

Influence of HCl Acid Treated Rice Husk Ash (RHA) on Physical Properties of Porcelain in Triaxial Composition

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Abstract

In South-East Asia and some part of Sub-Sahara region in Africa and Nigeria, rice industries are one of the most important agricultural industries. In Thailand it is estimated that the total waste generated by rice industries was around 10 million tons in one year. Rice Husk Ash (RHA) was collected from local farmers and heated with furnace at 800°C for 24 hours. The ash was treated with HCl acid to enhance the purity and remove unwanted materials. To maintain triaxial porcelain structure, 50 wt. % clay, 25 wt. % feldspar and 25 wt. % quartz was used. Quartz was replaced with treated RHA and mixed homogeneously in a Labkorean Ball Mill machine (model RS-1S) for 12 hours at a speed of 250 revolutions per minute. The mixed powder was dry pressed into pallets at mould pressure of 91 MPa and sintered at a temperature of 1050 °C, 1100 °C and 1150 °C for 2 hours soaking time, at heating rate of 5 °C per minute using electric furnace (Protherm Furnace PLF 130/15). The highest value of compressive strength was determined to be 490 MPa, X-ray Fluorescence Machine (XRF) revealed that, RHA has similar chemical composition with quartz with SiO₂ as dominant composition which was further confirmed by the structural analysis through X-ray Diffraction Analysis (XRD). The bulk density was reported to have maximum value at 1100 °C sintering temperature as 2.45g/cm³.

Keywords: Rice Husk Ash (RHA), Porcelain, Quartz, XRD, XRF and SEM

1. Introduction

Rice industries has been remarkably growing over the past few decades, thus becoming a vital agricultural industry. This agricultural-based industry has contributed to the socio-economic growth in Nigeria. Several million hectares is cultivated with rice in Nigeria, which produce about millions of tons of rice husk annually (Jamo et al., 2015). It was further argued that, out of this huge amount of rice husk produced, only few percentages of rice are extracted from the total biomass in the plantation, the remaining is a large amount of waste such as rice husk (Ece & Nakagawa, 2002).

As the rice plantations increased yearly, the production of RHA is expected to also increase. The researchers argued that, since RHA is rich in silica, recent research pave way for the production of sustainable materials and also reduce the environmental pollution caused by RHA disposal (Lima et al., 2018). Research by Noh et al., (2017), shows that, more than 100,000 tons of rice husk has been disposed yearly and this figure is likely to increase annually, thus it requires ultimate disposal. This ash is cheap and available in abundance, also looking at the properties of rice husk ash it can be used as substitution for quartz in porcelain production (Noh et al., 2017).

RHA, silica fume, fly ash (FA) and palm oil ash are waste product that are pozzolanic materials that incorporating such ashes in self-compacting concrete is proven to enhances material and energy conservation. Use of these materials reduces heat hydration and increase durability performance, job site productivity, construction

sustainability and its cost efficient. Furthermore, the waste is environmentally friendly as their incorporation as cementitious supplementary materials lead to saving resources and energy due to reducing greenhouse gas emission as a result of fuel combustion in the furnace, limestone calcination, process of transportation and manufacturing related to production of ordinary Portland cement (Rincón, 2015)

According to Awal & Hussin, (2011), incorporation of pozzolans such as RHA, either naturally occurring or artificially made into concrete has been in practice since the early civilization. Besides its economic advantages, the main reason for their use is that they can give useful modification or enhancements to concrete and porcelain properties. Some researchers have studied the use of agricultural wastes as constituents in normal weight concrete and cement mortar, namely rice-husk ash, sawdust ash, and POFA. The study has revealed that the agricultural waste ashes could be used as pozzolanic materials due to high amount of silica in amorphous form (Kitouni & Harabi, 2011).

