂Al₃Si₃O₁₀] serves as flux. These three constituents place porcelain tile in the phase system [(K₂Na₂)O·2Al₂O₃·SiO₂] in terms of oxide constituents, hence the term triaxial porcelains (Olupot, 2006).

The main phase composition of a porcelain body is constituted by a heterogeneous glassy matrix and needle shaped mullite crystals together with some quartz grains and closed irregular shaped pores. Mullite crystals, which are derived from the solid-state decomposition of the clay reacting with feldspar, are endowed with excellent mechanical, creep, thermal and chemical properties. Because of the complex interplay between raw materials, processing routes and the kinetics of the firing process, porcelains represent some of the most complicated ceramic systems (Lopez et al, 2011). The present study wishes to explore the Effects of Mould Pressure on the Substitution of Quartz by Palm Oil Fuel Ash (POFA) in Porcelain Composition.

MATERIALS AND METHODS
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 ºC for 24 h. The untreated POFA was then ground in a ball mill to reduce the particle size to
improve reactivity. The milling time was approximately one and half hours at 200 rev/min. It was then sieved using a set of sieves (50 µm) to remove the particles coarser than 50 µm. The untreated POFA was heated at a temperature of 600 ºC for 1.5 h in an electric furnace. After the heat treatment, the color of treated POFA turned from light brown to grayish red (Figure 3) when the unburned residue was removed.

Porcelain powder was grounded separately in a ball mill. The powder was sieved using sieve shaker and dried in an oven. The POFA was gradually incorporated into the body of porcelain powder from 0 wt% to 25 wt%. The composition was mixed using a ball mill for 1.5 h. The mixed powder was pressed into pellets at pressure of 31 MPa, 61 MPa, 91 MPa, 121 MPa All the pellets were sintered at a temperature of 1100 ºC for 2 h soaking time, at a heating rate of 50 ºC per minute.

The physical properties of the pellets such as volume shrinkage, porosity, and bulk density were determined. The chemical composition of the POFA was studied using X-Ray Fluorescence (XRF) while the crystalline structure of the POFA was identified through XRD and the microstructural features were studied by FESEM.

### Table 1: Body composition with progressive replacement of quartz by RHA in a standard whiteware composition (wt %)

<table>
<thead>
<tr>
<th>Mix Number</th>
<th>Clay</th>
<th>Feldspar</th>
<th>Quartz</th>
<th>POFA</th>
</tr>
</thead>
<tbody>
<tr>
<td>AP1</td>
<td>50</td>
<td>25</td>
<td>25</td>
<td>0</td>
</tr>
<tr>
<td>AP2</td>
<td>50</td>
<td>25</td>
<td>20</td>
<td>5</td>
</tr>
<tr>
<td>AP3</td>
<td>50</td>
<td>25</td>
<td>15</td>
<td>10</td>
</tr>
<tr>
<td>AP4</td>
<td>50</td>
<td>25</td>
<td>10</td>
<td>15</td>
</tr>
<tr>
<td>AP5</td>
<td>50</td>
<td>25</td>
<td>5</td>
<td>20</td>
</tr>
<tr>
<td>AP6</td>
<td>50</td>
<td>25</td>
<td>0</td>
<td>25</td>
</tr>
</tbody>
</table>

The X-Ray Fluorescence (XRF) analysis is proficient in analyzing material contents inside POFA, hence the amount of SiO$_2$ can be observed. The presence of various compounds within porcelain and POFA sample can be seen in Table 1. This table shows the result of XRF analysis of porcelain and POFA. It is evident that SiO$_2$ is the major composition in all the raw materials viz: POFA, clay, feldspar and quartz with 66.91 wt% and 69.30 wt%, 72.70 wt% and 99.40 wt%, respectively.

### Table 2: Chemical analysis of POFA and Porcelain

<table>
<thead>
<tr>
<th>Sample</th>
<th>SiO$_2$</th>
<th>Al$_2$O$_3$</th>
<th>Fe$_2$O$_3$</th>
<th>CaO</th>
<th>K$_2$O</th>
<th>P$_2$O$_5$</th>
<th>MgO</th>
<th>SO$_3$</th>
<th>Na$_2$O</th>
<th>MnO</th>
<th>TiO$_2$</th>
<th>CO$_2$</th>
<th>LOI</th>
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<tbody>
<tr>
<td>POFA</td>
<td>69.91</td>
<td>6.44</td>
<td>5.72</td>
<td>5.36</td>
<td>3.20</td>
<td>2.44</td>
<td>0.33</td>
<td>0.19</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>2.30</td>
<td></td>
</tr>
<tr>
<td>Clay</td>
<td>69.30</td>
<td>24.30</td>
<td>0.27</td>
<td>-</td>
<td>2.44</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>0.70</td>
<td>0.36</td>
</tr>
<tr>
<td>Feldspar</td>
<td>72.70</td>
<td>16.40</td>
<td>0.40</td>
<td>0.30</td>
<td>2.42</td>
<td>6.87</td>
<td>0.29</td>
<td>-</td>
<td>0.10</td>
<td>-</td>
<td>0.32</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Quartz</td>
<td>99.40</td>
<td>0.22</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>0.10</td>
<td>0.28</td>
</tr>
</tbody>
</table>