Porcelains are generally vitreous ceramic materials that are baked at higher temperatures. The triaxial porcelain consists of feldspar, clays mineral and quartz which after firing developed glassy, residual quartz and mullite (Lerdprom et al., 2017). Due to its high mechanical strength, durability, low water absorption and translucency, porcelains apart from household they are extensively used for scientific and engineering applications (Lerdprom et al., 2016). Porcelain tile that were previously used for floor are now used as ornaments in houses and internal wall, thus a slightly different properties are required for these applications such as mechanical designs that are directly related to the body microstructure (Rambaldi et al., 2017).

Porcelain is a traditional ceramic material which is also known as „whitewares“ ceramic due to its white color. Porcelain is systematically produced by three main raw materials of clay, feldspar, and quartz with a ratio of 50:25:25 referred as triaxial porcelain. It is comprised of plastic materials, fluxing agent and inert materials (Acchar et al., 2017) Although it has been established since 20th century, porcelain is still categorized as higher complexities ceramic in phase development (Carty & Senapati, 1998). Generally, quartz (SiO_2) and mullite ($3\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2$) are the main crystalline phase in porcelain. Iqbal and Lee reported that the equilibrium relation between different phases of the raw material changes due to the vitrification process which required higher temperature thus lead to the development of crystalline phase in porcelain (Lima et al., 2018). Literally, the compositional of raw materials of porcelain are widely presented in $\text{K}_2\text{O}-\text{Al}_2\text{O}_3-\text{SiO}_2$ phase system (Carty & Senapati, 1998; Iqbal & Lee, 2000).

The plasticity characteristic in clay or well known in industry as plastic clay or ball clay helps in giving shape in the forming process (Kingery et al., 1977). The kaolinite $\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4$ (clay mineral) contain of hydroxyl group dehydroxylated at 450–550 °C and transform into metakaolin, $2(\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2)$ (Iqbal & Lee, 2000). Above 900 °C, the aluminosilicate spinel ($2\text{Al}_2\text{O}_3 \cdot 3\text{SiO}_2$) phase form by the decompositions of metakaolin. Some of the researchers reported it as the $\gamma\text{-Al}_2\text{O}_3$ phase (Carty & Senapati, 1998). The aluminosilicate phase gives rise to the primary mullite above the temperature of 900 °C which can be seen as the agglomerate of small ($< 0.5 \mu\text{m}$) mullite crystal (Martín-Márquez et al., 2010). There are two types of mullite which are primary mullite and secondary mullite (Duval et al., 2008). The transformation of primary to secondary mullite forms through the reaction of clay relicts with feldspar melts at around 1200 °C and referred as elongated needle-shaped ($> 1 \mu\text{m}$) mullite crystal which is called as secondary mullite (Martín-Márquez et al., 2010). However, some of the researcher reported that all the primary mullite transformed to Secondary mullite at around the sintering temperature of 1350 – 1400 °C due to complete dissolution of quartz grains. This behavior allows the formation of porcelain microstructure consisting of glass, mullite and quartz (Carty & Senapati, 1998; Iqbal & Lee, 2000; Kingery et al., 1977). Dana and Das reported over the sintering temperature of 1300 °C, the microcracks commonly occurred around quartz grain on the porcelain microstructure due to large thermal expansion coefficient mismatch between quartz and silicate glass matrix (Dana & Das, 2004). Generally, quartz (filler) is an inert material with high melting point ($< 1300 \text{ °C}$) which could reduce the shrinkage and prevent the porcelain bodies to distort during firing process (Martín-Márquez et al., 2008). Meanwhile, feldspar as a flux consists of alkali and alkaline earth metal giving higher vitrification in porcelain. In addition, feldspar minerals also could influence the crystallization behavior where it could improve the mechanical strength of the porcelain. In

emphasizing the strength of the porcelain, several researchers used waste product as part of the raw materials of porcelain in order to improvise its quality. In fact, the waste materials influenced the crystalline phase behavior of porcelain. In the present study, feldspar partially replaced by fly ash and sintered in various sintering temperature. The aim of this study is to investigate the phase changes and mechanical behavior of the porcelain by the substitution of fly ash compositions according to the different sintering temperature (Martín-Márquez et al., 2008).

2. Materials and Methods

Rice Husk Ash (RHA) was collected from local farmers and heated with furnace at 800°C for 24 hours. The ash was treated with HCl acid to enhance the purity and remove unwanted materials. To maintain triaxial porcelain structure, 50 wt. % clay, 25 wt. % feldspar and 25 wt. % quartz was used. Quartz was replaced with treated RHA and mixed homogeneously in a Labkorean Ball Mill machine (model RS-1S) for 12 hours at a speed of 250 revolutions per minute. The mixed powder was dry pressed into pallets at mould pressure of 91 MPa and sintered at a temperature of 1050 °C, 1100 °C and 1150 °C for 2 hours soaking time, at heating rate of 5 °C per minute using electric furnace (Protherm Furnace PLF 130/15). The compressive strength was determined using Universal Testing Machine (UTM), and X-ray Fluorescence Machine (XRF) was used to study the chemical composition of the new material formed, the structure was studied through X-ray Diffraction Analysis.

3. Results and Discussion

Energy dispersive X-ray was first use to check the elemental composition of rice husk ash (RHA) and the feasibility of using it as quartz replacement, this is because, based on the literature, it has been reported that, the major chemical composition of quartz is Si (Jamo et al., 2015). Figure 1 below shows a graph of elemental composition of RHA.

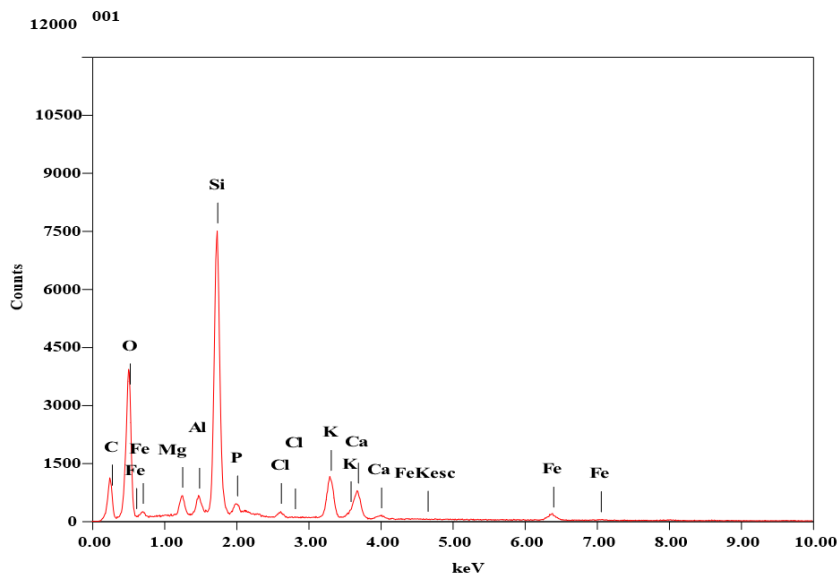


Figure 1: A graph of Elemental Composition of Rice Husk Ash (RHA)

When beam of X-rays is directed towards a material a fraction will be transmitted through while some will be absorbed, the interaction of X-ray with matter that resulted in absorption is known as fluorescent radiation, thus X-ray fluorescence analysis was used by different researchers as an efficient tool to analyze the chemical composition of RHA and quartz. To determine the feasibility of using rice husk ash as quartz replacement, RHA was investigated using X-ray Fluorescence analysis (XRF) and the result is presented in table 1 below. The table succinctly revealed the analogy of rice husk ash with quartz which is a good omen that RHA can be used as quartz replacement owing to their similar chemical composition. Furthermore, other chemical

compounds that play a vital role in the porcelain production such as Al_2O_3 , Fe_2O_3 , P_2O_5 , CaO and others are clearly in good agreement with other findings as regards to the composition of RHA and quartz. Table 1 shows the chemical composition of RHA and Quartz.