Figure 1 and table 3 show the X-ray patterns and the XRD analysis of the samples containing POFA pressed at different MP's. The main mineralogical phases of the raw materials are identifiable. The major phases identified are quartz hexagonal (ICDD 046-1045), mullite orthorhombic (ICDD 089-2645) and cristobalite tetragonal (ICDD 082-0512). As can be seen from the Figure there is not a significant change in the peaks, so also from the quantitative analysis (Table 3). This shows that the crystalline phases are independent of MP.
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Figure 1: The XRD curves of the mixed samples containing POFA pressed at different MP

Table 3: Quantitative result of mixed samples containing POFA

<table>
<thead>
<tr>
<th>Pressure (MPa)</th>
<th>Quartz (%)</th>
<th>Mullite (%)</th>
<th>Cristobalite (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>31</td>
<td>31.9</td>
<td>38.2</td>
<td>25.1</td>
</tr>
<tr>
<td>61</td>
<td>33.0</td>
<td>37.1</td>
<td>25.0</td>
</tr>
<tr>
<td>91</td>
<td>33.7</td>
<td>38.5</td>
<td>25.1</td>
</tr>
<tr>
<td>121</td>
<td>33.3</td>
<td>39.4</td>
<td>23.9</td>
</tr>
</tbody>
</table>

Figure 2 shows FESEM micrographs of a surface of the body mixes pressed at different MP. The existence of pores can be seen from the Figure. The porosity consists of interconnected pores with irregular shape (Figure 2a). The micrographs, show an increase in densification with increasing MP. The high porosity is clearly visible at lower MP and it is confirmed by FESEM observation. At the MP of 61 MPa (Figure 2b) densification began to take place. As MPa increases to 91 MPa (Figure 2c) densification reaches peak with least pores noticeable. It is because of the presence of POFA coupled with the MP that reduces the porosity via viscous-phase sintering which reduces deformation. At the MP of 121 MPa (Figure 2d) micro-cracks were developed. This could be attributed to over-compaction.
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Figure 2: FESEM of the body mixes samples sintered containing POFA pressed at pressed of (a) 31 MPa (b) 61 MPa (c) 91 MPa (d) 121 MPa 1000X

Figure 3 shows the bulk density of the sintered samples containing POFA. The bulk density increases with MP and with the gradual substitution of quartz by POFA. With the values of 2.30 g/cm$^3$, 2.40 g/cm$^3$, 2.45 g/cm$^3$ and 2.35 g/cm$^3$ on 15 wt% of POFA maximum bulk density was achieved at MP of 31 MPa, 61 MPa, 91 MPa and 121 MPa, respectively. The bulk density decreases after reaching maximum. In terms of MP, bulk density increases from 2.10 g/cm$^3$ to 2.45 g/cm$^3$ as the MP increases from 31 MPa to 91 MPa. While between the MP of 91 MPa and 121 MPa the bulk density decreases.

The increase in bulk density could be as a result of glassy formation which brings about increase in densification and compaction. The more the POFA is used to substitute quartz the more the glassy formation. This is in conformity with the FESEM results. Moreover, in the values of bulk density was noticed at an MP of 121 MPa, this may as a result of increase in values of porosity, since bulk density is inversely proportional to bulk density (Galos, 2011; Youssef and Ghazal, 2011).

Figure 3: Effect of MP on bulk density of bodymixes with different percentage of POFA
The result of Vickers micro-hardness for porcelain samples with different POFA content is shown in Figure 4. The maximum Vickers micro-hardness was achieved with values of 706 HV, 1015 HV, 1115 HV and 868 HV on 15 wt% of POFA at a MP of 31 MPa, 61 MPa, 91 MPa and 121 MPa, respectively. In line with the studies carried out by Daguano et al. (2012), the result of hardness is on the increase with MP and also with increase of quartz substitution by POFA. The hardness gradually increases with MP and with the substitution of quartz by POFA. The result of the Vickers micro-hardness is directly related to the result of bulk density and FESEM as shown in Figure 3. It was found out that the increase in hardness is related to densification, as a result of lower porosity (Kordani et al., 2012). Similarly, the Vickers hardness increases from 400 HV to 1115 HV as the MP increases from 31 MPa 91 MPa. Consequently, a decrease in the values of Vickers hardness was noticed at a pressure of 121 MPa. This may be as a result of a decrease in bulk density and increase in porosity values.

CONCLUSION
From the experimental results and discussion, the following conclusions can be drawn.

The bulk density and hardness was found to increases with increase in mould pressure and the substitution of quartz by POFA. Porcelain ceramic containing up to 15 wt% of treated POFA gives high bulk density (2.45 g/cm3) least porosity (2.20 %). The highest bulk density and least porosity took place at a mould pressure of 91 MPa. The increase in the physical properties is dependent upon microstructure changes.

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REFERENCES

Figure Error! No text of specified style in document.: Effect of MP on Vickers hardness of bodymixes with different percentage of POFA
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hardness and Fracture Toughness of Amorphous and Partially Crystallized 3CaO· P₂O₅ SiO₂–MgO Bioglasses", Materials Science and Engineering, A (533), 26-32.