Table 1. The chemical composition of RHA and Quartz

Chemical Compounds (wt.%)	RHA	Quartz
SiO_2	54.32	56.16
Al_2O_3	4.74	4.71
Fe_2O_3	5.20	5.16
K_2O	5.62	5.36
C	6.21	7.43
MgO	1.88	2.32
P_2O_5	6.54	5.73
Cl	3.25	4.61
CaO	5.62	6.47
SO_3	0.90	0.94
TiO_2	0.30	0.29
ZnO	0.01	0.01
LOI	0.09	0.07

Bulk density is an important factor that determined the physical property of a porcelain material in the ceramic industries. Bulk density was plotted against sintering temperature and presented in figure 2 below. It can be noticed from the figure that, the highest density was achieved at 1100 °C sintering temperature with acid treated RHA as 2.45 g/cm³, for heat treated and untreated samples at the same sintering temperatures the values were 2.42 g/cm³ and 2.43 g/cm³ respectively. As the temperature increased, the density decreased and this could be related to the achievement of complete densification and subsequent porosity presence. Similarly, as the temperature increased to 1150°C the values of bulk densities for acid treated, heat treated and untreated sample were found to be 2.42 g/cm³, 2.41 g/cm³ and 2.42 g/cm³ respectively, which indicate the optimum temperature was achieved and over firing took place. Figure 2 shows the bulk density for acid treated, heat treated and untreated samples.

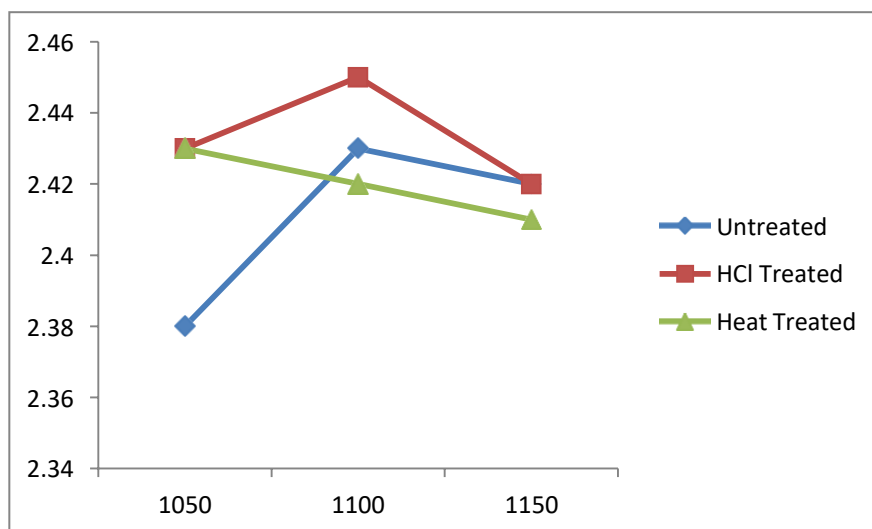


Figure 2. Bulk density for acid treated, heat treated and untreated samples.

Compressive strength was plotted against sintering temperature and the result is presented in figure 3. It is important to note that, compressive strength portray a similar pattern with bulk density. The highest value of compressive strength was achieved at 1100°C as 490 MPa for acid treated RHA, similarly for untreated and heat treated the values were 290 MPa and 350 MPa respectively. As the sintering temperature increased to 1150°C the values of compressive strength for HCl acid treated, untreated and heat treated were found to be 470 MPa, 450 MPa and 300 MPa respectively, indicating the maximum value with HCl acid treated sample. This pattern is clear indication that, treating RHA with HCl improved its purity and enhances the production of silicate material that helps in the production of glassy material and reduces porosity presence. Figure 3 shows the graph of compressivestrength versus sintering temperature.

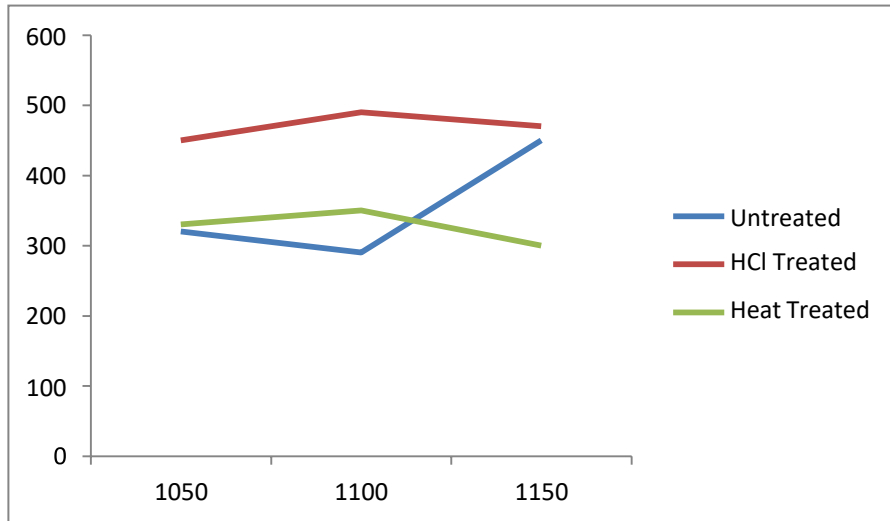


Figure 3. The graph of compressive strength versus sintering temperature

Figure 4 shows the XRD result. From the figure, it is further revealed that the main component of the porcelain was SiO₂ which is verified by chemical composition in XRF result presented in table 1, where SiO₂ is predominantly the main percentage of chemical composition. The crystalline phases in this pattern were quartz and mullite. Mullite is a stable phase in standard temperature that forms from the quartz. The acid treated sample has more mullite phases than other samples. In addition, the acid treatment sample has a cristoballite phase. The sample that has a higher composition of quartz cause the dissolution of SiO₂. Figure 4 shows the XRD pattern for HCl acid treated, heat treated and untreated samples.

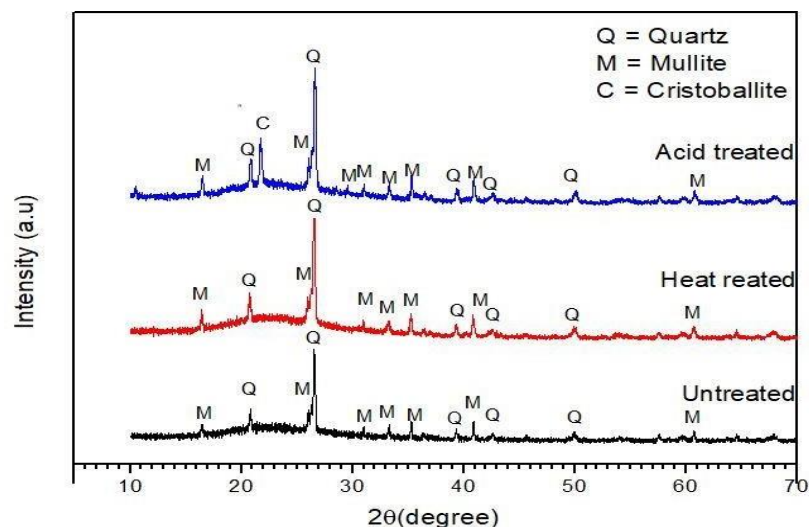


Figure 4. The XRD pattern for HCl acid treated, heat treated and untreated samples.

Figure 5 shows the result of Scanning Electron Microscopy. It can be deduced from the micrographs that, the acid treated sample has closed porosity and open porosities. Open porosity appears due to incomplete densification which has fine and interconnected pores with irregular shapes while closed porosity was isolated and it is spherical because it was larger. Besides that, Figure 5 additionally shows there was a glassy phase in the microstructure, and this could decrease the strength of the sample, as can be seen, the untreated sample has more glassy phases than the treated samples. Figure 5 shows the result of Scanning Electron Microscopy (SEM) for untreated, acid treated and heat-treated sample.

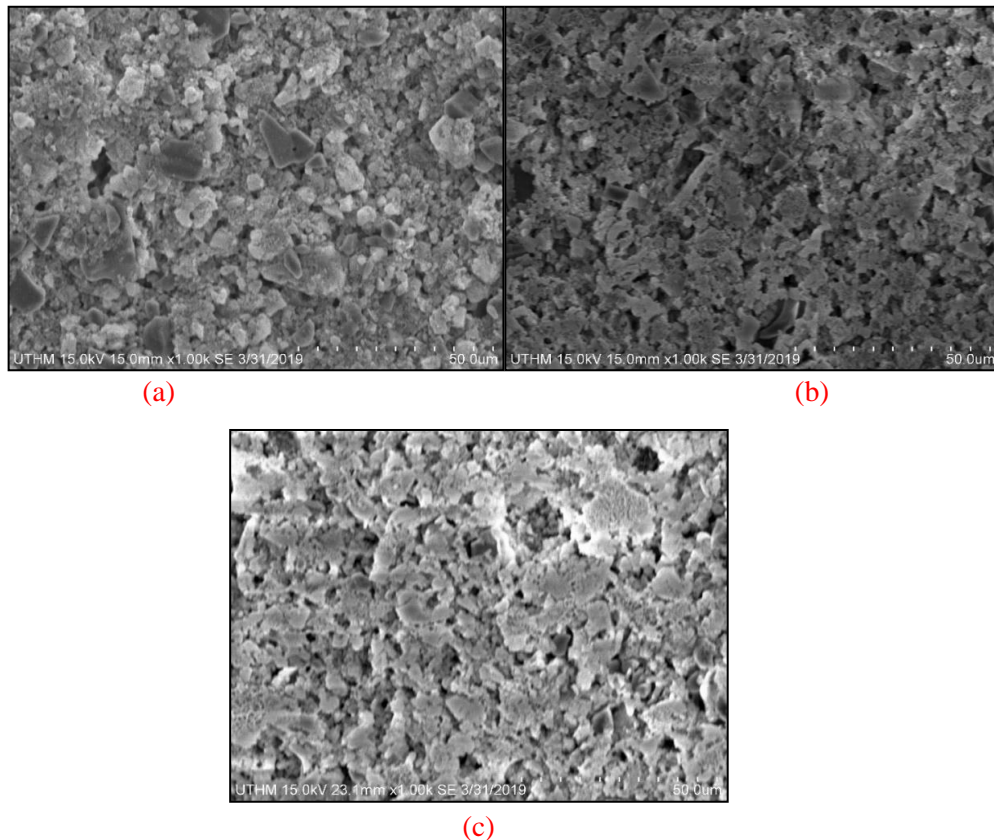


Figure 5 shows the result of Scanning Electron Microscopy (SEM) for untreated, acid treated and heat-treated sample.

4. Conclusion

The effect treated Rice Husk Ash (RHA) on the production of porcelain has been studied, based on the results of this research, it can be concluded that: treatment plays a vital role in the physical properties of Rice husk ash and porcelain. It can be noted that, the major chemical composition of RHA is SiO_2 which indicates that, it can be used as replacement of quartz. Furthermore, the bulk density was found to increase due to HCl acid treatment with highest value of 2.45 g/cm^3 at 1100°C . Similarly, the compressive strength also increased to 490 MPa for HCl acid treated from 290 and 350 for untreated and heat treated indicating overall maximum. Thus, Rice Husk Ash (RHA) is a good candidate for the replacement of quartz in the production of porcelain and HCl acid plays a vital role in enhancing and improving the physical and mechanical properties of porcelain.

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